A modal analysis of segregation of inertial particles in turbulence

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Abstract

An asymptotic solution is derived for prediction of segregation of heavy inertial particles in spatially and temporally varying flows with implications in particle-laden turbulence. The Lyapunov exponent is employed to identify regimes of particle segregation or mixing, where the Lyapunov exponent is negative or positive, respectively. The general solution, which classifies as the Fredholm integral equation of the second kind, is a function of the spectrum of the second invariant of the velocity gradient tensor of the underlying flow. We introduce a one-dimensional canonical flow oscillating at a single frequency to investigate the behavior of the Lyapunov exponent under a wide range of flow conditions. Through this canonical flow, we show that the Lyapunov exponent is a highly nonlinear function of the oscillation amplitude and frequency. In a straining regime, i.e. where the norm of the strain-rate is greater than the norm of rotation-rate tensor, the Lyapunov exponent can be positive or negative (either mixing and segregation can occur), whereas it is always positive in a rotating regime (only mixing is plausible). The minimum requirement for trajectory crossing is also predicted. The trajectory crossing never occurs in a rotating regime, whereas it occurs in a straining regime if the Lyapunov exponent is positive. Our analysis shows the Lyapunov exponent, normalized by the particle relaxation time, is \(-1/2\) at the minimum and grows linearly at small and nonlinearly with a power of 1/2 at large oscillation amplitudes. The direct numerical simulations confirm these predictions. The extension of our analysis to multimodal excitation is discussed and applied to a two-dimensional synthetic straining and a three-dimensional isotropic forced turbulent flow. The implication of our analysis to the realistic flow conditions is demonstrated by these examples. We show the results are analogous to that of the one-dimensional case, underscoring the relevance of our canonical setting and the fact that particle segregation arises from the general rather than particular solution of the Stokes drag equation. In comparison to two preexisting models, our model has a more general form and reproduces them under further simplifying assumptions. In contrast to these models that are valid only at small Stokes numbers, our model is valid at small and large Stokes numbers.

1. Introduction

The characterization, prediction, and design of a wide range of applications rely on understanding the dynamics of dispersed particles in turbulent flows in general and particle segregation in particular. The formation of planets and planetesimals in our early solar system is hypothesized to be a result of particle segregation \([1,2,3]\). The accurate prediction of weather relies on proper modeling of droplet coalescence in the clouds, a phenomenon with a direct connection to the problem of particle segregation \([4,5,6]\). The design of a particle-based solar receiver intended for the endothermic chemical reaction with high temperature requirement can be achieved through particles segregation \([7,8,9,10]\). Apart from the few preceding examples, numerous areas in science and engineering, ranging from sediment transport in environmental flows to pharmaceutical applications, also benefit from the better characterization of particle segregation \([11,12]\).

In a physically realistic scenario, particle segregation is governed by multiple non-dimensional parameters such as the density ratio of particle to carrier flow, the particle volume fraction, the size of particles relative to the characteristic length scale of the flow, Froude number, Stokes number, Reynolds number, Knudsen number, particle shape effects, and anisotropy of the underlying flow. While a wide range of behaviors is observed in different regions of this multidimensional parameter space, particle segregation is primarily
governed by a small subset of parameters. Hence, we focus on the subset of parameters with the largest influence on particle segregation in this study. Namely, we neglect the effect of particles on the flow and particle-particle interactions. These assumptions are particularly relevant to regimes with low volume and mass fractions. The finite-size effects are neglected by adopting a point-particle approach, which is to consider only particles that are much smaller than the smallest structure of the flow. The effect of flow inertia, thermal fluctuations, and body forces on the particle motion are neglected considering regimes with low Reynolds and Knudsen and high Froude numbers, respectively. To make the problem analytically tractable, we focus on the regime at which particles are much denser than the fluid and the flow is isotropic. In such a simplified regime, particle segregation can be quantified as a function of two dimensionless parameters that are the particle Stokes number and the flow Reynolds number.

The Stokes number represents the inertial of particles and is defined as the ratio of particle relaxation time $\tau = \rho_p d^2_p/(16 \rho f \nu)$ to the Kolmogorov time scale of the flow $\tau_\eta = (\nu/\epsilon)$, where $\rho_p$ is the particle density, $d_p$ is the particle diameter, $\rho_f$ is the fluid density, $\nu$ is the fluid kinematic viscosity, and $\epsilon$ is the mean volumetric dissipation rate. The Reynolds number, which can be considered as the ratio of largest to smallest structure of the flow, determines the range of time scales that a particle encounters along its trajectory. Particle segregation, as a function of the Stokes and Reynolds numbers, has been studied in the past extensively. It has been observed experimentally [13, 14, 15, 16, 25], simulated numerically [17, 18, 19, 20, 21, 22], and described analytically [23, 24, 25, 26, 27]. In this study, we focus on its analytical description and employ numerical simulations for verification purposes.

The particle motion subjected to the foregoing simplifications is well characterized by the Stokes drag. Denoting the position of a particle by $x(t)$ and the flow velocity at the particle location by $u(x, t)$, the dimensionless equation of the motion of a particle subjected to the Stokes drag is

$$\ddot{x} = u(x, t) - \dot{x},$$  \hspace{1cm} (1)

where $(\dot{\bullet}) := d(\bullet)/dt$. We employed $\tau$, $L$, and $U = L/\tau$ as the time, length, and velocity scales to nondimensionalize corresponding parameters in Eq. (1). All the following equations are also nondimensionalized based on $\tau$ and $L$. $\tau$ is the particle relaxation time, defined above, and $L$ is a characteristic length scale. The following formulations are independent of the choice of $L$, hence its choice is arbitrary. Parameters normalized based on the flow time scale rather than the particle relaxation time are distinguished by the subscript $\eta$. The only exception is $\tau_\eta$ which denotes the Kolmogorov time scale rather than particle relaxation time normalized by the flow time scale. Although Eq. (1) appears to be a simple ordinary differential equation, it behaves nonlinearly due to the dependence of $u$ on $x$. It is this nonlinear behavior that gives rise to particle segregation, which is a complex and nonlinear phenomenon.

The degree to which particles segregate in a turbulent flow strongly depends on the Stokes number. Depending on their Stokes number, particles may homogeneously disperse in space or preferentially concentrate in certain regions of the flow. For $St = \tau/\tau_\eta \ll 1$ particles become neutral fluid tracers and experience minimum segregation. There is also minimal segregation at the limit of $St \gg 1$, where particles follow a ballistic trajectory uncorrelated with the underlying flow. The maximum segregation is achieved when $St \approx 1$. The non-monotonic variation of particle segregation versus $St$ has been observed experimentally and numerically. However, this is yet to be described analytically.

One of the first analytical relations for quantification of particle segregation was obtained by [24] and [23]. This relation, which hereafter is referred to as RM, is obtained by approximating the acceleration of particles with that of the fluid. Following RM derivation by approximating $\ddot{x} \approx Du/Dt$ and assuming $\nabla \cdot u = 0$, i.e., flow being incompressible, and taking the divergence of Eq. (1), we obtain

$$\mathcal{C}(St) = \nabla \cdot \ddot{x} = - \text{tr} (\nabla u \nabla u) = ||\Omega||^2 - ||S||^2,$$  \hspace{1cm} (2)

where $\Omega$ and $S$ are the dimensionless rotation-rate and strain-rate and $\overline{\langle \dot{\bullet} \rangle}$ denotes Lagrangian time-averaging along the trajectory of a particle. The RM relates the degree of preferential concentration, here defined as $\mathcal{C}$, to the divergence of particle velocity field. From a Lagrangian perspective, $\mathcal{C}$ is the exponential rate at which the volume of a cloud of particles changes over a long period. In this description, the cloud is a collection of inertial particles that are within an infinitesimal distance from each other. The volume

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increase for $C > 0$ signifies the regimes of particle mixing (to be distinguished from diffusion induced mixing) and the volume decrease for $C < 0$ signifies the regimes of particle segregation. The distinction between this Lagrangian notion and the Eulerian definition though divergence operation becomes important when particles trajectories cross and is not a well defined function of the spatial coordinates $^{[26]}$. In what follows, we adopt the more general Lagrangian definition of $C$ for quantification of the particle segregation.

Approximating the acceleration of particles with that of flow limits the validity of RM to $St < 1$. The accuracy of RM in predicting the first- and second-order statistics of $C$ has been shown for $St < 1$ in homogeneous turbulence $^{[28, 29]}$ as well as synthetic flows $^{[30]}$. For $St \geq 1$, RM predicts an unbounded $C$ proportional to $St$ and fails to capture the non-monotonic behavior of $C(St)$. Additionally, Eq. $^{(2)}$ suggests that particles are repelled from the rotation-dominated regions, where $\Vert \Omega \Vert > \Vert S \Vert$, and preferentially concentrate in regions with higher strain-rate. These generally accepted qualitative assessments will be examined thoroughly in this study.

In an earlier attempt, we derived an alternative relationship for $C$ that is a first-order correction to RM $^{[29]}$. In that study, we linearized Eq. $^{(1)}$ and expressed $u$ in the Fourier space to obtain an eigenvalue problem for the Lyapunov exponents of pairs of inertial particles. The sum of these exponents (in three dimensions $\lambda_1$, $\lambda_2$, and $\lambda_3$) is equal to the divergence of the particle velocity field, $C$. We showed that $C$ can be expressed as

$$C(St) = \int_{-\infty}^{\infty} \frac{\rho^Q(\omega; St)}{1 + \omega^2} d\omega, \tag{3}$$

where

$$\rho^Q(\omega; St) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho^Q(t; St)e^{-i\omega t} dt = \frac{1}{2\pi} \int_{-\infty}^{\infty} tr \left(-\nabla u(t')\nabla u(t' + t)\right) e^{-i\omega t} dt. \tag{4}$$

$\rho^Q$ is half of the autocovariance of the Q-criterion, defined in the literature for identification of the vortical regions $^{[31, 32]}$. Its Fourier transformation $\tilde{\rho}^Q(\omega) = \tilde{\rho}^{\Omega}(\omega) - \tilde{\rho}^{\Omega}(\omega)$, where $\tilde{\rho}^{\Omega}$ and $\tilde{\rho}^{\Omega}$ are the spectrum of the norm of rotation-rate and strain-rate tensors, respectively. The sign of $\tilde{\rho}^{\Omega}$ indicates dominance of flow rotation-rate ($\tilde{\rho}^{\Omega} > 0$) or strain-rate ($\tilde{\rho}^{\Omega} < 0$), which are associated with elliptic and hyperbolic regions of the flow, respectively. Note these functions are computed along the trajectory of the particles, are different from a Eulerian averaged quantity, and thus dependent on $St$.

The primary assumption associated with Eq. $^{(3)}$, which we refer to as SL hereafter, is that the Lyapunov exponents $\lambda_j$ are much smaller than 1. This assumption is valid at relatively small $St$ in which $\lambda_j$ are also small. Comparing Eqs. $^{(2)}$ and $^{(3)}$, SL can be considered as the filtered RM which accounts for unresponsiveness of particles to high frequency fluctuations. As $St \to 0$, SL exactly reproduces RM. At this limit, $\omega^2$ in the denominator of Eq. $^{(3)}$ can be neglected, thus reproducing Eq. $^{(2)}$ exactly. Both SL and RM are a linear function of rotation-rate and strain-rate, predicting a stronger segregation proportional to $\Vert S \Vert - \Vert \Omega \Vert$. A comparison against the reference numerical results over a wide range of $St$ showed that SL and RM provide accurate predictions for small $St$. At higher $St$, although SL remains bounded and provides a better prediction than RM does, it still deviates from the reference due to the underlying assumption of $|\lambda_j| \ll 1$.

The objective of this study is to derive an analytical relationship for $C(St; \tilde{\rho}^{\Omega})$ that is valid at both small and large $St$. In what follows, we present a step-by-step derivation of an asymptotic solution, extracted from the linearized form of Eq. $^{(1)}$. Then, we compare the prediction of RM, SL, and our solution to the reference. For this purpose, we first consider a one-dimensional unimodal oscillatory flow. Through this case, we test the accuracy of the proposed solution and discuss its implications. Then, we extend our analysis to more complex multidimensional flows, oscillating at a continuous range of frequencies. We consider a two-dimensional synthetic straining flow and three-dimensional isotropic turbulence for this purpose. Prediction of RM, SL, and the present analysis will be compared at a wide range of Stokes numbers, studying their outcome in regimes of particle segregation and mixing. As a byproduct of our analysis, we establish a relationship between the second moments of the finite-time Lyapunov exponent of inertial particles pairs and their respective Lyapunov exponents.
2. Analytical derivation

In this section, the derivation of our asymptotic solution to Eq. (1) is presented. To begin, we define the trajectory of a particle that is located at \( X \) at time \( t = 0 \) as \( x = x(X, t) \). The trajectory of a nearby particle initially located at \( X + \delta X \) is then described by \( x(X + \delta X, t) = x(X, t) + (\partial x/\partial X)\delta X \). The deformation tensor associated with this motion is

\[
J := \frac{\partial x}{\partial X}.
\]

(5)

The determinant of \( J \), denoted by \(|J|\), represents the volume of a cloud of particles relative to its initial volume that undergone a linear deformation characterized by \( J \). Then, the finite-time exponential rate of change of the volume of the cloud is

\[
C_t := \frac{\ln |J(t + t')| - \ln |J(t')|}{t}.
\]

(6)

As shown in Appendix A, \( C_t \) is related to \( C \) by

\[
C = \lim_{t \to \infty} C_t.
\]

(7)

Based on Eqs. (6) and (7), \( C \) can be computed once \( J \) is determined. To obtain a relationship for \( J \), we take the derivative of Eq. (1) with respect to \( X \) and employ the chain rule to obtain

\[
\ddot{J} + \dot{J} = \nabla u J.
\]

(8)

To derive this equation, we employed \( \dot{J} = \partial \dot{x}/\partial X \) and \( \ddot{J} = \partial \ddot{x}/\partial X \), which hold true since \( X \neq X(t) \). Also, \( \nabla u \), which is a short-hand notation for \( \partial u/\partial x \), is expressed in terms of \( x \) and thus tractable by computing the gradient of fluid velocity along the trajectory of the cloud. The tensor \( \nabla u \) is a general function of time and as a result Eq. (8) is not a constant coefficient ordinary differential equation (ODE) to be integrated directly. On the other hand, \(|J|\) exponentially grows or decays indefinitely in time for \( C \neq 0 \). As discussed in the next section, this exponential variation hinders its accurate numerical computation. In this study we make use of

\[
F := \dot{J}J^{-1},
\]

(9)

a transformation that produces a more tractable constant coefficient ODE with favorable numerical properties. Based on this transformation,

\[
\dot{F} = \ddot{J}J^{-1} - F^2
\]

(10)

and hence from Eq. (8)

\[
\dot{F} + F^2 + F = \nabla u,
\]

(11)

which classifies as the Riccati equation in a tensorial form.

Equation (11), in contrast to Eq. (8), is nonlinear and has constant coefficients. It is expressed in terms of \( F \), which is an instantaneous rate of deformation of the cloud normalized by its size, and is independent of the arbitrary choice of \( X \). As a result, its determinant \(|F|\) is a statistically stationary variable for sufficiently long \( t \). To show that, we employ Jacobi’s formula,

\[
\text{tr}(F) = |J|^{-1} \frac{d}{dt} |J|,
\]

(12)

along with Eqs. (6) and (7) to obtain

\[
C = \text{tr}(F),
\]

(13)

in which \( \text{tr}(\bullet) \) is the trace operator. This simple relationship indicates that \( C \), which is the sum of the Lyapunov exponents, is the time average of the sum of the eigenvalues of \( F \). In other words, eigenvalues of \( F \) are the Lyapunov exponents associated with the pairs of inertial particles.

In a turbulent flow \( \nabla u \), which is computed along the trajectory of particles, is a bounded aperiodic function of time. \( F \) that is actuated by \( \nabla u \) is also aperiodic but may contain singular points at which
one of its eigenvalues becomes unbounded. In three dimensions, the singular points of $|F|$ corresponding to incidents of a spherical cloud collapsing to a planar ellipse. As a result the volume of the cloud $|J| \to 0$ and hence $|F| \to \infty$. For these singular points to occur, the particle Stokes number must be sufficiently high for the particle trajectories to cross. The occurrence of these singular points inhibits accurate numerical computation of the second moment of $|F|$. Thus, to circumvent this issue, we seek an alternative formula that is solely based on well-posed mean quantities.

The eigenvalues of $F$, denoted by $\lambda_i$, represent the instantaneous rate of expansion or contraction of cloud in the principal directions. To compute the sum of the eigenvalues, we take the trace of Eq. (11) to obtain

$$\sum_{i=1}^{N_{sd}} (\dot{q}_i + q_i^2 + q_i) = \nabla \cdot \mathbf{u}, \quad (14)$$

where $N_{sd}$ denotes the number of spatial dimensions. Averaging Eq. (14) over a period much longer than the integral time scale of $q_i$, the first term will be much smaller than the remaining terms. For a divergence-free flow or more specifically under less-restrictive condition $\nabla \cdot \mathbf{u} = 0$, the right-hand side of Eq. (14) will be zero, yielding

$$\sum_{i=1}^{N_{sd}} (\overline{q_i^2} + \overline{\dot{q}_i}) = 0, \quad (15)$$

where $\overline{\overline{q}}_i$ is the time averaged of the eigenvalues of $F$ and is equal to the Lyapunov exponents $\lambda_i$. Additionally, all $q_i$’s and $\lambda_i$’s are equal in a homogeneous flow. Thus

$$(q')^2 = - (\lambda^2 + \lambda), \quad (16)$$

where $q'$ denotes fluctuation of $q$, which is a representative $q_i$, around its time-averaged mean $\lambda$.

**Remarks on Eq. (16):**

1. Despite the singular points due to the collapse of clouds, $q'$ remains finite as long as the Lyapunov exponents are finite. To explain this apparent contradiction, one need to note that when a cloud collapses, both the real and imaginary parts of $q \to \infty$. The growth rate of these two components is such that the second moment of $q$ remains bounded. As a result, $(q')^2 = \overline{q^2} - \lambda^2$ remains bounded.

2. When $q$ and consequently $\lambda$ are real, $(q')^2 > 0$ if only if $-1 < \lambda < 0$. As shown later in the next section real $(\lambda)$ is always larger than $-1/2$ and the lower limit is always satisfied. $\lambda$ can also be imaginary and its real part may become larger than zero. These two conditions can occur simultaneously, particularly for $St \gg 1$, leading to $(q')^2 > 0$ when $\lambda > 0$. Also, $(q')^2 < 0$ is a plausible scenario that should be interpreted bearing in mind that $q$ is imaginary in general.

3. The sum of the finite-time Lyapunov exponents $C'$, computed by averaging $\sum q_i$ over a finite period $t$, also fluctuate due to the turbulence intermittency. Although the amplitude of these fluctuations depends on $t$, $\sqrt{\overline{\overline{q}}_i}$ is independent of $t$ for sufficiently large $t$. Due to the correlation between $q_i$’s, one should not employ Eq. (10) to estimate the normalized fluctuations of $C'$. For instance as $St \to 0$ in a divergence-free flow, $(\sum q_i) \to 0$ while individual $|q_i|$ remain finite. At this limit, $C'$ and its fluctuation tend to zero while $q'$ remains finite. As will be shown in Section 4.2 the asymptotic behavior of $q'$ and fluctuation of $C'$ also differ at the limit of large $St$.

Next, we solve Eq. (11) for $F$ to find an analytical estimate for $C$ via Eq. (10). Tensor $\nabla \mathbf{u}$ is a general function of time. To find an asymptotic solution for Eq. (11), we express $\nabla \mathbf{u}$ as a set of harmonic functions using the Fourier transformation. Defining

$$G(\omega) := \frac{1}{2\pi} \int_{-\infty}^{\infty} \nabla \mathbf{u}(t) e^{-i\omega t} dt, \quad (17)$$

\[ ^1 \lambda_i \text{ are based on } q_i(t) \text{ which are not sorted in a descending or ascending order by permuting } i \text{ but are ordered in association with } q_i(t - \delta t) \text{ such that } |q_i(t) - q_i(t - \delta t)| \text{ is minimized.} \]
Eq. (11) can be written as
\[ \dot{F} + F^2 + F = \sum_\omega G(\omega)e^{i\omega t}. \] (18)

Note \( \omega \in (-\infty, \infty) \) is implied in all summations without explicit bounds. Next, we consider an asymptotic solution to Eq. (18) with the following form
\[ F = \lambda I + \sum_\omega \epsilon_1(\omega)e^{i\omega t} + \sum_\omega \epsilon_2(\omega)e^{2i\omega t} + \cdots, \] (19)

where \( I \) is the identity tensor, \( \lambda \) is a representative Lyapunov exponent, and tensor \( \epsilon_i(\omega) \) is the \( i \)th-order oscillatory response of \( F \) to \( G \). For a given generic flow, the Lyapunov exponents are not equal. However, only a representative exponent is considered in Eq. (19) because the flow is assumed to be isotropic with no preferred direction. In this case, \( C \) can be computed (from Eqs. (19) and (13)) as
\[ C = N_{sd}\lambda. \] (20)

Neglecting the higher-order terms in Eq. (19), i.e., assuming \( \epsilon_i \approx 0 \) for all \( i > 1 \), and substituting it in Eq. (18) yields
\[ (\lambda + \lambda^2)I + \sum_\omega (1 + 2\lambda + \hat{i}\omega)\epsilon_1(\omega)e^{i\omega t} + \sum_\omega \sum_{\omega_n} \epsilon_1(\omega_n)\epsilon_1(\omega_m)e^{i(\omega_n + \omega_m)t} = \sum_\omega G(\omega)e^{i\omega t}. \] (21)

Assuming \( |\epsilon_1| \ll 1 \), all the time-dependent terms in the second summation can be neglected compared to those in the first summation. The remaining time-independent terms are retained as they might be comparable to the first term. Thus, keeping only the terms with \( \omega_n = -\omega_m \) yields
\[ (\lambda + \lambda^2)I + \sum_\omega (1 + 2\lambda + \hat{i}\omega)\epsilon_1(\omega)e^{i\omega t} + \sum_\omega \epsilon_1(\omega)\epsilon_1(-\omega) = \sum_\omega G(\omega)e^{i\omega t}. \] (22)

For Eq. (22) to hold,
\[ \epsilon_1(\omega) = (1 + 2\lambda + \hat{i}\omega)^{-1}G(\omega), \]
\[ \lambda + \lambda^2 + N_{sd}^{-1} \sum_\omega \text{tr} \{\epsilon_1(\omega)\epsilon_1(-\omega)\} = 0, \] (23)

which is a necessary but not a sufficient condition. The additional condition necessary for Eq. (22) to hold is \( \sum_\omega \text{tr} \{\epsilon_1(\omega)\epsilon_1(-\omega)\} \) to be a diagonal matrix, which occurs when the non-identical entries of the velocity gradient tensor are uncorrelated. From Eq. (23)
\[ \lambda + \lambda^2 + N_{sd}^{-1} \sum_\omega \frac{\text{tr} \{G(\omega)G(-\omega)\}}{(1 + 2\lambda)^2 + \omega^2} = 0. \] (24)

Using the convolution theorem, Eq. (24) is expressed in terms of a continuous integral as
\[ \lambda + \lambda^2 - N_{sd}^{-1} \int_{-\infty}^{\infty} \frac{\tilde{\rho}^Q(\omega)}{(1 + 2\lambda)^2 + \omega^2}d\omega = 0, \] (25)

where \( \tilde{\rho}^Q \) is the spectrum of Q-criterion defined in Eq. (4).

Remarks on Eq. (25):

1. This equation, which will be further developed in Section 3 and 4, classifies as the Fredholm integral equation of the second kind. Evaluating the integral that appears in this equation requires a knowledge of \( \lambda \), which itself is the solution. Hence, obtaining an explicit expression for \( \lambda \) relies on further simplification and requires additional assumptions.
2. In relation to this equation, RM and SL expressions are special forms that can be reproduced exactly by adopting further assumptions. Linearizing Eq. (25) at the limit of $|\lambda| \ll 1$ and using Eq. (20) to express it in terms of $C$ exactly reproduces SL (Eq. (3)). Furthermore, neglecting $\omega^2$ in the denominator reduces the integral to $\rho^2(0) = \|\Omega\|^2 - \|S\|^2$, exactly reproducing RM (Eq. (2)).

3. No length scale appears in this equation, justifying our earlier arbitrary choice of $L$. The choice of time scale, on the other hand, affects terms with $\omega$, $\lambda$, and $\rho^2$. These parameters are all normalized by the particle relaxation time $\tau$ in the present form of this equation. In most physical scenarios, $\rho^2$ is governed primarily by the underlying flow rather than $\tau$ and a natural choice for re-normalization of Eq. (20) is $\tau_\eta$. If normalized based on $\tau_\eta$, $St$ will appear in the denominator of the integral, compatible with the notion that the large $St$ particles barely respond to the flow fluctuations.

4. Contrary to RM and SL, $\lambda$ or $C$ vary nonlinearly as $\rho^2$ is changed. Scaling $\rho^2$ by a factor of $k$, clouds contract or expand $k$ times faster only if $k \to 0$. In the limit $k \to \infty$ and if $\lambda \neq -1/2$, $\lambda$ will scale as $k^{1/4}$. The numerical results of Section 3.1 confirm this asymptotic prediction.

5. Similar to RM and SL, the only term that appears in this equation which relates $\lambda$ to the underlying flow is $\rho^2$, which is computed from the second invariant of the velocity gradient tensor. Thus, as expected, this equation is invariant under Galilean transformation and rotation of the coordinate system. The absence of other flow-related parameters suggests that particle segregation, although complex, solely depends on the $Q$-criterion.

In what follows, we simplify Eq. (25) to find an explicit expression for $\lambda$ for a case in which the underlying flow oscillates at a single frequency.

3. Analysis of unimodal excitation

In this section, we consider the case in which $\rho^2$ is a single-harmonic oscillatory function of time. This model problem is studied for its fundamental importance in revealing the response of Eq. (25) to the entire parameter space. This investigation will provide us with a picture of various possibilities that may emerge in more complex flows.

We begin by considering two inertial particles that are confined to move along a straight line in one dimension. These two particles are subjected to the Stokes drag that is proportional to their relative velocity to the fluid velocity. The fluid velocity is prescribed to vary linearly along the line, producing a constant velocity gradient in space. The velocity gradient is prescribed to be a harmonic function with a given frequency and amplitude. Analytically, this flow is represented by

$$\nabla u := \sqrt{-2\Phi} \cos(\omega t)$$

where $\Phi$ and $\omega$ are the root square of amplitude and frequency of the oscillations, respectively. Note that both straining and rotating flow can be represented in this setting using $\Phi < 0$ and $\Phi > 0$, respectively. Interpreting Eq. (26) is simple in the straining regime, since $u$ is real and varies linearly along the line with all fluid particles moving away or toward a single point. Its interpretation in the rotation regime, on the other hand, requires imagining the line to have an oscillatory rotational motion with all fluid particles oscillating along concentric arches. From Eq. (11), one can proof that the rotating regime described in Eq. (26) with $\Phi > 0$ produces a Lyapunov exponent that is identical to that of a forced vortex defined by $u = [x_2, -x_1] \sqrt{2\Phi} \cos(\omega t)$ in two dimensions. In contrast to the one-dimensional case, $\nabla u$ is real in the two-dimensional case, allowing one to carry out the computations in the real plane.

Scenarios that may occur in the foregoing setting are shown schematically in Figure 1. In the straining regime, particles may diverge (cases I and II) or converge (cases III and IV) to each other in the long term. This convergence or divergence may occur while particles crossover (cases I and III) or otherwise (cases II and IV). In the rotating regime, there are two possible scenarios, which are divergence (case V) or convergence (case VI) of particles. Some of these cases may never occur in reality (e.g. case VI for heavy particles). However, regardless of the scenario, the rate at which the distance between two particles changes is exponential. This exponential behavior is caused by the difference between the underlying flow velocity at the location of particles being proportional to their distance. The exponential rate at which these particles
Figure 1: Possible relative motion of two particles in a one-dimensional straining (left) and rotating (right) flow. They diverge with crossover (I), diverge without crossover (II), converge with crossover (III), converge without crossovers (IV), diverge while rotating (V), and converge while rotating (VI). In cases I to IV curves correspond to the location of particles as a functions time, whereas in cases V and VI, curves are the position of particles in an imaginary plane.

converge or diverge to each other as \( t \to \infty \) is by definition the Lyapunov exponent \( \lambda \). \( \lambda < 0 \) (cases III, IV, and VI) represents the regimes at which particles segregate (particles occupy less space over time). \( \lambda > 0 \) (cases I, II, and V) represents the regimes at which particles mix (particles occupy more space over time). Our goal in this section is to find an analytical relationship for \( \lambda \) that delineates between the above scenarios.

The setup described above can be characterized using Eq. (25) by taking \( N_{sd} = 1 \) and based on Eq. (26)

\[
\tilde{\rho}^Q(\omega') = N_{sd}\delta(\omega - \omega'),
\]

where \( \delta \) is the Kronecker delta function. The sign of \( \tilde{\rho}^Q \) reaffirms our earlier distinction between the rotating (\( \Phi > 0 \)) and straining (\( \Phi < 0 \)) regimes. Substituting \( \tilde{\rho}^Q \) into Eq. (25) simplifies it to

\[
\lambda + \lambda^2 - \frac{\Phi}{(1 + 2\lambda)^2 + \omega^2} = 0.
\]

In total, there are two non-dimensional parameters that appear in Eq. (28), which are \( \Phi \) and \( \omega \). Since \( \Phi \) and \( \omega \) are normalized by \( \tau^2 \) and \( \tau \), respectively, the effect of Stokes number is embedded in both parameters. Equation (27) and consequently Eq. (28) also represent multidimensional isotropic flows as long as \( \tilde{\rho}^Q \) contains only a single frequency.

Next, we find a relationship for \( \lambda = \lambda(\Phi, \omega) \) using Eq. (28). According to Eq. (28), \( \lambda \) is one of the roots of a fourth-order polynomial. Closer examination shows that this polynomial has two real roots for \( \Phi > -\omega^2/4 \) or \( \Phi > 0 \), no real roots for \( \Phi < -(\omega^2 + 1)^2/16 \) or \( -(\omega^2 + 1)^2/16 < \Phi < -\omega^2/4 \), and four real roots otherwise (Figure 2). Out of four roots of this polynomial, only one is of physical relevance. It is that root that determines \( \lambda \) and the occurrence of particle crossovers. When there is no crossover, the distance between particles remains positive and \( \lambda \) remains real. In case of crossover, \( |J| \) changes sign and \( \lambda \) becomes imaginary (note the dependence of \( \lambda \) on \( \ln(|J|) \)).

The roots of Eq. (28) can be analytically computed by converting it to a depressed quadratic form and taking

\[
\gamma := 1 + 2\lambda.
\]

With this change of variable, Eq. (28) becomes

\[
\gamma^4 + (\omega^2 - 1)\gamma^2 - 4\Phi - \omega^2 = 0.
\]
\[ \Phi = -\left(\omega^2 + 1\right)/16 \]

No contraction or expansion is expected in the absence of flow. Thus, \( \lambda = 0 \) which is \( \gamma = 1 \) condition must be satisfied when \( \Phi = 0 \). Therefore, from the two possible solutions in Eq. (31), only the root corresponding to the plus sign is admissible. Computing \( \lambda \) from Eqs. (29) and (31) and imposing the condition \( \lambda(\Phi = 0, \omega) = 0 \) for one more time gives

\[ \lambda = -\frac{1}{2} + \frac{\sqrt{2}}{4} \sqrt{1 - \omega^2 + \sqrt{(\omega^2 + 1)^2 + 16\Phi}}. \]

Based on this equation \( \lambda \) can be complex for certain combination of \( \Phi \) and \( \omega \), pertaining to the occurrence of particle crossovers. The real and imaginary part of \( \lambda(\Phi, \omega) \) are plotted in Figure 3.

Remarks on Eq. (32):

1. \( \text{real}(\lambda) \) represents the rate of expansion (\( \text{real}(\lambda) > 0 \)) or contraction (\( \text{real}(\lambda) < 0 \)). Since the second term in Eq. (32) is always positive, the strongest possible rate of contraction is \(-1/2\). For \( \omega < 1 \), the minimum value of real(\( \lambda(\Phi) \)) occurs at the discriminant curve \( \Phi = -(\omega^2 + 1)^2/16 \), at which real(\( \lambda(\Phi) \)) is not differentiable. For \( \omega > 1 \), the minimum occurs in a region enclosed between \(-\omega^2/4 < \Phi < -\omega^2/4 \), where real(\( \lambda(\Phi, \omega) \)) is constant and equal to the global minimum \(-1/2 \) (Figure 3).
2. No expansion or contraction is predicted for two cases. The first is the trivial case in which \( \Phi = 0 \). The second case occurs at \( \Phi = -(\omega^4 + 10\omega^2 + 9)/16 \). In the latter case only real(\( \lambda(\Phi) \)) = 0 and imag(\( \lambda(\Phi) \)) \neq 0, indicating pure oscillation of the volume of the clouds between positive and negative values (a case between I and III in Figure 1).
3. In the rotation dominated regimes \( \Phi > 0 \) where the flow is elliptical, the clouds of particles always experience pure expansion \( \text{real}(\lambda) > 0 \). No crossovers occurs in this regime (Figure 3).
4. Contrary to the contraction-rate, the expansion-rate is unbounded. In general real(\( \lambda(\Phi) \)) is proportional to \( \Phi \) for \( |\Phi| \ll 1 \) and to \( |\Phi|^{1/4} \) for \( |\Phi| \gg 1 \).
5. As discussed above, imag(\( \lambda(\Phi) \)) should not be interpreted as the mean rate of rotation of clouds. imag(\( \lambda(\Phi) \)) \neq 0 is the byproduct of \( |J| < 0 \), occurring in strong-straining flows as clouds fully collapse and turn inside-out. In the 1D setting, imag(\( \lambda(\Phi) \)) \neq 0 when two particles crossover each other and exchange sides on the line.
6. Particle crossover occurs when \( \Phi < \max \left[ -\left( \omega^2 + 1 \right)^2/16, \min \left( -\omega^2/4, -1/4 \right) \right] \) (blue region in Figure 2). The maximum of \( \text{imag}(\lambda) \) occurs at \( \Phi = -\left( \omega^2 + 1 \right)^2/16 \) for \( \omega > 1 \), at which \( \text{imag}(\lambda) \) is not differentiable.

7. Out of six scenarios in Figure 1 only four is predicted to occur in the 1D setting. In a straining regime cases I, III, and IV are possible (Figure 2) while in a rotating regime only case V occurs. In other words, particles have to crossover to diverge in a straining flow, whereas they may or may not crossover when they converge. Thus, in a straining flow, particle mixing occurs only if particle trajectories cross, whereas they can segregate regardless of the occurrence of crossovers.

In the next section, we compare the prediction of Eq. (25) to the numerical results.

3.1. Numerical validation

To validate the present analysis and compute \( \lambda(\Phi, \omega) \), we numerically integrate Eq. (11), which is an equivalent form of Eqs. (1) and (8). Computations are performed based on Eq. (11) for its desirable numerical properties, avoiding the ill-conditioning issue associated with the exponential growth of \(|J|\). Given the setup of the unimodal excitation case, \( \nabla u \) is prescribed based on Eq. (26) for different values of \( \omega \) and \( \Phi \). Since this source term is imaginary for \( \Phi > 0 \), the integration is performed in the complex plane. To ensure stability and accuracy, we adopt the implicit Euler time integration scheme. Using Eq. (11), \( F \) is computed on a set of discrete time points such that

\[
\frac{F_{h}^{n+1} - F_{h}^{n}}{\Delta t^n} + F_{h}^{n+1} + (F_{h}^{n+1})^2 - \nabla u_{h}^{n+1} = 0, \tag{33}
\]

in which subscript \( h \) denotes a variable in the discrete setting, \( n \) is the time stepping index, and \( \Delta t^n \) is the time step size. The initial condition for Eq. (33) is \( F(t = 0) = F_{h}^{0} = I \). From Eq. (33)

\[
F_{h}^{n+1} = \sqrt{\left( \frac{1}{2} + \frac{1}{2\Delta t^n} \right)^2 I + \frac{1}{\Delta t^n} F_{h}^{n} + \nabla u_{h}^{n+1} - \left( \frac{1}{2} + \frac{1}{2\Delta t^n} \right) I} = 0. \tag{34}
\]
\[ \omega = 10 \]

\[ \lambda = 0.09 \]

\[ \lambda = -0.1 \]

\[ \phi = -10 \]

\[ \omega = 0.1 \]

\[ \omega = 1 \]

\[ \omega = 10 \]

\[ \lambda = 0.09 \]

\[ \lambda = -0.1 \]

**Figure 4:** The time variation of the distance between two particles \(|J|\) subjected to an oscillatory velocity gradient (Eq. (26)) at three frequencies. Curves are obtained from the numerical integration of Eq. (11). The time-averaged slope of these curves provides a numerical estimate for \(\lambda\). The spikes in these curves correspond to the singular point of \(F\) associated with particle crossovers.

In the case of multidimensional flows, the radicand in Eq. (34) is a tensor. Hence, the computation of the root-square would involve an eigenvalue decomposition. Two other challenges that may arise in the numerical integration of Eq. (34) are the singularity and slow dynamics of \(F\). These two challenges must be addressed for an accurate computation of \(\lambda\) or \(C\).

The singularity issue emerges from the homogeneous form of Eq. (11), which in one dimension is \(\dot{F} = -F^2 + F\). Once \(F < -1\), one can show that \(F\) grows to \(-\infty\) and becomes singular (spikes in Figure 4). A singularity point corresponds to the occurrence of particle crossover or \(|J|\) change of sign. In the discrete setting, \(|F_h| \rightarrow -\infty\) at the singular points as \(\Delta t \rightarrow 0\), inhibiting the numerical accuracy of the time integration scheme. To bound the error, in our calculations \(\Delta t^n\) is adapted based on an error estimator. Since the error is proportional to \(\ddot{F}_h\), \(\Delta t^{n+1}\) is reduced if it is larger than a given minimum and \(|F_n^{n+1} + F_{n-1}^{n} - 2F_n^{n}|\) is larger than a given tolerance. The tolerance and the minimum time step size are \(10^{-5}\) and \(2\pi \times 10^{-6}/\omega\), respectively, in our calculations (\(\omega\) is the excitation frequency of Eq. (34)). Conversely, the time step is increased if the estimated error is smaller that \(10^{-7}\) and \(\Delta t < 2\pi \times 10^{-2}/\omega\).

The slow dynamics of \(F\) occurs when \(|F_h|\) in the Fourier space contains frequencies much lower than \(\omega\). The nonlinear term in this equation usually generates only super-harmonics \(\omega, 2\omega, 3\omega, \ldots\). For certain combination of \(\omega\) and \(\Phi\), however, sub-harmonics at frequencies much lower than \(\omega\) are also present in the solution. The presence or absence of sub-harmonics has implications on the period of integration. In the absence of sub-harmonics, it is sufficient to integrate Eq. (34) for two cycles, i.e., for \(4\pi/\omega\). The first cycle is discarded to remove the effect of the initial condition and the second cycle is averaged to compute \(\lambda\). However, in the presence of sub-harmonics, the integration period must be extended to accommodate for the smallest frequency. To capture all frequencies in the solution, we follow a dynamic procedure by adjusting the number of computed cycles for any given \(\omega\) and \(\Phi\). In this dynamic method, after discarding the first cycle, Eq. (34) is integrated for two sets of cycles with each set containing \(N_c\) cycles. Starting with \(N_c = 1\), \(\lambda\) is computed from two sets of cycles and compared to be within a given tolerance. If the difference is larger, \(N_c\) is doubled and the integration is continued for another \(N_c\) cycles. The result is compared to the average of the previous \(N_c\) cycles. This process is repeated until it converges. \(N_c\) was as large as \(2^{14}\) for the considered discrete set of \(\omega\) and \(\Phi\).

Once \(F(t; \omega, \Phi)\) is calculated, \(\ln(|J|)\) can be computed from Eq. (9) as the time integral of \(F\). The behavior of \(\ln(|J|)\) as a function of time widely varies depending on \(\omega \) and \(\Phi\) (Figure 4). The slope of these curves is equivalent to \(\text{tr}(\dot{F})\), which based on Eq. (13) is \(C\). Since \(N_{sd} = 1\), this slope is identical to \(\lambda\) (Eq. (20)) and provides a numerical estimate for it. We refer to \(\lambda\) or \(C\) obtained from this procedure as...
Figure 5: The rate of expansion or contraction real (\(\lambda\)) (left) and crossover imag (\(\lambda\)) (right) of a particle cloud obtained from the numerical integration of Eq. (11) for a flow excited at frequency \(\omega\) with amplitude \(\Phi\). Dashed lines are predicted discriminants from our analysis and replicated from Figure 3.

The numerically computed \(\lambda\) is shown in Figure 5. This figure shows that \(\lambda\) is a highly nonlinear function of \(\omega\) and \(\Phi\). For certain combination of \(\omega\) and \(\Phi\), \(\lambda\) is not differentiable or has a very sharp gradient. These results confirm the possibility of expansion (mixing) in straining flows, where \(\Phi < 0\) and real (\(\lambda\)) > 0. They also confirm the existence of a bound on the contraction-rate that never drops below \(-1/2\). The asymptotic behaviors \(\lambda \propto |\Phi|^{1/4}\) as \(|\Phi| \to \infty\) and \(\lambda \propto \Phi\) when \(\Phi \ll 1\) are also confirmed by the numerical results. These numerical observations are in full agreement with the prediction of our analysis (Figures 3 and 5).

The numerically computed imag (\(\lambda\)) is also nonzero for certain combination of \(\omega\) and \(\Phi\). In those cases, imag (\(\lambda\)) can be either positive or negative depending on the initial condition. Its magnitude, however, is unique and depends on the rate of crossovers. According to Figures 3 and 5 there is an excellent agreement between the numerical and analytical results in terms of \(\omega\) and \(\Phi\) at which imag (\(\lambda\)) \(\neq 0\), viz. the blue region in Figure 2. The agreement between the numerical and analytical results shows our analysis correctly predicts the regimes of particle crossover.

Discrepancies between the numerical and analytical results are also observed, specifically for \(\Phi \lesssim -1\) and \(\omega \lesssim 1\) where imag (\(\lambda\)) \(\neq 0\) (blue region in Figure 2). A higher degree of non-linearity is observed in the numerical result for \(\Phi < -(\omega^2 + 10\omega^2 + 9)/16\). Additionally for \(\omega > 1\), while our analysis predicts real (\(\lambda\)) = \(-1/2\) for \(-(\omega^2 + 1)^2/16 < \Phi < -\omega^2/4\), the numerical result shows a broader range of \(\Phi\) producing real (\(\lambda\)) = \(-1/2\). Fitting a curve to the numerical result shows real (\(\lambda\)) = \(-1/2\) for \(-(\omega^2 + 1)(|\omega| + 1/\sqrt{2})^2/16 < \Phi < \omega^2/4\), which includes \(\Phi\) slightly lower than the analytical prediction (Figures 6). In overall, however, the discriminant curves obtained from our analysis provide a good approximation for the values at which the numerical solution changes sign or reaches a plateau.

\(\Phi\) and \(\omega\) are normalized by the particle relaxation time \(\tau\) and thus, are proportional to St. This correspondence implies that \(\Phi \gg 1\) and \(\omega \gg 1\) represent particles with high St. As a result, much of the

-10 -5 0 5 10
0 2 4 6 8 10
-0.5 -0.4 -0.3 -0.2 -0.1 0 0.1 0.2 0.3 0.4 0.5

\(0 < \omega < 10\) and \(\Phi \in [-10, 10]\).
depicted parameter space in Figures 3 and 5 is relevant to high St regime. Our analysis captures the general trend of $\lambda(\Phi, \omega)$ in this part of parameter space. However, extremely narrow valleys of parameter space with $\text{real}(\lambda) < 0$ (e.g. $\Phi = -10$ and $\omega = 1$) are missing in the analytical result (Figure 6). The pattern produced by these valleys resemble a fractal structure. The distance between valleys reduces and they become shallower as $\omega \to 0$. For a given $\omega < 1$, these valleys appear only at $\Phi < -\left(\omega^2 + 1\right)(|\omega| + 1/\sqrt{2})^2/16$ (Figure 6). These valleys may override the extremum of $\text{real}(\lambda)$ predicted at $\Phi = -\left(\omega^2 + 1\right)^2/16$ by our analysis. As discussed in more details in Section 3.4 the predicted extremum agrees with the numerical result when $\omega \to 0$. Neglecting the local extremum associated with the valleys at finite $\omega < 1$, the remainder of $\text{real}(\lambda(\omega, \Phi))$ is a smooth envelope that has an extremum at the predicted $\Phi = -\left(\omega^2 + 1\right)^2/16$.

### 3.2. Higher order expansions

To explain the difference between the analytical prediction and the numerical result, we need to revisit the underlying assumptions of Eq. (32). Equation (32) is an exact solution of Eq. (25) that was asymptotically obtained from Eq. (11) when higher order terms, i.e., $\epsilon_i$ for $i > 1$, are negligible. Neglecting these higher order terms constitutes one of the primary approximation of our analysis. The higher order terms modify $\lambda$ in two ways. The first is to interact with the lower order terms and alter their amplitude. The second is to directly contribute to $\lambda$ through contraction of $\epsilon_i(\omega)$ and $\epsilon_i(-\omega)$. In either case, $|\epsilon_i|$ provides a measure of the significance of those higher order terms. Therefore, we evaluate the primary approximation of our analysis by investigating the decay rate of $|\epsilon_i|$ versus $i$.

The first three terms of the asymptotic expansion are derived and provided in Table 1. As the order of expansion $n$ increases, additional terms appear in the lower order terms as a result of the interaction between higher order terms. All these additional terms are produced by the nonlinear term in Eq. (11) that turns into a convolution in the Fourier space. Further analysis of these terms shows that the decay rate of $|\epsilon_i|$ depends primarily on $|\Phi|/\omega$. In general $|\epsilon_i| \propto (|\Phi|/\omega)^i$ if $|\Phi|/\omega < 1$ and in the worst-case scenario $|\epsilon_1| \approx |\epsilon_2| \approx \cdots \approx |\epsilon_n|$. In the latter case, the asymptotic form in Eq. (19) will not converge. To show this behavior in practice, $\epsilon_i$ is derived by continuing Table 1 beyond $n = 3$. $\epsilon_i$ and $\lambda$ are then calculated iteratively for several values of $\omega$ and $\Phi$. The decay rate of $|\epsilon_i|$ versus $i$ is then computed and shown in Figure 6.
This figure confirms that $|\epsilon_i|$ may not decay monotonically if $|\Phi|/\omega \ll 1$, as is the case of $\Phi = 10$ and $\omega = 0.1$. Non-converging $|\epsilon_i(\omega, \Phi)|$ is particularly observed where the prediction of Eq. (25) disagrees with the reference numerical result. However, neglecting the higher order terms accounts for some the observed discrepancies.

Table 1: The leading order terms in the asymptotic solution of Eq. (11). The solution of the first order expansion ($n = 1$) for $\lambda$ is already provided in Eq. (25). For $n > 1$, $\epsilon_i$ and $\lambda$ must be calculated iteratively. For arbitrary matrices $A$ and $B$, $[AB]^n := (AB + BA)/2$.

For certain combination of $\omega$ and $\Phi$, including higher order terms does not produce a better estimate for $\lambda$. In these cases, $|\epsilon_i|$ may not even decay for $i > 1$, suggesting that the assumed form in Eq. (19) is incomplete. This form assumes that an excitation at $\omega$ would generate a solution oscillating at $\omega$, $2\omega$, $3\omega$, $\cdots$, producing only super-harmonics. The numerical simulation shows, however, that the solution may oscillate at lower frequencies, producing sub-harmonics. A closer examination shows that the amplitude of sub-harmonics can far exceed that of super-harmonics. It is these sub-harmonics that cause highly nonlinear behaviors that are missing in the prediction of present analysis. Identifying the nontrivial patterns of these sub-harmonics and thereby accounting for their contribution to $\lambda$ remains as a topic for the future studies.
3.3. Comparison against other models

As alluded to earlier, the present analysis is a more general form of our former [29] and Maxey’s [23] analysis. For the case of one-dimensional unimodal excitation, SL is simplified by combining Eqs. (3), (20), and (27) as

$$\lambda = \Phi \frac{1 + \omega^2}{1}$$  \hspace{1cm} (35)

and similarly, RM is simplified by combining Eqs. (2), (20), and (27) as

$$\lambda = \Phi$$  \hspace{1cm} (36)

These two relationships can be derived from Eq. (32) or Eq. (30) under the assumptions of $|\lambda| \ll 1$ and $|\omega| \ll 1$ as well.

RM and SL are valid only within the regimes compatible with their underlying assumptions (Figure 8). RM is derived for small St, which translates to $\omega \ll 1$ and $|\Phi| \ll 1$ (unless $|\Phi| \approx 0$, where there is no flow and $\lambda \approx 0$). SL is derived for small real ($\lambda$), which translates to $\omega \gg 1$ or $|\Phi| \approx 1$. Present analysis, on the other hand, provides a reasonable approximation at all $\omega$ and $\Phi$. To highlight these differences, the predictions of all models are compared against the reference numerical results in Figures 8 and 9. Figure 9(c) confirms RM and SL predictions are invalid at small $\omega$, except for $\Phi \ll 1$. These two relations predict $\lambda = -5$ at $\omega = 0$ and $\Phi = -5$, whereas $\lambda = 0.3$ and 0.29 from the numerical calculations and our analysis, respectively. At higher $\omega$, SL prediction approaches the numerical solution for a wide range of $|\Phi|$, while RM prediction is wrong everywhere except $|\Phi| \approx 0$ (Figure 9(e) and Figure 8).

The nonlinear behavior of real ($\lambda$) at high St regime is captured only by our analysis. According to Eqs. (35) and (36), RM and SL are both linear functions of $\Phi$. Hence, they both fail to predict nonlinear behavior of real ($\lambda$) at higher $|\Phi|/\omega$ (Figures 5 and 8). Neither RM or SL predicts the lower bound on the contraction rate ($\min(\lambda) \geq -0.5$), possibility of expansion in straining flows ($\lambda > 0$ for $\Phi \ll -\omega^4$), and the asymptotic variation of expansion or contraction rate at high amplitude oscillations ($\lambda \propto |\Phi|^{1/4}$ for $|\Phi| \gg 1$). These two models also fail to predict particle crossovers that occur in strong-straining flows where imag ($\lambda$) $\neq 0$.

3.4. One dimensional straining and rotating flows

All the derived equations so far have been normalized based on the particle relaxation time $\tau$. The choice of $\tau$ for the time scale led to compact equations in which only an oscillation amplitude and frequency appears. This normalization translates to experiments in which a single class of particles is reused in a variety of flows. Often in practice, however, we encounter multiple classes of particles in one particular flow. To predict the
trends observed in the latter case, the previous results must be re-normalized by a flow-dependent time scale. In this section, we re-normalize parameters to show the effect of Str on $C$ in a one-dimensional pure straining and rotating flow.

The Lyapunov exponent $\lambda$ is related to the underlying flow in Eq. (25) through $\rho^Q$. Its dimensional counterpart, $\rho^\eta$ is computed based on $\|S_d\|$ and $\|\Omega_d\|$, where subscript d denotes dimensional variables. Assuming the time scales of $\|S_d\|$ and $\|\Omega_d\|$ to be the same, the Kolmogorov time scale $\tau_\eta = \|S_d\|^{-1}$ is the most natural choice as a flow time scale for normalizing $\rho^\eta$. Employing subscript $\eta$ to distinguish parameters that are normalized based on $\tau_\eta$, we have $\rho^\eta = St\rho^\eta$, $\lambda = St\lambda_\eta$, $C = StC_\eta$, and $\Phi = St^2\Phi_\eta$.

The problem to be studied in this section is similar to that of Section 3, where particles are placed in a one-dimensional flow with an oscillatory gradient according to Eq. (26). However, instead of exploring a 2D parameter space, we focus only on the case of $\omega \ll 1$. This condition corresponds to a flow oscillating at a frequency much lower than the inverse of the Stokes number, i.e., $\omega_\eta \ll 1/St$. Although this simplified flow might appear to be irrelevant and unrepresentative of practical scenarios, we show in Section 4.2 that it is closely connected to the three dimensional turbulence. The simple setting of this one-dimensional problem allows delineating the role of underlying flow from that of the governing equation of particle motion (i.e. Eq. (1)) in particle segregation.

Based on the foregoing setting, we neglect $\omega$ in Eq. (32) and use Eq. (20) with $N_{sd} = 1$ to express $C$ in a re-normalized form as

$$C_\eta = -\frac{1}{2St} + \frac{1}{4St} \sqrt{2 + 2\sqrt{1 + 16St^2\Phi_\eta}}.$$  

Moreover, $\Phi_\eta = \|\Omega_\eta\|^2 - \|S_\eta\|^2$ (see Eq. (26)). Hence, to represent a rotating and straining flow, we take $\Phi_\eta = 1$ and $-1$, respectively. With this choice, two flows are equivalent and $\tau_\eta = 1$.

For $St \in [10^{-2}, 10^2]$, Eq. (37) is evaluated and the results are plotted in Figure 10. To compute the reference results, we follow the procedure described in Section 3.1 using $\omega = 10^{-4}$. Since $\omega \approx 0$, the
Figure 10: The rate of contraction or expansion of clouds $C_\eta$ (left) and particle crossover (right) as a function of $St$ in a low frequency oscillatory one-dimensional flow. RM and SL (dash-dotted) and the present analysis (dashed) are compared against the reference numerical computation (solid) for a straining (black) and rotating (red) flow. Inset: Same plot in the logarithmic scale for $St \leq 1$ with the curves associated with the straining flow inverted. Prediction of SL is identical to RM and is $C_\eta = St\Phi_\eta$. In this case study, note $\lambda_\eta = C_\eta$ since $N_{sd} = 1$. $\text{imag}(C_\eta)$ that represents the rate of particle crossover is also shown in this figure. To better understand the behavior of particles in the straining flow, it is necessary to distinguish between two regimes in which real $(C_\eta)$ decreases and increases with $St$. In the first regime occurring at $St < \sqrt{1/32}$, $\text{imag}(C_\eta) = 0$ and real $(C_\eta)$ decrease almost linearly versus $St$. No particle crossover occurs in this regime and the increase in the particle inertia is met with proportionally stronger slippage, leading to a faster rate at which particles get close to each other (case IV in Figure 1). Further increase in $St$ leads to the second regime, in which particles have enough inertia to cross over each other (cases I and III in Figure 1). The maximum rate of convergence is obtained at the onset of crossovers, before particles begin to overshoot each other. For $St > \sqrt{1/32}$, the relative velocity of particles at the moment of crossover increases with $St$, such that $St \geq 3/4$ their mean distance, rather than decreasing, begins to increase over time. As $St$ is increased beyond $St \approx 3.6$, the rate at which particles diverge decreases, i.e., $d($real$(C_\eta))/dSt < 0$ for $St > 3.6$. Due to the high inertia of particles at this limit, particles hardly respond to the oscillations of the underlying flow and as a result tend to maintain their initial position, lowering their divergence rate. The decrease in $\text{imag}(C_\eta)$ at this limit also supports the above argument by suggesting fewer incidents of crossovers for particles with higher inertia. The asymptotic behavior of real $(C_\eta)$ as $St \to \infty$ is $St^{-1/2}$, which is in agreement with Eq. (37).

The behavior of particles in the rotating flow is less complicated as there is no particle crossover (case V in Figure 1). The distance between particles always increase over time at $O(St)$ rate at $St \ll 1$ and $O(St^{-1/2})$ at $St \gg 1$. The explanation provided for the asymptotic behavior of particles in the straining flow at these two limits also applies to the rotating flow. At small $St$, particles follow fluid tracers and their distance barely changes over time. At high $St$, particles barely respond to the underlying flow oscillations and maintain their initial position. In either of these two limits, there is a minimal change in their distance analogous to the straining flow. Since real $(C_\eta(St))$ is linear at low $St$, the magnitude of real $(C_\eta)$ for straining and rotating flows is equal up to the leading order term. At the nonlinear regime of large $St$, however, the leading order terms have a similar exponent, i.e. real $(C_\eta(St)) \sim St^{-1/2}$, but different magnitude.

In overall, there is a good agreement between the prediction of our analysis and the reference result. The discrepancy occurs primarily at the onset of crossovers in the straining flow, where real $(C_\eta)$ is minimized and $St \approx 0.25$. To further analyze the source of this discrepancy, we reproduced a cut of Figure 6 at $\omega \approx 0$. 

\[ \text{Figure 10: The rate of contraction or expansion of clouds } C_\eta \text{ (left) and particle crossover (right) as a function of } St \text{ in a low frequency oscillatory one-dimensional flow. RM and SL (dash-dotted) and the present analysis (dashed) are compared against the reference numerical computation (solid) for a straining (black) and rotating (red) flow. Inset: Same plot in the logarithmic scale for } St \leq 1 \text{ with the curves associated with the straining flow inverted. Prediction of SL is identical to RM and is } C_\eta = St\Phi_\eta. \text{ In this case study, note } \lambda_\eta = C_\eta \text{ since } N_{sd} = 1. \text{ } \text{imag}(C_\eta) \text{ that represents the rate of particle crossover is also shown in this figure. To better understand the behavior of particles in the straining flow, it is necessary to distinguish between two regimes in which real } (C_\eta) \text{ decreases and increases with } St. \text{ In the first regime occurring at } St < \sqrt{1/32}, \text{imag} (C_\eta) = 0 \text{ and real } (C_\eta) \text{ decrease almost linearly versus } St. \text{ No particle crossover occurs in this regime and the increase in the particle inertia is met with proportionally stronger slippage, leading to a faster rate at which particles get close to each other (case IV in Figure 1). Further increase in } St \text{ leads to the second regime, in which particles have enough inertia to cross over each other (cases I and III in Figure 1). The maximum rate of convergence is obtained at the onset of crossovers, before particles begin to overshoot each other. For } St > \sqrt{1/32}, \text{ the relative velocity of particles at the moment of crossover increases with } St, \text{ such that } St \geq 3/4 \text{ their mean distance, rather than decreasing, begins to increase over time. As } St \text{ is increased beyond } St \approx 3.6, \text{ the rate at which particles diverge decreases, i.e., } d($real$(C_\eta))/dSt < 0 \text{ for } St > 3.6. \text{ Due to the high inertia of particles at this limit, particles hardly respond to the oscillations of the underlying flow and as a result tend to maintain their initial position, lowering their divergence rate. The decrease in } \text{imag}(C_\eta) \text{ at this limit also supports the above argument by suggesting fewer incidents of crossovers for particles with higher inertia. The asymptotic behavior of real } (C_\eta) \text{ as } St \to \infty \text{ is } St^{-1/2}, \text{ which is in agreement with Eq. (37).} \text{ The behavior of particles in the rotating flow is less complicated as there is no particle crossover (case V in Figure 1). The distance between particles always increase over time at } O(St) \text{ rate at } St \ll 1 \text{ and } O(St^{-1/2}) \text{ at } St \gg 1. \text{ The explanation provided for the asymptotic behavior of particles in the straining flow at these two limits also applies to the rotating flow. At small } St, \text{ particles follow fluid tracers and their distance barely changes over time. At high } St, \text{ particles barely respond to the underlying flow oscillations and maintain their initial position. In either of these two limits, there is a minimal change in their distance analogous to the straining flow. Since real } (C_\eta(St)) \text{ is linear at low } St, \text{ the magnitude of real } (C_\eta) \text{ for straining and rotating flows is equal up to the leading order term. At the nonlinear regime of large } St, \text{ however, the leading order terms have a similar exponent, i.e. real } (C_\eta(St)) \sim St^{-1/2}, \text{ but different magnitude.} \text{ In overall, there is a good agreement between the prediction of our analysis and the reference result. The discrepancy occurs primarily at the onset of crossovers in the straining flow, where real } (C_\eta) \text{ is minimized and } St \approx 0.25. \text{ To further analyze the source of this discrepancy, we reproduced a cut of Figure 6 at } \omega \approx 0.\]
in Figure 11. Based on Eq. (37), \(\lambda(\Phi < 0)\) in this figure correspond to \(StC_\eta(-St^2)\) for the straining flow in Figure 11. As discussed earlier and shown in Figure 11, the predicted extremum of real (\(\lambda\)) coincides with that of the reference, viz. \(\Phi = -1/16\) (i.e. \(\Phi = -((\omega^2 + 1)/16\) at \(\omega = 0\)). From the transformation of \(\Phi\) to \(St\), the extremum of real \((C_\eta(St))\) occurs at \(St = \sqrt{-\Phi}\) with \(\Phi\) satisfying real \((\lambda(\Phi)) = 2\Phi[d(\text{real}(\lambda))/d\Phi]\). Therefore, although the extremum of real (\(\lambda\)) is correctly predicted, the extremum of real \((C_\eta)\) is shifted to higher \(St\) (Figure 11). This shift is a result of the error in prediction of real (\(\lambda\)) at the extremum. Underpredicting real \((C_\eta)\) or real (\(\lambda\)) is attributed to the contribution of sub-harmonics in the solution as elaborated in the discussion pertaining to Figure 6.

4. Extension to multimodal excitation

In a physically realistic turbulent flow, excitation is not at a single frequency but involves a continuous range of frequencies. To analyze a multimodal excitation, all modes that appear in Eq. (25) must be retained. With the transformation introduced in Eq. (29), Eq. (25) can be expressed as

\[
\frac{N_{sd}}{4} (\gamma^2 - 1) - \int_{-\infty}^{\infty} \frac{\hat{\rho}^Q(\omega; St)}{\gamma^2 + \omega^2} d\omega = 0,
\]

which provides a generic relationship for \(\gamma\) in the form of an eigenvalue problem. An iterative approach can be adopted for computing \(\gamma\) from this equation, as the evaluation of the integral requires a prior knowledge of \(\gamma\). Our goal in this section is to find an alternative form of Eq. (38) that allows explicit evaluation of \(\gamma\).

The analytical form of \(\hat{\rho}^Q(\omega; St)\) is not known in general. It is attainable numerically through the Lagrangian sampling of fluid velocity gradient along the trajectory of a particle. If the analytical form of \(\hat{\rho}^Q(\omega)\) were to be available and \(\hat{\rho}^Q \to 0\) as \(|\omega| \to \infty\), Cauchy’s integral formula could be employed to express

\[
\int_{-\infty}^{\infty} \frac{\hat{\rho}^Q(\omega; St)}{\gamma^2 + \omega^2} d\omega = \frac{\pi}{\gamma} \hat{\rho}^Q(\hat{\gamma}; St).
\]

Depending on the form of \(\hat{\rho}^Q(\hat{\gamma}; St)\), an explicit relationship for \(\gamma\) can be obtained from this expression.

Another possible scenario that may arise is a design problem formulated as finding a specific \(\hat{\rho}^Q(\omega; St)\) when a desirable \(\gamma(St)\) is given. Designing a hydrodynamic particle separator by enhancing segregation
of a particular class of particles in a polydisperse distribution is an instance of such scenario. The present formulation can be instrumental in solving this inverse problem by expressing Eq. (38) as a Fredholm integral equation of the first kind with a Kernel function \((\gamma^2 + \omega^2)^{-1}\).

An explicit relationship for \(\gamma\) is needed for cases that do not classify as the two previous scenarios. To find this explicit relationship, we express the integral in Eq. (38) in a form that is independent of \(\gamma\) by transforming it to a polynomial, using the Taylor series expansion. Dividing the integration interval to \(|\omega| > |\gamma|\) and \(|\omega| < |\gamma|\) and expanding the denominator lead to

\[
\int_{|\omega|<|\gamma|} \frac{\hat{\rho}^Q}{\gamma^2 + \omega^2} d\omega = \gamma^{-2}\pi_0 - \gamma^{-4}\pi_2 + \gamma^{-6}\pi_4 - \cdots, \tag{40}
\]

and

\[
\int_{|\omega|>|\gamma|} \frac{\hat{\rho}^Q}{\gamma^2 + \omega^2} d\omega = \pi_2^+ - \gamma^2\pi_4^- + \gamma^4\pi_6^- - \cdots, \tag{41}
\]

in which

\[
\pi_i^+(\text{St}) := \int_{|\omega|>|\gamma|} \omega^i \hat{\rho}^Q(\omega; \text{St}) d\omega,
\]

are the moments of the spectrum of the Q-criterion. Note \(\pi^+\) and \(\pi^-\) are computed by integrating over \(|\omega| > |\gamma|\) and \(|\omega| < |\gamma|\) intervals, respectively. Substituting Eqs. (40) and (41) into Eq. (38) produces

\[
\frac{N_{\text{sd}}}{4} (\gamma^2 - 1) + \sum_{i=-\infty}^{-1} (-1)^i \gamma^{2i} \pi_{2i-2}^- - \sum_{i=0}^{\infty} (-1)^i \gamma^{2i} \pi_{2i-2}^+ = 0. \tag{43}
\]

To obtain a manageable expression, the summations are truncated by neglecting the higher order terms. Only keeping the terms with \(-1 \leq i \leq 1\) in Eq. (43), it can be shown

\[
\gamma^2 \approx \frac{1}{2} \left( N_{\text{sd}} + 4\pi_2^- \right)^{-1} \left( N_{\text{sd}} + 4\pi_2^+ + \sqrt{(N_{\text{sd}} + 4\pi_2^+)^2 + 16\pi_0^- (N_{\text{sd}} + 4\pi_2^-)} \right). \tag{44}
\]

Using this equation, \(\lambda\) can be calculated as \(\lambda = (\sqrt{2} - 1)/2\) if the moments defined in Eq. (42) are available.

Evaluating Eq. (44) still requires a prior knowledge of \(\gamma\) as the integrals in Eq. (42) depend on \(\gamma\). We present two approaches to obtain a fully explicit relationship for \(\lambda\). In the first approach, we factor out \(\gamma^2\) from the denominator of the integral in Eq. (38). That leaves \(1 + (\omega/\gamma)^2\) in the denominator. Assuming

\[
1 + (\omega/\gamma)^2 \approx 1 + \omega^2, \tag{45}
\]

Eq. (38) simplifies to

\[
\frac{N_{\text{sd}}}{4} \gamma^2 (\gamma^2 - 1) - \int_{-\infty}^{\infty} \frac{\hat{\rho}^Q(\omega; \text{St})}{1 + \omega^2} d\omega = 0. \tag{46}
\]

Following steps that were taken to derive Eq. (42), it can be shown

\[
\lambda(\text{St}) \approx -\frac{1}{2} + \frac{\sqrt{2}}{4} \sqrt{1 + \sqrt{1 + 16N_{\text{sd}}^{-1}}} , \tag{47}
\]

with

\[
\pi_{\omega}(\text{St}) := \int_{-\infty}^{\infty} \frac{\hat{\rho}^Q(\omega; \text{St})}{1 + \omega^2} d\omega. \tag{48}
\]

The second approach is based on the assumption \(|\hat{\rho}^Q(\omega) \geq |\gamma|\| \ll |\hat{\rho}^Q(\omega) \leq |\gamma|\|\), which corresponds to assuming \(|\hat{\rho}^Q|\) decays rapidly at higher frequencies. Similar to the assumption associated with the first approach, this assumption is valid at regimes of St with \(\lambda \neq -1/2\) or \(|\gamma| \approx 1\). Taking only the first term \(i = -1\) in the Taylor series expansion

\[
\frac{N_{\text{sd}}}{4} (\gamma^2 - 1) - \gamma^{-2} \int_{-\infty}^{\infty} \hat{\rho}^Q(\omega) d\omega = 0. \tag{49}
\]

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Similar to the derivation of Eq. (410), Eq. (411) can be rearranged to obtain

$$\lambda(St) \approx -\frac{1}{2} + \frac{\sqrt{2}}{4} \sqrt{1 + \sqrt{1 + 16N_{sd}^{-1}\bar{I}_0}},$$

(50)

where

$$\bar{I}_0(St) := \int_{-\infty}^{\infty} \rho^Q(\omega; St) d\omega = \rho^Q(0; St) = \text{tr} \left( -\nabla u(t) \nabla u(t) \right).$$

(51)

The above two expressions for $\lambda$ are very similar with the distinction that Eq. (47) filters higher frequency oscillations. While Eq. (50) can be computed from a single time snapshot, Eq. (47) requires a series of snapshots in time as it is computed from $\rho^Q(t)$ rather than $\rho^Q(0)$. Similar to the case of unimodal excitation, these relationships are a more general form of RM and SL expressions. One can verify that Eqs. (2) and (3) are the simplified form of Eqs. (50) and (47) when $|\lambda| \ll 1$, respectively. In fact, the estimate of RM and SL for $C$ is equal to $\bar{I}_0$ and $\bar{I}_\omega$ from Eqs. (51) and (45), respectively. Therefore, the outcome of present analysis can be considered as a correction to RM and SL analyses according to

$$C(St) = N_{sd} \left[ -\frac{1}{2} + \frac{\sqrt{2}}{4} \sqrt{1 + \sqrt{1 + 16N_{sd}^{-1}\hat{\lambda}(St)}} \right],$$

(52)

in which $\hat{\lambda}$ is the estimate obtained from RM or SL. To prevent any potential confusion in the interpretation of the results presented in the following sections, we only report $C$ computed based on Eq. (47) and compare it against RM and SL. In other words, reported $C$ are obtained from Eq. (2) for RM, Eq. (3) for SL, and Eq. (47) with $C = N_{sd}\lambda$ for the present analysis (or Eq. (52) with $\hat{\lambda}$ from SL). The prediction of Eq. (50), although not discussed, is very similar to Eq. (47) with an slight underprediction at $St \approx 1$ and overprediction at $St \gg 1$.

Equation (47) predicts an approximate rate of expansion or contraction of the cloud of particles (i.e., Lyapunov exponent of inertial particles) when they are subjected to a continuous spectrum of oscillatory fluid motions. This is in contrast to Eq. (52) that was derived for a unimodal excitation. Although it relies on additional assumptions, Eq. (47) prediction of $\lambda$ behavior remains similar to that of Eq. (52). The rate at which $|\lambda|$ increases as $|\bar{I}_0| \to \infty$ is identical in both cases. The lower bound on real $\lambda$ is also identical between the two. Because of its underlying assumption, however, Eq. (47) is not expected to produce accurate predictions when real $\lambda \approx -1/2$ that is the regime of strong particle segregation.

4.1. Two dimensional random mixing flow

To examine the validity of Eq. (47) and its underlying assumptions, we use two test cases: 1) a two-dimensional synthetic flow and 2) a forced isotropic turbulence. To allow for a step-by-step validation, we first consider the simpler case of a two-dimensional synthetic flow through a corner or near a stagnation point. In this flow, the velocity gradient tensor $\nabla u$ is diagonal and the two Lyapunov exponents are statistically identical. As a result, this case is compatible with the assumption of identical $\lambda$ in Eq. (19). In contrast to the previous section, particles are exposed to a continuous range of frequencies. Hence, this example provides a suitable substrate for evaluating the error associated with the assumption in Eq. (19).

The full description of the flow under consideration is provided in reference [30]. Here, we only provide details that are necessary for reproducing the results. The flow in the entire two-dimensional plane is constructed by repeating the flow defined in a $[-\pi/2, \pi/2] \times [-\pi/2, \pi/2]$ square box (highlighted in Figure 12). At a given point $\hat{x}$ in the box, the fluid velocity is $u_{x1} = \hat{x} S_\eta$ and $u_{x2} = -\hat{x} S_\eta$ where $S_\eta$ is the flow strain-rate. Note that subscript $\eta$ is adopted to distinguish $u_\eta$, which is normalized based on the characteristic time scale of the flow, from $u$ defined in the previous section, which is normalized based on the particle relaxation time $\tau$.

To ensure homogeneity in space, the point of origin is shifted and the strain-rate is adjusted randomly throughout the simulation. The random shift $\Delta$ is sampled from $U(-\pi, \pi) \times U(-\pi, \pi)$, where $U(a, b)$ denotes a uniform distribution with support $[a, b]$. The strain-rate $S_\eta$ is also sampled from $U(0, S_0)$. $S_0 = 6/\pi$ is computed such that the time-averaged of the flow kinetic energy is unity [31]. Once a flow is generated, it
remains unchanged for a large eddy turnover time $T_\eta$. At time $t_\eta = T_\eta$, a new flow is generated by computing new $\Delta$, $S_\eta$, and $T_\eta$. At each interval, $T_\eta$ is sampled from an exponential distribution under the assumption of unit decay time, i.e., $T_\eta \sim \log(U(0, 1))$. Note, $\tau_\eta$ is one in this example.

To summarize, the fluid velocity $u_\eta$ at the location of particle $x$ is
\begin{equation}
\begin{aligned}
u_{\eta 1} &= (-1)^{\xi_x} \xi_y (\hat{x}_1 - \pi \xi_x) S_\eta, \\
\nu_{\eta 2} &= (-1)^{\xi_x + 1} (\hat{x}_2 - \pi \xi_y) S_\eta,
\end{aligned}
\end{equation}
where
\begin{equation}
\begin{aligned}
\xi_x &:= \frac{\hat{x}_1}{|\hat{x}_1|} \left( \frac{|\hat{x}_1|}{\pi} + \frac{1}{2} \right), \\
\xi_y &:= \frac{\hat{x}_2}{|\hat{x}_2|} \left( \frac{|\hat{x}_2|}{\pi} + \frac{1}{2} \right),
\end{aligned}
\end{equation}
$\hat{x} := x - \Delta$, and $\lfloor \cdot \rfloor$ is the floor function. Having $u_\eta$, the location of particle is computed by integrating $\ddot{x}_\eta = (\nu_{\eta} - \dot{x}_\eta)/S_{\eta}$ in time.

In our computation, we consider 101 classes of particle with $\text{St} = \tau/\tau_\eta$ ranging from 0.01 to 100 with one particle in each class. Particles are traced for $t_\eta = 1000$ and statistics are collected between $800 < t_\eta < 1000$.

The second-order Crank Nickelson time integration scheme is adopted with a time step size of 0.02. Randomly sampled $T_\eta$ is rounded to the nearest time step to obtain statistics at uniform intervals. To ensure statistical convergence, this process is repeated for 100,000 flow realizations. Reducing the time step size and increasing the number of flow realizations had a negligible effect on the reported results.

From the time history of the velocity gradients along the trajectory of the particle, $\rho Q(t_\eta)$ and $\tilde{\rho} Q(\omega_\eta)$ are computed. The results are shown in Figure 13 for $\text{St} \in [0.01, 100]$. The minimal spread of curves in Figure 13 indicates: 1) $\tau_\eta$ is a better choice for normalization than $\tau$ as plotting $\tilde{\rho} Q = \tilde{\rho} Q_{\text{St}}$ versus $\omega = \omega_{\text{St}}$ results in a much larger spread and 2) the influence of $\text{St}$ on the particles trajectory is minimal. In this flow, particles with higher and lower $\text{St}$ experience higher levels of low- and high-frequency oscillations, respectively.

Several behaviors of the Q-criterion can be inferred from the construction procedure of this synthetic flow. $\tilde{\rho} Q_{\eta}$ is strictly negative regardless of $\omega_\eta$ and $\text{St}$ (Figure 13). Since the velocity gradient tensor is diagonal, $\text{tr}(\nabla u_\eta \nabla u_\eta) > 0$ and from Eq. (4), $\int \rho Q_{\eta} d\omega < 0$. This condition will not hold in a turbulent flow because particles may travel through elliptic regions where $\|\Omega_\eta\|^2 > \|S_\eta\|^2$.

While constructing the flow under consideration, $S_0$ was imposed to be constant and independent of $\text{St}$. Hence, all particle experience a random strain-rate $S_\eta \in [0, S_0]$ over time regardless of their $\text{St}$. From this independence, we can analytically compute $\langle \rho Q(t_\eta = 0) \rangle = -2\langle S_\eta \rangle = -2S_0^2/3 = -24/\pi^2$. This analytical estimate is compatible with Figure 13 that shows $-\pi^2/24\langle \rho Q_{\eta} \rangle$ has an intercept of 1, regardless.
of $\text{St}$. Moreover, the decay rate of $\langle \rho Q \rangle$ versus $t_\eta$ can be estimated from the distribution of $T_\eta$. Since $T_\eta$ is sampled from an exponential distribution with a decay rate of unity, the likelihood of $S(t'_{\eta})$ being identical to $S(t_{\eta} + t_{\eta})$ is $\exp(-t_{\eta})$ for a fixed point in space. Hence, for a fixed point in space or a particle that has a trajectory uncorrelated with the flow $\langle S(t'_{\eta}) S(t'_{\eta} + t_{\eta}) \rangle = \langle S^2_{\eta} \rangle \exp(-t_{\eta})$, which translates to $\langle \rho Q(t_{\eta}) \rangle = \langle \rho Q(0) \rangle \exp(-t_{\eta})$. Provided that the trajectory of particles becomes uncorrelated with the underlying flow as $\text{St} \to \infty$, this exponential decay is best observed at the largest simulated $\text{St}$ (Figure 13). From the analytical estimate of $\langle \rho Q \rangle = -24/\pi^3 \exp(-t_{\eta})$, one can obtain $\langle \tilde{\rho} Q \rangle = -24/\pi^3 (1 + \omega^2_{\eta})$. Similar to $\rho Q$, this analytical estimate is most relevant at $\text{St} \gg 1$ (Figure 13). At smaller $\text{St}$, particles move quickly between adjacent repeating-patterns, experiencing a strain-rate with an opposite sign. As a result, the autocorrelation function deviates from the analytically predicted exponential decay and becomes negative.

From $\tilde{\rho} Q$ (Figure 13), $C_\eta$ is computed using RM, SL, and the present analysis (Figure 14). The exact value of $C_\eta$ is also computed numerically as a reference for comparison by integrating Eq. (11) for $F$ in two dimensions. Since $\nabla u$ is diagonal, integration is performed in each dimension separately, using the scheme that was described in Section 3.1. The result of these direct computations is shown as the reference in Figure 14. A more detailed discussion on the physical interpretation of the reference results and RM are provided in reference [30]. Here, we emphasis on statistics relevant to the comparison and verification of our analysis.

The prediction of RM, SL, and the present analysis are based on $\tilde{\rho} Q$, which is computed numerically at different $\text{St}$ (Figure 13). An alternative is to use the analytical estimate

$$\langle \rho Q \rangle = -\frac{24}{\pi^3 (1 + \omega^2_{\eta})},$$

(55)

to obtain a prediction that is independent of the numerical results. Based on Eq. (55), from RM (Eq. (3))

$$\langle C_\eta \rangle = -\frac{24 \text{St}}{\pi^2},$$

(56)

from SL (Eq. (3))

$$\langle C_\eta \rangle = -\frac{24 \text{St}}{\pi^2 (1 + \text{St})},$$

(57)
The rate of contraction or expansion of particle clouds \( \text{real}(\mathbf{C}_\eta) \) (left) and crossovers (right) as a function of \( \text{St} \) for the random straining flow (Figure 12). The prediction of RM (dotted green), SL (dashed-dotted blue), and the present analysis (dashed red), obtained based on the numerically computed \( \tilde{\rho}Q_\eta \), are compared against the reference results (solid black). Using the analytical estimate of \( \tilde{\rho}Q_\eta \) (Eq. 55), the prediction of RM (green pluses) that is Eq. (56), SL (blue crosses) that is Eq. (57), and the present analysis (red dots) that is Eq. (58) are also shown for comparison. Inset is the same plot for \( \text{St} < 1 \) in the logarithmic scale.

The accuracy of these three equations relies on the accuracy of the underlying models as well as the analytical estimate in Eq. (55). As discussed above, the estimate is accurate for \( \text{St} \gg 1 \). Its integral is also exact. Hence, the estimation provided in Eq. (56) is identical to RM prediction. The estimation of Eqs. (57) and (58), however, agrees with SL and the present analysis, respectively, at very large \( \text{St} \) (Figure 14). They also agree at \( \text{St} \ll 1 \), since \( \mathbf{C}_\eta \) depends primarily on the integral of \( \tilde{\rho}Q_\eta \) at \( \text{St} \ll 1 \). Better estimates can be obtained if the deviation of \( \tilde{\rho}Q_\eta \) from the exponential decay is considered for \( \text{St} \approx 1 \) (Figure 13). We observed that replacing \( (1 + \text{St}) \) by an empirical estimate \( (1 + 2\text{St}) \) in the denominator of Eqs. (57) and (58) accounts for this deviation and produces much better estimates at \( \text{St} \approx 1 \) (not shown in Figure 14).

Comparing the results of various analyses with the reference results shows that while RM and SL are valid only at small \( \text{St} \), the present analysis is valid at small and large \( \text{St} \). The validity of RM at small \( \text{St} \) has been established in the literature [33, 34, 28]. It was also observed in Section 5.4 where all models collapsed with the reference results at \( \text{St} \ll 1 \). As discussed earlier, SL and the present analysis are more general formulation and reduce to RM at low \( \text{St} \). Hence, finding the prediction of all analyses to agree with the reference at low \( \text{St} \) was expected from their asymptotic behavior. Note that \( \mathbf{C}_\eta \propto \text{St} \) at \( \text{St} \ll 1 \) is a direct consequence of the independence of \( \tilde{\rho}Q_\eta(0) \) from \( \text{St} \).

The agreement between the present analysis and the reference results at large \( \text{St} \) is remarkable (dashed red and black curves in Figure 14) considering the numerous underlying assumptions. The agreement at large \( \text{St} \) is a result of \( \lambda_\eta > 0 \), which is compatible with the assumption of \( \gamma \approx 1 \) in Eq. (45), and \( \nabla \mathbf{u} \) being diagonal, which is compatible with the assumption of single \( \lambda \) in Eq. (19). The prediction of our analysis is not accurate at \( \text{St} \approx 1 \), where \( \mathbf{C}_\eta \) is minimized. This disagreement is partly a consequence of \( 2\lambda + 1 = \gamma < 1 \), which undermines the assumption \( 1 + (\omega/\gamma)^2 \approx 1 + \omega^2 \). The error associated with this assumption is particularly significant because \( \tilde{\rho}Q_\eta \) is relatively large at intermediate \( \omega_\eta \) at \( \text{St} \approx 1 \) and...
source of disagreement is the presence of sub-harmonics in the solution that are not included in our analysis. The discrepancy produced by these sub-harmonics is already discussed for the one-dimensional straining flow with unimodal excitation (Section 3.4).

Particles segregation is quantitatively analogous in this two-dimensional synthetic flow and the one-dimensional straining flow from Section 3.4. Considering $C_\eta (St)$ in Figures 10 and 14, the onset of particle crossovers and maximum contraction rate occurs at $St \approx 0.2$ and the maximum rate of crossover occurs at $St \approx 0.5$ in both flows. The maximum rate of contraction is different by a factor of two ($\approx 0.55$ versus $\approx 0.3$), indicating the extremum of real ($\lambda_\eta$) is almost the same in both flows. For $St \ll 1$, real ($C_\eta$) $\propto St$ in both flows as well. This similarity is observed despite the fact that the underlying flow is significantly different between the two cases. There is unimodal oscillation at $\omega \approx 0$ in one flow, whereas the excitation spectrum is continuous in the other flow. The excitation amplitude is not a function of $St$ in one flow, whereas it varies in the other flow. This quantitative similarity shows that the segregation phenomenon is the general response of Eq. (1) to an oscillatory motion and is relatively independent of the characteristics of the oscillations as long as $||S|| > ||\Omega||$. The three-dimensional turbulence results in the next section also support this statement.

To obtain a measure of intermittency, the normalized second moment of $C_\eta$ is computed. Without normalization, the standard deviation of $C_\eta$, denoted by $(C_\eta')'$, will vary depending on the integration period $t_\eta$. Contrary to $C_\eta'$, $F_\eta$ is a stationary random variable with a variance independent of $t_\eta$. From this independence and basic properties of variance, one can show that the variance of $C_\eta'$, which is the time-averaged of $|F_\eta|$ over period $t_\eta$, scales as $t_\eta^{-1/2}$ for $t_\eta$ longer than the integral time scale of $C_\eta$. Thus, $\sqrt{t_\eta (C_\eta')'}$ is computed as a quantity that is independent of $t_\eta$ and plotted in Figure 15.

Similar to the first moment, the present analysis provides the best estimate for the second moment of $C_\eta$. All models provide accurate approximation for $St \ll 1$. For $St \gg 1$, while both RM and SL predictions are irrelevant, our analysis is relatively accurate with a slight difference in the asymptotic variation. Variation of $\sqrt{t_\eta (C_\eta')'}$ as $St \to \infty$ is proportional to $St^{-1/3}$ from the reference, whereas it is proportional to $St^{-1/4}$ from our analysis, leading to overestimation of standard deviation (Figure 15). This overestimation, which also occurs at $St \approx 1$, is primarily due to the wider spread of predicted $C_\eta$ that is manifested in Figure 10.

A fundamental difference between the synthetic flow considered here and a physically realistic flow is the existence of discontinuities in the velocity field. Note that $\partial u_i/\partial x_j$ for $i \neq j$ is not defined on the patterns boundaries. As a particle crosses over these boundaries, the tangential component of velocity switches sign,
producing a singular value at the off-diagonal entries of $\nabla u$ and a spike in the rotation rate tensor. In our computations, we remove these singular points by assuming the off-diagonal entries of $\nabla u$ to be always zero. The role of these discontinuities in particle segregation, which may have little physical relevance, is yet to be determined. In the next example, we will consider an isotropic turbulent flow which is continuous in space but presents the challenge of three-dimensionality and intermittency.

### 4.2. Three dimensional isotropic turbulence

As the last case study, we consider a triply periodic homogeneous isotropic turbulence as the background flow. A stationary turbulence is maintained by adding a forcing term to the momentum equation that is proportional to the velocity $\frac{32}{4}$. The forcing term is dynamically computed at each time step to prevent variation of $\tau_\eta$ and thus $St \[29, 36\]$. The maximum deviation of $\tau_\eta$ from the target Kolmogorov time scale is 0.3%. The Reynolds number based on the Taylor microscale is $Re_\lambda = 100$. A second order spatial discretization on a $256^3$ grid and 4th order Runge-Kutta time integration scheme are employed. An in-house solver with a specialized linear solver is used in these computations $[37]$. Special care has been taken in interpolating quantities at the location of particles from the Eulerian grid. In particular, the interpolation scheme is designed to correctly translate the incompressibility condition to the Lagrangian velocity gradient tensor. Additionally, the Lagrangian gradients are kept $C^0$ continuous by interpolating from a pre-constructed continuous Eulerian field $[29]$. The particles trajectory is computed using Eq. (1). 86 classes of particles are considered with $St = 2^{p/4}$, $p \in \{-32, \ldots, 53\}$. At each Stokes number, $10^4$ randomly seeded particles were simulated for several large eddy turnover time to allow development of clusters. Starting with this time-evolved distribution, we record the velocity gradient tensor at the position of each particle for $800\tau_\eta$ with $\tau_\eta$ intervals. Based on $\nabla u_\eta(t)$, $\hat{\rho}^{S\eta}$ is computed using Eq. (4) at each $St$. The number of particles and integration period are verified to be sufficient for achieving statistical convergence. The results of these calculations at few Stokes numbers are shown in Fig. 16.

In a turbulent flow, in contrast to the two cases studied so far, $\nabla u_\eta$ has a full rank and $\|S_\eta\|$ and $\|\Omega_\eta\|$ are nonzero simultaneously. These parameters, computed along the trajectory of particles, exhibit complex behaviors due to their dependence on $St$ and preferential concentration of particles. These dependencies are briefly mentioned here for the sake of completeness and discussed in more details in reference $[29]$. In the homogeneous turbulence under consideration, $\rho^{S\eta}(t; St)$ and $\rho^{\Omega\eta}(t; St)$ (the autocovariance functions of the norm of strain and rotation tensors, respective) are both strictly positive functions. $\rho^{S\eta}$ is relatively
independent of St and exponentially decays with time, which is analogous to the random straining flow (Figure 13). \( \rho^{\Omega_1} \), on the other hand, varies significantly versus St. As a result, \( \rho^{\Omega_1} = \rho^{\Omega_1} - \rho^{S_1} \) strongly depends on St.

For \( St \lesssim 1 \), \( \rho^{\Omega_1} \) undergoes an increasing-decreasing trend in time. \( \rho^{\Omega_1} \) being negative at \( t_\eta \ll 1 \) and \( St \ll 1 \) is due to the smaller value of \( \rho^{\Omega_1} \). Hence, particles with small St tend to centrifuge out of rotational regions with the short time constant and follow slow vortical features since \( \rho^{\Omega_1} > 0 \) at \( \omega_\eta \ll 1 \) and \( St < 1 \) (Figure 10).

For \( St \gg 1 \), particles are not responsive to the velocity fluctuations and follow a trajectory that is uncorrelated with the flow. As a result, particles distribute uniformly in space and the Lagrangian and Eulerian statistics become almost identical. Additionally, one can show that the Eulerian strain-rate and rotation-rate autocovariance functions are equal in a periodic domain. Therefore, \( \rho^{S_1} \) and \( \rho^{\Omega_1} \) converge to the same value, leading to \( \rho^{\Omega_1} \rightarrow 0 \) and \( \rho^{S_1} \rightarrow 0 \) as \( St \rightarrow \infty \).

In Section 3.1, we demonstrated that the particle clouds only contract in a straining regime as a rotating regime only leads to cloud expansion. Thus, in a three-dimensional turbulence, where regions of higher rotation-rate and strain-rate coexist in space, particles tend to accumulate in regions of higher strain-rate. The preferentially concentrate of particles in the straining regions, which occurs at all St, is supported by the numerical results showing the dominance of \( \rho^{S_1}(t = 0) \) over \( \rho^{\Omega_1}(t = 0) \). This dominance, which measures the preferential concentration of particles in the straining regions, is most noticeable at \( St \approx 1 \) (where \( \rho^{\Omega_1}(0) \) has an extremum) and asymptotes to zero at large and small Stokes numbers.

From \( \rho^{\Omega_1} \), \( C_1^i \) is computed for RM, SL, and the present analysis. The method described in Section 3.1 is employed to compute the reference quantities. Treating each of the individual \( 10^4 \) particles as an independent sample, we construct the PDF of \( C_1^i \) (Figure 17). In terms of accuracy, the overall trend is similar to the previous case studies with the present analysis being the closest to the reference followed by SL and RM. The only exception is \( St = 1 \) where the PDF from the present analysis is extremely skewed and shows a second unphysical peak at \( C_1^i \approx -0.5 \).

The ensemble-averaged of \( C_1^i \) is computed from the PDFs and shown in Figure 18. Since RM and SL are a linear function of \( \langle \rho^{\Omega_1} \rangle \), these ensemble-averaged quantities can be computed directly from the results shown in Figure 16. For the present analysis, however, \( \langle C_1^i(\rho^{\Omega_1}) \rangle \) is slightly different from \( C_1^i(\rho^{\Omega_1}(\rho^{\Omega_1})) \). Their difference depends on the integration period \( t_\eta \) and asymptotes to zero as \( t_\eta \rightarrow \infty \).

The behavior of all three models remains similar to the two previous cases. All models collapse with the reference for \( St \ll 1 \), whereas for \( St \gg 1 \), their prediction widely varies. Among the three models, only the present analysis captures the expansion of clouds at \( St \gg 1 \). (real (\( C_1^i \)) > 0 is predicted at \( St \gg 1 \) despite the fact that \( \rho^{\Omega_1} < 0 \) at all frequencies (Figure 16). Prediction of expansion in a straining regime stems from the nonlinear behavior observed in Figure 3 showing real(\( \lambda \)) > 0 for \( \Phi < -\omega_4 + 10\omega_2^2 + 0)/16. \)

Despite capturing the overall trend, the present analysis is not in full quantitative agreement with the reference at high St. The disagreement can be attributed to the assumption of: 1) single \( \lambda \) in Eq. (19) and replacing a full rank matrix with a diagonal matrix, 2) \( \gamma \approx 1 \) in Eq. (15), and 3) excluding sub-harmonics from our analysis. We expect the predictions to be more accurate at St beyond the range considered in this study. The prediction of Eq. (50), computed from a single snapshot of the flow, is relatively similar to Eq. (17) (i.e. the result associated with the present analysis) in Figure 18. The slight difference between their prediction is at \( St \approx 1 \) and \( St \gg 1 \) where Eq. (50) under- and over-predicts Eq. (17), respectively.

As mentioned earlier, the accumulation of particles in the straining region of a turbulent flow leads to \( \| \mathbf{S}_\eta \| > \| \Omega_0 \| \). As a result, the behavior of \( C_1(St) \) in the isotropic turbulence (Figure 18) is similar to the straining flows (Figures 10 and 11) rather than the rotating flow (Figure 11). In these straining flows, \( C(St) \) follows the same trend. Increasing St from zero, real (\( C_1 \)) decreases till the onset of crossover. The trend is reversed once \( \text{imag}(C_1) \neq 0 \) up to a Stokes number at which \( C_1 > 0 \). For larger St, real (\( C_1 \)) changes non-monotonically with \( C_1 \rightarrow 0 \) as \( St \rightarrow \infty \). Provided this similarity, the physical interpretations that were provided in the previous sections also applies to the isotropic turbulent flow. Additionally, this similarity confirms our earlier hypothesis that the behavior of \( C_1(St) \) is primarily determined based on the governing equations of particle motion rather than the underlying flow as long as \( \| \mathbf{S}_\eta \| > \| \Omega_0 \| \).

Despite the close comparison, there are also differences between these cases. In the turbulent flow, maximum contraction rate occurs at a higher St and is less significant (Figures 18 and 14). The maximum
Figure 17: The PDF of finite-time contraction-rate $\mathcal{C}_t^{\eta}$ at different Stokes numbers obtained based on RM (dotted green), SL (dashed-dotted blue), the present analysis (dashed red), and the reference numerical results (solid black) from the three-dimensional isotropic turbulence at $Re_\lambda = 100$. While all models provide good approximation at low St, only the present analysis agrees with the reference results at high St.
rate of crossover is also lower in the turbulent flow. The presence of rotating regions in the turbulent flow (which were absent in the other cases) reduces the rate of segregation and crossovers by diminishing the effect of straining regions, thereby producing these quantitative differences.

Another important difference between these cases is the asymptotic behavior of \( \text{real} (\langle C_\eta \rangle) \) at \( St \ll 1 \). \( \text{real} (\langle C_\eta \rangle) \) is linear for one- and two-dimensional straining flows (Figures 10 and 14), whereas it is superlinear for the turbulent flow. From RM, \( \text{real} (\langle C_\eta \rangle) \) can be approximated as \( St \rho Q^\eta(t = 0) \) at small \( St \). Therefore, the rate of growth of \( \text{real} (\langle C_\eta \rangle) \) simply depends on the rate of growth of \( \rho Q^\eta(t = 0) \) versus \( St \). \( \rho Q^\eta(t = 0) \) was independent of \( St \) in the previous two cases, hence the linear rate, whereas it is proportional to \( St \) in the present case, hence the superlinear rate. The rate at which \( \rho Q^\eta(t = 0) \) grows versus \( St \) in an isotropic turbulent flow can be an artifact of the periodic boundary condition imposed for computational consideration (see our earlier discussion on \( \rho Q^\eta(t = 0) = 0 \) for uniformly distributed particles on a periodic domain). This potential artifact caused by the space confinement must be removed in the future for a more realistic assessment of the asymptotic behavior of \( \text{real} (\langle C_\eta \rangle) \) at small \( St \) in a turbulent flow.

To show the effect of turbulence intermittency on the particle segregation, we computed the second moment PDFs of \( C_\eta^t \) (Figure 19). As discussed earlier, the plotted moments are normalized by \( \sqrt{t_\eta} \) to ensure their independence from sampling period \( t \). Among the available models, the present analysis provides the best estimation for \( (C_\eta^t)' \). All models collapse with the direct computations at the limit of small \( St \) and deviate from it as \( St \) increases. For \( St > 1 \), RM and SL predictions linearly increase with \( St \) while the present analysis remains bounded. Similar to the two-dimensional case, a slope of \(-1/3\) is observed in this regime of Stokes number.

4.3. On the moments of the finite-time Lyapunov exponent

In this section, we verify the prediction of Eq. (16) using the three cases considered in Sections 3.4, 4.1, and 1.2. The standard deviation of the finite-time Lyapunov exponent \( q_\eta \) is computed numerically for the above three cases. As discussed earlier in Section 2, the standard deviations of \( q_\eta \) remains finite despite the fact \( \text{real} (q_\eta) \to \infty \) at crossovers, which was explained by \( \text{imag} (q_\eta) \to \infty \). The finite second moment of \( q_\eta \) allows achieving statistical convergence in these direct computations. To obtain an estimate of \( q_\eta' \) from Eq. (16), the Lyapunov exponent \( \lambda_\eta \) is extracted from \( C_\eta \) (Figures 10, 14, and 18). This process is repeated.

Figure 18: The rate of expansion or contraction of particle clouds (left) and their crossover (right) as a function of Stokes number based on RM (dotted green), SL (dashed-dotted blue), the present analysis (dashed red), and the reference numerical simulations (solid black). The underlying flow is a three-dimensional stationary isotropic turbulence at \( Re_\lambda = 100 \). Inset is the same plot for \( St \leq 1 \) in a logarithmic scale. Lines with a slope of 1 and 2 are shown for reference.
Figure 19: The normalized standard deviation of the rate of contraction as a function of Stokes number, obtained from RM (dotted green), SL (dashed-dotted blue), the present analysis (dashed red), and the reference numerical simulations (solid black). Results corresponds to the three-dimensional isotropic turbulence at $Re_\lambda = 100$.

twice by extracting $\lambda_\eta$ from the reference results and the prediction of the present analysis. The results of these computations are shown in Figure 20.

Contrary to the fluctuation of $C_\eta$ that tends to zero for small St (Figures 15 and 19), $q_\eta'$ asymptotes to a nonzero constant as $St \to 0$. $q_\eta'$ remains finite in this regime of St because of the finite instantaneous fluctuations, which in the case of $C_\eta$ are either canceled by the opposite fluctuation of $q_\eta'$ in multiple directions or the vanishing fluctuation of $\overline{q_\eta}$. Although at a different rate, the fluctuation of both $C_\eta$ and $q_\eta$ tend to zero at large St. As noted earlier, due to the fundamental difference between these two parameters, $q_\eta'$ should not be used to study intermittency in a particle-laden turbulent flow.

The excellent agreement between direct computations of $q_\eta'$ and its estimation using the reference $\lambda_\eta$ validate Eq. (16). Employing the prediction of present analysis instead of the reference results produces accurate estimation for the one-dimensional case. For two- and three-dimensional cases, however, the agreement vanishes, particularly for $St \gtrsim 1$. An accurate prediction of $q_\eta'(St)$ from $\lambda_\eta(St)$ relies on having a model that captures not only the first order term correctly but also the higher order terms. For instance, adopting RM or SL for the one-dimensional case, in which $\lambda_\eta$ is predicted to be $-St$, produces $q_\eta'(St) = 0$. From Eq. (37), the prediction of the present analysis is $(q_\eta')^2 = \frac{(1 - \sqrt{1 - 16St^2})}{(8St^2)}$, which asymptotes to 1 for $St \ll 1$ and to $1/(8St^2)$ for $St \gg 1$. These predictions are in full agreement with the direct computations, showing that the present analysis captures at least the first and second order behavior of $\lambda_\eta(St)$ correctly (Figure 20).

To derive Eq. (16), we employed the assumption of long-term incompressibility of the underlying flow along the trajectory of particles, viz. $\nabla \cdot \mathbf{u} = 0$. This condition is satisfied in an incompressible flow, where $\nabla \cdot \mathbf{u} = 0$, or when the long-term variation of $\nabla \cdot \mathbf{u}$ is negligible. For the three cases shown in Figure 20 the incompressibility condition is satisfied instantaneously for the two- and three-dimensional flows. $\nabla \cdot \mathbf{u} \neq 0$ for the one-dimensional case, nevertheless a good agreement is observed in Figure 20. It is the long-term variation of $\nabla \cdot \mathbf{u}$ that vanishes in this case since the flow is purely oscillatory (Eq. (26)).

5. Conclusions

We derived an asymptotic solution (Eq. (25)) for the Lyapunov exponents of inertial particles subjected to oscillatory fluid motion. Our analysis is aimed at predicting the rate of expansion or contraction of clouds
Figure 20: Verifying Eq. (16) using the three model problems: the one-dimensional straining flow (a), the two-dimensional random straining flow (b), and the three-dimensional isotropic turbulence (c). The standard deviation of the finite-time Lyapunov exponents $q'$ is computed directly (circles), based on the Lyapunov exponent obtained from the reference results (solid black), and based on the Lyapunov exponents predicted by the present analysis (dashed red). Note that y-axis is logarithmic in (a) and (b) and linear in (c). The decay rate in (c) for $St > 1$ is also second-order.
of inertial particles, and also their rate of crossovers. We employed the sum of the Lyapunov exponents, i.e. the rate of change of volume of a cloud of particles in three dimensions, to characterize regimes of preferential concentration. We showed that our solution is more general and reproduces the pre-established models in literature ([23] and [29]). Consistent with the previous models, the only flow-related parameter that appears in our model is the spectrum of the second invariant of the velocity gradient tensor, viz. the Q-criterion, underscoring its fundamental role in segregation of inertial particles. We employed a canonical setup with unimodal excitation to investigate the behavior of the Lyapunov exponent under a wide range of flow conditions. Only the expansion with no crossovers was observed in a rotating regime, whereas both the contraction and expansion with the possibility of crossovers was observed in a straining regime. In a straining regime, the expansion and crossover occur for a sufficiently large oscillation amplitude. Additionally, a $-1/2$ bound on the rate of contraction (normalized by the particle relaxation time) was found. Our analysis also showed the Lyapunov exponent is linearly proportional to the Q-criterion at low oscillation amplitude and its power of $1/4$ at high oscillation amplitude. These observations, which are confirmed by the numerical simulations, are not captured by the other models. Other available models capture only the linear regime, where oscillation amplitude is small. Discrepancies were also observed between our analysis and the reference results. Neglecting the contribution of the sub- and super-harmonics, considering a diagonal form in Eq. (19) for $\lambda$, and employing the assumption in Eq. (45) are deemed to be the primary sources of discrepancies.

Following this canonical setting, we extended our analysis to complex multidimensional flows, in which a continuous range of frequencies is present. We considered two cases for validation: 1) a two-dimensional random straining synthetic flow and 2) a three-dimensional isotropic forced turbulence. Despite the added complexity, both cases produced results analogous to that of the one-dimensional straining regime with unimodal excitation. In all cases, the contraction rate was proportional to $St$ at small $St$ with an extremum around the onset of crossovers. For larger $St$, the rate of contraction decreases till net expansion is observed at $St \approx 1$. Beyond that $St$, the rate of expansion reaches a maximum and then asymptotes to zero as $St \to \infty$. While all models correctly capture the linear trend at low $St$, only the present analysis provides a good prediction of the subsequent nonlinear behaviors at higher Stokes numbers. Additionally, only our analysis captures the occurrence of particle crossovers at high $St$ and also the non-monotonic variation of the standard deviation of the rate of expansion or contraction versus $St$.

Appendix A. Derivation of Eq. (7)

To relate $\mathcal{C}$ to $\mathcal{C}^t$, the Eulerian form of Eq. (2) must be expressed in terms of Lagrangian quantities. Since the gradient operator can be expressed as $\nabla = (\partial/\partial x)(\partial X/\partial x)$, we have

$$\nabla \cdot \dot{x} = \text{tr} \left( \frac{\partial \dot{x}}{\partial x} \frac{\partial X}{\partial x} \right) = \text{tr} \left( J J^{-1} \right),$$

where $\text{tr}(\bullet)$ is the trace operator. From Jacobi’s formula

$$\text{tr} \left( J J^{-1} \right) = |J|^{-1} \frac{d |J|}{dt},$$

and as a result

$$\nabla \cdot \dot{x} = \frac{d (\ln |J|)}{dt}.$$

From Eqs. (A.3) and (6)

$$\frac{d (t \mathcal{C}^t)}{dt} = \nabla \cdot \dot{x},$$

which in combination with Eq. (2) gives

$$\mathcal{C} = \frac{d (t \mathcal{C}^t)}{dt} = \lim_{t \to \infty} \mathcal{C}^t,$$

completing the derivation.
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