

Modal coupling of nonlinear inviscid axisymmetric droplet shape oscillations

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We investigate the nonlinear axisymmetric shape oscillations of inviscid droplets, focusing on the modal coupling between different oscillation modes. The research builds upon the geometrically exact nonlinear theory for droplet shape oscillations by Plümacher *et al.* [*Phys. Fluids* **32**, 067104 (2020)], which allows for large deformations and superimposed initial deformations. The theory rests on the potential flow assumption and the usual momentum jump condition at the droplet surface including the surface tension. Using the unified transform method of A. S. Fokas [*Proc. R. Soc. London Ser. A* **453**, 1411 (1997)], the governing equations are transformed into a system of integrodifferential equations defined on a unit sphere. These equations are then solved numerically with a Galerkin method that uses spherical harmonics and exhibits overall spectral convergence. The frequency shift and the extended time spent in the prolate state within an oscillation period are analyzed for different initial deformation modes and show good agreement with existing literature. The results demonstrate that for large initial deformations, the modal coupling significantly influences the oscillation dynamics, particularly in the interaction between even and odd modes. To study modal coupling in detail, superimposed initial deformations are investigated in which a large-amplitude even mode is superimposed with various individual small-amplitude odd modes. It is shown that the initially excited large-amplitude even mode remains nearly unaffected by the superimposed small-amplitude odd modes. The initially superimposed small-amplitude odd mode, however, is altered by the large-amplitude even mode without being significantly amplified or damped. This nonlinear modal coupling is not present in single even initial deformation modes, where it is shown that an even initial deformation mode cannot excite odd modes.

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I. INTRODUCTION

Droplet shape oscillations are of crucial importance in various fields of science and technology, e.g., in cloud physics [1], mass transfer operations in chemical engineering [2], and containerless processing in a microgravity environment [3]. The deviation of a droplet from its spherical shape leads to an increase in surface area, which in turn can lead to an increase in mass, momentum,

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and energy transfer across the droplet surface. The study of droplet shape oscillations dates back to Savart [4] and Plateau [5], where the free shape oscillations of droplets caused by the breakup of a liquid jet were investigated. Strutt (Lord Rayleigh) [6] considered infinitesimally small shape oscillations of an inviscid droplet about its spherical shape in a vacuum and treated the surface tension as a restoring force. A fundamental result is the Rayleigh angular frequency of oscillation ω_ℓ^R as

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

with the surface tension σ , the droplet density ϱ , the undeformed droplet radius R_0 , and the degree of initial deformation mode ℓ , respectively. The respective Rayleigh period, indicated with the superscript R , is defined by $\tau_\ell^R = (2\pi)/\omega_\ell^R$ and the Rayleigh frequency by $f_\ell^R = 1/\tau_\ell^R$. Apparently, higher initial deformation modes ℓ have higher frequencies, i.e., shorter oscillation periods. Lamb [7,8] generalized Strutt's results by considering a viscous sphere surrounded by another liquid medium with a different density. It was shown that for a negligible viscosity, the Rayleigh frequency of oscillation as given in Eq. (1) was proved, while the oscillation amplitude for a small capillary number is damped as $\exp(-\lambda t)$, where $\lambda = (\nu/R_0^2)(\ell-1)(2\ell+1)$, with ν being the kinematic viscosity of the droplet.

The first step toward a more fundamental theory was taken by Chandrasekhar [9]. By normal-mode analysis for small-amplitude shape oscillations of a liquid globe with self-gravitation forces neglecting surface tension, he numerically solved a linearized version of the Navier-Stokes equations and showed that if the relation $\omega_\ell^R R_0^2/\nu$ is large, then a damped oscillation occurs. For smaller values of $\omega_\ell^R R_0^2/\nu$, an aperiodic decay was observed. For small viscosity, Reid [10] considered surface tension as the restoring force instead of self-gravitation and reproduced the damping rate discovered by Lamb [8] and Chandrasekhar [9]. Miller and Scriven [11] considered both the viscous and inertial effects of a viscous ambient medium surrounding a droplet undergoing shape oscillations and examined different interfacial conditions. For shape oscillations of a viscous liquid droplet in a vacuum or gas of negligible density and viscosity, their results were in good agreement with the results of Reid [10].

The first computer simulations for slightly viscous large-amplitude droplet shape oscillations were carried out using a Marker-and-Cell finite-difference method by Foote [12] and Alonso [13]. Foote [12] showed that for larger deformations of the initial deformation mode $\ell = 2$, the droplet spends considerably more time in the prolate configuration than in the oblate and predicted a linear behavior between the aspect ratio and the asymmetry between prolate and oblate state. Furthermore, it was shown therein that the oscillation frequency decreases with increasing aspect ratio L/W (with length L and width W) of the viscous droplet, and demonstrated that there should be a functional relation between the increase in oscillation frequency and the aspect ratio. A further analysis was performed by Prosperetti [14], where small-amplitude (linearized) shape oscillations of free droplets, gas bubbles and droplets in a viscous medium were investigated. It was illustrated that the motion consists of modulated damped oscillations with the damping parameter and frequency approaching only asymptotically the results of the normal-mode analysis. For small viscosity ν , the angular frequency $\bar{\omega}_\ell$ was characterized by $\bar{\omega}_\ell = \sqrt{(\omega_\ell^R)^2 - \lambda^2}$ with the damping parameter λ .

Experimental analyses of large-amplitude shape oscillations of droplets were performed by Trinh and Wang [15]. They used acoustic radiation pressure forces to trap a droplet in a stable position, deform its equilibrium shape and drive it into shape oscillation. A frequency decrease with increasing amplitude of the droplet deformation and an asymmetry of the prolate and oblate times for the two-lobed (mode $\ell = 2$) initial deformation mode were also confirmed. These results compared well with Tsamopoulos and Brown [16], who analyzed moderate-amplitude axisymmetric shape oscillations of inviscid and incompressible liquid droplets and bubbles. Based on the method of Poincaré and Lindstedt for approximating the time-periodic solutions of the nonlinear differential equation, they expanded Strutt's analysis to the second order. It was stated that the frequency

of oscillation was found to decrease with the square of the amplitude. Further experiments were performed by Wang *et al.* [17] for nonlinear shape oscillations of two different low-viscosity droplets in different states of motion in a micro-gravity environment of a space shuttle flight. For the classical case of the shape oscillations of a low-viscosity droplet, the frequency shift agrees well with the predictions of the inviscid nonlinear theory of Tsamopoulos and Brown [16].

Natarajan and Brown [18,19] investigated the internal resonance between different deformation modes in inviscid nonlinear droplet shape oscillations. It was observed that the nonlinear shape oscillations of inviscid drops with surface tension exhibit complex behaviors, including instability and stochastic motions, driven by cubic and quadratic resonances influenced by the interactions of different oscillatory modes. A numerical boundary-integral method was developed by Lundgren and Mansour [20] to investigate mode coupling in large-amplitude shape oscillations with small viscosity. They stated that small viscosity has a relatively large effect on resonant-mode coupling phenomena. Becker *et al.* [21] presented an inviscid theoretical model for the prediction of the surface tension from large-amplitude shape oscillations of low-viscosity droplets. Patzek *et al.* [22] conducted an analysis of mode interaction and frequency shift using Fourier spectral analysis on the surface of a deformed droplet experiencing large-amplitude shape oscillations. They attributed the observed frequency decrease to the presence of higher harmonics. Basaran [23] analyzed the axisymmetric shape oscillations of a viscous droplet immersed in a dynamically inactive medium at medium-to-large amplitudes. A Galerkin/Finite-Element technique, an implicit predictor-corrector method, and Newton's method were employed to solve the resulting system of nonlinear algebraic equations. The results were compared with the few published predictions made by the Marker-and-Cell method and experiments. It was shown that viscous droplets [released from initially two-lobed ($\ell = 2$) configurations] spend less time in prolate form than inviscid droplets. Another numerical simulation of the shape oscillations of viscous droplets with large amplitude was performed by Meradji *et al.* [24]. The obtained results were in excellent agreement with those of Basaran [23]. Later, Smith [25] studied large-amplitude shape oscillations (about 20% of the droplet radius) of incompressible viscous droplets for a small Ohnesorge number, which is a dimensionless number relating the viscous force to the surface tension force.

The weakly nonlinear theory (WNLT) by Zrnić and Brenn [26] investigated the shape oscillations of an inviscid liquid droplet in vacuum. Their theory is related to that proposed by Tsamopoulos and Brown [16], but differs significantly in that it incorporates the third-order mode, thereby enabling the representation of the quasiperiodicity observed in droplet shape oscillations. Furthermore, Zrnić and Brenn [27] recently investigated the shape oscillations of viscoelastic droplets with the WNLT. Compared to the Newtonian case, the theory shows an amplification or damping of the nonlinear behavior and an increased mode coupling. Smuda *et al.* [28] compared both the analytical WNLT with the numerical method of the extended discontinuous Galerkin method (XDG). Analytical insights into the quasiperiodic time behavior of the shape oscillations were given by the WNLT. In the case of large initial deformations, the XDG is used to calculate the shape oscillations and damping behavior as well as the conversion of kinetic and surface energies during the shape oscillations with very low volume loss (0.11%), which is not possible with the WNLT. The foundation of this current work is based on the geometrically exact nonlinear theory (GNLT) proposed by Plümacher *et al.* [29], who applied the unified transform method developed by Fokas [30,31], Fokas and Pelloni [32,33] to simplify the free boundary value problem into a system of integrodifferential equations. This approach enabled a dimensional reduction, as the state of the droplet flow is uniquely characterized by the velocity potential on the surface q and the local droplet radius $\mathcal{R}$, both of which depend only on the polar and azimuthal angles θ and φ on a sphere and time t .

The theory allows for the simulation of droplet shape oscillations with reasonably large deformations. In particular, it enables the calculation of superimposed initial deformation modes. A case of special interest involves a large-amplitude even mode superimposed with a small-amplitude odd mode. Since the calculations are performed at zero viscosity, resulting in undamped shape oscillations, and the numerical methods employed admit spectral accuracy, we have the opportunity to study the modal coupling between even and odd modes without any further disturbing effect. One

of the questions addressed in this research is whether small-amplitude odd modes are amplified over time or remain finitely small, and how the modal coupling between odd and even modes operates. Introducing viscosity would complicate the analysis, since the small-amplitude odd mode would be rapidly damped, making an analysis over several oscillation periods hardly feasible. For this reason, we are neglecting viscous stresses in our study.

To explore this and other related questions, the mathematical problem of the GNLT is formulated in Sec. II, while the simulation model is described in detail in Sec. III. Furthermore, the precision of the simulation code is tested in Sec. III in three distinct ways: first, by a temporal and spatial convergence test, second, by ensuring that the droplet volume remains virtually constant, even at large amplitudes; third, by verifying that, in the absence of viscosity, the total energy remains constant. In Sec. IV, the axisymmetric case is explored, with droplet shape oscillations up to the sixth initial deformation mode ($\ell = 6$). These findings are physically verified by the motion of the north pole of the droplet and compared with the well-established literature. Furthermore, the timescales of the droplet shape oscillations are presented, including detailed analyses of the frequency shift and the extended time in the prolate state. Last, investigations of modal coupling and power spectrum are detailed. The shape oscillations of a large-amplitude even mode ($\ell = 2$) superimposed with small-amplitude odd modes ($\ell = 3$) and ($\ell = 5$) are analyzed. The conclusion is given in Sec. V.

II. FORMULATION OF THE PROBLEM AND MODEL EQUATIONS

The equation system governing an incompressible, inviscid droplet shape oscillations in vacuum is briefly introduced, while a detailed derivation was given by Plümacher *et al.* [29]. In contrast to Plümacher *et al.* [29], where a weak viscosity dependency at the surface boundary was used, this is omitted in this work, to better investigate the effects of mode excitation and modal coupling, as stated in the Introduction.

For an incompressible fluid, the continuity equation is given as

$$\nabla \cdot \mathbf{u} = 0 \quad \text{in } \mathcal{D}(t), \quad (2)$$

and the corresponding Euler momentum equations take the form

$$\mathbf{u}_{,t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\frac{1}{\varrho} \nabla p \quad \text{in } \mathcal{D}(t), \quad (3)$$

where $\mathbf{u}$ is the velocity, p is the pressure field, and ϱ the density. Both equations are defined of the time-dependent droplet domain $\mathcal{D}(t)$, which is depicted in Fig. 1. The momentum jump condition on the droplet surface $\mathcal{S}(t)$, as shown in Fig. 1, can be formulated as

$$-[[p]] \mathbf{n} = \sigma \kappa \mathbf{n} \quad \text{on } \mathcal{S}(t), \quad (4)$$

where σ is the surface tension without the Marangoni effect ($\sigma = \text{const}$), $\kappa = \nabla \cdot \mathbf{n}$ is twice the mean curvature and $\mathbf{n}$ is the normal unit vector of the droplet surface. The pressure jump $[[p]]$ is defined as $[[p]] = p^o - p^i$, where p^o is the pressure outside the droplet and p^i is the pressure inside the droplet. For a dynamically negligible outer medium, p^o is zero. Since p^i is the fluid pressure at the droplet surface and can be replaced by p , Eq. (4) can be reformulated as follows:

$$p = \sigma \kappa \quad \text{on } \mathcal{S}(t). \quad (5)$$

For an inviscid droplet shape oscillation, a potential flow is assumed with $\mathbf{u} = \nabla \phi$, where ϕ is the velocity potential. With this, the continuity equation (2) is reformulated into the Laplace equation for the velocity potential

$$\Delta \phi = 0 \quad \text{in } \mathcal{D}(t), \quad (6)$$

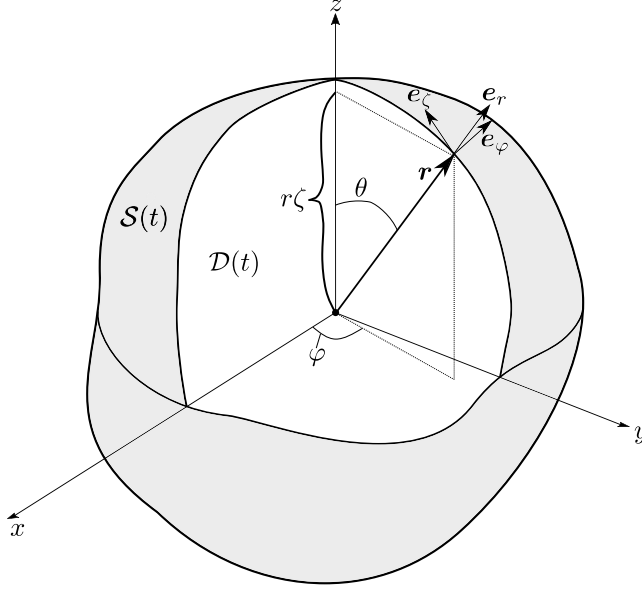


FIG. 1. General droplet shape described in the spherical coordinate system (r, θ, φ) and the considered coordinate system (r, ζ, φ) with the modified polar coordinate $\zeta = \cos(\theta)$ by Plümacher *et al.* [29]. The corresponding local basis vectors are given by $(\mathbf{e}_r, \mathbf{e}_\zeta, \mathbf{e}_\varphi)$ with $\mathbf{e}_\zeta = -\mathbf{e}_\theta$. The droplet surface is denoted by $\mathcal{S}(t)$ and the bulk droplet domain is denoted by $\mathcal{D}(t)$.

and the Euler equations (3) reduce to the Bernoulli equation

$$\phi_{,t} + \frac{1}{2}|\nabla\phi|^2 + \frac{p}{\rho} + C = 0 \quad \text{in } \mathcal{D}(t), \quad (7)$$

with an integration constant C . Substituting Eq. (5) into the Bernoulli equation (7) leads to the modified Bernoulli equation

$$\phi_{,t} + \frac{1}{2}|\nabla\phi|^2 + \frac{\sigma}{\rho}\kappa + C = 0 \quad \text{on } \mathcal{S}(t), \quad (8)$$

which is only valid on the time-dependent droplet surface $\mathcal{S}(t)$. Figure 1 shows a general droplet shape in a spherical coordinate system (r, θ, φ) with the modified polar coordinate $\zeta = \cos(\theta)$ and the azimuthal angle φ . With this coordinate system, the surface of the droplet is explicitly parametrized by

$$\mathbf{r}(\zeta, \varphi, t) = \mathcal{R}(\zeta, \varphi, t)(\sqrt{1 - \zeta^2} \cos(\varphi)\mathbf{e}_x + \sqrt{1 - \zeta^2} \sin(\varphi)\mathbf{e}_y + \zeta\mathbf{e}_z), \quad (9)$$

where $\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$ denote the standard orthogonal Cartesian unit vectors, sharing the same origin as the coordinates (r, ζ, φ) . The corresponding local basis vectors are given by $(\mathbf{e}_r, \mathbf{e}_\zeta, \mathbf{e}_\varphi)$. The function $\mathcal{R}(\zeta, \varphi, t)$, which depends on two spatial variables (ζ, φ) and time t , defines the distance between the coordinate origin and the surface points of the droplet.

The objective is to transform the Laplace equation (6) and the modified Bernoulli equation (8) defined in the time-dependent droplet domain $\mathcal{D}(t)$ and its surface $\mathcal{S}(t)$, respectively, into a system of integrodifferential equations, defined on the unit sphere $\mathbb{S}$. Starting with the Laplace equation (6), which is defined within the droplet domain $\mathcal{D}(t)$, the transformation onto the unit sphere is achieved using the unified transform method. For this purpose, Eq. (6) is rewritten into an equivalent system of equations in divergence form (*local relation*). With Green's second identity, a function ϕ that

TABLE I. Mathematical operators on the unit sphere $\mathbb{S}$.

Induced on the unit sphere $\mathbb{S}$		
Operator	Symbol	Definition (with $\eta = \sqrt{1 - \zeta^2}$)
Gradient	$\nabla_{\mathbb{S}} f$	$\eta f_{,\zeta} \mathbf{e}_{\zeta} + \frac{1}{\eta} f_{,\varphi} \mathbf{e}_{\varphi}$
Laplacian	$\Delta_{\mathbb{S}} f$	$\eta^2 f_{,\zeta\zeta} - 2\zeta f_{,\zeta} + \frac{1}{\eta^2} f_{,\varphi\varphi}$
Hessian	$\mathbf{H}_{\mathbb{S}}^f$	$\begin{pmatrix} \eta^2 f_{,\zeta\zeta} - \zeta f_{,\zeta} & f_{,\zeta\varphi} + \frac{\zeta}{\eta^2} f_{,\varphi} \\ f_{,\zeta\varphi} + \frac{\zeta}{\eta^2} f_{,\varphi} & \frac{1}{\eta^2} f_{,\varphi\varphi} - \zeta f_{,\zeta} \end{pmatrix}$
Integral over unit sphere	$\int_{\mathbb{S}} f d\Omega$	$\int_0^{2\pi} \int_{-1}^1 f d\zeta d\varphi$
Inner product	$\langle f, g \rangle$	$\int_0^{2\pi} \int_{-1}^1 f g d\zeta d\varphi$

satisfies the identity

$$\nabla \cdot (v^{(k)} \nabla \phi - \phi \nabla v^{(k)}) = 0 \quad \text{in } \mathcal{D}(t) \quad (10)$$

is equivalent to satisfying the Laplace equation (6), if for $v^{(k)}$ the set of regular solid harmonics $r^{\ell} Y_{\ell}^m$ is chosen for all $k = (\ell, m)$. The parameter k (*mode number*) is given by the integer pair (ℓ, m) , where ℓ is the *degree* and m is the *order* of the spherical harmonics Y_{ℓ}^m . A detailed overview of the derivation and definition of spherical harmonics can be found in Appendix A. The application of the divergence theorem (Gauss' theorem) transforms the *local relation* (10), defined on the bounded droplet domain $\mathcal{D}(t)$, into the *global relation*

$$0 = \int_{\mathcal{S}(t)} (v^{(k)} \nabla \phi - \phi \nabla v^{(k)}) \cdot \mathbf{n} dA \quad \text{for all } k = (\ell, m), \quad (11)$$

with $\ell \geq 0$, $|m| \leq \ell$. We will briefly show that this integrodifferential equation can be expressed in a form on the unit sphere $\mathbb{S}$. For this purpose, we express the corresponding outward pointing unit normal $\mathbf{n}$ as

$$\mathbf{n} = \frac{N}{|N|} = \frac{\mathbf{e}_r - \nabla_{\mathbb{S}} \mathcal{R}}{\sqrt{1 + \frac{|\nabla_{\mathbb{S}} \mathcal{R}|^2}{\mathcal{R}^2}}}, \quad (12)$$

where $N = \mathbf{e}_r - \nabla_{\mathbb{S}} \mathcal{R}$ is the normal vector and $|N|$ its Euclidean norm. The subscript $\mathbb{S}$ means that the operator is induced on the unit sphere, which is not to be confused with the surface of the droplet $\mathcal{S}(t)$. All relevant mathematical operators with respect to the unit sphere $\mathbb{S}$ are listed in Table I. The surface area element on $\mathcal{S}(t)$ is given by

$$\mathbf{n} dA = \mathcal{R}^2 N d\Omega, \quad (13)$$

where $d\Omega = d\zeta d\varphi$ denotes the element on the unit sphere. Evaluating $v^{(k)}$ and $\nabla v^{(k)}$ on the droplet surface $\mathcal{S}(t)$ leads to

$$v^{(k)} = \mathcal{R}^{\ell} Y_{\ell}^m, \quad \nabla v^{(k)} = \mathcal{R}^{\ell-1} (\ell Y_{\ell}^m \mathbf{e}_r + \nabla_{\mathbb{S}} Y_{\ell}^m). \quad (14)$$

Using the definition of the surface normal N together with the kinematic condition that couples the surface motion to the fluid motion leads to

$$\nabla \phi \cdot N = \frac{\partial \phi}{\partial n} |N| = \mathcal{R}_{,t} \mathbf{e}_r \cdot N = \mathcal{R}_{,t}. \quad (15)$$

Finally, the velocity potential ϕ is evaluated on the droplet surface $\mathcal{R}$, thereby introducing the velocity potential on the surface q as

$$q(\zeta, \varphi, t) = \phi(r, \zeta, \varphi, t)|_{r=\mathcal{R}(\zeta, \varphi, t)} = \phi(\mathcal{R}(\zeta, \varphi, t), \zeta, \varphi, t). \quad (16)$$

Using relations (12)–(16), Plümacher *et al.* [29] reformulated expression (11) into a set of equations depending on q and $\mathcal{R}$, both expressed as functions of ζ and φ . The global relation now reads

$$\mathcal{F}_\ell^m(\mathcal{R}_{,t}, \mathcal{R}, q) := \int_{\mathbb{S}} \mathcal{R}^\ell [(\mathcal{R}^2 \mathcal{R}_{,t} - \ell \mathcal{R} q) Y_\ell^m + q \nabla_{\mathbb{S}} Y_\ell^m \cdot \nabla_{\mathbb{S}} \mathcal{R}] d\Omega = 0 \quad \forall \ell \geq 0, |m| \leq \ell. \quad (17)$$

This set of equations is now defined on the unit sphere $\mathbb{S}$ and comprises a set of infinitely many equations, one for each mode number (ℓ, m) . After deriving the transformed form of the Laplace equation, the next step is to transform the modified Bernoulli equation onto the unit sphere $\mathbb{S}$. Following Ref. [29], each term in Eq. (8) is transformed individually, starting with the temporal term

$$\phi_{,t} = q_{,t} - \frac{\mathcal{R}_{,t}}{\alpha} (\mathcal{R}_{,t} \mathcal{R}^2 + \nabla_{\mathbb{S}} \mathcal{R} \cdot \nabla_{\mathbb{S}} q), \quad (18)$$

with $\alpha = \mathcal{R}^2 + |\nabla_{\mathbb{S}} \mathcal{R}|^2$. The convective term is given by

$$\frac{1}{2} |\nabla \phi|^2 = \frac{1}{2} \left(\frac{1}{\alpha^2} [\alpha \mathcal{R}^2 \mathcal{R}_{,t}^2 + (\nabla_{\mathbb{S}} \mathcal{R} \cdot \nabla_{\mathbb{S}} q)^2 + \mathcal{R}^2 |\nabla_{\mathbb{S}} q|^2 + (\nabla_{\mathbb{S}} q \times \nabla_{\mathbb{S}} \mathcal{R})^2 (1 + |N|^2)] \right), \quad (19)$$

with

$$|N| = \sqrt{1 + \frac{|\nabla_{\mathbb{S}} \mathcal{R}|^2}{\mathcal{R}^2}}. \quad (20)$$

Twice the mean curvature κ is expressed as

$$\kappa = \frac{|N|}{\alpha^2} \lambda \quad \text{and} \quad \lambda = [\nabla_{\mathbb{S}} \mathcal{R} \cdot (\mathbf{H}_{\mathbb{S}}^{\mathcal{R}} \cdot \nabla_{\mathbb{S}} \mathcal{R}) - \alpha \Delta_{\mathbb{S}} \mathcal{R} + 3\mathcal{R} |\nabla_{\mathbb{S}} \mathcal{R}|^2 + 2\mathcal{R}^3], \quad (21)$$

where the Hessian $\mathbf{H}_{\mathbb{S}}^{\mathcal{R}}$, as well as $\Delta_{\mathbb{S}} f$ are defined in Table I. Substituting the above relations into Eq. (8), Plümacher *et al.* [29] obtained the final form of the modified Bernoulli equation expressed in terms of $\mathcal{R}$ and q :

$$\begin{aligned} G(\mathcal{R}_{,t}, q_{,t}, \mathcal{R}, q) := & q_{,t} - \frac{\mathcal{R}_{,t}}{\alpha} \left[\frac{\mathcal{R}^2 \mathcal{R}_{,t}}{2} + \nabla_{\mathbb{S}} \mathcal{R} \cdot \nabla_{\mathbb{S}} q \right] + \frac{\sigma}{\varrho} \kappa - C + \frac{1}{2\alpha^2} \left[(\nabla_{\mathbb{S}} \mathcal{R} \cdot \nabla_{\mathbb{S}} q)^2 \right. \\ & \left. + \mathcal{R}^2 |\nabla_{\mathbb{S}} q|^2 + (\nabla_{\mathbb{S}} q \times \nabla_{\mathbb{S}} \mathcal{R})^2 \left(\frac{|\nabla_{\mathbb{S}} \mathcal{R}|^2}{\mathcal{R}^2} + 2 \right) \right] = 0, \quad \text{on } \mathbb{S}. \end{aligned} \quad (22)$$

Both Eqs. (17) and (22) are the reduced formulation of the initial problem of an oscillating droplet shape in terms of the two unknowns q and $\mathcal{R}$ defined on the unit sphere $\mathbb{S}$ with the spatial variables ζ and φ . The integration constant C is defined as $C = \frac{2\sigma}{\varrho \mathcal{L}}$, following the approach of Plümacher *et al.* [29], where $\mathcal{L}$ denotes the characteristic length scale of the droplet and is chosen as the zeroth-order spherical harmonic amplitude. The governing equations are written in nondimensional form using the dimensionless variables

$$\tilde{\mathcal{R}} = \frac{\mathcal{R}}{\mathcal{L}}, \quad \tilde{q} = \frac{q T}{\mathcal{L}^2}, \quad \tilde{\kappa} = \kappa \mathcal{L}, \quad \tilde{C} = \frac{C T^2}{\mathcal{L}^2} = 2 \quad \text{and} \quad \tilde{t} = \frac{t}{T}, \quad (23)$$

with $T = \sqrt{\varrho \mathcal{L}^3 / \sigma}$. All surface differential operators are defined on the unit sphere $\mathbb{S}$ and are therefore dimensionless. As a result, the dimensionless operators are the same as their dimensional ones $\tilde{\nabla}_{\mathbb{S}} = \nabla_{\mathbb{S}}$, $\tilde{\Delta}_{\mathbb{S}} = \Delta_{\mathbb{S}}$ and $\tilde{H}_{\mathbb{S}}^{\mathcal{R}} = H_{\mathbb{S}}^{\mathcal{R}}$. After nondimensionalization, the governing equations retain

the same structure but are expressed in terms of the dimensionless variables, leading to the following two equations (24) and (25):

$$\tilde{\mathcal{F}}_\ell^m(\tilde{\mathcal{R}}_{,t}, \tilde{\mathcal{R}}, \tilde{q}) := \int_{\mathbb{S}} \tilde{\mathcal{R}}^\ell [(\tilde{\mathcal{R}}^2 \tilde{\mathcal{R}}_{,t} - \ell \tilde{\mathcal{R}} \tilde{q}) Y_\ell^m + \tilde{q} \tilde{\nabla}_{\mathbb{S}} Y_\ell^m \cdot \tilde{\nabla}_{\mathbb{S}} \tilde{\mathcal{R}}] d\Omega = 0 \quad \forall \ell \geq 0, |m| \leq \ell, \quad (24)$$

$$\begin{aligned} \tilde{G}(\tilde{\mathcal{R}}_{,t}, \tilde{q}_{,t}, \tilde{\mathcal{R}}, \tilde{q}) := & \tilde{q}_{,t} - \frac{\tilde{\mathcal{R}}_{,t}}{\tilde{\alpha}} \left[\frac{\tilde{\mathcal{R}}^2 \tilde{\mathcal{R}}_{,t}}{2} + \tilde{\nabla}_{\mathbb{S}} \tilde{\mathcal{R}} \cdot \tilde{\nabla}_{\mathbb{S}} \tilde{q} \right] + \tilde{\kappa} - \tilde{C} + \frac{1}{2\tilde{\alpha}^2} \left[(\tilde{\nabla}_{\mathbb{S}} \tilde{\mathcal{R}} \cdot \tilde{\nabla}_{\mathbb{S}} \tilde{q})^2 \right. \\ & \left. + \tilde{\mathcal{R}}^2 |\tilde{\nabla}_{\mathbb{S}} \tilde{q}|^2 + (\tilde{\nabla}_{\mathbb{S}} \tilde{q} \times \tilde{\nabla}_{\mathbb{S}} \tilde{\mathcal{R}})^2 \left(\frac{|\tilde{\nabla}_{\mathbb{S}} \tilde{\mathcal{R}}|^2}{\tilde{\mathcal{R}}^2} + 2 \right) \right] = 0, \quad \text{on } \mathbb{S}. \end{aligned} \quad (25)$$

For simplicity, the tilde notation is omitted in the following and all variables are considered dimensionless.

III. NUMERICAL PROCEDURE

The system of equations (24) and (25) is numerically solved with a highly specialized code using the Galerkin method in a space spanned by spherical harmonics. The principal idea is to approximate the unknown variables $\mathcal{R}$ and q by a truncated series of basis functions [34]. Since spherical harmonics Y_ℓ^m have already been chosen to express the system of equations (24), it is a straightforward choice to use them also as the basis to represent the two unknowns $\mathcal{R}$ and q , i.e.,

$$\begin{aligned} \mathcal{R}(\zeta, \varphi, t) &\approx \mathcal{R}_L(\zeta, \varphi, t) = \sum_{\ell=0}^L \sum_{m=-\ell}^{\ell} a_\ell^m(t) Y_\ell^m(\zeta, \varphi), \\ q(\zeta, \varphi, t) &\approx q_L(\zeta, \varphi, t) = \sum_{\ell=0}^L \sum_{m=-\ell}^{\ell} b_\ell^m(t) Y_\ell^m(\zeta, \varphi), \end{aligned} \quad (26)$$

where L is the truncation degree. As we have currently limited our solver to the axisymmetric case, the coordinate φ does not appear. Therefore, the *order* m drops out and the ansatz reduces to

$$\begin{aligned} \mathcal{R}(\zeta, t) &\approx \mathcal{R}_L(\zeta, t) = \sum_{\ell=0}^L a_\ell(t) Y_\ell(\zeta), \\ q(\zeta, t) &\approx q_L(\zeta, t) = \sum_{\ell=0}^L b_\ell(t) Y_\ell(\zeta). \end{aligned} \quad (27)$$

To obtain a compact notation, the time-dependent coefficients $a_\ell(t)$ and $b_\ell(t)$ are combined in a degrees-of-freedom vector $\mathbf{x}(t)$, i.e.,

$$\mathbf{x}(t) := (a_0(t), \dots, a_L(t), b_0(t), \dots, b_L(t)). \quad (28)$$

Note that Eq. (24) is already a weak formulation. Therefore, ansatz (26) can be substituted into the left-hand side of Eq. (24) to obtain a semidiscrete form (discrete in space, continuous in time)

$$F_\ell(\dot{\mathbf{x}}(t), \mathbf{x}(t)) := \mathcal{F}_\ell(\partial_t \mathcal{R}_L, \mathcal{R}_L, q_L) = 0 \quad \text{for} \quad 0 \leq \ell \leq L. \quad (29)$$

In contrast, Eq. (25) is a strong formulation. A semidiscrete, weak form is obtained by substituting ansatz (26) into the left-hand side of Eq. (25) and multiplying it with the test functions Y_ℓ . To ensure manageable, integrable functions, this expression is multiplied by $\mathcal{R}$ before integration over the unit

sphere $\mathbb{S}$. Thereby, one obtains the semidiscrete form

$$G_\ell(\dot{\mathbf{x}}(t), \mathbf{x}(t)) := \int_{\mathbb{S}} \mathcal{R} G(\partial_t \mathcal{R}_L, \partial_t q_L, \mathcal{R}_L, q_L) Y_\ell d\Omega = 0 \quad \text{for } 0 \leq \ell \leq L. \quad (30)$$

Note that the semidiscrete, highly nonlinear system of equations (29) and (30) contains $2(L+1)$ equations with $2(L+1)$ unknown coefficients $a_\ell(t)$ and $b_\ell(t)$. It can be summarized as

$$\mathbf{H}_L(\dot{\mathbf{x}}, \mathbf{x}) := (F_0(\dot{\mathbf{x}}, \mathbf{x}), \dots, F_L(\dot{\mathbf{x}}, \mathbf{x}), G_0(\dot{\mathbf{x}}, \mathbf{x}), \dots, G_L(\dot{\mathbf{x}}, \mathbf{x}))^T = 0. \quad (31)$$

For numerical simulations, the time derivatives are approximated using the second-order backward differentiation formula (BDF2), $\dot{\mathbf{x}}(t) \approx (3\mathbf{x}^{(i)} - 4\mathbf{x}^{(i-1)} + \mathbf{x}^{(i-2)})/(2\Delta t)$, where the superscript $(\dots)^{(i)}$ denotes the time step i . For the first time step, the first-order backward differentiation formula (BDF1) with a much smaller step size of $0.01\Delta t$ is applied to reduce the start-up error of the lower-order scheme. The subsequent time step lengths are doubled at each time step in a start-up sequence using a BDF2 formula with a variable time step by Hairer *et al.* [35] until the target time step Δt length is reached. To solve this system of equations, the initial values of the unknowns, $\mathbf{x}^{(0)}$, must be established. The initial shape and the velocity potential on the surface are thus necessarily given, leading to a fully discrete system of time-discretized equations (denoted by the calligraphy $\mathcal{H}$)

$$\mathcal{H}(\mathbf{x}^{(i)}) := \mathbf{H}_L\left(\frac{3\mathbf{x}^{(i)} - 4\mathbf{x}^{(i-1)} + \mathbf{x}^{(i-2)}}{2\Delta t}, \mathbf{x}^{(i)}\right) = 0. \quad (32)$$

To find the numerical solution $\mathbf{x}^{(i)}$, Newton's method is applied with ψ as the tolerance. The integration is performed numerically using the computer algebra system Maple. When the tolerance is reached, the approximate solution of Eq. (32) is obtained. To assure numerical convergence of Eq. (32), the initial estimation $\mathbf{x}_0^{(i)}$ must be chosen as close as possible to the exact solution. A well-estimated initial value can save a large number of Newton steps and computation time. If the time step Δt is sufficiently small, then a well-estimated initial value for each time step (i) can be found by a linear extrapolation from the last two time steps $\mathbf{x}_0^{(i)} = 2\mathbf{x}^{(i-1)} - \mathbf{x}^{(i-2)}$. However, for the first time step, the initial state is given as $\mathbf{x}_0^{(1)} = \mathbf{x}^{(0)}$. For Newton's method, the following equation has to be solved for each time step:

$$[\partial \mathcal{H}(\mathbf{x}_k^{(i)})] \delta_k^{(i)} = -\mathcal{H}(\mathbf{x}_k^{(i)}), \quad (33)$$

where $\partial \mathcal{H}(\mathbf{x}_k^{(i)})$ denotes the Jacobian matrix and $\delta_k^{(i)}$ is the correction value after the Newton step k . The derivatives in the Jacobian matrix are approximated with FDM. To ensure that the initial guess is close to the radius of convergence, the Armijo condition (see Ref. [36]) is introduced

$$|\mathcal{H}(\mathbf{x}_{k+1}^{(i)})|_2^2 < (1 - 0.002 \sigma_p) |\mathcal{H}(\mathbf{x}_k^{(i)})|_2^2, \quad (34)$$

with

$$\mathbf{x}_{k+1}^{(i)} = \mathbf{x}_k^{(i)} + \sigma_p \delta_k^{(i)}, \quad (35)$$

and $\sigma_p = 1/2^{p-1}$ the damping term vector and $|\dots|_2^2$ the square of the L_2 -norm. If the Armijo condition is not satisfied, then the initial guess is not within the radius of convergence of the Newton method. Then, the index p increases by one, so the correction value $\delta_k^{(i)}$ is further attenuated by σ_{p+1} . This procedure is repeated until the Armijo condition is satisfied. Thus, a Newton step is performed and the next Newton step is calculated in the same way. When the tolerance of the Newton procedure ψ is reached, the time step (i) is performed. The initial values are specified by

$$\mathcal{R}_L(\zeta, 0) = 1 + a_\ell(0) Y_\ell(\zeta) \quad \ell = 2, \dots, 6, \quad q_L(\zeta, 0) = 0. \quad (36)$$

The first condition of Eq. (36) represents an initial deviation of the ℓ -th order spherical harmonics Y_ℓ with an amplitude $a_\ell(0)$ with $\ell = 2, 3, 4, 5$ or 6 from their unit spherical shape (the zeroth-order

TABLE II. Simulation parameters.

Name	Symbol	Value
Time step (nondimensionalized)	Δt	0.0018
Truncation degree	L	12
Tolerance of Newton's method	ψ	10^{-9}

spherical harmonic with $\ell = 0$). The mode $\ell = 1$, representing a rigid translation of the droplet, is not taken into account in the initial shape deformation. The second condition of Eq. (36), a zero initial velocity potential at the surface of the droplet, implies a zero initial velocity throughout the droplet. This can be observed by determining the kinetic energy of the entire droplet. The total kinetic energy is defined as

$$E_{\text{kin}}(t) = \frac{\rho}{2} \int_{\mathcal{D}(t)} |\mathbf{u}|^2 \, d\mathbf{v}. \quad (37)$$

Using the identity for the potential flow assumption,

$$\nabla \cdot (\phi \mathbf{u}) = \nabla \cdot (\phi \nabla \phi) = (\nabla \phi)^2 + \underbrace{\phi \Delta \phi}_{=0} = |\mathbf{u}|^2, \quad (38)$$

and inserting the expressions ϕ and $\nabla \phi$ from Plümacher *et al.* [29] into Eq. (37), we obtain the kinetic energy

$$E_{\text{kin}}(t) = \frac{\rho}{2} \int_{\mathcal{S}} q \mathcal{R}^2 \mathcal{R}_{,t} \, d\Omega, \quad (39)$$

expressed by the surface quantities $\mathcal{R}(\zeta, t)$ and $q(\zeta, t)$. This shows that a vanishing initial velocity potential at the surface of the droplet, $q = 0$, corresponds to a vanishing total initial kinetic energy, $E_{\text{kin}} = 0$, i.e., a vanishing initial velocity throughout the entire droplet.

The selected numerical parameters for the time step Δt , the truncation degree L , and the tolerance of Newton's method are shown in Table II. These parameters have been chosen to ensure sufficiently accurate numerical results. The choice of L and Δt can be verified, for example, by temporal and spatial convergence studies, as illustrated in Fig. 2 with the relative error

$$\mathcal{E}_{L,\Delta t} = \sqrt{\frac{1}{N} \sum_i^N \frac{\int_{\mathcal{S}} (\mathcal{R}(\zeta, \varphi, t_i) - \mathcal{R}_{\text{ref}}(\zeta, \varphi, t_i))^2 \, d\Omega}{\int_{\mathcal{S}} (\mathcal{R}_{\text{ref}}(\zeta, \varphi, t_i))^2 \, d\Omega}} 100\%, \quad (40)$$

where $\mathcal{R}_{\text{ref}}(\zeta, \varphi, t_i)$ is the reference solution. Figure 2(a) presents the relative errors with different truncation degrees L but a fine time step $\Delta t = 0.0018$, denoted by $\mathcal{E}_L$, in which the case $L = 14$ is used as the reference solution $\mathcal{R}_{\text{ref}}(\zeta, \varphi, t_i)$. Figure 2(b) shows the relative errors with different time steps Δt but a large cutoff degree of $L = 12$, denoted by $\mathcal{E}_{\Delta t}$, in which the case $\Delta t = 0.0009$ is used as the reference solution $\mathcal{R}_{\text{ref}}(\zeta, \varphi, t_i)$. For both cases, the initial deformation mode $\ell = 2$ with different amplitudes $a_2(0) = 0.4, 0.5, 0.6, 0.7$ is considered and a clear convergence behavior is observed. The spatial convergence exhibits an exponential rate of $\mathcal{O}(e^{-0.48L})$, while the temporal convergence shows an order of $\mathcal{O}(\Delta t^{1.5})$ below two. These are consistent with the expected behaviors of a spectral method and a BDF2 scheme, but a BDF1 for the first time step. These convergence studies justify our choice of $L = 12$ and $\Delta t = 0.0018$, which ensure a relative numerical error of $\mathcal{E}_{L,\Delta t} < 0.0001\%$. Additionally, the validity of these choices is confirmed below by examining the conservation of volume and energy of the oscillating droplet.

Figure 3(a) displays the relative volume deviation $\mathcal{E}_V(t)$ over two oscillation periods for different initial deformation modes $\ell = 2, 3, 4, 5, 6$ and distinct amplitudes $a_2(0) = 0.8$, $a_3(0) = 0.5$, $a_4(0) = 0.3$, $a_5(0) = 0.2$, $a_6(0) = 0.1$, respectively. Figure 3(b) shows the maximum values

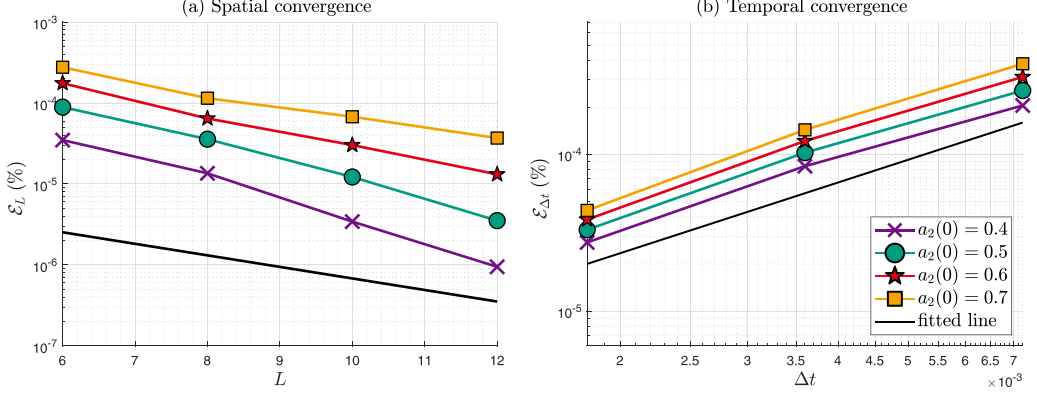


FIG. 2. Spatial and temporal convergence plots for an initial deformation mode $\ell = 2$ with different amplitudes $a_2(0) = 0.4, 0.5, 0.6, 0.7$. (a) Spatial convergence for the relative spatial error $\mathcal{E}_L$ with a fixed time step of $\Delta t = 0.0018$. The highest truncation degree $L = 14$ has been used as the reference solution. The black line indicates a fitted exponential decay of $O(e^{-0.48L})$. (b) Temporal convergence with the relative temporal error $\mathcal{E}_{\Delta t}$ with a fixed truncation degree of $L = 12$. The finest time step $\Delta t = 0.0009$ has been used for the reference solution. The black line indicates a fitted convergence of $O(\Delta t^{1.5})$.

$\mathcal{E}_{V,\max}$ of the relative volume deviation within the first two oscillation periods for different amplitudes. The volume loss due to numerical errors is less than 0.0005% in all simulations, i.e., extremely small.

Observing the energy conservation is another method to estimate the accuracy of the numerical procedure. Under the potential flow assumption, the total energy $E_{\text{total}}(t)$ should remain constant. This total energy consists of the potential energy $E_{\text{pot}}(t)$ and the kinetic energy $E_{\text{kin}}(t)$. The formula for the usable potential energy is $E_{\text{pot}}(t) = (A(t) - A_{\text{sph}}) \sigma$, where

$$A(t) = 2\pi \int_{-1}^1 \mathcal{R}^2 \sqrt{1 + \frac{1}{\mathcal{R}^2} \left(\frac{\partial \mathcal{R}}{\partial \zeta} \right)^2 (1 - \zeta^2)} d\zeta \quad (41)$$

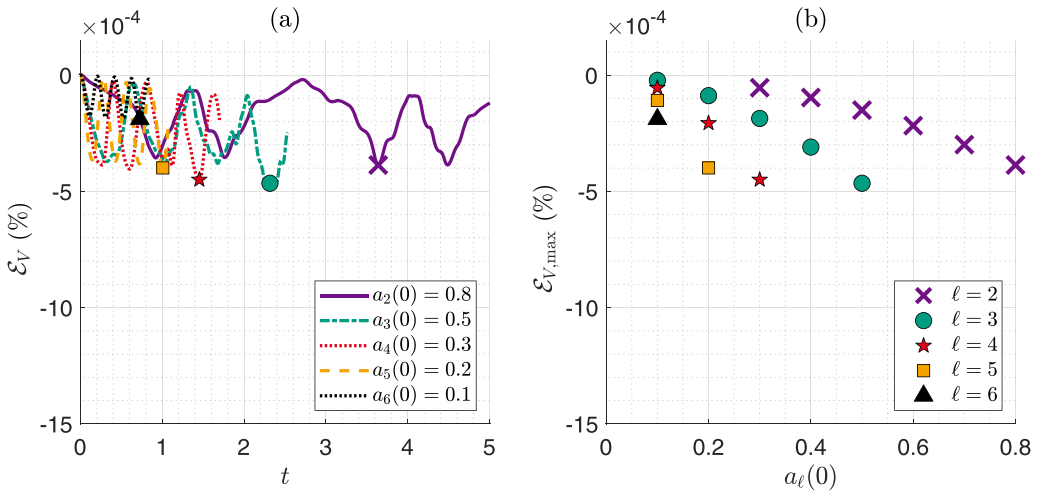


FIG. 3. (a) Relative volume deviation $\mathcal{E}_V(t) = \frac{V(t) - V(0)}{V(0)} 100\%$ for initial deformation modes $\ell = 2, \dots, 6$ with the respective initial amplitudes $a_2(0) = 0.8, a_3(0) = 0.5, a_4(0) = 0.3, a_5(0) = 0.2, a_6(0) = 0.1$ and (b) the maximum deviation $\mathcal{E}_{V,\max}$ within the first two oscillation periods.

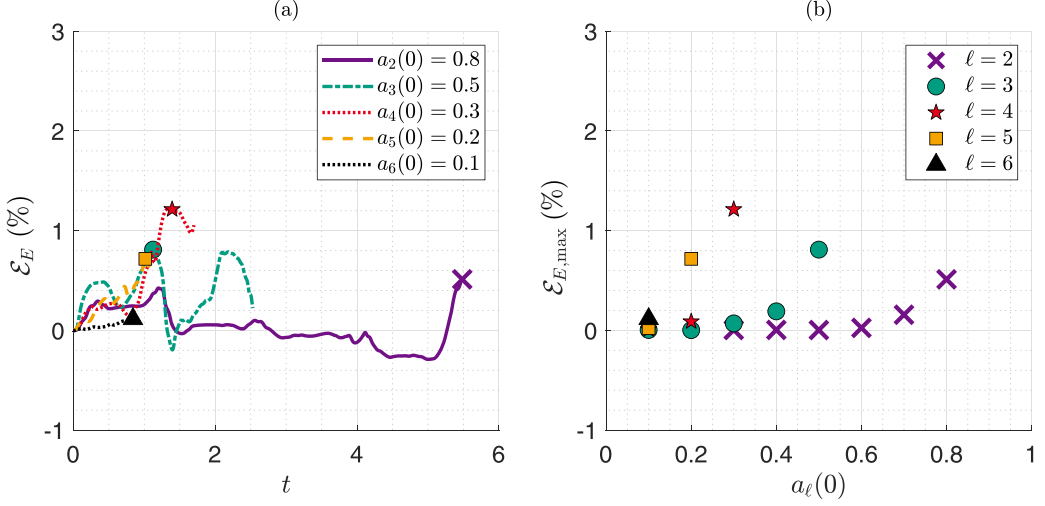


FIG. 4. (a) Relative deviation in total energy $\mathcal{E}_E(t)$ for initial deformation modes $\ell = 2, \dots, 6$ with the respective initial amplitudes $a_2(0) = 0.8$, $a_3(0) = 0.5$, $a_4(0) = 0.3$, $a_5(0) = 0.2$, $a_6(0) = 0.1$ and (b) the maximum deviation $\mathcal{E}_{E,\max}$ within the first two oscillation periods.

is the surface area of the oscillating droplet and A_{sph} is the surface area of its undeformed spherical shape having the same volume as the deformed droplet. Figure 4 shows the relative deviation of the total energy

$$\mathcal{E}_E(t) = \frac{E_{\text{pot}}(t) + E_{\text{kin}}(t) - E_{\text{pot}}(0)}{E_{\text{pot}}(0)} 100\% \quad (42)$$

for the initial deformation modes $\ell = 2, 3, 4, 5, 6$, as given in Eq. (36). In Fig. 4(a), the energy deviations over time t are displayed over two oscillation periods and in Fig. 4(b), their maximum energy deviation $\mathcal{E}_{E,\max}$ over different amplitudes. All maximum deviations are within 2%, which is sufficiently precise for our analysis.

IV. RESULTS AND DISCUSSION

In this section, axisymmetric droplet shape oscillations are presented up to the sixth initial deformation mode ($\ell = 6$). The motion of the droplet's north pole is compared with the existing literature. Timescales for the shape oscillations are also presented, with detailed analyses of the frequency shifts and the extended time in states of elongation. The results available in the literature were successfully validated and further extended by considering initial deformations ($\ell = 5$) and ($\ell = 6$).

Furthermore, the modal coupling of droplet shape oscillations and their respective power spectra are analyzed for superimposed initial deformations. To the best of the authors' knowledge, this is the first time that a large-amplitude even mode ($\ell = 2$) has been superimposed with a small-amplitude odd mode ($\ell = 3$ or $\ell = 5$).

A. Droplet shape oscillations

The shape oscillations with relatively large amplitudes for various initial deformation modes ℓ are displayed in Figs. 5–9, for the first two oscillation periods with the initial shape shown in Figs. 5(a), 6(a), 7(a), 8(a), and 9(a). Strongly nonlinear effects can be observed. First, for each single initial deformation mode ℓ , different additional modes are excited. The number of bulges present indicates the active modes. For example, in Figs. 5(e), 5(g), 5(h), 5(n),..., the droplet displays more

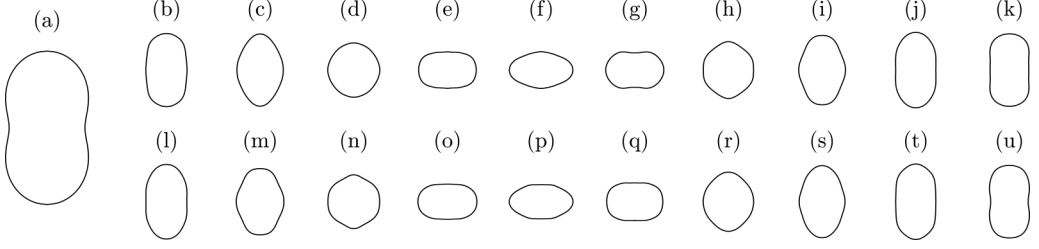


FIG. 5. Shape oscillations for the initial deformation mode $\ell = 2$ with amplitude $a_2(0) = 0.8$ at 21 time instants with equal time intervals $\Delta t = T/10$ with the period T in the first two oscillation periods. Panel (a) shows the initial configuration. Panels (b)–(k) in the upper row depict the first oscillation period and panels (l)–(u) in the lower row depict the second oscillation period. The oscillation video can be viewed as Supplemental Material [37].

than two bulges, indicating that modes higher than $\ell = 2$ are excited for an initial deformation mode $\ell = 2$. Nonlinear effects can also be confirmed by the nonperiodicity (or quasiperiodicity) of the droplet shape oscillations, showing that each following oscillation deviates slightly from the previous one. This nonperiodicity arises due to the excitation of various modes with their distinct frequencies. This can be identified in Figs. 5 and 6 by comparing the droplet shapes between the first and second oscillation periods, i.e., by comparing the columns of the first and second row vertically. For example, in Figs. 6(i) and 6(s), the droplet shapes are quite different, indicating the quasiperiodicity. Another clearly observable effect is that an initially symmetric droplet with respect to the horizontal symmetry plane, i.e., an even initial deformation mode, maintains its symmetry over all the time, as shown in Figs. 5, 7, and 9. This is due to the fact that an even initial deformation mode excites only even modes, as also noted by Ref. [26]. The videos of all shape oscillations can be viewed as Supplemental Material [37].

B. Droplet's north pole oscillation

In droplet dynamics, the relative motion of the droplet's north pole with respect to the droplet center serves as an important reference for observing and analyzing nonlinear effects. The relative position of the droplet's north pole is defined by

$$\mathcal{N}(t) = \frac{\mathcal{R}^N(t) - \mathcal{R}_{\text{Sph}}}{\mathcal{R}_{\text{Sph}}}. \quad (43)$$

The north pole position is denoted as $\mathcal{R}^N(t) = \mathcal{R}(\zeta = 1, t)$, where $\zeta = \cos(\theta) = 1$ corresponds to the north pole at $\theta = 0$. The radius of an undeformed, i.e., spherical droplet, having the same volume as the investigated deformed droplet is denoted with $\mathcal{R}_{\text{Sph}}$.

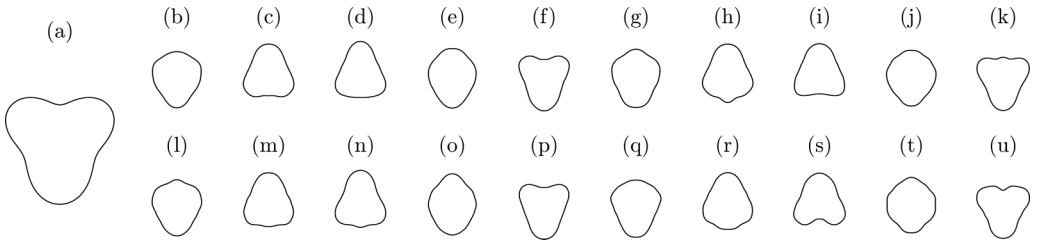


FIG. 6. Shape oscillations as in Fig. 5, but for the initial deformation mode $\ell = 3$ with amplitude $a_3(0) = 0.5$. The oscillation video can be viewed as Supplemental Material [37].

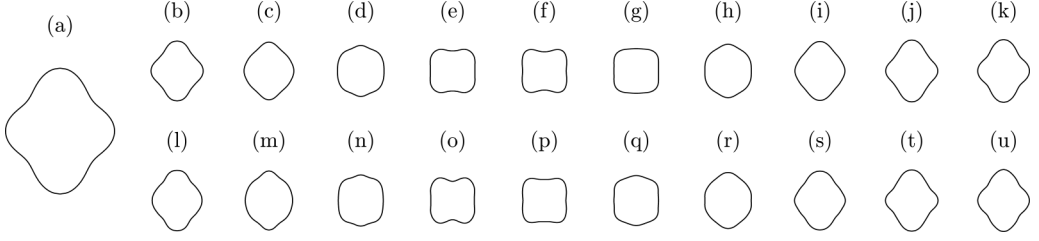


FIG. 7. Shape oscillations as in Fig. 5, but for the initial deformation mode $\ell = 4$ with amplitude $a_4(0) = 0.3$. The oscillation video can be viewed as Supplemental Material [37].

In Fig. 10, the relative position of the droplet's north pole for the initial deformation mode ($\ell = 4$) is compared with two other studies with individual values of amplitude $a_4(0)$. Zrnić and Brenn [26] applied a weakly nonlinear theory to reach the approximate solution up to the third order for inviscid droplets, while Lundgren and Mansour [20] calculated the shape oscillations numerically using a boundary-integral method for slightly viscous droplets. The results from the present GNLT show very good agreement with the literature for small and medium initial amplitudes.

Several essentially nonlinear effects can be observed from Fig. 10. First, the relative position $\mathcal{N}(t)$ of the north pole for the initial deformation mode ($\ell = 4$) is an overlap of various excited even modes of different frequencies with a base frequency f_4 , modified from its corresponding Rayleigh frequency f_4^R due to finite oscillation amplitudes. It is noticeable that every third peak of $\mathcal{N}(t)$ is slightly lower than the previous two. A closer observation of Eq. (1) reveals that the frequency f_4^R is three times that of f_2^R . Hence, the lower third peak in $\mathcal{N}(t)$ should be the result of the superposition of the oscillations with the two different frequencies f_4 and f_2 .

Second, the oscillations persist longer in the elongated, prolate form (τ_4^P defined in Fig. 10) than in the oblate shape. This effect increases with increasing amplitude. Third, the oscillation period τ_4 increases with larger amplitudes. The reason for the last two phenomena is the different pressure distribution at the poles and at the equator of the droplet during the shape oscillation. This pressure difference due to the different surface curvatures is the driving force for the droplet shape oscillation. In the prolate state, the curvature of the droplet is greatest at its poles, while in the oblate phase, it is greatest at the equator. Because the poles in the prolate phase cover a smaller area than the equator in the oblate phase, the restoring force in the prolate phase of the oscillation is weaker. Furthermore, the rebound of the droplet in the prolate state is hindered and slowed by the strong action of surface tension at the equator [17]. As a result, the droplet spends more time in the prolate form, the period length increases, and consequently the oscillation frequency decreases.

C. Timescales of the drop shape oscillations

The last two phenomena described previously, i.e., the frequency shift and the extended time in the prolate state, are analyzed in detail in this subsection. For this purpose, we compare the

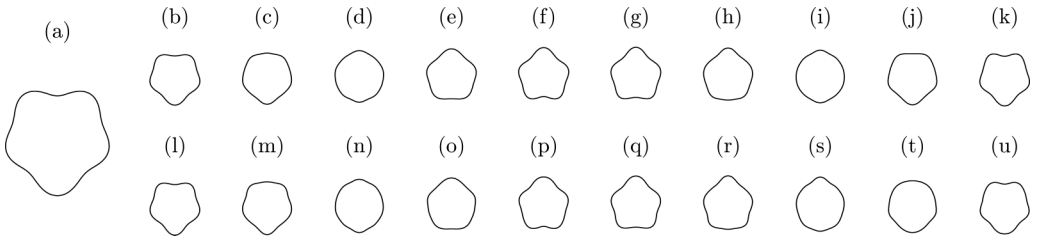


FIG. 8. Shape oscillations as in Fig. 5, but for the initial deformation mode $\ell = 5$ with amplitude $a_5(0) = 0.2$. The oscillation video can be viewed as Supplemental Material [37].

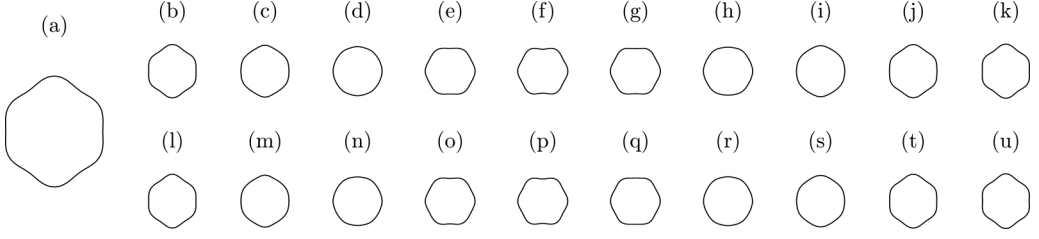


FIG. 9. Shape oscillations as in Fig. 5, but for the initial deformation mode $\ell = 6$ with amplitude $a_6(0) = 0.1$. The oscillation video can be viewed as Supplemental Material [37].

results of the GNLT with the literature. The first experimental study of large-amplitude droplet shape oscillations, conducted by Trinh and Wang [15], investigated these oscillations in an immiscible hot liquid. Foote [12] performed one of the first numerical studies based on an extension of the Marker-and-Cell method for an Ohnesorge number of 0.028. Meradji *et al.* [24] and Basaran [23] numerically analyzed droplet shape oscillations with a small viscosity (Ohnesorge number = 0.01), while the numerical study by Patzek *et al.* [22] was based on an inviscid droplet shape oscillation. Theoretical results using a weakly nonlinear analysis were performed by Tsamopoulos and Brown [16] and Zrnić and Brenn [26] for inviscid droplet shape oscillations.

All these results are displayed in Fig. 11 for comparison. Figure 11(a) shows the relative decrease in angular frequency with increasing initial aspect ratio compared to the Rayleigh oscillation frequency, while Fig. 11(b) illustrates the percentage of the time that the droplet spends in the prolate form compared to the oscillation period. Both values are displayed as functions of the initial droplet aspect ratio L/W (with length L and width W) for an initial deformation mode $\ell = 2$. The aspect ratio is directly related to the amplitude $a_2(0)$. A large amplitude of $a_2(0)$ corresponds to a large aspect ratio. Since two oscillation periods are investigated and the first and second period are not fully identical, a mean value for the two oscillation periods is taken for the present GNLT. The results of the GNLT agree qualitatively well with the literature, although rather different methods

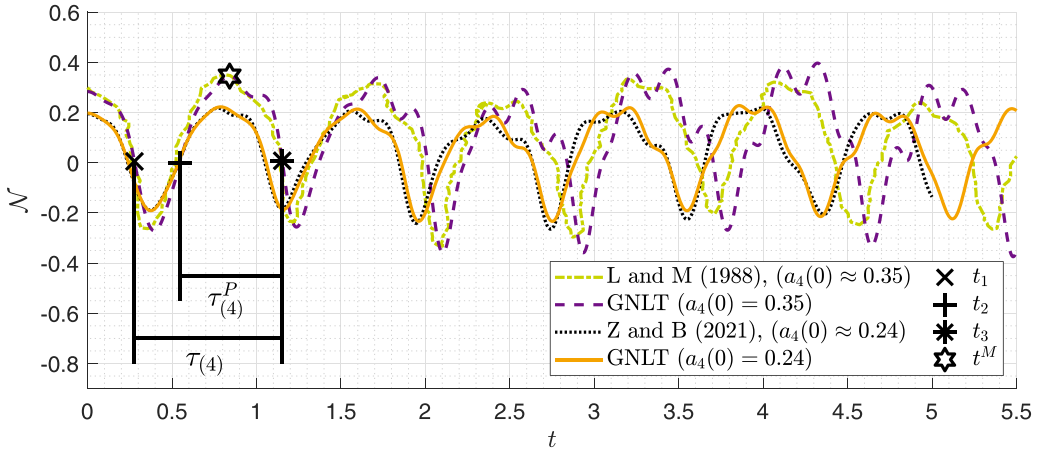


FIG. 10. Relative position N of the droplet's north pole as a function of time for the initial deformation mode $\ell = 4$ and comparison with the results from Zrnić and Brenn [26] [abbreviated as Z and B (2021)] and Lundgren and Mansour [20] [L and M (1988)]. The timestamps t_i ($i = 1, 2, 3$) denote the first three zero crossings of N . The period length of N is measured with $\tau_4 = t_3 - t_1$ and the time in the elongated, prolate form (positive half-period) is measured with $\tau_4^P = t_3 - t_2$. The timestamp of the first local maximum (subscript M) is defined as t^M .

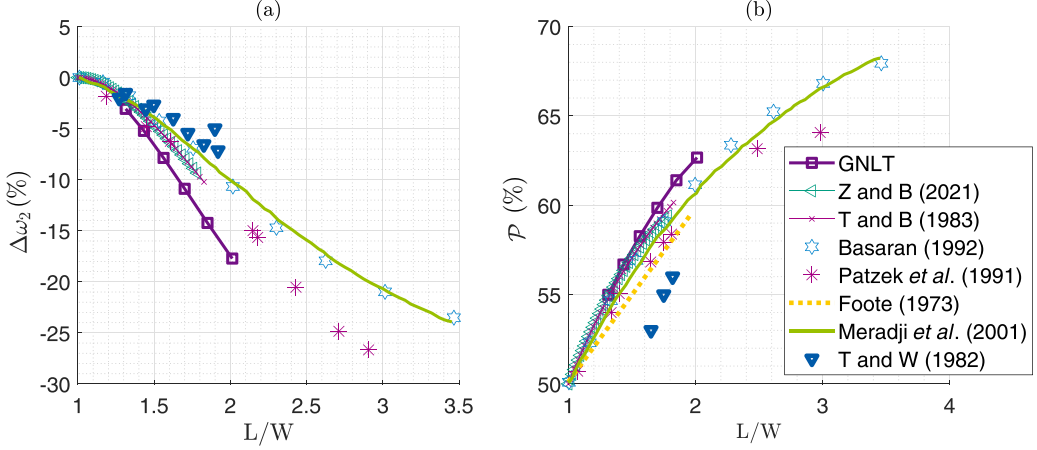


FIG. 11. (a) Relative frequency shift $\Delta\omega_\ell = \frac{\omega_\ell - \omega_\ell^R}{\omega_\ell^R} 100\%$, for $\ell = 2$ with ω_ℓ^R as the Rayleigh angular frequency and ω_ℓ as the frequency of the current oscillation. (b) Relative time in the elongated, prolate form $P = (\tau_\ell^P / \tau_\ell) 100\%$, for $\ell = 2$ and with τ_ℓ^P as the time in the prolate form and τ_ℓ as the period length. Zrnić and Brenn [26] is abbreviated with Z and B (2021). Tsamopoulos and Brown [16] is abbreviated with T and B (1983), Trinh and Wang [15] with T and W (1982).

were employed. As expected, the differences are also clearly recognisable, as different conditions were used.

For small-to-moderate aspect ratios (up to $L/W = 1.7$), all results are similar and the present GNLT results are close to the weakly nonlinear theories of Zrnić and Brenn [26] and Tsamopoulos and Brown [16]. A larger deviation occurs for large aspect ratios, as their analyses are only applicable for relatively small aspect ratios. In other results, droplet's viscosity plays an important role, then deviations are more obvious.

Figure 12 extends the GNLT results shown in Fig. 11 from the initial deformation mode $\ell = 2$ to higher initial deformation modes $\ell = 3, 4, 5, 6$ with different initial deformation amplitudes $a_\ell(0)$, respectively. In Fig. 11(a), the relative frequency shift, and in Fig. 11(b), the prolate times are displayed over the amplitude. For higher initial deformation modes with the same initial deformation amplitude, the asymmetry of the prolate and oblate oscillation and the frequency shift become more obvious. These results are in good agreement with the findings of the WNLT by Zrnić and Brenn [26], which have been multiplied with the appropriate factor (A14), for lower-order initial deformation modes and small to moderate amplitudes, as indicated in Fig. 12 as well. The GNLT extend their results to higher-order initial deformation modes $\ell = 5$ and 6 and larger amplitudes.

D. Modal coupling and power spectrum of a large-amplitude even mode superimposed with a small-amplitude odd mode

In the GNLT, the time-dependent coefficients $a_\ell(t)$, $b_\ell(t)$, which correspond to some extent to the amplitudes of the mode ℓ , are calculated explicitly, making clearly identifiable which modes are excited and which are not. Given this and the very small allowed energy deviation, it is particularly interesting to investigate the behavior of the coefficients under a superposition of a large-amplitude even mode and a small-amplitude odd mode as an initial deformation. This is of particular interest since it is well known that single even modes excite only even modes, while odd modes can excite both even and odd modes. Accordingly, the present work addresses for the first time the modal coupling between odd and even modes and investigate whether odd modes persist, vanish, or amplify over time for superimposed initial deformations. To examine these phenomena, the initial

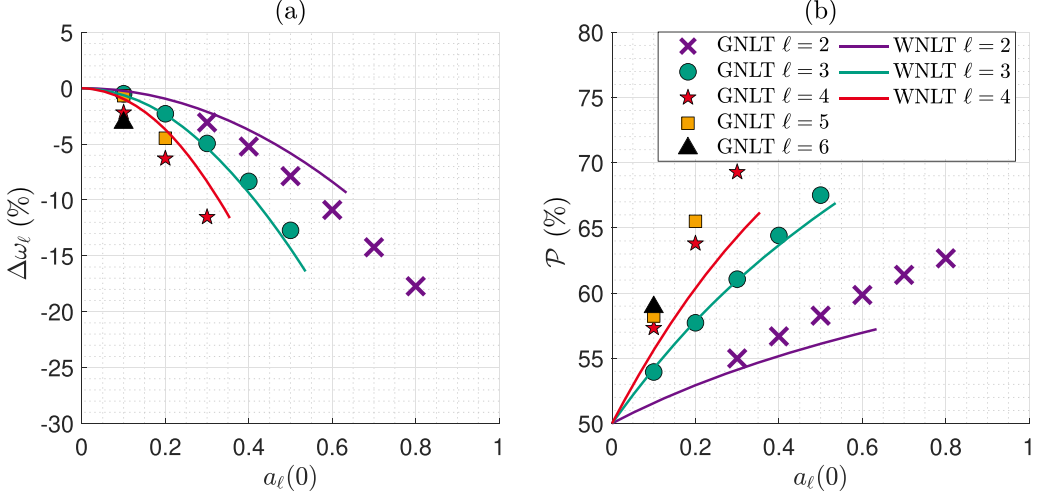


FIG. 12. (a) Relative frequency shift $\Delta\omega_\ell = \frac{\omega_\ell - \omega_\ell^R}{\omega_\ell} 100\%$ and (b) the relative time in the elongated, prolate form $\mathcal{P}$ in % from the present GNLt. For comparison, the results of the WNLT by Zrnić and Brenn [26] are also provided. Both values are given as a function of the amplitude $a_\ell(0)$ of the respective initial deformation mode ℓ over two oscillation periods.

deformation forms are defined as follows:

$$\mathcal{R}_L(\zeta, 0) = 1 + a_2(0)Y_2(\zeta) + a_\ell(0)Y_\ell(\zeta), \quad \text{for } \ell = 3, 5, \quad q_L(\zeta, 0) = 0, \quad (44)$$

where the large-amplitude even mode is chosen as $a_2(0) = 0.5, 0.6$ and the small-amplitude odd modes are chosen as $a_3(0) = 0.15, 0.1, 0.05$ and $a_5(0) = 0.05, 0.03$, respectively. To verify numerical convergence, the first 5 oscillations are observed with a total energy deviation of less than 2%.

First, the addressed questions are analyzed exemplarily in the context of the second and third modes, based on the superimposed initial deformation with the even mode amplitude $a_2(0) = 0.6$, which is symmetric about the horizontal plane, and the odd mode amplitude $a_3(0) = 0.1$, which is not symmetric with respect to the horizontal plane. The shape oscillations of this superimposed initial deformation are depicted in Fig. 13, for the two oscillation periods with the initial configuration shown in Fig. 13(a). It can be seen that the droplet is slightly asymmetric with respect to the horizontal plane due to the superimposed small-amplitude odd mode, which excites further odd and even modes. It is apparent that this slight asymmetry is maintained throughout the shape oscillations, indicating a stable state, i.e., nonvanishing or even growing odd modes. This stability is consistent for all five calculated oscillation periods (see Fig. 22 in Appendix B). The persistence of the coupled

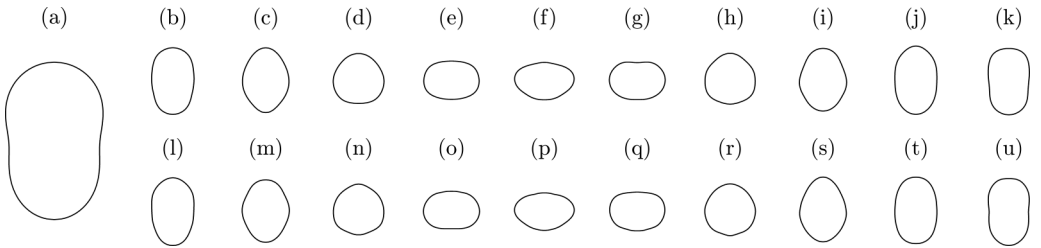


FIG. 13. Shape oscillations as in Fig. 5, but for the superimposed initial deformation ($a_2(0) = 0.6, a_3(0) = 0.1$). The oscillation video can be viewed as Supplemental Material [37].

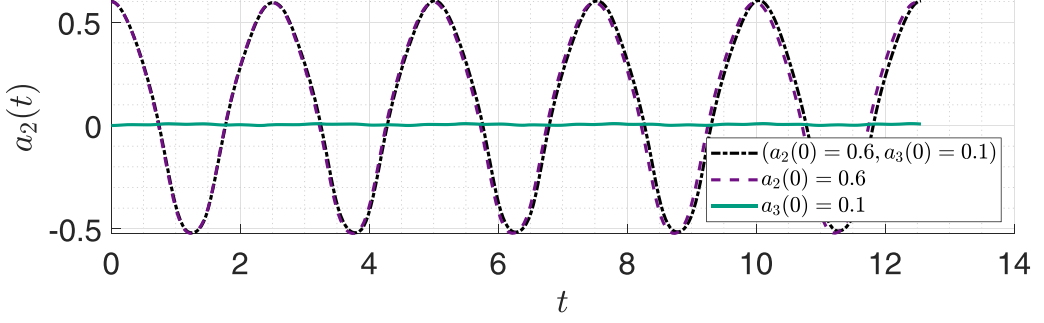


FIG. 14. The time-dependent coefficient $a_2(t)$ examined for three different initial deformations: the superimposed case with $(a_2(0) = 0.6, a_3(0) = 0.1)$ and the two single-mode initial deformations each for $a_2(0) = 0.6$ and $a_3(0) = 0.1$.

modes can be well demonstrated by the time-dependent coefficients $a_2(t)$ and $a_3(t)$ of the initially excited second- and third-order deformation modes in the ansatz (27).

In Fig. 14 the time-dependent coefficient $a_2(t)$ for the superimposed initial deformation with $(a_2(0) = 0.6, a_3(0) = 0.1)$ is compared with the single-mode initial deformations with $a_2(0) = 0.6$ and $a_3(0) = 0.1$. In Fig. 14 it is noticeable that for the small single odd initial deformation mode $a_3(0) = 0.1$ the even mode is only slightly excited, corresponding to a very small amplitude $a_2(t)$. To examine the coupling between the leading modes in more detail, a spectral analysis of the time-dependent coefficients $a_2(t)$ is performed and shown in Fig. 15. The frequency f_2 is dominant, while another frequency near $2f_2^R$ or f_3^R , both of which are very close to each other, is also excited. We strongly believe that this is the frequency $2f_2$, as this frequency does also appear in the single even initial deformation ($a_2(0) = 0.6$), where the third deformation mode ($\ell = 3$) is not excited.

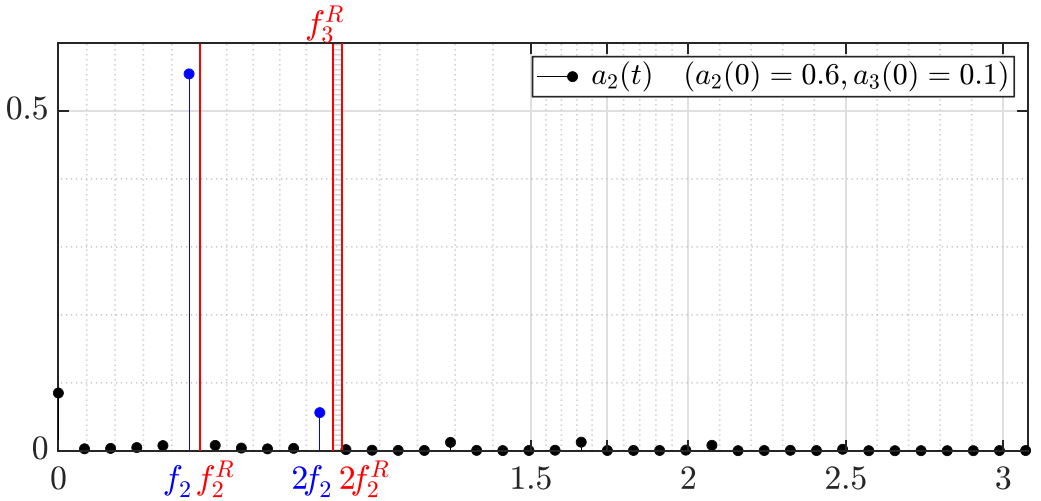


FIG. 15. Power spectrum of the time-dependent coefficient $a_2(t)$ for the superimposed initial deformation $(a_2(0) = 0.6, a_3(0) = 0.1)$. The ordinate represents the power density associated with each frequency, indicating the contribution of different frequencies to the temporal evolution of $a_2(t)$. The abscissa shows the frequency, nondimensionalized with respect to the capillary timescale. The Rayleigh frequencies are marked by red vertical lines labeled as f_i^R ($i = 2, 3$). Two dominant peaks in the power density, highlighted by blue lines, correspond to the modified base frequencies f_2 and $2f_2$.

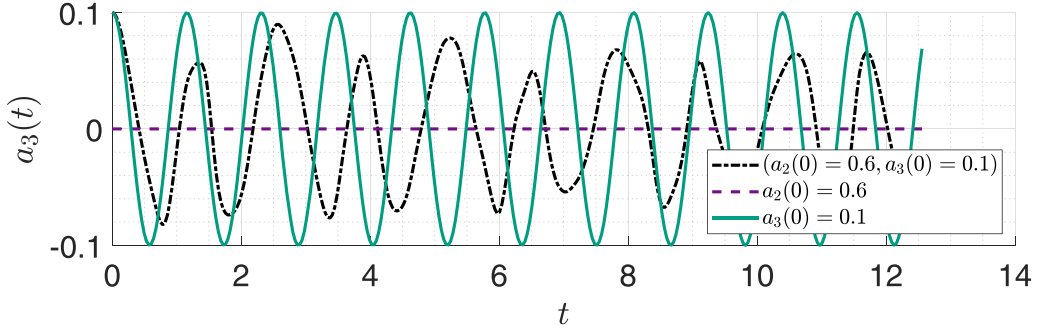


FIG. 16. Time-dependent coefficient $a_3(t)$ examined for three different initial deformations: The superimposed case with $(a_2(0) = 0.6, a_3(0) = 0.1)$ and the two single-mode initial deformations each for $a_2(0) = 0.6$ and $a_3(0) = 0.1$.

This shows again that the superimposed small-amplitude third mode has no significant influence on large-amplitude second mode. It should be noted that the left-most peak for a zero order frequency but with a certain power density represents the time-averaged part. This is the result of nonlinear effects, which lead to asymmetry in the prolate and oblate shapes during droplet oscillation, causing a nonoscillating, nonzero time-averaged contribution.

The interaction between the odd and even modes in the superimposed initial deformation with $(a_2(0) = 0.6, a_3(0) = 0.1)$ is further analyzed by the time-dependent coefficient $a_3(t)$ in Fig. 16 by comparing the superimposed initial deformation with the corresponding single-mode initial deformations, respectively, with $a_2(0) = 0.6$ and $a_3(0) = 0.1$. First, it is evident that the responses of the corresponding odd mode resulting from the superimposed initial deformation $(a_2(0) = 0.6, a_3(0) = 0.1)$ and the single odd initial deformation $a_3(0) = 0.1$ differ significantly in both frequency and amplitude. For the single odd initial deformation, a purely sinusoidal oscillation is observed, characteristic of linear behavior. In contrast, the superimposed initial deformation produces a nonlinear response, deviating from a sinusoidal oscillation, where the dominant even mode exerts a strong influence on the smaller odd mode, although a single even mode does not excite odd modes, as shown in Fig. 16. This behavior is also reflected in Fig. 17, which depicts the power spectrum of the time-dependent odd mode amplitude $a_3(t)$ for the superimposed initial deformation. As expected, the most dominant peak appears at the modified base frequency f_3 , corresponding to the third deformation mode. In addition, the frequency f_2 , associated with the influence of the large-amplitude second mode, also contributes noticeably to the third mode amplitude, indicating nonlinear coupling between the modes.

The statement that the small-amplitude odd mode is strongly influenced by the large-amplitude even mode, whereas the reverse influence is small, is heavily related to the surface curvature. The droplet's shape is mainly determined by the even mode due to its large amplitude. During oscillation, the droplet alternates between a prolate and oblate form. As shown in Fig. 18, in the oblate form, the highest curvature is at the equator and covers a relatively large area. In the prolate form, the highest curvature is located at the poles, which corresponds to a relatively small area. Therefore, the restoring force is higher in the oblate form than in the prolate form.

This asymmetry in the restoring force significantly affects all modes, including the small-amplitude odd mode. Thus, the large-amplitude even mode exerts a strong influence on the small-amplitude odd mode, even though a single even initial deformation mode cannot excite odd modes. In contrast, the deformation caused by the small-amplitude odd mode is too weak to produce meaningful curvature changes and therefore has little effect on the large-amplitude even mode.

The three observations, the persistence of slight asymmetry in the droplet shape with respect to the horizontal plane, the minimal influence on the large-amplitude even mode, and the significant effect on the small-amplitude odd mode due to nonlinear coupling, can also be observed when the

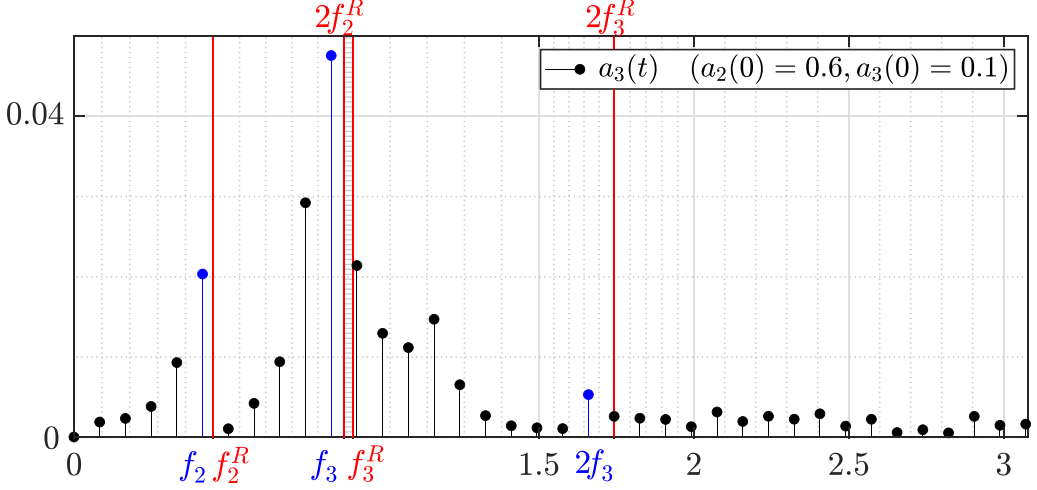


FIG. 17. Power spectrum of the time-dependent coefficient $a_3(t)$ for the superimposed initial deformation ($a_2(0) = 0.6$, $a_3(0) = 0.1$) as in Fig. 15. Three dominant peaks in the power density, highlighted by blue lines, correspond to the modified base frequencies f_2 , f_3 , and $2f_3$.

fifth mode is superimposed onto the large-amplitude second mode. For the superimposed initial deformation with ($a_2(0) = 0.6$, $a_5(0) = 0.05$), the first two oscillations are depicted in Fig. 19, with the initial configuration shown in panel (a). The slight asymmetry in the droplet shape with respect to the horizontal plane persists over the two oscillation periods. This asymmetry remains consistent also across all five calculated oscillations (see Fig. 23 in Appendix B). Figure 20 shows the power spectrum of the corresponding time-dependent coefficient $a_2(t)$ for the superimposed initial deformation with ($a_2(0) = 0.6$, $a_5(0) = 0.05$). It is clearly observable that only the base frequency f_2 and its multiples occur in the second mode. There is no peak at f_5 , indicating that the fifth mode has no significant influence on the second mode amplitude.

Furthermore, Fig. 21 shows the power spectrum of the coefficient $a_5(t)$ of the fifth mode, which reveals a broader range of excited frequencies. As expected, the modified base frequency f_5 is the

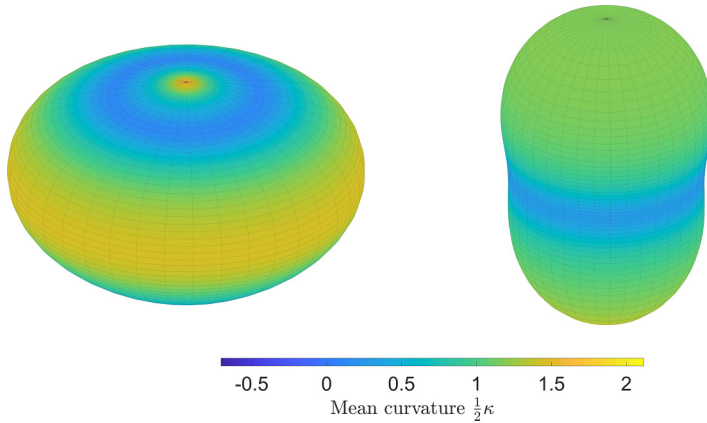


FIG. 18. The mean curvature $\frac{1}{2}\kappa$ (nondimensionalized) for the oblate form (left) and the prolate form (right) for the superimposed initial deformation ($a_2(0) = 0.6$, $a_3(0) = 0.1$). The oscillation video can be viewed as Supplemental Material [37].

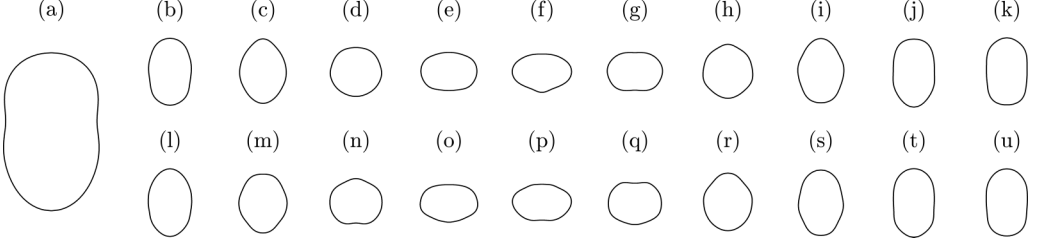


FIG. 19. Shape oscillations as in Fig. 5, but for the superimposed initial deformation ($a_2(0) = 0.6$, $a_5(0) = 0.05$). The oscillation video can be viewed as Supplemental Material [37].

dominant one, but several multiples of the second mode frequency f_2 also appear, confirming that the dominant even second mode exerts a strong influence on the smaller odd mode, which in turn is due to nonlinear modal coupling and the resulting pressure effects associated with surface curvature during oscillation.

V. CONCLUSIONS

The GNLT developed by Plümacher *et al.* [29] can be used to represent droplet shape oscillations for reasonably large-amplitude deformations. A new and highly specialized solver, based on the spectral method and embedded within a Galerkin framework, has been developed to solve the resulting integrodifferential equations. Neglecting viscous stresses in the momentum balance, droplet shape oscillations have been simulated for different initial deformation modes ($\ell = 2, 3, 4, 5, 6$) with different amplitudes. With a truncation degree of $L = 12$ for the spectral series of spherical harmonics, highly precise results have been achieved. With the GNLT, it is possible to describe the quasiperiodicity, the relative frequency change and the relative time in the prolate form of droplet shape oscillations. Good agreement with the known literature has been demonstrated. All described phenomena based on modal coupling become more apparent at larger amplitudes. To examine modal

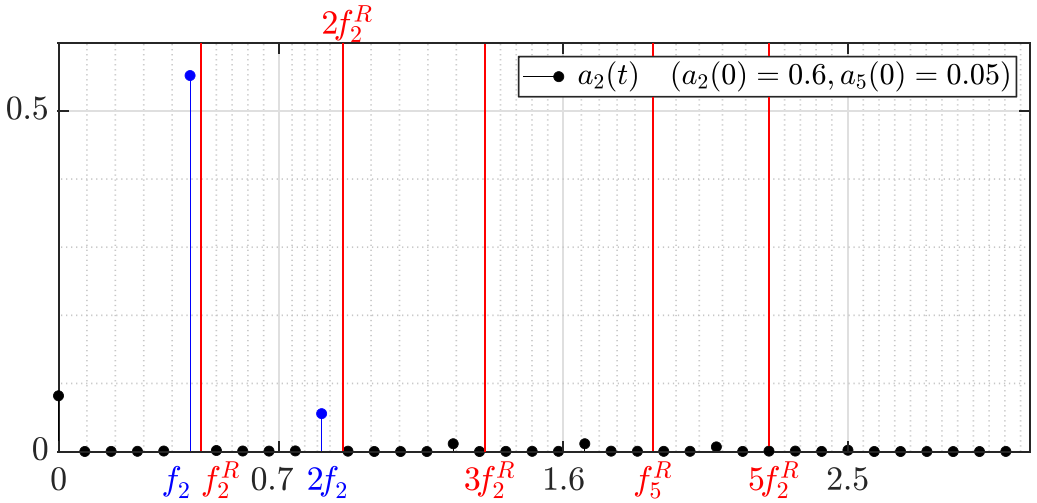


FIG. 20. Power spectrum of the time-dependent coefficient $a_2(t)$ for the superimposed initial deformation ($a_2(0) = 0.6$, $a_5(0) = 0.05$) as in Fig. 15. The Rayleigh frequencies are marked by red vertical lines labeled as f_i^R ($i = 2, 5$). Two dominant peaks in the power density, highlighted by blue lines, correspond to the modified base frequencies f_2 and $2f_2$.

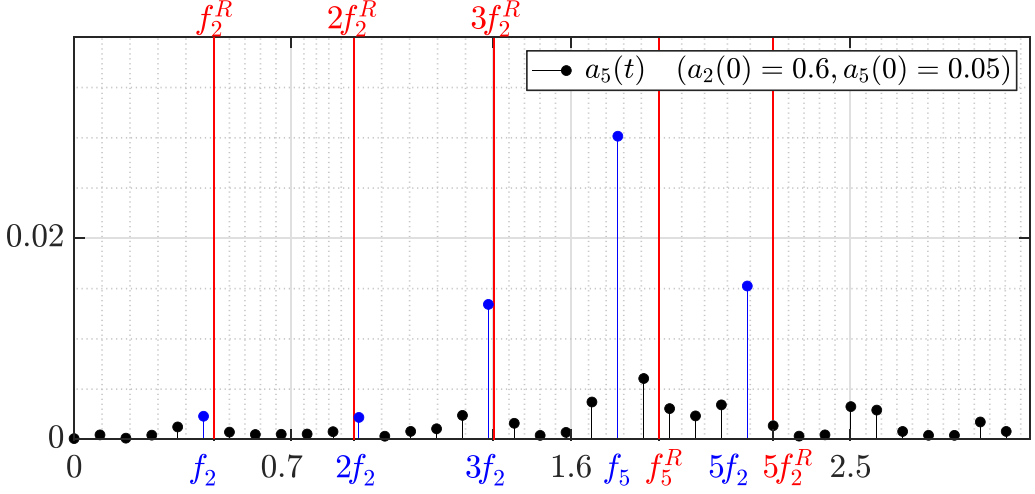


FIG. 21. Power spectrum of the time-dependent coefficient $a_5(t)$ for the superimposed initial deformation ($a_2(0) = 0.6$, $a_5(0) = 0.05$) as in Fig. 15. The Rayleigh frequencies are marked by red vertical lines labeled as f_i^R ($i = 2, 5$). Dominant peaks in the power density, highlighted by blue lines, correspond to the modified base frequencies f_2 , $2f_2$, $3f_2$, f_5 , and $5f_2$.

coupling in detail, a study has been conducted on the interaction between a large-amplitude even mode and a small-amplitude odd mode. This study presents the first detailed examination of this specific configuration. The axisymmetric initial deformation with a slight asymmetry with respect to the horizontal plane by superimposing a small-amplitude odd mode (third or fifth mode) on a large-amplitude even mode (second mode) was maintained throughout the shape oscillations, indicating a nondamping coupling. Furthermore, it has been shown that the large-amplitude even mode is nearly unaffected by the small-amplitude odd modes, whereas the small-amplitude odd modes are significantly influenced by the large-amplitude even mode due to nonlinear modal coupling and the resulting restoring forces arising from pressure differences linked to surface curvature during the oscillations.

ACKNOWLEDGMENT

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DATA AVAILABILITY

The data that support the findings of this article are openly available [38].

APPENDIX A: SPHERICAL HARMONICS

The spherical harmonics Y_ℓ^m are special functions defined on the surface of a sphere. They result as the solution of the angular part to Laplace's equation in spherical coordinates (r, θ, φ) . The definition from Olver *et al.* [39] is chosen. The Laplace equation in spherical harmonics is given by

$$\Delta f = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial f}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial f}{\partial \theta} \right) + \frac{1}{r^2 \sin^2 \theta} \frac{\partial^2 f}{\partial \varphi^2} = 0. \quad (\text{A1})$$

With separation of variables

$$f(r, \theta, \varphi) = R(r)Y(\theta, \varphi), \quad (\text{A2})$$

Eq. (A1) can be written into two independent differential equations

$$\frac{1}{R} \frac{\partial}{\partial r} \left(r^2 \frac{\partial R}{\partial r} \right) = \ell(\ell + 1) \quad (\text{A3})$$

and

$$\frac{1}{Y} \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial Y}{\partial \theta} \right) + \frac{1}{Y} \frac{1}{\sin^2 \theta} \frac{\partial^2 Y}{\partial \varphi^2} = -\ell(\ell + 1). \quad (\text{A4})$$

It should be noted that Eq. (A4) represents the angular differential equation. Since spherical harmonics do not depend on the radial coordinate r , the radial differential equation (A3) is not considered further. However, separating variables for Eq. (A4) with

$$Y(\theta, \varphi) = \Theta(\theta)\Phi(\varphi) \quad (\text{A5})$$

and taking into account the requirement that the total solution be single-valued and periodic in φ , i.e., $\Phi(\varphi + 2\pi) = \Phi(\varphi)$ follows,

$$\frac{1}{\Phi} \frac{\partial^2 \Phi}{\partial \varphi^2} = -m^2 \quad (\text{A6})$$

and

$$\ell(\ell + 1) \sin^2 \theta + \frac{\sin \theta}{\Theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial \Theta}{\partial \theta} \right) = m^2. \quad (\text{A7})$$

The solution of Eq. (A6) is

$$\Phi^m(\varphi) = \exp(im\varphi). \quad (\text{A8})$$

For Eq. (A7), the substitution of

$$\zeta = \cos \theta \quad (\text{A9})$$

leads to the associated Legendre differential equation

$$(1 - \zeta^2) \frac{\partial^2}{\partial \zeta^2} \Theta - 2\zeta \frac{\partial}{\partial \zeta} \Theta + \left[\ell(\ell + 1) - \frac{m^2}{1 - \zeta^2} \right] \Theta = 0, \quad (\text{A10})$$

whose general solution is a linear combination of the associated Legendre functions of the first and second kind

$$\Theta_\ell^m(\zeta) = A P_\ell^m(\zeta) + B Q_\ell^m(\zeta). \quad (\text{A11})$$

Regularity of the spherical harmonics at the poles $\theta = 0$ and $\theta = \pi$ requires $B = 0$, because Q_ℓ^m diverges there. Hence, only P_ℓ^m remains

$$P_\ell^m(\zeta) = \frac{(-1)^m}{2^\ell \ell!} (1 - \zeta^2)^{m/2} \frac{\partial^{\ell+m}}{\partial \zeta^{\ell+m}} (\zeta^2 - 1)^\ell. \quad (\text{A12})$$

Since spherical harmonics are orthonormal,

$$\int_{\mathbb{S}} Y_\ell^m \cdot Y_{\ell'}^{m'} \cdot d\Omega = \delta_{\ell\ell'} \cdot \delta_{mm'}, \quad (\text{A13})$$

the normalization factor A given by

$$A = \sqrt{\frac{2\ell + 1}{4\pi} \cdot \frac{(\ell - m)!}{(\ell + m)!}} \quad (\text{A14})$$

is introduced. Assembling Eq. (A5) with Eqs. (A8) and (A12) and multiplying it with the normalization factor (A14), the spherical harmonics $Y_\ell^m(\zeta, \varphi)$ can be expanded to

$$Y_\ell^m(\zeta, \varphi) = \frac{(-1)^m}{2^\ell \ell!} \left(\frac{(\ell - m)!(2\ell + 1)}{4\pi(\ell + m)!} \right)^{1/2} \exp(im\varphi) (1 - \zeta^2)^{m/2} \frac{\partial^{\ell+m}}{\partial \zeta^{\ell+m}} (\zeta^2 - 1)^\ell. \quad (\text{A15})$$

The real parts of the spherical harmonics are given for $m < 0$:

$$Y_\ell^m(\zeta, \varphi) = \frac{(-1)^{|m|}}{2^\ell \ell!} \left(\frac{(\ell - |m|)!(2\ell + 1)}{2\pi(\ell + |m|)!} \right)^{1/2} \sin(|m|\varphi) (1 - \zeta^2)^{|m|/2} \frac{\partial^{\ell+|m|}}{\partial \zeta^{\ell+|m|}} (\zeta^2 - 1)^\ell, \quad (\text{A16})$$

for $m = 0$:

$$Y_\ell(\zeta) = \frac{1}{2^\ell \ell!} \left(\frac{\ell!(2\ell + 1)}{4\pi \ell!} \right)^{1/2} \frac{\partial^\ell}{\partial \zeta^\ell} (\zeta^2 - 1)^\ell, \quad (\text{A17})$$

and for $m > 0$:

$$Y_\ell^m(\theta, \varphi) = \frac{(-1)^m}{2^\ell \ell!} \left(\frac{(\ell - m)!(2\ell + 1)}{2\pi(\ell + m)!} \right)^{1/2} \cos(m\varphi) (1 - \zeta^2)^{m/2} \frac{\partial^{\ell+m}}{\partial \zeta^{\ell+m}} (\zeta^2 - 1)^\ell. \quad (\text{A18})$$

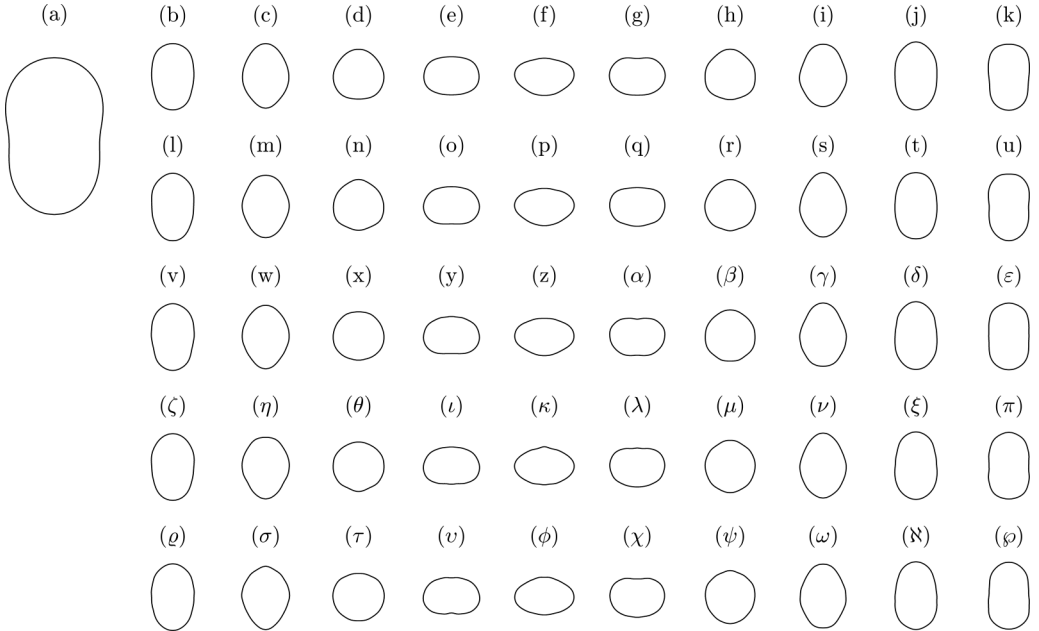


FIG. 22. Shape oscillations for the initial deformation modes $\ell = 2$, $\ell = 3$ with amplitudes ($a_2(0) = 0.6$, $a_3(0) = 0.1$) at 51 time instants with equal time intervals $\Delta t = T/10$ with the period T in the first five oscillation periods. The oscillation video can be viewed as Supplemental Material [37].

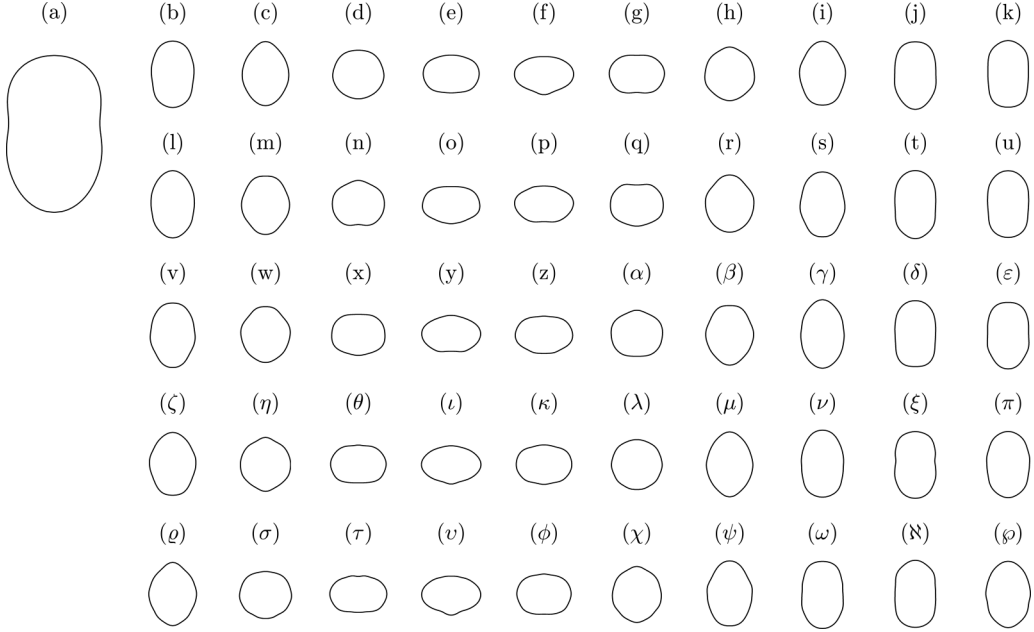


FIG. 23. Shape oscillations for the initial deformation modes $\ell = 2$, $\ell = 5$ with amplitudes ($a_2(0) = 0.6$, $a_5(0) = 0.05$) at 51 time instants with equal time intervals $\Delta t = T/10$ with the period T in the first five oscillation periods. The oscillation video can be viewed as Supplemental Material [37].

APPENDIX B: SHAPE OSCILLATIONS OF DROPLETS WITH A SUPERIMPOSED INITIAL DEFORMATION

Figures 22 and 23 show the long-term shape oscillations of two superimposed initial deformations. The large-amplitude even mode $a_2(0) = 0.6$ is combined with small-amplitude odd modes $a_3(0) = 0.1$ and $a_5(0) = 0.05$, respectively. These extended time sequences confirm that the qualitative behavior observed in Sec. IV continues beyond the initial two oscillation periods.

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