Mixing of spherical bubbles with time-dependent radius in incompressible flows

Vicente Pérez-Muñuzuri* and Daniel Garaboa-Paz

Group of Nonlinear Physics, Faculty of Physics, University of Santiago de Compostela, E-15782 Santiago de Compostela, Spain

(Received 4 August 2015; revised manuscript received 16 December 2015; published 12 February 2016)

The motion of contracting and expanding bubbles in an incompressible chaotic flow is analyzed in terms of the finite-time Lyapunov exponents. The viscous forces acting on the bubble surface depend not only on the relative acceleration but also on the time dependence of the bubble volume, which is modeled by the Rayleigh-Plesset equation. The effect of bubble coalescence on the coherent structures that develop in the flow is studied using a simplified bubble merger model. Contraction and expansion of the bubbles is favored in the vicinity of the coherent structures. Time evolution of coalescence bubbles follows a Lévy distribution with an exponent that depends on the initial distance between bubbles. Mixing patterns were found to depend heavily on merging and on the time-dependent volume of the bubbles.

DOI: 10.1103/PhysRevE.93.023107

I. INTRODUCTION

The advection of finite-size or inertial solid particles in open, unsteady incompressible flows was initially assessed independently by Maxey and Riley [1] and Gatignol [2], and more recently in the context of compressible fluids [3]. Examples of inertial particles include sedimentation processes [4], turbulent flows [5], rain generation [6], composite materials [7], volcanic ash transport [8], and the formation of planetesimals in the early solar system [9], and other processes as transport of dust from soil erosion, combustion, or the mixing of sprays. Finite-size or inertial particle dynamics in fluid flows can differ markedly from Lagrangian particle dynamics, in both their motion and clustering behavior (see Ref. [10] for a review).

The case of bubbles with a time-dependent radius has received less attention because of its increased complexity although wide range of applications. A number of papers have been devoted to study the temporal dynamics of gas bubbles in strong acoustic fields [11], the effect of high local temperatures and pressures developed inside collapsing bubbles that may be used to induce chemical reactions [12], and many other applications including water treatment (oxygenation and purification) and medicine (microbubbles bursting), to mention just a few.

Nevertheless, predicting the motion of bubbles in turbulent flows is a key problem in fluid mechanics that has a bearing on a wide range of applications from oceanography to chemical engineering, although few studies have taken into account the motion of bubbles with a time-dependent volume [13,14].

On the other hand, the mixing and dispersion of Lagrangian and inertial particles have been widely studied in terms of the finite-time Lyapunov exponents (FTLEs) [15,16]. They have been used extensively to identify persisting transport patterns, geometric separatrices, or coherent structures, from trajectories of Lagrangian or inertial particles in the flow.

To our knowledge, the formation of coherent structures in a bubbly flow has not been analyzed yet. A time-dependent volume changes the stresses on the bubble interface, thus modifying its trajectory in the flow. With that purpose, in this paper we use the FTLEs to study the formation of coherent structures in a chaotic flow with inertial contracting and expanding bubbles. The effect of bubbles merging on the formation of coherent transport structures is also considered.

II. MODEL

The motion of bubbles in nonuniform incompressible flows can be modeled by the momentum equation [10,17]

$$\frac{\rho_b}{\rho_f} \frac{dv_i}{dt} = \frac{D}{d} U_i + (\rho_b - \rho_f) g_i + \rho_f C_L (U_i \times \omega_i)$$

$$+ \frac{9 \rho_f v^2}{R^2} U_i + \frac{\rho_f}{2R^3} \left( \frac{d(R^3 U_i)}{dt} + 2R^3 \frac{dU_i}{dt} \right),$$

(1)

where \(i\) is the \(i\) component of the fluid velocity, \(v_i\) is the velocity of the bubble, \(U_i\) that of the fluid, \(U_i = \omega_i \times \omega_i\) the relative velocity between the fluid and the bubble, \(\omega_i\) the fluid vorticity, \(C_L = 0.5\) the lift coefficient for a sphere, \(\rho_b/\rho_f\) the density of the bubble gas or the fluid it displaces, \(v = \mu/\rho_f\) the kinematic viscosity, and \(g_i = \delta_{i3} g\), the gravity. The five terms on the right represent, respectively, the force exerted by the undisturbed flow, the buoyancy, the lift force, the Stokes drag, and the viscous force. In the last term, the effect of a spherical bubble with a time-dependent radius \(R(t)\) has been considered [13]. Other corrections, such as the history forces [13,18], are not included here. The derivative \(D/\text{Dt}\) is taken along the path of a fluid element, whereas the derivative \(d/dt\) is taken along the trajectory of the particle.

The instantaneous bubble radius \(R(t)\) is calculated from the well-known Rayleigh-Plesset equation [19] of bubble dynamics:

$$R \frac{d^2 R}{dt^2} + \frac{3}{2} \left( \frac{dR}{dt} \right)^2 = \frac{P_f - P}{\rho_f}. $$

(2)

Further generalizations of this equation to a compressible fluid [20] have been published, but for the purpose of this study we will keep on a first-order approach, since the bubbly flow is mainly driven by the momentum equation (1). In Eq. (2), \(dR/dt\) and \(d^2R/dt^2\) are the bubble wall velocity and acceleration, respectively, \(P_f\) is the pressure in the fluid at the bubble interface, and \(P\) is the pressure field imposed by the
flow. The pressure at the bubble interface is given by

$$P_f(R) = P_v + P_g = \frac{2\gamma}{R} - \frac{4\mu}{R} \frac{dR}{dt},$$  \hspace{1cm} (3)$$

in which the first two terms are the internal pressure of the bubble related to the partial pressure due to vapor content $P_v$ and gas content $P_g$, respectively, and the last terms account for the interface curvature effect and the viscous stress at the interface. Surface tension is given by $\gamma$. The gas pressure inside the bubble changes as the bubble contracts or expands. As the total amount of gas in the bubble remains constant, the bubble radius and gas pressure are related by $P_g = P_g(0) R^3/R_0^3$, where $a = 1$ for an isothermal process, or equal to the ratio of specific heats for an adiabatic process. The external pressure at depth $x_3$ is calculated in terms of the hydrostatic relationship $P = P_0 + \rho_f g x_3$, while the vapor pressure is considered to remain constant.

For our simulations we choose the well-known Arnold-Beltrami-Childress (ABC) flow ($u_i = dx_i/dt$) \cite{21}, defined as

$$dx_1/dt = A \sin(x_1) + C \cos(x_2),$$

$$dx_2/dt = B \sin(x_1) + A \cos(x_3),$$

$$dx_3/dt = C \sin(x_2) + B \cos(x_1).$$  \hspace{1cm} (4)

This flow is divergenceless and is an exact, three-dimensional, steady-state solution of the Euler equation with the Beltrami property $\mathbf{u} = \nabla \times \mathbf{u}$. For the parameter configuration $A = \sqrt{3}$, $B = \sqrt{2}$, and $C = 1$, the flow generates chaotic streamlines \cite{21}. Other 3D geophysical flows could have been selected, but the FTLE fields for the ABC flow have been profusely studied for Lagrangian particles \cite{22} while the chaotic dynamics has been analyzed for inertial particles \cite{23}. To be consistent with the rest of the units employed along the paper, SI units have been used for the velocity and distances.

In order to characterize the transport of bubbles, we introduce the finite-time Lyapunov exponents, that measure, at a given location, the maximum stretching rate of an infinitesimal fluid parcel over the interval $[t_0, t_0 + \tau]$ starting at $\mathbf{x}(t_0) = \mathbf{x}_0$ and finishing at $\mathbf{x}(t_0 + \tau)$ \cite{15}. The integration time $\tau$ must be predefined and it has to be long enough to allow trajectories to explore the coherent structures present in the flow. The FTLE fields $\sigma$ are computed along their trajectories in the flow as \cite{24}

$$\sigma(\mathbf{x}_0, t_0, \tau) = \frac{1}{\tau} \log \sqrt{\mu_{\max}[C(\mathbf{x}_0)]},$$  \hspace{1cm} (5)$$

where $\mu_{\max}$ is the maximum eigenvalue of the right Cauchy-Green deformation tensor $C = F \cdot F$, and $F(\mathbf{x}_0) = \nabla[\mathbf{x}(t_0 + \tau)]$. Repelling (attracting) coherent structures for $\tau > 0$ ($\tau < 0$) can be thought of as finite-time generalizations of the stable (unstable) manifolds of the system. These structures govern the stretching and folding mechanism that controls flow mixing.

For the numerical experiments, initially a cluster of $N = 300 \times 300$ bubbles with radius $R_0$ was regularly spaced in the domain $(x_1, x_2) \in [0, 2\pi] \times [0, 2\pi]$ m, $\delta_0 = 2\pi/N$. Although ABC flow is $2\pi$ periodic, bubbles in the $(x_1, x_2)$ plane move in all directions without constraints, and to avoid particles to reach the upper layer where $P = P_0$, initially bubbles are located deeply enough so they cannot reach the upper boundary during the integration time. The trajectories of these bubbles are calculated by integrating the equations above using a fourth-order Runge-Kutta scheme with a fixed time step $\Delta t = 10^{-3}$ s and initial conditions $v_i = u_i$, $R = R_0$, and $dR/dt = 0$.

Finally, a very viscous fluid was considered to avoid (i) surface deformation of the bubbles and (ii) their break during their motion, so they keep their spherical shape although their volume may change. In terms of the Weber number $We = 2R_0 \rho_0 u/\gamma$, i.e., inertial forces were supposed to be smaller than surface tension forces, so the spherical shape of the bubbles was preserved. To analyze the effect of the Reynolds number $Re = 2R_0 |u|/v$ on the motion of the bubbles, fluid parameters $\rho_f$, $\gamma$, and $v$ vary with temperature, and so does the density of the bubble gas $\rho_0$. Table I summarizes the set of physical constants and their values used in our simulations.

<table>
<thead>
<tr>
<th>Bubble</th>
<th>Air</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_f$ (Pa)</td>
<td>0.03 $\times 10^5$</td>
</tr>
<tr>
<td>$P_0$ (Pa)</td>
<td>1.01 $\times 10^5$</td>
</tr>
<tr>
<td>$\rho_f$ (kg m$^{-3}$)</td>
<td>$P_0/R(T)$</td>
</tr>
<tr>
<td>$\rho_0$ (kg m$^{-3}$)</td>
<td>$a_1 t^3 + a_2 t^2 + a_3 t + a_4$</td>
</tr>
<tr>
<td>$\gamma$ (Nm$^{-1}$)</td>
<td>29.06 $\times 10^{-3} - 2.67 \times 10^{-5} T$</td>
</tr>
<tr>
<td>$v$ (m$^2$ s$^{-1}$)</td>
<td>$v^0 \exp[\beta(1/T - 1/T_0^0)]$</td>
</tr>
</tbody>
</table>

### III. RESULTS

We have studied the motion of bubbles with different radius in a viscous flow in terms of the FTLE fields. To that end, two situations have been considered; in the first one, bubbles do not interact with each other, while in the second case, bubbles can merge and coalesce into a single bubble. The rebound of bubbles was not considered here.

The FTLE fields (Fig. 1) change dramatically as the Reynolds number increases, either by increasing (decreasing) the flow temperature (viscosity) or increasing the initial bubble’s radius. Decreasing the bubbles’ radius, the inertial effects diminish and the FTLE field approaches that of the Lagrangian particles (Fig. 2). Increasing the Reynolds number, the inertial effects wrinkle the coherent structures otherwise clearly defined by the Lagrangian particles. Inertial effects become more important as the bubbles’ volume increases, folding and mixing the coherent structures. For the largest Re value shown here, the coherent structures have been largely modified.

The values of the bubbles’ radius at the end of the integration time $\tau$ are shown in Fig. 3. Note that upward (downward) motions favor a larger (smaller) radius than $R_0$. Bubbles tend to concentrate inside of the vortices, as they are lighter than the surrounding flow, and then they are transported upward or downward by the flow depending on their corresponding...
Mixing of spherical bubbles with time-dependent radius.

**Fig. 1.** XY field of finite-time Lyapunov exponents $\sigma (s^{-1})$ for different bubbles’ initial radius $R_0$ and Reynolds numbers. Darker (red) colors indicate larger values. Local maxima in the plot are repelling coherent structures, responsible for the flow stretching in their vicinity. Integration time $\tau = 5 \text{ s}$. (a) $R_0 = 10^{-4} \text{ m}$, Re = 0.9; (b) $R_0 = 10^{-3} \text{ m}$, Re = 25; (c) $R_0 = 10^{-2} \text{ m}$, Re = 92; (d) $R_0 = 10^{-2} \text{ m}$, Re = 253.

For larger initial radius, inertial effects compensate the flow transport and new transport barriers develop, trapping bubbles at different levels with different radius. See, for example, for the bottom-right panel in Fig. 3, $R_0 = 10^{-2} \text{ m}$, the formation of bubbles with intermediate values of $R/R_0$, if compared with other cases with smaller initial radius. On the other hand, in the neighborhood of the coherent structures, $R/R_0$ attains minimum values as the bubbly flow moves to deeper depths, thus decreasing the bubbles’ volume.

The effect of bubble contraction and expansion is analyzed in Fig. 4. To that end, $R(t)$ is kept constant along the simulations and equal to its initial value $R_0$. In other words, the Rayleigh-Plesset equation (2) was not solved. The left figure shows the FTLEs calculated for a constant radius, while the right figure shows the differences between both cases. Note that the main differences are near the shear regions where the coherent structures develop and the effects of a time-dependent radius are more important, as mentioned above. Additionally, increasing the integration time from $\tau = 5 \text{ s}$ in Fig. 1 to $\tau = 10 \text{ s}$ allows one to observe the formation of new coherent structures. As mixing inside the vortices favors downward and upward motions, the effect of not considering a time-dependent radius is clearly visible there.

To assess quantitatively the effects of a time-varying bubble volume on the FTLEs, we perform systematic simulations for different values of the Reynolds number Re, both by varying the initial radius $R_0$ and the fluid temperature $T$. To characterize the resulting FTLE fields, it is convenient to calculate the mean FTLE value $\bar{\sigma}$ and the percentage of bubbles with $\sigma < 0$. These negative values characterize groups of bubbles that cluster together [3]. Results are shown in Fig. 5. Increasing the Reynolds number leads to an increase of mixing and a decrease of clustering. The larger mean FTLE values correspond to an increase in variability (note the increasing number of coherent structures with Re shown in Fig. 1) of final bubble positions. Thus, the increasing dispersion does not favor cluster formation.

**A. Bubble merging: Lévy distribution**

The effect of coalescence bubble motion has been investigated in terms of the formation of coherent structures in the ABC flow. Coalescence is known to depend on mass transfer,

**Fig. 2.** XY field of Lagrangian finite-time Lyapunov exponents $\sigma (s^{-1})$ for the ABC flow. Darker (red) colors indicate larger values. Same color scale as in Fig. 1. Integration time $\tau = 5 \text{ s}$.

**Fig. 3.** XY field of bubble’s radius $R/R_0$ for different initial radius $R_0$ and Reynolds numbers. Parameters as in Fig. 1.

**Fig. 4.** FTLE $(s^{-1}) x y$ field (left panel) considering a constant bubbles’ radius $R_0 = 5 \times 10^{-3} \text{ m}$, and the difference with the FTLE field considering a time-dependent radius $R(t)$ (right panel). Re = 0.9 and $\tau = 10 \text{ s}$.
surface tension, van der Waals forces, surface diffusion, electrostatic and double-layer forces, and on the surrounding flow conditions, among other effects (for a review see [25]). However, our purpose in this paper is to investigate the transport of coalescence bubbles in a chaotic flow, assuming that the coalescence time is much shorter than the flow time scale. With that purpose, we have used a very simple procedure to merge two bubbles into one. After collision, the new resulting spherical bubble has a volume equal to the total volume of the bubbles just prior to the merging, while its radius velocity is the mean of the merging velocities. By doing so, we assume that the time required for the internal pressure gas to reach equilibrium is much shorter than the time the external liquid flow needs to respond to changes in the bubble pressure. So, the new internal pressure is assumed to obtain its equilibrium state immediately, and thus the shape of the new bubble is considered to become spherical immediately as well. In order for the numerical procedure to calculate the FTLEs to be stable, a virtual bubble is created along the new one with similar properties, so the total number of bubbles remains constant.

The main results for bubble merging are shown in Fig. 6. The number of coalescence bubbles increases with decreasing initial distance between bubbles $\delta_0/R_0$ (mesh resolution to bubble size ratio) (a) as merging is favored mostly for initial times. On the other hand, the FTLE fields obtained considering merging do not differ qualitatively from those shown in Fig. 1. However, the standard deviation of the difference between the FTLE fields calculated with and without considering merging increases with decreasing $\delta_0/R_0$ (b). After merging, subsequent evolution of the coalescence bubbles modified their trajectories with respect to their parents, thus increasing the spatial FTLE variability.

Finally, we analyze the time evolution of the number of coalescence bubbles [Fig. 6(c)]. $N_B \sim t^x$. Three different regimes have been identified for any parameter used along this paper. Initially, during some period of time $t_0$ bubbles do not merge together. This period of time decreases with decreasing initial separation between bubbles $\delta_0/R_0$. Later, up to a critical time, the number of merging bubbles grows with time with an exponent $x_1$ (dashed line), while above that time bubbles merge (dash-dotted line) with an exponent $x_2 < x_1$. For the last case, the probability to merge decreases due to flow dispersion.

Coalescence of bubbles occurs suddenly and does not happen continuously in time [Fig. 6(c)]. This behavior can be described statistically by two elementary functions, the increasing factor of coalescence bubble distribution $p(\Delta N)$ and the waiting time distribution $q(t_w)$:

$$p(\Delta N) \propto (\Delta N)^{-\alpha}, \quad q(t_w) \propto (t_w)^{-\beta}. \quad (6)$$

Bubbles are assumed to merge at discrete intervals of time, increasing the number of coalescence bubbles by a factor $\Delta N$. The time intervals without merging are known as waiting times $t_w$. Power-law waiting times with tail exponent $\beta > 1$ have been observed in the wait between solar flares [26] or the wait between earthquakes [27], among others. Figure 6(d) shows the exponents $\alpha$ and $\beta$ as a function of the ratio $\delta_0/R_0$ obtained by fitting a power law to the Lévy distributions given by Eq. (6) [28]. The exact value of the exponents can be affected by the finite length of the integration time $\tau$, which underestimates the occurrence of long waiting times. The use of larger values of $\tau$ attempts to locate the initial position of the particles deeper, thus avoiding bubble concentration at the free surface due to buoyancy. Thus, the higher pressure values make the numerical solution of the Rayleigh-Plesset equation (2) more difficult because of its stiffness.

Decreasing the initial distance between bubbles $(\delta_0/R_0 \to 0)$ increases the coalescence rate and the population of coalescence bubbles. In terms of the distribution probabilities (6), $q(t_w)$ has a short tail ($\beta \to \infty$), while $p(\Delta N)$ has a heavy tail ($\alpha \to 0$). In the opposite limit, for very dilute flows $(\delta_0/R_0 \to \infty)$, the merging probability is nearly zero, so very large waiting times [heavy tails for $q(t_w)$] and small $\Delta N$ values [short tails for $p(\Delta N)$] are attained.
IV. CONCLUSIONS

Through numerical experiments, we have provided some results about the influence of contraction and expansion of bubbles on the FTLEs and the coherent structures developed in the flow. The influence of a time-dependent radius is more important in the vicinity of these structures as bubbles move in the vertical direction. The FTLE variability increases with the Reynolds number, while new coherent structures develop trapping bubbles during their motion.

On the other hand, the effect of merging on the mixing and dispersion of bubbles favors an increase of the FTLE variability with the number of coalescence bubbles. The time evolution of this increase was found to follow a Lévy distribution with exponents that depend on the initial distance between bubbles. We expect that no significant differences should be observed if a more refined merging model is used.

Mixing patterns in bubbly flows have been shown to depend heavily on the time-dependent volume of the bubbles, above all for increasing Reynolds number. Such phenomena deserve further numerical and experimental studies. For example, dispersion of injected CO₂ emissions deep into the ocean [29], or the gas exchange in bubble column reactors [30], could be optimized in terms of the mixing patterns depicted by their respective FTLE fields.

ACKNOWLEDGMENTS

The authors would like to acknowledge the support of Ministerio de Economía y Competitividad and Xunta de Galicia under Research Grants No. CGL2013-45932-R and No. GPC2015/014, respectively, and the contribution of the COST Action No. MP1305. The computational part of this work was done in the Supercomputing Center of Galicia, CESGA.