Model for the dynamics of two interacting axisymmetric spherical bubbles undergoing small shape oscillations

Eru Kurihara,^{a)} Todd A. Hay,^{b)} Yurii A. Ilinskii, Evgenia A. Zabolotskaya, and Mark F. Hamilton

Applied Research Laboratories, The University of Texas at Austin, Austin, Texas 78713–8029

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Interaction between acoustically driven or laser-generated bubbles causes the bubble surfaces to deform. Dynamical equations describing the motion of two translating, nominally spherical bubbles undergoing small shape oscillations in a viscous liquid are derived using Lagrangian mechanics. Deformation of the bubble surfaces is taken into account by including quadrupole and octupole perturbations in the spherical-harmonic expansion of the boundary conditions on the bubbles. Quadratic terms in the quadrupole and octupole amplitudes are retained, and surface tension and shear viscosity are included in a consistent manner. A set of eight coupled second-order ordinary differential equations is obtained. Simulation results, obtained by numerical integration of the model equations, exhibit qualitative agreement with experimental observations by predicting the formation of liquid jets. Simulations also suggest that bubble-bubble interactions act to enhance surface mode instability. © 2011 Acoustical Society of America. [DOI: 10.1121/1.3626137]

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I. INTRODUCTION

High-speed photographs have revealed that a bubble moving in the proximity of an interface or a neighboring bubble may undergo rapid translation and form a liquid jet.^{1–3} Beginning with Plesset, who investigated single bubble shape oscillations arising from the Rayleigh–Taylor instability, a number of theoretical studies have focused on modeling the dynamics of stationary aspherical bubbles in high intensity sound fields.^{4,5} Formation of a liquid jet by a collapsing gas bubble was investigated numerically by Plesset and Chapman,⁶ who predicted the formation of a high-speed liquid jet resulting from bubble collapse near a rigid wall in an incompressible and inviscid liquid. Lauterborn and Bolle⁷ as well as others^{8,9} performed experiments studying the collapse of laser-generated gas bubbles in the vicinity of a wall, and their observations agree qualitatively with the solution obtained by Plesset.⁶ Tomita and Shima⁸ investigated bubbles excited by shock waves and showed that the resulting liquid jets can cause pitting on an adjacent surface. The effects of viscous dissipation and molecular fluctuations on surface deformation have also been thoroughly considered.^{5,10,11} The shape stability of an isolated bubble undergoing translational motion was investigated by Feng and Leal,¹² Reddy and Szeri,¹³ and Doinikov,¹⁴ who demonstrated that bubble translation tends to enhance shape oscillations. Testud-Giovanneschi *et al.*⁹ demonstrated through high-speed photographs of coupled bubbles that the bubbles may undergo aspherical deformation and translation.

Analytical models for two or many interacting bubbles have been developed, some of which account for shape

deformation.^{15–20} In addition, shock-wave induced bubble collapse and jetting near a liquid-solid interface have been studied via direct solution of the Navier–Stokes equations.^{21,22} Blake *et al.* studied bubble deformation near a rigid boundary²³ and a free surface.²⁴ Modeling the system with the boundary element method (BEM), they found that the liquid jet is directed away from the free surface. Chahine and Duraiswami²⁵ also applied the BEM to bubble surface deformations. By introducing a cut into the domain so that the bubble surface remains simply connected, Best²⁶ and Chang *et al.*²⁷ adapted the BEM to model the toroidal shape that is often assumed by the bubbles after the liquid jet penetrates the bubble wall. In recent years various numerical techniques such as the level set²⁸ and front tracking methods²⁹ have also been applied to model bubble dynamics.

In the present work, Lagrangian mechanics is used to derive dynamical equations which describe the motion of two interacting nominally-spherical bubbles undergoing small shape deformations immersed in a viscous liquid. The velocity potential in the liquid is expressed as an expansion in spherical harmonics. Quadrupole and octupole modes are included as perturbations of the bubble surface to quadratic order, and viscous damping of the surface modes is also taken into account. Doinikov's model¹⁴ for a single translating bubble includes an arbitrary number of surface modes and is also accurate to quadratic order in the mode amplitudes, but neglects viscous damping of the modes. The present theory can, in principle, be extended to include modes of higher order than the octupole, but the resulting dynamical equations would be unwieldy for a two bubble system and it was found that truncation at the octupole is sufficient to capture the dynamics qualitatively, for example, describing the direction of jetting. Fujikawa and Takahira^{19,20} developed analytical models for arbitrary numbers of interacting bubbles, but retained only linear terms in the surface perturbation amplitudes. Furthermore, their model is presented as a

^{a)}Present address: Department of Mechanical and Energy Systems Engineering, Oita University, Oita 870-1192, Japan.

^{b)}Author to whom correspondence should be addressed. Electronic mail: hayta@arlut.utexas.edu

system of coupled first-order ordinary differential equations for the state variables and velocity potential coefficients. In contrast, quadratic terms in the quadrupole and octupole amplitudes are retained in the present work, and a standard set of eight coupled second-order ordinary differential equations is derived.

We first calculate the kinetic and potential energies of the system, the dynamical equations and the viscous loss terms in Sec. II. Numerical simulations are then presented in Sec. III. The simulations illustrate the free and forced dynamics of the system, and demonstrate that the model qualitatively reproduces observed bubble behavior, such as the onset of liquid jets in neighboring bubbles. Finally, the effect of bubble interaction on the stability of the surface amplitudes is discussed.

II. THEORY

The geometry of the problem is illustrated in Fig. 1. Two bubbles are immersed in an infinite incompressible liquid and positioned along the x axis at locations X_i ($i = 1, 2$). We assume $X_2 > X_1$ and denote the distance separating the bubbles by $D = X_2 - X_1$. Due to the absence of any other confining surfaces in the liquid, symmetry about the x axis may be assumed, and the surface of i th bubble in its local spherical coordinate system may be written as a multipole expansion of the form

$$\mathbf{r}_{si}(t, \theta_i) = \left(A_{i0}(t)P_0(\cos \theta_i) + \sum_{n=2}^{\infty} A_{in}(t)P_n(\cos \theta_i) \right) \mathbf{e}_{ri}, \quad (1)$$

where t is time, $\mathbf{e}_{ri}$ are unit vectors pointing radially outward from each bubble center at angles θ_i with the x axis, $P_n(\cos \theta_i)$ are Legendre polynomials of order n , and $A_{in}(t)$ are time-dependent expansion coefficients. The present analysis can be extended to nonaxisymmetric surface distortions but, as mentioned in Ref. 14, doing so would make the resulting expressions extremely bulky without changing the fundamental dynamics. The focus of the present work is *qualitative* description of shape deformation in coupled bubble dynamics, including the onset of jet formation. Accounting for the latter requires retention of a spherical harmonic in the expression for the bubble surface that is antisymmetric about $\theta_i = \pi/2$, the first of which is the octupole mode ($n = 3$). Therefore we truncate Eq. (1) at $n = 3$, to obtain

$$\begin{aligned} \mathbf{r}_{si}(t, \theta_i) &= [A_{i0}(t)P_0(\cos \theta_i) + A_{i2}(t)P_2(\cos \theta_i) \\ &\quad + A_{i3}(t)P_3(\cos \theta_i)] \mathbf{e}_{ri} \\ &= [R_i(t) + A_{i2}(t)P_2(\cos \theta_i) + A_{i3}(t)P_3(\cos \theta_i)] \mathbf{e}_{ri} \end{aligned} \quad (2)$$

FIG. 1. Geometry and coordinate system for two interacting spherical bubbles with shape perturbations.

for the surfaces of the bubbles, where $A_{i0}(t)$ [denoted $R_i(t)$ hereafter] is the time-dependent amplitude of the monopole mode, and $A_{i2}(t)$ and $A_{i3}(t)$ are the time-dependent amplitudes of the quadrupole and octupole modes, respectively. The quadrupole and octupole modes account for deformation of the bubble surface from a spherical shape. Truncation of Eq. (1) at the octupole mode keeps the model simple while still accounting for initiation of a jet. The dipole ($n = 1$) mode corresponds to translation of the local coordinate system, and is therefore omitted from Eq. (2) but is included in the velocity boundary condition for the bubbles.

The dynamical equations will be derived as expansions in $A_{in}/R_i = O(\varepsilon)$ for $n = 2, 3$, and $R_i/D = O(\mu)$. It is assumed that both ε and μ are small compared to unity. Terms up to order $O(\varepsilon^2\mu^2)$ will be retained. The motion of the bubbles is described by the radial velocities $\dot{R}_i$ [later replaced by the velocity of an “effective radius” defined by Eq. (11)], translational velocities $U_i = \dot{X}_i$, and the quantities A_{in} , where overdots denote differentiation with respect to time. Lagrange’s equations describing the dynamics of the system are

$$\frac{d}{dt} \left(\frac{\partial \mathcal{L}}{\partial \dot{q}} \right) = \frac{\partial \mathcal{L}}{\partial q} - \frac{\partial F}{\partial \dot{q}}, \quad (3)$$

where q is a generalized coordinate (taking the value R_i , X_i , A_{i2} , or A_{i3}), F is the dissipation function, and

$$\mathcal{L}(q, \dot{q}) = \mathcal{K}(q, \dot{q}) - \mathcal{V}(q) \quad (4)$$

is the Lagrangian of the system, where $\mathcal{K}$ and $\mathcal{V}$ are the kinetic and potential energies, respectively.

We now proceed to calculate the kinetic and potential energies, as well as the dissipation function. Since the external acoustic excitations considered in the numerical simulations correspond to wavelengths that are large relative to the bubble radii and separation distance, it is sufficient to take acoustic excitation into account through the dynamical equation for the monopole mode alone, e.g., the primary Bjerknes force in the dynamical equation for the dipole (translation) mode is ignored.

A. Kinetic energy

The motion of both the liquid surrounding the bubbles and the gas inside the bubbles contribute to the kinetic energy of the system. However, the gas density is small compared to that of the liquid, and therefore kinetic energy of the gas may be neglected. The liquid is assumed to be irrotational, such that its motion may be described by the gradient of a scalar velocity potential ϕ satisfying Laplace’s equation

$$\nabla^2 \phi = 0. \quad (5)$$

For a liquid at rest at infinity the kinetic energy may be expressed as an integral over the liquid volume V_{liq} as

$$\mathcal{K} = \frac{\rho}{2} \int_{V_{\text{liq}}} |\nabla \phi|^2 dV_{\text{liq}}. \quad (6)$$

After application of Green’s first identity and Eq. (5) the volume integral in Eq. (6) can be transformed into an integral over the bubble surfaces S_i , and the kinetic energy written

$$\begin{aligned}\mathcal{K} &= -\frac{\rho}{2} \sum_{i=1}^2 \int_{S_i} \phi(r_{si}, \theta_i) [\dot{r}_{si} + U_i P_1(\cos \theta_i)] dS_i \\ &= -\pi \rho \sum_{i=1}^2 \int_0^\pi \phi(r_{si}, \theta_i) [\dot{r}_{si} + U_i P_1(\cos \theta_i)] r_{si}^2 \sin \theta_i d\theta_i. \quad (7)\end{aligned}$$

As Eq. (7) shows, calculation of the kinetic energy requires that the velocity potential be known. While an expression

for the velocity potential that satisfies Eq. (5) exactly is not available, an approximate expression may be found which satisfies Eq. (5) to desired orders of ε and μ . In the present model terms up to $O(\varepsilon^2 \mu^2)$ in the kinetic energy and dynamical equations are retained, and the procedure used to derive ϕ is outlined in the Appendix. Substitution of Eq. (A9) for the velocity potential and Eq. (2) for r_{si} into Eq. (7) yields

$$\begin{aligned}\mathcal{K} &= 2\pi \rho \sum_{i=1}^2 R_i^2 \left\{ R_i \dot{R}_i^2 + \frac{1}{6} R_i U_i^2 + \frac{R_j^2}{D} \dot{R}_i \dot{R}_j + \frac{1}{2D^2} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) (R_i \dot{R}_j U_i - R_j \dot{R}_i U_j) - \frac{3}{10} A_{i2} U_i^2 + \frac{3}{10D^2} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) (A_{j2} \dot{R}_i U_j - A_{i2} \dot{R}_j U_i) \right. \\ &\quad + \frac{\dot{A}_{i2}^2 R_i}{15} + \frac{\dot{A}_{i3}^2 R_i}{28} + \frac{3}{35} U_i (3A_{i2} \dot{A}_{i3} - 2\dot{A}_{i2} A_{i3}) + \frac{1}{15} \frac{A_{i2}^2}{R_i} \dot{R}_i^2 + \dot{R}_i \left(\frac{2}{3} A_{i2} \dot{A}_{i2} + \frac{3}{7} A_{i3} \dot{A}_{i3} \right) \\ &\quad + \frac{1}{5} \frac{A_{j2}}{D} \dot{R}_i (\dot{R}_j A_{j2} + 2R_j \dot{A}_{j2}) + \frac{1}{7} \frac{A_{j3}}{D} \dot{R}_i (\dot{R}_j A_{j3} + 2R_j \dot{A}_{j3}) + \frac{1}{35} \frac{U_i}{R_i} \left(\frac{113}{10} A_{i2}^2 U_i - \frac{8}{7} A_{i3}^2 U_i - 12A_{i2} A_{i3} \dot{R}_i \right) \\ &\quad + \frac{1}{5} \frac{R_j^2 A_{i2}}{R_i D} \dot{R}_j \left(\frac{A_{i2}}{R_i} \dot{R}_i + 2\dot{A}_{i2} \right) + \frac{1}{7} \frac{R_j^2 A_{i3}}{R_i D} \dot{R}_j \left(\frac{A_{i3}}{R_i} \dot{R}_i + 2\dot{A}_{i3} \right) \\ &\quad + \frac{6}{35} \frac{R_j}{D^2} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \left[R_j A_{i3} \dot{R}_j \dot{A}_{i2} - R_j A_{j3} \dot{R}_i \dot{A}_{j2} - 2A_{j2} A_{j3} \dot{R}_i \dot{R}_j \left(1 - \frac{R_j}{R_i} \right) \right] + \frac{27}{70} \frac{R_j^2}{D^2} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) (A_{i2} \dot{A}_{i3} \dot{R}_j - A_{j2} \dot{A}_{j3} \dot{R}_i) \\ &\quad + \frac{U_i R_i}{D^2} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \left[R_j \left(\frac{A_{j2} \dot{A}_{j2}}{5} + \frac{A_{j3} \dot{A}_{j3}}{7} \right) + \dot{R}_j \left(\frac{A_{j2}^2}{10} + \frac{A_{j3}^2}{14} \right) \right] - \frac{U_i R_j^3}{R_i D^2} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \left[\frac{A_{i2} \dot{A}_{i2}}{5} + \frac{A_{i3} \dot{A}_{i3}}{7} + \frac{\dot{R}_i}{R_i} \left(\frac{A_{i2}^2}{10} + \frac{A_{i3}^2}{14} \right) \right] \\ &\quad \left. + \frac{1}{35} \frac{R_j}{D^2} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \left[\frac{183}{10} \left(A_{i2}^2 U_i \dot{R}_j \frac{R_j}{R_i} - A_{j2}^2 U_j \dot{R}_i \right) + \frac{1}{7} \left(173A_{j3}^2 U_j \dot{R}_i - 53A_{i3}^2 U_i \dot{R}_j \frac{R_j}{R_i} \right) \right] \right\} + O(\varepsilon^2 \mu^3) + O(\varepsilon^3 \mu^2), \quad (8)\end{aligned}$$

where $\mathbf{e}_{ij}$ is the unit vector pointing along the x axis from bubble i to bubble j and $\mathbf{e}_x$ is the unit vector pointing along the x axis in the direction of increasing x . The index j takes the value 2 when $i = 1$ or the value 1 when $i = 2$. Therefore, the quantity $\mathbf{e}_{ij} \cdot \mathbf{e}_x$ is

$$\mathbf{e}_{ij} \cdot \mathbf{e}_x = \begin{cases} 1 & \text{when } i = 1, j = 2, \\ -1 & \text{when } i = 2, j = 1. \end{cases} \quad (9)$$

B. Potential energy

We account for potential energy due to changes in bubble volume and due to surface tension. The potential energy due to volume change is

$$\mathcal{V}_{gi} = V_i \left(P_0 + \frac{P_i}{\kappa - 1} \right) = \frac{4}{3} \pi R_{ei}^3 \left(P_0 + \frac{P_i}{\kappa - 1} \right), \quad (10)$$

where P_i is the pressure in the liquid at the bubble wall, κ is the polytropic exponent of the gas, P_0 is the equilibrium pressure at infinity, V_i is the bubble volume, and

$$R_{ei} \equiv \left(\frac{3V_i}{4\pi} \right)^{1/3} \quad (11)$$

is defined to be the effective radius. An expression for R_{ei} may be found by calculating the bubble volume:

$$\begin{aligned}V_i &= \frac{2\pi}{3} \int_0^\pi r_{si}^3 \sin \theta_i d\theta_i \\ &= \frac{2\pi}{3} \int_0^\pi [R_i + A_{i2} P_2(\cos \theta_i) + A_{i3} P_3(\cos \theta_i)]^3 \sin \theta_i d\theta_i \\ &= \frac{4\pi}{3} R_i^3 \left[1 + \frac{3}{R_i^2} \left(\frac{A_{i2}^2}{5} + \frac{A_{i3}^2}{7} \right) \right] + O(\varepsilon^3). \quad (12)\end{aligned}$$

The amplitude of the monopole mode may therefore be approximated to order ε^2 as

$$R_i = R_{ei} - \frac{1}{R_{ei}} \left(\frac{A_{i2}^2}{5} + \frac{A_{i3}^2}{7} \right), \quad (13)$$

which is substituted in Eq. (8) to obtain an expression for the kinetic energy in terms of R_{ei} .

Potential energy due to surface tension is given by $\mathcal{V}_{\sigma i} = \sigma S_i$,³⁰ where σ is the surface tension of the liquid and

$$\begin{aligned}S_i &= 2\pi \int_0^\pi r_{si}^2 \sqrt{1 + \frac{1}{r_{si}^2} \left(\frac{\partial r_{si}}{\partial \theta_i} \right)^2} \sin \theta_i d\theta_i \\ &\simeq 2\pi \int_0^\pi \left[r_{si}^2 + \frac{1}{2} \left(\frac{\partial r_{si}}{\partial \theta_i} \right)^2 \right] \sin \theta_i d\theta_i \\ &= 4\pi R_{ei}^2 \left[1 + \frac{2}{5} \left(\frac{A_{i2}}{R_{ei}} \right)^2 + \frac{5}{7} \left(\frac{A_{i3}}{R_{ei}} \right)^2 \right] + O(\varepsilon^3) \quad (14)\end{aligned}$$

is the surface area of the i th bubble. The potential energy due to surface tension is therefore

$$\mathcal{V}_{oi} = \sigma \mathcal{S}_i = 4\pi\sigma \left(R_{ei}^2 + \frac{2}{5}A_{i2}^2 + \frac{5}{7}A_{i3}^2 \right) + O(\varepsilon^3), \quad (15)$$

and the total potential energy of the system is

$$\mathcal{V} = \sum_{i=1}^2 (\mathcal{V}_{gi} + \mathcal{V}_{oi}). \quad (16)$$

C. Viscous dissipation

In treating viscous losses we will assume that the vorticity generated by viscous stresses remains local to the bubble surface and is negligible outside a small boundary layer region surrounding each bubble.^{5,10} Following Ref. 5 we include viscous losses due to vorticity to order $O(\delta\varepsilon)$, where δ is the ratio of the boundary layer thickness to the bubble radius. This is discussed in more detail in Sec. II D. The flow field outside the boundary layer can be considered potential flow, and viscous losses in this region may be included to $O(\varepsilon^2\mu^2)$ via a dissipation function.^{11,30} For the present case the dissipation function is

$$\begin{aligned} F = & \frac{\nu}{2} \sum_{i=1}^2 \int_{S_i} \nabla v_{si}^2 \cdot d\mathbf{S}_i \\ = & \frac{\nu}{2} \sum_{i=1}^2 \left\{ -8R_{ei}\dot{R}_{ei}^2 + 12A_{i2}U_i^2 - 6R_{ei}U_i^2 - \frac{16}{3}R_{ei}\dot{A}_{i2}^2 \right. \\ & - \frac{32}{15}\dot{R}_{ei}A_{i2}\dot{A}_{i2} - 5R_{ei}\dot{A}_{i3}^2 - \frac{20}{7}\dot{R}_{ei}A_{i3}\dot{A}_{i3} + \frac{8}{3}A_{i2}^2\dot{R}_{ei}^2 \\ & + 4\frac{A_{i3}^2}{R_{ei}}\dot{R}_{ei}^2 + \frac{36}{35}U_i \left(16A_{i3}\dot{A}_{i2} - 19\dot{A}_{i3}A_{i2} + 18\frac{A_{i3}A_{i2}}{R_{ei}}\dot{R}_{ei} \right) \\ & - \frac{6U_i^2}{7R_{ei}} \left(18A_{i3}^2 + \frac{397}{25}A_{i2}^2 \right) - 12\dot{R}_{ej}\frac{R_{ej}^2}{D^2}(\mathbf{e}_{ij} \cdot \mathbf{e}_x) \\ & \times \left[U_i \left(R_{ei} - 2A_{i2} + \frac{18A_{i2}^2}{7R_{ei}} \right) + \frac{A_{i2}}{35} \left(57\dot{A}_{i3} - 54\frac{A_{i3}}{R_{ei}}\dot{R}_{ei} \right. \right. \\ & \left. \left. - 48\frac{A_{i3}}{A_{i2}}\dot{A}_{i2} + \frac{397}{5}\frac{A_{i2}}{R_{ei}}U_i \right) \right] \right\}, \quad (17) \end{aligned}$$

where ν is the kinematic viscosity of the liquid, and $\mathbf{v}_{si}$ is the liquid velocity at the bubble surface, given by Eq. (A4).

D. Dynamical equations for two interacting bubbles

Eight coupled dynamical equations are obtained by substituting Eqs. (8) [after substitution of Eq. (13) for R_i], (16), and (17) into Eq. (3) with $q = R_{ei}, X_i, A_{i2}$, or A_{i3} . The dynamical equation for the effective radii R_{ei} is

$$\begin{aligned} R_{ei}\ddot{R}_{ei} + \frac{3}{2}\dot{R}_{ei}^2 = & \frac{P_i - P_0}{\rho} - \nu \left[4\frac{\dot{R}_{ei}}{R_{ei}} - 3(\mathbf{e}_{ij} \cdot \mathbf{e}_x)\frac{R_{ej}U_j}{D^2} \right] \\ & + \frac{1}{4}U_i^2 - \frac{R_{ej}}{D} \left(R_{ej}\ddot{R}_{ej} + 2\dot{R}_{ej}^2 \right) + \frac{R_{ej}^2(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{2D^2} \\ & \times [R_{ej}\dot{U}_j - \dot{R}_{ej}(5U_j + U_i)] - Q_{m1} - Q_{m2}, \quad (18) \end{aligned}$$

where the pressure in the liquid at each bubble surface is

$$P_i = \left(P_0 + \frac{2\sigma}{R_{0i}} \right) \left(\frac{R_{0i}}{R_{ei}} \right)^{3\kappa} - \frac{2\sigma}{R_{ei}} - p_{ac}(t), \quad (19)$$

contributions of the surface modes have been collected in Q_{m1} and Q_{m2} , and $p_{ac}(t)$ is the acoustic pressure due to an external source. With $Q_{m1} = Q_{m2} = 0$, Eq. (18) is equivalent to the models derived by Takahira *et al.*²⁰ and Harkin *et al.*¹⁸ at orders μ and μ^2 , respectively. Viscous dissipation is represented at leading order by the term $4\nu\dot{R}_{ei}/R_{ei}$ in Eq. (18).³¹ The terms of orders ε and $\varepsilon\mu^2$ are

$$\begin{aligned} Q_{m1} = & \frac{3}{10}A_{i2}U_i^2 + 6\nu U_j(\mathbf{e}_{ij} \cdot \mathbf{e}_x)\frac{A_{j2}}{D^2} + \frac{3R_{ej}}{5D^2}(\mathbf{e}_{ij} \cdot \mathbf{e}_x) \\ & \times \left[\frac{\dot{R}_{ej}}{R_{ei}}(R_{ei}U_jA_{j2} + R_{ej}U_iA_{i2}) + \frac{1}{2}R_{ej}(U_j\dot{A}_{j2} + A_{j2}\dot{U}_j) \right], \quad (20) \end{aligned}$$

and the terms of orders ε^2 and $\varepsilon^2\mu^2$ are

$$\begin{aligned} Q_{m2} = & \nu \left\{ \frac{1}{R_{ei}^2} \left(\frac{8}{15}A_{i2}\dot{A}_{i2} - \frac{4A_{i2}^2}{3R_{ei}}\dot{R}_{ei} + \frac{5}{7}A_{i3}\dot{A}_{i3} - 2\frac{\dot{R}_{ei}A_{i3}^2}{R_{ei}} - \frac{162A_{i3}A_{i2}}{35}U_i \right) \right. \\ & - \frac{(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{7D^2} \left[\frac{1191A_{j2}^2}{25}U_j + 54\frac{A_{j3}^2}{R_{ej}}U_j - \frac{144}{5}A_{j3}\dot{A}_{j2} + \frac{171}{5}\dot{A}_{j3}A_{j2} - \frac{162A_{j3}A_{j2}}{5}\dot{R}_{ej} + \frac{162R_{ej}^2A_{i3}A_{i2}}{5R_{ei}^3}\dot{R}_{ej} \right] \Big\} \\ & - \frac{1}{15}A_{i2}\ddot{A}_{i2} - \frac{1}{6}\dot{A}_{i2}^2 - \frac{1}{14}A_{i3}\ddot{A}_{i3} - \frac{1}{8}\dot{A}_{i3}^2 - \frac{1}{R_{ei}^2}(\dot{R}_{ei}^2 + 2R_{ei}\ddot{R}_{ei}) \left(\frac{A_{i2}^2}{15} + \frac{A_{i3}^2}{14} \right) \\ & - \frac{2\dot{R}_{ei}}{R_{ei}} \left(\frac{2}{15}A_{i2}\dot{A}_{i2} + \frac{1}{7}A_{i3}\dot{A}_{i3} \right) - \frac{3A_{i2}}{7R_{ei}} \left(\frac{2}{5}A_{i3}\dot{U}_i + U_i\dot{A}_{i2} \right) - \frac{3U_i^2}{70R_{ei}^2} \left(\frac{13}{5}A_{i2}^2 - \frac{17}{14}A_{i3}^2 \right) \\ & + \frac{(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{D^2} \left\{ -\frac{3A_{i2}R_{ej}^2}{7R_{ei}}\dot{R}_{ej}\dot{A}_{i3} + \frac{12A_{i2}A_{i3}}{35R_{ei}}(R_{ej}^2\ddot{R}_{ej} + 2R_{ej}\dot{R}_{ej}^2) - \frac{3}{70}R_{ej}^2(4A_{j3}\ddot{A}_{j2} + 13\dot{A}_{j2}\dot{A}_{j3} + 9A_{j2}\ddot{A}_{j3}) \right. \\ & - \frac{3}{35}R_{ej}\dot{R}_{ej}(13A_{j2}\dot{A}_{j3} + 8A_{j3}\dot{A}_{j2}) - \frac{12}{35}A_{j2}A_{j3}(R_{ej}\ddot{R}_{ej} + \dot{R}_{ej}^2) - \frac{39}{175} \left(\frac{A_{i2}^2R_{ej}^2}{R_{ei}^2}\dot{R}_{ej}U_i + R_{ej}A_{j2}^2\dot{U}_j + U_jA_{j2}^2\dot{R}_{ej} + 2R_{ej}U_jA_{j2}\dot{A}_{j2} \right) \\ & \left. + \frac{331}{490} \left(\frac{A_{i3}^2R_{ej}^2}{R_{ei}^2}\dot{R}_{ej}U_i + R_{ej}A_{j3}^2\dot{U}_j + U_jA_{j3}^2\dot{R}_{ej} + 2R_{ej}U_jA_{j3}\dot{A}_{j3} \right) \right\}. \quad (21) \end{aligned}$$

The dynamical equation for translational motion is given by Eq. (3) with $q = X_i$:

$$\begin{aligned} R_{ei}\dot{U}_i + 3U_i\dot{R}_{ei} = & -\frac{18\nu}{R_{ei}} \left[U_i + (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \frac{R_{ej}^2\dot{R}_{ej}}{D^2} \right] \\ & - \frac{3\dot{R}_{ei}R_{ej}^2\dot{R}_{ej}}{D^2} [3(\mathbf{e}_{ij} \cdot \mathbf{e}_x) + 2] \\ & - \frac{3(\mathbf{e}_{ij} \cdot \mathbf{e}_x)R_{ei}R_{ej}}{D^2} (R_{ej}\ddot{R}_{ej} + 2\dot{R}_{ej}^2) \\ & - Q_{d1} - Q_{d2}. \end{aligned} \quad (22)$$

The term proportional to ν in Eq. (22) accounts for the drag force on a sphere with a time-dependent radius. Formally this expression for the drag force is only valid for $\text{Re}_i \gg 1$ or $\text{Re}_i\mathcal{U}_i \gg 1$,^{32,33} where $\text{Re}_i = \nu R_{ei}|U_i|$ and $\mathcal{U}_i = |\dot{R}_{ei}/U_i|$ are the Reynolds number and ratio of radial to translational velocities of the bubble, respectively. However, this expression is reasonably accurate for bubbles oscillating at MHz

frequencies and we will therefore use it for all simulations considered here.³² Viscous drag was included in an identical manner in the translational equation of motion for a single bubble undergoing shape distortions in Ref. 14.

In Eq. (22) terms of orders ε and $\varepsilon\mu^2$ are collected in

$$\begin{aligned} Q_{d1} = & -\frac{36\nu A_{i2}}{R_{ei}^2} \left[U_i + (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \frac{R_{ej}^2\dot{R}_{ej}}{D^2} \right] \\ & - \frac{9}{5R_{ei}} (2\dot{R}_{ei}U_iA_{i2} + R_{ei}\dot{U}_iA_{i2} + R_{ei}U_i\dot{A}_{i2}) \\ & - \frac{(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{D^2} \left[\frac{9R_{ej}A_{i2}}{5} (R_{ej}\ddot{R}_{ej} + 2\dot{R}_{ej}^2) \right. \\ & \left. + 9R_{ej}^2\dot{R}_{ej} \left(\frac{\dot{A}_{i2}}{5} + \frac{2A_{i2}\dot{R}_{ei}}{5R_{ei}} \right) \right], \end{aligned} \quad (23)$$

and terms of orders ε^2 and $\varepsilon^2\mu^2$ are collected in

$$\begin{aligned} Q_{d2} = & \nu \left\{ \frac{9}{35R_{ei}^2} \left(114\dot{A}_{i3}A_{i2} - 96A_{i3}\dot{A}_{i2} - 108\frac{A_{i3}A_{i2}}{R_{ei}}\dot{R}_{ei} \right) + \frac{18}{7R_{ei}^3} \left(\frac{397}{25}A_{i2}^2 + 18A_{i3}^2 \right) \left[U_i + (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \frac{R_{ej}^2\dot{R}_{ej}}{D^2} \right] \right\} \\ & + \frac{234A_{i2}}{175R_{ei}^2} (\dot{R}_{ei}U_iA_{i2} + R_{ei}\dot{U}_iA_{i2} + 2R_{ei}U_i\dot{A}_{i2}) - \frac{153A_{i3}}{245R_{ei}^2} (\dot{R}_{ei}U_iA_{i3} + R_{ei}\dot{U}_iA_{i3} + 2R_{ei}U_i\dot{A}_{i3}) \\ & + \frac{9}{35} (3A_{i2}\ddot{A}_{i3} - 2A_{i3}\ddot{A}_{i2} + \dot{A}_{i2}\dot{A}_{i3}) + \frac{18\dot{R}_{ei}}{35R_{ei}} (A_{i2}\dot{A}_{i3} - 4A_{i3}\dot{A}_{i2}) - \frac{36A_{i2}A_{i3}}{35R_{ei}^2} (R_{ei}\ddot{R}_{ei} + \dot{R}_{ei}^2) \\ & + \frac{3(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{35D^2} \left[\frac{78A_{i2}^2}{5R_{ei}} (R_{ej}^2\ddot{R}_{ej} + 2R_{ej}\dot{R}_{ej}^2) + \frac{78R_{ej}^2\dot{R}_{ej}}{5R_{ei}} \left(\frac{A_{i2}^2\dot{R}_{ei}}{R_{ei}} + 2A_{i2}\dot{A}_{i2} \right) - \frac{331A_{i3}^2}{7R_{ei}} (R_{ej}^2\ddot{R}_{ej} + 2R_{ej}\dot{R}_{ej}^2) \right. \\ & \left. - \frac{331R_{ej}^2\dot{R}_{ej}}{7R_{ei}} \left(\frac{A_{i3}^2\dot{R}_{ei}}{R_{ei}} + 2A_{i3}\dot{A}_{i3} \right) \right]. \end{aligned} \quad (24)$$

Equation (23) shows that, at order ε , the translational motion of the bubble is independent of the octupole mode. In the absence of viscosity, bubble interaction, and surface distortions, i.e., for uncoupled spherical bubbles in free pulsation and translation, the right-hand side of Eq. (22) vanishes.

Setting $q = A_{i2}$ in Eq. (3) yields the dynamical equation for the quadrupole mode which, once arranged in the canonical form for a harmonic oscillator, is

$$\ddot{A}_{i2} + \frac{1}{R_{ei}} \left[3\dot{R}_{ei} + \frac{8\nu}{R_{ei}} \left(5 - 16\frac{\delta_{i2}}{R_{ei}} \right) \right] \dot{A}_{i2} + \Omega_{i2}^2 A_{i2} = F_{i2} + F_{i2\varepsilon}, \quad (25)$$

where δ_{i2} is a boundary layer thickness associated with vorticity, the square root of the quantity

$$\begin{aligned} \Omega_{i2}^2 = & -\frac{1}{R_{ei}} \left[\ddot{R}_{ei} + \frac{117}{35} \frac{U_i}{R_{ei}} \left(U_i + \frac{2R_{ej}^2\dot{R}_{ej}(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{D^2} \right) \right. \\ & \left. - \frac{12\sigma}{R_{ei}^2\rho} - \frac{8\nu}{R_{ei}^2} \left(1 + \frac{4\delta_{i2}}{R_{ei}} \right) \dot{R}_{ei} \right] \end{aligned} \quad (26)$$

when evaluated at equilibrium is the natural angular frequency of the quadrupole mode in the linear approximation, and the forcing functions are

$$F_{i2} = -\frac{9U_i}{4R_{ei}} \left(U_i + \frac{2R_{ej}^2\dot{R}_{ej}(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{D^2} \right) \quad (27)$$

and

$$\begin{aligned} F_{i2\varepsilon} = & \frac{9}{7R_{ei}} \left\{ U_i \left(\frac{5}{2}\dot{A}_{i3} + \frac{48\nu}{R_{ei}^2}A_{i3} \right) + A_{i3}\dot{U}_i \right. \\ & + \frac{R_{ej}(\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{D^2} \left[\frac{5}{2}R_{ej}\dot{R}_{ej}\dot{A}_{i3} \right. \\ & \left. \left. - A_{i3} \left(R_{ej}\ddot{R}_{ej} + 2\dot{R}_{ej}^2 + 2\dot{R}_{ei}R_{ej}\frac{R_{ej}}{R_{ei}} - 48\nu\dot{R}_{ej}\frac{R_{ej}}{R_{ei}^3} \right) \right] \right\}. \end{aligned} \quad (28)$$

At leading order Eq. (27) shows that the quadrupole mode is excited by translational motion. Terms accounting for contributions of the quadrupole and octupole modes to excitation

of the quadrupole mode are of order ε and are collected in $F_{i2\varepsilon}$. There are no contributions at order ε^2 . If terms of order μ^2 are neglected, Eq. (25) coincides with the equation derived by Plesset for surface modes in the case of a single stationary bubble.⁴ Equation (25) shows that the quadrupole mode depends on the radial, dipole, and octupole modes. Previous work^{19,20} has shown that if order ε^2 terms are discarded, inhomogeneous terms in the dynamical equation for the n th mode ($n \geq 2$) are influenced only by lower order modes. Therefore, terms containing the octupole amplitude in Eq. (28) result from the nonlinear boundary condition, Eqs. (A2) and (A3).

Setting $q = A_{i3}$ in Eq. (3) gives the equation for the octupole mode:

$$\ddot{A}_{i3} + \frac{1}{R_{ei}} \left[3\dot{R}_{ei} + \frac{10\nu}{R_{ei}} \left(7 - 30 \frac{\delta_{i3}}{R_{ei}} \right) \right] \dot{A}_{i3} + \Omega_{i3}^2 A_{i3} = F_{i3\varepsilon}, \quad (29)$$

where δ_{i3} is a boundary layer thickness associated with vorticity, the coefficient of A_{i3} is

$$\Omega_{i3}^2 = \frac{2}{R_{ei}} \left\{ \frac{20\sigma}{R_{ei}^2 \rho} - \ddot{R}_{ei} + \frac{U_i}{35R_{ei}} \left[51U_i + 211 \frac{R_{ej}^2}{D^2} \dot{R}_{ej} (\mathbf{e}_{ij} \cdot \mathbf{e}_x) \right] + 10\nu \frac{\dot{R}_{ei}}{R_{ei}^2} \left(6 \frac{\delta_{i3}}{R_{ei}} + 1 \right) \right\} \quad (30)$$

and the forcing function is

$$F_{i3\varepsilon} = -\frac{6}{R_{ei}} \left\{ U_i \left(\dot{A}_{i2} + 2\dot{R}_{ei} \frac{A_{i2}}{R_{ei}} + \frac{114\nu}{5R_{ei}^2} A_{i2} \right) + \frac{3}{5} \dot{U}_i A_{i2} + 27 \frac{R_{ej} (\mathbf{e}_{ij} \cdot \mathbf{e}_x)}{5D^2} \left[\frac{5}{54} \dot{A}_{i2} R_{ej} \dot{R}_{ej} + A_{i2} (R_{ej} \ddot{R}_{ej} + 2\dot{R}_{ej}^2 + 2 \frac{R_{ej}}{R_{ei}} \dot{R}_{ei} \dot{R}_{ej} + \nu \frac{38 R_{ej}}{9 R_{ei}^3} \dot{R}_{ej}) \right] \right\}. \quad (31)$$

Unlike the dynamical equation for the quadrupole mode, the right-hand side of Eq. (29) does not contain any terms independent of the quadrupole or octupole modes. In other words, if the octupole mode is initially zero it can only be excited after excitation of the quadrupole mode. Plesset's result⁴ is recovered from Eq. (29) if terms of order μ^2 are neglected. Lamb's expression³⁴ for the square of the natural frequency of the n th mode for $n \neq 0$,

$$\omega_n^2 = (n-1)(n+1)(n+2) \frac{\sigma}{\rho R_{ei}^3}, \quad (32)$$

is also recovered from Ω_n^2 in Eqs. (26) and (30) after evaluating those expressions at equilibrium. Previous research^{19,20} has shown that terms accounting for bubble interaction are of order μ^n in the dynamical equation for the n th mode [see Eqs. (A10) and (A11)]. In the present model, however, terms of order μ^2 appear in Eqs. (30) and (31) as a result of the nonlinear boundary condition.

Note that, similar to the forcing function F_{i2} given by Eq. (27), terms of leading order in Eq. (31) are proportional

to the translational velocity or acceleration. Doinikov¹⁴ noted that for a single bubble translational or octupole (i.e., odd-order mode) motion is able to generate all other modes, even if they are initially zero. Even-order modes, however, are only capable of generating other even order modes. These observations also apply to the current two-bubble model. In addition, Eqs. (27) and (28) show that quadrupole motion may be induced by the volume oscillations of a neighboring bubble interacting with either the dipole or octupole mode. Similarly Eq. (31) shows that the octupole mode may be generated by the volume oscillation of a neighboring bubble interacting with the quadrupole mode.

Following Ref. 5, we approximate the viscous damping due to vorticity through a boundary layer approximation. In Eqs. (25) and (29) parameters δ_{i2} and δ_{i3} represent the thicknesses of the boundary layers surrounding bubble i which correspond to the quadrupole and octupole modes, respectively. For large bubbles the boundary layer thickness is approximated by the diffusion length $\sqrt{\nu/\omega}$, whereas for small bubbles we introduce a cutoff thickness $R_{0i}/2n$ associated with surface mode n . Thus the boundary layer thickness is⁵

$$\delta_{in} = \min \left(\sqrt{\frac{\nu}{\omega}}, \frac{R_{0i}}{2n} \right). \quad (33)$$

Terms accounting for vorticity in the present model are obtained by applying Eq. (13) to the terms containing the boundary layers in Eqs. (12) and (13) of Ref. 5, and performing a binomial expansion in the surface mode amplitudes.

III. SIMULATIONS

In this section numerical simulations obtained by integrating Eqs. (18), (22), (25), and (29) using a standard backward differentiation routine³⁵ are presented. The equilibrium radii of the bubbles were assumed to be on the order of a few microns. We assume the liquid to be water with density $\rho = 1000 \text{ kg/m}^3$, surface tension $\sigma = 0.073 \text{ N/m}$, and kinematic viscosity $\nu = 1 \times 10^{-6} \text{ m}^2/\text{s}$. The gas contained in the bubbles is assumed to be air with polytropic exponent $\kappa = 1.4$. Examples of free and forced dynamics of the system are presented below. In all cases the amplitudes of the quadrupole and octupole modes are less than 15% of the equilibrium radii of the bubbles.

A. Free response (in-phase)

The free response of two identical bubbles given identical initial conditions (leading to in-phase oscillations) is illustrated in Fig. 2 and movie 1.³⁶ In addition, still frames from movie 1 (Ref. 36) are shown in Fig. 3. The initial conditions are $R_{0i} = 7 \mu\text{m}$, $R_i(0)/R_{0i} = 1.5$, $D(0)/R_{0i} = 10$, with both bubbles at rest. Amplitudes of the monopole ($n=0$), quadrupole ($n=2$), and octupole ($n=3$) modes for bubbles 1 and 2 are shown in Figs. 2(a) and 2(b), respectively. In Fig. 3 and movie 1 (Ref. 36) bubble 1 is shown on the left, and bubble 2 on the right. The amplitudes of the monopole and quadrupole modes are represented by the solid and dashed lines, respectively. Time is normalized by

FIG. 2. Time evolution of the normalized multipole amplitudes [parts (a) and (b)] and separation distance [part (c)] for in-phase free response. Both bubbles are initially at rest with $R_{0i} = 7 \mu\text{m}$, $R_i(0)/R_{0i} = 1.5$, and initial separation distance $D(0)/R_{0i} = 10$.

the natural period of the monopole mode $T_0 = 2\pi/\omega_0$, where³¹

$$\omega_0^2 = \frac{1}{\rho R_{0i}^2} \left[3\kappa \left(P_0 + \frac{2\sigma}{R_{0i}} \right) - \frac{2\sigma}{R_{0i}} - \frac{4\rho\nu^2}{R_{0i}^2} \right]. \quad (34)$$

Figures 2(a), 2(b) show that the quadrupole and octupole amplitudes tend to be largest during bubble collapse. Figure 3 shows the initial conditions of the system at $t/T_0 = 0$ and at points of bubble collapse occurring at $t/T_0 = 4.1$ and 7.5. As discussed in Sec. II D, the dynamical equations possess a sequential structure. Namely, higher order modes are principally excited through the interaction of lower order modes. Note that while the monopole and quadrupole amplitudes of the bubbles are in phase, the fact that the octupole amplitudes A_{13} (left bubble) and A_{23} (right bubble) are oscillating in anti-phase indicates that the jets starting to form on the bubbles are directed toward one another. These opposing jets can be seen in Fig. 3 at $t/T_0 = 7.5$. Figure 2(c) illustrates

FIG. 3. (Color online) Still frames from movie 1 (Ref. 36). Both bubbles are initially at rest with $R_{0i} = 7 \mu\text{m}$, $R_i(0)/R_{0i} = 1.5$, and initial separation distance $D(0)/R_{0i} = 10$.

the time evolution of the separation distance D . As the bubbles move closer together their translational velocities increase. The translational (secondary Bjerknes) force acting between the bubbles is attractive as a result of their in-phase pulsation.^{18,37} The monopole mode decays with time due to viscous damping, while the quadrupole and octupole modes increase as the bubbles move closer together. The dynamics is qualitatively consistent with Fig. 2 of Pishchalnikov *et al.*²

B. Free response (antiphase)

The free response of two identical bubbles given non-identical initial conditions is illustrated in Fig. 4 and in movie 2.³⁶ The initial conditions of the bubbles are $R_{0i} = 6.5 \mu\text{m}$ with $R_1(0)/R_{01} = 1.5$ and $R_2(0)/R_{02} = 0.65$, respectively, and $D(0)/R_{0i} = 5$. In Figs. 4(a) and 4(b), solid, dashed, and dotted lines denote the amplitudes of the monopole, quadrupole, and octupole modes, respectively, with Fig. 4(a) corresponding to bubble 1 and Fig. 4(b) corresponding to bubble 2. As illustrated in Fig. 4(c), antiphase

FIG. 4. Time evolution of the normalized multipole amplitudes [parts (a) and (b)] and separation distance [part (c)] for the antiphase free response. Both bubbles are initially at rest with $R_{0i} = 6.5 \mu\text{m}$, $D(0)/R_{0i} = 5$, $R_1(0)/R_{01} = 1.5$, and $R_2(0)/R_{02} = 0.65$.

oscillations generate a repulsive translational force between the bubbles. Both the translation velocity and the surface mode amplitudes decrease with time as the bubbles move farther apart. In addition, the bubbles form jets pointing away from their mutual center.

C. Free response (nonidentical bubbles)

The free response of two nonidentical bubbles is illustrated in Fig. 5 and movie 3.³⁶ The initial conditions are $R_{01} = 4.5 \mu\text{m}$, $R_{02} = 8.5 \mu\text{m}$, $R_1(0)/R_{01} = 1.5$, $R_2(0)/R_{02} = 1.5$, $D(0)/R_{01} = 10$, with both bubbles at rest. Amplitudes of the monopole ($n=0$), quadrupole ($n=2$), and octupole ($n=3$) modes for bubble 1 and 2 are shown in Figs. 5(a) and 5(b), respectively, where bubble 1 is taken to be on the left and bubble 2 on the right. Time is normalized by the natural period of bubble 1. The amplitudes of the monopole and quadrupole modes are represented by the solid and dashed lines, respectively. Figure 5(c) shows the separation distance

FIG. 5. Time evolution of the normalized multipole amplitudes [parts (a) and (b)] and separation distance [part (c)] for the anti-phase free response. Both bubbles are initially at rest with $R_{01} = 4.5 \mu\text{m}$, $R_{02} = 8.6 \mu\text{m}$, $D(0)/R_{01} = 10$, $R_1(0)/R_{01} = 1.5$, and $R_2(0)/R_{02} = 1.5$.

between the bubbles. Examination of Eq. (22) reveals that to leading order the translational force acting on the bubbles is determined by the instantaneous phase difference between the volume oscillation amplitudes. In-phase pulsation results in an attractive force while antiphase pulsation results in a repulsive force. In this case the natural frequencies of the bubbles are different and therefore the translational force oscillates between being attractive and repulsive [Fig. 5(c)].

D. Forced response (identical bubbles)

Next we consider the dynamics of two identical bubbles driven by a sinusoidal external acoustic source, i.e.,

$$p_{ac}(t) = p_0 \sin \omega t, \quad (35)$$

where p_0 and $\omega = 2\pi f$ are the amplitude and angular frequency, respectively, of the acoustic pressure perturbation.

FIG. 6. Time evolution of the normalized multipole amplitudes [parts (a) and (b)] and separation distance [part (c)]. The system is driven by a sinusoidal acoustic pressure with amplitude 20 kPa and frequency 0.623 MHz. The bubbles are initially at rest and equilibrium with $R_{0i} = 5 \mu\text{m}$ and $D(0)/R_{0i} = 10$.

In the following simulations time is normalized by the period of the driving frequency, i.e., $T_{ac} = 1/f$.

Figure 6 and movie 4 (Ref. 36) illustrate the dynamics of two identical bubbles [$R_{0i} = 5 \mu\text{m}$ and $D(0)/R_{0i} = 10$] which are both initially at rest and at equilibrium. The bubbles are driven by a sinusoidal acoustic pressure with amplitude 20 kPa and frequency $f = 0.623$ MHz. This frequency is below the natural frequency (0.72 MHz) of the bubbles but, due to the high amplitude of the external source, the bubbles respond more strongly at this drive frequency than at their natural frequency.³⁸ This particular drive frequency was chosen so that visible surface oscillations are obtained.

Initially only the monopole mode is excited but, after approximately the third rebound, the quadrupole and octupole modes are excited via interaction between the bubbles. The resulting formation of opposing jets is illustrated in movie 4.³⁶ An attractive translational force, due to the in-phase pulsation, is also observed in Fig. 6(c).

FIG. 7. Time evolution of the normalized multipole amplitudes [parts (a) and (b)] and separation distance [part (c)]. The system is driven by a sinusoidal acoustic pressure with amplitude 70 kPa and frequency 0.65 MHz. The bubbles are initially at rest and equilibrium, with $R_{01} = 5 \mu\text{m}$, $R_{02} = 4 \mu\text{m}$, and $D(0)/R_{01} = 7$.

E. Forced response (nonidentical bubbles)

Figure 7 and movie 5 (Ref. 36) illustrate the dynamics of two nonidentical bubbles with equilibrium radii $R_{01} = 5 \mu\text{m}$ and $R_{02} = 4 \mu\text{m}$, driven by an external acoustic source of the form given by Eq. (35) with $p_0 = 70$ kPa and $f = 0.65$ MHz. The bubbles are initially at rest and at equilibrium, and separated by distance $D(0)/R_{01} = 7$. Similar to the situation in Sec. III C, the natural frequencies of the bubbles are different and the dynamics is more complicated. Comparison of the monopole amplitudes illustrated in parts (a) and (b) of Fig. 7 show that for the first two cycles the bubbles pulsate approximately in-phase and, according to Eq. (22), the bubbles move closer together during this time period [Fig. 7(c)]. Thereafter however, the pulsation of the second bubble is out of phase with that of the first and the bubbles begin to move apart. Although the amplitude of the excitation pressure is larger here than in the previous case, the amplitudes

of the surface modes are smaller due to the larger average separation distance.

F. Stability analysis

Under sufficiently strong external acoustic excitation the surface modes may grow without bound.³⁹ In certain cases the surface of the bubble, as given by the model, may form a multiply-connected region, and thus the solution loses physical meaning. In this section we explore surface mode growth of two interacting bubbles due to the parametric instability, which causes a gradual increase in the surface mode amplitudes for weakly nonlinear dynamics. Our parameters are identical to those used by Brenner to analyze the stability of a single isolated bubble.³⁹

The system is excited by a sinusoidal pressure source of variable amplitude and fixed frequency $f=26.5$ kHz. As done in Ref. 40 the amplitude of each surface mode during one acoustic period is calculated as

$$B_{in}(t) = \int_t^{t+T_{ac}} |A_{in}(\tau)| d\tau, \quad (36)$$

where $T_{ac} = 37.7 \mu\text{s}$ is the period of the applied acoustic field. Two parameters, the pressure source amplitude and the equilibrium radii of the bubbles, are varied. The amplitude of the applied acoustic pressure is varied between 0.8 and 1.2 atm, and the equilibrium radii of the bubbles are varied between 3 and 7 μm . Therefore, the natural frequency of the monopole mode [see Eq. (34)] is on the order of 1 MHz, which is much larger than the driving frequency. For parametrically unstable oscillations, the amplitude of the surface mode grows with time, and thus

$$B_{in}(T_0) < B_{in}(t) \quad \text{for} \quad T_0 < t. \quad (37)$$

In order to evaluate the stability of the system for each parameter set (combination of pressure amplitude and bubble radius), we integrate the dynamical equations numerically for $0 \leq t \leq 3T_{ac}$ and then compare the values of $B_{i2}(t)$ and $B_{i3}(t)$ via Eq. (36) at $t = 2T_{ac}$ and $t = 3T_{ac}$. In each case the bubbles are initially at rest and at equilibrium. If $B_{in}(3T_{ac})$ is greater than $B_{in}(2T_{ac})$ then the parameter set is declared unstable.

Diagrams of the dependence of the shape stability on equilibrium radius and pressure amplitude of the external excitation for large $[D(0)/R_{0i} = 100]$ and small $[D(0)/R_{0i} = 20]$ separation distances are shown in Figs. 8(a) and 8(b), respectively. Parameters in the black region result in stable surface modes, and those in the white region result in unstable modes. Since the amplitudes of the quadrupole and octupole modes are initially zero, surface oscillations are generated solely by bubble-bubble interactions. For $D(0)/R_{0i} = 100$, Fig. 8(a) shows that the bubble interaction is weak and the results agree with the case of a single isolated bubble. The surface modes are unstable if either the excitation amplitude or equilibrium radius is large.

For a smaller separation distance of $D(0)/R_{0i} = 20$, Fig. 8(b) shows that the stronger bubble-bubble interaction enlarges the parameter space for unstable surface modes.

FIG. 8. Dependence of the shape stability for two interacting bubbles on initial equilibrium radius and excitation pressure amplitude. The initial separation distance is either $D(0)/R_{0i} = 100$ [part (a)] or $D(0)/R_{0i} = 20$ [part (b)]. The frequency of acoustic excitation is $f = 26.5$ kHz. Black and white regions correspond to stable and unstable dynamics, respectively. In the gray region, bubble collision occurs before the completion of three acoustic cycles.

Moreover, in some cases bubble collision occurs during the first three driving cycles, thus preventing evaluation of Eq. (36). The parameter space for which bubble collision occurs is indicated by the gray region in Fig. 8(b).

Bubble collision will eventually occur for any initial separation distance, even if both the equilibrium radius and excitation amplitude are in the stable region indicated in Fig. 8. This is due to the in-phase pulsation of the bubbles, which results in an attractive translational (secondary Bjerknes) force. Figure 9 illustrates the dependence of the collision time on the bubble's equilibrium radius. The collision time decreases with increasing bubble radius, and large excitation amplitudes also shorten the time to collision.

IV. SUMMARY

Dynamical equations describing the motion of two translating aspherical bubbles in a viscous, incompressible

FIG. 9. Dependence of collision time on equilibrium radius of the bubbles, and acoustic pressure amplitude. The bubbles are initially separated by a distance $D(0)/R_{0i} = 20$. Solid, dashed, and dotted lines correspond to pressure amplitudes of 0.9, 1.0, and 1.1 atm, respectively.

fluid are derived using Lagrangian mechanics. Quadrupole and octupole spherical harmonics are included to model deformation of the nominally spherical bubble surfaces. Numerical integration of the model equations shows that nonlinear bubble interaction plays an important role in the initiation and growth of the surface modes. As has been observed in experiments, simulations performed with the model equations show that bubble-bubble interaction is capable of inducing liquid jets which point either toward or away from the mutual center of the bubbles. The stability of the surface modes is also investigated numerically. If the separation distance between the bubbles is sufficiently large the results are consistent with the stability diagrams for a single bubble. However, if the bubbles are sufficiently close, their stronger interaction leads to an enlargement of the parameter space corresponding to unstable surface modes.

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APPENDIX: DERIVATION OF THE VELOCITY POTENTIAL

In this section the derivation of the velocity potential in a liquid containing two aspherical bubbles is outlined. The velocity potential in a liquid containing a single bubble (e.g., bubble 1) may be expressed as

$$\phi = \sum_{n=0}^3 \frac{a_n R_1^{(n+1)}}{r_1^{(n+1)}} P_n(\cos \theta_1). \quad (\text{A1})$$

The boundary condition on the bubble surface is

$$\nabla_1 \phi(r_1, \theta_1) \cdot \mathbf{N}_1 = \mathbf{v}_{s1} \cdot \mathbf{N}_1, \quad (\text{A2})$$

evaluated at $\mathbf{r}_1 = \mathbf{r}_{s1}$, where ∇_1 is the gradient operator in the local coordinate system of bubble 1, $\mathbf{v}_{s1}$ is the velocity at the bubble surface, and $\mathbf{N}_1$ is a vector normal to the bubble surface. For the present case r_{si} is independent of the azimuthal angle φ_i . The normal vector is therefore

$$\mathbf{N}_1 = \frac{\partial \mathbf{r}_{s1}}{\partial \theta_1} \times \frac{\partial \mathbf{r}_{s1}}{\partial \varphi_1} = r_{s1}^2 \sin \theta_1 \mathbf{e}_{r1} - r_{s1} \sin \theta_1 \frac{\partial r_{s1}}{\partial \theta_1} \mathbf{e}_{\theta 1} \quad (\text{A3})$$

and the bubble surface velocity is

$$\begin{aligned} \mathbf{v}_{s1} &= \dot{\mathbf{r}}_{s1} + U_1 P_1(\cos \theta_1) \mathbf{e}_{r1} \\ &= [\dot{R}_1 P_0(\cos \theta_1) + U_1 P_1(\cos \theta_1) + \dot{A}_{13} P_2(\cos \theta_1) \\ &\quad + \dot{A}_{13} P_3(\cos \theta_1)] \mathbf{e}_{r1}. \end{aligned} \quad (\text{A4})$$

Evaluation of Eq. (A2) on the bubble surface yields expressions for the coefficients a_n in Eq. (A1):

$$a_0 = -R_1 \dot{R}_1 - \frac{A_{12}}{5} \left(2\dot{A}_{12} + \dot{R}_1 \frac{A_{12}}{R_1} \right) - \frac{A_{13}}{7} \left(2\dot{A}_{13} + \dot{R}_1 \frac{A_{13}}{R_1} \right), \quad (\text{A5})$$

$$\begin{aligned} a_1 &= -\frac{1}{2} R_1 U_1 + \frac{3}{10} A_{12} \left(U_1 - \frac{9}{7} \dot{A}_{13} \right) \\ &\quad + \frac{U_1}{35 R_1} \left(\frac{173}{7} A_{13}^2 - \frac{183}{10} A_{12}^2 \right) \\ &\quad - \frac{6}{35} A_{13} \left(\dot{A}_{12} + 2\dot{R}_1 \frac{A_{12}}{R_1} \right), \end{aligned} \quad (\text{A6})$$

$$\begin{aligned} a_2 &= -\frac{R_1}{3} \dot{A}_{12} - 2A_{12} \left(\frac{\dot{R}_1}{3} + \frac{\dot{A}_{12}}{7} \right) + \frac{A_{13}}{7} \left(3U_1 - \frac{11}{9} \dot{A}_{13} \right) \\ &\quad - \frac{2\dot{R}_1}{7R_1} \left(A_{12}^2 + \frac{5}{9} A_{13}^2 \right) + \frac{167}{245 R_1} U_1 A_{12} A_{13}, \end{aligned} \quad (\text{A7})$$

$$\begin{aligned} a_3 &= -\frac{R_1}{4} \dot{A}_{13} - A_{13} \left(\frac{\dot{R}_1}{2} + \frac{\dot{A}_{12}}{5} \right) - \frac{A_{12}}{10} \left(9U_1 + \frac{17}{6} \dot{A}_{13} \right) \\ &\quad - \frac{3U_1}{5R_1} \left(\frac{3}{5} A_{12}^2 - \frac{61}{77} A_{13}^2 \right) - \frac{17}{30} \frac{A_{12} A_{13} \dot{R}_1}{R_1}. \end{aligned} \quad (\text{A8})$$

The velocity potential in a liquid containing two aspherical bubbles is

$$\phi = \sum_{n=0}^3 \frac{a_{1n} R_1^{(n+1)}}{r_1^{(n+1)}} P_n(\cos \theta_1) + \sum_{n=0}^3 \frac{a_{2n} R_2^{(n+1)}}{r_2^{(n+1)}} P_n(\cos \theta_2), \quad (\text{A9})$$

where the boundary condition and vector normal to the surface of each bubble are given by Eqs. (A2) and (A3) with subscript 1 replaced by i , for $i = 1, 2$. The procedure outlined

in the Appendix of Ref. 41 may now be used to derive the velocity potential coefficients a_{in} for the two bubble geometry, given Eqs. (A5)–(A8). In order to evaluate Eq. (A2) the velocity potential given by Eq. (A9) must be expressed in the local spherical coordinate system of either bubble, (r_1, θ_1) or (r_2, θ_2) , using

$$\frac{P_n(\cos \theta_2)}{r_2^{(n+1)}} = \frac{1}{n!D^{n+1}} \sum_{m=0}^3 \frac{(-1)^m (m+n)!}{m!} \left(\frac{r_1}{D}\right)^m P_m(\cos \theta_1), \quad (\text{A10})$$

$$\frac{P_n(\cos \theta_1)}{r_1^{(n+1)}} = \frac{(-1)^n}{n!D^{n+1}} \sum_{m=0}^3 \frac{(m+n)!}{m!} \left(\frac{r_2}{D}\right)^m P_m(\cos \theta_2). \quad (\text{A11})$$

Note that at order μ^2 Eqs. (A10) and (A11) are zero for the quadrupole ($n=2$) and octupole ($n=3$) modes. The resulting expressions for the velocity potential coefficients are

$$a_{10} = -R_1 \dot{R}_1 - \frac{A_{12}^2 \dot{R}_1}{5R_1} - \frac{A_{13}^2 \dot{R}_1}{7R_1} - \frac{2A_{12} \dot{A}_{12}}{5} - \frac{2A_{13} \dot{A}_{13}}{7}, \quad (\text{A12})$$

$$\begin{aligned} a_{11} = & -\frac{R_1 U_1}{2} + \frac{3A_{12} U_1}{10} - \frac{183A_{12}^2 U_1}{350R_1} + \frac{173A_{13}^2 U_1}{245R_1} \\ & - \frac{12A_{13} A_{12} \dot{R}_1}{35R_1} - \frac{6A_{13} \dot{A}_{12}}{35} - \frac{27A_{12} \dot{A}_{13}}{70} \\ & - \frac{R_1 R_2^2 \dot{R}_2}{2D^2} - \frac{183A_{12}^2 R_2^2 \dot{R}_2}{350R_1 D^2} + \frac{173A_{13}^2 R_2^2 \dot{R}_2}{245R_1 D^2} \\ & + \frac{3A_{12} R_2^2 \dot{R}_2}{10D^2} - \frac{A_{22}^2 R_1 \dot{R}_2}{10D^2} - \frac{A_{23}^2 R_1 \dot{R}_2}{14D^2} \\ & - \frac{A_{22} R_1 R_2 \dot{A}_{22}}{5D^2} - \frac{A_{23} R_1 R_2 \dot{A}_{23}}{7D^2}, \end{aligned} \quad (\text{A13})$$

$$\begin{aligned} a_{12} = & -\frac{R_1 \dot{A}_{12}}{3} - \frac{2A_{12} \dot{R}_1}{3} + \frac{3A_{13} U_1}{7} - \frac{2A_{12} \dot{A}_{12}}{7} \\ & - \frac{2A_{12}^2 \dot{R}_1}{7R_1} - \frac{11A_{13} \dot{A}_{13}}{63} - \frac{10A_{13}^2 \dot{R}_1}{63R_1} + \frac{167A_{13} A_{12} U_1}{245R_1} \\ & + \frac{3A_{13} R_2^2 \dot{R}_2}{7D^2} + \frac{167A_{13} A_{12} R_2^2 \dot{R}_2}{245R_1 D^2}, \end{aligned} \quad (\text{A14})$$

$$\begin{aligned} a_{13} = & -\frac{R_1 \dot{A}_{13}}{4} - \frac{A_{13} \dot{R}_1}{2} - \frac{9A_{12} U_1}{10} - \frac{A_{13} \dot{A}_{12}}{5} - \frac{17A_{12} \dot{A}_{13}}{60} \\ & - \frac{17A_{13} A_{12} \dot{R}_1}{30R_1} - \frac{9A_{12}^2 U_1}{25R_1} + \frac{183A_{13}^2 U_1}{385R_1} - \frac{9A_{12} R_2^2 \dot{R}_2}{10D^2} \\ & - \frac{9A_{12}^2 R_2^2 \dot{R}_2}{25R_1 D^2} + \frac{183A_{13}^2 R_2^2 \dot{R}_2}{385R_1 D^2}. \end{aligned} \quad (\text{A15})$$

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