

Nonlinear mode coupling and excitation in nonaxisymmetric droplet shape oscillations

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Nonaxisymmetric droplet shape oscillations are investigated by extending the numerical framework developed in Akbari *et al.* [*Phys. Rev. Fluids* **11**, 033602 (2026)], which is based on a Galerkin method using spherical harmonics, to fully three-dimensional configurations. The study focuses on the role of shape symmetry and mode excitation of inviscid droplet shape oscillation. The numerical results confirm that the oscillation frequency is independent of the azimuthal wave number, in agreement with classical theoretical predictions. The discrete geometric symmetries of both the initial droplet deformation and the initial velocity potential are demonstrated to be preserved throughout the oscillation. Consequently, only modes compatible with these symmetries are excited. When the initial velocity potential breaks the symmetries of the droplet deformation, leading to a reduction in these symmetries, additional modes are excited compared to those for a zero initial velocity. A similar result is observed in the droplet shape oscillations of a single initial deformation mode and a superposition of two initial deformation modes. In the former case, only modes that satisfy the symmetries of the initial deformation are excited. In the latter case, the superposition reduces the overall symmetry of the droplet shape, leading to additional mode excitation through symmetry breaking. These results highlight the fundamental role of symmetry preservation in nonlinear droplet oscillations. By exploiting these symmetry properties, the computational effort could be reduced without compromising numerical accuracy, as modes that do not occur can be excluded from the solution approach prior to the simulation.

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I. INTRODUCTION

The scientific investigation of droplet shape oscillations and dynamics goes back more than one and a half centuries, which reflects the high relevance in science and nature. Droplet shape oscillations play an essential role in cloud microphysics and can also provide valuable insights for remotely estimating rainfall rates [1] and improving the accuracy of radar-based precipitation measurements [2,3]. Droplet shape oscillations are also relevant in mass transfer operations in chemical engineering [4], as the change in droplet surface area has a direct influence on the transport processes of mass, momentum, and energy across the droplet [5].

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Most studies in the literature have focused on axisymmetric droplet shape oscillations. One of the first studies was conducted by Lord Rayleigh [6], who analyzed small-amplitude oscillations of an inviscid droplet in a vacuum around its spherical equilibrium shape. He treated surface tension as a restoring force and derived the angular frequency of inviscid droplet shape oscillations

$$\omega_\ell^R = \sqrt{\frac{\sigma \ell(\ell+2)(\ell-1)}{\varrho R_0^3}}, \quad (1)$$

which is one of his fundamental results. Here σ is the surface tension, ϱ the droplet density, R_0 the undeformed droplet radius, and ℓ is the degree of initial deformation mode, respectively. Lamb [7,8] extended Rayleigh's work to viscous droplets in another fluid. This confirmed the Rayleigh frequency in the limit of negligible viscosity and demonstrated the exponential damping of the oscillation amplitude,

$$\eta_A(t) \sim \exp(-\lambda t), \quad (2)$$

where $\lambda = (\nu/R_0^2)(\ell-1)(2\ell+1)$ is the damping parameter and ν is the kinematic viscosity of the droplet. A more fundamental theoretical treatment was first developed by Chandrasekhar [9]. By applying a normal-mode analysis to small-amplitude shape oscillations of a liquid globe with self-gravitation forces, while neglecting surface tension, he derived the characteristic governing equations and solved the linearized Navier-Stokes equations. He showed that, when the relevant parameter $\omega_\ell^R R_0^2/\nu$ is large, damped oscillations occur, whereas, for smaller values, aperiodic decay is observed.

For low viscosity, Reid [10] treated surface tension rather than self-gravitation as the restoring force and obtained the same damping rate as Lamb [8] and Chandrasekhar [9]. Subsequently, Miller and Scriven [11] incorporated the viscous and inertial effects of a surrounding viscous medium on an oscillating droplet and investigated several interfacial conditions. In the limiting case of a viscous liquid droplet oscillating in a vacuum or in a gas with negligible density and viscosity, their predictions aligned well with those of Reid [10].

A more detailed analysis was carried out by Prosperetti [12], who investigated the small-amplitude, linearized shape oscillations of free droplets, gas bubbles, and droplets immersed in a viscous medium. The study showed that the motion takes the form of modulated damped oscillations, with both the damping parameter and the frequency approaching the corresponding normal-mode results only asymptotically. For small viscosity ν , the angular frequency $\bar{\omega}_\ell$ was expressed as

$$\bar{\omega}_\ell = \sqrt{(\omega_\ell^R)^2 - \lambda^2}, \quad (3)$$

with the damping parameter λ . Natarajan and Brown [13,14] examined internal resonances among different deformation modes in nonlinear shape oscillations of inviscid droplets. They demonstrated that, in the presence of surface tension, such oscillations can exhibit complex dynamics, including instabilities and stochastic motion, due to cubic and quadratic resonances associated with interactions between oscillatory modes.

One of the first numerical simulations was performed by Foote [15]. He developed a modified marker-and-cell computational method and provided consistent results with wind tunnel observations of oscillating droplets reported by Montgomery [16]. In contrast to linear-theory predictions, these studies showed that the fundamental resonant frequency decreases as the initial deformation amplitude increases. Foote [15] also found that the droplet spends more time in the prolate, elongated state than in the oblate state, which was attributed to the asymmetry of the restoring force during the prolate and oblate phases. These nonlinear effects were later further confirmed experimentally by Trinh and Wang [17], who used acoustic radiation pressure to trap a droplet in a stable position, deform its equilibrium shape, and induce shape oscillations. Their experiments confirmed the decrease in oscillation frequency with increasing deformation amplitude and also

revealed an asymmetry between the prolate and oblate phases for the two-lobed initial deformation mode, $\ell = 2$.

Patzek *et al.* [18] conducted an analysis of mode interaction and frequency shift using Fourier spectral analysis on the surface of an inviscid deformed droplet experiencing large-amplitude shape oscillations. They attributed the observed frequency decrease to the presence of higher harmonics. Extending these investigations to viscous droplets, Basaran [19] numerically analyzed large-amplitude axisymmetric shape oscillations of droplets immersed in a dynamically inactive medium using a Galerkin finite-element method. Their results were in agreement with the published predictions obtained using the marker-and-cell method. It was demonstrated that viscous droplets remain in the prolate state for a shorter duration than inviscid droplets. Another numerical simulation for investigating large-amplitude shape oscillations of viscous droplets was conducted by Meradji *et al.* [20], showing good agreement with the results of Basaran [19]. These findings, along with other numerical investigations by Smith [21] and Lundgren and Mansour [22], were confirmed by the experimental studies Wang *et al.* [23] and Becker *et al.* [24], as well as by the analytical results of Tsamopoulos and Brown [25] and Zrnić and Brenn [26].

While most of the literature focuses on axisymmetric droplet shape oscillations, few studies have addressed nonaxisymmetric cases. In particular, Pozrikidis [27] developed a numerical method to simulate the three-dimensional oscillations of an inviscid liquid droplet in a vacuum. The difficulty was that unavoidable numerical instabilities in the distribution of the velocity potential on the droplet surface were encountered, which were addressed with smoothing algorithms. The results demonstrated that the numerical procedure provides a successful simulation method, particularly for small to moderate interfacial deformations, confirming the theoretical prediction by Lord Rayleigh [6] that the oscillation frequencies are independent of the azimuthal wave number (m for harmonic order). Azuma and Yoshihara [28] investigated forced three-dimensional droplet shape oscillations experimentally and theoretically, where the forcing produced arbitrary surface shapes rather than free shape oscillations. Both Pozrikidis [27] and Azuma and Yoshihara [28] offered only limited insights into asymmetric droplet shape oscillations. Since nonlinear mode coupling and excitation have not been systematically investigated, the present study aims to address this.

This work builds upon Plümacher *et al.* [29], who applied the unified transform method developed by Fokas [30,31] and Fokas and Pelloni [32,33] to transform the free boundary value problem describing the droplet shape oscillation in a time-dependent droplet domain into a system of integrodifferential equations on a unit sphere S and derived the so-called geometrically exact nonlinear theory (GNLT). Based on GNLT, Akbari *et al.* [34] developed a highly accurate and specific numerical framework to investigate axisymmetric droplet shape oscillations and modal coupling. The present study extends the analysis of Akbari *et al.* [34] to nonaxisymmetric droplet shape oscillations. First, a brief overview of the physical principles and modeling assumptions as well as the solution techniques is given. Following this, the shape oscillations of nonaxisymmetric droplets are investigated, with a particular focus on mode excitation by various initial nonaxisymmetric deformation modes. Modal coupling is further analyzed using a superimposed initial deformation, in which a small-amplitude perturbation mode is superimposed on a nonaxisymmetric base deformation.

II. PHYSICAL MODEL AND SOLUTION TECHNIQUES

A. Model assumptions and governing equations

The governing equations can be formulated based on the continuity and momentum balance equations within the droplet, as well as the jump condition at its surface. The following assumptions are made: incompressibility (droplet fluid density $\rho = \text{const}$), constant surface tension ($\sigma = \text{const}$), absence of an external medium and viscous stresses, and negligible gravity. Using the unified transform method of Fokas [30], Plümacher *et al.* [29] derived a system of integrodifferential equations describing nonlinear droplet shape oscillations with large amplitudes. In particular, these

equations are defined on the surface of a unit sphere $\mathbb{S}$, which greatly simplifies the corresponding numerical simulation due to the time-independent domain.

The unknown variables are the velocity potential $q(\theta, \phi, t)$ on the droplet surface and the droplet radius $\mathcal{R}(\theta, \phi, t)$. These variables depend only on the polar angle θ and azimuthal angle ϕ of the spherical coordinate system, as well as on time t . Using $T = \sqrt{\varrho \mathcal{L}^3 / \sigma}$ as the characteristic time and $\mathcal{L}$ as the characteristic length scale of the droplet, Akbari *et al.* [34] nondimensionalized the governing equations. The resulting nondimensional governing equations are as follows:

$$\mathcal{F}_\ell^m(\mathcal{R}_{,t}, \mathcal{R}, q) := \int_{\mathbb{S}} \mathcal{R}^\ell [(\mathcal{R}^2 \mathcal{R}_{,t} - \ell \mathcal{R} q) Y_\ell^m + q \nabla_{\mathbb{S}} Y_\ell^m \cdot \nabla_{\mathbb{S}} \mathcal{R}] d\Omega = 0 \quad \forall \ell \geq 0, |m| \leq \ell, \quad (4)$$

and

$$\begin{aligned} G(\mathcal{R}_{,t}, q_{,t}, \mathcal{R}, q) := & q_{,t} - \frac{\mathcal{R}_{,t}}{\alpha} \left[\frac{\mathcal{R}^2 \mathcal{R}_{,t}}{2} + \nabla_{\mathbb{S}} \mathcal{R} \cdot \nabla_{\mathbb{S}} q \right] + \kappa - C \\ & + \frac{1}{2\alpha^2} \left[(\nabla_{\mathbb{S}} \mathcal{R} \cdot \nabla_{\mathbb{S}} q)^2 + \mathcal{R}^2 |\nabla_{\mathbb{S}} q|^2 \right. \\ & \left. + (\nabla_{\mathbb{S}} q \times \nabla_{\mathbb{S}} \mathcal{R})^2 \left(\frac{|\nabla_{\mathbb{S}} \mathcal{R}|^2}{\mathcal{R}^2} + 2 \right) \right] = 0, \quad \text{on } \mathbb{S}, \end{aligned} \quad (5)$$

with

$$\begin{aligned} \kappa &= \frac{|N|}{\alpha^2} \lambda, \quad |N| = \sqrt{1 + \frac{|\nabla_{\mathbb{S}} \mathcal{R}|^2}{\mathcal{R}^2}}, \quad \alpha = \mathcal{R}^2 + |\nabla_{\mathbb{S}} \mathcal{R}|^2, \\ \lambda &= [\nabla_{\mathbb{S}} \mathcal{R} \cdot (\mathbf{H}_{\mathbb{S}}^{\mathcal{R}} \cdot \nabla_{\mathbb{S}} \mathcal{R}) - \alpha \Delta_{\mathbb{S}} \mathcal{R} + 3\mathcal{R} |\nabla_{\mathbb{S}} \mathcal{R}|^2 + 2\mathcal{R}^3]. \end{aligned} \quad (6)$$

Here κ is the dimensionless total curvature of the droplet and Y_ℓ^m denotes spherical harmonics of the degree ℓ ($\ell \geq 0$) and order m ($|m| \leq \ell$). The operator $\nabla_{\mathbb{S}}$ represents the surface gradient on the unit sphere $\mathbb{S}$. The operator $\Delta_{\mathbb{S}}$ denotes the Laplace-Beltrami operator, and $\mathbf{H}_{\mathbb{S}}^{\mathcal{R}}$ denotes the surface Hessian of $\mathcal{R}$. The nondimensional integration constant is set to $C = 2$. Different choices do not affect the droplet dynamics and only add a spatially uniform offset to the surface velocity potential, as demonstrated in Plümacher *et al.* [29]. The integral $\int_{\mathbb{S}}(\cdot) d\Omega$ is evaluated on the unit sphere $\mathbb{S}$ with respect to the surface area element $d\Omega = \sin(\theta) d\theta d\phi$.

Equation (4) has been derived from the continuity equation and leads to the infinite system of integrodifferential equations, one for each spherical harmonic mode $\{\ell, m\}$. Equation (5) has been derived from the momentum balance equations within the droplet and the jump condition on the droplet surface. Both Eqs. (4) and (5) are evaluated on the surface of the unit sphere $\mathbb{S}$. A detailed derivation of the governing equations can be found in Plümacher *et al.* [29] and their nondimensionalization in Akbari *et al.* [34].

B. Numerical solution

Plümacher *et al.* [29] solved the governing equations (4) and (5) using the Poincaré expansion method up to second order. Although the theory provides an explicit analytical solution, avoiding the need for numerical computation, its validity is limited to small-amplitude oscillations. To ensure high accuracy even for large deformations, Akbari *et al.* [34] developed a highly specialized numerical code based on the Galerkin method. This numerical method uses spherical harmonics as both test and ansatz functions. The original implementation was designed for axisymmetric cases, allowing for certain simplifications that are no longer valid when two spherical variables (θ, ϕ) are considered for the present nonaxisymmetric droplet shape oscillations. Nevertheless, the core numerical approach remains the same and is outlined only briefly in the following.

First, both governing equations need to be formulated in their weak forms to apply the Galerkin method. While Eq. (4) is already expressed in weak form, the second equation, Eq. (5), must also

be transformed into a weak form. This is done by multiplying by a test function Y_ℓ^m and integrating over the unit sphere surface $\mathbb{S}$, resulting in

$$G_\ell^m := \int_{\mathbb{S}} G(\partial_t \mathcal{R}, \partial_t q, \mathcal{R}, q) Y_\ell^m d\Omega = 0 \quad \text{for } \forall \ell \geq 0 \text{ and } |m| \leq \ell. \quad (7)$$

The two unknowns in the system are the droplet radius $\mathcal{R}(\theta, \phi, t)$ and the surface velocity potential $q(\theta, \phi, t)$. These unknowns are approximated using truncated spherical harmonic expansions

$$\begin{aligned} R(\theta, \phi, t) &\approx a_0^0(t)Y_0^0 + \sum_{\ell=1}^L \sum_{m=-\ell}^{\ell} a_\ell^m(t)Y_\ell^m(\theta, \phi), \\ q(\theta, \phi, t) &\approx b_0^0(t)Y_0^0 + \sum_{\ell=1}^L \sum_{m=-\ell}^{\ell} b_\ell^m(t)Y_\ell^m(\theta, \phi), \end{aligned} \quad (8)$$

with truncation degree L . The time-dependent modal coefficients, $\{a_\ell^m(t), b_\ell^m(t)\}$, are determined numerically. To clarify the physical meaning of the contributions of the zeroth-order spherical harmonics, we have explicitly isolated these terms, $a_0^0(t)Y_0^0$ and $b_0^0(t)Y_0^0$, in the solution approach (8). Both are functions of time only. The contribution of the term $a_0^0(t)Y_0^0$ corresponds to a central pulsation, i.e., a spherically symmetric expansion or contraction of the droplet, rather than a physical oscillation of its shape. Instead, it is considered a compensating contribution that ensures the droplet's volume is conserved during nonlinear deformations because spherical harmonics are generally not volume conserving. The time-dependent contribution $b_0^0(t)Y_0^0$ to the velocity potential has no physical significance and is determined by the initial conditions and the choice of the integration constant C arising in (5).

The numerical implementation of the surface integrals, $\int_{\mathbb{S}}(\cdot)d\Omega$, arising in the governing equations (4) and (7), is carried out using MATLAB's adaptive quadrature routine `integral2` (see Refs. [35,36]). Rather than using a fixed quadrature grid, this routine recursively subdivides the integration domain. Local quadrature errors are estimated from the difference between higher- and lower-order embedded Gauss-Kronrod formulas. Regions that exceed the prescribed tolerance are refined until the error criterion is satisfied. In the present computations, the integrals are evaluated as `integral2(H,0,pi,0,2*pi,'AbsTol',1e-12)`, where H denotes the integrand, and the absolute tolerance is set to 10^{-12} . Thus, the nonlinear integrals are computed using adaptive, error-controlled quadrature rather than a fixed, low-order quadrature rule. This reduces integration errors and mitigates the aliasing of unresolved nonlinear contributions into the retained Galerkin modes.

The temporal discretization is performed using the second-order backward differentiation formula (BDF2), but a BDF1 for the first time step. The resulting nonlinear system is solved at each time step using a Newton scheme. The initial guess for the Newton scheme is obtained by a linear extrapolation of the previous time steps. The system can now be solved with suitable initial conditions. The following initial conditions are considered:

$$\begin{aligned} q(\theta, \phi, 0) &= 0, \\ \mathcal{R}(\theta, \phi, 0) &= 1 + a_2^m(0)Y_2^m(\theta, \phi), \quad m = -2, -1, 0, 1, 2. \end{aligned} \quad (9)$$

The first condition assumes that the droplet is initially at rest, since a vanishing surface velocity potential corresponds to zero kinetic energy of the entire droplet, as shown by Akbari *et al.* [34]. The second condition describes an initial shape deformation of the droplet in the form of the spherical harmonics Y_2^m of degree $\ell = 2$ and different orders $m = -2, -1, 0, 1, 2$ with the same initial amplitude $a_2^m(0) = 0.5$ superimposed on the unit spherical shape (the zeroth-order spherical harmonic), respectively. This indicates that the amplitude of the zeroth-order spherical harmonic has been selected as the characteristic length $\mathcal{L}$ in the nondimensionalization process. As mentioned above, the mode with $\ell = 0$ is not an independent shape oscillation of an incompressible droplet but

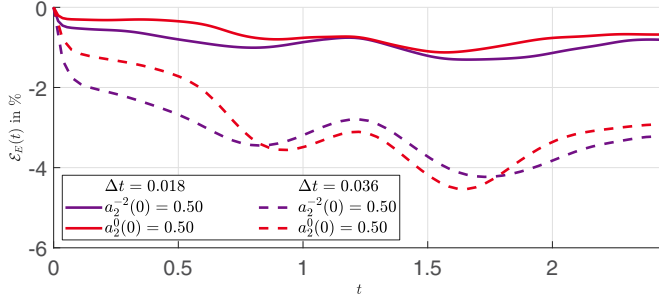


FIG. 1. Relative energy deviation for two different initial deformation modes, $Y_2^{-2}(\theta, \phi)$ and $Y_2^0(\theta, \phi)$, with amplitudes of $a_2^{-2}(0) = 0.5$ and $a_2^0(0) = 0.5$, using different time steps, $\Delta t = 0.036$ and $\Delta t = 0.018$, respectively.

serves only as a volume-correction term. The modes with $\ell = 1$ represent rigid-body translations of the droplet and are therefore also excluded from the initial shape deformation.

To assess the influence of the time step size, we consider the relative energy deviation,

$$\mathcal{E}_E(t) = \frac{E_{\text{pot}}(t) + E_{\text{kin}}(t) - E_{\text{pot}}(0)}{E_{\text{pot}}(0) + E_{\text{kin}}(0)} \times 100\%, \quad (10)$$

with the potential energy $E_{\text{pot}}(t)$ and the kinetic energy $E_{\text{kin}}(t)$. The potential energy is expressed as $E_{\text{pot}}(t) = (A(t) - A_{\text{sph}})\sigma$, where $A(t)$ is the instantaneous surface area of the oscillating droplet and A_{sph} denotes the surface area of the corresponding undeformed sphere with the same volume as the deformed droplet. The total kinetic energy of the oscillating droplet is given by Akbari *et al.* [34] as $E_{\text{kin}} = \frac{\rho}{2} \int_{\mathbb{S}} q \mathcal{R}^2 \mathcal{R}_{,i} d\Omega$. In the absence of viscous stresses and external forces, the total energy should be conserved; thus, the relative energy deviation should be zero. To examine energy conservation during droplet oscillation, we compare two different time step sizes, $\Delta t = 0.036$ and $\Delta t = 0.018$, for two different initial deformation modes, $a_2^{-2}(t)Y_2^{-2}$ and $a_2^0(t)Y_2^0$ with amplitudes of $a_2^{-2}(0) = 0.5$ and $a_2^0(0) = 0.5$, respectively. Figure 1 shows the relative energy deviation, using a truncation degree of $L = 6$. For a time step size of $\Delta t = 0.036$, the total energy can deviate by up to 4.5 % during oscillation. Using a time step size of $\Delta t = 0.018$ achieves a significant improvement, with the energy deviation remaining below 1.2 % for both initial deformations with a chosen truncation degree of $L = 6$. This study indicates that the time step size of $\Delta t = 0.018$ introduces negligible numerical diffusion. This demonstrates the reliability of the numerical solution, as the energy deviation is the most critical factor, as shown in Akbari *et al.* [34]. For the following simulations, the time step size of $\Delta t = 0.018$ is used.

The initial droplet shapes specified in (9) are depicted in Fig. 2. A closer look shows that the initial deformation modes $a_2^{-2}(0) = 0.5$ and $a_2^2(0) = 0.5$, as well as $a_2^{-1}(0) = 0.5$ and $a_2^1(0) = 0.5$, have identical shapes in pairs. For example, a rotation of the deformation for $a_2^{-2}(0) = 0.5$ by

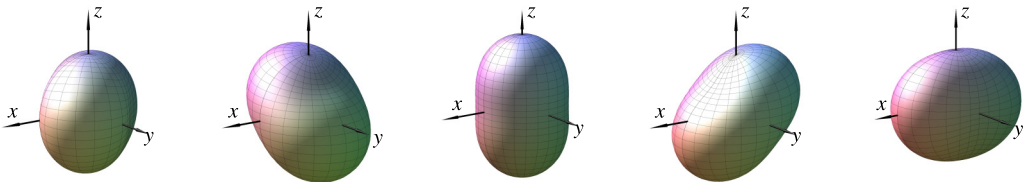


FIG. 2. Initial deformations for $\mathcal{R}(\theta, \phi, 0) = 1 + a_2^m(0)Y_2^m(\theta, \phi)$ with amplitude $a_2^m(0) = 0.5$, shown from left to right for $m = -2, \dots, 2$. Their corresponding oscillation videos can be viewed as Supplemental Material [37] for three different viewing angles.

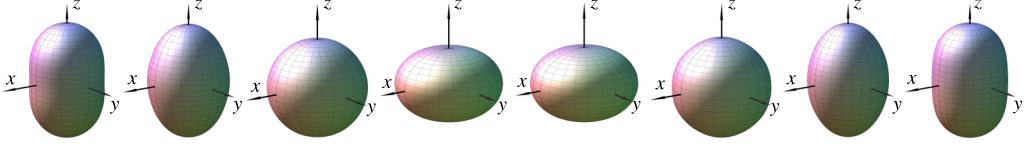


FIG. 3. Shape oscillations for the initial deformation mode $\{m, \ell\} = \{0, 2\}$ with amplitude $a_2^0(0) = 0.5$ at eight different time steps within one oscillation period. The oscillation videos can be viewed as Supplemental Material [37] for three different viewing angles.

$\Delta\phi = -45^\circ$ around the z axis results in the deformation for $a_2^2(0) = 0.5$, while a rotation of $\Delta\phi = -90^\circ$ for $a_2^{-1}(0) = 0.5$ yields the droplet shape of $a_2^1(0) = 0.5$. This discrete symmetry between the positive and negative order m of the spherical harmonics Y_ℓ^m arises from the inherent rotational properties of Y_ℓ^m [see Eq. (11)]. Since the initial deformations are the same for both positive and negative values of m , except for a rotation by a given angle around z axis, their oscillation behavior must also be identical. This provides another test for both the numerical method and the underlying theory. The modified numerical code developed by Akbari *et al.* [34] has proven to be highly accurate.

III. SIMULATION RESULTS

A. Droplet shape oscillations

The shape oscillations for the initial deformations (9) with $a_2^m(0) = 0.5$, for $m = 0, 1, 2$ are shown in Figs. 3, 4, and 5, respectively. The remaining initial deformations for negative values of $m = -1, -2$ are not displayed, as their oscillation behavior, apart from almost invisible small numerical errors, is practically identical, to those of the corresponding positive m cases, but rotated by a certain angle around the z axis. It is evident that the axisymmetric initial deformation mode $\{m, \ell\} = \{0, 2\}$ remains axisymmetric throughout the oscillation, whereas the modes $\{m, \ell\} = \{1, 2\}$ and $\{m, \ell\} = \{2, 2\}$ exhibit clearly nonaxisymmetric oscillatory behavior. Furthermore, the simulation results reveal that the oscillation frequency is affected by the degree ℓ and the deformation amplitude a_2^m , but not by the order m . Thus, although the nonaxisymmetric modes $Y_\ell^{m \neq 0}$ with $\{m, \ell\} = \{1, 2\}$ and $\{m, \ell\} = \{2, 2\}$ differ visibly in shape from the axisymmetric mode $Y_\ell^{m=0}$ with $\{m, \ell\} = \{0, 2\}$, they exhibit the same oscillation frequency due to the same degree ℓ and deformation amplitude a_2^m . These results are consistent with the general theory of droplet shape oscillations, as discussed by Landau and Lifshitz [38] for inviscid fluids and by Prosperetti [39] for viscous fluids.

B. Modal excitation based on initial deformation modes

Of particular interest in this work is the investigation of modal excitation resulting from a given nonaxisymmetric initial deformation, $a_\ell^m(0)$ ($m \neq 0$). To the authors' knowledge, this has not been

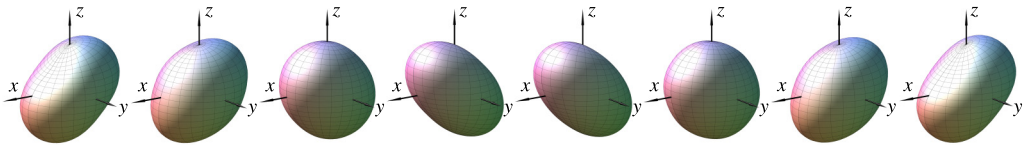


FIG. 4. Shape oscillations for the initial deformation mode $\{m, \ell\} = \{1, 2\}$ with amplitude $a_2^1(0) = 0.5$ at eight different time steps within one oscillation period. The oscillation videos can be viewed as Supplemental Material [37] for three different viewing angles.

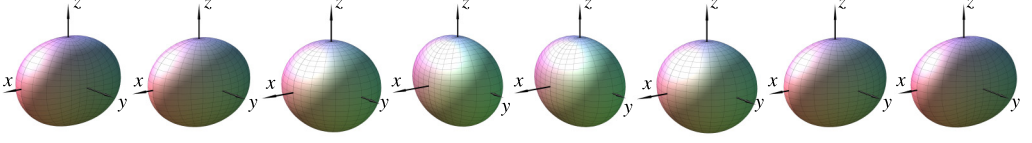


FIG. 5. Shape oscillations for the initial deformation mode $\{m, \ell\} = \{2, 2\}$ with amplitude $a_2^2(0) = 0.5$ at eight different time steps within one oscillation. The oscillation videos can be viewed as Supplemental Material [37] for three different viewing angles.

studied before. The numerical simulation results provide the time-dependent coefficients $a_\ell^m(t)$ and $b_\ell^m(t)$, as shown in the solution approaches (8), at discrete time points. This enables the identification of which modes are excited and which are not. The latter being characterized by corresponding coefficients that are practically zero within the numerical tolerance of Newton's method. Numerical results show that the modal coupling patterns, i.e., which modes are excited and which are not, depend on the initial deformation and are illustrated in Fig. 6, for the four nonaxisymmetric initial deformation modes above mentioned, respectively.

In Fig. 6, the ordinate corresponds to the degree ℓ , while the abscissa corresponds to the order m . All modes up to sixth degree are presented. Due to the properties of the spherical harmonics shown in the solution ansatz (8), the representation naturally forms a pyramid. Gray circles indicate modes that are not excited ($a_\ell^m \approx 0$, $b_\ell^m \approx 0$), while blue squares denote excited modes ($a_\ell^m \neq 0$, $b_\ell^m \neq 0$). The initial deformation mode is marked with a dark blue diamond. Black outlined squares represent modes that were also predicted by the theory of Plümacher *et al.* [29]. It is evident that the analytical predictions of Plümacher *et al.* [29] align perfectly with the present numerical results. However, since their theory only includes modes up to second order, all higher-order excited modes observed in the present numerical results cannot be predicted. For the axisymmetric initial deformation mode

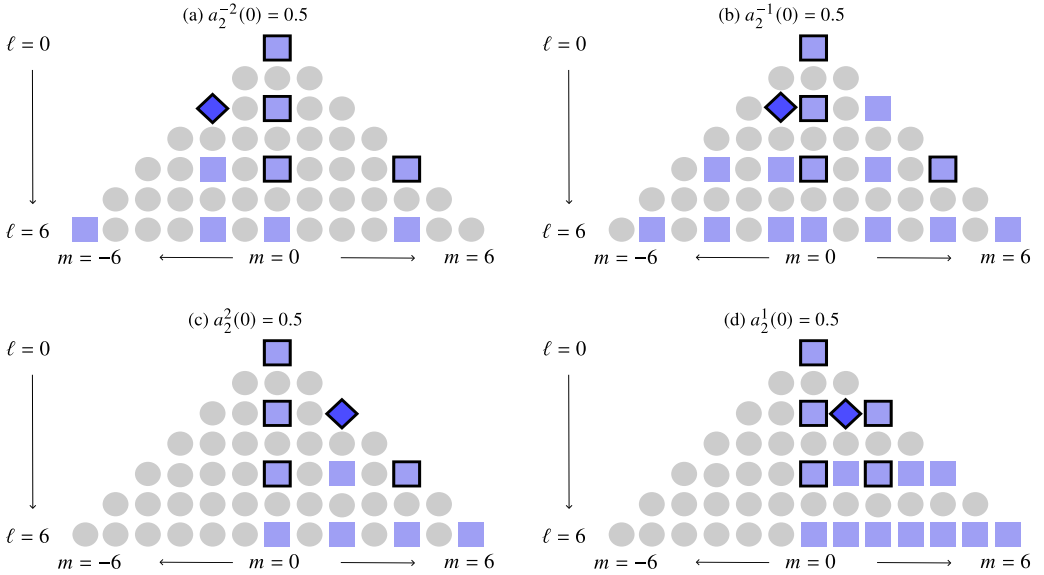


FIG. 6. Excited modes by a single initial deformation mode $\mathcal{R}(\theta, \phi, 0) = 1 + a_2^m(0)Y_2^m(\theta, \phi)$ for (a) $m = -2$, (b) $m = -1$, (c) $m = 2$, and (d) $m = 1$, respectively. The ordinate corresponds to the degree ℓ and the abscissa is the order m . The initial deformation mode is marked with a dark blue diamond. Blue squares denote excited modes. Squares outlined in black indicate modes that are also predicted by the theory of Plümacher *et al.* [29]. Gray circles indicate unexcited modes.

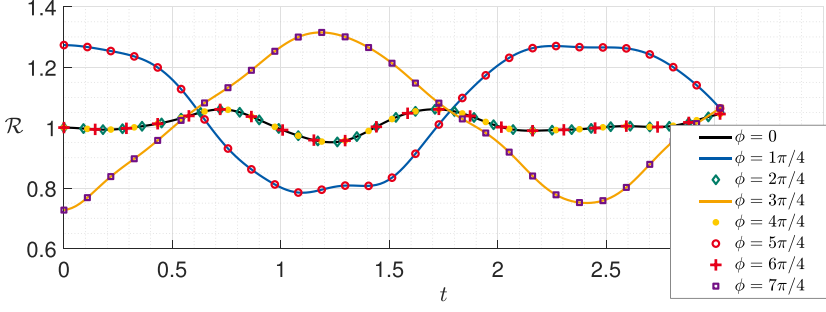


FIG. 7. Time series of the nondimensionalized equatorial positions $\mathcal{R}(\theta = \pi/2, \phi = k\pi/4, t)$ for $k = 0, \dots, 7$ over the nondimensional time t for an initial deformation $\mathcal{R}(\theta, \phi, 0) = 1 + 0.5 Y_2^{-2}(\theta, \phi)$.

$\{m, \ell\} = \{0, 2\}$, not shown in Fig. 6, the results of Akbari *et al.* [34] and Zrnić and Brenn [26] are reproduced, in which case only axisymmetric even modes are excited, i.e., with $m = 0$ and even values of ℓ .

From these patterns (Fig. 6) of excited modes, it can be seen that only some specific modes are excited by a single initial deformation mode. The principle behind this excitation will be analyzed in Sec. III D. Here we first observe some symmetry behavior in the mode excitation patterns between the initial deformation modes with the positive and negative values of m . As discussed above, the initial deformations for $a_2^{-2}(0) = 0.5$ and $a_2^2(0) = 0.5$, as well as for $a_2^{-1}(0) = 0.5$ and $a_2^1(0) = 0.5$, are each equivalent up to a rigid-body rotation by an angle $\Delta\phi = -45^\circ$ and $\Delta\phi = -90^\circ$, respectively. In fact, a rotation around the z axis by an arbitrary angle $\Delta\phi$ leads to the following transformation characteristics for the oscillation modes, i.e., the spherical harmonics:

$$\begin{pmatrix} Y_\ell^m(\theta, \phi + \Delta\phi) \\ Y_\ell^{-m}(\theta, \phi + \Delta\phi) \end{pmatrix} = \begin{pmatrix} \cos(m\Delta\phi) & -\sin(m\Delta\phi) \\ \sin(m\Delta\phi) & \cos(m\Delta\phi) \end{pmatrix} \begin{pmatrix} Y_\ell^m(\theta, \phi) \\ Y_\ell^{-m}(\theta, \phi) \end{pmatrix}. \quad (11)$$

For example, applying a rotation by an angle of $\Delta\phi = -45^\circ$ to all excited modes in the modal pattern in Fig. 6(a) for initial deformation mode $\{m, \ell\} = \{-2, 2\}$, exactly the modal pattern in Fig. 6(c) for initial deformation mode $\{m, \ell\} = \{2, 2\}$ is generated. The same holds for modal patterns in Fig. 6(b) for $\{m, \ell\} = \{-1, 2\}$ and Fig. 6(d) for $\{m, \ell\} = \{1, 2\}$, but for a rotation angle of $\Delta\phi = -90^\circ$. This means that these rotational invariances occurring in the initial deformation modes are maintained in the corresponding excited modes.

C. Time series for initial deformation $\mathcal{R}(\theta, \phi, 0) = 1 + 0.5 Y_2^{-2}(\theta, \phi)$

The time-dependent superposition behavior of the different modes excited by a single initial deformation mode can also be observed from the oscillatory motions of selected specific positions on the droplet surface. Figure 7 shows the oscillations of eight nondimensional equatorial positions $\mathcal{R}(\theta = \pi/2, \phi = k\pi/4, t)$ for $k = 0, \dots, 7$ over the nondimensional time t for an initial deformation $\mathcal{R}(\theta, \phi, 0) = 1 + 0.5 Y_2^{-2}(\theta, \phi)$. On the one hand, the oscillations are not harmonic due to superposition of different excited modes. On the other hand, the droplet oscillation exhibits certain discrete symmetries. It is evident that the oscillation behavior of several corresponding positions at the equator is exactly the same in groups. These are the cases for $\{\phi = \pi/4, \phi = 5\pi/4\}$, $\{\phi = 3\pi/4, \phi = 7\pi/4\}$ and $\{\phi = 0, \phi = 2\pi/4, \phi = 4\pi/4, \phi = 6\pi/4\}$. These discrete symmetries of the droplet shape oscillation by an initial deformation mode are analyzed in the following.

D. Symmetry preservation of a droplet oscillation

As previously observed in the numerical results, a single initial deformation mode Y_2^m can excite certain harmonic modes, as shown in Fig. 6, for $m = -2, -1, 1, 2$, respectively. Furthermore,

TABLE I. Symmetries of initial deformations $\mathcal{R} = 1 + a_2^m(0)Y_2^m(\theta, \phi)$ for $m = -2, -1, 1, 2$. A cross indicates that a given symmetry is absent, whereas a tick denotes its presence.

Symmetries	$Y_2^{-2}(\theta, \phi)$ $\frac{\sqrt{15}}{4\sqrt{\pi}} \sin^2(\theta)\sin(2\phi)$	$Y_2^{-1}(\theta, \phi)$ $-\frac{\sqrt{15}}{4\sqrt{\pi}} \sin(2\theta)\sin(\phi)$	$Y_2^1(\theta, \phi)$ $-\frac{\sqrt{15}}{4\sqrt{\pi}} \sin(2\theta)\cos(\phi)$	$Y_2^2(\theta, \phi)$ $\frac{\sqrt{15}}{4\sqrt{\pi}} \sin^2(\theta)\cos(2\phi)$
$\phi \rightarrow \phi + \pi$	✓	✗	✗	✓
$\theta \rightarrow \theta + \pi$	✓	✓	✓	✓
$\phi \rightarrow -\phi$	✗	✗	✓	✓
$\theta \rightarrow \pi - \theta$	✓	✗	✗	✓
$\phi \rightarrow \phi + \frac{\pi}{2}$	✗	✗	✗	✗
$\phi \rightarrow \phi + \epsilon, \quad \forall \epsilon$	✗	✗	✗	✗
$\phi \rightarrow \pi - \phi$	✗	✓	✗	✓
$\phi \rightarrow -\phi + \frac{\pi}{2}$	✓	✗	✗	✗
$\phi \rightarrow -\phi - \frac{\pi}{2}$	✓	✗	✗	✗
$\phi \rightarrow -\phi, \theta \rightarrow -\theta$	✗	✓	✗	✓
$\phi \rightarrow \pi - \phi$	✗	✗	✓	✓
$\theta \rightarrow \pi - \theta$	✗	✓	✗	✓
$\phi \rightarrow -\phi$	✗	✓	✗	✓
$\theta \rightarrow \pi - \theta$	✗	✓	✗	✓

the oscillation motion superimposed by these excited modes exhibits certain symmetries, as can be seen in Fig. 7 for $m = -2$ as an example. These oscillation behaviors can be associated with the symmetry properties of these oscillation modes. The corresponding discrete symmetries for these employed initial deformation modes are summarized in Table I. Possible discrete symmetries are listed in the leftmost column. The top row shows the spherical harmonics corresponding to different initial deformations. A cross indicates the absent of a given symmetry, while a tick denotes its presence. As shown, each initial deformation exhibits its own symmetry pattern. Comparing these symmetric patterns with the modes that are excited by corresponding initial deformation modes in Fig. 6 leads to the fundamental finding that mode excitation is based on symmetry conservation. The symmetries of an initial deformation, as shown in Table I, are preserved throughout the oscillation of the droplet. As a result, only a limited number of modes compatible with the symmetries of the initial deformation can be excited. The numerical results show that all and only those modes that exhibit all symmetries of the initial deformation are excited, as shown in Fig. 6. For the sake of simplicity, the discrete symmetries of these excited modes, which contain all symmetries of the initial deformation but go beyond them, are not given here.

E. Modal coupling induced by superimposed initial deformation and velocity-potential modes

Another important aspect of the present study is to investigate the shape oscillation behavior of a droplet when a base deformation mode is superimposed with either (i) another deformation mode or (ii) a nonzero initial velocity-potential mode. The first approach (i) considers the superimposition of a small-amplitude perturbation mode that is not naturally excited by the base mode during oscillation. This approach allows us to investigate whether further additional modes are triggered, how their amplitudes evolve over time, and whether the underlying symmetries of the system are preserved. To explore these, the following initial conditions are considered:

$$q(\theta, \phi, 0) = 0, \\ \mathcal{R}(\theta, \phi, 0) = 1 + a_2^{-1}(0)Y_2^{-1}(\theta, \phi) + a_3^m(0)Y_3^m(\theta, \phi), \quad m = -3, -1, 1, 3. \quad (12)$$

As can be seen in Fig. 6(b), these superimposed modes Y_3^m are not expected to be excited by the base mode Y_2^{-1} , since each mode Y_3^m contains only partial symmetries of mode Y_2^{-1} , but not all of them.

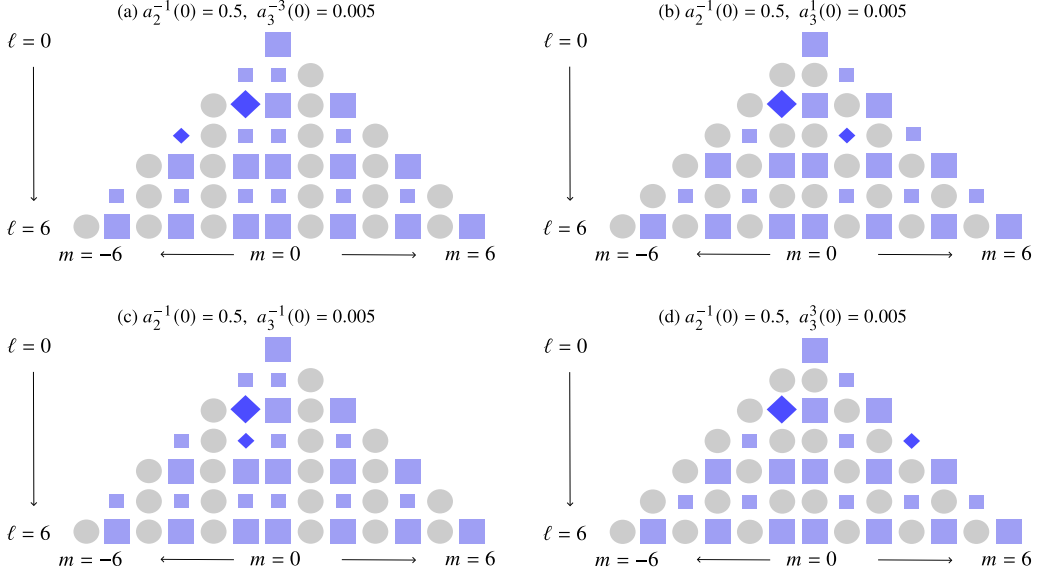


FIG. 8. Modal coupling scheme for superimposed initial deformations $\mathcal{R}(\theta, \phi, 0) = 1 + a_2^{-1}(0)Y_2^{-1}(\theta, \phi) + \frac{1}{100}a_3^m(0)Y_3^m(\theta, \phi)$, with $m = -3, -1, 1, 3$. The initial deformation modes are marked with a dark blue diamond. Blue squares denote excited modes. The reduced symbol size reflects a small amplitude. The gray circles indicate unexcited modes.

Here the base deformation mode Y_2^{-1} is chosen with a large amplitude $a_2^{-1}(0) = 0.5$, while a small-amplitude perturbation mode Y_3^m , each with $m = -3, -1, 1, 3$, is superimposed with a much smaller amplitude $a_3^m(0) = 0.005$. The amplitude of the perturbation mode is chosen to be two orders of magnitude smaller than that of the dominant base deformation mode, such that it is practically not visible in the droplet shape. Numerical results show that with the addition of much smaller perturbation modes, more modes are excited during the droplet oscillation, as illustrated in the corresponding excited mode patterns in Fig. 8. From the time evolution of the corresponding mode coefficients, not shown here, it is evident that the superimposed small-amplitude initial modes and the additional modes excited by them remain weak and do not grow during the oscillation.

This reflects a weak coupling between the two superimposed initial modes. The reason behind the additional modes excited by the superimposed initial deformation, more than those excited only by the single base mode, is that the additional perturbation modes break some of the discrete symmetries of the initial base deformation, as indicated in Table II, even if the amplitude of the superimposed mode is vanishingly small. Since certain discrete symmetries of the base deformation are broken, fewer symmetry constraints remain during the droplet shape oscillation. For example, in the case of the base mode Y_2^{-1} superimposed by the mode Y_3^1 or Y_3^3 , only one of four discrete symmetries is still present. As a result, more modes are excited because fewer restrictions need to be satisfied. Nevertheless, the amplitudes of the additionally excited modes remain small. Interestingly, it could be observed that all modes are excited during oscillation if the initial deformation of the droplet shape is a superposition of modes, e.g., Y_2^{-2} , Y_2^{-1} , and Y_3^{-3} , since no common symmetries exists between them, as shown in Tables I and II. Corresponding results are not presented here.

The second approach (ii) considers the superposition of different nontrivial initial velocity-potential modes on the base deformation mode. This allows us to study how symmetry-breaking initial velocity fields influence the modal dynamics, trigger additional oscillatory modes, and modify the subsequent evolution of the droplet shape. To this end, a series of cases are investigated in which different initial velocity potentials, $q(\theta, \phi, 0)$, are superimposed on the base deformation,

TABLE II. Discrete symmetries of superimposed initial deformation modes $\mathcal{R}(\theta, \phi, 0) = 1 + a_2^{-1}(0)Y_2^{-1}(\theta, \phi) + \frac{1}{100}a_3^m(0)Y_3^m(\theta, \phi)$, for $m = -3, -1, 1, 3$. A cross indicates that a given symmetry is absent, whereas a tick denotes its presence.

Symmetries	$Y_3^{-3}(\theta, \phi)$ $\frac{\sqrt{70}}{8\sqrt{\pi}} \sin(3\phi) \sin^3(\theta)$	$Y_3^{-1}(\theta, \phi)$ $\frac{\sqrt{42}}{8\sqrt{\pi}} \sin(\phi) \sin(\theta)$ $(-1 + 5 \cos^2(\theta))$	$Y_3^1(\theta, \phi)$ $\frac{\sqrt{42}}{8\sqrt{\pi}} \cos(\phi) \sin(\theta)$ $(-1 + 5 \cos^2(\theta))$	$Y_3^3(\theta, \phi)$ $\frac{\sqrt{70}}{8\sqrt{\pi}} \cos(3\phi) \sin^3(\theta)$
$\theta \rightarrow \theta + \pi$	✗	✗	✗	✗
$\phi \rightarrow \pi - \phi$	✓	✓	✗	✗
$\phi \rightarrow -\phi$	✓	✓	✗	✗
$\theta \rightarrow -\theta$				
$\phi \rightarrow -\phi$	✗	✗	✓	✓
$\theta \rightarrow \pi - \theta$				

$\mathcal{R}(\theta, \phi, 0)$,

$$q(\theta, \phi, 0) = b_2^m(0)Y_2^m(\theta, \phi), \quad m = -2, -1, 0, 1, 2,$$

$$\mathcal{R}(\theta, \phi, 0) = 1 + a_2^{-1}(0)Y_2^{-1}(\theta, \phi), \quad (13)$$

with the amplitudes of $b_2^m(0) = 0.25$ and $a_2^{-1}(0) = 0.5$. Due to the nonzero initial velocity potential, the droplet-shape evolution is significantly altered. Figure 9 illustrates the time series of the nondimensional equatorial positions, $\mathcal{R}(\theta = \pi/2, \phi = k\pi/4, t)$ with $k = 0, \dots, 7$, for the initial deformation $\mathcal{R}(\theta, \phi, 0) = 1 + 0.5Y_2^{-1}(\theta, \phi)$. Figure 9(a) shows the case with a zero initial velocity potential $q(\theta, \phi, 0) = 0$, and Fig. 9(b) includes $q(\theta, \phi, 0) = b_2^{-2}(0)Y_2^{-2}(\theta, \phi)$. The resulting oscillation response differs considerably between the two cases, as evidenced by the altered phase and amplitude behavior of the equatorial positions. Furthermore, in the case of zero initial velocity, as shown in Fig. 9(a), the equatorial positions can be grouped into three sets: $\{\phi = 0, \phi = \pi\}$, $\{\phi = \pi/2, \phi = 3\pi/2\}$, and $\{\phi = \pi/4, \phi = 3\pi/4, \phi = 5\pi/4, \phi = 7\pi/4\}$. The oscillations of the equatorial positions in each set are equivalent. This behavior is consistent with the shape symmetry of the initial deformation mode, $Y_2^{-1}(\theta, \phi)$. In the case of an initial velocity potential mode, $Y_2^{-2}(\theta, \phi)$, as shown in Fig. 9(b), there are four such groups, each with equivalent equatorial positions. This behavior arises because the initial velocity potential, $q(\theta, \phi, 0) = 0.25Y_2^{-2}(\theta, \phi)$, breaks some of the discrete symmetries of the initial deformation $\mathcal{R}(\theta, \phi, 0) = 1 + 0.5Y_2^{-1}(\theta, \phi)$, and splits the

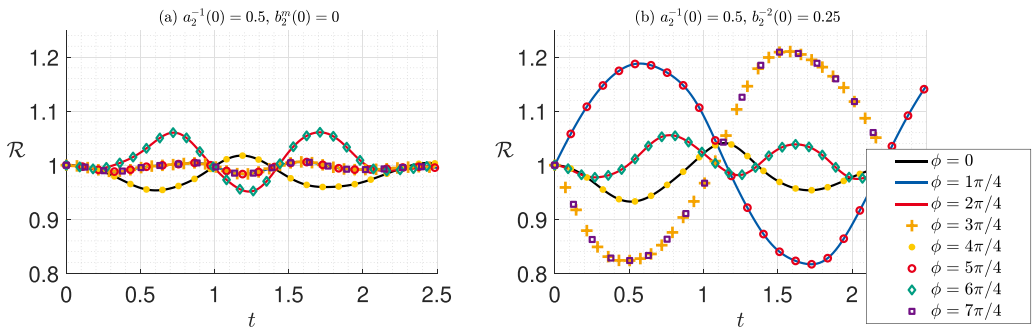


FIG. 9. Time series of the nondimensionalized equatorial positions, $\mathcal{R}(\theta = \pi/2, \phi = k\pi/4, t)$ for $k = 0, \dots, 7$, with the same initial deformation, $\mathcal{R}(\theta, \phi, 0) = 1 + a_2^{-1}Y_2^{-1}(\theta, \phi)$, but with different initial velocity potentials, $q(\theta, \phi, 0) = 0$ in panel (a) and $q(\theta, \phi, 0) = b_2^{-2}Y_2^{-2}(\theta, \phi)$ in panel (b).

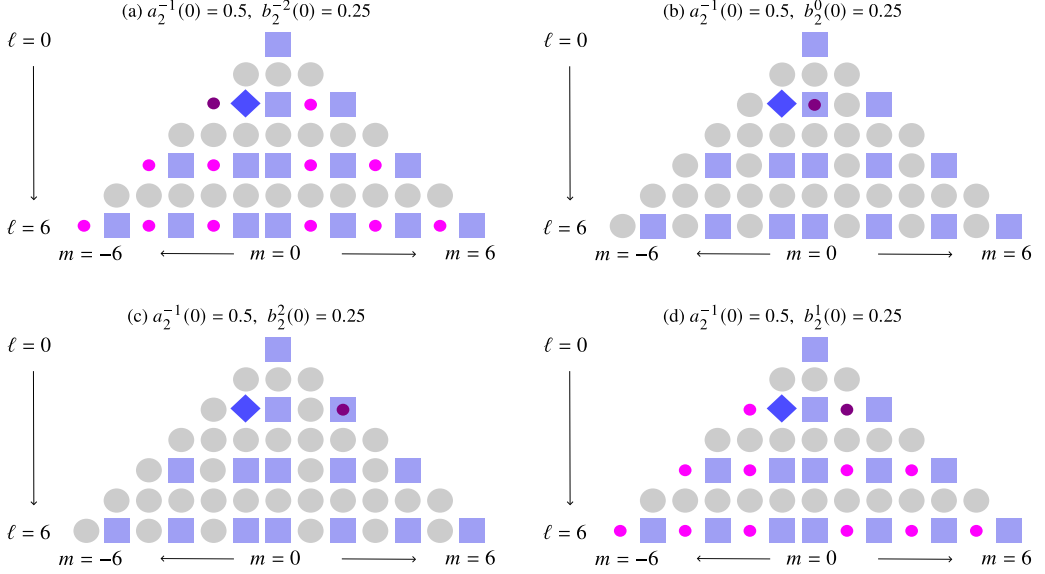


FIG. 10. Modal coupling scheme for the initial deformation, $\mathcal{R}(\theta, \phi, 0) = 1 + a_2^{-1}(0)Y_2^{-1}(\theta, \phi)$, and the initial velocity potential $b_2^m(0)Y_2^m(\theta, \phi)$ with $m = -2$ in panel (a), $m = 0$ in panel (b), $m = 2$ in panel (c), and $m = 1$ in panel (d), respectively. The initial deformation modes are marked with dark blue diamonds. Blue squares denote excited modes. Small violet circles indicate the initial velocity potential and pink circles reflect the additional excited modes due to the superimposed initial velocity potential. Gray circles indicate unexcited modes.

group $\{\phi = \pi/4, \phi = 3\pi/4, \phi = 5\pi/4, \phi = 7\pi/4\}$ into two distinct pairs, $\{\phi = \pi/4, \phi = 5\pi/4\}$ and $\{\phi = 3\pi/4, \phi = 7\pi/4\}$.

As more discrete symmetries are broken, more modes can be excited. This is evident in the excited mode patterns shown in Fig. 10. As shown in Eq. (13), different initial velocity potentials, $q(\theta, \phi, 0) = 0.25 Y_2^m(\theta, \phi)$ with $m = -2, 0, 1, 2$, marked with a small violet circle in Fig. 10, are superimposed on the base deformation, $\mathcal{R}(\theta, \phi, 0) = 1 + 0.5 Y_2^{-1}(\theta, \phi)$. If the initial velocity potential contains all the symmetries of the initial droplet deformation, the nonzero initial velocity will not cause symmetry breaking; thus, no further modes will be excited. This is the case in Figs. 10(b) and 10(c), for which the initial velocity potential modes, Y_2^0 and Y_2^2 , contain all the symmetries of the initial deformation mode, Y_2^{-1} , respectively. In these cases, the discrete symmetries of the base deformation are not broken, and thus all modes excited by the initial velocity potential are also excited by the initial deformation. However, when an initial velocity potential mode breaks the symmetries of the initial droplet deformation, additional modes are excited by introducing a nonzero initial velocity. This is evident in Figs. 10(a) and 10(d), where these additional modes are indicated by small pink circles.

IV. CONCLUSION

In this study, nonaxisymmetric droplet shape oscillations were analyzed. For this purpose, the numerical method developed in Ref. [34] for the investigation of axisymmetric droplet oscillations was extended to the analysis of nonaxisymmetric cases. This approach uses spherical harmonic expansions to directly resolve temporal evolution of individual modal amplitudes to be resolved directly. This makes it possible to identify the modal excitation patterns generated by prescribed nonaxisymmetric initial deformations and initial velocity potentials.

The numerical results of the droplet shape oscillations revealed the central finding that the discrete symmetries of the initial droplet state are preserved throughout the oscillation. Consequently, only modes that are compatible with the common symmetries of the initial deformation and the initial velocity potential can be excited. For a single initial deformation mode with no initial velocity, this leads to restricted excitation patterns that align with the second-order analytical predictions of Plümacher *et al.* [29], where such predictions are available, and extend them to higher-order modes.

The role of symmetry breaking was further demonstrated by superimposing a base deformation mode with an additional, small-amplitude mode that cannot be excited by the base deformation. In this case, the excited modes are those that exhibit all the common symmetries of the two superimposed initial deformation modes. As can be seen, the superimposed small-amplitude mode causes the symmetry breaking of the single initial deformation mode and the additional excited modes. This mode barely decays or grows during the oscillation, indicating weak modal coupling. A similar mechanism was observed when introducing nonzero initial velocity-potential modes. Additional modes are only excited when the initial velocity potential breaks the symmetry of the initial droplet shape.

Knowledge gained from modal excitation provides valuable insights into the oscillation behavior. Simulations show that the oscillation frequency is independent of the order m for a given degree ℓ and deformation amplitude. Therefore, axisymmetric and nonaxisymmetric deformation modes with the same degree ℓ exhibit the same frequency despite their different shapes. Beyond its physical relevance, the observation that symmetry preservation determines which modes can be excited has direct numerical implications. Taking symmetry preservation into account allows us to eliminate known nonexcitable modes from the solution approach. This will significantly reduce computational effort without compromising numerical accuracy.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [40].

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