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Nonlinear Drop Shape Oscillations

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“If you want to find the secrets of the universe, think in terms of energy, frequency and vibration.” Nikola Tesla (1856 - 1943)

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Abstract

Nonlinear axisymmetric shape oscillations of a viscoelastic, viscous and inviscid liquid drop in a vacuum are investigated theoretically for their relevance in transport processes across the drop surface, and for fundamental interest. The theoretical approach applied is weakly nonlinear analysis that is carried out to third order for the modes of initial deformation up to $m = 4$, but not limited to it. The analysis covers the range of the drop Ohnesorge number between zero, and one for Newtonian liquids and viscoelastic material behavior, selected according to experimental data. With increasing surface deformation for the fundamental mode of initial deformation $m = 2$, the oscillations develop an excess time of the prolate shape against time in oblate shape, a frequency decrease below the linear value and quasi-periodicity of the motion. In contrast to the inviscid case, the analysis for the Newtonian drop reveals the frequency decrease and the quasi-periodicity already in the second-order approximation. The nonlinear phenomena are positively validated against relevant literature, and the results are extended to the higher-order initial modes for the first time. The results for the inviscid drop reveal mode coupling as the source causing the quasi-periodic motion of the drop. The next result highlighted is that, during the oscillations, slightly viscous drops at strong deformations may never reach the spherical shape. The analysis reveals damped oscillatory behaviour for higher-order initial modes even at super-critical Ohnesorge number. This is due to coupling to lower-order modes, which the linear theory cannot predict. The analysis quantifies the effects of viscosity and timescales of viscoelastic material behaviour, measured in Ohnesorge and Deborah numbers, respectively.

Zusammenfassung

Nichtlineare achsensymmetrische Formschwingungen viskoelastischer, viskoser oder nicht viskoser Flüssigkeitstropfen im Vakuum werden theoretisch untersucht. Motivation sind die Bedeutung der Schwingungen für Transportprozesse über die Tropfenoberfläche und grundsätzliches wissenschaftliches Interesse. Die angewandte theoretische Methode ist die schwach nichtlineare Analyse, die bis zur dritten Näherungsordnung durchgeführt wird. Der Tropfen ist zu Anfang der Bewegung entsprechend einem Legendre-Polynom des Grades m verformt, wobei m einen Schwingungsmodus bedeutet. Moden der Anfangsverformung bis zu $m = 4$ werden untersucht, die Theorie ist hierauf aber nicht begrenzt. Die Analyse umfasst Ohnesorge-Zahlen des Tropfens zwischen null und eins für Newtonsche Flüssigkeiten und für viskoelastisches Materialverhalten. Diese Werte wurden nach experimentellen Daten ausgewählt. Bei der Grundmode $m = 2$ verbringen die Tropfen mit zunehmender Verformung während einer Schwingungsperiode immer mehr Zeit in der "prolaten" als in der "oblaten" Form, und die Schwingungsfrequenz sinkt unter den Wert aus der linearen Analyse. Ferner entsteht eine Quasi-Periodizität der Bewegung. Im Gegensatz zum reibungsfreien Fall zeigt die Analyse für den Newtonschen Tropfen die Frequenzabnahme und die Quasi-Periodizität bereits in der Näherung zweiter Ordnung. Die nichtlinearen Phänomene werden für $m = 2$ positiv gegen die Literatur validiert, und die Ergebnisse werden erstmals auf die höheren Anfangsmoden $m \geq 3$ erweitert, für die es in der existierenden Literatur keine Ergebnisse zur Validierung gibt. Die Ergebnisse zeigen für den reibungsfreien Tropfen, dass die quasi-periodische Bewegung des Tropfens durch die Kopplung verschiedener Verformungsmoden entsteht. Ein weiteres wichtiges Ergebnis ist, dass leicht viskose Tropfen bei starken Verformungen bei der Schwingung niemals die Kugelform erreichen. Ferner ergibt die Analyse

gedämpfte Schwingungen bei Anfangsmoden höherer Ordnung auch bei überkritischer Ohnesorge-Zahl, für die die lineare Analyse aperiodisches Verhalten vorhersagt. Ursache ist die Kopplung mit Moden niedrigerer Ordnung, für die ein schwingendes Verhalten des Tropfens vorliegt. Für viskoelastische Tropfen quantifiziert die Analyse die Auswirkungen der Viskosität und der Zeitskalen des viskoelastischen Materialverhaltens, gemessen in Ohnesorge- und Deborah-Zahlen.

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Nomenclature

Latin Symbols

a	equilibrium drop radius
a_{km}	second-order coefficient
A	oscillation amplitude
A_{p_r}	particular solutions of the radial velocity
A_{p_θ}	particular solutions of the polar velocity
a_1, a_2	coefficients in second-order coefficient calculation
A_{21l}	second-order velocity coefficient
b_{km}	second-order coefficient
b_1, b_2	coefficients in second-order coefficient calculation
B_{21l}	second-order velocity coefficient with index l
c_{km}	second-order coefficient
C	second-order coefficient
C_{1m}, C_{2m}	first-order coefficients
C_{3l}, C_{4l}	second-order coefficients
d_{lkm}	second-order coefficient
D_{21l}	second-order pressure coefficient
$\mathbf{D}$	rate of deformation tensor
$\mathbf{D}_1$	first-order rate of deformation tensor
$\mathbf{D}_2$	second-order rate of deformation tensor

Nomenclature

∇ $\mathbf{D}$	upper convected derivative of tensor $\mathbf{D}$
De_1	Deborah number for stress relaxation time
De_2	Deborah number for deformation retardation time
E_k	drop kinetic energy
E_s	operator
E_{surf}	drop surface energy
E_{tot}	drop total energy (linear combination of E_k and E_{surf})
f	oscillation frequency
f_{ar}, f_{br}	radially dependent functions in second-order radial velocity
$f_{a\theta}, f_{b\theta}$	radially dependent functions in second-order polar velocity
f_{km}	second-order coefficients
f_r, f_θ, f_p	radially dependent first-order functions
G_{2kl}	second-order coefficient
h	summation index
H	second-order coefficient
j	summation index
j_m	spherical Bessel function of the first kind of the order m
j_k	spherical Bessel function of the first kind of the order k
j_h	spherical Bessel function of the first kind of the order h
k	summation index
K	second-order coefficient
l	summation index
L	second-order coefficient
m	oscillation mode number
$\vec{n}$	normal unit vector

Oh	Ohnesorge number
Oh_v	frequency dependent Ohnesorge number
Oh_0	Ohnesorge number with μ_0
p	pressure
P_m, P_l, P_h	Legendre polynomial of degrees m, l and h
P_m^1	Legendre function of order 1 and degree m
p_0	capillary pressure inside the drop
p_1	first-order pressure
p_2, p_{21}, p_{22}	second-order pressure
p_3, p_{31}, p_{32}	third-order pressure
$\mathcal{P}$	modified pressure
$\mathcal{P}_{21,H}, \mathcal{P}_{31,H}$	homogeneous solutions of modified pressure
$\mathcal{P}_{21,I}, \mathcal{P}_{31,I}$	particular solutions of modified pressure
q	complex argument in spherical Bessel function
Q_h, Q_k, Q_m	ratio of Bessel functions of order h, k and m
r	radial coordinate
r_s	drop shape function
$R_{k_{31}}$	second-order kinematic BC right-hand side
$R_{z_{31}}$	second-order zero-normal BC right-hand side
$R(\eta_0, t)$	relative volume deviation
R_1	second-order kinematic BC right-hand side
R_2	second-order zero-shear BC right-hand side
R_3	second-order zero-normal BC right-hand side
t	time
t_{km}, t_{km}^*	second-order coefficient with index k and m

Nomenclature

u_r	radial velocity
u_{r1}	first-order radial velocity
u_{r2}, u_{r21}, u_{r22}	second-order radial velocity
u_{r3}, u_{r31}, u_{r32}	third-order radial velocity
u_θ	polar velocity
$u_{\theta1}$	first-order polar velocity
$u_{\theta2}, u_{\theta21}, u_{\theta22}$	second-order polar velocity
$u_{\theta3}, u_{\theta31}, u_{\theta32}$	third-order polar velocity
$\vec{v}_1$	first-order velocity vector
$\vec{v}_2$	second-order velocity vector
V	drop volume
V_s	spherical drop volume
w	glycerol mass fraction
x	variable with polar dependency = $\cos \theta$
y_m	spherical Bessel function of the second kind
y_0	equilibrium drop height or width
$y(t)$	time dependent distance
z_m	drop centre of mass for initial mode m

Greek symbols

α_m	complex angular frequency of mode m
$\alpha_{2k,0}, \alpha_{3h,0}$	Rayleigh angular frequency for mode k and h
α_i	imaginary part of the complex angular frequency
$\alpha_{m,i}$	imaginary part of the complex angular frequency
$\alpha_{m,r}$	real part of the complex angular frequency

$\alpha_{res,i}$	imaginary part of the resultant complex angular frequency
α_r	real part of the complex angular frequency
$\alpha_{r,C}$	α_r by Chandrasekhar (1959)
β_1	frequency dependent factor
δ_{2j}, δ_{2l}	second-order surface deformation function
δ_{3j}, δ_{3h}	third-order surface deformation function
$\delta_{3i}^*, \delta_{34}^*$	third-order surface deformation function with secular terms
δ_0, δ_{kl}	second-order coefficients
$\delta_{l_1 l_2}$	Kronecker delta
$\epsilon_{0m}, \epsilon_{km}$	second-order coefficients
η	surface deformation function
η_0	initial deformation parameter
η_1	first-order surface deformation function
$\eta_2, \eta_{21}, \eta_{22}$	second-order surface deformation function
$\eta_3, \eta_{31}, \eta_{32}$	third-order surface deformation function
$\hat{\eta}_1$	first-order surface deformation amplitude
$\hat{\eta}_{22k}$	second-order surface deformation amplitude
$\hat{\eta}_{32h}$	third-order surface deformation amplitude
θ	polar angle
λ_1	dimensional stress relaxation time
λ_2	dimensional deformation retardation time
μ	drop dynamic viscosity
μ^*	measured drop dynamic viscosity
μ_0	drop zero-shear stress dynamic viscosity
ν, ν_0, ν_1, ν_2	coefficient in strained time coordinate

Nomenclature

ρ	drop density
σ	drop surface tension
σ^*	measured drop surface tension
τ	strained time
$\tau_{\phi\phi}$	normal azimuthal extra stress
$\tau_{\theta\theta}$	normal polar extra stress
$\tau_{r\theta}$	extra shear stress
τ_{rr}	normal radial extra stress
$\boldsymbol{\tau}$	extra stress tensor
$\boldsymbol{\tau}_1$	first-order extra stress tensor
$\boldsymbol{\tau}_2, \boldsymbol{\tau}_{21}, \boldsymbol{\tau}_{22}$	second-order extra stress tensor
$\boldsymbol{\tau}_3, \boldsymbol{\tau}_{31}, \boldsymbol{\tau}_{32}$	third-order extra stress tensor
$\overset{\nabla}{\boldsymbol{\tau}}$	upper-convected derivative of extra stress tensor
φ	azimuthal angle
φ_0	reference phase angle
ψ, ψ_1, ψ_2	stream functions
ω_m	dimensional complex angular frequency
$\omega_{m,i}$	imaginary part of ω_m
$\omega_{m,r}$	real part of ω_m
$\omega_{m,0}$	dimensional Rayleigh frequency for m
Ω_m	non-dimensional complex angular frequency for m

Superscripts and subscripts

LA	large amplitude oscillations
------	------------------------------

(n)	negative time dependency
(nn)	time dependency according to $\exp[-(\alpha_m^{(n)} + \alpha_{2k}^{(n)})t]$
(np)	time dependency according to $\exp[-(\alpha_m^{(n)} + \alpha_{2k}^{(p)})t]$
(p)	positive time dependency
(pn)	time dependency according to $\exp[-(\alpha_m^{(p)} + \alpha_{2k}^{(n)})t]$
(pnn)	time dependency according to $\exp[-(\alpha_m^{(p)} + 2\alpha_m^{(n)})t]$
(pp)	time dependency according to $\exp[-(\alpha_m^{(p)} + \alpha_{2k}^{(p)})t]$
(ppn)	time dependency according to $\exp[-(2\alpha_m^{(p)} + \alpha_m^{(n)})t]$
SA	small amplitude oscillations
(v)	purely damped time dependency
(1)	presents (p) or (n) time dependency
(2)	presents (ppn) or (pnn) time dependency
(3)	presents (pp) or (nn) time dependency
(4)	presents (pn) or (np) time dependency
$1kl, \dots, 3kl$	1, 2 and 3 distinguish coefficient
$1km, \dots, 5km$	1, 2, ..., 5 distinguish coefficient
$2k$	second-order with subscript k
$21, 21j, 21l$	second-order contribution 21
$22, 22k$	second-order contribution 22
$3h$	third-order with subscript h
$3j$	third-order with subscript j
$31, 31h, 31j$	third-order contribution 31
$31hk, 31jk$	third-order contribution 31
$32, 32h$	third-order contribution 32

1. Introduction

The introduction provides an overview of literature on drop shape oscillations. It shows the related literature on drop shape oscillations, from their beginning till the present day describing the elementary findings of this fundamental physical phenomenon. Motivation for the current work on drop shape oscillations is presented in the second section. The third section lays out the organization of the thesis and emphasizes its objectives.

1.1. Scientific interest in drop shape oscillations

Drop shape oscillations have been a subject of scientific investigations for a long period of time, and this is due to fundamental interest and various biological and physical applications. The fields of interest-span vary in the same way as the length scale of the systems based on the droplets. Therefore the investigation of drop shape oscillations finds its purpose in fields such as container-less materials processing, separation processes in chemical engineering, in-air microfluidics and in different sub-fields of meteorology. The drop shape oscillations are of fundamental interest, since the drop surface interaction with the ambient gas phase induces gradients of velocity, temperature and species concentration, influencing the transport of momentum, mass and heat across the drop surface. The drop shape oscillations in the light of application processes are used for measuring liquid rheological and drop capillary behaviours, which are challenging topics. They benefit from the predictions known for small-amplitude drop shape oscillations. The motivation arises when one considers the effects that a drop exhibits with moderate and large amplitude oscillations. An example is seen in experimental studies based on drop shape oscillations using ultrasonic levitation (Trinh and Zwern, 1982) and a microgravity field

(Holt et al., 1997; Wang, Anilkumar, and Lee, 1996). Finding a method that describes correctly nonlinear effects produced by moderate amplitude oscillations for different liquid rheological fluid behaviour may be of great interest. Further motivation will be discussed in the next section of the current chapter; now a historical study on drop shape oscillations is presented.

In a context of geophysical theories, Thomson (1863) studied the case with gravity as a restoring force on the interior of the earth and the tidal forces acting on it due to the moon and the sun. He considered a case of infinitesimally small oscillations of an inviscid drop in a dynamically negligible outer medium and described the flow as a potential field and used spherical harmonics, which he called modes. From his study, frequencies are derived for single mode oscillation. Darwin (1879) extended Thomson's analysis by considering the effects of low viscosity. In the appendix of his paper on the capillary phenomena of jets, Rayleigh (1879) presented an analysis of the linear shape oscillations of an inviscid drop in a vacuum around a spherical equilibrium state. One result is the equation for the dimensional angular frequency that reads

$$\omega_{m,0} = \sqrt{m(m-1)(m+2)} \sqrt{\frac{\sigma}{\rho a^3}}, \quad (1.1)$$

for a mode m , density ρ , surface tension σ and radius a of the drop in the ambient vacuum. The initial deformation mode m has natural values $m = 2, 3, 4, \dots$ and the corresponding modes are related to the shapes which are shown in the Figure 1.1. Analogous to the extension of Thomson's work by Darwin, Rayleigh's work is continued by Lamb (1881, 1932). He generalised Rayleigh's result by accounting for the drop viscosity and the density of the ambient medium. He predicted the threshold Ohnesorge number $Oh = \mu/(\sigma a \rho)^{1/2}$ of the drop for the onset of aperiodic motion. Oscillating drops reduce the frequency with the increase of liquid viscosity until the threshold Ohnesorge number. Further increase of viscosity yields aperiodic movement as a reaction to perturbation.

Chandrasekhar (1959) analysed small drop shape oscillations applied to a viscous, self-gravitating globe in a vacuum. In his work, the linearised Navier-Stokes equations are solved taking gravity into account, and the solutions of the velocity field and stress tensor are determined for the globe oscillation.

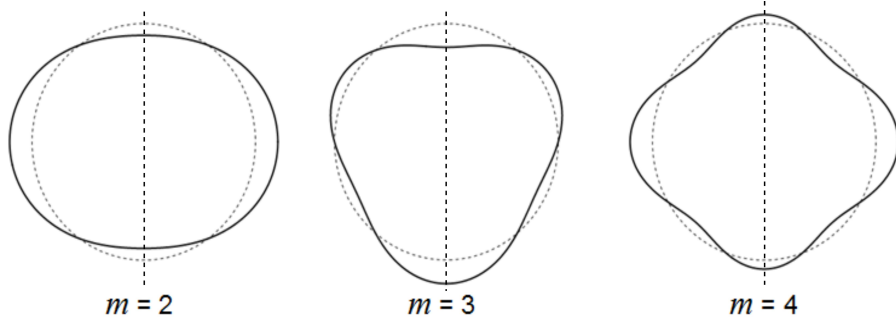


Figure 1.1.: The first three deformation modes. The undeformed spherical drop shape in equilibrium is presented with the dashed lines.

The characteristic equation is derived for the globe, which determines the complex angular oscillation frequency. Further, the analysis by Reid (1960) showed that the results could be applicable for drops, where the restoring force towards the spherical state is due to surface tension rather than self-gravitation. First known extensive experimental investigations on the effect of droplet oscillations on mass transfer is by Rose and Kintner (1966). These authors studied mass transfer from organic liquid-water mixture droplets oscillating in an ambient water phase and quantified the degree of mass transport from the droplet to the ambient medium. The prediction of mass transfer across the interface between a droplet and its host medium with the influence from adsorbed substances was studied by Gottier, Amundson, and Flumerfelt (1986). Further analysis on linear drop shape oscillations is done by Miller and Scriven (1968). They accounted for both the viscous and the inertial influences from the ambient medium that surrounds the viscous oscillating drop. Prosperetti studied the impact of the initial conditions on the oscillations. He analysed drop shape oscillations as an initial-value problem (Prosperetti, 1980a) and showed that the normal-mode approach might miss the fact that, in a range of Ohnesorge numbers, oscillations starting aperiodically may become periodic after some time. The most important results from the papers highlighted are the angular frequency and damping rate of the oscillations, as well as the time-dependent description of the drop shape evolution during the oscillations.

Foote (1973) and Alonso (1974) made first computational investigations

of nonlinear drop shape oscillations with large amplitudes. They used the marker-and-cell finite-difference technique, which could numerically simulate the time-dependent free-surface flows for weak liquid viscosity. In the former work, Foote (1973) showed the oscillation frequency decrease with increasing aspect ratio of the drops with small viscosity. The same work initially addressed the drops with a Legendre polynomial of a certain degree m or with shapes defined as oblate spheroids. Alonso (1974) used the former initial shape for his simulations of atomic nuclei and drop oscillations. She observed a drop with initially large deformations and electrical charges. Results address fissions in large-amplitude oscillations and merging after collisions, and they show kinetic and Coulomb energies during the oscillations and the surface area as time functions.

Early experiments on oscillations of liquid droplets were carried out on neutrally buoyant droplets immersed in an immiscible host liquid. Studies by Marston (1980) and Marston and Apfel (1980) showed the effects of the acoustic radiation pressure to be suitable for driving droplet shape oscillations - even for superheated droplets placed in an ambient fluid. Trinh and Zwern (1982) and Trinh and Wang (1982) made the first experimental studies of the small- and large-amplitude shape oscillations of drops, respectively, in an immiscible host liquid and levitated free liquid drops in air. They showed, for the two-lobed oscillation mode, that the frequency decreases with increasing drop deformation and that the drop spends more time in the prolate shape than in the oblate shape. The data measured were found consistent with the theoretical predictions by Marston (1980).

The well-known analytical work by Tsamopoulos and Brown (1983) found motivation in previous numerical and experimental studies, and they investigated nonlinear oscillations of inviscid drops and bubbles for the initial deformation modes $m = 2, 3$ and 4 . They used a method of weakly nonlinear analysis, with the Poincaré-Lindstedt expansion of the drop shape, the velocity potential and the oscillation frequency. Similar to the computational studies by Foote (1973) and Alonso (1974), Tsamopoulos and Brown found that the drop shapes deviated from the linear results, and angular frequencies decreased with increasing oscillation amplitude (in the case of the two-lobed shape of a maximum prolate aspect ratio 1.4 by approximately

5%). In the two-lobed oscillation mode, an increase of oscillation amplitude affects the motion. It loses the sinusoidal dependency on time and gives rise to an asymmetry of times during one oscillation period spent in the prolate and the oblate states. An extension to the classical work (Tsamopoulos and Brown, 1983) was the investigation of the resonant oscillations of a charged drop (Tsamopoulos and Brown, 1984). Their study led to predictions of inviscid charged-drop resonance for different modes of deformation. The internal resonance study was continued by Natarajan and Brown (1987) based on different modes of deformation in inviscid three-dimensional non-linear oscillations. Lundgren and Mansour (1988) worked on a numerical boundary-integral method to investigate the mode coupling in axisymmetric large-amplitude shape oscillations of large drops. They gave insight into instabilities due to large drop surface deformations and resonant energy transfer to higher-order frequencies in the limit of low liquid viscosity. Furthermore, work by Becker, Hiller, and Kowalewski (1991) found that nonlinear effects in the oscillating drops arise when the amplitude of the fundamental mode exceeds approximately 10% of the undeformed drop radius. The phenomena are studied both experimentally and theoretically. The latter inviscid model is applicable for determining the surface tension from large-amplitude shape oscillations of low-viscosity droplets.

Patzek et al. (1991) and Basaran (1992) based their work on a numerical approach and studied the medium- and large-amplitude shape oscillations of inviscid and viscous drops, respectively. The former work used a Fourier spectral analysis and detected mode coupling of the deformed drop surface and related the frequency decrease to higher harmonics. The latter work by Basaran (*ibid.*) predicted more intense mode coupling in drops of finite viscosity than with only an infinitesimal amount of viscosity. Both works confirmed earlier numerical and theoretical results concerning the frequency decrease and the excess time in the prolate state as opposed to the oblate. In general, the drop tends to spend more time in the prolate than in the oblate state, and the effect increases with an increasing drop aspect ratio. Furthermore, Patzek et al. (1991) and Basaran (1992) found that the initial state of the drop (deformed and at rest, or spherical and disturbed by some pressure impulse) widely does not affect the frequency decrease, but

considerably influences the mode coupling and time asymmetry found in the oscillatory motion. Viscous drops spend more time in the prolate form than inviscid drops when released from an initial static deformed state, but they spend less time in the prolate form than inviscid drops when released from a steady acoustic drive (Patzek et al., 1991).

Later, an analysis of nonlinear viscous droplet oscillations solved with the Navier-Stokes equations of an incompressible fluid with a mode expansion was done by Becker, Hiller, and Kowalewski (1994). Their results agreed remarkably well with Becker, Hiller, and Kowalewski (1991) and confirmed that viscosity has a more marked effect on mode coupling compared with earlier analysis. Furthermore, they stated that the resonant mode interaction remains for asymptotically decreasing amplitudes of the fundamental deformation mode. Meradji et al. (2001) reproduced the results of Lundgren and Mansour (1988), Patzek et al. (1991) and Basaran (1992) with the CFD software *Fluid Dynamics Application Package* (FIDAP).

Mashayek and Ashgriz (1998) investigated nonlinear shape oscillations of viscous liquid drops with and without initial internal circulation. The authors concluded that the internal circulation in a drop released from an even-mode shape results in the transfer of energy between even modes, while for an initial odd mode, energy transfer exists between both even and odd modes. Experiments performed in the microgravity environment of a space shuttle flight were conducted by Wang, Anilkumar, and Lee (1996). The experimental data describe nonlinear shape oscillations of two different low-viscosity drops in different states of motion. The authors measured the frequency shift and time asymmetry of the oscillations of one (non-rotating) drop, and their findings agreed well with the theory by Tsamopoulos and Brown (1983).

Shi and Apfel (1995) based their work on the boundary integral method for the study of a deformed drop and found good correlation between their numerical results and experimental data on the frequency shift, both in micro-gravity and on the ground, within the accuracy of the experimental data. Three-dimensional oscillations with large amplitude were studied by Azuma and Yoshihara (1999). These authors used an electrically induced force to drive the oscillations. The electric charges reduce the surface ten-

sion of the droplet against its host medium, and their variation in time induces the oscillations. Small-amplitude axisymmetric shape deformation of a viscoelastic liquid drop in microgravity are theoretically analysed by Khismatullin and Nadim (2001). They performed an asymptotic analysis in the low- and high-viscosity limits and for the cases of low, moderate and large elasticities, and concluded that elastic effects give rise to a type of shape oscillation that does not depend on the surface tension. Trends of frequency and decay rate of shape oscillations are represented quantitatively well by the theory of viscous potential flow applied to oscillating drops and bubbles produced by Padrino, Funada, and Joseph (2008). Agreement is obtained for certain ranges of deformation modes and dimensionless viscosity, and is good only for large deformation modes and large viscosity since the theory is in contrast to the classical inviscid theory.

As a result of a modulation equation for nonlinear and large-amplitude oscillations of viscous drops, Smith (2010) confirmed the existence of nonlinear phenomena for amplitudes exceeding 10% of the undeformed drop radius and the impact of viscosity on the mode coupling. His further conclusions are abrupt changes in acceleration of the inviscid bulk compared to small-amplitude deformations that occur for amplitudes exceeding 20% of the drop radius. The technique allows the oscillation frequency to vary with time and reveals a quasi-periodic time history of the shape motion. For $m = 2$, the frequency decrease agrees very well with Tsamopoulos and Brown (1983). Plümacher et al. (2020) studied nonlinear drop shape oscillations of arbitrary amplitude, using the unified transform method by Fokas, which reduces the dependency of the formulation to two spatial and one time variable by a new system of equations on a time-independent domain. The authors showed in comparison drop shapes to first order for different times within one oscillation period. The oscillations are limited to small Ohnesorge number and short time, since the flow is assumed potential and the viscous effects are accounted for only at the drop surface. The work by Plümacher et al. (ibid.) is an extension of the viscous potential flow theory by Padrino, Funada, and Joseph (2008) to fully three-dimensional oscillations and to the second order.

Binary collisions of liquid droplets may cause their deformation into flat, pancake-like shapes, which then relax and return to deformed states

only slightly deviating from the spherical shape. The transition is due to viscous damping. Investigations into oscillations in this context are sparse in the literature. The investigation Brenn and Frohn (1993) presented an experimental method for characterizing shape oscillations of droplets upon collisions which may be used for determining material properties.

The nonlinear behaviour of capillary jets breaking up into droplets may cause the formation of smaller (satellite) droplets between the main droplets in the resulting droplet stream. These satellites may move relative to the main droplets, eventually taking up contact with them and merging, either backward or forward, depending on the orientation of the relative motion. The shapes of the droplets right after the formation of the common surface of the merging droplets of different size exhibit three lobes, which can be produced by droplet streams with satellites Brenn and Frohn (*ibid.*).

As an alternative to droplet streams, shape oscillations of individual droplets may be introduced by levitating a droplet in an acoustic resonator at ultrasonic frequencies, and amplitude-modulating the ultrasound at a frequency around the two-lobed resonance frequency of the droplet. Upon switching off the modulation, the droplet returns to its equilibrium shape performing damped oscillations. The oscillations are characterized by angular frequency and damping rate. Their values may yield information on the values of two material properties relevant for the oscillations, such as surface tension against the ambient medium and dynamic viscosity. Predominantly applied to Newtonian liquids, the oscillating drop method was also applied to the measurement of a time scale of polymeric liquids for which there is no established measuring method (Brenn and Teichtmeister, 2013). The basis is the solution of the characteristic equation for the argument of spherical Bessel functions involved in the equation. The argument is complex and yields two pieces of information, from which one extracts the values of the zero-shear dynamic viscosity and the deformation retardation time of the polymeric droplet liquid. This method has proven applicable to various liquids, with a preference for solutions in solvents with moderate vapour pressure so that the droplet volume changes only slightly during the experiment. Once the oscillations become large, one must account for the nonlinear effects and they should be introduced in calculations when extracting material properties

from the oscillating drop method.

The option of measuring liquid physical properties using oscillating drops has led to the development of the related experimental method. Kovalchuk et al. (2001) provided a comprehensive review of oscillating drop and bubble techniques. Liquid material properties measured are the dynamic viscosity (Egry et al., 1998; Perez et al., 2000) as well as the liquid surface tension against the ambient gas (Hiller and Kowalewski, 1989) and the interfacial tension between immiscible liquids (Hsu and Apfel, 1985). For viscoelastic drops, surface rheological properties may be measured (Apfel et al., 1997; Aske, Orr, and Sjöblom, 2002; Tian, Holt, and Apfel, 1995), while for Newtonian drops, dynamic surface tension has also been verified (Yang et al., 2014). Zhang, Harris, and Basaran (1994) investigated a growing-drop technique for measuring the dynamic surface tension of the drop liquid in contact with an ambient gas. Shape oscillations of the pendant drop prevent the measurements in the first 20 ms after formation of the interface.

The oscillating drop method is used for determining the transient shear viscosity and the dynamic surface tension of shear-thinning fluids by Yang et al. (2014). They performed measurements on free-falling drops and reported the dynamic properties of two types of shear-thinning fluids. They also showed that the dynamic viscosity greatly affects the liquid behaviour at high shear. The study by Brenn and Plohl (2015) used the damped drop shape oscillation method for measuring the deformation retardation time of polymeric liquids. In their studies they relied on the work by Brenn and Teichtmeister (2013) and showed that the values of the deformation retardation time deviate strongly from the values typically used in simulations of viscoelastic liquid flow. The latter study analytically analysed small-amplitude axisymmetric shape oscillations of viscoelastic drops in a gas using a Jeffreys model for the rheological constitutive equation of the liquid. They quantified the influence of the stress relaxation and deformation retardation time scales, and suggested an experimental method for determining one of the time scales by measuring damped drop shape oscillations. Ponce-Torres et al. (2017) studied the breakup of surfactant-laden drops, concluding from shape oscillations of satellite droplets on their surfactant content. The drop shape oscillations induced by acoustic drop levitation are used for measuring rheo-

logical material behaviour of blood in the work by Hosseinzadeh, Brugnara, and Holt (2018). The technique makes it possible to assess blood viscosity including its changes and can be used for monitoring blood rheology in sickle cell and other haematological diseases. The most recent studies on soft viscoelastic drops were carried out by Tamim and Bostwick (2021). They researched free and forced oscillations on a soft viscoelastic drop deriving a characteristic relationship for the former oscillations using Kelvin-Voigt and Maxwell models which are applicable to soft gels and polymeric fluids, respectively. The frequency-response diagram from their theoretical prediction is used in conjunction with drop-oscillation experiments as a “drop vibration rheometer”, which is proposed for future experiments. Conclusions from these findings are that the nonlinear shape oscillations for viscoelastic drops are still sparse in literature. For further understanding of the measured nonlinear drop shape oscillations described above, one needs a reliable and general theoretical method for describing the time evolution of the drop shapes for different liquid material behaviour.

1.2. Motivation for the study

The present work is motivated by the interest in and the need for a detailed analysis of the time behaviour of nonlinear drop shape oscillations, taking into account the excitation of modes higher than the mode of initial deformation, and the quasi-periodic motion resulting from mode coupling. It aims to elaborate on drops with different liquid characteristics, accounting for the complex rheological behaviour of viscoelastic liquids across viscous to inviscid drops. Even though the inviscid drops are highly investigated, a proper explanation of the quasi-periodic phenomenon of oscillating drops with large amplitudes was missing. The method of weakly nonlinear analysis is applied to investigate nonlinear axisymmetric shape oscillations of viscoelastic, viscous and inviscid liquid drops in a vacuum. We employ the method for the viscoelastic and Newtonian drop, for which there is no previous literature. Modes of initial deformation up to $m = 4$ are addressed in the study, but the analysis is not limited to it. As in Tsamopoulos and Brown (1983), our analysis is based on series expansions of the flow field

variables and the drop shape with respect to a deformation parameter. The nonlinear phenomena of both an asymmetry of the times spent in different states of drop deformation and an oscillation frequency decrease with increasing drop deformation are revealed for a wide range of drop Ohnesorge numbers relevant to technical applications. The present work on an inviscid drop reveals the quasi-periodicity of the drop shape oscillations and is thus substantially different from the findings of Tsamopoulos and Brown (*ibid.*). Oscillation frequencies arise from the general solutions of homogeneous differential equations and from the particular solutions of their inhomogeneous counterparts. The latter introduce frequencies from products of solutions of the respective lower-order approximations. The results apply to the modelling of transport processes across the oscillating drop surface, such as the quantification of the drag coefficient, as well as the rates of heat and mass transfer, where the time behaviour of the oscillatory drop motion, as presently developed, is needed. Applications in production techniques, such as containerless materials processing based on single drops (Pandey, Prabhakaran, and Basu, 2019), inkjet printing (Clavert, 2001), in air-microfluidics (Baumgartner, Brenn, and Planchette, 2020; Kamperman et al., 2018) and spray cooling (Breitenbach, Roisman, and Tropea, 2018; Kim, 2007) benefit from the results.

1.3. Objectives and work organization

The study deals with axisymmetric drop shape oscillations for different material behavior of the drop. The shape oscillations are investigated in separate chapters for the corresponding liquid material behaviors of viscoelastic, viscous and inviscid. For different liquid material behaviour, the goal is to develop solutions of the governing equations, subject to boundary conditions and initial conditions, using the weakly nonlinear method. Thus, the thesis starts with the investigation of viscoelastic drops, continues with Newtonian liquids with an overview of experimental measurements, and concludes with inviscid liquid drops.

The thesis is organized as follows:

Chapter 2 presents the equations of motion together with the boundary and initial conditions for a viscoelastic oscillating drop. It describes the weakly nonlinear method and the Oldroyd-B model, which is selected as the appropriate rheological model. Furthermore, the solutions of the first orders are developed and completed with selection of the complex angular frequency according to experiments. The chapter finishes with the conclusions.

Chapter 3 continues with the investigation of the nonlinear shape oscillations of Newtonian drops and therefore presents the governing equations as the Navier-Stokes equations, together with boundary and initial conditions. The next section refers to the equations, which are developed up to the third-order of approximation using the weakly nonlinear method. The solutions are derived and results shown in four subsections. Before the concluding section, a new section is introduced describing the settings of a measurement campaign and measured experimental data. The conclusions of the current chapter come at the end.

Chapter 4 narrows the study to inviscid liquids only. The method of deriving governing equations and finding solutions to them is similar to the methods explained in the previous chapters. The procedure of deriving the equations and finding solutions of respective orders is explained in the second section. Further, the results are divided into three subsections. The first shows the relative drop volume in time that is used as the verification of results found. The second shows the drop shapes compared with classical work for different instances in time, and the last subsection studies the time scales of the drop oscillations varying the initial mode and amplitudes. The conclusions are at the end of the chapter.

Chapter 5 at the end shows the conclusions from the work as a whole.

2. Viscoelastic drop shape oscillations

The current chapter outlines the study of viscoelastic drop oscillations. Non-linear solutions for drop shape oscillations of viscoelastic liquid drops are still sparse in the relevant literature. One of the goals of the thesis is to investigate the shape oscillations of viscoelastic drops and propose linear solutions with a method of determining angular frequency and damping rate, which influence time dependencies in higher orders of approximation. The Oldroyd-B model is employed to characterise the liquid material behaviour. In the first section, the governing equations derived by the weakly nonlinear method for the respective order of approximation are obtained. The second section shows the solutions of the first-order equations and presents the method for selecting complex conjugate angular frequencies, which determine the number of initial conditions in every order of approximation. The selection is conducted according to experiments. The chapter ends with the conclusions and outlook.

2.1. Governing equations of the oscillating drop

The present chapter outlines the weakly nonlinear analysis of shape oscillations of a viscoelastic liquid drop as sketched in figure 2.1. The drop is assumed to be axisymmetric with respect to the azimuthal coordinate ϕ of the spherical coordinate system. The liquid is treated as incompressible, and the dynamic influence from an ambient medium is neglected, i.e. we analyse the drop motion in a vacuum. Body forces are not accounted for, since the Froude number is large. The problem is formulated in spherical coordinates to account for its geometry.

2. Viscoelastic drop shape oscillations

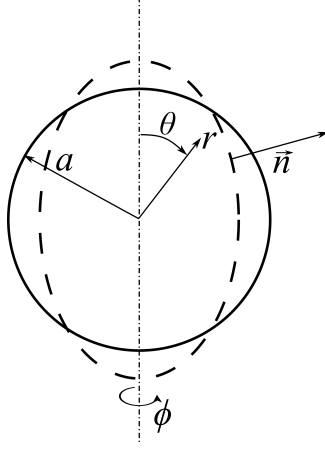


Figure 2.1.: Geometrical sketch of a liquid drop under the initial mode of deformation $m = 2$.

The equations of motion with their initial and boundary conditions are non-dimensionalized with the undeformed drop radius a , the capillary time scale $(\rho a^3 / \sigma)^{1/2}$, the capillary pressure σ/a and the viscous stress $\mu_0(\sigma / \rho a^3)^{1/2}$ for length, time, pressure and stress respectively. Here, ρ is the liquid density, σ the air-liquid interfacial tension and μ_0 the zero-shear liquid dynamic viscosity of the drop. The drop surface is described as the place where $r_s(\theta, t) = 1 + \eta(\theta, t)$, with the non-dimensional deformation η against the undisturbed spherical shape (cf. Figure 2.1). For the problem at hand, the equation of continuity and the two components of the momentum equation in the radial and polar angular directions, (r) and (θ) , read

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_r) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_\theta \sin \theta) = 0, \quad (2.1)$$

$$\begin{aligned} \frac{\partial u_r}{\partial t} + u_r \frac{\partial u_r}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_r}{\partial \theta} - \frac{u_\theta^2}{r} = - \frac{\partial p}{\partial r} \\ + Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \tau_{rr}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{r\theta} \sin \theta) - \frac{\tau_{\theta\theta} + \tau_{\phi\phi}}{r} \right], \end{aligned} \quad (2.2)$$

$$\begin{aligned} \frac{\partial u_\theta}{\partial t} + u_r \frac{\partial u_\theta}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_\theta}{\partial \theta} + \frac{u_r u_\theta}{r} = -\frac{1}{r} \frac{\partial p}{\partial \theta} + \\ + Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \tau_{r\theta}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{\theta\theta} \sin \theta) + \frac{\tau_{r\theta}}{r} - \frac{\cot \theta}{r} \tau_{\phi\phi} \right]. \end{aligned} \quad (2.3)$$

The ϕ -component of the momentum equation vanishes due to the axisymmetric drop shape and the absence of swirl. The characteristic dimensionless parameter measuring viscosity influences is the Ohnesorge number, formulated as $Oh_0 = \mu_0 / (\sigma a \rho)^{1/2}$, with the zero-shear liquid dynamic viscosity μ_0 . The viscoelastic behaviour upon rates of deformation is represented by the Oldroyd-B model. Its liquid material is characterised by the nondimensional rheological constitutive equation (RCE)

$$\boldsymbol{\tau} + De_1 \overset{\nabla}{\boldsymbol{\tau}} = 2 \left(\boldsymbol{D} + De_2 \overset{\nabla}{\boldsymbol{D}} \right), \quad (2.4)$$

where $\boldsymbol{\tau}$ and $\boldsymbol{D}$ are the extra-stress and rate-of-deformation tensors, respectively, the latter defined with the velocity gradient tensor $\vec{\nabla} \vec{v}$ as

$$\boldsymbol{D} = \frac{1}{2} \left(\vec{\nabla} \vec{v} + \vec{\nabla} \vec{v}^T \right). \quad (2.5)$$

The Deborah numbers $De_1 = \lambda_1 (\sigma / \rho a^3)^{1/2}$ and $De_2 = \lambda_2 (\sigma / \rho a^3)^{1/2}$ represent the nondimensional stress relaxation and deformation retardation times of the liquid, respectively. The upper-convected derivative $\overset{\nabla}{\boldsymbol{A}}$ of a tensor $\boldsymbol{A}$ is given as

$$\overset{\nabla}{\boldsymbol{A}} = \frac{d\boldsymbol{A}}{dt} - \vec{\nabla} \vec{v} \cdot \boldsymbol{A} - \boldsymbol{A} \cdot \vec{\nabla} \vec{v}^T, \quad (2.6)$$

where $d\boldsymbol{A}/dt$ is the material derivative.

The above set of equations is solved subject to initial and boundary conditions. The kinematic boundary condition describes the material rate of deformation of the drop surface which equals the radial velocity component at the place of the deformed surface, i.e.,

$$u_r = \frac{d\eta}{dt} = \frac{\partial \eta}{\partial t} + \frac{u_\theta}{r} \frac{\partial \eta}{\partial \theta} \quad \text{at } r = 1 + \eta. \quad (2.7)$$

Since we assume that, in a vacuum, the dynamic viscosity is negligible, the first dynamic boundary condition states that the shear stress at the

2. Viscoelastic drop shape oscillations

drop surface is zero i.e, momentum cannot be transferred across the drop boundary at an appreciable rate. The second dynamic boundary condition forms the stress normal to the drop surface, which is composed from the flow-induced pressure and the viscoelastic normal stress. The normal stress differs across the interface by the stress due to the surface tension. Hence, the zero-shear stress boundary condition reads

$$(\vec{n} \cdot \boldsymbol{\tau}) \times \vec{n} = \vec{0} \quad \text{at } r = 1 + \eta, \quad (2.8)$$

where one gives the outward unit normal vector $\vec{n}$ as

$$\vec{n} = \frac{1}{|\vec{\nabla} F|} \vec{\nabla} F \quad \text{with } F = r - 1 - \eta(\theta, t) = 0, \quad (2.9)$$

and the extra stress tensor in (2.8) is the one for the incompressible Oldroyd-B fluid. The corresponding normal stress boundary condition reads

$$-p + Oh_0 (\vec{n} \cdot \boldsymbol{\tau}) \cdot \vec{n} + \left(\vec{\nabla} \cdot \vec{n} \right) = 0 \quad \text{at } r = 1 + \eta. \quad (2.10)$$

We obtain the divergence of the normal unit vector on the drop surface in this equation as

$$\left(\vec{\nabla} \cdot \vec{n} \right) = \frac{1}{r} \frac{2 + 3 \left(\frac{1}{r} \frac{\partial \eta}{\partial \theta} \right)^2}{\left[1 + \left(\frac{1}{r} \frac{\partial \eta}{\partial \theta} \right)^2 \right]^{3/2}} - \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left[\frac{\frac{\partial \eta}{\partial \theta}}{\left(1 + \left(\frac{1}{r} \frac{\partial \eta}{\partial \theta} \right)^2 \right)^{1/2}} \sin \theta \right], \quad (2.11)$$

at $r = 1 + \eta$.

These equations are analyzed in a weakly nonlinear form. It implies that the two velocity components, the pressure and the extra stress tensor in the flow field, as well as the deformed interface shape, are expanded in power series with respect to the small deformation parameter η_0 . We therefore compose the expansions of the flow field properties, for example u_r , p , $\boldsymbol{\tau}$, $\boldsymbol{D}$

and η , as

$$u_r = u_{r1}\eta_0 + u_{r2}\eta_0^2 + u_{r3}\eta_0^3 \dots, \quad (2.12)$$

$$p = p_0 + p_1\eta_0 + p_2\eta_0^2 + p_3\eta_0^3 \dots, \quad (2.13)$$

$$\tau = \tau_1\eta_0 + \tau_2\eta_0^2 + \tau_3\eta_0^3 \dots, \quad (2.14)$$

$$D = D_1\eta_0 + D_2\eta_0^2 + D_3\eta_0^3 \dots, \quad (2.15)$$

$$\eta = \eta_1\eta_0 + \eta_2\eta_0^2 + \eta_3\eta_0^3 \dots. \quad (2.16)$$

For convergence of these series expansions, the deformation parameter η_0 must be small. One essential difference between the present weakly nonlinear analysis and the linear one is that the boundary conditions are satisfied on the deformed drop surface, not on the undeformed spherical shape as in the linear case. Accounting for this, one expands the deformed shapes in Taylor series, which still allow for the functions in the boundary conditions to be evaluated on the undeformed drop surface, for example for u_r , p and τ ,

$$u_r|_{r=1+\eta} = u_r|_{r=1} + \left. \frac{\partial u_r}{\partial r} \right|_{r=1} \eta + \dots, \quad (2.17)$$

$$p|_{r=1+\eta} = p|_{r=1} + \left. \frac{\partial p}{\partial r} \right|_{r=1} \eta + \dots, \quad (2.18)$$

$$\tau|_{r=1+\eta} = \tau|_{r=1} + \left. \frac{\partial \tau}{\partial r} \right|_{r=1} \eta + \dots. \quad (2.19)$$

These approaches are substituted into the equations of motion (2.1) - (2.3), into the RCE (2.4) and into the boundary conditions (2.7), (2.8) and (2.10). Representing the flow properties and their derivatives as given in (2.12) through (2.19), one obtains sets of first-, second- and third-order equations of motion, with the boundary conditions, consisting of all the terms with the deformation parameter η_0 to the first, second and third powers, respectively.

An initially imposed surface deformation against the spherical shape induces the drop motion. A Legendre polynomial of degree m with the amplitude η_0 governs the initial deformation, and the surface is initially at rest. One calculates the volume of the deformed drop, which leads to the

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expression

$$\begin{aligned}
 r_s(\theta, 0) &= 1 + \eta(\theta, 0) \\
 &= \left(1 + \frac{3\eta_0^2}{2m+1} + \frac{1}{2}\eta_0^3 \int_{-1}^1 P_m(\cos \theta)^3 d(\cos \theta) \right)^{-1/3} + \eta_0 P_m(\cos \theta) \\
 &= 1 + \eta_0 P_m(\cos \theta) - \eta_0^2 \frac{1}{2m+1} - \frac{\eta_0^3}{6} \int_{-1}^1 P_m(\cos \theta)^3 d(\cos \theta) \mp \dots,
 \end{aligned} \tag{2.20}$$

for the initial non-dimensional drop shape. The factor in front of η_0 to the respective power corresponds to the approximation order needing the correction so that the surface shape represents the correct drop volume. In addition, for the solution development of the problem of viscoelastic and Newtonian drop shape oscillations, the values of the angular frequencies for modes $m = 0$ and $m = 1$ are needed, which, in contrast to all the other modes, are not given by the solution of the characteristic equation of the drop. To overcome this problem, one uses the requirement that the centre of mass of the drop remains at its initial position

$$\begin{aligned}
 z_m &= \frac{\pi}{2V} \int_{-1}^1 \left[1 + 4\eta_0\eta_1 + \eta_0^2 (6\eta_1^2 + 4\eta_2) \right. \\
 &\quad \left. + \eta_0^3 (4\eta_1^3 + 12\eta_1\eta_2 + 4\eta_3) + \dots \right] x dx,
 \end{aligned} \tag{2.21}$$

at all times (where $x = \cos \theta$). This statement is physically justified because there is no resultant force to move the drop. We will come back to this later in the course of the development of the solutions.

2.1.1. First-order equations

To obtain the first-order equations with their boundary and initial conditions, the above series expansions are introduced into the respective equations, and all the terms with the parameter η_0 to the first power are collected. The first-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_{r1}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 1} \sin \theta) = 0, \tag{2.22}$$

$$\frac{\partial u_{r1}}{\partial t} = -\frac{\partial p_1}{\partial r} + \quad (2.23)$$

$$+ Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \tau_{rr1}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{r\theta 1} \sin \theta) - \frac{\tau_{\theta\theta 1} + \tau_{\phi\phi 1}}{r} \right] ,$$

$$\frac{\partial u_{\theta 1}}{\partial t} = -\frac{1}{r} \frac{\partial p_1}{\partial \theta} + \quad (2.24)$$

$$+ Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \tau_{r\theta 1}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{\theta\theta 1} \sin \theta) + \frac{\tau_{r\theta 1}}{r} - \frac{\cot \theta}{r} \tau_{\phi\phi 1} \right] .$$

The RCE of first order becomes

$$\tau_1 + De_1 \frac{\partial \tau_1}{\partial t} = 2 \left(\mathbf{D}_1 + De_2 \frac{\partial \mathbf{D}_1}{\partial t} \right) . \quad (2.25)$$

2.1.2. First-order boundary and initial conditions

As the boundary conditions of first order, to be satisfied at $r = 1$, we obtain

$$u_{r1} = \frac{\partial \eta_1}{\partial t} , \quad (2.26)$$

$$\tau_{r\theta 1} = 0 , \quad (2.27)$$

$$-p_1 + Oh_0 \tau_{rr1} - \left(2\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right) = 0 , \quad (2.28)$$

respectively. Furthermore, the initial conditions of first order are

$$\eta_1(\theta, 0) = P_m(\cos \theta) , \quad \frac{\partial \eta_1}{\partial t}(\theta, 0) = 0 . \quad (2.29)$$

The first initial condition determines the initial shape of the deformed drop which is given by a Legendre polynomial of order m , and the second one states that the drop surface is initially at rest.

2.1.3. Second-order equations

The second-order equations with their boundary and initial conditions are obtained as all the terms from the expansion with the parameter η_0 to the

second power. The second-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_{r2}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 2} \sin \theta) = 0, \quad (2.30)$$

$$\begin{aligned} \frac{\partial u_{r2}}{\partial t} - Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \tau_{rr2}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{r\theta 2} \sin \theta) - \frac{\tau_{\theta\theta 2} + \tau_{\phi\phi 2}}{r} \right] \\ + \frac{\partial p_2}{\partial r} = \frac{u_{\theta 1}^2}{r} - u_{r1} \frac{\partial u_{r1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{r1}}{\partial \theta}, \end{aligned} \quad (2.31)$$

$$\begin{aligned} \frac{\partial u_{\theta 2}}{\partial t} - Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \tau_{r\theta 2}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{\theta\theta 2} \sin \theta) + \frac{\tau_{r\theta 2}}{r} - \frac{\cot \theta}{r} \tau_{\phi\phi 2} \right] \\ + \frac{1}{r} \frac{\partial p_2}{\partial \theta} = -u_{r1} \frac{\partial u_{\theta 1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} - \frac{u_{r1} u_{\theta 1}}{r}. \end{aligned} \quad (2.32)$$

The RCE of second order may be written in the form

$$\begin{aligned} \boldsymbol{\tau}_2 + De_1 \frac{\partial \boldsymbol{\tau}_2}{\partial t} = 2 \left(\mathbf{D}_2 + De_2 \frac{\partial \mathbf{D}_2}{\partial t} \right) \\ - De_1 \left[\left(\vec{v}_1 \cdot \vec{\nabla} \right) \boldsymbol{\tau}_1 - \vec{\nabla} \vec{v}_1 \cdot \boldsymbol{\tau}_1 - \boldsymbol{\tau}_1 \cdot \vec{\nabla} \vec{v}_1^T \right] \\ + 2De_2 \left[\left(\vec{v}_1 \cdot \vec{\nabla} \right) \mathbf{D}_1 - \vec{\nabla} \vec{v}_1 \cdot \mathbf{D}_1 - \mathbf{D}_1 \cdot \vec{\nabla} \vec{v}_1^T \right]. \end{aligned} \quad (2.33)$$

2.1.4. Second-order boundary and initial conditions

The second-order kinematic boundary condition, as well as the boundary conditions stating zero shear and normal stresses, to be satisfied at $r = 1$, are

$$u_{r2} - \frac{\partial \eta_2}{\partial t} = \frac{u_{\theta 1}}{r} \frac{\partial \eta_1}{\partial \theta} - \eta_1 \frac{\partial u_{r1}}{\partial r}, \quad (2.34)$$

$$\tau_{r\theta 2} = -\eta_1 \frac{\partial \tau_{r\theta 1}}{\partial r} - (\tau_{rr1} - \tau_{\theta\theta 1}) \frac{1}{r} \frac{\partial \eta_1}{\partial \theta}, \quad (2.35)$$

$$-p_2 + Oh_0 \tau_{rr2} - \left(2\eta_2 + \frac{\partial \eta_2}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_2}{\partial \theta^2} \right) = \eta_1 \frac{\partial p_1}{\partial r} \quad (2.36)$$

$$-Oh_0 \left[\eta_1 \frac{\partial \tau_{rr1}}{\partial r} - 2 \frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \tau_{r\theta 1} \right] - \left(2\eta_1^2 + 2\eta_1 \frac{\partial \eta_1}{\partial \theta} \cot \theta + 2\eta_1 \frac{\partial^2 \eta_1}{\partial \theta^2} \right),$$

respectively. Furthermore, the initial conditions of second order are

$$\eta_2(\theta, 0) = -\frac{1}{2m+1}, \quad \frac{\partial \eta_2}{\partial t}(\theta, 0) = 0. \quad (2.37)$$

The first initial condition ensures volume conservation, the second one contributes to the fact that the drop surface is initially at rest.

Steered by the work by Zrnić, Berglez, and Brenn (2022) and Zrnić and Brenn (2021b), which analyse the viscous and inviscid cases, we conclude that solving the sets of first- and second-order equations, together with their boundary and initial conditions, will reveal two known nonlinear effects (except for $m = 2$ in the inviscid case). These effects are a frequency change and an excess time the drop spends in an elongated state, which can be represented as functions of the initial deformation amplitude. In the inviscid and viscous cases, the analysis had to be extended to the third order of approximation in order to represent these effects. Therefore, the extension to the third order is also done in the present chapter.

2.1.5. Third-order equations

The third-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 u_{r3} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 3} \sin \theta) = 0, \quad (2.38)$$

$$\frac{\partial u_{r3}}{\partial t} - Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \tau_{rr3} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{r\theta 3} \sin \theta) - \frac{\tau_{\theta\theta 3} + \tau_{\phi\phi 3}}{r} \right] \quad (2.39)$$

$$\begin{aligned} & + \frac{\partial p_3}{\partial r} = 2 \frac{u_{\theta 1} u_{\theta 2}}{r} - u_{r1} \frac{\partial u_{r2}}{\partial r} - u_{r2} \frac{\partial u_{r1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{r2}}{\partial \theta} - \frac{u_{\theta 2}}{r} \frac{\partial u_{r1}}{\partial \theta}, \\ & \frac{\partial u_{\theta 3}}{\partial t} - Oh_0 \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \tau_{r\theta 3} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{\theta\theta 3} \sin \theta) + \frac{\tau_{r\theta 3}}{r} - \frac{\cot \theta}{r} \tau_{\phi\phi 3} \right] \\ & + \frac{1}{r} \frac{\partial p_3}{\partial \theta} = -u_{r1} \frac{\partial u_{\theta 2}}{\partial r} - u_{r2} \frac{\partial u_{\theta 1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 2}}{\partial \theta} - \frac{u_{\theta 2}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} \\ & - \frac{u_{r1} u_{\theta 2}}{r} - \frac{u_{r2} u_{\theta 1}}{r}. \end{aligned} \quad (2.40)$$

The RCE of third order reads

$$\begin{aligned} \boldsymbol{\tau}_3 + De_1 \frac{\partial \boldsymbol{\tau}_3}{\partial t} - 2 \left(\mathbf{D}_3 + De_2 \frac{\partial \mathbf{D}_3}{\partial t} \right) = -De_1 \left[\left(\vec{v}_1 \cdot \vec{\nabla} \right) \boldsymbol{\tau}_2 \right. \\ \left. + \left(\vec{v}_2 \cdot \vec{\nabla} \right) \boldsymbol{\tau}_1 - \vec{\nabla} \vec{v}_1 \cdot \boldsymbol{\tau}_2 - \vec{\nabla} \vec{v}_2 \cdot \boldsymbol{\tau}_1 - \boldsymbol{\tau}_1 \cdot \vec{\nabla} \vec{v}_2^T - \boldsymbol{\tau}_2 \cdot \vec{\nabla} \vec{v}_1^T \right] \end{aligned} \quad (2.41)$$

$$\begin{aligned}
 & + 2De_2 \left[\left(\vec{v}_1 \cdot \vec{\nabla} \right) \mathbf{D}_2 + \left(\vec{v}_2 \cdot \vec{\nabla} \right) \mathbf{D}_1 - \vec{\nabla} \vec{v}_1 \cdot \mathbf{D}_2 - \vec{\nabla} \vec{v}_2 \cdot \mathbf{D}_1 \right. \\
 & \left. - \mathbf{D}_1 \cdot \vec{\nabla} \vec{v}_2^T - \mathbf{D}_2 \cdot \vec{\nabla} \vec{v}_1^T \right].
 \end{aligned}$$

2.1.6. Third-order boundary and initial conditions

The third-order kinematic boundary condition, as well as the boundary conditions stating zero shear and normal stresses to be satisfied at $r = 1$, are

$$\begin{aligned}
 u_{r3} - \frac{\partial \eta_3}{\partial t} &= \frac{u_{\theta 1}}{r} \frac{\partial \eta_2}{\partial \theta} + \frac{u_{\theta 2}}{r} \frac{\partial \eta_1}{\partial \theta} + \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) \frac{\partial \eta_1}{\partial \theta} \eta_1 - \frac{\partial u_{r2}}{\partial r} \eta_1 \\
 & - \frac{\partial u_{r1}}{\partial r} \eta_2 - \frac{1}{2} \frac{\partial^2 u_{r1}}{\partial r^2} \eta_1^2, \tag{2.42}
 \end{aligned}$$

$$\begin{aligned}
 \tau_{r\theta 3} &= -\eta_1 \frac{\partial \tau_{r\theta 2}}{\partial r} - \eta_2 \frac{\partial \tau_{r\theta 1}}{\partial r} - (\tau_{rr2} - \tau_{\theta\theta 2}) \frac{1}{r} \frac{\partial \eta_1}{\partial \theta} - \\
 & - (\tau_{rr1} - \tau_{\theta\theta 1}) \frac{1}{r} \frac{\partial \eta_2}{\partial \theta} - \left[\frac{\partial}{\partial r} \left(\frac{\tau_{rr1}}{r} \right) - \frac{\partial}{\partial r} \left(\frac{\tau_{\theta\theta 1}}{r} \right) \right] \frac{1}{2} \frac{\partial \eta_1^2}{\partial \theta} - \\
 & - \frac{1}{2} \frac{\partial^2 \tau_{r\theta 1}}{\partial r^2} \eta_1^2 + \tau_{r\theta 1} \left(\frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \right)^2, \tag{2.43}
 \end{aligned}$$

$$\begin{aligned}
 -p_3 + Oh_0 \tau_{rr3} &- \left(2\eta_3 + \frac{\partial \eta_3}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_3}{\partial \theta^2} \right) = \frac{\partial p_1}{\partial r} \eta_2 + \frac{\partial p_2}{\partial r} \eta_1 \\
 & + \frac{1}{2} \frac{\partial^2 p_1}{\partial r^2} \eta_1^2 - Oh_0 \left\{ \frac{\partial \tau_{rr1}}{\partial r} \eta_2 + \frac{\partial \tau_{rr2}}{\partial r} \eta_1 - \frac{2}{r} \frac{\partial \eta_1}{\partial \theta} \tau_{r\theta 2} - \frac{2}{r} \frac{\partial \eta_2}{\partial \theta} \tau_{r\theta 1} - \right. \\
 & - \frac{\partial \eta_1^2}{\partial \theta} \frac{\partial}{\partial r} \left(\frac{\tau_{r\theta 1}}{r} \right) - \left(\frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \right)^2 (\tau_{rr1} - \tau_{\theta\theta 1}) + \frac{1}{2} \frac{\partial^2 \tau_{rr1}}{\partial r^2} \eta_1^2 \left. \right\} - \\
 & - \eta_1^3 - 2\eta_1 \left(\eta_2 + \frac{\partial \eta_2}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_2}{\partial \theta^2} \right) \\
 & - 2\eta_2 \left(\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right) - \frac{1}{2} \left(\frac{\partial \eta_1}{\partial \theta} \right)^2 \left(\frac{\partial \eta_1}{\partial \theta} \cot \theta + 3 \frac{\partial^2 \eta_1}{\partial \theta^2} \right) \\
 & + 3\eta_1^2 \left(\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right), \tag{2.44}
 \end{aligned}$$

respectively. Furthermore, the initial conditions of third order are

$$\eta_3(\theta, 0) = -\frac{1}{3} \begin{pmatrix} m & m & m \\ 0 & 0 & 0 \end{pmatrix}^2, \quad \frac{\partial \eta_3}{\partial t}(\theta, 0) = 0, \quad (2.45)$$

where the first initial condition is formulated using a 3- j symbol from Wigner (1993). Solving the sets of equations up to third order, together with their initial and boundary conditions, will reveal the weakly nonlinear drop oscillation dynamics.

2.2. Solutions of the governing equations

The first-order equations describe the linear problem. Well-known linear solutions, together with the characteristic equation of the viscoelastic drop in a vacuum, are due to Khismatullin and Nadim (2001) and Brenn and Teichtmeister (2013). Similar solutions are obtained in this section. Since only two-dimensional flow fields are investigated, we apply the method of the Stokesian streamfunction for determining the first-order velocity field. The streamfunction $\psi(r, \theta, t)$ is defined by its relations to the two velocity components u_{r1} and $u_{\theta 1}$ as (Bird, Stewart, and Lightfoot, 1962)

$$u_{r1} = -\frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta} \quad \text{and} \quad u_{\theta 1} = \frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r}. \quad (2.46)$$

These first-order velocity components satisfy the continuity equation identically.

The first-order drop surface shape is governed by the Legendre polynomial of the initial deformation. The solution is therefore sought in the form

$$\eta_1(\theta, t) = \hat{\eta}_1 P_m(\cos \theta) e^{-\alpha_m t}, \quad (2.47)$$

with the first-order initial surface amplitude $\hat{\eta}_1$ and the first-order complex angular frequency α_m for the deformation mode m .

For finding the first-order solutions of the equations of motion, we first solve the RCE of first order. Given the exponential dependency of all the flow

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field variables on time via $\exp(-\alpha_m t)$, we find for the extra-stress tensor of first order

$$\boldsymbol{\tau}_1 = 2 \frac{1 - De_2 \alpha_m}{1 - De_1 \alpha_m} \mathbf{D}_1 =: 2\beta_1 \mathbf{D}_1. \quad (2.48)$$

This means that, at first order, the extra-stress tensor of the viscoelastic fluid differs from the Newtonian material by just a frequency-dependent factor β_1 in front of the rate-of-deformation tensor. This fluid is therefore formally identical to a Newtonian one, so that all the first-order results obtained in our previous paper (Zrnić, Berglez, and Brenn, 2022) apply here, only with the Ohnesorge number Oh in those equations replaced by $Oh_v := \beta_1 Oh_0$. We therefore apply those previous results as follows.

Taking the curl of the first-order momentum equation in a vectorial form using equations (2.23) and (2.24), we obtain the fourth-order partial differential equation

$$\left(\frac{1}{Oh_v} \frac{\partial}{\partial t} - E_s^2 \right) (E_s^2 \psi) = 0, \quad (2.49)$$

with the operator (Bird, Stewart, and Lightfoot, 1962)

$$E_s^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin \theta}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \right). \quad (2.50)$$

The resulting stream function ψ consists of two contributions ψ_1 and ψ_2 (Tomotika, 1935) represented by

$$\psi_1 = C_{1m} r^{m+1} \sin^2 \theta P'_m(\cos \theta) e^{-\alpha_m t}, \quad (2.51)$$

$$\psi_2 = C_{2m} q r j_m(qr) \sin^2 \theta P'_m(\cos \theta) e^{-\alpha_m t}, \quad (2.52)$$

where $P'_m(\cos \theta)$ is the first-order derivative of the Legendre polynomial P_m with respect to its argument and j_m is a spherical Bessel function of the first kind and order m . In its argument we have defined $q = \sqrt{\alpha_m / Oh_v}$. The radial and angular components of the first-order velocity vector follow as derivatives of the stream function

$$u_{r1} = - \left[C_{1m} r^{m-1} + C_{2m} q^2 \frac{j_m(qr)}{qr} \right] m(m+1) P_m(\cos \theta) e^{-\alpha_m t}, \quad (2.53)$$

and

$$u_{\theta 1} = \left[C_{1m}(m+1)r^{m-1} + C_{2m}q^2 \left((m+1)\frac{j_m(qr)}{qr} - j_{m+1}(qr) \right) \right] \sin \theta P'_m(\cos \theta) e^{-\alpha_m t}, \quad (2.54)$$

respectively. The two integration constants C_{1m} and C_{2m} are determined by the first-order kinematic and zero shear stress boundary conditions and read

$$C_{1m} = \frac{\hat{\eta}_1 \alpha_m}{m(m+1)} \left[1 + \frac{2(m^2-1)}{2qj_{m+1}(q)/j_m(q) - q^2} \right], \quad (2.55)$$

$$C_{2m} = -\frac{2(m-1)\hat{\eta}_1 \alpha_m}{mq[2qj_{m+1}(q) - q^2j_m(q)]}. \quad (2.56)$$

The first-order pressure field is obtained by integration of one component of the momentum equation as

$$p_1 = -C_{1m}(m+1)\alpha_m r^m P_m(\cos \theta) e^{-\alpha_m t}. \quad (2.57)$$

With the velocity and pressure fields known, the zero normal stress boundary condition of first order (2.28) yields the characteristic equation of the drop, which reads

$$\frac{\alpha_{m,0}^2}{\alpha_m^2} = \frac{2(m^2-1)}{q^2 - 2qj_{m+1}/j_m} - 1 + \frac{2m(m-1)}{q^2} \left[1 + \frac{2(m+1)j_{m+1}/j_m}{2j_{m+1}/j_m - q} \right]. \quad (2.58)$$

This equation determines the complex angular frequency α_m , where we have denoted $\alpha_{m,0} = [m(m-1)(m+2)]^{1/2}$. The equation is identical to the results of Lamb (1881) and Chandrasekhar (1959). The spherical Bessel functions are taken at the value q of their arguments. We can express the square q^2 of the argument of the spherical Bessel functions in a non-dimensional form as

$$q^2 = \alpha_{m,0} \frac{\Omega_m}{Oh_0} \frac{1 - \Omega_m De_1 \alpha_{m,0}}{1 - \Omega_m De_2 \alpha_{m,0}}, \quad (2.59)$$

where $\Omega_m = \alpha_m / \alpha_{m,0}$. This form is identical to Brenn and Teichtmeister (2013). For $De_1 = 0$ and $De_2 = 0$, equation (2.59) reduces to the statement for the Newtonian case.

In the next section, the solutions of the characteristic equation will be shown and their importance will be addressed. The method of their selection with the use of experiments will be proposed.

2.3. Discussion of the characteristic equation and outlook

Khismatullin and Nadim (2001) point out that there exists an infinite manifold of solutions of the characteristic equation of a viscoelastic drop for a given range of Ohnesorge numbers and fixed Deborah numbers. One manifold of solutions of the complex angular frequencies for different range of Deborah and Ohnesorge numbers are relieved in the work by Brenn and Teichtmeister (2013), but not their complex conjugated part. In the method for measuring the deformation retardation time of viscoelastic liquids, Brenn and Plohl (2015) found a large number of solutions for the argument q , in agreement with the finding by Khismatullin and Nadim.

The characteristic equation is transcendental and highly nonlinear. We show an analysis of the equation for the case corresponding to a 2 mm 0.5 wt% Praestol 2500 solution drop. Measured liquid drop properties of aqueous solutions of the polyacrylamide Praestol 2500 are given in table 2.1. A similar analysis for a 1.97 mm 0.3 wt% Praestol 2500 solution drop was conducted by Brenn and Plohl (ibid.). First, we define a function as

$$F(q) = \frac{2(m^2 - 1)}{q^2 - 2qj_{m+1}/j_m} - 1 + \frac{2m(m - 1)}{q^2} \left[1 + \frac{2(m + 1)j_{m+1}/j_m}{2j_{m+1}/j_m - q} \right], \quad (2.60)$$

which is the right-hand side of the characteristic equation (2.58). We plot the real and imaginary parts of $F(q)$ as functions of the real part of the argument q in figures 2.2 (a) and (b), respectively. The values of the argument q are computed using equation (2.59) and ranged such that the imaginary part is fixed to a constant value (0.0236) corresponding to the 2 mm 0.5 wt% Praestol 2500 solution drop. The values of the real part of the argument q are between 0 and 40. The horizontal red dashed line presents the left-hand side of the characteristic equation (2.58) for the corresponding case. The

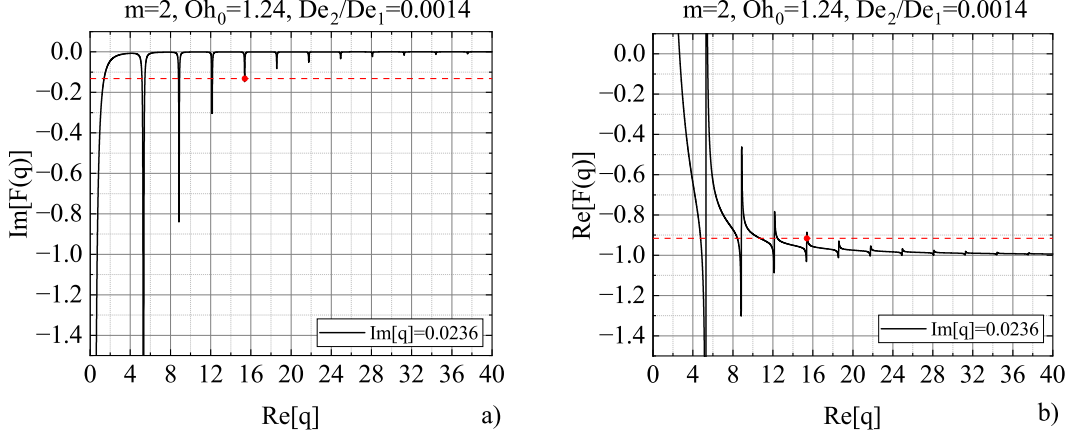


Figure 2.2.: The function $F(q)$ for different values of its real part of the argument q for $m = 2$. (a) The imaginary and (b) the real parts of the function. The horizontal lines at $Im[F(q)] = -0.132$ and $Re[F(q)] = -0.916$ represent the left hand side of equation (2.58) for a 2 mm 0.5 wt% Praestol 2500 solution drop studied.

intersections between dashed and solid lines represent the solutions of the characteristic equation for the corresponding case and because of them we obtain more than one set of complex conjugate pairs of angular frequency. The intersections indicated with red circles present the complex conjugate pair of numbers, which correspond to the values from the experiments that are shown with black squares in figures 2.3 (e) and (f) and 2.4 (a) and (b). The spikes in figures 2.2 (a) and (b) originate from the value of the imaginary part of the argument q . The function $F(q)$ exhibits sharp peaks for values of the imaginary part of q which are much smaller than 1. The same observation was pointed out by Brenn and Plohl (ibid.).

In figure 2.3, we show the first five sets of complex conjugate pairs of angular frequencies as the solutions of the characteristic equation and the complex conjugate pairs selected according to the experimental data in Brenn and Plohl (ibid.). The experiments are based on droplet oscillations, which were analysed using an ultrasonic resonator for levitating single drops. The description of the experimental method for determining the complex angular frequency is detailed in ibid. Due to the small solute contents, the density ρ for all solute mass fractions equals 1000 kg/m^3 . Measured surface tension σ , droplet radius a , stress relaxation time λ_1 and deformation retardation time

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λ_2 from table 2.1 and the density are used to compute the Deborah numbers.

For the SMF of 0.1 wt%, 0.3 wt% and 0.5 wt%, the values of De_1 are 8, 22.1 and 33.9, respectively. The values of De_2 one obtains by the ratios of Deborah numbers in figure 2.3. The solutions of the characteristic equation are defined as nondimensional complex angular frequencies Ω_2 because the drop shape oscillations are excited in the fundamental initial deformation mode $m = 2$. The frequency and the damping rate of the drop shape oscillations are presented by the imaginary and the real parts of the complex angular frequency, respectively. In figure 2.3, five sets of nondimensional complex conjugate angular frequencies Ω_2 for the fundamental initial deformation mode, the range of Ohnesorge numbers between 0.03 and 3, and SMF of 0.1, 0.3 and 0.5 wt% are shown with dashed lines.

Since the characteristic equation yields an infinite number of roots for fixed Deborah numbers, one must select solutions appropriate for representing the shape oscillations. We compare the solutions with experimental results. For the 0.1, 0.3 and 0.5 wt% aqueous Praestol 2500 solution drops in table 2.1, the measured frequencies are $f = 114.5$ Hz, $f = 129.9$ Hz and $f = 113.1$ Hz, respectively, and the damping rates are $\alpha_{2,r} = 35.2$ s⁻¹, $\alpha_{2,r} = 36$ s⁻¹ and $\alpha_{2,r} = 51$ s⁻¹, respectively. Therefore the complex angular frequencies $\alpha_2^* = \alpha_{2,r} + i2\pi f$ are known. The uncertainties of the measured oscillation frequency f and the damping rate $\alpha_{2,r}$ are $\pm 1\%$ and $\pm 10\%$, respectively. The measured nondimensional complex angular frequency Ω_2 one formulates as $\alpha_2^*/\alpha_{2,0}^*$, where $\alpha_{2,0}^*$ is the dimensional Rayleigh frequency. The uncertainties of the measured radius a , the surface tension σ and the density ρ are $\pm 0.6\%$, $\pm 0.5\%$ and $\pm 0.05\%$, respectively. This results in the uncertainty of about $\pm 2\%$ of the squared Rayleigh frequency $\alpha_{2,0}^2$ for $m = 2$. The Ohnesorge

Table 2.1.: Measured properties of the aqueous Praestol 2500 solutions and measured drop radius.

SMF [wt%]	μ_0 [Pa s]	λ_1 [s]	λ_2 [s]	σ [N/m]	a [m]
0.1	0.017	0.033	$0.4 \cdot 10^{-4}$	0.072	$1.07 \cdot 10^{-3}$
0.3	0.043	0.078	$0.9 \cdot 10^{-4}$	0.073	$0.97 \cdot 10^{-3}$
0.5	0.35	0.14	$0.19 \cdot 10^{-3}$	0.074	$1.08 \cdot 10^{-3}$

2.3. Discussion of the characteristic equation and outlook

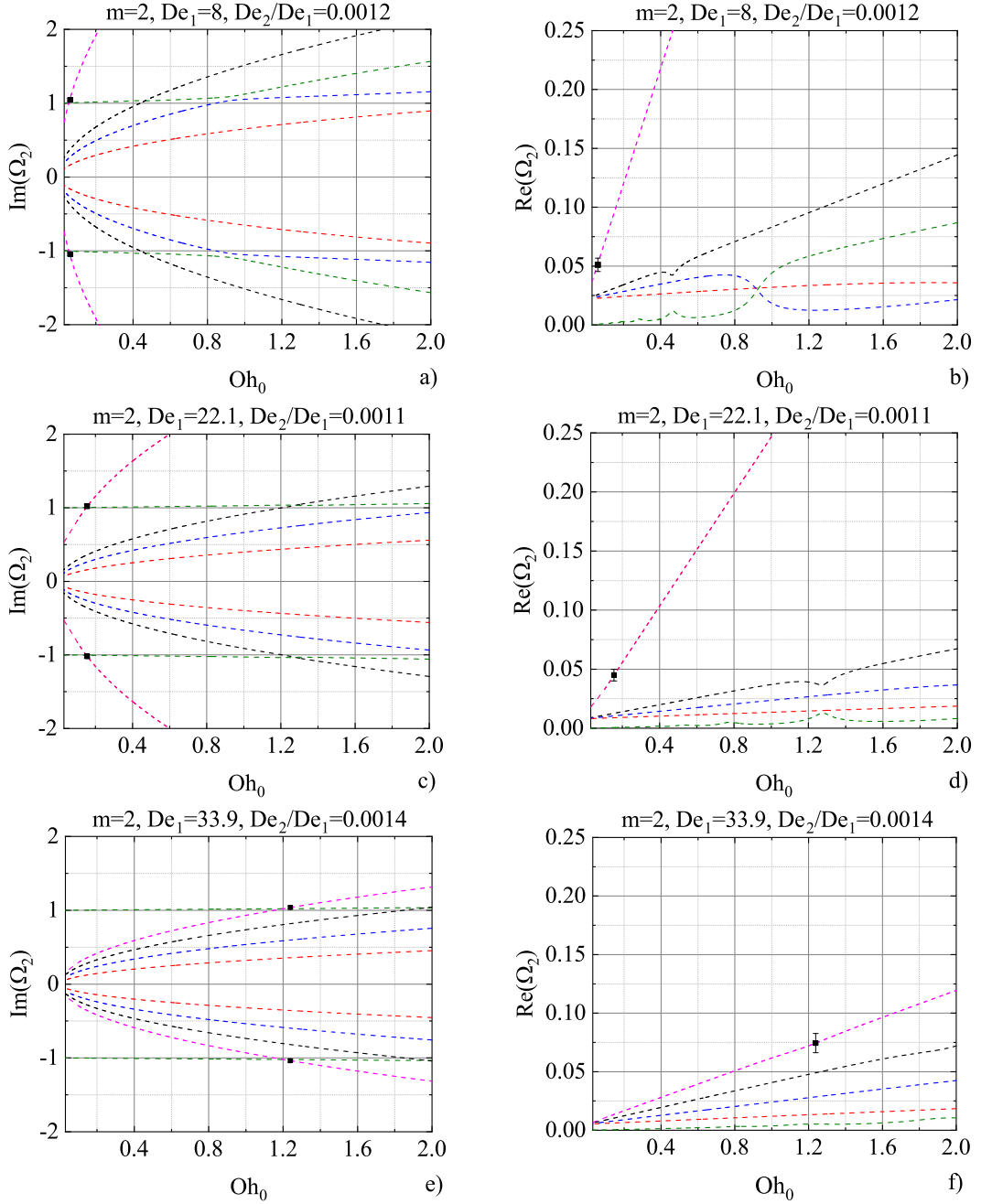


Figure 2.3.: Solutions of the characteristic equation for various polymer solutions with different values of De_1 and De_2/De_1 , corresponding to 2 mm drops of Praestol 2500 solutions in water with mass fractions of 0.1 wt% (a and b), 0.3 wt% (c and d) and 0.5 wt% (e and f), varying the Ohnesorge number. a), c) and e) - imaginary and b), c) and f) - real parts of the nondimensional frequency Ω_2 .

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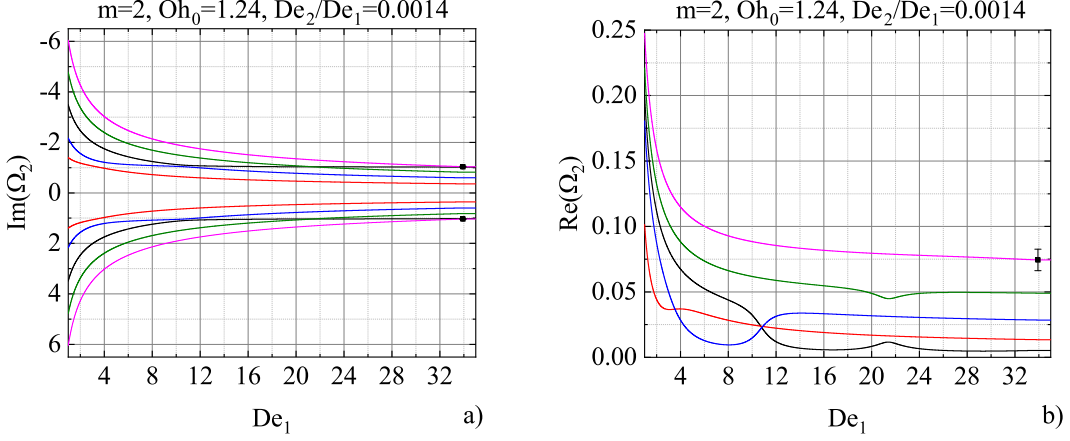


Figure 2.4.: Solutions of the characteristic equation for viscoelastic liquids as functions of De_1 with $Oh_0 = 1.24$ and $De_2/De_1 = 0.0014$, corresponding to a 2 mm drop of a Praestol 2500 solution in water with mass fraction of 0.5 wt%. a) - imaginary and b) - real parts of the nondimensional frequency Ω_2 .

numbers for the SMF of 0.1 wt%, 0.3 wt% and 0.5 wt% are 0.06, 0.16 and 1.24, respectively. The results with the uncertainties for the corresponding Ohnesorge numbers are shown by the black squares and error bars in figure 2.3. The comparison yields perfect agreement between the measured and the calculated (pink dashed line) nondimensional complex conjugate angular frequencies. For drops with different Oh_0 , but the same $m = 2$ and De_2/De_1 , therefore, the values of Ω_2 can be taken from those lines.

In figure 2.4, we show the solutions of the characteristic equation for the range of the Deborah number De_1 between 1 and 35, keeping constant Ohnesorge number $Oh = 1.24$ and Deborah number ratio $De_2/De_1 = 0.0014$, which correspond to a 2 mm 0.5 wt% Praestol 2500 solution drop. The representation shows five sets of complex conjugate pairs of angular frequency, which are presented with different colour lines. The imaginary and real parts of the solution assume high values as the Deborah number De_1 goes to zero. For the comparison with presented analytical solutions, the values of the experimental results and their uncertainties are plotted with the black squares and error bars, respectively. The agreement is again excellent.

Traces of the drop north pole obtained from the experiments and linear

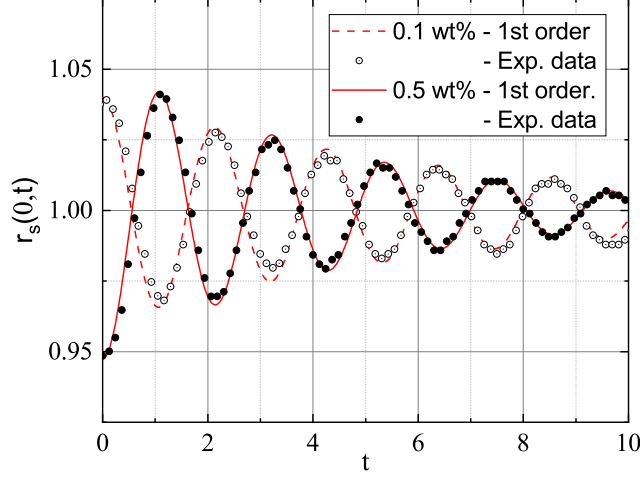


Figure 2.5.: Traces of the drop north pole in time for two different polymer solutions obtained from the linear theory and experimental data.

solutions using the first-order surface deformation (2.61) are shown in figure 2.5. From the complex conjugate angular frequency for the corresponding liquid, we deduce frequency and damping rate of the linear solutions. According to the experimental data at initial time, the surface deformation amplitudes and phase angles are chosen. Time and length scale for the experimental data are nondimensionalized with the capillary time scale and droplet radius, respectively. Since the experimental and theoretical data agree very well for different solute mass fractions, we select only one complex conjugate solution from an infinite number for every SMF. The selection consequently limits the number of initial conditions to two, for the complete first-order solutions.

The existence of pairs of solutions of the characteristic equation is accounted for by the first-order solutions. We therefore re-formulate the first-order deformation of the drop surface as

$$\eta_1(\theta, t) = \left(\hat{\eta}_1^{(p)} e^{-\alpha_m^{(p)} t} + \hat{\eta}_1^{(n)} e^{-\alpha_m^{(n)} t} \right) P_m(\cos \theta). \quad (2.61)$$

The first-order initial conditions (2.61) state that initially the drop surface shape is governed by the function $P_m(\cos \theta)$ and that the surface is at rest.

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These conditions reveal the following expressions for the amplitudes $\hat{\eta}_1^{(p)}$ and $\hat{\eta}_1^{(n)}$:

$$\hat{\eta}_1^{(p)} = -\frac{\alpha_m^{(n)}}{\alpha_m^{(p)} - \alpha_m^{(n)}} \quad \text{and} \quad \hat{\eta}_1^{(n)} = \frac{\alpha_m^{(p)}}{\alpha_m^{(p)} - \alpha_m^{(n)}}. \quad (2.62)$$

We denote the complex angular frequencies with positive and negative imaginary parts by superscripts (p) and (n) , respectively. The first-order stream function, velocity components and pressure are then obtained as

$$\psi(r, \theta, t) = \frac{r^{m+1}}{m(m+1)} \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)} \left(1 + \frac{2(m^2-1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right) e^{-\alpha_m^{(p)} t} \right. \quad (2.63)$$

$$+ \hat{\eta}_1^{(n)} \alpha_m^{(n)} \left(1 + \frac{2(m^2-1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right) e^{-\alpha_m^{(n)} t} \Big] \sin^2 \theta P'_m(\cos \theta) +$$

$$+ \left[\frac{-2(m-1) \hat{\eta}_1^{(p)} \alpha_m^{(p)}}{mq^{(p)} j_m(q^{(p)}) (2q^{(p)} Q_m^{(p)} - q^{(p)2})} q^{(p)} r j_m(q^{(p)} r) e^{-\alpha_m^{(p)} t} \right.$$

$$+ \left. \frac{-2(m-1) \hat{\eta}_1^{(n)} \alpha_m^{(n)}}{mq^{(n)} j_m(q^{(n)}) (2q^{(n)} Q_m^{(n)} - q^{(n)2})} q^{(n)} r j_m(q^{(n)} r) e^{-\alpha_m^{(n)} t} \right]$$

$$\times \sin^2 \theta P'_m(\cos \theta) = \psi_1^{(p)} + \psi_1^{(n)} + \psi_2^{(p)} + \psi_2^{(n)},$$

$$u_{r1}(r, \theta, t) = -\hat{\eta}_1^{(p)} \alpha_m^{(p)} e^{-\alpha_m^{(p)} t} \left[r^{m-1} + \frac{2(m^2-1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right. \quad (2.64)$$

$$\times \left(r^{m-1} - \frac{j_m(q^{(p)} r)}{j_m(q^{(p)})} \frac{1}{r} \right) \Big] P_m(\cos \theta)$$

$$- \hat{\eta}_1^{(n)} \alpha_m^{(n)} e^{-\alpha_m^{(n)} t} \left[r^{m-1} + \frac{2(m^2-1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right.$$

$$\times \left(r^{m-1} - \frac{j_m(q^{(n)} r)}{j_m(q^{(n)})} \frac{1}{r} \right) \Big] P_m(\cos \theta)$$

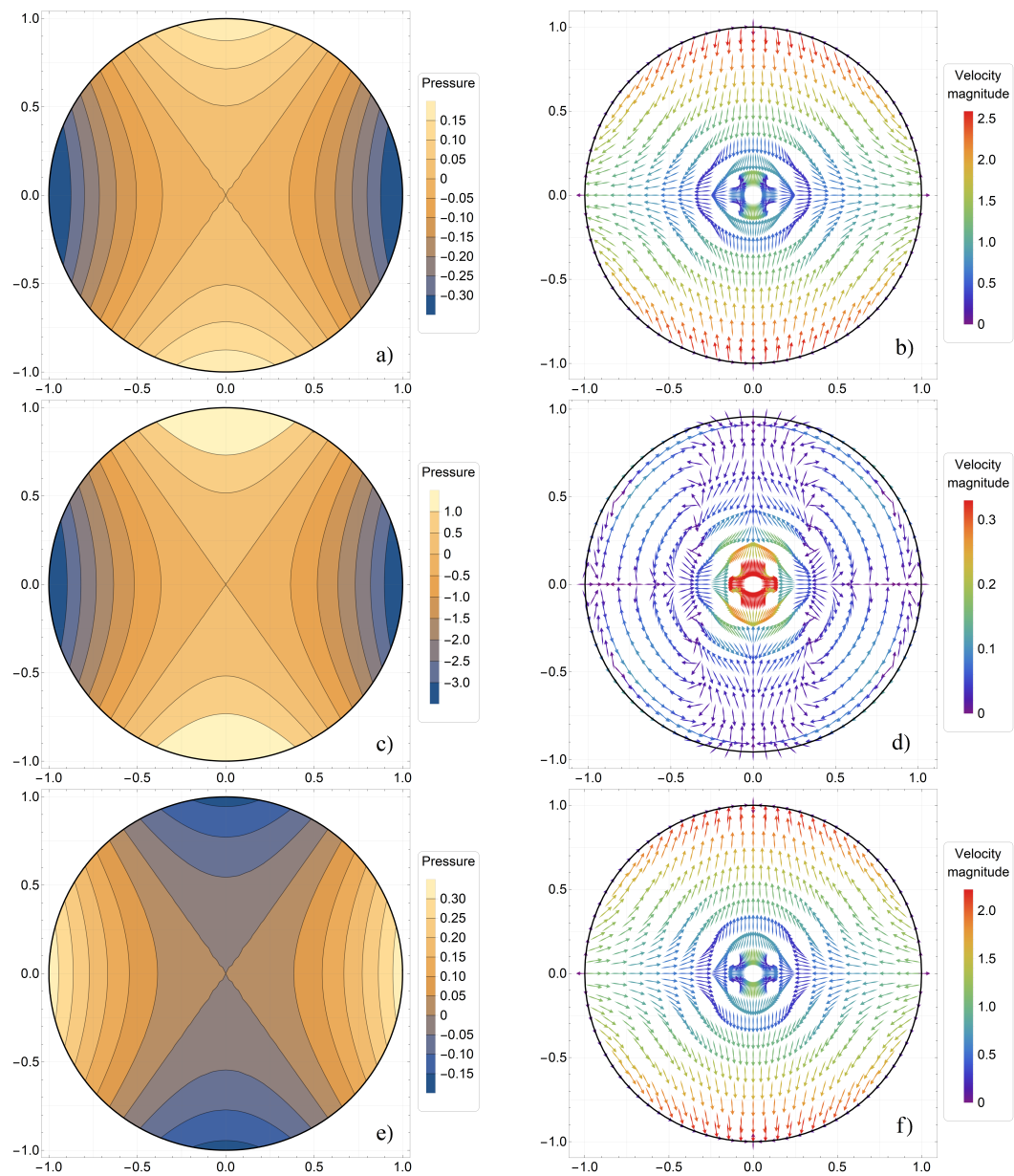
$$\begin{aligned}
 &= - \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)} e^{-\alpha_m^{(p)} t} f_r^{(p)}(r) + \hat{\eta}_1^{(n)} \alpha_m^{(n)} e^{-\alpha_m^{(n)} t} f_r^{(n)}(r) \right] P_m(\cos \theta) , \\
 u_{\theta 1}(r, \theta, t) &= - \frac{\hat{\eta}_1^{(p)} \alpha_m^{(p)}}{m} e^{-\alpha_m^{(p)} t} \left[\left(1 + \frac{2(m^2 - 1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right) r^{m-1} \right. \\
 &\quad \left. - \frac{2(m-1)q^{(p)}}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \left((m+1) \frac{j_m(q^{(p)} r)}{j_m(q^{(p)})} \frac{1}{q^{(p)} r} - \frac{j_{m+1}(q^{(p)} r)}{j_m(q^{(p)})} \right) \right] \\
 &\quad \times P_m^1(\cos \theta) - \frac{\hat{\eta}_1^{(n)} \alpha_m^{(n)}}{m} e^{-\alpha_m^{(n)} t} \left[\left(1 + \frac{2(m^2 - 1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right) r^{m-1} \right. \\
 &\quad \left. - \frac{2(m-1)q^{(n)}}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \left((m+1) \frac{j_m(q^{(n)} r)}{j_m(q^{(n)})} \frac{1}{q^{(n)} r} - \frac{j_{m+1}(q^{(n)} r)}{j_m(q^{(n)})} \right) \right] \\
 &\quad \times P_m^1(\cos \theta) = - \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)} e^{-\alpha_m^{(p)} t} f_\theta^{(p)}(r) + \right. \\
 &\quad \left. + \hat{\eta}_1^{(n)} \alpha_m^{(n)} e^{-\alpha_m^{(n)} t} f_\theta^{(n)}(r) \right] P_m^1(\cos \theta) ,
 \end{aligned} \tag{2.65}$$

$$\begin{aligned}
 p_1(r, \theta, t) &= - \frac{r^m}{m} \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)2} e^{-\alpha_m^{(p)} t} \left(1 + \frac{2(m^2 - 1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right) \right. \\
 &\quad \left. + \hat{\eta}_1^{(n)} \alpha_m^{(n)2} e^{-\alpha_m^{(n)} t} \left(1 + \frac{2(m^2 - 1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right) \right] P_m(\cos \theta) \\
 &= - \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)2} e^{-\alpha_m^{(p)} t} f_p^{(p)}(r) + \hat{\eta}_1^{(n)} \alpha_m^{(n)2} e^{-\alpha_m^{(n)} t} f_p^{(n)}(r) \right] P_m(\cos \theta) ,
 \end{aligned} \tag{2.66}$$

where we have introduced the function $Q_m = j_{m+1}(q)/j_m(q)$, formulated with $q^{(p)}$ and $q^{(n)}$ depending on the two frequencies $\alpha_m^{(p)}$ and $\alpha_m^{(n)}$ in q . The functions ψ_1 , ψ_2 , f_r , f_θ and f_p , defined for each oscillation frequency, are introduced to facilitate the expressions of the flow quantities. With these findings we conclude the complete first-order solutions, which are formally identical to the Newtonian one obtained in Zrnić, Berglez, and Brenn (2022). One of the differences is the Ohnesorge number, which is dependent on the frequency and on two viscoelastic time scales.

In a meridional section, we show the nondimensional pressure field and velocity vectors. The values of the pressure field are presented with a contour

2. Viscoelastic drop shape oscillations



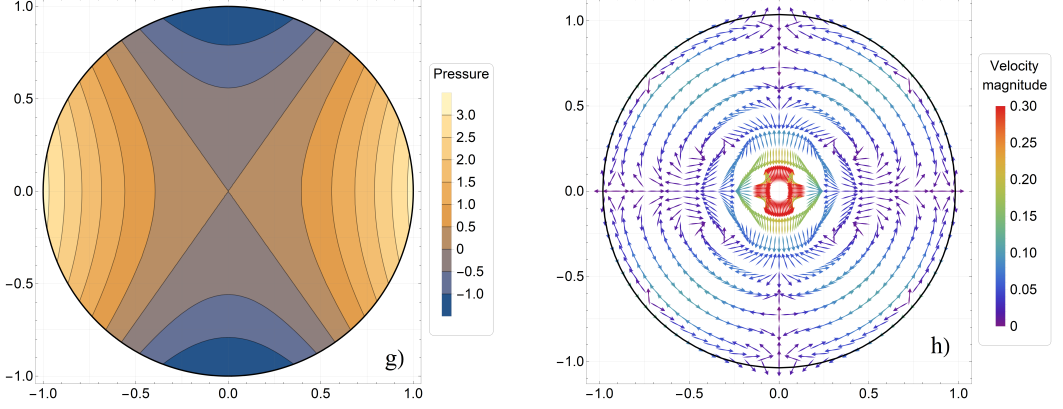


Figure 2.6.: The nondimensional pressure and velocity fields in a meridional section of the drop for $t = 0.548$ (a and b), $t = 1.063$ (c and d) representing the maximum oblate state, $t = 1.611$ (e and f), and $t = 2.126$ (g and h) representing the maximum prolate state.

plot and the vector magnitude with a colour. The nondimensional first-order pressure, radial and polar velocity fields are defined by equations (2.66), (2.64) and (2.65), respectively. We show results for the fundamental initial mode of deformation $m = 2$, initial deformation parameter $\eta_0 = 0.05$ and solute mass fraction of 0.1 wt%, which with table 2.1 defines the Ohnesorge and two Deborah numbers that equal 0.061, 8.03 and 0.0096, respectively. From the characteristic equation in (2.58), the complex conjugate angular frequency for the corresponding case is shown in figure 2.3 (a and b) with the black filled squares and equals $0.051 \pm 1.045i$. The pressure and velocity fields are represented for four time instances: for the maximum oblate and prolate states, and for the spherical states before and between them. In figure 2.6 (a and b), the fields are shown for the spherical state, which, at the time $t = 0.548$, is before the maximum oblate form. At that time instance, the drop surface is moving as it is compressed from the north and south poles. In the region close to the north and south poles, the velocity vectors are pointing to the drop centre, and the velocity magnitudes and the pressure values are the greatest. The opposite state, the spherical state before the maximum prolate form, is at $t = 1.611$ and is shown in figure 2.6 (e and f). The vectors are pointing in the opposite direction in comparison with figure

2.6 (a and b). Figures e) and f) show that the drop surface is moving as it is compressed in the equator region. The maximum oblate and prolate states at times $t = 1.063$ and $t = 2.126$, respectively, show small values of the pressure and large velocity magnitudes near the drop centre. In comparison with the spherical states, the pressure contour presents substantially larger pressure values near the equator, north and south poles. The pressure distribution is similar for all time instances.

2.4. Conclusions

A weakly nonlinear analysis is carried out for axisymmetric drop shape oscillations in a vacuum, and the governing equations with boundary and initial conditions to the third order of approximation are derived. The Oldroyd-B model is used for describing the rheological behaviour of the viscoelastic fluid upon deformation. We derived the first-order solutions, known as linear solutions, with the characteristic equation for the drop shape oscillations. The characteristic equation yields an infinite number of solutions, which are known as complex angular oscillation frequencies. The equation is formally identical to the Newtonian form, except that the Ohnesorge number depends on the oscillation frequency and on the two viscoelastic time scales, which are the stress relaxation and deformation retardation times. These time scales are defined with two Deborah numbers. From the infinite number of complex angular frequencies for the range of Ohnesorge numbers and fixed Deborah numbers, one must select an appropriate one.

The appropriate solutions are selected according to experimental data to determine time dependencies in the higher-order solutions and the number of initial conditions in the analysis. The solutions of the characteristic equation agree well with the experimental data from damped drop shape oscillations of aqueous Praestol 2500 solution drops in an acoustic levitator. We select only one complex conjugate solution, and doing that, define the number of initial conditions in the higher orders of approximation. Steered by our work Zrnić, Berglez, and Brenn (2022) and the fact that the characteristic equation for the range of Ohnesorge numbers and fixed Deborah numbers gives a pair of complex conjugate angular frequencies, one can predict that the structure

of the solutions in the higher-orders will be similar to the ones from the viscous case, which is presented in the next chapter. The first difference from the viscous case is that the viscosity is dependent on the frequency, which one concludes from the definition of the Ohnesorge number, and the second is the presence of two time scales that are contained in the solutions via argument q .

At the end of this section, the nondimensional pressure field and velocity vectors are shown in a meridional section of the drop for the fundamental initial mode of deformation $m = 2$, initial deformation parameter $\eta_0 = 0.05$ and solute mass fraction of 0.1 wt%. The pressure and velocity fields are represented for four time instances at the maximum oblate and prolate states, and in the spherical states before and between the maximum deformations. The pressure distribution is similar for all time instances, but values vary according to the motion.

3. Newtonian drop shape oscillations

The current chapter encompasses the study of Newtonian drop shape oscillations. The first section demonstrates the governing equations derived by the weakly nonlinear method for the respective order of approximation, and the second section gives the solutions to the equations. In the third section we arrive at a discussion of the results, and it is divided into four subsections. Before showing the result highlights, the first subsection includes the validation of the method applied. The second subsection presents the deformed surface shapes for different initial modes of deformation. The result highlights are discussed in the third subsection, which investigates the time scales of the drop oscillations and reveals unexpected oscillatory phenomena at supercritical Ohnesorge numbers and higher initial deformation modes. The next section shows the experimental method applied for measuring time scales of Newtonian drops and verifies the measurements with the related literature. The chapter ends with the conclusions. Many parts of the work in this chapter originate from the work by [D. Zrnić, P. Berglez, and G. Brenn, "Weakly nonlinear shape oscillations of a Newtonian drop", *Physics of Fluids* 34, 043103 (2022)], with permission from AIP Publishing.

3.1. Governing equations of the oscillating drop

The method for deriving the following equations up to the third-order of approximation corresponds to that from the previous chapter. The current chapter shows the viscous drop shape oscillations as sketched in figure 2.1, and the weakly nonlinear theory approach applied here makes similar assumptions, which will be pointed out below. The liquid material behaviour

3. Newtonian drop shape oscillations

is characterized as Newtonian. We define the stress tensor $\boldsymbol{\tau}$ in the spherical coordinate system as in appendix A of the work by Brenn (2017), the general description of the momentum equations (2.2)-(2.3) can be reduced to

$$\begin{aligned} \frac{\partial u_r}{\partial t} + u_r \frac{\partial u_r}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_r}{\partial \theta} - \frac{u_\theta^2}{r} = -\frac{\partial p}{\partial r} \\ + Oh \left[\frac{1}{r^2} \frac{\partial^2}{\partial r^2} (r^2 u_r) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\partial u_r}{\partial \theta} \sin \theta \right) \right], \end{aligned} \quad (3.1)$$

$$\begin{aligned} \frac{\partial u_\theta}{\partial t} + u_r \frac{\partial u_\theta}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_\theta}{\partial \theta} + \frac{u_r u_\theta}{r} = -\frac{1}{r} \frac{\partial p}{\partial \theta} + \\ + Oh \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_\theta}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} (u_\theta \sin \theta) \right) + \frac{2}{r^2} \frac{\partial u_r}{\partial \theta} \right], \end{aligned} \quad (3.2)$$

The set of these equations, together with the continuity equation, is subjected to the boundary and initial conditions. Both initial and boundary conditions are derived in the same manner as in chapter 2. The kinematic, zero-shear stress and zero-normal stress boundary conditions are given in equations (2.7), (2.8) and (2.10), respectively.

The two velocity components, the pressure in the flow field and the deformed interface shape, analysed in a weakly nonlinear form, are expanded in power series for the small deformation parameter η_0 . Since the equations (2.1), (3.1) and (3.2) differ from the general ones in the previous chapter only by the diffusion term, one formulates the expansions of the flow field properties, for u_r , p and η , in the same manner as in chapter 2. They are given in equation (2.12). These series expansions converge only if the deformation parameter η_0 is small.

Unlike the linear analysis, the boundary conditions are satisfied on the deformed drop surface, and the values of the deformed shape are represented by Taylor expansions. Equation (2.19) presents the expansion for u_r and p . Further, the flow properties and their derivatives given in (2.12) through (2.19) are substituted into the equations of change (2.1), (3.1) and (3.2) and into the boundary conditions (2.7), (2.8) and (2.10). In this way, sets of first-, second- and third-order equations of motion with the boundary conditions are obtained, consisting of all the terms with the deformation parameter η_0

to the first, second and third powers, respectively. All the benefits of such analysis are explained in chapter 2.

The initial conditions are given with the initial non-dimensional drop shape by equation (2.20), while the centre of mass of the drop is given in equation (2.21). The reasons for constructing both conditions have already been explained and justified in the previous chapter. Further details on the development of the centre of mass are delivered in the course of the solution development.

3.1.1. First-order equations

The first-order equations with their boundary and initial conditions are derived introducing the series expansions into the respective equations. All the terms with the parameter η_0 to the first power are collected. The first-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_{r1}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 1} \sin \theta) = 0, \quad (3.3)$$

$$\frac{\partial u_{r1}}{\partial t} = -\frac{\partial p_1}{\partial r} + Oh \left[\frac{1}{r^2} \frac{\partial^2}{\partial r^2} (r^2 u_{r1}) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\partial u_{r1}}{\partial \theta} \sin \theta \right) \right], \quad (3.4)$$

$$\begin{aligned} \frac{\partial u_{\theta 1}}{\partial t} = & -\frac{1}{r} \frac{\partial p_1}{\partial \theta} \\ & + Oh \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_{\theta 1}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 1} \sin \theta) \right) + \frac{2}{r^2} \frac{\partial u_{r1}}{\partial \theta} \right]. \end{aligned} \quad (3.5)$$

3.1.2. First-order boundary and initial conditions

Three boundary conditions are set: the rate of radial drop surface displacement, which equals the radial surface velocity, the shear and the normal stresses at the drop surface, which are zero because of the ambient vacuum. Deriving these three boundary conditions of first order, to be satisfied at

3. Newtonian drop shape oscillations

$r = 1$, one obtains

$$u_{r1} = \frac{\partial \eta_1}{\partial t} , \quad (3.6)$$

$$r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} = 0 , \quad (3.7)$$

$$-p_1 + 2Oh \frac{\partial u_{r1}}{\partial r} - \left(2\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right) = 0 , \quad (3.8)$$

respectively. Furthermore, the initial conditions of first order are

$$\eta_1(\theta, 0) = P_m(\cos \theta) , \quad \frac{\partial \eta_1}{\partial t}(\theta, 0) = 0 . \quad (3.9)$$

The first initial condition determines the initial shape of the deformed drop which is given by a Legendre polynomial of degree m , and the second one accounts for the initial quiescent state of the drop surface.

3.1.3. Second-order equations

The second-order equations with their boundary and initial conditions are extracted collecting all the terms from the expansion with the parameter η_0 to the second power. The second-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 u_{r2} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 2} \sin \theta) = 0 , \quad (3.10)$$

$$\begin{aligned} \frac{\partial u_{r2}}{\partial t} - Oh \left[\frac{1}{r^2} \frac{\partial^2}{\partial r^2} \left(r^2 u_{r2} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\partial u_{r2}}{\partial \theta} \sin \theta \right) \right] + \frac{\partial p_2}{\partial r} = \frac{u_{\theta 1}^2}{r} \\ - u_{r1} \frac{\partial u_{r1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{r1}}{\partial \theta} , \end{aligned} \quad (3.11)$$

$$\begin{aligned} \frac{\partial u_{\theta 2}}{\partial t} - Oh \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_{\theta 2}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 2} \sin \theta) \right) \right. \\ \left. + \frac{2}{r^2} \frac{\partial u_{r2}}{\partial \theta} \right] + \frac{1}{r} \frac{\partial p_2}{\partial \theta} = -u_{r1} \frac{\partial u_{\theta 1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} - \frac{u_{r1} u_{\theta 1}}{r} , \end{aligned} \quad (3.12)$$

3.1.4. Second-order boundary and initial conditions

The second-order kinematic boundary condition and the boundary conditions stating zero shear and normal stresses, to be satisfied at $r = 1$, are

$$u_{r2} - \frac{\partial \eta_2}{\partial t} = \frac{u_{\theta 1}}{r} \frac{\partial \eta_1}{\partial \theta} - \eta_1 \frac{\partial u_{r1}}{\partial r}, \quad (3.13)$$

$$\begin{aligned} r \frac{\partial}{\partial r} \left(\frac{u_{\theta 2}}{r} \right) + \frac{1}{r} \frac{\partial u_{r2}}{\partial \theta} = & -\eta_1 \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} \right) \\ & - 2 \left(r \frac{\partial}{\partial r} \left(\frac{u_{r1}}{r} \right) - \frac{1}{r} \frac{\partial u_{\theta 1}}{\partial \theta} \right) \frac{1}{r} \frac{\partial \eta_1}{\partial \theta}, \end{aligned} \quad (3.14)$$

$$\begin{aligned} -p_2 + 2Oh \frac{\partial u_{r2}}{\partial r} - \left(2\eta_2 + \frac{\partial \eta_2}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_2}{\partial \theta^2} \right) = & \eta_1 \frac{\partial p_1}{\partial r} \\ & - 2Oh \left[\eta_1 \frac{\partial^2 u_{r1}}{\partial r^2} - \frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} \right) \right] \\ & - \left(2\eta_1^2 + 2\eta_1 \frac{\partial \eta_1}{\partial \theta} \cot \theta + 2\eta_1 \frac{\partial^2 \eta_1}{\partial \theta^2} \right), \end{aligned} \quad (3.15)$$

respectively. Furthermore, the initial conditions of second order are

$$\eta_2(\theta, 0) = -\frac{1}{2m+1}, \quad \frac{\partial \eta_2}{\partial t}(\theta, 0) = 0. \quad (3.16)$$

The first initial condition ensures drop volume conservation, and the second one ensures that the drop surface is initially in a quiescent state.

In the later development, it can be seen that solving the sets of first- and second-order equations, together with their boundary and initial conditions, reveals two known nonlinear effects. The first is a frequency change, and the second is an excess time the drop spends in an elongated state, both as functions of the initial deformation amplitude. The analysis must be extended to the third order to represent these effects with good agreement with other literature.

3.1.5. Third-order equations

The third-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_{r3}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 3} \sin \theta) = 0, \quad (3.17)$$

$$\begin{aligned} \frac{\partial u_{r3}}{\partial t} - Oh \left[\frac{1}{r^2} \frac{\partial^2}{\partial r^2} (r^2 u_{r3}) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\partial u_{r3}}{\partial \theta} \sin \theta \right) \right] + \frac{\partial p_3}{\partial r} = 2 \frac{u_{\theta 1} u_{\theta 2}}{r} \\ - u_{r1} \frac{\partial u_{r2}}{\partial r} - u_{r2} \frac{\partial u_{r1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{r2}}{\partial \theta} - \frac{u_{\theta 2}}{r} \frac{\partial u_{r1}}{\partial \theta}, \end{aligned} \quad (3.18)$$

$$\begin{aligned} \frac{\partial u_{\theta 3}}{\partial t} - Oh \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_{\theta 3}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 3} \sin \theta) \right) \right] \\ + \frac{2}{r^2} \frac{\partial u_{r3}}{\partial \theta} \Bigg] + \frac{1}{r} \frac{\partial p_3}{\partial \theta} = -u_{r1} \frac{\partial u_{\theta 2}}{\partial r} - u_{r2} \frac{\partial u_{\theta 1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 2}}{\partial \theta} - \frac{u_{\theta 2}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} \\ - \frac{u_{r1} u_{\theta 2}}{r} - \frac{u_{r2} u_{\theta 1}}{r}. \end{aligned} \quad (3.19)$$

3.1.6. Third-order boundary and initial conditions

The third-order kinematic boundary condition, as well as the boundary conditions stating zero shear and normal stress, to be satisfied at $r = 1$, are

$$\begin{aligned} u_{r3} - \frac{\partial \eta_3}{\partial t} = \frac{u_{\theta 1}}{r} \frac{\partial \eta_2}{\partial \theta} + \frac{u_{\theta 2}}{r} \frac{\partial \eta_1}{\partial \theta} + \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) \frac{\partial \eta_1}{\partial \theta} \eta_1 \\ - \frac{\partial u_{r2}}{\partial r} \eta_1 - \frac{\partial u_{r1}}{\partial r} \eta_2 - \frac{1}{2} \frac{\partial^2 u_{r1}}{\partial r^2} \eta_1^2, \end{aligned} \quad (3.20)$$

$$\begin{aligned} r \frac{\partial}{\partial r} \left(\frac{u_{\theta 3}}{r} \right) + \frac{1}{r} \frac{\partial u_{r3}}{\partial \theta} = \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} \right) \left(\frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \right)^2 \\ - 2 \left\{ \left(r \frac{\partial}{\partial r} \left(\frac{u_{r2}}{r} \right) - \frac{1}{r} \frac{\partial u_{\theta 2}}{\partial \theta} \right) \frac{1}{r} \frac{\partial \eta_1}{\partial \theta} + \left(r \frac{\partial}{\partial r} \left(\frac{u_{r1}}{r} \right) - \frac{1}{r} \frac{\partial u_{\theta 1}}{\partial \theta} \right) \frac{1}{r} \frac{\partial \eta_2}{\partial \theta} \right. \\ \left. + r \frac{\partial}{\partial r} \left[\frac{1}{r} \left(r \frac{\partial}{\partial r} \left(\frac{u_{r1}}{r} \right) - \frac{1}{r} \frac{\partial u_{\theta 1}}{\partial \theta} \right) \right] \frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \eta_1 \right\} \\ - \frac{1}{2} \frac{\partial^2}{\partial r^2} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} \right) \eta_1^2 - \eta_1 \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 2}}{r} \right) + \frac{1}{r} \frac{\partial u_{r2}}{\partial \theta} \right) \\ - \eta_2 \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} \right), \end{aligned} \quad (3.21)$$

$$\begin{aligned}
 & -p_3 + 2Oh \frac{\partial u_{r3}}{\partial r} - \left(2\eta_3 + \frac{\partial \eta_3}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_3}{\partial \theta^2} \right) = \frac{\partial p_1}{\partial r} \eta_2 + \frac{\partial p_2}{\partial r} \eta_1 \\
 & - 2Oh \left\{ \frac{\partial^2 u_{r1}}{\partial r^2} \eta_2 + \frac{\partial^2 u_{r2}}{\partial r^2} \eta_1 - \left(\frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \right)^2 \left(r \frac{\partial}{\partial r} \left(\frac{u_{r1}}{r} \right) - \frac{1}{r} \frac{\partial u_{\theta 1}}{\partial \theta} \right) \right. \\
 & - r \frac{\partial}{\partial r} \left(\frac{1}{r} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} \right) \right) \frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \eta_1 + \frac{1}{2} \frac{\partial^3 u_{r1}}{\partial r^3} \eta_1^2 \\
 & \left. - \frac{1}{r} \frac{\partial \eta_2}{\partial \theta} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) + \frac{1}{r} \frac{\partial u_{r1}}{\partial \theta} \right) - \frac{1}{r} \frac{\partial \eta_1}{\partial \theta} \left(r \frac{\partial}{\partial r} \left(\frac{u_{\theta 2}}{r} \right) + \frac{1}{r} \frac{\partial u_{r2}}{\partial \theta} \right) \right\} \\
 & - \eta_1^3 - 2\eta_1 \left(\eta_2 + \frac{\partial \eta_2}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_2}{\partial \theta^2} \right) \\
 & - 2\eta_2 \left(\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right) - \frac{1}{2} \left(\frac{\partial \eta_1}{\partial \theta} \right)^2 \left(\frac{\partial \eta_1}{\partial \theta} \cot \theta + 3 \frac{\partial^2 \eta_1}{\partial \theta^2} \right) \\
 & + 3\eta_1^2 \left(\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right) + \frac{1}{2} \frac{\partial^2 p_1}{\partial r^2} \eta_1^2 , \tag{3.22}
 \end{aligned}$$

respectively. Furthermore, the initial conditions of third order are

$$\eta_3(\theta, 0) = -\frac{1}{3} \begin{pmatrix} m & m & m \\ 0 & 0 & 0 \end{pmatrix}^2, \quad \frac{\partial \eta_3}{\partial t}(\theta, 0) = 0, \tag{3.23}$$

where the first initial condition contains a 3- j symbol known as the Wigner 3- j symbol from Wigner (1993). Solutions of the sets of equations up to third order, together with their initial and boundary conditions, will reveal the weakly nonlinear drop oscillation dynamics.

3.2. Solutions of the governing equations

The current section describes the method of finding the solutions to the above equations for the corresponding order of approximation. For the sake of clarity, parts of the derivations and coefficient calculations are shown in appendix A. References to the appendix are given in the derivations in the text below. The first section shows the first-order solutions, which are well-known solutions of the linear system. They are obtained using the Stokesian stream

function, and the oscillation frequency is found by solving the characteristic equation emerging from the zero normal stress boundary condition. The second- and third-order solutions, shown in the second and third subsection, respectively, are obtained from the pressure first, formulating the Poisson equation for a modified pressure, and the pressure solutions are divided on two counterparts which satisfy the homogeneous and inhomogeneous equations. The velocity fields are solved in a straightforward manner using the governing equations and the known pressure field. The surface deformations and amplitudes are determined using boundary conditions. The method of finding the solutions described below are found in the work by Zrnić, Berglez, and Brenn (2022).

3.2.1. First-order solutions

The well-known linear solutions satisfy the first-order equations. These solutions, together with the characteristic equation of the viscous drop in a vacuum, are due to Lamb (1881, 1932) and Chandrasekhar (1959). For determining the first-order velocity and pressure fields, the method of the Stokesian stream function is applied because only two-dimensional flow fields are investigated. The relations to the two velocity components u_{r1} and $u_{\theta 1}$ as in Bird, Stewart, and Lightfoot (1962) are defined through the stream function $\psi(r, \theta, t)$

$$u_{r1} = -\frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta} \quad \text{and} \quad u_{\theta 1} = \frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r}. \quad (3.24)$$

These first-order velocity components satisfy the continuity equation identically.

The Legendre polynomial of the initial deformation governs the first-order drop surface deformation, and therefore the shape is sought in the form

$$\eta_1(\theta, t) = \hat{\eta}_1 P_m(\cos \theta) e^{-\alpha_m t}, \quad (3.25)$$

where the first-order initial surface amplitude is $\hat{\eta}_1$ and the first-order complex angular frequency is α_m for the deformation mode m .

The fourth-order partial differential equation is obtained by taking the curl of the first-order momentum equation (3.4) and (3.5) in a vectorial form and

reads

$$\left(\frac{1}{Oh} \frac{\partial}{\partial t} - E_s^2 \right) (E_s^2 \psi) = 0, \quad (3.26)$$

with the operator E_s from Bird, Stewart, and Lightfoot (ibid.) written as

$$E_s^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin \theta}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \right). \quad (3.27)$$

The resulting stream function ψ consists of two contributions $\psi_1 + \psi_2$ (Tomotika, 1935) represented by

$$\psi_1 = C_{1m} r^{m+1} \sin^2 \theta P'_m(\cos \theta) e^{-\alpha_m t}, \quad (3.28)$$

$$\psi_2 = C_{2m} q r j_m(qr) \sin^2 \theta P'_m(\cos \theta) e^{-\alpha_m t}, \quad (3.29)$$

where the first-order derivative of the Legendre polynomial P_m with respect to its argument is $P'_m(\cos \theta)$ and a spherical Bessel function of the first kind and order m is j_m . In the latter, the argument contains q that equals $\sqrt{\alpha_m / Oh}$. The radial and angular components of the first-order velocity vector follow as derivatives of the stream function

$$u_{r1} = - \left[C_{1m} r^{m-1} + C_{2m} q^2 \frac{j_m(qr)}{qr} \right] m(m+1) P_m(\cos \theta) e^{-\alpha_m t}, \quad (3.30)$$

and

$$\begin{aligned} u_{\theta 1} = & - \left[C_{1m} (m+1) r^{m-1} \right. \\ & \left. + C_{2m} q^2 \left((m+1) \frac{j_m(qr)}{qr} - j_{m+1}(qr) \right) \right] P_m^1(\cos \theta) e^{-\alpha_m t}, \end{aligned} \quad (3.31)$$

respectively. The product $-\sin \theta P'_m(\cos \theta)$ is represented as the Legendre function $P_m^1(\cos \theta)$. Further, the first-order kinematic and zero shear stress boundary conditions determine the two integration constants C_{1m} and C_{2m} , which read

$$C_{1m} = \frac{\hat{\eta}_1 \alpha_m}{m(m+1)} \left[1 + \frac{2(m^2 - 1)}{2q j_{m+1}(q) / j_m(q) - q^2} \right] \quad (3.32)$$

$$\text{and} \quad C_{2m} = - \frac{2(m-1) \hat{\eta}_1 \alpha_m}{mq [2q j_{m+1}(q) - q^2 j_m(q)]}. \quad (3.33)$$

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The first-order pressure field is obtained from the integration of one component of the momentum equation as

$$p_1 = -C_{1m}(m+1)\alpha_m r^m P_m(\cos \theta) e^{-\alpha_m t}. \quad (3.34)$$

Once the velocity and pressure fields are known, the zero normal stress boundary condition of first order (3.8) yields the characteristic equation of the drop, which reads

$$\frac{\alpha_{m,0}^2}{\alpha_m^2} = \frac{2(m^2 - 1)}{q^2 - 2qj_{m+1}/j_m} - 1 + \frac{2m(m-1)}{q^2} \left[1 + \frac{2(m+1)j_{m+1}/j_m}{2j_{m+1}/j_m - q} \right]. \quad (3.35)$$

The derivations arrive at the same equation as Lamb (1881) and Chandrasekhar (1959). It determines the complex angular frequency α_m , knowing that $\alpha_{m,0} = [m(m-1)(m+2)]^{1/2}$. The spherical Bessel functions are taken at the value q of their arguments. The expression for the square q^2 of the argument of the spherical Bessel functions is the same as in equation (2.59), but with $De_1 = 0$ and $De_2 = 0$.

For the whole range of Ohnesorge numbers studied here, the characteristic equation of the Newtonian drop exhibits pairs of solutions. The solutions are depicted in figure 3.1 for the first three modes $m = 2, 3$ and 4 . Starting from the initial deformation, drops perform damped shape oscillations with Ohnesorge numbers below the critical value (of 0.7665 for mode $m = 2$, Prosperetti (1980a)). In this range, the characteristic equation of the drop produces complex conjugate solutions. For Ohnesorge numbers greater than the critical value, the drop aperiodically returns to its equilibrium shape. In this case, the oscillation frequency is zero, while the rate of decay is described by two real solutions, of which the stronger one dies out more quickly and therefore is not visible in the experiments. Prosperetti (ibid.) found a range of Ohnesorge numbers where a drop starts from an aperiodic motion but eventually develops damped oscillations. Aperiodic motion and damped oscillation correspond to surface motion obtained by the solution types of the dispersion relation for the jet (Renoult et al., 2018), where, however, the aperiodic behaviour makes initial deformations grow, while the periodic behaviour is also a damped shape oscillation for the jet. The parameter varied for the jet, however, is the wavenumber of the surface deformation,

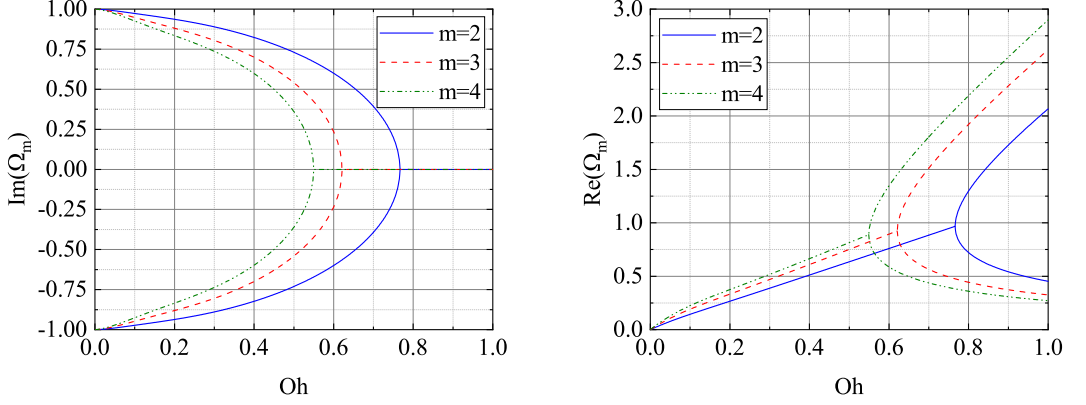


Figure 3.1.: Solutions of the characteristic equation for different Ohnesorge numbers. The left diagram represents the imaginary and the right the real part of the nondimensional frequency Ω_m , defined as $\alpha_m/\alpha_{m,0}$. The critical Ohnesorge number is value 0.7665 for mode $m = 2$ (Prosperetti, 1980a). The figures originate from Zrnić, Berglez, and Brenn (2022).

i.e. a geometric parameter of the jet shape, not a material property like the Ohnesorge number.

Since the characteristic equation yields a pair of solutions, the first-order deformation of the drop surface demands re-formulation as

$$\eta_1(\theta, t) = \left(\hat{\eta}_1^{(p)} e^{-\alpha_m^{(p)} t} + \hat{\eta}_1^{(n)} e^{-\alpha_m^{(n)} t} \right) P_m(\cos \theta). \quad (3.36)$$

The drop surface shape is given by the first-order initial conditions (3.9). The conditions describe the shape by a function $P_m(\cos \theta)$, where the state of the surface is quiescent. These two conditions bring about the amplitudes $\hat{\eta}_1^{(p)}$ and $\hat{\eta}_1^{(n)}$ that read

$$\hat{\eta}_1^{(p)} = -\frac{\alpha_m^{(n)}}{\alpha_m^{(p)} - \alpha_m^{(n)}} \quad \text{and} \quad \hat{\eta}_1^{(n)} = \frac{\alpha_m^{(p)}}{\alpha_m^{(p)} - \alpha_m^{(n)}}. \quad (3.37)$$

The complex angular frequencies with positive and negative imaginary parts are denoted by superscripts (p) and (n) , respectively. Therefore the first-order

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stream function and radial velocity component are written as

$$\begin{aligned}
 \psi(r, \theta, t) = & \frac{r^{m+1}}{m(m+1)} \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)} \left(1 + \frac{2(m^2-1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right) e^{-\alpha_m^{(p)} t} \right. \\
 & + \hat{\eta}_1^{(n)} \alpha_m^{(n)} \left(1 + \frac{2(m^2-1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right) e^{-\alpha_m^{(n)} t} \left. \right] \sin^2 \theta P'_m(\cos \theta) + \\
 & + \left[\frac{-2(m-1) \hat{\eta}_1^{(p)} \alpha_m^{(p)}}{mq^{(p)} j_m(q^{(p)}) (2q^{(p)} Q_m^{(p)} - q^{(p)2})} q^{(p)} r j_m(q^{(p)} r) e^{-\alpha_m^{(p)} t} \right. \\
 & + \left. \frac{-2(m-1) \hat{\eta}_1^{(n)} \alpha_m^{(n)}}{mq^{(n)} j_m(q^{(n)}) (2q^{(n)} Q_m^{(n)} - q^{(n)2})} q^{(n)} r j_m(q^{(n)} r) e^{-\alpha_m^{(n)} t} \right] \\
 & \times \sin^2 \theta P'_m(\cos \theta) = \psi_1^{(p)} + \psi_1^{(n)} + \psi_2^{(p)} + \psi_2^{(n)},
 \end{aligned} \tag{3.38}$$

$$\begin{aligned}
 u_{r1}(r, \theta, t) = & -\hat{\eta}_1^{(p)} \alpha_m^{(p)} e^{-\alpha_m^{(p)} t} \left[r^{m-1} + \frac{2(m^2-1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right. \\
 & \times \left(r^{m-1} - \frac{j_m(q^{(p)} r)}{j_m(q^{(p)})} \frac{1}{r} \right) \left. \right] P_m(\cos \theta) \\
 & - \hat{\eta}_1^{(n)} \alpha_m^{(n)} e^{-\alpha_m^{(n)} t} \left[r^{m-1} + \frac{2(m^2-1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right. \\
 & \times \left(r^{m-1} - \frac{j_m(q^{(n)} r)}{j_m(q^{(n)})} \frac{1}{r} \right) \left. \right] P_m(\cos \theta) \\
 = & - \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)} e^{-\alpha_m^{(p)} t} f_r^{(p)}(r) + \hat{\eta}_1^{(n)} \alpha_m^{(n)} e^{-\alpha_m^{(n)} t} f_r^{(n)}(r) \right] P_m(\cos \theta).
 \end{aligned} \tag{3.39}$$

For the sake of readability of the thesis, the first-order polar velocity component $u_{\theta 1}$ and pressure p_1 are given in appendix A under section A. Introduced in the first-order solutions, the function $Q_m = j_{m+1}(q)/j_m(q)$ contains spherical Bessel functions with the arguments $q^{(p)}$ and $q^{(n)}$ that depend on the two frequencies $\alpha_m^{(p)}$ and $\alpha_m^{(n)}$, respectively. To facilitate the expressions of the flow quantities for use in the following sections, the functions ψ_1 , ψ_2 , f_r , f_θ and f_p are constructed for each oscillation frequency.

3.2.2. Second-order solutions

The second-order solutions are developed from pressure. The general solution for the second-order pressure is formulated as a sum

$$p_2(r, \theta, t) = p_{21}(r, \theta, t) + p_{22}(r, \theta, t) \quad (3.40)$$

of two contributions, where the inhomogeneous system and the homogeneous system of second-order equations are solved by the pressures with the subscript "21" and "22", respectively.

The solutions are sought for the subscript "21" first. The derivations start with forming the divergence of the vectorial momentum equation, where the second-order velocities are eliminated from the second-order equations of motion (3.11)-(3.12) with the use of continuity equation (3.10). Ultimately, the differential equation for the second-order pressure p_{21} reads

$$\begin{aligned} \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial p_{21}}{\partial r} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\sin \theta}{r} \frac{\partial p_{21}}{\partial \theta} \right) = \\ - \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \left(u_{r1} \frac{\partial u_{r1}}{\partial r} + \frac{u_{\theta 1}}{r} \frac{\partial u_{r1}}{\partial \theta} - \frac{u_{\theta 1}^2}{r} \right) \right] \\ - \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left[\left(u_{r1} \frac{\partial u_{\theta 1}}{\partial r} + \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} + \frac{u_{r1} u_{\theta 1}}{r} \right) \sin \theta \right], \end{aligned} \quad (3.41)$$

which can be re-written in a compact form using the Lamé identity

$$\Delta p_{21} = -\nabla \cdot \left[\vec{\nabla} \vec{v}_1^2 / 2 - \vec{v}_1 \times \left(\vec{\nabla} \times \vec{v}_1 \right) \right]. \quad (3.42)$$

The present flow field allows re-writing equation (3.42) as

$$\Delta \left[p_{21} + \vec{v}_1^2 / 2 \right] = \frac{1}{Oh} \nabla \cdot \left[\frac{1}{r^2 \sin^2 \theta} \frac{\partial \psi_2}{\partial t} \vec{\nabla} \psi \right]. \quad (3.43)$$

It shows a Poisson equation for the modified pressure $\mathcal{P}_{21} = p_{21} + \vec{v}_1^2 / 2$ (Renoult et al., 2018). The structure of the polar and time dependencies of the right-hand side of equation (3.43) are visible from the products of first-order solutions $\vec{v}_1^2$. Exponential functions of $-2\alpha_m t$ appear in both groups of

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terms. The stream function ψ and its constituent ψ_2 are introduced into the right-hand side of equation (3.43) to write

$$\begin{aligned} \Delta \mathcal{P}_{21} = & -q^2 C_{2m}^2 \left\{ \left[\frac{q^2}{(qr)^2} \left(\frac{d(qrj_m)}{dr} \right)^2 - \frac{q^4}{(qr)^2} (qrj_m)^2 + \right. \right. \\ & + \frac{C_{1m}}{C_{2m}} (m+1) r^{m-2} \frac{d(qrj_m)}{dr} \left. \right] (1-x^2) \left(\frac{dP_m(x)}{dx} \right)^2 + \\ & + \left[\frac{q^4}{(qr)^4} (qrj_m)^2 + \frac{C_{1m}}{C_{2m}} r^{m-3} (qrj_m) \right] (m(m+1)P_m(x))^2 \left. \right\} e^{-2\alpha_m t}, \end{aligned} \quad (3.44)$$

where $x = \cos \theta$. The quantity q and the coefficients C_{1m} and C_{2m} represent the influence of either $\alpha_m^{(p)}$ or $\alpha_m^{(n)}$ that correspond to the dependencies on time according to $\exp(-2\alpha_m^{(p)} t)$ or $\exp(-2\alpha_m^{(n)} t)$. The products of terms with $\exp(-\alpha_m^{(p)} t)$ and $\exp(-\alpha_m^{(n)} t)$ define a third contribution to the right-hand side, resulting in the dependency on time according to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$. The third contribution to the right-hand side of equation (3.43) reads

$$\begin{aligned} \Delta \mathcal{P}_{21}^{(pn)} = & - \left\{ \left[C_{2m}^{(p)} C_{2m}^{(n)} \frac{q^{(p)2} + q^{(n)2}}{r^2} \frac{d(q^{(p)} r j_m^{(p)})}{dr} \frac{d(q^{(n)} r j_m^{(n)})}{dr} \right. \right. \\ & - 2 C_{2m}^{(p)} C_{2m}^{(n)} \frac{q^{(p)2} q^{(n)2}}{r^2} (q^{(p)} r j_m^{(p)}) (q^{(n)} r j_m^{(n)}) \\ & + C_{1m}^{(n)} C_{2m}^{(p)} (m+1) q^{(p)2} r^{m-2} \frac{d(q^{(p)} r j_m^{(p)})}{dr} \\ & + C_{1m}^{(p)} C_{2m}^{(n)} (m+1) q^{(n)2} r^{m-2} \frac{d(q^{(n)} r j_m^{(n)})}{dr} \left. \right] \left(\frac{dP_m(x)}{dx} \right)^2 (1-x^2) \\ & + \left[C_{2m}^{(p)} C_{2m}^{(n)} \frac{q^{(p)2} + q^{(n)2}}{r^4} (q^{(p)} r j_m^{(p)}) (q^{(n)} r j_m^{(n)}) \right. \\ & + C_{1m}^{(n)} C_{2m}^{(p)} q^{(p)2} r^{m-3} (q^{(p)} r j_m^{(p)}) \\ & + C_{1m}^{(p)} C_{2m}^{(n)} q^{(n)2} r^{m-3} (q^{(n)} r j_m^{(n)}) \left. \right] (m(m+1)P_m(x))^2 \left. \right\} e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t}. \end{aligned} \quad (3.45)$$

When solving the Poisson equation, one firstly determines the general solution of the homogeneous equation, which, for the time dependency according to $\exp(-2\alpha_m^{(p)}t)$, reads

$$\mathcal{P}_{21,H} = - \sum_{l=0}^L D_{21l}^{(p)} r^l P_l(\cos \theta) e^{-2\alpha_m^{(p)}t} . \quad (3.46)$$

All the summations which arise in the solutions henceforth extend to an upper summation index symbolised by an upper-case letter. Since closed-form particular solutions $\mathcal{P}_{21,I}(r, \theta, t)$ for the right-hand sides in equations (3.44) and (3.45) seem to be out of reach, the particular solutions are therefore to be sought in the form of power series. The right-hand sides are approximated by series expansions of the spherical Bessel functions and the Legendre polynomials for a given value of the mode m of initial deformation. With an increase in the order of the approximation, the approximation accuracy rises. The details on the accuracy of the approximations and further development of the solutions in series expansion form of the Poisson equation for all time dependencies is given in section B of appendix A.

The solutions for the modified pressure are composed from the general solutions of the homogeneous and particular solutions of the inhomogeneous differential equation as

$$\mathcal{P}_{21}(r, \theta, t) = \mathcal{P}_{21,H}(r, \theta, t) + \mathcal{P}_{21,I}(r, \theta, t) . \quad (3.47)$$

The second-order pressure contribution p_{21} builds from all the solutions for the three different time dependencies reading

$$p_{21}(r, \theta, t) = p_{21}^{(p)}(r, \theta, t) + p_{21}^{(n)}(r, \theta, t) + p_{21}^{(pn)}(r, \theta, t) . \quad (3.48)$$

The following contributions to the solution emerge through the use of the modified pressure definition $\mathcal{P}_{21} = p_{21} + \bar{v}_1^2/2$ and for the positive time

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dependency reads

$$\begin{aligned}
 p_{21}^{(p)}(r, \theta, t) = & -e^{-2\alpha_m^{(p)}t} \left\{ \sum_{l=0}^L D_{21l}^{(p)} r^l P_l(x) - \left[\sum_{k=0}^N \sum_{l=0}^{2m} G_{2kl}^{(p)} \left(q^{(p)} r \right)^{2k+2m+2} P_l(x) \right. \right. \\
 & + \left. \sum_{k=0}^N \sum_{l=0}^{2m-1} \delta_{kl}^{(p)} \left(q^{(p)} r \right)^{2k+2m} P_l(x) + \sum_{k=1}^N \delta_{k(2m)}^{(p)} \left(q^{(p)} r \right)^{2k+2m} P_{(2m)}(x) \right] \Big\} \\
 & - \frac{1}{2} \left(\hat{\eta}_1^{(p)} \right)^2 \alpha_m^{(p)2} \left[f_r^{(p)2}(r) P_m(\cos \theta)^2 + f_\theta^{(p)2}(r) P_m^1(\cos \theta)^2 \right] e^{-2\alpha_m^{(p)}t} .
 \end{aligned} \tag{3.49}$$

For the negative time dependency and for the dependency on $\exp[(\alpha_m^{(p)} + \alpha_m^{(n)})t]$ the solutions are given in section B of appendix A. The coefficients D_{21l} for each time dependency are unknown and their determination is presented in a separate section C in appendix A. The first part of the solution "21", the terms in curly brackets, is the solution of the Poisson equation for the modified pressure, and the second contribution is the dynamic pressure from the first-order velocity field.

The corresponding second-order velocities are determined from the pressure field and the equations of motion (3.10)-(3.12). One can use the definition of the modified pressure $\mathcal{P}_{21}$ and the radial momentum equation (3.11) and obtains

$$\begin{aligned}
 -\frac{r^2}{Oh} \frac{\partial u_{r21}}{\partial t} + \frac{\partial^2}{\partial r^2} (r^2 u_{r21}) + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u_{r21}}{\partial \theta} \right) = & \frac{r^2}{Oh} \frac{\partial \mathcal{P}_{21}^{(p)}}{\partial r} + \frac{r^2}{Oh} \frac{\partial \mathcal{P}_{21}^{(n)}}{\partial r} \\
 & + \frac{r^2}{Oh} \frac{\partial \mathcal{P}_{21}^{(pn)}}{\partial r} - \frac{1}{Oh^2 \sin^2 \theta} \left(\frac{\partial \psi_2^{(p)}}{\partial t} \frac{\partial \psi^{(p)}}{\partial r} + \frac{\partial \psi_2^{(n)}}{\partial t} \frac{\partial \psi^{(n)}}{\partial r} \right. \\
 & \left. + \frac{\partial \psi_2^{(p)}}{\partial t} \frac{\partial \psi^{(n)}}{\partial r} + \frac{\partial \psi_2^{(n)}}{\partial t} \frac{\partial \psi^{(p)}}{\partial r} \right) .
 \end{aligned} \tag{3.50}$$

The second-order radial momentum equations for all time dependencies are shown in section C of appendix A. Equations (A.27) and (A.30) shown in section C for the second-order radial velocity u_{r21} are solved. For each

time dependency, the solutions u_{r21} consist of homogeneous and particular solutions of the partial differential equations. The second-order radial velocity u_{r21} reads

$$u_{r21}(r, \theta, t) = u_{r21}^{(p)}(r, \theta, t) + u_{r21}^{(n)}(r, \theta, t) + u_{r21}^{(pn)}(r, \theta, t) , \quad (3.51)$$

where for the positive time dependency the second-order radial velocity u_{r21} is

$$\begin{aligned} u_{r21}^{(p)}(r, \theta, t) = & - \sum_{l=0}^L \left[A_{21l}^{(p)} r^{l-1} + B_{21l}^{(p)} q^{(p)2} \frac{j_l(\sqrt{2}q^{(p)}r)}{q^{(p)}r} \right] l(l+1) P_l(x) e^{-2\alpha_m^{(p)}t} \\ & + \left\{ \sum_{k=0}^N \sum_{l=0}^{2m} \left[K_{1kl}^{(p)} \left(q^{(p)}r \right)^{2k+2m+1} + K_{2kl}^{(p)} \left(q^{(p)}r \right)^{2k+2m+3} \right] P_l(x) \right. \\ & + \sum_{k=0}^N \sum_{l=0}^{2m-1} K_{3kl}^{(p)} \left(q^{(p)}r \right)^{2k+2m+1} P_l(x) \\ & \left. + \sum_{k=1}^N K_{3k(2m)}^{(p)} \left(q^{(p)}r \right)^{2k+2m+1} P_{(2m)}(x) \right\} e^{-2\alpha_m^{(p)}t} \\ = & - \sum_{l=0}^L \left(A_{21l}^{(p)} f_{ar}^{(p)} + B_{21l}^{(p)} f_{br}^{(p)} \right) P_l(\cos \theta) e^{-2\alpha_m^{(p)}t} + A p_{r21}^{(p)}(r, \theta, t) . \end{aligned} \quad (3.52)$$

The solutions for the other time dependencies are given in section C of appendix A. With the radial velocity component u_{r21} known, the continuity equation (3.10) determines the second-order polar velocity component $u_{\theta21}$, which is composed as

$$u_{\theta21}(r, \theta, t) = u_{\theta21}^{(p)}(r, \theta, t) + u_{\theta21}^{(n)}(r, \theta, t) + u_{\theta21}^{(pn)}(r, \theta, t) . \quad (3.53)$$

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The first term on the right reads

$$\begin{aligned}
 u_{\theta 21}^{(p)}(r, \theta, t) = & - \left\{ \sum_{l=0}^L \left[A_{21l}^{(p)} (l+1) r^{l-1} \right. \right. \\
 & + B_{21l}^{(p)} q^{(p)2} \left((l+1) \frac{j_l(\sqrt{2}q^{(p)}r)}{q^{(p)}r} - \sqrt{2}j_{l+1}(\sqrt{2}q^{(p)}r) \right) \left. \right] P_l^1(x) \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m} \left[L_{1kl}^{(p)} (q^{(p)}r)^{2k+2m+1} + L_{2kl}^{(p)} (q^{(p)}r)^{2k+2m+3} \right] \frac{P_{l+1}(x) - P_{l-1}(x)}{\sqrt{1-x^2}} \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m-1} L_{3kl}^{(p)} (q^{(p)}r)^{2k+2m+1} \frac{P_{l+1}(x) - P_{l-1}(x)}{\sqrt{1-x^2}} \\
 & + \sum_{k=1}^N L_{3k(2m)}^{(p)} (q^{(p)}r)^{2k+2m+1} \frac{P_{(2m+1)}(x) - P_{(2m-1)}(x)}{\sqrt{1-x^2}} \left. \right\} e^{-2\alpha_m^{(p)}t} = \\
 = & - \sum_{l=0}^L \left(A_{21l}^{(p)} f_{a\theta}^{(p)} + B_{21l}^{(p)} f_{b\theta}^{(p)} \right) P_l^1(\cos \theta) e^{-2\alpha_m^{(p)}t} + A p_{\theta 21}^{(p)}(r, \theta, t) ,
 \end{aligned} \tag{3.54}$$

while the second and third contributions are presented in section D of appendix A.

The second-order deformation of the drop surface η_{21} follows from the structure of the above solutions and reads

$$\eta_{21}(\theta, t) = \sum_{l=0}^L \left(H_{21l}^{(p)} e^{-2\alpha_m^{(p)}t} + H_{21l}^{(n)} e^{-2\alpha_m^{(n)}t} + H_{21l}^{(pn)} e^{-\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)t} \right) P_l(\cos \theta) , \tag{3.55}$$

where the coefficients H_{21l} are unknown and different for the different time dependencies. Section F of appendix A details the determination of these coefficients. The deformed drop shape is governed by all Legendre polynomials in the sum. The reason for the latter is that pressure and surface deformation are coupled in the zero normal-stress boundary condition (3.15).

The homogeneous forms of equations (3.11)-(3.12) determine the second contributions to the second-order solutions, with subscript "22". Since the set of equations has the same structure as at first order, the deformation amplitude $\hat{\eta}_1$ of the drop surface is replaced by $\hat{\eta}_{22k}$ and the complex angular

frequency α_m by α_{2k} (number "2" in the subscript indicates the second-order solution, and k a deformation mode number). The replaced variables $\hat{\eta}_{22k}$ and α_{2k} contain the sum with the summation index k , which defines the deformation mode in second order approximation, concluding that all possible deformation modes k influence the results.

The second-order pressure, velocity and drop surface deformation for the contribution "22" are found as linear combinations of eigen-solutions of the underlying differential equations with the summation index k . The form of contribution "22" is similar to the first-order solutions, and the radial velocity component reads

$$\begin{aligned}
 u_{r22}(r, \theta, t) = & - \sum_{k=0}^K \hat{\eta}_{22k}^{(p)} \alpha_{2k}^{(p)} \left[r^{k-1} + \frac{2(k^2 - 1)}{2q_{2k}^{(p)} Q_k^{(p)} - q_{2k}^{(p)2}} \right. \\
 & \times \left(r^{k-1} - \frac{j_k(q_{2k}^{(p)} r)}{j_k(q_{2k}^{(p)})} \frac{1}{r} \right) \left. \right] P_k(\cos \theta) e^{-\alpha_{2k}^{(p)} t} \\
 & - \sum_{k=0}^K \hat{\eta}_{22k}^{(n)} \alpha_{2k}^{(n)} \left[r^{k-1} + \frac{2(k^2 - 1)}{2q_{2k}^{(n)} Q_k^{(n)} - q_{2k}^{(n)2}} \right. \\
 & \times \left(r^{k-1} - \frac{j_k(q_{2k}^{(n)} r)}{j_k(q_{2k}^{(n)})} \frac{1}{r} \right) \left. \right] P_k(\cos \theta) e^{-\alpha_{2k}^{(n)} t} = \\
 & = - \sum_{k=0}^K \left[\hat{\eta}_{22k}^{(p)} \alpha_{2k}^{(p)} e^{-\alpha_{2k}^{(p)} t} f_{2r}^{(p)}(r) + \hat{\eta}_{22k}^{(n)} \alpha_{2k}^{(n)} e^{-\alpha_{2k}^{(n)} t} f_{2r}^{(n)}(r) \right] P_k(\cos \theta),
 \end{aligned} \tag{3.56}$$

while the polar velocity component and pressure field are shown in section E of appendix A. The parameter q_{2k} in the homogeneous solutions "22" equals $\sqrt{\alpha_{2k}/Oh}$. The function f_{2r} is defined for each oscillation frequency and is introduced to keep the expressions of the flow field quantities compact. The following subsections will use them. The deformation of the drop surface for solution "22" is given as

$$\eta_{22}(\theta, t) = \sum_{k=0}^K \left(\hat{\eta}_{22k}^{(p)} e^{-\alpha_{2k}^{(p)} t} + \hat{\eta}_{22k}^{(n)} e^{-\alpha_{2k}^{(n)} t} \right) P_k(\cos \theta). \tag{3.57}$$

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The solutions of a new characteristic equation formed by the zero-normal stress boundary condition (3.15) of the second-order homogeneous system are the complex conjugate angular frequencies α_{2k} . The new characteristic equation is formally equal to the first-order equation (3.35), but, as the solution requires, it has to be solved for all the modes k .

The second-order solutions are complete with known coefficients A_{21l} , B_{21l} , H_{21l} for each time dependency, and the amplitudes $\hat{\eta}_{22k}^{(p)}$ and $\hat{\eta}_{22k}^{(n)}$. We use the three second-order boundary conditions (3.13)-(3.15) for the calculation of the three unknown coefficients A_{21l} , B_{21l} and H_{21l} , which is detailed in section F of appendix A. The two amplitudes $\hat{\eta}_{22k}^{(p)}$ and $\hat{\eta}_{22k}^{(n)}$ are determined using the initial conditions (3.16) (Renoult et al., 2018). The two contributions to the second-order deformation of the drop surface are mutually dependent, since their sum must satisfy the second-order initial condition for every summation index k in the solutions. The initial conditions are applied, and a set of two equations with the two unknown coefficients $\hat{\eta}_{22k}^{(p)}$ and $\hat{\eta}_{22k}^{(n)}$ is obtained. The first initial condition is written

$$\sum_{k=0}^K \left(H_{21k}^{(p)} + H_{21k}^{(n)} + H_{21k}^{(pn)} + \hat{\eta}_{22k}^{(p)} + \hat{\eta}_{22k}^{(n)} \right) P_k(x) = -\frac{1}{2m+1}. \quad (3.58)$$

Since P_0 is a constant, the term in the sum with $k = 0$ satisfies the condition with its right-hand side. For all the other indexes $k \geq 1$, the corresponding Legendre polynomials $P_k(x)$ depend on the polar angle. In that case, in order that the condition be satisfied, the sum of all the coefficients and amplitudes in the round brackets must then be zero. For $k = 0$, the calculation of the coefficients $H_{210}^{(p)}$, $H_{210}^{(n)}$ and $H_{210}^{(pn)}$ gives that

$$H_{210}^{(p)} + H_{210}^{(n)} + H_{210}^{(pn)} = -\frac{1}{2m+1}. \quad (3.59)$$

From this result, which follows from (3.58), the amplitudes $\hat{\eta}_{220}^{(p)}$ and $\hat{\eta}_{220}^{(n)}$ follow to be zero.

The requirement for a stationary drop center of mass for all times states that, at second order,

$$\int_0^\pi \left[6\eta_1^2(\theta, 0) + 4\eta_2(\theta, 0) \right] \cos \theta \sin \theta d\theta = 0, \quad (3.60)$$

from which condition we derive that

$$\int_{-1}^1 \left(\eta_{21}(x, 0) + \sum_{k=0}^K \left(\hat{\eta}_{22k}^{(p)} + \hat{\eta}_{22k}^{(n)} \right) P_k(x) \right) x dx = -3 \begin{pmatrix} m & m & 1 \\ 0 & 0 & 0 \end{pmatrix}^2. \quad (3.61)$$

All the integrals in the sum are zero, except that for $k = 1$. From the boundary conditions, the calculation of the coefficients $H_{211}^{(p)}$, $H_{211}^{(n)}$ and $H_{211}^{(pn)}$ shows that

$$H_{211}^{(p)} + H_{211}^{(n)} + H_{211}^{(pn)} = -3 \begin{pmatrix} m & m & 1 \\ 0 & 0 & 0 \end{pmatrix}^2. \quad (3.62)$$

With this result, it follows from (3.61) that the amplitudes $\hat{\eta}_{221}^{(p)}$ and $\hat{\eta}_{221}^{(n)}$ are zero.

This analysis shows that in the solutions "22" the cases $k = 0$ and $k = 1$ drop out. Becker, Hiller, and Kowalewski (1994) correlate these cases with a volumetric pulsation and a translatory motion of the drop, respectively. The former case is impossible for incompressible liquid, and since there is no resultant force acting on the drop that could induce a translatory motion, the latter case must be excluded from the solutions. These results cohere with the problem we are solving.

For the values $k \geq 2$, the second-order initial conditions (3.16) determine the two amplitudes $\hat{\eta}_{22k}^{(p)}$ and $\hat{\eta}_{22k}^{(n)}$. With H_{21k} , which are the same as H_{21l} in (3.55) for $l \geq 2$, we write

$$\begin{aligned} \hat{\eta}_{22k}^{(p)} = & \frac{1}{\alpha_{2k}^{(p)} - \alpha_{2k}^{(n)}} \left[\alpha_{2k}^{(n)} \left(H_{21k}^{(p)} + H_{21k}^{(n)} + H_{21k}^{(pn)} \right) \right. \\ & \left. - \left(2\alpha_m^{(p)} H_{21k}^{(p)} + 2\alpha_m^{(p)} H_{21k}^{(p)} + (\alpha_m^{(p)} + \alpha_m^{(n)}) H_{21k}^{(pn)} \right) \right], \end{aligned} \quad (3.63)$$

$$\begin{aligned} \hat{\eta}_{22k}^{(n)} = & \frac{1}{\alpha_{2k}^{(n)} - \alpha_{2k}^{(p)}} \left[\alpha_{2k}^{(p)} \left(H_{21k}^{(p)} + H_{21k}^{(n)} + H_{21k}^{(pn)} \right) \right. \\ & \left. - \left(2\alpha_m^{(p)} H_{21k}^{(p)} + 2\alpha_m^{(p)} H_{21k}^{(p)} + (\alpha_m^{(p)} + \alpha_m^{(n)}) H_{21k}^{(pn)} \right) \right]. \end{aligned} \quad (3.64)$$

3.2.3. Third-order solutions

The general solution for the third-order pressure is sought as a sum of two contributions, reading

$$p_3(r, \theta, t) = p_{31}(r, \theta, t) + p_{32}(r, \theta, t) . \quad (3.65)$$

The subscript "31" indicates the solution of the third-order equations including the nonlinear first- and second-order term products, while the subscript "32" denotes the solutions of the homogeneous system.

The solutions are sought for the subscript "31" first. Following the same procedure as in the second-order approximation, the divergence of the vectorial momentum equation of third order (3.18) and (3.19) is taken, and using the continuity equation (3.17), the third-order velocities are eliminated. The result is the differential equation for the third-order pressure p_{31} that is written as

$$\begin{aligned} \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial p_{31}}{\partial r} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\sin \theta}{r} \frac{\partial p_{31}}{\partial \theta} \right) = & - \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \left(u_{r1} \frac{\partial u_{r2}}{\partial r} + u_{r2} \frac{\partial u_{r1}}{\partial r} \right. \right. \\ & + \frac{u_{\theta 1}}{r} \frac{\partial u_{r2}}{\partial \theta} + \frac{u_{\theta 2}}{r} \frac{\partial u_{r1}}{\partial \theta} - 2 \frac{u_{\theta 1} u_{\theta 2}}{r} \left. \right) \Big] - \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left[\left(u_{r1} \frac{\partial u_{\theta 2}}{\partial r} + u_{r2} \frac{\partial u_{\theta 1}}{\partial r} \right. \right. \\ & + \frac{u_{\theta 2}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} + \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 2}}{\partial \theta} + \frac{u_{r2} u_{\theta 1}}{r} + \frac{u_{r1} u_{\theta 2}}{r} \left. \right) \sin \theta \Big] , \end{aligned}$$

which are re-written in a compact form as

$$\Delta p_{31} = -\nabla \cdot \left[\left(\vec{u}_1 \cdot \vec{\nabla} \right) \vec{u}_2 + \left(\vec{u}_2 \cdot \vec{\nabla} \right) \vec{u}_1 \right] . \quad (3.66)$$

The equation is re-written using the dot product rule to yield

$$\Delta \mathcal{P}_{31} = \nabla \cdot \left[\vec{u}_1 \times \left(\vec{\nabla} \times \vec{u}_2 \right) + \vec{u}_2 \times \left(\vec{\nabla} \times \vec{u}_1 \right) \right] , \quad (3.67)$$

where one defines the modified pressure as $\mathcal{P}_{31} = p_{31} + \vec{u}_1 \cdot \vec{u}_2$. Due to long terms on the right-hand side of equation (3.67), the detailed representation is omitted. The solutions are found following an identical procedure as in

the second-order calculation. Therefore, the critical step in finding the third-order solutions is to present the products of spherical Bessel functions and products of Legendre functions as series expansions. In this way, one is able to turn the Poisson equation into an ordinary differential equation. Those mathematical procedures are carried out and verified using the software MATHEMATICA. Finally, the work yields the solution for the modified pressure that reads

$$\begin{aligned} \mathcal{P}_{31}(r, \theta, t) = & - \sum_{h=0}^H \left[D_{31h}^{(p)} e^{-3\alpha_m^{(p)} t} + D_{31h}^{(n)} e^{-3\alpha_m^{(n)} t} + D_{31h}^{(ppn)} e^{-(2\alpha_m^{(p)} + \alpha_m^{(n)}) t} \right. \\ & + D_{31h}^{(pnm)} e^{-(\alpha_m^{(p)} + 2\alpha_m^{(n)}) t} + \sum_{k=0}^K D_{31hk}^{(pp)} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(p)}) t} + \sum_{k=0}^K D_{31hk}^{(nn)} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(n)}) t} \\ & \left. + \sum_{k=0}^K D_{31hk}^{(pn)} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(n)}) t} + \sum_{k=0}^K D_{31hk}^{(np)} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(p)}) t} \right] r^h P_h(x) + \mathcal{P}_{31,I}(r, \theta, t). \end{aligned}$$

Terms in the brackets containing coefficients D_{31h} and D_{31hk} present the homogeneous solutions, and $\mathcal{P}_{31,I}$ the particular solutions of the equation (3.67). The details for the latter solutions are not presented, since they contain a large number of terms. We identify all possible time dependencies in the terms in brackets by looking at the product of the first- and second-order solutions in the nonlinear terms in the momentum equations. Solutions are grouped and verified in the software MATHEMATICA by plugging them back into the equation (3.67). They are presented for all time dependencies in a general form as

$$\begin{aligned} \mathcal{P}_{31,I}(r, \theta, t) = & \mathcal{P}_{31}^{(p)}(r, \theta, t) + \mathcal{P}_{31}^{(n)}(r, \theta, t) + \mathcal{P}_{31}^{(ppn)}(r, \theta, t) + \mathcal{P}_{31}^{(pnm)}(r, \theta, t) \\ & + \mathcal{P}_{31}^{(pp)}(r, \theta, t) + \mathcal{P}_{31}^{(nn)}(r, \theta, t) + \mathcal{P}_{31}^{(pn)}(r, \theta, t) + \mathcal{P}_{31}^{(np)}(r, \theta, t). \end{aligned}$$

Substituting the definition of the modified pressure $\mathcal{P}_{31}$ into the radial momentum equation (3.18), one obtains

$$\begin{aligned} \frac{r^2}{Oh} \frac{\partial u_{r31}}{\partial t} - \frac{\partial^2}{\partial r^2} (r^2 u_{r31}) - \frac{\partial}{\partial x} \left((1 - x^2) \frac{\partial u_{r31}}{\partial x} \right) = & - \frac{r^2}{Oh} \frac{\partial \mathcal{P}_{31}}{\partial r} \\ & + \frac{r^2}{Oh} \left[\frac{u_{\theta 1}}{r} \left(\frac{\partial (r u_{\theta 2})}{\partial r} - \frac{\partial u_{r2}}{\partial \theta} \right) + \frac{u_{\theta 2}}{r} \left(\frac{\partial (r u_{\theta 1})}{\partial r} - \frac{\partial u_{r1}}{\partial \theta} \right) \right]. \end{aligned} \quad (3.68)$$

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One derives the solutions u_{r31} for each time dependency following an identical procedure as in the second-order approximation. Therefore the terms with products of spherical Bessel functions and their derivatives on the right-hand sides are approximated by their series expansions. Also, the series expansions in Legendre polynomials approximate the different products of Legendre functions. Due to the lengthy terms involved, they are produced, verified and stored in the software MATHEMATICA. A detailed representation of the derivations and details of these particular solutions is omitted here. The third-order radial velocity in a general form reads

$$u_{r31}(r, \theta, t) = u_{r31}^{(p)} + u_{r31}^{(n)} + u_{r31}^{(ppn)} + u_{r31}^{(pnn)} + u_{r31}^{(pp)} + u_{r31}^{(nn)} + u_{r31}^{(pn)} + u_{r31}^{(np)}, \quad (3.69)$$

where the terms for the time dependencies (p) and (pp) read

$$u_{r31}^{(p)}(r, \theta, t) = - \sum_{h=0}^H \left[A_{31h}^{(p)} r^{h-1} + B_{31h}^{(p)} q^{(p)2} \frac{j_h(\sqrt{3}q^{(p)}r)}{q^{(p)}r} \right] \times h(h+1)P_h(x)e^{-3\alpha_m^{(p)}t} + Ap_{r31}^{(p)}(r, \theta, t), \quad (3.70)$$

$$u_{r31}^{(pp)}(r, \theta, t) = - \sum_{h=0}^H \sum_{k=0}^K \left[A_{31hk}^{(pp)} r^{h-1} + B_{31hk}^{(pp)} \frac{j_h(Q_k^{(pp)}r)}{r} \right] \times h(h+1)P_h(x)e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(p)})t} + Ap_{r31}^{(pp)}(r, \theta, t), \quad (3.71)$$

and terms with different time dependencies are given in section G of appendix A. The particular solutions are denoted as $Ap_{r31}(r, \theta, t)$ for every time dependency, and they describe solutions of the inhomogeneous equation (3.68). The solutions consist of pairs of terms with the unknown coefficients A_{31h} and B_{31h} , as well as A_{31hk} and B_{31hk} , under sums with respect to the summation indexes h and k . The relations between the coefficients A_{31h} and A_{31hk} , and D_{31h} and D_{31hk} are given by section G of appendix A.

The continuity equation (3.17) and the radial velocity component yield the velocity component in the polar direction for the various time dependencies. In a general form it is written as

$$u_{\theta31}(r, \theta, t) = u_{\theta31}^{(p)} + u_{\theta31}^{(n)} + u_{\theta31}^{(ppn)} + u_{\theta31}^{(pnn)} + u_{\theta31}^{(pp)} + u_{\theta31}^{(nn)} + u_{\theta31}^{(pn)} + u_{\theta31}^{(np)}, \quad (3.72)$$

where the solutions for the time dependencies (p) and (pp) are

$$u_{\theta 31}^{(p)}(r, \theta, t) = Ap_{\theta 31}^{(p)}(r, \theta, t) - \sum_{h=0}^H (h+1)P_h^1(x) \left[A_{31h}^{(p)} r^{h-1} + B_{31h}^{(p)} q^{(p)2} \left(\frac{j_h(\sqrt{3}q^{(p)}r)}{q^{(p)}r} - \sqrt{3} \frac{j_{h+1}(\sqrt{3}q^{(p)}r)}{(h+1)} \right) \right] e^{-3\alpha_m^{(p)}t}, \quad (3.73)$$

$$u_{\theta 31}^{(pp)}(r, \theta, t) = Ap_{\theta 31}^{(pp)}(r, \theta, t) - \sum_{h=0}^H \sum_{k=0}^K (h+1)P_h^1(x) \left[A_{31hk}^{(pp)} r^{h-1} + B_{31hk}^{(pp)} \left(\frac{j_h(Q_k^{(pp)}r)}{r} - Q_k^{(pp)} \frac{j_{h+1}(Q_k^{(pp)}r)}{(h+1)} \right) \right] e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(p)})t}, \quad (3.74)$$

and the solutions with different time dependencies are given in section H of appendix A. The term $Ap_{\theta 31}(r, \theta, t)$ for every time dependency comes from the corresponding particular solution of the inhomogeneous radial momentum equation. The contribution “31” of the deformation amplitude of the drop shape in the third order has the form

$$\begin{aligned} \eta_{31}(\theta, t) = & \sum_{h=0}^H \left[H_{31h}^{(p)} e^{-3\alpha_m^{(p)}t} + H_{31h}^{(n)} e^{-3\alpha_m^{(n)}t} + H_{31h}^{(ppn)} e^{-(2\alpha_m^{(p)} + \alpha_m^{(n)})t} \right. \\ & + H_{31h}^{(pnn)} e^{-(\alpha_m^{(p)} + 2\alpha_m^{(n)})t} + \sum_{k=0}^K H_{31hk}^{(pp)} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(p)})t} + \sum_{k=0}^K H_{31hk}^{(nm)} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(n)})t} \\ & \left. + \sum_{k=0}^K H_{31hk}^{(pn)} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(n)})t} + \sum_{k=0}^K H_{31hk}^{(np)} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(p)})t} \right] P_h(x). \end{aligned} \quad (3.75)$$

The third-order boundary conditions (3.20)-(3.22) determine the set of unknown coefficients A_{31h} , B_{31h} and H_{31h} , as well as A_{31hk} , B_{31hk} and H_{31hk} for each time dependency and summation index h and k . The procedure is identical to the calculation of the corresponding second-order coefficients. The coefficients, due to the lengthy terms involved, are only possible to calculate and verify using the software MATHEMATICA. The details of the calculations are therefore omitted.

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The homogeneous forms of the equations (3.17)-(3.19) determine the second contributions to the third order, with subscript "32". These equations have the same structure as the second contribution to the second-order solutions, denoted with subscript "22". Therefore the complex angular frequencies α_{2k} are replaced by α_{3h} and the deformation amplitudes of the drop surface $\hat{\eta}_{22k}$ by $\hat{\eta}_{32k}$. The solution structure is similar to the second-order contributions "22". Finally, section I of appendix A gives the pressure field and polar velocity component, while the radial velocity component reads

$$\begin{aligned}
 u_{r32}(r, \theta, t) = & - \sum_{h=0}^H \hat{\eta}_{32h}^{(p)} \alpha_{3h}^{(p)} e^{-\alpha_{3h}^{(p)} t} \left[r^{h-1} + \frac{2(h^2 - 1)}{2q_{3h}^{(p)} Q_h^{(p)} - q_{3h}^{(p)2}} \right. \\
 & \times \left. \left(r^{h-1} - \frac{j_h(q_{3h}^{(p)} r)}{j_h(q_{3h}^{(p)})} \frac{1}{r} \right) \right] P_h(\cos \theta) \\
 & - \sum_{h=0}^H \hat{\eta}_{32h}^{(n)} \alpha_{3h}^{(n)} e^{-\alpha_{3h}^{(n)} t} \left[r^{h-1} \frac{2(h^2 - 1)}{2q_{3h}^{(n)} Q_h^{(n)} - q_{3h}^{(n)2}} \right. \\
 & \times \left. \left(r^{h-1} - \frac{j_h(q_{3h}^{(n)} r)}{j_h(q_{3h}^{(n)})} \frac{1}{r} \right) \right] P_h(\cos \theta) ,
 \end{aligned} \tag{3.76}$$

where the new function $Q_h = j_{h+1}(q_{3h})/j_h(q_{3h})$ and the new parameter $q_{3h} = \sqrt{\alpha_{3h}/O\hbar}$ are introduced. The deformation of the drop surface for solution "32" is obtained as

$$\eta_{32}(\theta, t) = \sum_{h=0}^H \left(\hat{\eta}_{32h}^{(p)} e^{-\alpha_{3h}^{(p)} t} + \hat{\eta}_{32h}^{(n)} e^{-\alpha_{3h}^{(n)} t} \right) P_h(\cos \theta) . \tag{3.77}$$

The complex conjugate angular frequencies α_{3h} are obtained as solutions of a new characteristic equation emerging from the zero-normal stress boundary condition (3.22) of the third-order homogeneous system. The new characteristic equation is formally equal to equation (3.35) from first order, but it has to be solved for all summation indexes h , as the solution requires.

The third-order solutions are fully characterised if the constants $\hat{\eta}_{32h}^{(p)}$ and $\hat{\eta}_{32h}^{(n)}$ for every h are known. The three initial conditions (3.23) determine these

coefficients. The relationship between the two contributions to the third-order drop surface deformation is due to the fact that their sum needs to satisfy the third-order initial condition for every summation index in the solution. The initial deformation is prescribed by a mode m . The first initial condition can be written as

$$\sum_{h=0}^H \left(H_{31h}^{(p)} + H_{31h}^{(n)} + H_{31h}^{(ppn)} + H_{31h}^{(pnn)} + H_{31h}^{(pp)} + H_{31h}^{(nn)} \right) \quad (3.78)$$

$$+ H_{31h}^{(pn)} + H_{31h}^{(np)} + \hat{\eta}_{32h}^{(p)} + \hat{\eta}_{32h}^{(n)} \Big) P_h(x) = -\frac{1}{3} \begin{pmatrix} m & m & m \\ 0 & 0 & 0 \end{pmatrix}^2, \quad (3.79)$$

where $x = \cos\theta$. The term in the sum with $h = 0$ satisfies the condition with its right-hand side, since P_0 is a constant. For all the other indexes $h \geq 1$, the corresponding Legendre polynomials $P_h(x)$ depend on the polar angle, so that the sum of all the coefficients and amplitudes in the round brackets must then be zero, in order that the condition is satisfied. For $h = 0$, the calculation of the coefficients H_{310} for all the time dependencies from the three boundary conditions yields that

$$\begin{aligned} & H_{310}^{(p)} + H_{310}^{(n)} + H_{310}^{(ppn)} + H_{310}^{(pnn)} + H_{310}^{(pp)} + H_{310}^{(nn)} \\ & + H_{310}^{(pn)} + H_{310}^{(np)} = -\frac{1}{3} \begin{pmatrix} m & m & m \\ 0 & 0 & 0 \end{pmatrix}^2. \end{aligned} \quad (3.80)$$

Consequently, the amplitudes $\hat{\eta}_{320}^{(p)}$ and $\hat{\eta}_{320}^{(n)}$ are zero.

The requirement that the drop center of mass is always stationary states that, at third order,

$$\int_0^\pi \left(\eta_3 + 3\eta_1\eta_2 + \eta_1^3 \right) \cos\theta \sin\theta d\theta = 0, \quad (3.81)$$

from which equation we derive

$$\begin{aligned} & \int_{-1}^1 \left(\eta_{31}(x, 0) + \sum_{h=0}^H \left(\hat{\eta}_{32h}^{(p)} + \hat{\eta}_{32h}^{(n)} \right) P_h(x) \right) x dx = \\ & - 2 \sum_{j=0}^{2m} \begin{pmatrix} m & m & j \\ 0 & 0 & 0 \end{pmatrix}^2 \begin{pmatrix} m & 1 & j \\ 0 & 0 & 0 \end{pmatrix}^2, \end{aligned} \quad (3.82)$$

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where $x = \cos \theta$. All the integrals in the sum are zero, except that for $h = 1$. The calculation of the coefficients H_{311} from the boundary conditions yields that

$$\begin{aligned} H_{311}^{(p)} + H_{311}^{(n)} + H_{311}^{(ppn)} + H_{311}^{(pnn)} + H_{311}^{(pp)} + H_{311}^{(nn)} \\ + H_{311}^{(pn)} + H_{311}^{(np)} = -2 \sum_{j=0}^{2m} \begin{pmatrix} m & m & j \\ 0 & 0 & 0 \end{pmatrix}^2 \begin{pmatrix} m & 1 & j \\ 0 & 0 & 0 \end{pmatrix}^2. \end{aligned} \quad (3.83)$$

Consequently, the amplitudes $\hat{\eta}_{321}^{(p)}$ and $\hat{\eta}_{321}^{(n)}$ are zero. This means that the cases $h = 0$ and $h = 1$, which correspond to a volumetric pulsation and a translatory motion of drop (Becker, Hiller, and Kowalewski, 1994), respectively, drop out from the solutions "32" of the homogeneous system of third-order equations. This property of the solutions is equivalent to that at second order.

The first and second third-order initial conditions define the other amplitudes for $h \geq 2$. The third-order surface deformation amplitude for positive time dependency reads

$$\begin{aligned} \hat{\eta}_{32h}^{(p)} = \frac{1}{\alpha_{3h}^{(p)} - \alpha_{3h}^{(n)}} \left[\alpha_{3h}^{(n)} \left(H_{31h}^{(p)} + H_{31h}^{(n)} \right. \right. \\ + \sum_{k=0}^K \left(H_{31hk}^{(pp)} + H_{31hk}^{(nn)} + H_{31hk}^{(pn)} + H_{31hk}^{(np)} \right) + H_{31h}^{(ppn)} + H_{31h}^{(pnn)} \Big) \\ - \left(3\alpha_m^{(p)} H_{31h}^{(p)} + 3\alpha_m^{(n)} H_{31h}^{(n)} + (2\alpha_m^{(p)} + \alpha_m^{(n)}) H_{31h}^{(ppn)} + (\alpha_m^{(p)} + 2\alpha_m^{(n)}) H_{31h}^{(pnn)} \right. \\ + \sum_{k=0}^K (\alpha_m^{(p)} + 2\alpha_{2k}^{(p)}) H_{31hk}^{(pp)} + \sum_{k=0}^K (\alpha_m^{(n)} + 2\alpha_{2k}^{(n)}) H_{31hk}^{(nn)} \\ \left. \left. + \sum_{k=0}^K (\alpha_m^{(p)} + \alpha_{2k}^{(n)}) H_{31hk}^{(pn)} + \sum_{k=0}^K (\alpha_m^{(n)} + \alpha_{2k}^{(p)}) H_{31hk}^{(np)} \right) \right], \end{aligned} \quad (3.84)$$

where the deformation amplitude $\hat{\eta}_{32h}^{(n)}$ is given in section I of appendix A. We end up with complete third-order solutions. The following section will use the above solutions to study the problem of an oscillating Newtonian drop.

3.3. Results and discussion

This section evaluates the solutions derived above. The conservation of the drop volume for varying deformation parameter η_0 and for Ohnesorge numbers investigated in the present study is verified first. In this way the maximum allowable values of η_0 for varying Ohnesorge number ensuring a volume accuracy to 2 % are determined. The second section presents the drop surface shapes at maximum deformation during one period. Then, for the mode of initial deformation $m = 2$, the results of the nonlinear behaviour are compared with data from existing literature, and the results of the analysis are extended to higher modes. Further inspection considers the influences of the initial mode of deformation on mode coupling, on the oscillation frequency, and on the damping rate. At the end of the section, the results show that shape oscillations may occur for supercritical Ohnesorge numbers for higher-order modes of initial drop deformation, where the linear analysis predicts aperiodic motion.

3.3.1. Volume inspection

All the field variables in the weakly nonlinear theory are presented as truncated power series of a small deformation parameter, as shown in equation (2.12) and the following. Since the drop surface shape is also included, the non-dimensional drop volume may deviate from its exact value of $V_s = 4\pi/3$, i.e., the drop volume is not accurately conserved (Renoult et al., 2018; Zrnić and Brenn, 2021b). Before showing any results using the complete solutions up to the third order, the relative volume deviation is analytically determined for the validation of the method. The drop volume deviation is largest at different instants of time, which appear in every oscillation period and depend on the deformation parameter η_0 . The time of the largest deformation in one oscillation period is denoted with t_{max} . The non-dimensional drop volume $V(t)$ is determined as

$$\begin{aligned} V(t) &= \frac{2\pi}{3} \int_{-1}^1 r_s^3(\theta, t) d \cos \theta \\ &= \frac{2\pi}{3} \int_{-1}^1 \left[1 + \eta_1(\theta, t)\eta_0 + \eta_2(\theta, t)\eta_0^2 + \eta_3(\theta, t)\eta_0^3 \right]^3 d \cos \theta . \end{aligned} \quad (3.85)$$

3. Newtonian drop shape oscillations

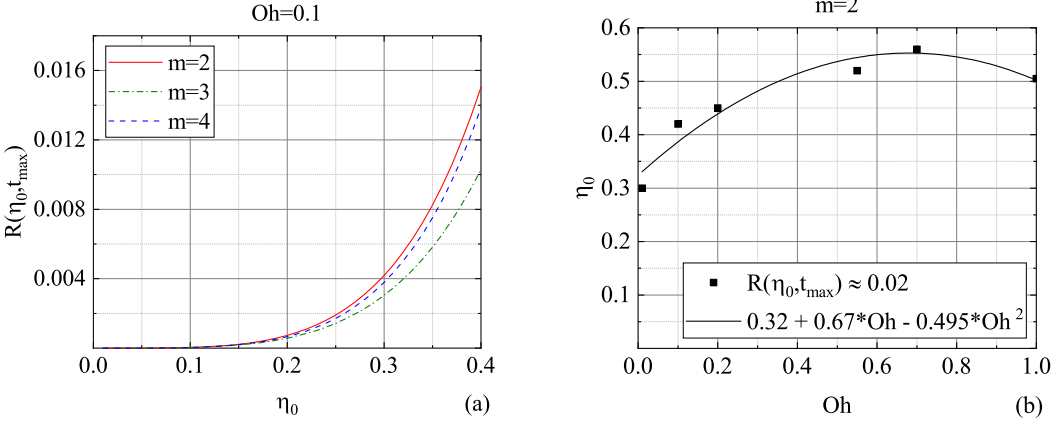


Figure 3.2.: (a) For initial modes of deformation $m = 2, 3$ and 4 and $Oh = 0.1$, maximum relative drop volume deviating from the exact value as a function of the deformation parameter η_0 . (b) Limiting the relative drop volume deviation to 2%, maximum allowable deformation parameter as a function of the Ohnesorge number for the initial deformation mode $m = 2$. The figures originate from Zrnić, Berglez, and Brenn (2022).

Therefore the relative volume deviation from the exact value is

$$\frac{V(t) - V_s}{V_s} = \frac{1}{2} \int_{-1}^1 \left[r_s^3(\theta, t) - 1 \right] d \cos \theta =: R(\eta_0, t), \quad (3.86)$$

where the values of R are all positive, oscillate in time and increase with η_0 . One obtains figure 3.2 (a) by using the equation (3.86) and varying the modes of initial deformation $m = 2, 3$ and 4 at the instant of time $t = t_{\max}$ of largest volume deviation. The figure shows volume deviations by not more than 0.5% from the exact value for all three cases, deformation parameters $\eta_0 \leq 0.3$ and $Oh = 0.1$. The volume deviation also depends on the Ohnesorge number that influences the velocity field in the drop. Changes in velocity change the surface deformation, since they are coupled via boundary conditions, and consequently change the resultant drop volume.

Figure 3.2 (b) is reproduced plotting the maximum allowable deformation parameter as a function of the Ohnesorge number, keeping the relative volume deviation at 2%. The trend of this maximum allowable deformation parameter with the Ohnesorge number varies in the range between $Oh = 0.1$ and 1 and is shown with a quadratic fit of the data points represented

with the solid line. One concludes that, with this method, relatively large oscillatory drop deformations can be covered for a wide range of Ohnesorge numbers.

Another validation is performed for the weakly nonlinear results up to the second order of approximation for initial modes of deformation $m = 2, 3$ and 4 . The analysis, however, also allows for modes $m > 4$. The conservation of the drop volume as a function of deformation parameter and time is analyzed, where the non-dimensional drop volume equals $V(t) = (2\pi/3) \int_{-1}^1 r_s^3(x, t) dx$. The relative volume deviation from the exact value is defined in the equation (3.86). The top diagram in figure 3.3 presents the relative volume deviation using the solutions up to the second order of approximation for the initial deformation modes $m = 2, 3$ and 4 at the instant of time $t = t_{max}$ that defines the largest values of relative volume deviation. The values of R are positive, increase with η_0 and oscillate in time. Figure 3.3 shows the relative volume deviation increasing with the deformation parameter η_0 and with the initial deformation mode m . For $\eta_0 = 0.4$, R at t_{max} is around 2.5%, 0.4% and 0.8% for $m = 2, 3$ and 4 , respectively.

The calculation of the energy contents of the drop in time is another essential aspect used for the validation of the weakly nonlinear solutions. The energy conservation gives insight into the transformation of the mechanical energy forms during the oscillation and shows the dissipation of mechanical energy. The fields dealing with transport processes across the drop surface may find these predictions relevant. The sum of the kinetic E_k and surface energies E_{surf} equals the total energy E_{tot} . Kinetic and surface energies are computed via relations

$$E_{surf} = \frac{1}{2} \int_0^\pi r_s \sqrt{r_s^2 + \left(\frac{dr_s}{d\theta} \right)^2} \sin \theta d\theta, \quad (3.87)$$

$$E_k = \frac{1}{4} \int_\theta \int_{r=0}^{r=r_s(\theta, t)} \vec{v}^2 r^2 \sin \theta dr d\theta, \quad (3.88)$$

where r_s defines the position of the surface of the drop and depends on time t and polar angle θ , r is the radial coordinate and $\vec{v}^2$ is the scalar product of the velocity vector with itself, $\vec{v} \cdot \vec{v}$. The corresponding energies are shown

3. Newtonian drop shape oscillations

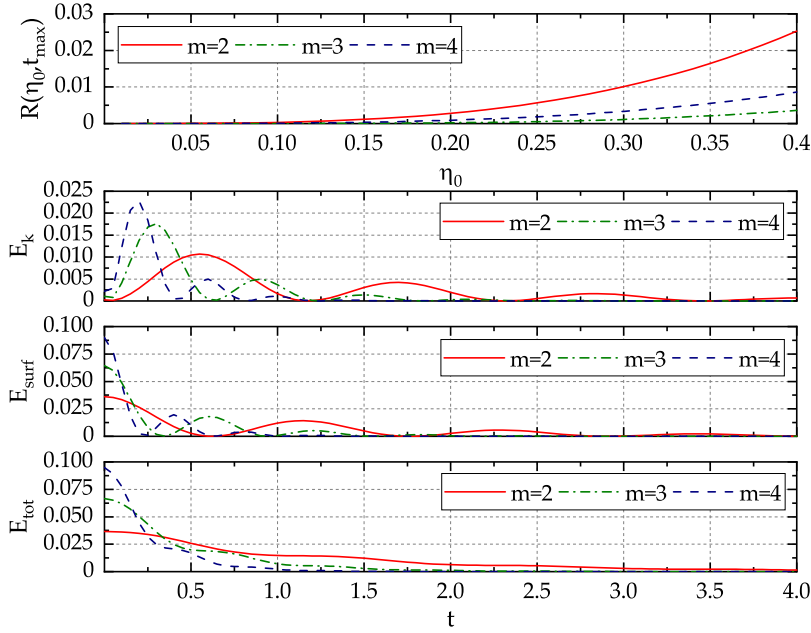


Figure 3.3.: Top figure - maximum relative drop volume deviation as a function of the deformation parameter η_0 . From top to bottom: kinetic, surface and total energies as functions of time for $Oh = 0.1$, $\eta_0 = 0.3$. The figures originate from Zrnić and Brenn (2021a).

in figure 3.3 for the Ohnesorge number 0.1. The values in the diagrams and relations in equation (3.87) are non-dimensional, relating to the surface energy $4\pi a^2\sigma$. Since the field variables contain truncated series expansions, the calculation of the energies is limited to second order of approximation. The droplet is most deformed at the initial time, containing the maximum surface energy. The initial conditions ensure a quiescent drop surface, but the bulk of the drop is not restricted by conditions and therefore is in motion. The kinetic and surface energies are continuously transformed into each other. The sum of the former and latter energies reveals the total energy as a function of time. The total energy decreases monotonically in time due to dissipation. The maximum decrease rate is achieved at the time instance when the rate of dissipation is highest. For the highest deformation mode studied at the beginning of the motion, the dissipation rate is greatest.

3.3.2. Deformed shapes of the drop surface

The present analysis shows the deformed drop surface shapes up to third order and presents the drop shapes for different instances of time during the drop's motion. The meridional sections of deformed drops for the initial deformation modes $m = 2, 3$ and 4 are given in figures 3.4, 3.5 and 3.6, respectively. The first three states of maximum deformation are shown, since the drop shapes are presented for the first oscillation period. The related non-dimensional time is described in the definition $r_s(\theta, t)$ for every figure. Each figure depicts the results from the present weakly nonlinear theory (WNL) to first order (Lamb) and to third order. The first initial condition is derived from the volume conservation (2.20), from which follows the volume correction in every order of approximation. For $t = 0$, constant corrections of the shape between the first- and third-order results are present along the circumference of the meridional section in the direction of the polar angle. The equal differences appear at the initial states for every initial deformation mode m . Table 3.1 lists the degrees of Legendre polynomials contributing to the field variables of second and third order for the various modes of initial deformation. The results presented in all figures in this subsection correspond to the Ohnesorge number of 0.1.

Figure 3.4 displays the deformed shapes for initial mode $m = 2$, which describes the well-known prolate-to-oblate oscillations with an initial aspect ratio of the drop length / width $(L/W) = 1.76$. The drop aspect ratio L/W is defined as $(r_s(0, t) + r_s(\pi, t)) / 2 r_s(\pi/2, t)$. Since the drop shape oscillations are damped by viscous stress, the maximum deformed state is at the initial time. The second prolate state is therefore less deformed than the first. The

m	Second-order approximation	Third-order approximation
2	0, 2, 4	0, 2, 4, 6
3	0, 2, 4, 6	1, 3, 5, 7, 9
4	0, 2, 4, 6, 8	0, 2, 4, 6, 8, 10, 12

Table 3.1.: For the modes of initial deformation $m = 2, 3$ and 4 , degrees of Legendre polynomials contributing to the solutions at second and third order.

3. Newtonian drop shape oscillations

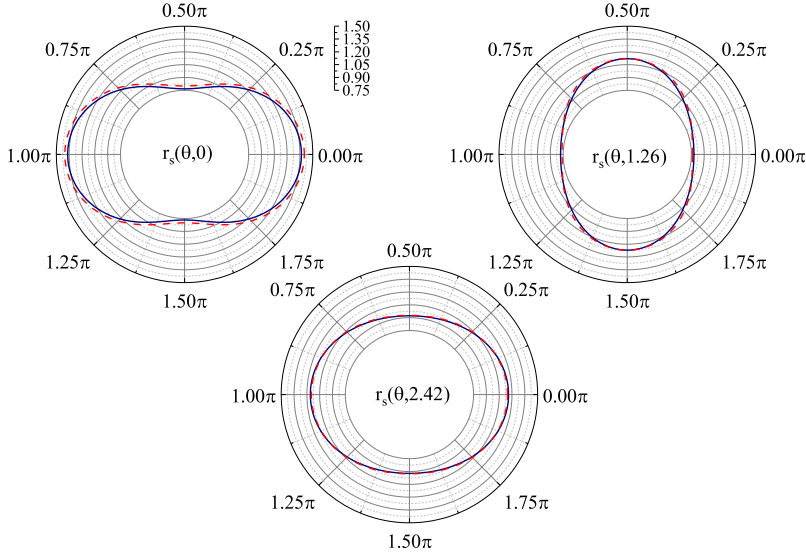


Figure 3.4.: The first three maximum deformed drop shapes, axisymmetric around the abscissa axis ($0 - \pi$), for the initial deformation mode $m = 2$ with $\eta_0 = 0.4$ and $Oh = 0.1$. The results present the WNL of first order (---) and third order (—). The figures originate from Zrnić, Berglez, and Brenn (2022).

differences between the first- and third-order results decreases with ongoing time. The shapes collapse at small oscillation amplitudes. The collapse is caused by the fact that higher-order modes are of low influence at small deformations.

The three- and four-lobed drop shapes in their first three maximum deformed states are shown in figures 3.5 and 3.6. As shown in table 3.1, the surface deformation includes Legendre polynomials of even and of odd degrees for $m = 3$. The surface deformation includes only Legendre polynomials of even degrees, for $m = 4$. With increasing initial deformation mode, the number of higher-order modes increases, thus increasing the number of terms in the series expansion of the drop surface shape. Therefore, looking at meridional sections of higher initial modes $m > 2$ one concludes that a greater number of terms markedly change the drop shapes compared with the first-order results.

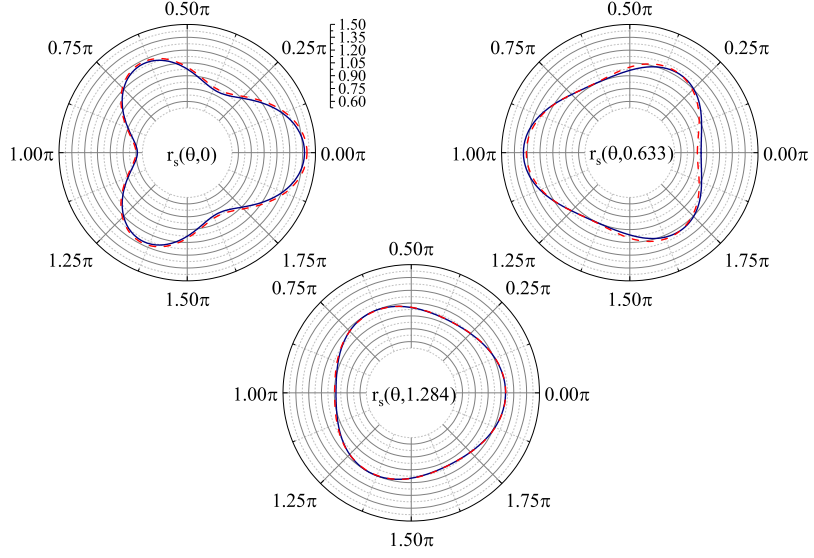


Figure 3.5.: The first three maximum deformed drop shapes, axisymmetric around the abscissa axis ($0 - \pi$), for the initial deformation mode $m = 3$ with $\eta_0 = 0.4$ and $Oh = 0.1$. The figures originate from Zrnić, Berglez, and Brenn (2022).

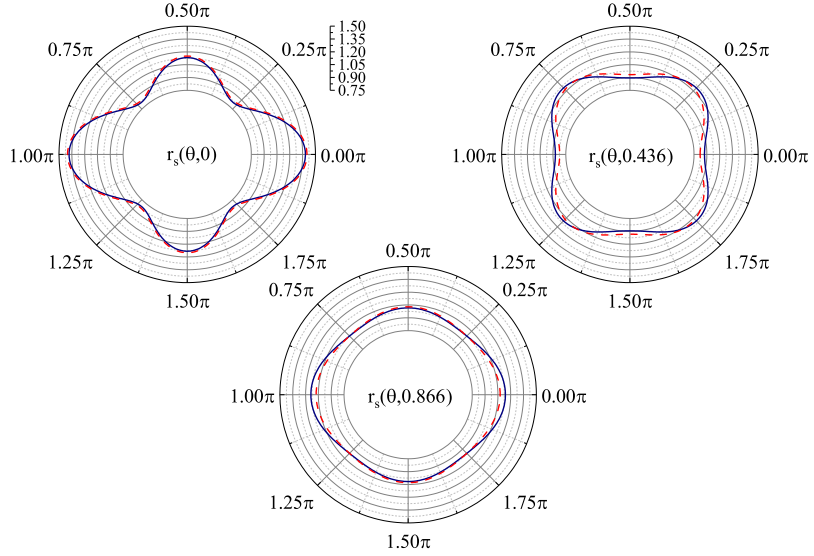


Figure 3.6.: The first three maximum deformed drop shapes, axisymmetric around the abscissa axis ($0 - \pi$), for the initial deformation mode $m = 4$ with $\eta_0 = 0.4$ and $Oh = 0.1$. The figures originate from Zrnić, Berglez, and Brenn (2022).

3. Newtonian drop shape oscillations

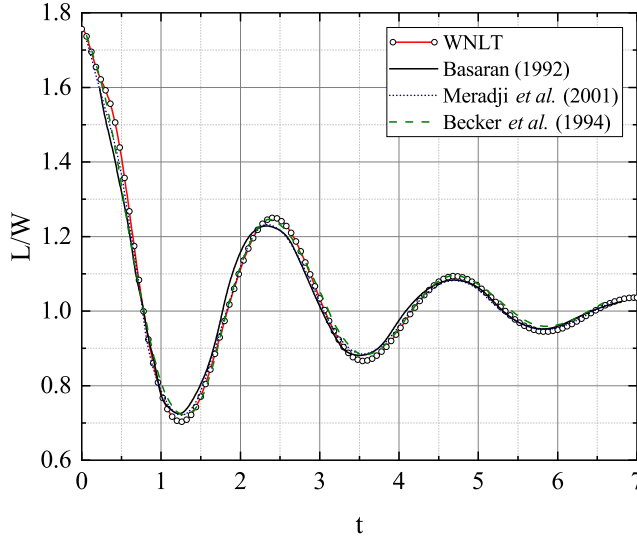


Figure 3.7.: Aspect ratio L/W of a drop as a function of time from the WNLT with $\eta_0 = 0.4$, $Oh = 0.1$ and $m = 2$, compared against results from existing literature. The figure originates from Zrnić, Berglez, and Brenn (2022).

Figure 3.7 shows a validation of the drop surface deformation. The figure depicts numerical results for the drop aspect ratio L/W as a function of time by Basaran (1992), Becker, Hiller, and Kowalewski (1994) and Meradji et al. (2001) in comparison to the present WNLT. The data from literature is extracted from the corresponding diagrams in the papers using a MATLAB routine. For the smoothness of the curves between the extracted data points, b-splines are used. The comparison shows good agreement between the results from the WNLT and the literature data. Comparison of Basaran (1992) and the WNLT results gives the largest deviations, which occur immediately after the start of the motion. It may be explained by the differences between the initial states of the drop bulk of those two works. The difference is that in the WNLT initial fluid motion is present in contrast to the numerical work, which starts an oscillation from the quiescent state.

3.3.3. The drop shape oscillation time scales

This subsection studies two well-known effects often demonstrated for the motion of the two-lobed deformation mode $m = 2$ appearing in nonlinear drop-shape oscillations. The first effect is the asymmetry of the times a drop spends in the prolate and in the oblate states of deformation during one oscillation period. The second effect is a decrease of the oscillation frequency with increasing drop deformation. These two nonlinear effects are predicted by the WNLT and analyzed and validated against existing literature. The WNLT results of time asymmetry between different states of deformation, as well as the WNLT results of oscillation frequency, are extracted from traces of the drop north pole position ($\theta = 0$) as a function of time. The oscillation frequency is denoted $\alpha_{res,i}$, where subscript i indicates the imaginary part. It is determined from the first oscillation period, which is defined in this case as the time elapsed between the first two maximum deformed states, i.e., between the first two maximum prolate shapes for the mode of initial deformation $m = 2$. From the same oscillation period, the excess time spent in the prolate state is determined. The frequency change is computed relative to the first-order oscillation frequency $\alpha_{m,i}$.

Figure 3.8 (a) presents the prolate excess-time effect for the mode of initial deformation $m = 2$. Experimental results by Trinh and Wang (1982), and numerical results by Basaran (1992) and Meradji et al. (2001) are plotted for comparison. The Ohnesorge number of the drop equals 0.1. Trinh and Wang (1982) performed experiments on neutrally buoyant silicon oil and carbon tetrachloride mixture drops in distilled water and measured the percentage of time the drop spent in prolate shapes. In the results by Trinh et al., the corrections of non-dimensional frequencies due to the liquid/liquid system are explained in Zrnić and Brenn (2021b). The results by Basaran (1992) and Meradji et al. (2001) in figure 3.8 (a) are obtained by numerical simulation of the time evolution of large-amplitude drop oscillations with $Oh = 0.1$. They applied different methods. Basaran (1992) used the Galerkin-finite element method, and Meradji et al. (2001) the CFD software *Fluid Dynamics Application Package* (FIDAP).

The WNLT results as functions of the aspect ratio L/W are also shown in figure 3.8 (a). Moreover, the WNLT results are near the numerical data

3. Newtonian drop shape oscillations

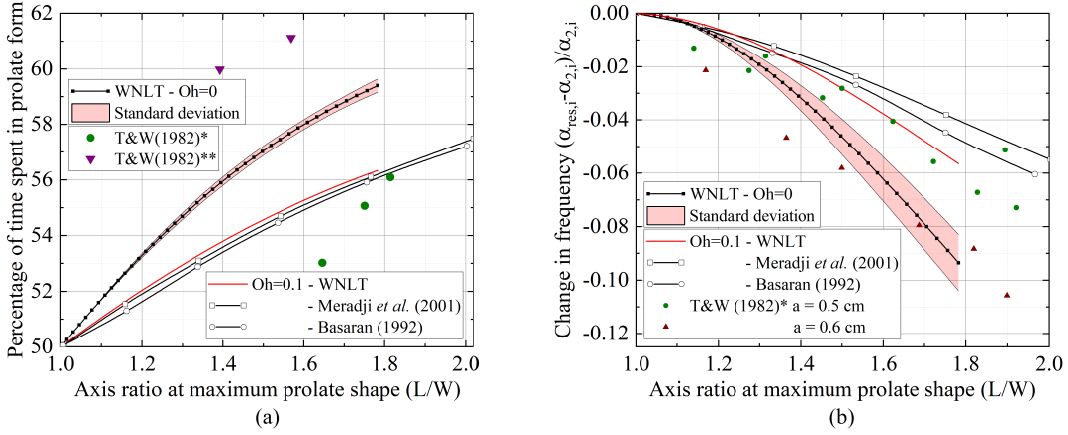


Figure 3.8.: (a) Percentage of the oscillation period spent in prolate shape and (b) decrease of the angular frequency as functions of the drop aspect ratio L/W for the fundamental initial mode. The values are obtained from the first oscillation. Experimental data by Trinh & Wang (1982) for the initial steady prolate drive of the drop is denoted as T&W(1982)*, and for the initial static prolate drop shape as T&W (1982)**. WNLt results with $Oh = 0$ are by Zrnić and Brenn (2021b). The figures originate from Zrnić, Berglez, and Brenn (2022).

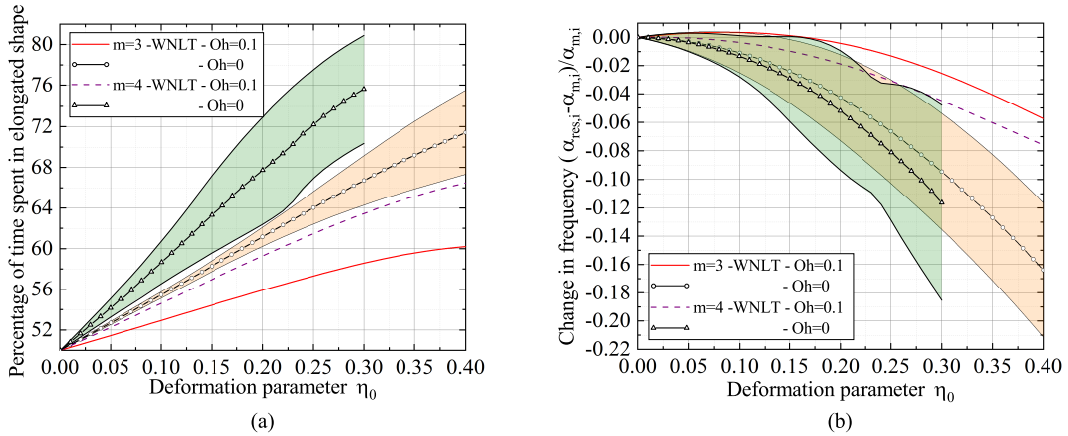


Figure 3.9.: (a) Percentage of the oscillation period spent in elongated shape, and (b) change of the angular oscillation frequency as functions of the deformation parameter η_0 for the modes $m = 3$ and $m = 4$. The values are obtained from the first oscillation. WNLt results with $Oh = 0$ are by Zrnić and Brenn (2021b). The figures originate from Zrnić, Berglez, and Brenn (2022).

from Basaran (1992) and Meradji et al. (2001). Comparing the deviation of the WNLT results with the experimental data by Trinh and Wang (1982), one concludes less deviation for the initially steady prolate driven drop than for the static case. The difference may be explained with the velocity field in the drop bulk that is present both in the WNLT and in the experiment with the initial steady prolate drive of the drop. The influence of the initial drop state on the oscillations is evident from the differences between the two groups of experimental data in figure 3.8 (a).

In figure 3.8 (b) the frequency decrease with increasing drop deformation is shown. The numerical data by Basaran (1992) and Meradji et al. (2001), together with the WNLT results, show the same trend. However, the relative deviation predicted by the WNLT is 16% greater than Basaran's and 29% greater than Meradji et al.'s results at the aspect ratio around $L/W = 1.775$. It is due to a difference in initial motion of the drop bulk, where the numerical simulations treat the drop as initially deformed and at rest, while the WNLT predicts motion in the drop bulk initially. The initial state of motion in the drop has been a subject of discussion by Mashayek and Ashgriz (1998) and Trinh and Wang (1982). The latter experimental work analysed effects of the initial drop state on the shape oscillations. The difference between the WNLT and the numerical data increases with the initial drop deformation. The effect is explained by the fact that the initial velocity in the drop increases with the drop deformation (Basaran, 1992). The second reason is the way of measuring the time period of the first oscillation, which Basaran (ibid.) does in the following manner: plotting the north pole's trace in time, Basaran defines the prolate time with a first half-prolate part of the oscillation, i.e., the prolate time is twice the first half-prolate time. He states on page 190 that the prolate time of the first cycle is defined to be twice the time that it takes the drop to relax from the initial state to the spherical shape. In contrast to that, the period of the first oscillation of the WNLT is taken differently, e.g. the period $t_{p,1}$ in figure 3.10 (a). The prolate time for the corresponding period is the time when the radial coordinate of the north pole is greater than 1.

In figs. 3.9 (a) and (b), excess time and frequency decrease are presented as functions of the drop deformation parameter η_0 for the higher modes of

initial deformation $m = 3$ and $m = 4$. Firstly, the percentage of the oscillation period the drop north pole ($\theta = 0$) spends at greater distances from the drop centre than in the spherical state is depicted in figure 3.9 (a). An elongation of the drop in the direction of the symmetry axis ($0 - \pi$) is indicated with the results in the corresponding figure. With an increase of the deformation parameter η_0 and the mode of initial deformation, the excess time increases. Secondly, the frequency change from the first oscillation period of the drop north pole ($\theta = 0$) divided by the imaginary part of the first-order oscillation frequency $\alpha_{m,i}$ for each corresponding mode m is determined and shown in figure 3.9 (b). At small deformation amplitudes, the frequency for the three-lobed initial deformation increases against the first-order result. The WNLT predicts that, for mode $m = 3$, values of the deformation parameter above 0.15 lead to a frequency decrease. The same trend of an increase at $m = 3$ is observed in the inviscid case, and in both cases, viscous and inviscid, it is specific to the north pole of the drop. This non-monotonic behaviour does not appear at its south pole. Greater frequency change than $m = 3$ is observed in the four-lobed initial deformation mode. A validation of these WNLT results is not possible because of the lack of data from the literature.

Figures 3.8 and 3.9 show the results of inviscid drops by Zrnić and Brenn (2021b) to quantify the difference between the viscous and the inviscid cases. The difference between the latter and the present work is due only to the drop liquid viscosity. Since quasi-periodicity of the oscillations is found in the inviscid case, the quantities depicted are based on the first ten oscillations, and their variations are represented by the standard deviations denoted with shaded areas. The comparison between the results show that both nonlinear effects are weaker for the viscous than for the inviscid drop.

Next, the time evolution of the periodic drop surface motion is studied. For the fundamental initial mode $m = 2$, the drop Ohnesorge number $Oh = 0.1$ and two different values of the deformation parameter η_0 , figure 3.10 (a) depicts the position of the drop north pole as a function of time. The evolution of the oscillation frequency is analysed, and the frequency is characterized by the oscillation period length, which is defined as the time elapsed between two extreme states of deformation or between two spherical states of the drop. Denoted by $t_{p,1} - t_{p,4}$ in figure 3.10 (a) and in

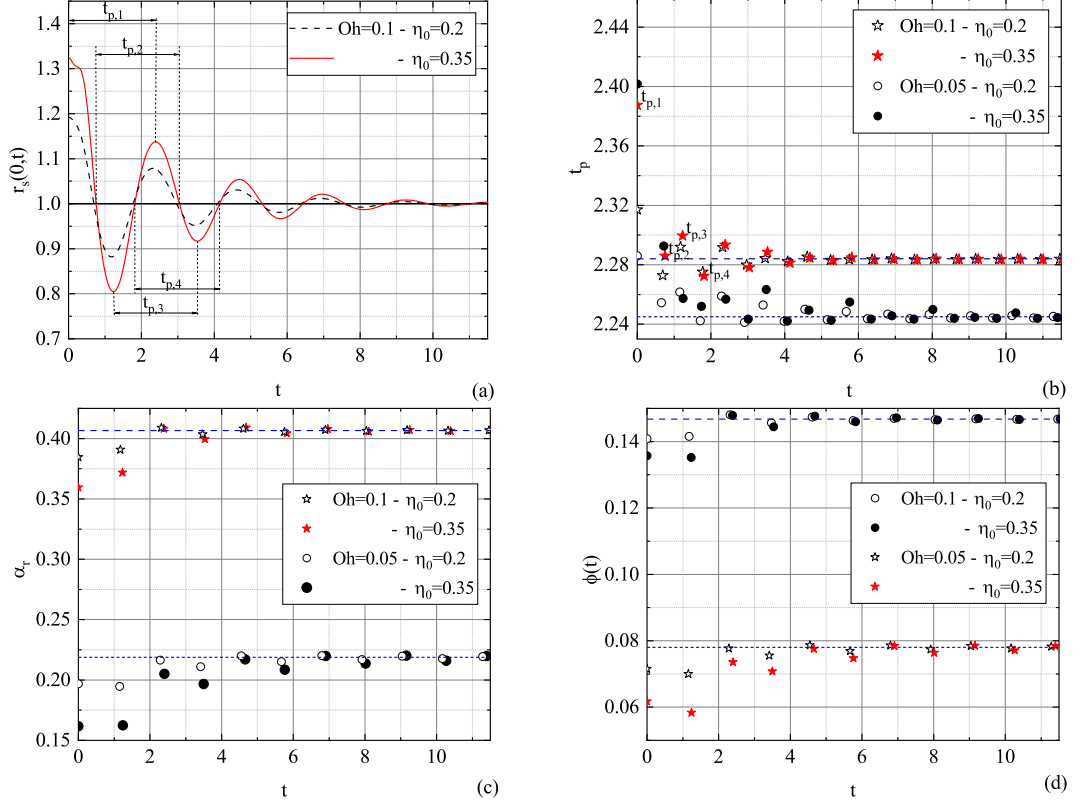


Figure 3.10.: (a) The WNLt results of the drop north pole position as a function of time for the deformation parameters $\eta_0 = 0.2$ and 0.4 and $Oh = 0.1$ for $m = 2$. Double arrows show periods extracted from different starting positions. (b) Period durations t_p measured from different starting points during the oscillation, as shown in (a), as functions of time t . (c) Damping rate determined for different pairs of displacement maxima or minima with ongoing time. (d) Loss angle $\phi(t)$ as a function of time. Dashed lines show the linear solutions. The figures originate from Zrnić, Berglez, and Brenn (2022).

figure 3.10 (b) marked next to the red star symbols, the first four periods for $\eta_0 = 0.35$ are shown. The corresponding evaluation of the oscillation period is applied to the complete oscillations as shown in figure 3.10 (a). One concludes from the results of this analysis that, the period length changes and leads to a resultant increase of the frequency with ongoing time. Prosperetti (1980a) already stated that a time dependency of the frequency for nonlinear drop oscillations is indicated by the change of the period length in time.

3. Newtonian drop shape oscillations

Our analysis is carried out for a viscous case $Oh = 0.1$ and a much less viscous case $Oh = 0.05$, and for two different deformation parameters each. In figure 3.10 (b) the results present the oscillation periods, which converge to the values of the linear case in a non-monotonic manner. Predicted by the linear theory, the non-dimensional oscillation period for the drop Ohnesorge number $Oh = 0.1$ is 2.284, and for $Oh = 0.05$ it is 2.245, depicted by the blue dashed and dotted horizontal lines, respectively. For the larger deformation parameter $\eta_0 = 0.35$ and at the initial time, the period is by 4.5 % and 6.9 % longer than the linear solution, i.e. the oscillation frequency is lower, for both $Oh = 0.1$ and $Oh = 0.05$. It refers to the noticeable dampening effect at large drop deformations. At deformation parameter $\eta_0 = 0.2$, the period length converges faster to the value of the linear case with ongoing time. For the case of $\eta_0 = 0.35$, the period length converges faster to the linear value for the larger Ohnesorge number. While observing the latter case one concludes a strong dependency of mode coupling on the drop liquid viscosity that was also found by Becker, Hiller, and Kowalewski (1994). The trend of the viscosity influence on mode coupling is consistent with the dampening, which is marked at a large Oh and expected from a physical point of view. On the other hand, for the less viscous drop, more oscillation cycles are needed for a return to the linear regime. One of the unexpected findings is that the period length suddenly increases, even during cycles with small maximum deformations. The reason is the beat between different modes of oscillation created by the mode coupling. One of the further consequences of mode coupling is the quasi-periodicity of the drop shape oscillations Zrnić and Brenn (2021b). This phenomenon is related to the beat caused by the interaction of various modes of oscillation excited in the course of the motion. However, as shown in figure 3.10 (b), it is damped as the viscosity increases and is manifested in much smaller fluctuations of the data with the higher Ohnesorge number than for the lower one.

After the analysis of the frequency based on the results of the oscillations depicted in figure 3.10 (a), the damping rate is studied on the same basis. Pairs of data points depicting two states of maximum positive or negative displacement from the spherical state characterize the damping rate. Thus, the two maximum positive or negative points define the beginning and the

end of an analysed oscillation. Starting times $t_{p,1}$ and $t_{p,3}$ in figure 3.10 (a) mark examples of such oscillations. Based on every pair of minimum or maximum values at an earlier time t_1 and a later time t_2 , the damping rate is deduced from the ratio of the surface point positions $r_s(0, t_1) = Ae^{-\alpha_r t_1}$ and $r_s(0, t_2) = Ae^{-\alpha_r t_2}$ as per $\alpha_r = \ln[(r_s(0, t_2)/r_s(0, t_1))]/(t_1 - t_2)$. Using these relations, the damping rate α_r is plotted against the starting time of an oscillation for drops with different Ohnesorge numbers and deformation parameters η_0 and shown in figure 3.10 (c). A marked difference between the results for the different deformation parameters 0.2 and 0.35 appears for the Ohnesorge number of 0.1, but both sets of results converge to the value of 0.407. This value is the real part of the complex angular frequency α_2 predicted by the linear theory, and it is depicted by the blue dashed horizontal line. For the drop with $Oh = 0.05$, the damping rate converges in time to the blue dotted horizontal line at the value of 0.219, which corresponds to the real part of the complex angular frequency α_2 for $Oh = 0.05$ calculated using the linear theory. Viscosity affects the damping rate differently in the nonlinear drop oscillations, which is concluded by comparing the phenomenon between the two Ohnesorge numbers of 0.1 and 0.05. As expected, the greater viscosity dampens the system more rapidly, so that, at $\eta_0 = 0.35$, the drop with $Oh = 0.1$ approaches the linear value faster than the drop with $Oh = 0.05$. In both cases, since the deformations decrease, their behaviour reflects the return of nonlinear damped drop-shape oscillations with ongoing time to a linear motion. The viscous influence on the drop shape oscillations is also reflected by the loss angle $\phi(t) = \arctan(\alpha_r/\alpha_i)$ of the motion. The results are plotted in figure 3.10 (d). For a given Oh , the loss angle increases by increasing the initial deformation of the drop and its Ohnesorge number. The loss angle with ongoing time converges to the linear values of 0.0780022 and 0.145754 for $Oh = 0.05$ and 0.1, respectively.

In contrast to figure 3.10 (c), where the motion of the drop north pole was analysed, figure 3.11 depicts the time evolution of the drop aspect ratio

$$\frac{r_s(0, t)}{r_s(\pi/2, t)}, \quad (3.89)$$

from $m = 2$ instead. Similarly, Basaran (1992) studied the trace of the aspect ratio in time as shown in figure 3.11 (a) for the drop Ohnesorge number

3. Newtonian drop shape oscillations

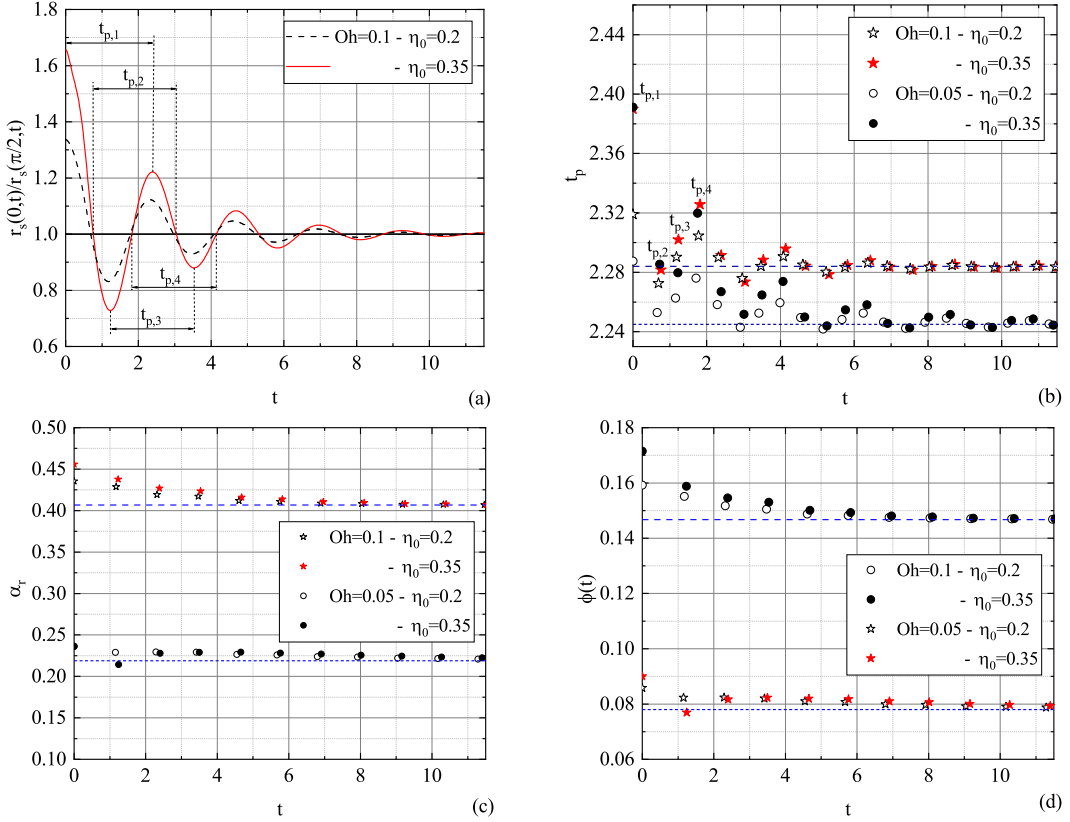


Figure 3.11.: (a) The drop aspect ratio from the present WNLT as a function of time for the deformation parameters $\eta_0 = 0.2$ and 0.35 and $Oh = 0.1$. Periods t_p starting from different instants in time are denoted with double arrows. (b) Periods t_p measured from different starting times during the oscillation, as shown in (a) as functions of time. (c) Damping rate determined from different pairs of maximum or minimum aspect ratio. (d) Loss angle $\phi(t)$. The linear solutions are marked with dashed lines. The figures originate from Zrnić, Berglez, and Brenn (2022).

$Oh = 0.1$. Figures 3.11 (b) - (d) show the evolutions of the oscillation period t_p , the damping rate α_r and the loss angle ϕ in time, respectively, each for $m = 2$ and with the initial deformation parameters $\eta_0 = 0.2$ and $\eta_0 = 0.35$ and for the drop Ohnesorge numbers $Oh = 0.05$ and $Oh = 0.1$. From the trace of the drop aspect ratio in figure 3.11 one can extract the oscillation

periods, and the damping rate reads

$$\alpha_r = \ln \left[\frac{\frac{r_s(0,t_2)}{r_s(\pi/2,t_2)} - 1}{\frac{r_s(0,t_1)}{r_s(\pi/2,t_1)} - 1} \right] / (t_1 - t_2). \quad (3.90)$$

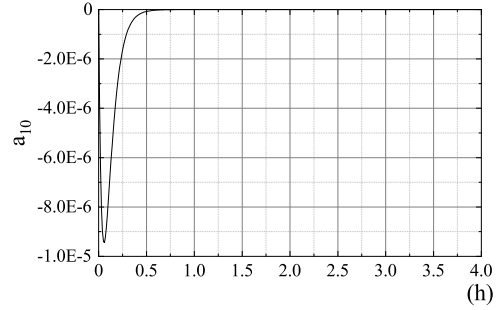
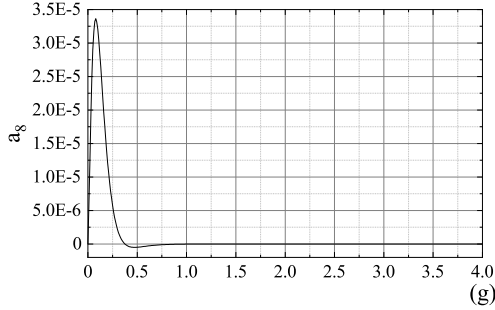
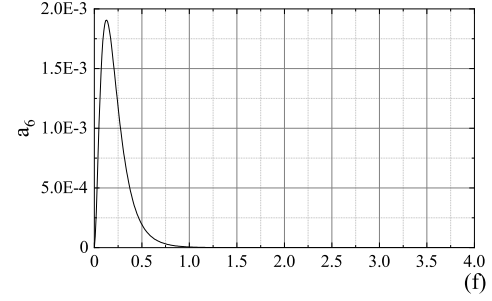
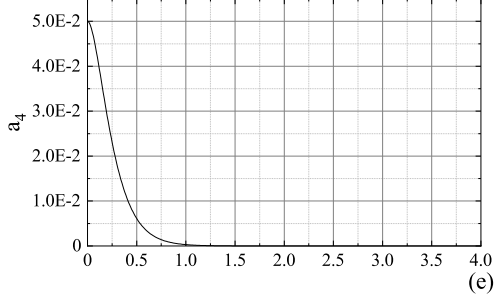
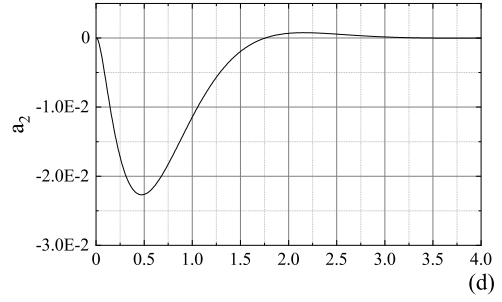
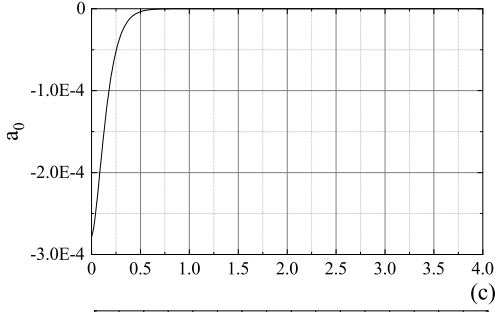
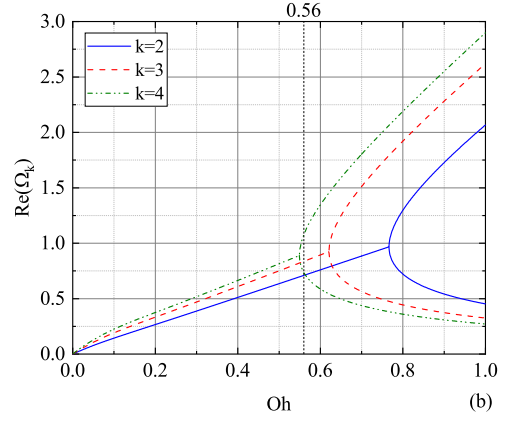
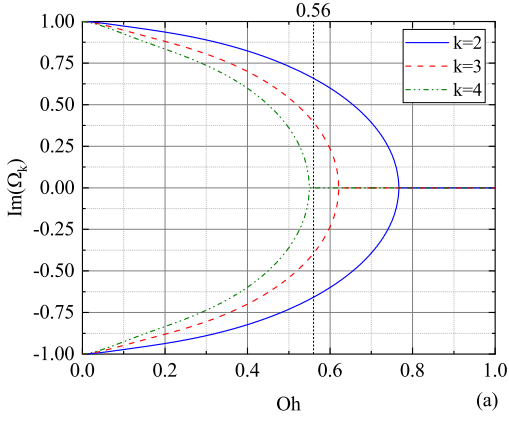
An interesting fact shown by these data is that the damping rate α_r , for the initial deformation parameters $\eta_0 = 0.2$ and $\eta_0 = 0.35$, decreases in time, which corresponds to the physical expectation. The difference between the data in the present figure 3.11 (c) and in figure 3.10 (c) is the geometrical drop property followed in time. The aspect ratio includes the effect from a phase difference between the surface points on the north pole and on the equator, which enters the ratio. On the other hand, the north pole position alone does not represent this effect. The position of a surface point on the equator shows the same increase of the damping rate in time. It is important to note that the damping rates measured from all positions converge in time, i.e. the damping rates decrease with ongoing time and approach the same linear result.

3.3.4. Oscillations from a supercritical state

Starting from an initial state of non-equilibrium, damped oscillators may return to their equilibrium state via damped oscillations or in an aperiodic manner. For the initially deformed drops presently studied, the drop Ohnesorge number Oh is the quantity that decides the behaviour of the drop. The solutions of the characteristic equation of the drop in figs. 3.12 (a) and 3.12 (b) show this behaviour. With increasing drop Oh , the oscillation frequency decreases, while the damping rate increases. At the cutoff value of Oh , the oscillation frequency reaches zero, and the damping rate bifurcates. For Ohnesorge numbers less than the cutoff value, therefore, the drop performs damped oscillations, while it behaves aperiodically for Oh greater than that.

Figures 3.12 (a) and 3.12 (b) show that the cutoff value of Oh depends on the mode of drop deformation. The cutoff value of Oh decreases with increasing mode number, while the corresponding damping rate increases. Therefore, values of Oh exist with damped oscillatory drop behaviour for a given mode, but, at the same time, with aperiodic behaviour of modes higher

3. Newtonian drop shape oscillations



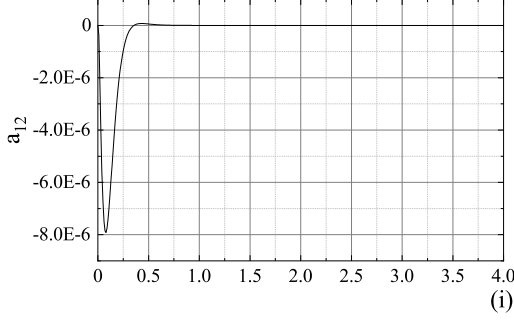


Figure 3.12.: The characteristic equation of drops with different Ohnesorge numbers for the modes of deformation 2, 3, 4 gives oscillation frequency (a) and damping rate (b) of viscous drops. $\Omega_k = \alpha_k / \alpha_{k,0}$. (c)-(i) The time-dependent coefficients in the series expansion of the drop surface shape for the initial mode of deformation $m = 4$, $Oh = 0.56$ and $\eta_0 = 0.05$. The figures originate from Zrnić, Berglez, and Brenn (2022).

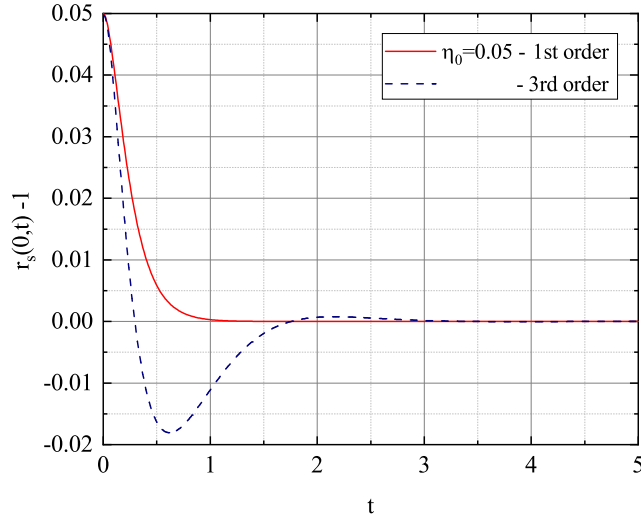


Figure 3.13.: The trace of the drop north pole deviation from the spherical state in time for the mode of initial deformation $m = 4$ and the drop Ohnesorge number $Oh = 0.56$ obtained by the first-order (linear) and third-order theory. The figure originates from Zrnić, Berglez, and Brenn (2022).

than that. For the example of $Oh = 0.56$ marked in figs. 3.12 (a) and 3.12 (b), a drop deformed according to the modes $m = 2$ or $m = 3$ performs damped oscillations, while for mode $m = 4$ it behaves aperiodically.

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Since nonlinear dynamics couples various modes, in which a mode m of initial deformation excites other modes k during the motion, a drop behaving aperiodically for a high mode of initial deformation may nonetheless show damped oscillatory behaviour due to the lower-order modes excited during the motion.

One such case with the mode $m = 4$ of initial drop deformation and the drop Ohnesorge number of 0.56 is studied here. The deformation parameter η_0 is set to the relatively small value of 0.05 in order to keep the relative volume deviation below 1%. The mode $m = 4$ couples with the even-numbered modes $k = 0, 2, 6, 8, 10$ and 12 only. In figs. 3.12 (c)-(i), the coefficients a_k of the series expansion for the drop surface shape related to the Legendre polynomials $P_k(x)$ are presented as functions of time. The linear result, which cannot account for mode coupling, would only predict aperiodic behaviour according to $m = 4$. The coefficients for all the modes appearing are oscillatory, except a_0 and a_4 , which are aperiodic. The higher the oscillatory mode, the more quickly it dies out. The two-lobed mode survives longest, thus dominating the motion. To the best of our knowledge, this phenomenon caused by the coupling of a high-order mode of initial drop deformation to lower-order modes has not been reported before (Zrnić, Berglez, and Brenn, 2022). Once the Ohnesorge number exceeds the cutoff value of all the modes, including $m = 2$, the drop returns to its equilibrium shape in an aperiodic manner.

Figure 3.13 represents the trace of the displacement of the drop north pole for the initial mode $m = 4$ and $Oh = 0.56$ as predicted by the linear and by the third-order theories. The solution of the characteristic equation of the drop yields the damping rates of 9.1292 and 6.24152 of the (linear) aperiodic motion, in which the latter dominates, since the motion dampened at the former rate disappears much more rapidly. An attempt could be made to calculate this value using equation (48) from Chandrasekhar (1959)

$$\alpha_{r,C} = \frac{m(2m+1)(m+2)}{2(2m^2+4m+3)} \frac{1}{Oh}, \quad (3.91)$$

assuming that $Oh = 0.56$ is large enough to allow for the application of this approximation for $\nu \rightarrow \infty$, and analysing oscillations at small deformations. The result from this equation, however, is 3.78151, thus underpredicting the

correct result by 39%. For the Chandrasekhar equation (3.91) to represent damping rates at $m = 4$ to an accuracy better than 10% of the correct value, the drop Ohnesorge number must be as high as 0.905. The linear results predict an aperiodic motion, presented by the solid red line in figure 3.13, while the nonlinear, third-order result represents an oscillatory motion induced by the coupling to modes of lower order than that of the initial deformation. We conclude that, even for small amplitudes, the linear description fails if motions for higher-order modes are described. These modes are coupled to other, both lower- and higher-order modes, and consequently yield a resultant oscillatory motion, even for Ohnesorge numbers which are supercritical for the mode of initial deformation.

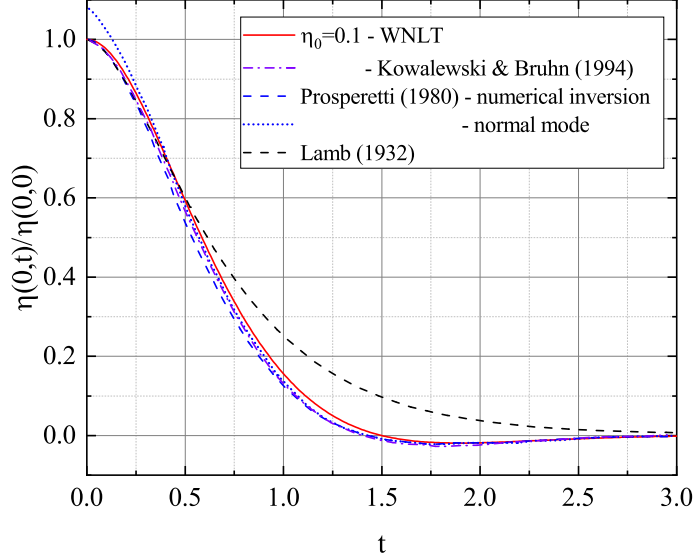


Figure 3.14.: Normalised drop deformation at the north pole for initial mode $m = 2$ and $Oh = 0.6$. The figures originate from supplementary material by Zrnić, Berglez, and Brenn (2022).

Another set of data was compared with the weakly nonlinear theory as a validation of the phenomenon found above. As seen in the previous figure, it is only partially true that, in the limit of small initial deformation amplitudes, the linear model gives the correct description of the motion. We plot the normalized drop north pole displacement for the fundamental mode $m = 2$

and $Oh = 0.6$ against time. We compare the present WNLT with results from linear theories by Prosperetti (1980a) and Lamb (1932), and with the nonlinear model by Kowalewski and Bruhn (1994). Prosperetti (1980a) presents results treating the drop oscillation as an initial-value problem governed by an integro-differential equation for the time-dependent coefficients in the expansion of the drop shape. Prosperetti's analysis showed that, for $m = 2$, in the range of $0.5657 < Oh < 0.7665$ of the drop Ohnesorge number, the linear normal-mode analysis of the motion may miss the fact that a motion starting out aperiodically may become periodic with ongoing time. The solution of the initial-value problem is depicted by the dashed blue line in figure 3.14. The normal-mode result by Prosperetti is depicted by the dotted line. The result by Lamb (1932) was extracted from Kowalewski and Bruhn (1994). The latter work discussed the fully viscous nonlinear model developed by Becker, Hiller, and Kowalewski (1994). As already seen by Kowalewski and Bruhn (1994), a considerable difference exists between the nonlinear and linear results, which is due to the large Ohnesorge number. It confirms the fact that, even for low deformation amplitudes, the linear theory fails in describing the motion for the fundamental initial deformation mode. The small difference between the results from the WNLT and by Kowalewski and Bruhn (*ibid.*) is due to different initial states of the drop, where in the latter work the initial rates of change of all the expansion coefficients for the drop shape are set to zero.

3.4. Experiments on Newtonian oscillating drops

Next to the theoretical study of nonlinear drop shape oscillations yielding the derivations and results detailed above, the present section deals with an experimental study on the same topic. The study is performed on Newtonian liquids, and it is divided into two parts. First is a short overview of the applied method and properties of the liquids used in the measurements. The second part depicts the results of the Newtonian drop shape oscillations with the focus on time scales. The experimental observations are essential for better understanding of the nonlinear effects related to the frequency and damping rate of the shape oscillations. This work was published in Zrnić

and Brenn (2019).

3.4.1. Overview of experimental method and liquids applied

The experimental investigation uses the technique of acoustic levitation (Yarin et al., 1999) for the damped shape oscillations of Newtonian liquid drops. An ultrasonic levitator makes individual drops levitate, i.e., the drop is contactlessly placed in a fixed position, and the method allows an investigation of various physical processes without any influences from contacting mechanical parts.

The drops consisting of the test liquids are produced with a thin syringe, and the drop diameters are in the range between 1 mm and 3 mm. At framing rates up to 4 kHz, a high-speed camera was used to record the images of the levitated drop under back-light illumination. The acquired images have the resolution of 300 Pxl/mm with the uncertainty of ± 2 Pxl.

The levitated drops are excited by amplitude-modulating the ultrasound near the resonance frequency of the base mode $m = 2$, and the amplitude of excitation was around 10% of the equilibrium drop radius. Since the author is interested in damped shape oscillations, the focus of the experiment starts once the modulation is switched off. The carrier signal driving the acoustic transducer is always active during the experiments, so that the drop remains levitated. Once the drop is placed in the acoustic levitator, the drop radius is measured. The author records the damped shape oscillation from which the height and width of the drop in time are postprocessed. Based on the experimental data, both the angular frequency and the damping rate of the drop shape oscillations are obtained. For further details on the experimental method the reader is referred to Brenn and Plohl (2015), Brenn and Teichtmeister (2013), and Yarin et al. (1999).

Aqueous glycerol solutions are used as the test liquids. The test solutions are prepared in demineralized water with different glycerol mass fractions. A capillary viscometer measures the dynamic viscosity μ^* , and a pendant-drop tensiometer measures the surface tension σ^* of the test solutions. The author determined the density of the solutions by weighing a known volume of the test liquids. The liquid properties measured at the temperature of 24°C are

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Table 3.2.: Properties of the studied aqueous glycerol solutions at 24°C. The table originates from Zrnić and Brenn (2019).

Glycerol mass fraction w [wt.%]	Density ρ [kg/m ³]	Surface tension σ^* [N/m]	Viscosity μ^* [mPa s]
20	1045 ±1%	0.069 ±4%	1.62 ±10%
30	1068 ±1%	0.067 ±4%	2.23 ±10%
50	1121.5 ±1%	0.065 ±4%	5.25 ±10%
65	1161 ±1%	0.063 ±4%	11.75 ±10%

listed in table 3.2 and are in good agreement with values known from the literature (Takamura, Fischer, and Morrow, 2021).

The height and width of the drop as functions of time are obtained from the recorded images of drop shape oscillations. In figure 3.15, the oscillation is depicted for a 20 wt.% solution drop with the equilibrium diameter of 2.05 mm. A general description of a damped oscillation (Brenn and Plohl, 2015) is

$$y(t) = y_0 + A \cos(\omega_{2,i}t + \varphi_0) \cdot e^{-\omega_{2,r}t}, \quad (3.92)$$

which represents the time-dependent distance $y(t)$ between the north and south poles of the drop, i.e., the drop height. The drop width is defined equivalently to the drop height. In equation (3.92), y_0 is the equilibrium drop height or the equilibrium drop width, $\omega_{2,i}$ is the angular oscillation frequency, t is time, A the oscillation amplitude at time $t = t_0$ from which on the data is modeled, φ_0 is a reference phase angle and $\omega_{2,r}$ is the damping rate for the fundamental mode $m = 2$. In figure 3.15 on the bottom, the oscillations were fitted in both directions of deformation. The author measured the average values for the frequency and the damping rate, and results measured in the case of large deviations between the values obtained in the equatorial and polar directions are discarded. Shown in figure 3.15 on the top, the first five and the rest of the oscillations are fitted separately. The first five oscillations correspond to large-amplitude (LA) oscillations and the rest to small-amplitude (SA) oscillations. From the fitting, the author measures the complex angular frequency $\omega_2 = \omega_{2,r} + i\omega_{2,i} = \omega_{2,r} + i2\pi \cdot f$ for the large ω_2^{LA} and small ω_2^{SA} amplitudes.

3.4. Experiments on Newtonian oscillating drops

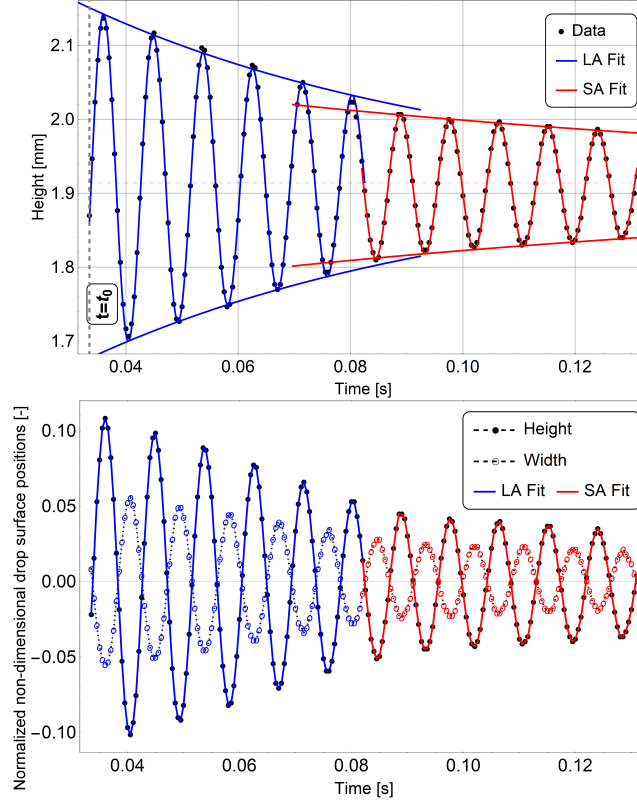


Figure 3.15.: Damped oscillations of a levitated drop with 20 wt.% aqueous glycerol solution. (Top) Drop height as a function of time for the large-amplitude (LA) and for the small-amplitude oscillations (SA) and (Bottom) normalized non-dimensional drop surface positions as functions of time, both with the fitting curves. The figures originate from Zrnić and Brenn (2019).

Figure 3.15 on the bottom presents the normalized non-dimensional drop surface positions as functions of time. In this figure, the oscillation amplitude in the equatorial direction is always smaller than the oscillation amplitude in the polar direction for reasons of the spheroidal drop geometry. The amplitude in the polar direction defines the drop oscillation amplitude in the present context.

Figure 3.15 shows the oscillations for the large- and small-amplitude regimes, where the following values are obtained. According to the first five oscillations, the droplet amplitude is initially about 11.6% of the equilibrium

3. Newtonian drop shape oscillations

drop radius, and in the last part about 6.9%. The measured complex angular frequencies are

$$\omega_2^{LA} = 15 \text{ s}^{-1} + i2\pi \cdot 113 \text{ Hz}, \quad \omega_2^{SA} = 7 \text{ s}^{-1} + i2\pi \cdot 113.8 \text{ Hz}. \quad (3.93)$$

Following from the analysis of the fitting results of the function for the drop height or width to the measured data, the determined complex angular frequency ω_2 has an uncertainty of $\pm 10\%$ in the damping rate, and of $\pm 1\%$ in the oscillation frequency. For both the SA oscillation and the LA oscillation, the frequencies are nearly the same. However, the damping rate in the case of the LA oscillation is nearly double the value of the SA oscillation.

The surface tension σ and the dynamic viscosity μ of the drop liquid may be determined from the measured complex angular frequency. The corresponding oscillating drop method predicts the material properties of the drop liquid, and this relies on the linear theory. The method holds good only if the initial oscillation amplitude is small, i.e., less than 10% of the equilibrium drop radius (Becker, Hiller, and Kowalewski, 1991). The liquid dynamic viscosity μ determines the damping rate and the drop surface tension σ determines the drop oscillation frequency (Lamb, 1881). The values are calculated as

$$\mu = \frac{\omega_{m,r} \rho a^2}{(m-1)(2m+1)}, \quad \sigma = \frac{3\pi \rho V f^2}{m(m-1)(m+2)}. \quad (3.94)$$

Prosperetti (1980a) pointed out that equation (3.94) is only valid for drops with small Ohnesorge number ($Oh < 0.1$).

3.4.2. Experimental measurement results

The results from the experiments with known liquid properties are studied in the current subsection. At least five oscillation experiments are performed with each of the glycerol solutions shown in table 3.2. Measurements are conducted at a room temperature of 24°C and atmospheric pressure. The characteristic equation (3.35) for the non-dimensional complex angular frequency $\Omega_2 = \omega_2/\omega_{2,0}$ is used to calculate ω_2 for the corresponding cases. The results of the characteristic equation are compared with the measured complex angular frequencies for the range of Ohnesorge numbers between

3.4. Experiments on Newtonian oscillating drops

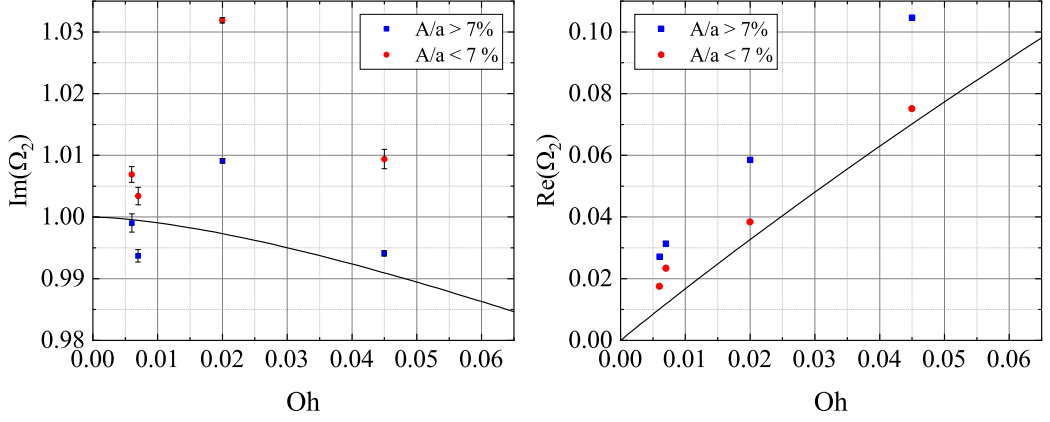


Figure 3.16.: Non-dimensional complex angular frequency Ω_2 presented as measured mean values (points) with its standard deviation (error bars) and calculated (solid line) values as a function of the Ohnesorge number. Imaginary part of Ω_2 is angular frequency and real is damping rate. Comparison of the results for the large-amplitude (LA) and small-amplitude (SA) oscillations. The threshold oscillation amplitude that separates the LA and the SA cases is set to 7% of the equilibrium radius.

Table 3.3.: Data for the 20 wt.% glycerol solution drop used for oscillation shown in figure 3.15. The equilibrium drop radius is 1.025 mm, the corresponding dimensional Rayleigh frequency is $\omega_{2,0} = 2\pi \cdot 115\text{Hz}$, and the Ohnesorge number is 0.006. The table originates from Zrnić and Brenn (2019).

	f [Hz]	$\omega_{2,r}$ [1/s]	$\text{Im}(\Omega_2)$ [—]	$\text{Re}(\Omega_2)$ [—]	A/a [%]	σ [N/m]	μ [mPa s]
Small-amplitude oscillations	113.8	7	1.02	0.01	6.9	0.072	1.52
Large-amplitude oscillations	113	15	1.013	0.022	11.6	0.071	3.33

3. Newtonian drop shape oscillations

0 and 0.07. Measured complex angular frequencies are shown as a mean value and standard deviation (error bars) based on at least five oscillation experiments of drops with the same glycerol mass fraction. Figure 3.16 shows the comparison and presents good agreement with the complex angular frequency extracted from the second part of the oscillation, i.e. at small-amplitude oscillations. Large-amplitude oscillations exhibit large deviations of the measured complex angular frequencies from the values calculated from equation (3.35). The right-hand figure 3.16 presents the damping rate, which exhibits large deviations from the linear case, while the left-hand figure 3.16 presents the non-dimensional oscillation frequency with smaller deviations from the linear case. For the latter, one makes no clear distinction between the small- and large-amplitude cases. Mainly it is due to uncertainties of the liquid properties entering the equation for the dimensional form of $\omega_{m,0}^2 = m(m-1)(m+2)\sigma/\rho a^3$ and due to the error of the measured oscillation frequency. The absolute uncertainty of the Rayleigh frequency $\omega_{m,0}$ is $\pm 8.5\%$. For the calculation, we use the uncertainties in table 3.2 for liquid properties and the uncertainty of the measured drop radius a that equals $\pm 4\%$. The right-hand figure 3.16 shows an increasing trend in the deviation of SA and LA cases of non-dimensional damping rates as the Ohnesorge number increases.

The threshold oscillation amplitude of $0.07a$ separating the LA and SA cases is set between the minimum oscillation amplitude extracted from the first part of it and the maximum amplitude extracted from the last part of the motion. The results presented in figure 3.16 are expected, since the characteristic equation (3.35) is a result of the linear theory.

The next results concern the oscillating drop method using equations (3.94) for the cases of small- and large-amplitude oscillations. The data are presented in table 3.3 for the damped drop oscillation shown in figure 3.15. As noted in the previous subsection, the values of the oscillation frequencies are similar for the small- and large-amplitude oscillations. As a consequence, the surface tension calculated by the second equation in (3.94) is nearly the same for both cases, and the values are close to the value σ^* from pendant drop tensiometry. About 4% is the absolute deviation between the values from the two methods. The value of absolute deviation is in the order of

3.4. Experiments on Newtonian oscillating drops

Table 3.4.: From equations (3.94), surface tension σ and dynamic viscosity μ of aqueous glycerol solution drops. Absolute deviations between values from the oscillating drop method with (LA) and (SA) oscillations and from table 3.2. The table originates from Zrnić and Brenn (2019).

Glycerol mass fraction w [wt.%]	SA oscillations		LA oscillations	
	Surface	Dynamic	Surface	Dynamic
	tension	viscosity	tension	viscosity
	[N/m] $\sigma \left(\frac{ \Delta\sigma }{\sigma^*} \right)$	[mPas] $\mu \left(\frac{ \Delta\mu }{\mu^*} \right)$	[N/m] $\left(\frac{ \Delta\sigma }{\sigma^*} \right)$	[mPas] $\mu \left(\frac{ \Delta\mu }{\mu^*} \right)$
20	0.069 (< 1%)	2.4 (48%)	0.068 (1.5%)	3.75 (132%)
30	0.068 (< 1%)	3.3 (48%)	0.066 (1.4%)	4.56 (105%)
50	0.068 (4.6%)	5.58 (6.8%)	0.067 (3.7%)	8.53 (53%)
65	0.064 (1.6%)	11.84 (< 1%)	0.062 (1.5%)	15.9 (35%)

the uncertainties of σ^* . By contrast, the damping rate for the LA oscillation is nearly double the value of SA, which doubles the value of the dynamic viscosity calculated by the first equation in (3.94) against the LA case. The difference between capillary viscometry and the values obtained by the oscillating drop method is around 6% for small-amplitude oscillations, and around 105% for large-amplitude oscillations. This result corresponds to the far stronger sensitivity of the damping rate against the deformation than seen for the oscillation frequency.

Table 3.4 presents the average values of surface tension and dynamic viscosity for the aqueous glycerol solutions measured by the oscillating drop method. For both small- and large-amplitude oscillations, the values of surface tension σ calculated with the second equation in (3.94) are in excellent agreement with the values σ^* from the pendant drop tensiometry. This is due to similar values of the oscillation frequencies for small- and large-amplitude oscillations. The maximum absolute deviation between the pendant drop tensiometry and the oscillating drop method is within 4.6%, which is around the order of the σ^* uncertainties. The values of the dynamic viscosity μ calculated using the first equation in (3.94), in contrast, show strong dependency on the oscillation amplitude. The maximum absolute

deviation between the oscillating drop method and capillary viscometry is around 48% in the case of small-amplitude oscillations and up to 132% in the case of large-amplitude oscillations. Comparing the calculated and measured values, the absolute deviation between the methods decreases with increasing glycerol mass fraction. These findings correspond to the theoretical results in figs. 3.10 and 3.11.

3.5. Conclusions

A theoretical analysis using the weakly nonlinear theory is conducted on axisymmetric shape oscillations of Newtonian liquid drops in a vacuum, and experimental measurements are performed on small- and large-amplitude shape oscillations of liquid drops. The goal was to reveal the detailed time behaviour of the nonlinear shape oscillations. In the theoretical work, the velocity and pressure fields in the drop, as well as the drop shape, are expanded in power series of a small deformation parameter η_0 , giving sets of equations of motion with their boundary and initial conditions for the different orders of approximation. The mode of initial drop deformation, the deformation parameter and the drop Ohnesorge number govern the problem of the oscillating drop. To the best of our knowledge, this is the first weakly nonlinear analysis of Newtonian drop shape oscillations (Zrnić, Berglez, and Brenn, 2022).

The initial deformation of the drop surface against the spherical state is governed by a Legendre polynomial of order $m = 2, 3$ or 4 , and the deformation parameter defines the oscillation amplitude. The truncated power series lead to volume deviations, which are calculated in non-dimensional form with respect to the correct value of $4\pi/3$. For the Ohnesorge number of 0.1 and deformation parameters $\eta_0 < 0.4$, the drop volume deviation is no larger than 1.6% for all the deformation modes $m = 2, 3$ and 4 . The drop volume deviates by no more than 2% for deformation parameters $\eta_0 < 0.4$ for drop Ohnesorge numbers between 0.1 and 1 .

The analysis yields the well-known first-order solution from Lamb (1881), including the characteristic equation for the complex angular frequency of the drop deduced by Chandrasekhar (1959) and Prosperetti (1977). The second-

and third-order approximations exhibit the nonlinear influences from the first order and from the first and second orders, respectively. The analysis yields known nonlinear effects in drop shape oscillations that are an asymmetry in the times the drop spends in different deformation states and a decrease of the oscillation frequency with increasing deformation. Another nonlinear phenomenon that was found for inviscid drops also (Zrnić and Brenn, 2021b) is quasi-periodicity of the shape oscillations. The work on inviscid drop oscillations is given in detail in the next chapter. Since Zrnić and Brenn (ibid.) established that the frequency decrease is a third-order effect for the fundamental initial deformation mode, the analysis for the Newtonian drop is taken to the third order. It also accounts for the quasi-periodic motion and for the frequency change for different Ohnesorge numbers and deformation parameters.

These effects are confirmed, correctly represented and verified with numerical results from relevant literature for the case $Oh = 0.1$. The method applied represents the quasi-periodic motion induced by nonlinearity for increasing deformation amplitude for a wide range of Ohnesorge numbers. The mode coupling is substantially affected by viscosity, as stated by Becker, Hiller, and Kowalewski (1994). The damping rate is also influenced by the nonlinear effects, where, in the large-amplitude regime, the damping rate decreases with the deformation when deduced from the north pole position of the drop in time. On the other hand, the damping rate increases with increasing deformation when deduced from the aspect ratio in time. The present study shows that mode coupling may lead to damped oscillatory drop behaviour despite the initiation by a deformation mode with a drop Ohnesorge number beyond cutoff. This case was shown for $m = 4$ and $Oh = 0.56$, which reveals damped oscillatory motion, while the linear theory predicts an aperiodic response. The oscillations are brought about by the lower-order modes excited by coupling with the initial mode of deformation.

The linear theory holds for initial oscillation amplitudes less than 10% of the equilibrium drop radius, according to Becker, Hiller, and Kowalewski (1991). The experimental results, however, showed that large deviations from the linear theory can occur for initial oscillation amplitudes $\geq 7\%$ of the equilibrium drop radius. From the experimental measurements of the

3. Newtonian drop shape oscillations

drop height in time we determine the damping rate from which, for large amplitudes, we obtain more pronounced deviations from the linear theory and the measured damping rates are higher than the values predicted by the linear theory. The oscillation frequency is practically unaffected by the deformation amplitude.

4. Inviscid drop shape oscillations

The present chapter outlines in detail nonlinear shape oscillations of inviscid drops. As in the previous chapter, the first two sections present the governing equations, which are derived by the weakly nonlinear method for the respective order of approximation. The results are presented in three subsections. The first subsection gives the validation of results found and investigates the volume for an inviscid liquid drop. The deformed surface shapes for three initial modes are shown in the second subsection. The third part investigates the time scales of the inviscid drop oscillations. It includes an analysis of nonlinear effects from the trace of the drop north pole in time and minimum drop surface area. It shows the relative frequency change against the linear cases and excess time spent in prolate state against oblate and the corresponding nonlinear effects for higher modes of initial drop deformation. Parts of the present analysis originate from Zrnić and Brenn (2021b).

4.1. Governing equations of the oscillating drop

The aim of the current study is to analyse inviscid liquid drop shape oscillations as sketched in figure 2.1. Most of the details on approach and assumptions of the weakly nonlinear method have already been described in chapter 2. To avoid repetition, only the differences between the current case and the previous chapters will be given here. The rest is assumed to be identical. Since the liquid material behavior is inviscid, the velocity field is irrotational and the Ohnesorge number equals zero. Due to the latter, the extra stress in the momentum balance is zero, and the continuity equation together with the components of the momentum equation in the radial (r)

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and polar angular (θ) directions are reduced to

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_r) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_\theta \sin \theta) = 0, \quad (4.1)$$

$$\frac{\partial u_r}{\partial t} + u_r \frac{\partial u_r}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_r}{\partial \theta} - \frac{u_\theta^2}{r} = -\frac{\partial p}{\partial r}, \quad (4.2)$$

$$\frac{\partial u_\theta}{\partial t} + u_r \frac{\partial u_\theta}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_\theta}{\partial \theta} + \frac{u_r u_\theta}{r} = -\frac{1}{r} \frac{\partial p}{\partial \theta}. \quad (4.3)$$

The set of equations is solved subject to boundary and initial conditions. The boundary and initial conditions are derived in the same manner as in chapter 2. The kinematic and zero-normal stress boundary conditions are given in equations (2.7) and (2.28), respectively, where for the latter the second term vanishes here since the Ohnesorge number equals zero.

In equations (4.1), (4.2) and (4.3), and in the boundary and initial conditions, the two velocity components and the pressure in the flow field, as well as the deformed interface shape, are expanded in power series of the small deformation parameter η_0 . We therefore formulate the expansions of the flow field properties for u_r , p and η as in the equations (2.12), (2.13) and (2.16). These series expansions converge only for small deformation parameter η_0 . The potential and the benefits of this approach are explained in chapters 2 and 3. The main difference in the approach compared to the case with viscous drops concerns the appearance of secular terms in the solutions for the surface deformation through the boundary conditions. Those terms arise from an integration with respect to time, and they are unphysical. To suppress them, the method of a strained time coordinate is applied (Yuen, 1968). The physical time t is related to the strained time τ as

$$\tau = \nu t. \quad (4.4)$$

The coefficient ν is expanded with respect to the deformation parameter η_0 as per

$$\nu = \nu_0 + \nu_1 \eta_0 + \nu_2 \eta_0^2 + \dots. \quad (4.5)$$

The method creates a new time coordinate, which contributes to a frequency decrease in the oscillatory motion of the drop, as will be seen later. For the fundamental initial deformation mode, the frequency decrease is visible

only when taking the approximation to the third order (Zrnić and Brenn, 2021b). The expansions given in (2.12), (2.13) and (2.16) are substituted into the governing equations (4.1) - (4.3). The same expansions together with the Taylor expansions for velocity (2.17) and pressure (2.18) are substituted into the boundary conditions (2.7) and (2.10). Equation (4.4) with the expansion (4.5) must be completely introduced into the governing equations, boundary and initial conditions to account for the time derivative. We collect all the terms with the deformation parameter η_0 to the first, second and third powers and obtain a set of first-, second- and third-order equations of motion, with the boundary and initial conditions, respectively. This procedure with its benefits is explained in chapter 2.

The initial conditions are derived from the volume conservation (3.85), which yields the initial non-dimensional drop shape in equation (2.20), and they state that the drop surface initially does not move in the radial direction. The difference between the viscous and inviscid cases is that the inviscid case does not exhibit a problem with the calculation of the complex angular frequency for the oscillation modes zero and one, since, according to Rayleigh (1879), these frequencies are zero.

4.1.1. First-order equations

The series expansions of the field variables are introduced into the equations of motion, and all the terms with the parameter η_0 to the first power are collected to obtain the first-order equations, with their boundary and initial conditions. The first-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_{r1}) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 1} \sin \theta) = 0, \quad (4.6)$$

$$\nu_0 \frac{\partial u_{r1}}{\partial \tau} = -\frac{\partial p_1}{\partial r}, \quad (4.7)$$

$$\nu_0 \frac{\partial u_{\theta 1}}{\partial \tau} = -\frac{1}{r} \frac{\partial p_1}{\partial \theta}. \quad (4.8)$$

4.1.2. First-order boundary and initial conditions

At $r = 1$, the kinematic and normal-stress boundary conditions of first order reads

$$u_{r1} = \nu_0 \frac{\partial \eta_1}{\partial \tau}, \quad (4.9)$$

$$-p_1 - \left(2\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right) = 0, \quad (4.10)$$

respectively. Furthermore, the initial conditions of first order are

$$\eta_1(\theta, 0) = P_m(\cos \theta) \quad \text{and} \quad \nu_0 \frac{\partial \eta_1}{\partial \tau}(\theta, 0) = 0. \quad (4.11)$$

The first initial condition determines the initial shape of the deformed drop, governed by a Legendre polynomial of order m . The corresponding shapes are shown in figure 1.1.

4.1.3. Second-order equations

To obtain the second-order equations with their boundary and initial conditions, all the terms from the expansion with the parameter η_0 to the second power are collected. The second-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 u_{r2} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 2} \sin \theta) = 0, \quad (4.12)$$

$$\nu_0 \frac{\partial u_{r2}}{\partial \tau} + \frac{\partial p_2}{\partial r} = \frac{u_{\theta 1}^2}{r} - \nu_1 \frac{\partial u_{r1}}{\partial \tau} - u_{r1} \frac{\partial u_{r1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{r1}}{\partial \theta}, \quad (4.13)$$

$$\nu_0 \frac{\partial u_{\theta 2}}{\partial \tau} + \frac{1}{r} \frac{\partial p_2}{\partial \theta} = -\nu_1 \frac{\partial u_{\theta 1}}{\partial \tau} - u_{r1} \frac{\partial u_{\theta 1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} - \frac{u_{r1} u_{\theta 1}}{r}. \quad (4.14)$$

4.1.4. Second-order boundary and initial conditions

The kinematic and zero normal-stress boundary conditions of second order to be satisfied at $r = 1$ are

$$u_{r2} - v_0 \frac{\partial \eta_2}{\partial \tau} = v_1 \frac{\partial \eta_1}{\partial \tau} + \frac{u_{\theta 1}}{r} \frac{\partial \eta_1}{\partial \theta} - \eta_1 \frac{\partial u_{r1}}{\partial r}, \quad (4.15)$$

$$-p_2 - \left(2\eta_2 + \frac{\partial \eta_2}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_2}{\partial \theta^2} \right) = \eta_1 \frac{\partial p_1}{\partial r} - 2\eta_1^2 - 2\eta_1 \frac{\partial \eta_1}{\partial \theta} \cot \theta - 2\eta_1 \frac{\partial^2 \eta_1}{\partial \theta^2}. \quad (4.16)$$

The initial conditions of second order are

$$\eta_2(\theta, 0) = -\frac{1}{2m+1} \quad \text{and} \quad v_0 \frac{\partial \eta_2}{\partial \tau}(\theta, 0) + v_1 \frac{\partial \eta_1}{\partial \tau}(\theta, 0) = 0. \quad (4.17)$$

For the deformed drop up to second order, the first initial condition ensures the volume conservation, while the second constrains the surface initially at rest. The sets of first- and second-order equations together with their boundary and initial conditions will reveal the nonlinear effect of unequal time spent in the prolate and oblate shapes as a function of the initial deformation amplitude. The frequency decrease only for initial deformation modes $m > 2$ is revealed. To find the frequency decrease for the two-lobed mode $m = 2$, the theory must therefore be taken to third order, as will be shown below.

4.1.5. Third-order equations

The third-order continuity and momentum equations read

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 u_{r3} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_{\theta 3} \sin \theta) = 0, \quad (4.18)$$

$$\begin{aligned} v_0 \frac{\partial u_{r3}}{\partial \tau} + \frac{\partial p_3}{\partial r} = & -v_1 \frac{\partial u_{r2}}{\partial \tau} - v_2 \frac{\partial u_{r1}}{\partial \tau} + 2 \frac{u_{\theta 1} u_{\theta 2}}{r} \\ & - u_{r1} \frac{\partial u_{r2}}{\partial r} - u_{r2} \frac{\partial u_{r1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{r2}}{\partial \theta} - \frac{u_{\theta 2}}{r} \frac{\partial u_{r1}}{\partial \theta}, \end{aligned} \quad (4.19)$$

$$\begin{aligned} v_0 \frac{\partial u_{\theta 3}}{\partial \tau} + \frac{1}{r} \frac{\partial p_3}{\partial \theta} = & -v_1 \frac{\partial u_{\theta 2}}{\partial \tau} - v_2 \frac{\partial u_{\theta 1}}{\partial \tau} - \frac{u_{r1} u_{\theta 2}}{r} - \frac{u_{r2} u_{\theta 1}}{r} \\ & - u_{r1} \frac{\partial u_{\theta 2}}{\partial r} - u_{r2} \frac{\partial u_{\theta 1}}{\partial r} - \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 2}}{\partial \theta} - \frac{u_{\theta 2}}{r} \frac{\partial u_{\theta 1}}{\partial \theta}. \end{aligned} \quad (4.20)$$

4.1.6. Third-order boundary and initial conditions

At $r = 1$, the third-order kinematic and zero normal-stress boundary conditions read

$$u_{r3} - \nu_0 \frac{\partial \eta_3}{\partial \tau} = \nu_1 \frac{\partial \eta_2}{\partial \tau} + \nu_2 \frac{\partial \eta_1}{\partial \tau} + \frac{u_{\theta 1}}{r} \frac{\partial \eta_2}{\partial \theta} + \frac{u_{\theta 2}}{r} \frac{\partial \eta_1}{\partial \theta} + \frac{\partial}{\partial r} \left(\frac{u_{\theta 1}}{r} \right) \frac{\partial \eta_1}{\partial \theta} \eta_1 - \frac{\partial u_{r2}}{\partial r} \eta_1 - \frac{\partial u_{r1}}{\partial r} \eta_2 - \frac{1}{2} \frac{\partial^2 u_{r1}}{\partial r^2} \eta_1^2, \quad (4.21)$$

$$-p_3 - \left(2\eta_3 + \frac{\partial \eta_3}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_3}{\partial \theta^2} \right) = \frac{\partial p_1}{\partial r} \eta_2 + \frac{\partial p_2}{\partial r} \eta_1 + \frac{1}{2} \frac{\partial^2 p_1}{\partial r^2} \eta_1^2 - \eta_1^3 - 2\eta_1 \left(\eta_2 + \frac{\partial \eta_2}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_2}{\partial \theta^2} \right) - 2\eta_2 \left(\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right) - \frac{1}{2} \left(\frac{\partial \eta_1}{\partial \theta} \right)^2 \left(\frac{\partial \eta_1}{\partial \theta} \cot \theta + 3 \frac{\partial^2 \eta_1}{\partial \theta^2} \right) + 3\eta_1^2 \left(\eta_1 + \frac{\partial \eta_1}{\partial \theta} \cot \theta + \frac{\partial^2 \eta_1}{\partial \theta^2} \right), \quad (4.22)$$

respectively. Furthermore, the third-order initial conditions are

$$\eta_3(\theta, 0) = -\frac{1}{3} \begin{pmatrix} m & m & m \\ 0 & 0 & 0 \end{pmatrix}^2 \quad \text{and} \quad \left(\nu_0 \frac{\partial \eta_3}{\partial \tau} + \nu_1 \frac{\partial \eta_2}{\partial \tau} + \nu_2 \frac{\partial \eta_1}{\partial \tau} \right) (\theta, 0) = 0, \quad (4.23)$$

where the first initial condition is described in terms of a $3-j$ symbol, which is explained in appendix B. The weakly nonlinear dependency of the angular oscillation frequency on the amplitude of the initial drop deformation will be revealed by solving the sets of equations up to third order together with their boundary and initial conditions. The effect of the frequency change becomes visible due to the strained time introduced for suppressing secular terms in solutions.

4.2. Solutions of the governing equations

The present section describes the solutions of the governing equations for the respective order of approximation shown above and is divided into three subsections. For the sake of clarity, parts of the derivations are shown in appendix C. References to the appendix will be made during the derivations in

the text below. First-order solutions, which are shown in the first subsection, are the well-known linear solutions, while second- and third-order solutions, which are presented in the second and third subsections, respectively, account for nonlinear phenomena. The method described in detail below can also be found in Zrnić and Brenn (2021b). It follows the same pattern as for the Newtonian drop shape oscillations detailed in the previous chapter, with the introduced strained time coordinate to suppress secular terms in the higher-order solutions. The strained time coordinate plays an essential role and helps to avoid unphysical behaviour.

4.2.1. First-order solutions

The linear problem is described with the first-order equations. The section recovers the well-known solutions of Rayleigh (1879) and Lamb (1881) for the inviscid drop in a vacuum. The Stokesian stream function is applied for determining the first-order velocity field, since we are investigating two-dimensional flow fields. The coefficient ν_0 is set to unity in the strained time definition. With the help of the stream function $\psi(r, \theta, \tau)$, the two velocity components u_{r1} and $u_{\theta 1}$ are defined as in Bird, Stewart, and Lightfoot (1962)

$$u_{r1} = -\frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta} \quad \text{and} \quad u_{\theta 1} = \frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r}, \quad (4.24)$$

where the continuity equation is satisfied using those two first-order velocity components.

The Legendre polynomial of the initial deformation governs the first-order drop surface deformation, and the solution is therefore sought in the form

$$\eta_1 = \hat{\eta}_1 P_m(\cos \theta) e^{-\alpha_m \tau}. \quad (4.25)$$

The first-order initial deformation amplitude is $\hat{\eta}_1$, and the first-order angular frequency is α_m for the deformation mode m .

The partial differential equation for the stream function is formed taking the curl of the vectorial first-order momentum equation with the components (4.7) and (4.8), which yields

$$\frac{\partial^2 \psi}{\partial r^2} + \frac{\sin \theta}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial \psi}{\partial \theta} \right) = 0, \quad (4.26)$$

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when substituting the stream function in the velocity components. The regular solution at $r = 0$ reads (Brenn, 2017)

$$\psi = C_{1m} r^{m+1} \sin^2 \theta P'_m(\cos \theta) e^{-\alpha_m \tau}, \quad (4.27)$$

where the prime describes the derivative of the Legendre polynomial P_m with respect to its argument. The first-order radial and angular velocity components follow as

$$u_{r1} = -C_{1m} r^{m-1} m(m+1) P_m(\cos \theta) e^{-\alpha_m \tau}, \quad (4.28)$$

and

$$u_{\theta 1} = -C_{1m} r^{m-1} (m+1) P_m^1(\cos \theta) e^{-\alpha_m \tau}, \quad (4.29)$$

respectively. The function P_m^1 is a Legendre function of degree m and order 1. Through the first-order kinematic boundary condition, the integration constant C_{1m} is determined, and it reads

$$C_{1m} = \frac{\alpha_m \hat{\eta}_1}{m(m+1)}. \quad (4.30)$$

Using the integration of one component of the momentum equation, the first-order pressure field is formed as

$$p_1 = -C_{1m} \alpha_m (m+1) r^m P_m(\cos \theta) e^{-\alpha_m \tau}. \quad (4.31)$$

Obtained in Rayleigh (1879), the characteristic equation of the drop follows from the first-order zero normal stress boundary condition (4.10), which determines the complex angular frequency α_m . Its complex conjugate solutions are

$$\begin{aligned} \alpha_m^{(p)} &= i \sqrt{m(m-1)(m+2)} =: i \alpha_{m,0}, \\ \alpha_m^{(n)} &= -i \sqrt{m(m-1)(m+2)} =: -i \alpha_{m,0}, \end{aligned} \quad (4.32)$$

where the positive and negative solutions are represented with the superscripts (p) and (n) respectively. The characteristic equation gives a pair of solutions, which have to be represented in the first-order solutions. Therefore, we formulate the first-order deformation of the drop surface as

$$\eta_1(\theta, \tau) = \left(\hat{\eta}_1^{(p)} e^{-\alpha_m^{(p)} \tau} + \hat{\eta}_1^{(n)} e^{-\alpha_m^{(n)} \tau} \right) P_m(\cos \theta). \quad (4.33)$$

The requirement of the first-order initial conditions (4.11) states that the initial drop surface shape is governed by the function $P_m(\cos \theta)$, and that the surface is at rest. These conditions reveal the deformation amplitudes

$$\hat{\eta}_1^{(p)} = -\frac{\alpha_m^{(n)}}{\alpha_m^{(p)} - \alpha_m^{(n)}} = \frac{1}{2} \quad \text{and} \quad \hat{\eta}_1^{(n)} = \frac{\alpha_m^{(p)}}{\alpha_m^{(p)} - \alpha_m^{(n)}} = \frac{1}{2}. \quad (4.34)$$

The first-order radial and polar velocity components are represented as

$$u_{r1}(r, \theta, \tau) = -\left(C_{1m}^{(p)} e^{-\alpha_m^{(p)} \tau} + C_{1m}^{(n)} e^{-\alpha_m^{(n)} \tau}\right) r^{m-1} m(m+1) P_m(\cos \theta), \quad (4.35)$$

and

$$u_{\theta 1}(r, \theta, \tau) = -\left(C_{1m}^{(p)} e^{-\alpha_m^{(p)} \tau} + C_{1m}^{(n)} e^{-\alpha_m^{(n)} \tau}\right) r^{m-1} (m+1) P_m^1(\cos \theta), \quad (4.36)$$

respectively. The first-order pressure field reads

$$p_1(r, \theta, \tau) = -\left(C_{1m}^{(p)} \alpha_m^{(p)} e^{-\alpha_m^{(p)} \tau} + C_{1m}^{(n)} \alpha_m^{(n)} e^{-\alpha_m^{(n)} \tau}\right) (m+1) r^m P_m(\cos \theta). \quad (4.37)$$

The first-order drop surface shape, which accounts for the complex conjugate frequencies in (4.32), forms velocity components and pressure field as

$$u_{r1}(r, \theta, \tau) = -\alpha_{m,0} r^{m-1} P_m(\cos \theta) \sin(\alpha_{m,0} \tau), \quad (4.38)$$

$$u_{\theta 1}(r, \theta, \tau) = -\frac{\alpha_{m,0}}{m} r^{m-1} P_m^1(\cos \theta) \sin(\alpha_{m,0} \tau), \quad (4.39)$$

$$p_1(r, \theta, \tau) = (m-1)(m+2) r^m P_m(\cos \theta) \cos(\alpha_{m,0} \tau), \quad (4.40)$$

$$\eta_1(\theta, \tau) = P_m(\cos \theta) \cos(\alpha_{m,0} \tau). \quad (4.41)$$

4.2.2. Second-order solutions

The second-order solutions are developed from pressure, where the general solution for the second-order pressure is sought as a sum

$$p_2(r, \theta, \tau) = p_{21}(r, \theta, \tau) + p_{22}(r, \theta, \tau) \quad (4.42)$$

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of two contributions. The subscript "21" indicates the solution of the system of second-order equations including the products of first-order terms, while the subscript "22" is for the solutions of the homogeneous system. Since in the second-order approximation the coefficient ν_1 is unknown, further steps will be taken to determine it.

Firstly, solutions with subscripts "21" are deduced, forming the divergence of the vectorial second-order momentum equation with the components (4.13) and (4.14) to eliminate the second-order velocity. The terms containing ν_1 disappear due to the use of the continuity equation (4.12). The corresponding procedure yields the differential equation for the second-order pressure p_{21}

$$\begin{aligned} \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial p_{21}}{\partial r} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\sin \theta}{r} \frac{\partial p_{21}}{\partial \theta} \right) = \\ - \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \left(u_{r1} \frac{\partial u_{r1}}{\partial r} + \frac{u_{\theta 1}}{r} \frac{\partial u_{r1}}{\partial \theta} - \frac{u_{\theta 1}^2}{r} \right) \right] \\ - \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left[\left(u_{r1} \frac{\partial u_{\theta 1}}{\partial r} + \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} + \frac{u_{r1} u_{\theta 1}}{r} \right) \sin \theta \right], \end{aligned} \quad (4.43)$$

which is re-written, taking into account the curl-free first-order velocity field and using the Lamé identity, in the form

$$\Delta \left[p_{21} + \bar{v}_1^2 / 2 \right] = 0. \quad (4.44)$$

This statement describes the fact that the modified pressure $\mathcal{P}_{21} = p_{21} + \bar{v}_1^2 / 2$ is potential. The time dependency of pressure p_{21} is determined taking into account the squared first-order velocity vector.

The solution of the Laplace equation for the modified pressure $\mathcal{P}_{21}$ that accounts for all the time dependencies arising from the square of the first-order velocity vector may be written as the contribution to the second-order pressure p_{21} as

$$\begin{aligned} p_{21}(r, \theta, \tau) = - \sum_{l=0}^L \left(C_{21l}^{(p)} e^{-2\alpha_m^{(p)} \tau} + C_{21l}^{(n)} e^{-2\alpha_m^{(n)} \tau} \right. \\ \left. + C_{21l}^{(pn)} e^{-(\alpha_m^{(p)} + \alpha_m^{(n)}) \tau} \right) r^l P_l(\cos \theta) - \frac{1}{2} (u_{r1}^2 + u_{\theta 1}^2). \end{aligned} \quad (4.45)$$

The coefficients $C_{21l}^{(pn)}$ which relate to the time-independent terms in p_{21} are set to zero for all l . After the calculation of the radial velocity in the next step, this treatment of these coefficients is justified. All the summations which emerge from the solutions henceforth extend to an upper summation index denoted by an upper-case letter. Knowing that the first-order velocity field is irrotational, the radial momentum equation (4.13) is re-written with the modified pressure for the contribution "21". One obtains

$$\frac{\partial u_{r21}}{\partial \tau} = -\frac{\partial \mathcal{P}_{21}}{\partial r} - \nu_1 \frac{\partial u_{r1}}{\partial \tau}. \quad (4.46)$$

The radial velocity component u_{r21} from this equation is calculated by integration with respect to time, revealing

$$u_{r21}(r, \theta, \tau) = -\sum_{l=0}^L \left(\frac{C_{21l}^{(p)}}{2\alpha_m^{(p)}} e^{-2\alpha_m^{(p)}\tau} + \frac{C_{21l}^{(n)}}{2\alpha_m^{(n)}} e^{-2\alpha_m^{(n)}\tau} \right) l r^{l-1} P_l(\cos \theta) - \nu_1 u_{r1}. \quad (4.47)$$

In the pressure p_{21} , non-zero coefficients $C_{21l}^{(pn)}$ would have produced linearly time-dependent secular terms, which would grow in time. One can prevent this growth by setting the coefficients $C_{21l}^{(pn)}$ in (4.45) for all l to zero. The velocity component $u_{\theta 21}$ is then obtained from the continuity equation and reads

$$u_{\theta 21}(r, \theta, \tau) = -\sum_{l=0}^L \left(\frac{C_{21l}^{(p)}}{2\alpha_m^{(p)}} e^{-2\alpha_m^{(p)}\tau} + \frac{C_{21l}^{(n)}}{2\alpha_m^{(n)}} e^{-2\alpha_m^{(n)}\tau} \right) r^{l-1} P_l^1(\cos \theta) - \nu_1 u_{\theta 1}.$$

Studying the work by Tsamopoulos and Brown (1983, 1984) one can write the surface deformation η_{21} in a general form as

$$\eta_{21}(\theta, \tau) = \sum_{l=0}^L \delta_{2l}(\tau) P_l(\cos \theta). \quad (4.48)$$

The unknown functions $\delta_{2l}(\tau)$ govern the time dependency, while all possible Legendre polynomials control the deformed shape. The explanation for the latter arises from the fact that the zero normal-stress boundary condition couples the pressure and surface deformation. Further, the boundary

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conditions (4.15) and (4.16) determine the functions $\delta_{2l}(\tau)$ for every l . In order to do this, u_{r21} and η_{21} from (4.47) and (4.48) are introduced into the kinematic boundary condition (4.15). The derivative of the latter is taken with respect to the strained time τ . Additionally, the pressure p_{21} and surface deformation η_{21} from (4.45) and (4.48), respectively, are substituted into the zero normal-stress boundary condition (4.16). These boundary conditions are evaluated at $r = 1$. Finally, one proceeds by multiplying the two equations with a Legendre polynomial P_j and integrating the results between the south and north poles of the drop. Multiplying the equation resulting from the zero-normal stress boundary condition by the natural number j , and subtracting the two resulting equations, eliminates the coefficients C_{21j} for all time dependencies. The result is the ordinary differential equation for $\delta_{2j}(\tau)$ (Tsamopoulos and Brown, 1983, 1984)

$$\begin{aligned} \ddot{\delta}_{2j}(\tau) + j(j-1)(j+2)\delta_{2j}(\tau) = & \left[- \int_{-1}^1 \frac{\partial}{\partial \tau} \left(\frac{u_{\theta 1}}{r} \frac{\partial \eta_1}{\partial \theta} - \eta_1 \frac{\partial u_{r1}}{\partial r} \right) \right]_{r=1} P_j dx \\ & - \nu_1 \left(\alpha_m^{(p)2} e^{-\alpha_m^{(p)} \tau} + \alpha_m^{(n)2} e^{-\alpha_m^{(n)} \tau} \right) \int_{-1}^1 P_m P_j dx \\ & + \int_{-1}^1 \left(Rz_{21} - \frac{1}{2} (u_{r1}^2 + u_{\theta 1}^2) \right) \left[j P_j dx \right] \frac{2j+1}{2}, \end{aligned} \quad (4.49)$$

where Rz_{21} is the right-hand side of the zero normal-stress boundary condition (4.16). Only the particular solutions of the differential equation (4.49) are of interest, because the homogeneous solutions will be accounted for by the second-order contribution "22". The particular solutions need to be found for every j between zero and L . Introducing back all particular solutions $\delta_{2j}(\tau)$ of (4.49) into (4.48), the final form is written as

$$\begin{aligned} \eta_{21}(\theta, \tau) = & \sum_{j=0}^L \left(H_{21j}^{(p)} e^{-2\alpha_m^{(p)} \tau} + H_{21j}^{(n)} e^{-2\alpha_m^{(n)} \tau} + H_{21j}^{(pn)} e^{-(\alpha_m^{(p)} + \alpha_m^{(n)}) \tau} \right) P_j(\cos \theta) \\ & + \frac{\nu_1}{4} \left[\left(1 + 2\alpha_m^{(p)} \tau \right) e^{-\alpha_m^{(p)} \tau} + \left(1 + 2\alpha_m^{(n)} \tau \right) e^{-\alpha_m^{(n)} \tau} \right] P_m, \end{aligned} \quad (4.50)$$

where the coefficients $H_{21j}^{(p)}$, $H_{21j}^{(n)}$ and $H_{21j}^{(pn)}$ are known. The second part of the solution (4.50), with v_1 in front, contains secular terms that grow in time and are unphysical (Zrnić and Brenn, 2021b). These terms must be suppressed by setting $v_1 = 0$. Using one of the boundary conditions (4.15) or (4.16), and with the known surface deformation η_{21} , the coefficients C_{21l} in u_{r21} , $u_{\theta 21}$ and p_{21} are readily determined for every time dependency and for all l between zero and L . The details of the calculation of C_{21l} are presented in section A of appendix C of the thesis. Now, the contributions "21" to the second-order solutions can be written in the form

$$u_{r21}(r, \theta, \tau) = \sum_{l=0}^L \frac{C_{21l}}{\alpha_{m,0}} \sin(2\alpha_{m,0}\tau) l r^{l-1} P_l(\cos \theta), \quad (4.51)$$

$$u_{\theta 21}(r, \theta, \tau) = \sum_{l=0}^L \frac{C_{21l}}{\alpha_{m,0}} \sin(2\alpha_{m,0}\tau) r^{l-1} P_l^1(\cos \theta), \quad (4.52)$$

$$p_{21}(r, \theta, \tau) = -2 \sum_{l=0}^L C_{21l} \cos(2\alpha_{m,0}\tau) r^l P_l(\cos \theta) - \frac{1}{2} (u_{r1}^2 + u_{\theta 1}^2), \quad (4.53)$$

$$\eta_{21}(\theta, \tau) = \sum_{l=0}^L \left(2H_{21l} \cos(2\alpha_{m,0}\tau) + H_{21l}^{(pn)} \right) P_l(\cos \theta). \quad (4.54)$$

The homogeneous forms of equations (4.12)-(4.14) determine the second contributions to the second-order solutions, with subscript "22". This set of equations has the same structure as at first order, where the complex angular frequency α_m is replaced by α_{2k} (with the number "2" in the subscript indicating the second-order solution, and k a deformation mode number) and the deformation amplitude of the drop surface $\hat{\eta}_1$ by $\hat{\eta}_{22k}$. Both replaced variables appear under a sum with the summation index k , concluding that all possible deformation modes k affect the results.

The contributions "22" to the second-order velocity, pressure and deformation are obtained as linear combinations of eigen-solutions of the underlying differential equations with the summation index k . Their forms are similar

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to the first-order solutions and read

$$u_{r22}(r, \theta, \tau) = - \sum_{k=0}^K \left(C_{22k}^{(p)} e^{-\alpha_{2k}^{(p)} \tau} + C_{22k}^{(n)} e^{-\alpha_{2k}^{(n)} \tau} \right) \times k(k+1) r^{k-1} P_k(\cos \theta) , \quad (4.55)$$

$$u_{\theta 22}(r, \theta, \tau) = - \sum_{k=0}^K \left(C_{22k}^{(p)} e^{-\alpha_{2k}^{(p)} \tau} + C_{22k}^{(n)} e^{-\alpha_{2k}^{(n)} \tau} \right) \times (k+1) r^{k-1} P_k^1(\cos \theta) , \quad (4.56)$$

$$p_{22}(r, \theta, \tau) = - \sum_{k=0}^K \left(\alpha_{2k}^{(p)} C_{22k}^{(p)} e^{-\alpha_{2k}^{(p)} \tau} + \alpha_{2k}^{(n)} C_{22k}^{(n)} e^{-\alpha_{2k}^{(n)} \tau} \right) \times (k+1) r^k P_k(\cos \theta) , \quad (4.57)$$

$$\eta_{22}(\theta, \tau) = \sum_{k=0}^K \left(\hat{\eta}_{22k}^{(p)} e^{-\alpha_{2k}^{(p)} \tau} + \hat{\eta}_{22k}^{(n)} e^{-\alpha_{2k}^{(n)} \tau} \right) P_k(\cos \theta) . \quad (4.58)$$

The solutions of the homogeneous zero normal-stress boundary condition are given as the two new complex conjugate angular frequencies α_{2k} for all summation indices k . They take the forms obtained in Rayleigh (1879)

$$\alpha_{2k}^{(p)} = i \sqrt{k(k-1)(k+2)} =: i \alpha_{2k,0} \quad \alpha_{2k}^{(n)} = -i \sqrt{k(k-1)(k+2)} =: -i \alpha_{2k,0} . \quad (4.59)$$

Due to the long expressions, the coefficients C_{22k} and the amplitudes $\hat{\eta}_{22k}$ for both time dependencies are presented in section A of appendix C.

The simplified forms of the contributions "22" to the second-order solutions are

$$u_{r22}(r, \theta, \tau) = -2 \sum_{k=0}^K \hat{\eta}_{22k} \alpha_{2k,0} r^{k-1} P_k(\cos \theta) \sin(\alpha_{2k,0} \tau) , \quad (4.60)$$

$$u_{\theta 22}(r, \theta, \tau) = -2 \sum_{k=1}^K \frac{\hat{\eta}_{22k} \alpha_{2k,0}}{k} r^{k-1} P_k^1(\cos \theta) \sin(\alpha_{2k,0} \tau) , \quad (4.61)$$

$$p_{22}(r, \theta, \tau) = 2 \sum_{k=0}^K \hat{\eta}_{22k} (k-1)(k+2) r^k P_k(\cos \theta) \cos(\alpha_{2k,0} \tau) , \quad (4.62)$$

$$\eta_{22}(\theta, \tau) = 2 \sum_{k=0}^K \hat{\eta}_{22k} P_k(\cos \theta) \cos(\alpha_{2k,0} \tau) . \quad (4.63)$$

4.2.3. Third-order solutions

The development of the third-order solutions is also started from pressure. The general solution for the third-order pressure is sought as a sum

$$p_3(r, \theta, \tau) = p_{31}(r, \theta, \tau) + p_{32}(r, \theta, \tau) \quad (4.64)$$

of two contributions. The subscript "31" indicates the solution of the third-order equations system including the products of first- and second-order terms, while the subscript "32" symbolizes the solutions of the homogeneous system of the governing equations with its boundary and initial conditions. As in the second-order approximation, the coefficient ν_2 in the strained time coordinate is unknown and needs to be determined.

The solutions with the subscript "31" are determined following the same procedure as in the second-order approximation. The divergence of the vectorial third-order momentum equation with the components (4.19) and (4.20) is taken, and the continuity equation (4.18) for elimination of the third-order velocities used. This yields the partial differential equation for the third-order pressure p_{31}

$$\begin{aligned} & \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial p_{31}}{\partial r} \right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left(\frac{\sin \theta}{r} \frac{\partial p_{31}}{\partial \theta} \right) = \\ & - \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \left(u_{r1} \frac{\partial u_{r2}}{\partial r} + u_{r2} \frac{\partial u_{r1}}{\partial r} + \frac{u_{\theta 1}}{r} \frac{\partial u_{r2}}{\partial \theta} + \frac{u_{\theta 2}}{r} \frac{\partial u_{r1}}{\partial \theta} - 2 \frac{u_{\theta 1} u_{\theta 2}}{r} \right) \right] \\ & - \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left[\left(u_{r1} \frac{\partial u_{\theta 2}}{\partial r} + u_{r2} \frac{\partial u_{\theta 1}}{\partial r} + \frac{u_{\theta 2}}{r} \frac{\partial u_{\theta 1}}{\partial \theta} + \frac{u_{\theta 1}}{r} \frac{\partial u_{\theta 2}}{\partial \theta} \right. \right. \\ & \left. \left. + \frac{u_{r2} u_{\theta 1}}{r} + \frac{u_{r1} u_{\theta 2}}{r} \right) \sin \theta \right], \end{aligned} \quad (4.65)$$

which we write in a compact form as

$$\Delta p_{31} = -\nabla \cdot \left[\left(\vec{u}_1 \cdot \vec{\nabla} \right) \vec{u}_2 + \left(\vec{u}_2 \cdot \vec{\nabla} \right) \vec{u}_1 \right]. \quad (4.66)$$

The equation is re-written using the dot product rule yielding

$$\Delta \mathcal{P}_{31} = \nabla \cdot \left[\vec{u}_1 \times \left(\vec{\nabla} \times \vec{u}_2 \right) + \vec{u}_2 \times \left(\vec{\nabla} \times \vec{u}_1 \right) \right]. \quad (4.67)$$

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The modified pressure is defined as $\mathcal{P}_{31} = p_{31} + \vec{u}_1 \cdot \vec{u}_2$. The right-hand side is zero, since the velocity fields affected are irrotational. The time dependencies of the third-order pressure p_{31} are determined following the same procedure as for the second-order modified pressure. Thus, all the time dependencies arising from the inner product of the first- and second-order velocity fields are taken into account and are written as

$$\begin{aligned}
 p_{31}(r, \theta, \tau) = & - \sum_{h=0}^H r^h P_h(x) \left(C_{31h}^{(p)} e^{-3\alpha_m^{(p)} \tau} + C_{31h}^{(n)} e^{-3\alpha_m^{(n)} \tau} \right. \\
 & + C_{31h}^{(ppn)} e^{-\left(2\alpha_m^{(p)} + \alpha_m^{(n)}\right) \tau} + C_{31h}^{(pnn)} e^{-\left(\alpha_m^{(p)} + 2\alpha_m^{(n)}\right) \tau} \\
 & + \sum_{k=0}^K C_{31hk}^{(pp)} e^{-\left(\alpha_m^{(p)} + \alpha_{2k}^{(p)}\right) \tau} + \sum_{k=0}^K C_{31hk}^{(nn)} e^{-\left(\alpha_m^{(n)} + \alpha_{2k}^{(n)}\right) \tau} \\
 & \left. + \sum_{k=0}^K C_{31hk}^{(pn)} e^{-\left(\alpha_m^{(p)} + \alpha_{2k}^{(n)}\right) \tau} + \sum_{k=0}^K C_{31hk}^{(np)} e^{-\left(\alpha_m^{(n)} + \alpha_{2k}^{(p)}\right) \tau} \right) - \vec{u}_1 \cdot \vec{u}_2.
 \end{aligned} \tag{4.68}$$

Following the same reasoning as in the second-order approximation, the coefficients $C_{31hm}^{(pn)}$ and $C_{31hm}^{(np)}$ relating to time-independent terms in p_{31} are set to zero for all h . Using the same procedure as in the previous approximation order, the contributions "31" to the velocity field are determined. When the radial momentum equation (4.19) is re-written and the definition of the third-order modified pressure $\mathcal{P}_{31}$ and the irrotational first- and second-order velocity fields are used, the following is obtained

$$\frac{\partial u_{r31}}{\partial \tau} = - \frac{\partial \mathcal{P}_{31}}{\partial r} - \nu_2 \frac{\partial u_{r1}}{\partial \tau}. \tag{4.69}$$

The radial velocity component u_{31} for every time dependency is easily obtained by integration of this momentum equation with respect to time,

since the modified pressure $\mathcal{P}_{31}$ is known. It yields

$$\begin{aligned}
 u_{r31}(r, \theta, \tau) = & -v_2 u_{r1} - \sum_{h=0}^H h r^{h-1} P_h(\cos \theta) \left(\frac{C_{31h}^{(p)}}{3\alpha_m^{(p)}} e^{-3\alpha_m^{(p)} \tau} + \frac{C_{31h}^{(n)}}{3\alpha_m^{(n)}} e^{-3\alpha_m^{(n)} \tau} \right. \\
 & + \frac{C_{31h}^{(ppn)}}{2\alpha_m^{(p)} + \alpha_m^{(n)}} e^{-(2\alpha_m^{(p)} + \alpha_m^{(n)}) \tau} + \frac{C_{31h}^{(pnn)}}{\alpha_m^{(p)} + 2\alpha_m^{(n)}} e^{-(\alpha_m^{(p)} + 2\alpha_m^{(n)}) \tau} \\
 & + \sum_{k=0}^K \frac{C_{31hk}^{(pp)}}{\alpha_m^{(p)} + \alpha_{2k}^{(p)}} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(p)}) \tau} + \sum_{k=0}^K \frac{C_{31hk}^{(nn)}}{\alpha_m^{(n)} + \alpha_{2k}^{(n)}} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(n)}) \tau} \\
 & \left. + \sum_{k=0 \setminus m}^K \frac{C_{31hk}^{(pn)}}{\alpha_m^{(p)} + \alpha_{2k}^{(n)}} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(n)}) \tau} + \sum_{k=0 \setminus m}^K \frac{C_{31hk}^{(np)}}{\alpha_m^{(n)} + \alpha_{2k}^{(p)}} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(p)}) \tau} \right). \quad (4.70)
 \end{aligned}$$

As seen at second order, the non-zero coefficients $C_{31hm}^{(pn)}$ and $C_{31hm}^{(np)}$ in the pressure p_{31} would have produced linearly time-dependent secular terms, which would increase in time. Since they are unphysical (Zrnić and Brenn, 2021b), they are suppressed with the same treatment as in the previous section. The continuity equation (4.18) determines the polar velocity component, which reads

$$\begin{aligned}
 u_{\theta 31}(r, \theta, \tau) = & -v_2 u_{\theta 1} - \sum_{h=0}^H r^{h-1} P_h^1(\cos \theta) \left(\frac{C_{31h}^{(p)}}{3\alpha_m^{(p)}} e^{-3\alpha_m^{(p)} \tau} + \frac{C_{31h}^{(n)}}{3\alpha_m^{(n)}} e^{-3\alpha_m^{(n)} \tau} \right. \\
 & + \frac{C_{31h}^{(ppn)}}{2\alpha_m^{(p)} + \alpha_m^{(n)}} e^{-(2\alpha_m^{(p)} + \alpha_m^{(n)}) \tau} + \frac{C_{31h}^{(pnn)}}{\alpha_m^{(p)} + 2\alpha_m^{(n)}} e^{-(\alpha_m^{(p)} + 2\alpha_m^{(n)}) \tau} \\
 & + \sum_{k=0}^K \frac{C_{31hk}^{(pp)}}{\alpha_m^{(p)} + \alpha_{2k}^{(p)}} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(p)}) \tau} + \sum_{k=0}^K \frac{C_{31hk}^{(nn)}}{\alpha_m^{(n)} + \alpha_{2k}^{(n)}} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(n)}) \tau} \\
 & \left. + \sum_{k=0 \setminus m}^K \frac{C_{31hk}^{(pn)}}{\alpha_m^{(p)} + \alpha_{2k}^{(n)}} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(n)}) \tau} + \sum_{k=0 \setminus m}^K \frac{C_{31hk}^{(np)}}{\alpha_m^{(n)} + \alpha_{2k}^{(p)}} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(p)}) \tau} \right). \quad (4.71)
 \end{aligned}$$

For formulating the surface deformation η_{31} , we proceed as in the second-order approximation. Steered by the work by Tsamopoulos and Brown (1983,

1984), the surface deformation is written in a general form as

$$\eta_{31}(\theta, \tau) = \sum_{h=0}^H \delta_{3h}(\tau) P_h(\cos \theta), \quad (4.72)$$

where the unknown functions $\delta_{3h}(\tau)$ govern the time dependency, and all possible Legendre polynomials the deformed shape.

The functions $\delta_{3h}(\tau)$ are determined by deriving the governing ordinary differential equation, following the same approach as in the second-order approximation. The method eliminates the coefficients C_{31j} and C_{31jk} for all time dependencies from the kinematic and zero normal-stress boundary conditions at third order, (4.21) and (4.22), and yields the ordinary differential equation (Tsamopoulos and Brown, 1983, 1984)

$$\begin{aligned} \ddot{\delta}_{3j}(\tau) + j(j-1)(j+2)\delta_{3j}(\tau) = & \frac{2j+1}{2} \left[- \int_{-1}^1 \frac{\partial Rk_{31}}{\partial \tau} \bigg|_{r=1} P_j dx \right. \\ & - \frac{\nu_2}{2} \left(\alpha_m^{(p)^2} e^{-\alpha_m^{(p)} \tau} + \alpha_m^{(n)^2} e^{-\alpha_m^{(n)} \tau} \right) \int_{-1}^1 P_m P_j dx \\ & \left. + \int_{-1}^1 (Rz_{31} - \vec{u}_1 \cdot \vec{u}_2) \bigg|_{r=1} j P_j dx \right], \end{aligned} \quad (4.73)$$

for $\delta_{3j}(\tau)$. The right-hand sides of the two boundary conditions (4.21) and (4.22) are Rk_{31} and Rz_{31} , respectively. Since these right-hand sides consist of many terms, a compact formulation of the equation is kept. The symbolic derivations and calculations were performed with the software MATHEMATICA. Since these general solution of the homogeneous equation is accounted for by the contribution "32", which solves the homogeneous system of equations, there is only interest in the particular solutions of equation (4.73). The solutions need to be found for every j between zero and H . All the particular solutions $\delta_{3j}(\tau)$ of the differential equation (4.73) are re-introduced into

m	Coefficients in η_{31}
2	$H_{314}^{(p)}, H_{314}^{(n)}, H_{312}^{(ppn)}, H_{312}^{(pnn)}, H_{3120}^{(pp)}, H_{3120}^{(nn)}, H_{3120}^{(pn)}, H_{3120}^{(np)}$
3	$H_{313}^{(ppn)}, H_{313}^{(pnn)}, H_{3130}^{(pp)}, H_{3130}^{(nn)}, H_{3130}^{(pn)}, H_{3130}^{(np)}$
4	$H_{314}^{(ppn)}, H_{314}^{(pnn)}, H_{3140}^{(pp)}, H_{3140}^{(nn)}, H_{3140}^{(pn)}, H_{3140}^{(np)}$

Table 4.1.: The coefficients H_{31j} and H_{31jk} in the surface deformation η_{31} for $m = 2, 3$ and 4 set to zero, since they relate to secular solutions.

(4.72), and therefore the final form is

$$\begin{aligned}
 \eta_{31}(\theta, \tau) = & \sum_{j=0}^H \left(H_{31j}^{(p)} e^{-3\alpha_m^{(p)} \tau} + H_{31j}^{(n)} e^{-3\alpha_m^{(n)} \tau} \right. \\
 & + H_{31j}^{(ppn)} e^{-(2\alpha_m^{(p)} + \alpha_m^{(n)}) \tau} + H_{31j}^{(pnn)} e^{-(\alpha_m^{(p)} + 2\alpha_m^{(n)}) \tau} \\
 & + \sum_{k=0}^K H_{31jk}^{(pp)} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(p)}) \tau} + \sum_{k=0}^K H_{31jk}^{(nn)} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(n)}) \tau} \\
 & \left. + \sum_{k=0}^K H_{31jk}^{(pn)} e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(n)}) \tau} + \sum_{k=0}^K H_{31jk}^{(np)} e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(p)}) \tau} \right) P_j(\cos \theta),
 \end{aligned} \tag{4.74}$$

where for all time dependencies the coefficients H_{31j} and H_{31jk} are known. The solutions of the corresponding coefficients in table 4.1 are set to zero because they exhibit secular terms. Since these solutions are unphysical and their structure would not correspond to those represented by the above equation, they must be suppressed. The phenomenon can be interpreted as a resonance, because the initial deformation of the mode m excites higher-order modes i . The secular terms appear in the three cases where

$$\left. \begin{aligned} & i(i-1)(i+2)/\alpha_m^2 \\ & i(i-1)(i+2)/4\alpha_m^2 \\ & i(i-1)(i+2)/9\alpha_m^2 \end{aligned} \right\} = -1. \tag{4.75}$$

Equation (4.73) then assumes the general form

$$\ddot{\delta}_{3i}^*(\tau) - c^2 \delta_{3i}^*(\tau) = A e^{-c\tau}, \tag{4.76}$$

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with the particular solution

$$\delta_{3i}^*(\tau) = -\frac{A}{4c^2} (1 + 2c\tau) e^{-c\tau}, \quad (4.77)$$

where the exceptional cases of (4.73) and their solutions are indicated with the asterisk. These solutions and coefficients in table 4.1 are shown and explained in Zrnić and Brenn (2021b). Due to their importance they are discussed here. The solutions exhibit a linear secular term in front of the exponential function which increases in time. Secular solutions relating to the first part of the statement (4.75), for which $i = m$, are due to functions of time on the right-hand sides of (4.73) leading to resonance with the frequency $\alpha_m^{(p)}$ or $\alpha_m^{(n)}$ appearing in the exponentials. They contain the coefficient ν_2 in a linear combination with other, known quantities, represented by the factor A in (4.77). For suppressing all these secular solutions, the values of the coefficient ν_2 are determined for each mode m of initial drop deformation in such a way that the solutions vanish. This results in a straining of time, and it can be seen as equivalent to a change of the oscillation frequency compared with the first-order motion. Except for $H_{314}^{(p)}$ and $H_{314}^{(n)}$, the corresponding coefficients are all those listed in table 4.1 for the various modes m .

The second part of the statement (4.75) appears only for $m = 5$ when $i = 8$, and for $m = 10$ when $i = 16$, corresponding to solutions with the coefficients $H_{3185}^{(pp)}$, $H_{3185}^{(nm)}$ and $H_{311610}^{(pp)}$, $H_{311610}^{(nn)}$, respectively. Since our analysis is restricted to modes less than $m = 5$ these cases are not treated here. The third part of the statement appears only for $m = 2$ when $i = 4$, with the corresponding two coefficients $H_{314}^{(p)}$ and $H_{314}^{(n)}$. The secular solutions $\delta_{34}^*(\tau)$ are due to functions of time on the right-hand sides of (4.73) leading to resonance with the frequency $3\alpha_m^{(p)}$ or $3\alpha_m^{(n)}$ appearing in the exponentials. In these secular solutions $\delta_{34}^*(\tau)$, the coefficient ν_2 does not occur, so that it cannot be used for suppressing them. Therefore, the coefficients $H_{314}^{(p)}$ and $H_{314}^{(n)}$ corresponding to this case are set to zero to suppress these secular solutions.

With the surface deformation η_{31} known, the coefficients C_{31j} and C_{31jk} are calculated using one of the boundary conditions (4.21) or (4.22) for all time dependencies. The procedure of this coefficient calculation is identical

to the second-order approximation. Due to large expressions, the details of this derivation are omitted. Simplified forms of the final representations are

$$\begin{aligned}
 p_{31}(r, \theta, \tau) = & -2 \sum_{h=0}^H r^h P_h(x) \left[C_{31h}^{(1)} \cos(3\alpha_{m,0}\tau) + C_{31h}^{(2)} \cos(\alpha_{m,0}\tau) \right. \\
 & + \sum_{k=0}^K C_{31hk}^{(3)} \cos((\alpha_{2k,0} + \alpha_{m,0})\tau) \\
 & \left. + \sum_{k=0 \setminus m}^K C_{31hk}^{(4)} \cos((\alpha_{2k,0} - \alpha_{m,0})\tau) \right] - \vec{u}_1 \cdot \vec{u}_2,
 \end{aligned} \tag{4.78}$$

$$\begin{aligned}
 u_{r31}(r, \theta, \tau) = & 2 \sum_{h=0}^H h r^{h-1} P_h(\cos \theta) \left[\frac{C_{31h}^{(1)}}{3\alpha_{m,0}} \sin(3\alpha_{m,0}\tau) + \frac{C_{31h}^{(2)}}{\alpha_{m,0}} \sin(\alpha_{m,0}\tau) \right. \\
 & + \sum_{k=0}^K \frac{C_{31hk}^{(3)}}{\alpha_{2k,0} + \alpha_{m,0}} \sin((\alpha_{2k,0} + \alpha_{m,0})\tau) \\
 & \left. + \sum_{k=0 \setminus m}^K \frac{C_{31hk}^{(4)}}{\alpha_{2k,0} - \alpha_{m,0}} \sin((\alpha_{2k,0} - \alpha_{m,0})\tau) \right] - \nu_2 u_{r1},
 \end{aligned} \tag{4.79}$$

$$\begin{aligned}
 u_{\theta 31}(r, \theta, \tau) = & 2 \sum_{h=0}^H r^{h-1} P_h^1(\cos \theta) \left[\frac{C_{31h}^{(1)}}{3\alpha_{m,0}} \sin(3\alpha_{m,0}\tau) + \frac{C_{31h}^{(2)}}{\alpha_{m,0}} \sin(\alpha_{m,0}\tau) \right. \\
 & + \sum_{k=0}^K \frac{C_{31hk}^{(3)}}{\alpha_{2k,0} + \alpha_{m,0}} \sin((\alpha_{2k,0} + \alpha_{m,0})\tau) \\
 & \left. + \sum_{k=0 \setminus m}^K \frac{C_{31hk}^{(4)}}{\alpha_{2k,0} - \alpha_{m,0}} \sin((\alpha_{2k,0} - \alpha_{m,0})\tau) \right] - \nu_2 u_{\theta 1},
 \end{aligned} \tag{4.80}$$

which describe the pressure (4.78), radial velocity (4.79) and polar velocity (4.80) "31".

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The related drop surface deformation reads

$$\begin{aligned} \eta_{31}(\theta, \tau) = 2 \sum_{h=0}^H P_h(\cos \theta) & \left[H_{31h}^{(1)} \cos(3\alpha_{m,0}\tau) + H_{31h}^{(2)} \cos(\alpha_{m,0}\tau) \right. \\ & \left. + \sum_{k=0}^K H_{31hk}^{(3)} \cos((\alpha_{2k,0} + \alpha_{m,0})\tau) + \sum_{k=0}^K H_{31hk}^{(4)} \cos((\alpha_{2k,0} - \alpha_{m,0})\tau) \right], \end{aligned} \quad (4.81)$$

where the definitions of the coefficients for the different time dependencies are given in section B of appendix C.

Since the contributions "31" to the third-order solutions are determined, the method enables us to proceed with the contributions "32". The contribution "32" relates to the homogeneous system of the third-order governing equations (4.18)-(4.20), and its solutions must satisfy the third-order homogeneous boundary conditions (4.21)-(4.22). The solutions are similar to the first order and the contributions "22" to the second-order solutions, and contain the linear combinations of eigen-solutions with index h . The velocity components, the pressure field and the deformation of the drop surface read

$$\begin{aligned} u_{r32}(r, \theta, \tau) = - \sum_{h=0}^H & \left(C_{32h}^{(p)} e^{-\alpha_{3h}^{(p)}\tau} + C_{32h}^{(n)} e^{-\alpha_{3h}^{(n)}\tau} \right) \\ & \times h(h+1)r^{h-1}P_h(\cos \theta), \end{aligned} \quad (4.82)$$

$$\begin{aligned} u_{\theta 32}(r, \theta, \tau) = - \sum_{h=0}^H & \left(C_{32h}^{(p)} e^{-\alpha_{3h}^{(p)}\tau} + C_{32h}^{(n)} e^{-\alpha_{3h}^{(n)}\tau} \right) \\ & \times (h+1)r^{h-1}P_h^1(\cos \theta), \end{aligned} \quad (4.83)$$

$$\begin{aligned} p_{32}(r, \theta, \tau) = - \sum_{h=0}^H & \left(\alpha_{3h}^{(p)} C_{32h}^{(p)} e^{-\alpha_{3h}^{(p)}\tau} + \alpha_{3h}^{(n)} C_{32h}^{(n)} e^{-\alpha_{3h}^{(n)}\tau} \right) \\ & \times (h+1)r^h P_h(\cos \theta), \end{aligned} \quad (4.84)$$

$$\eta_{32}(\theta, \tau) = \sum_{h=0}^H \left(\hat{\eta}_{32h}^{(p)} e^{-\alpha_{3h}^{(p)}\tau} + \hat{\eta}_{32h}^{(n)} e^{-\alpha_{3h}^{(n)}\tau} \right) P_h(\cos \theta). \quad (4.85)$$

We find two new complex conjugate angular frequencies α_{3h} from the homogeneous third-order zero normal-stress boundary condition that take the

form obtained in Rayleigh (1879). They are determined for all the summation indices h as

$$\begin{aligned}\alpha_{3h}^{(p)} &= i \sqrt{h(h-1)(h+2)} =: i \alpha_{3h,0} , \\ \alpha_{3h}^{(n)} &= -i \sqrt{h(h-1)(h+2)} =: -i \alpha_{3h,0} .\end{aligned}\tag{4.86}$$

The calculations of the coefficients C_{32h} and of the amplitudes $\hat{\eta}_{32h}$ are detailed in section C of appendix C of this thesis.

The third-order solutions with the contributions "32" are obtained in the simplified forms

$$u_{r32}(r, \theta, \tau) = -2 \sum_{h=0}^H \hat{\eta}_{32h} \alpha_{3h,0} r^{h-1} P_h(\cos \theta) \sin(\alpha_{3h,0} \tau) , \tag{4.87}$$

$$u_{\theta 32}(r, \theta, \tau) = -2 \sum_{h=1}^H \frac{\hat{\eta}_{32h} \alpha_{3h,0}}{h} r^{h-1} P_h^1(\cos \theta) \sin(\alpha_{3h,0} \tau) , \tag{4.88}$$

$$p_{32}(r, \theta, \tau) = 2 \sum_{h=0}^H \hat{\eta}_{32h} (h-1)(h+2) r^h P_h(\cos \theta) \cos(\alpha_{3h,0} \tau) , \tag{4.89}$$

$$\eta_{32}(\theta, \tau) = 2 \sum_{h=0}^H \hat{\eta}_{32h} P_h(\cos \theta) \cos(\alpha_{3h,0} \tau) . \tag{4.90}$$

The method finally gives the third-order nonlinear solutions that present the response of an inviscid oscillating drop to an initial deformation of its surface from the spherical shape governed by one initial deformation mode.

4.3. Results and discussion

This section presents the solutions developed by the weakly nonlinear analysis described in the previous section. The drop shapes are initially deformed according to different modes m . The section is divided into three parts. The first part describes the conservation of the drop volume as a function of deformation amplitude and time. The second part shows the drop surface shapes at maximum deformation during one period of oscillation and the position of the drop north pole as a function of time with its Fourier frequency spectrum for $m = 4$. In the same second part, due to its importance

in transport processes across surfaces, the drop surface area as a function of time is analysed. The final part quantifies frequency decrease and time spent in the elongated form of the drop shape with respect to the symmetry axis for varying initial deformation mode m . All the figures as well as part of the explanations are taken from Zrnić and Brenn (2021b).

4.3.1. Volume inspection

The weakly nonlinear theory represents all the field variables as truncated power series of a small deformation parameter as shown in equation (2.12). The drop surface shape is also expanded in power series. As a consequence, the non-dimensional drop volume may deviate from its exact value of $V_s = 4\pi/3$, i.e., the volume is not accurately conserved (Renoult et al., 2018). One way to validate the method is to determine analytically the relative volume deviation. The time during an oscillation period at which the largest drop volume deviation appears, depending on the deformation parameter, is denoted t_{max} . The non-dimensional drop volume $V(t)$ is determined as

$$V(t) = \frac{2\pi}{3} \int_{-1}^1 r_s^3(\theta, t) d \cos \theta, \quad (4.91)$$

where $r_s(\theta, t) = 1 + \eta_1(\theta, t)\eta_0 + \eta_2(\theta, t)\eta_0^2 + \eta_3(\theta, t)\eta_0^3$. The relative volume deviation from the exact value V_s is

$$\frac{V(t) - V_s}{V_s} = \frac{1}{2} \int_{-1}^1 [r_s^3(\theta, t) - 1] d \cos \theta =: R(\eta_0, t). \quad (4.92)$$

The values of R are all positive, oscillate in time and increase with η_0 . Figure 4.1 presents the results for the modes of initial deformation $m = 2, 3$ and 4 as functions of η_0 at the instant of time $t = t_{max}$ of largest volume deviation. The results show that the relative volume deviation for the fundamental mode $m = 2$ at the relatively large deformation $\eta_0 = 0.4$ is around 1%, while for $m = 3$ it is around 2.5%.

4.3.2. Deformed shapes of the drop surface

The present subsection describes the deformed drop surface shapes, and include the results of the analysis up to third order. In figures 4.2-4.4, the

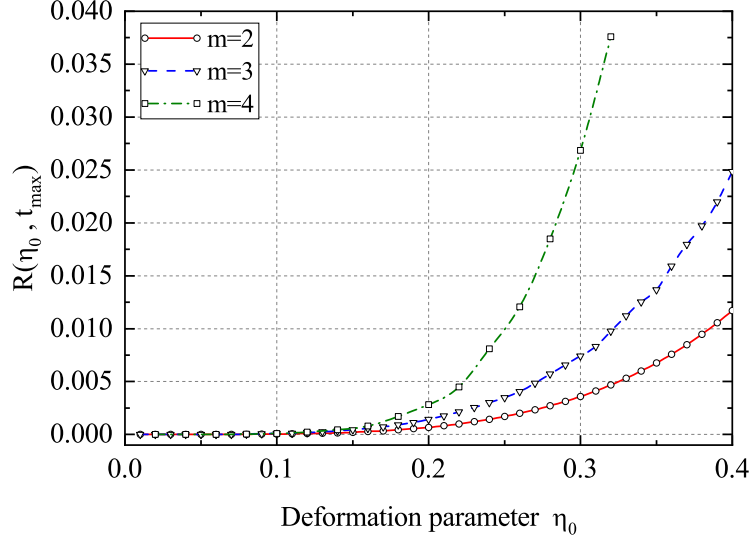


Figure 4.1.: Maximum relative drop volume deviation as a function of the deformation parameter η_0 for initial modes of deformation $m = 2, 3$ and 4 . The figure originates from Zrnić and Brenn (2021b).

results are validated against results from Tsamopoulos and Brown (1983) for the initial deformation modes $m = 2, 3$ and 4 . In these figures, the drop shapes are turned by 90 degrees compared with the sketch in figure 2.1, i.e, the drop axis of symmetry is horizontal. The figures show the results from the present weakly nonlinear theory (WNLT) to first order (Rayleigh) and to third order in states of maximum deformation for the deformation parameter $\eta_0 = 0.4$. In the same figures, the empty black circles indicate shapes that correspond to the work by Tsamopoulos and Brown (ibid.). They are plotted for validation of the WNLT results. We can conclude that the third-order results are in good agreement with the data by Tsamopoulos and Brown (ibid.).

Figure 4.2 shows the drop shapes for the fundamental mode of initial deformation, i.e. the prolate-to-oblate oscillation with a ratio of drop length to width $L/W = 1.76$. In the previous section, table 3.1 lists the degrees of Legendre polynomials contributing to the field variables of second and third orders for the various modes of initial deformation. In the third-order approximation, the coefficient $\nu_2 = -0.585187$ is found for the strained

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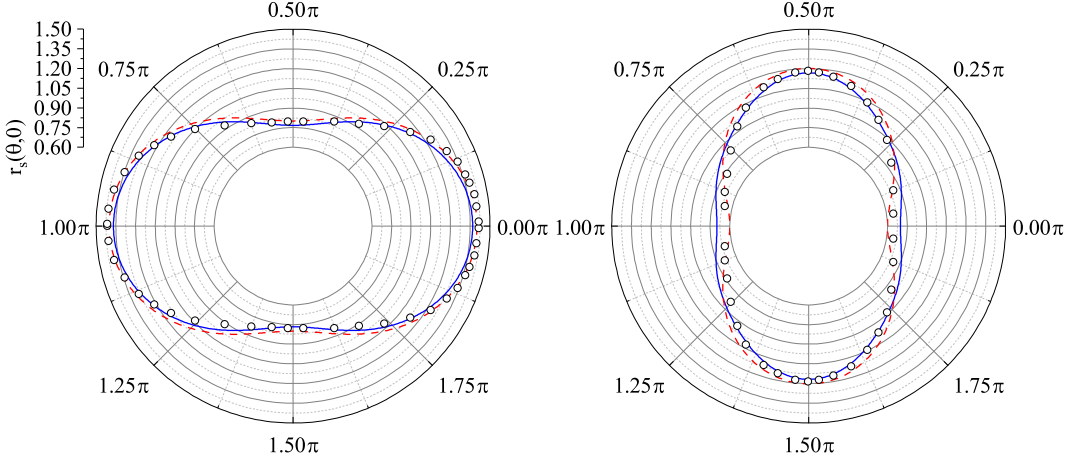


Figure 4.2.: Maximum deformed prolate and oblate states of a drop axisymmetric around the abscissa axis for the initial deformation mode $m = 2$ with $\eta_0 = 0.4$. Results show the Wnlt of first order (---) and third order (—) and the third-order result from Tsamopoulos and Brown (1983) presented as empty black circles. The figure originates from Zrnić and Brenn (2021b).

time coordinate. The theory yields corrections to the frequency for initial deformation modes $m > 2$ only.

The three- and four-lobed drop shapes in their states of maximum deformation are shown in figures 4.3 and 4.4, respectively. They agree well with the work by Tsamopoulos and Brown (1983). The degrees of the Legendre polynomials contributing to the third-order surface deformations for $m = 3$ are given in table 3.1 in the discussion section of the previous chapter. For the odd initial mode, even and odd degrees appear in the second and third orders of approximation, respectively, while for $m = 4$ only the even degrees from 0 to 12 contribute in both second and third order of approximation. At the rounded parts, the radii of curvature at third order are smaller than for the linear shape, but overall the third-order results follow the fundamental shapes of the first-order results. In the third-order calculation, for the initial deformation modes $m = 3$ and $m = 4$, the coefficient $\nu_2 = -0.988785$ and -1.398515 , respectively. Another interesting validation of the results is that Tsamopoulos (1990) determined frequency corrections which, in the present symbols, can be transcribed into the form $\nu = 1 + \nu_2 \epsilon^2 / 2$, where

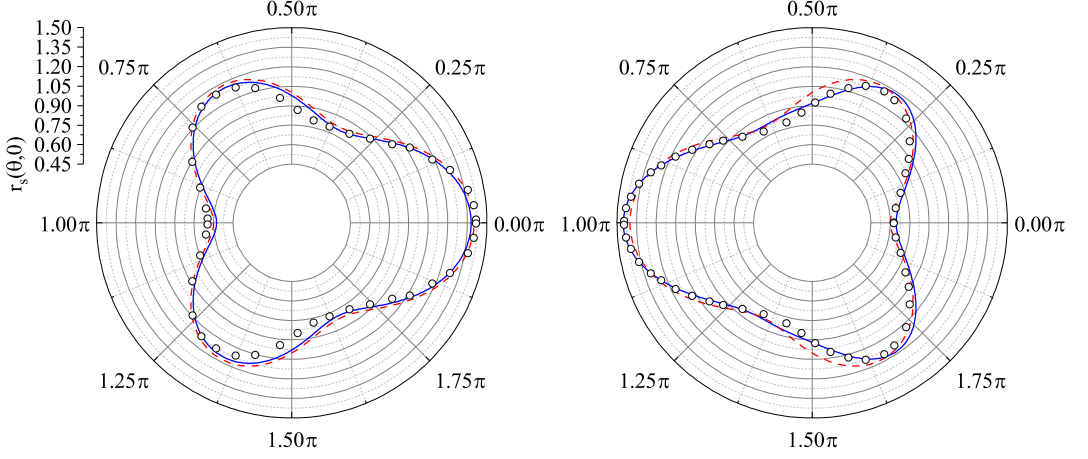


Figure 4.3.: Maximum deformed states of the drop for the initial deformation mode $m = 3$ with $\eta_0 = 0.4$, axisymmetric around the abscissa axis. Results show the WNL of first order (---) and third order (—) and the third-order result from Tsamopoulos and Brown (1983) presented as empty black circles. The figure originates from Zrnić and Brenn (2021b).

$\nu_2 \approx -1.17037$ for $m = 2$, $\nu_2 \approx -1.97757$ for $m = 3$ and $\nu_2 \approx -2.79703$ for $m = 4$. The symbol ϵ denotes the deformation parameter in the work by Tsamopoulos. The coefficients ν_2 by Tsamopoulos (ibid.), accounting for the factor of $1/2$ in the frequency series, are identical to the present results.

The position of the drop north pole relative to the undeformed state as a function of time for the initial deformation mode $m = 4$ and $\eta_0 = 0.2$ is shown in figure 4.5 (a). In the same figure, the third-order solutions are compared with the results of Tsamopoulos and Brown (1983). Figure 4.5 (b) depicts the Fourier power spectrum of the oscillation from the present theory. It shows the dominant mode and other modes excited by mode coupling. We notice that the dominant frequency is smaller than the Rayleigh frequency of 8.485 for $m = 4$, which is expected for the nonlinear oscillation. Further, with an increase of the deformation amplitude, the influence of mode coupling on the drop dynamics increases. Stronger mode coupling is experienced by the oscillations driven by initially higher modes, and this will be discussed in more detail later. Another phenomenon is the quasi-periodicity of the motion, which is one consequence of mode coupling and is visible in figure

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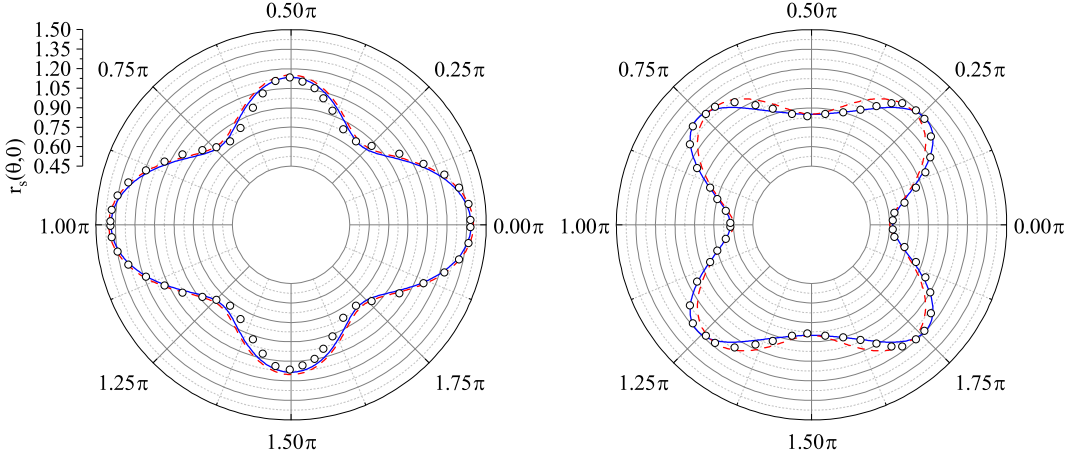


Figure 4.4.: Maximum deformed states of the drop for the initial deformation mode $m = 4$ with $\eta_0 = 0.4$, axisymmetric around the abscissa axis. Results show the WNL of first order (---) and third order (—) and the third-order result from Tsamopoulos and Brown (1983) presented as empty black circles. The figure originates from Zrnić and Brenn (2021b).

4.5 (a). Patzek et al. (1991) and Tsamopoulos and Brown (1983) excluded quasi-periodicity from their studies. The phenomenon explains the complex form of the deformation trace in time. Another validation is that the present results exhibit the same finding as in Basaran (1992) and Lundgren and Mansour (1988), who stated that every third deformation amplitude peak is lower than the previous two. We conclude that the frequency of the first oscillation agrees well with Tsamopoulos and Brown (1983) and differs in all the remaining oscillatory motion. Since the quasi-periodicity makes the oscillation frequency vary in time, the frequency variation will be presented and discussed in the following subsection.

In many applications, interest is focused on how the change of the drop surface area in time due to the shape oscillations influences transport processes of momentum, as well as heat and mass. Both the increase of the surface area and the velocity fields inside and outside the drop surface influence gradients of velocity, temperature and species concentration. Motivated with the importance for the prediction of drop surface area in time, this work shows figure 4.6 (a). It presents the relative surface area deviation from the

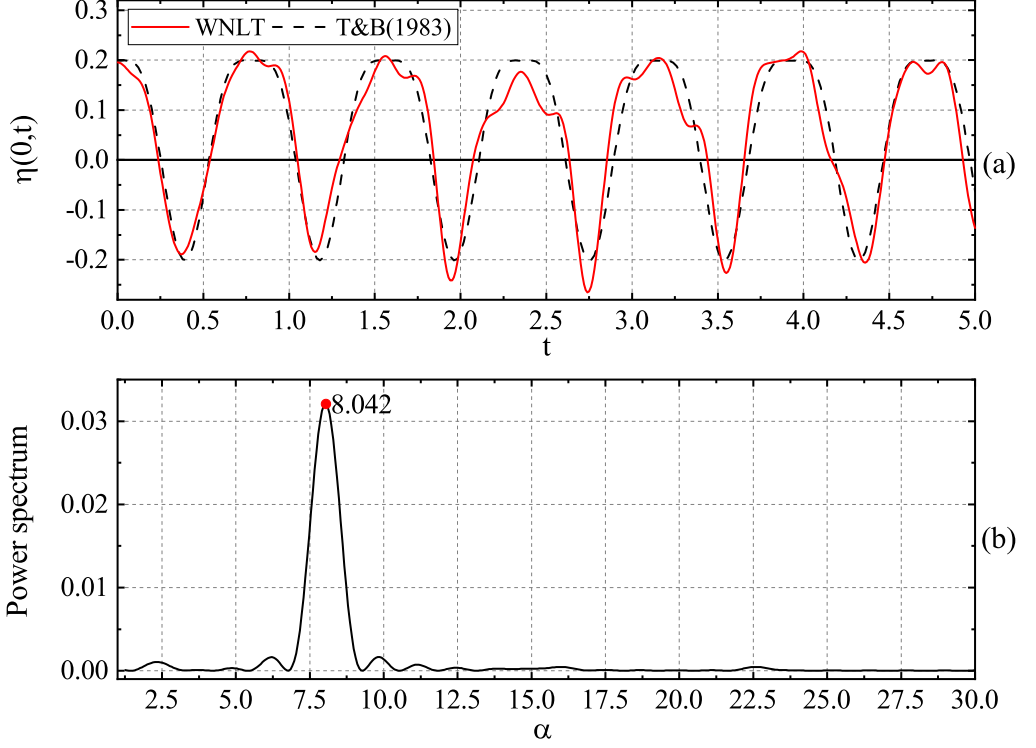


Figure 4.5.: (a) Position of the drop north pole relative to the undeformed state as a function of time from the present WNLt, compared against results by Tsamopoulos and Brown (1983). (b) Fourier power spectrum of the frequency for the oscillation in (a). The deformation parameter $\eta_0 = 0.2$ and initial mode $m = 4$. The figures originate from Zrnić and Brenn (2021b).

spherical state for the fundamental initial mode $m = 2$ as a function of time for the two different deformation parameters $\eta_0 = 0.1$ and $\eta_0 = 0.32$. The drop surface in the maximum deformed state for $\eta_0 = 0.32$ is larger than the spherical state by 3.8 %, 7 % and 11% for $m = 2$, $m = 3$ and $m = 4$, respectively. The trace of the calculated drop surface area for $\eta_0 = 0.1$ follows a sinusoidal shape, where the minimum values touch the value of the non-dimensional surface area of the spherical shape. With an increase of the deformation parameter to 0.32, the minimum surface areas increase, and they are always larger than spherical in the course of the oscillation. Another observation is the delay of the minimum surface area, which increases with

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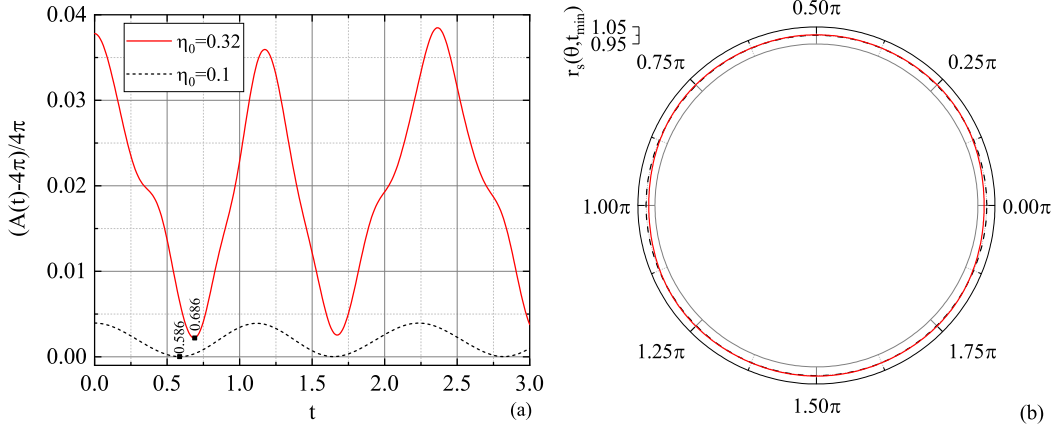


Figure 4.6.: (a) Relative surface area deviation from the spherical area as a function of time. (b) Drop meridional section at minimum surface area for $\eta_0 = 0.32$ (—), compared to the circular section for $\eta_0 = 0.1$ (----), both for fundamental initial mode. The figures originate from Zrnić and Brenn (2021b).

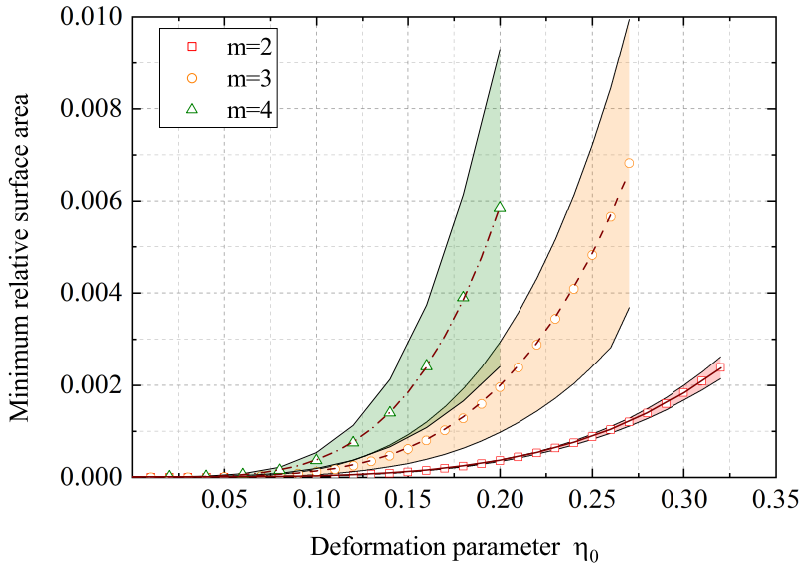


Figure 4.7.: Minimum relative drop surface area deviation from the spherical shape as a function of the deformation parameter η_0 for the initial deformation modes $m = 2, 3$ and 4 . The figure originates from the supplementary materials by Zrnić and Brenn (2021b).

the deformation parameter and, due to the quasi-periodicity of the motion, appears irregularly in every period. Figure 4.6 (b) shows the drop meridional section for $\eta_0 = 0.32$ at $t = 0.686$, i.e. in the state of minimum drop surface area, which is compared with the circular (minimum) shape for $\eta_0 = 0.1$. It confirms the result from Patzek et al. (1991) showing that drops oscillating nonlinearly at large amplitude never reach the spherical state during the oscillations. This effect cannot be represented by the linear solution and appears for large initial deformations only.

Another finding that concerns drop surface area is the data in Fig. 4.7, which show the minimum deviation of the drop surface area from the spherical state as a function of the deformation parameter η_0 for the modes of initial deformation $m = 2, 3$ and 4 . The lines are fit curves, showing the proportionality of the surface area deviations, which are empirically found according to the function $0.0142 (m \eta_0)^4$.

4.3.3. The drop shape oscillation time scales

One of the well-known nonlinear effects in drop shape oscillations is that, for the prolate-oblate motion of the fundamental initial mode, the drop spends more time in the prolate than in the oblate state of deformation. This phenomenon will be studied first, and figure 4.8 shows results for the corresponding initial deformation mode $m = 2$. In order to present the WNLТ results in comparison with results from literature, we depict the nonlinear effects with dependency on deformation parameter and aspect ratio in separate figures. Therefore, figure 4.8 (a) shows the time percentage spent in the prolate form as a function of the drop aspect ratio L/W , while figure 4.8 (b) shows it as a function of the deformation parameter η_0 . The solutions of Tsamopoulos and Brown (1983) are transcribed into the dependency on the deformation parameter, using their analytical results. In figures 4.8 (a) and (b), the prolate times in the WNLТ curves are determined from traces of the position of the drop north pole ($\theta = 0$) as a function of time, as in figure 4.5 (a). For depiction of the results in the corresponding figure, the times spent in the prolate state are measured for the first ten oscillations of the traces from the third-order calculations. Since the motion is quasi-periodic, the measured

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values differ in every oscillation. Thus, for every deformation parameter, every point on the diagram is a mean value and the standard deviation of the time spent in the prolate state. The WNLT results are shown as solid lines for the mean values and shaded areas for the standard deviation. The second-order results do not exhibit this phenomenon, so that their values are depicted by solid lines only. The data show that the present WNLT represents the time asymmetry of the $m = 2$ drop shape oscillations. The results from the present WNLT almost perfectly coincide with the results by Tsamopoulos and Brown (1983). At larger deformations, the WNLT shows slightly smaller excess times than the latter. The theory of Tsamopoulos and Brown (ibid.), however, does not represent the quasi-periodicity of the motion.

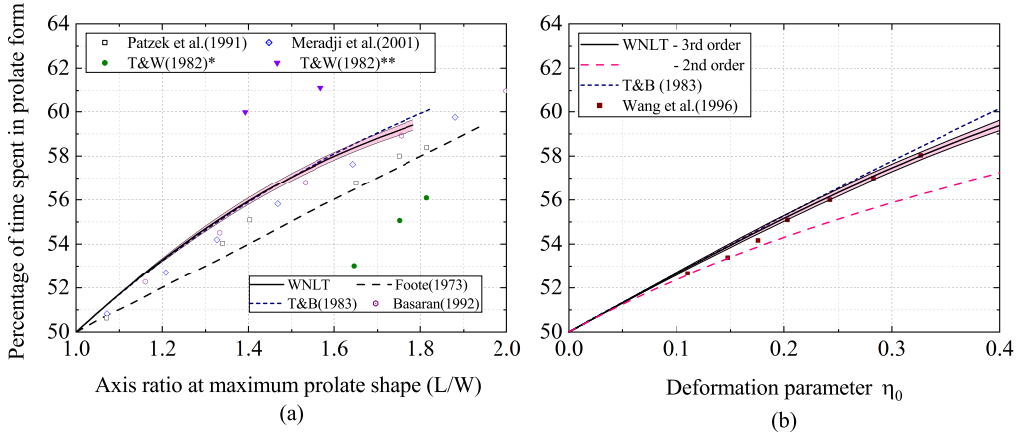


Figure 4.8.: Percentage of the oscillation period spent in the prolate shape (a) as a function of the aspect ratio L/W of the drop at maximum deformation, and (b) as a function of the deformation parameter η_0 for the fundamental initial deformation mode $m = 2$. In (a), experimental data by Trinh and Wang (1982) for initial steady prolate drive is denoted with T&W(1982)*, and for initial static prolate shape as T&W(1982)**. The standard deviations are presented with shaded areas. The figures originate from Zrnić and Brenn (2021b).

For validation, figure 4.8 shows experimental results by Trinh and Wang (1982), and numerical results by Foote (1973), Alonso (1974), Patzek et al. (1991), Basaran (1992) and Meradji et al. (2001). Trinh and Wang (1982) carried out experiments on neutrally buoyant silicone oil and carbon tetrachloride mixture drops in distilled water and measured the percentage of time spent

in prolate shapes. They acoustically excited the drops near the fundamental frequency to investigate the shape oscillations. When the excitation was turned off, the drop performed a free oscillation, and the oscillation frequency was also measured. Since the experiments were performed in an immiscible viscous liquid/liquid system, the calculation requires corrections for density and viscosity of the host fluid. The corrections can be done because the measurements are in the limits of moderate-amplitude oscillations and the drops are large ($a = 1$ cm). The calculation of the viscous correction of the measured frequency to an inviscid liquid/liquid system was derived by Prosperetti (1980b) (given in Fig. 14 on page 175 of that paper). The non-dimensional frequency is corrected with the viscosities of the inner and outer fluids, which have the same density. Foote (1973) and Alonso (1974), who solved the oscillating drop problem numerically, made the same corrections and confirmed the possibility of comparing the immiscible liquid/liquid system with free drop shape oscillations in a vacuum. Chandrasekhar (1959) states that the effect of the viscosity on the oscillation frequency is very small when the Ohnesorge number $\mu / (\sigma a \rho)^{1/2}$ is small. Both the data of Foote (1973) and Alonso (1974) satisfy this condition, because the Ohnesorge numbers are $1/35.4$ and $1/3.3$, respectively, as pointed out by Tsamopoulos and Brown. Further numerical work is due to Basaran (1992) and Meradji et al. (2001), who investigated the time evolution of small-viscosity drop oscillations ($Oh = 0.01$). Patzek et al. (1991) numerically studied free oscillations of inviscid liquid drops, and their results are also presented for comparison. From the latter three studies, the percentage of time spent in the prolate form of the two-lobed oscillations fits in the region between our inviscid WNLT and the small-viscosity results by Foote (1973). The initial state of the drop has an influence on mode coupling. This is clearly presented in figure 4.8 (a) by the data by Trinh and Wang (1982), who distinguished two initial states - steady prolate drive or static prolate shape. The time asymmetry between prolate and oblate states found in the oscillatory motion for these two initial states is quite different.

Trinh and Wang (ibid.) used the method for determining the time spent in the prolate form and the oscillation frequency for different deformation amplitudes. The same method was applied by Wang, Anilkumar, and Lee

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(1996), who acoustically positioned drops in a microgravity environment. They extracted the data for a glycerin/water drop with $Oh = 0.0125$ using a central averaging scheme to eliminate the role of viscosity in nonlinear effects. The experiments reported by Trinh and Wang (1982) can be compared with the inviscid case, because the results have $Oh \ll 0.1$. The excess times from the present second- and third-order solutions together with the results by Tsamopoulos and Brown (1983) and Wang, Anilkumar, and Lee (1996) are presented in figure 4.8 (b). The present third-order results coincide with the data by Tsamopoulos and Brown (1983) for small deformation parameters $\eta_0 < 0.15$. For higher deformation parameters, the presented third-order results agree very well with the data by Wang, Anilkumar, and Lee (1996).

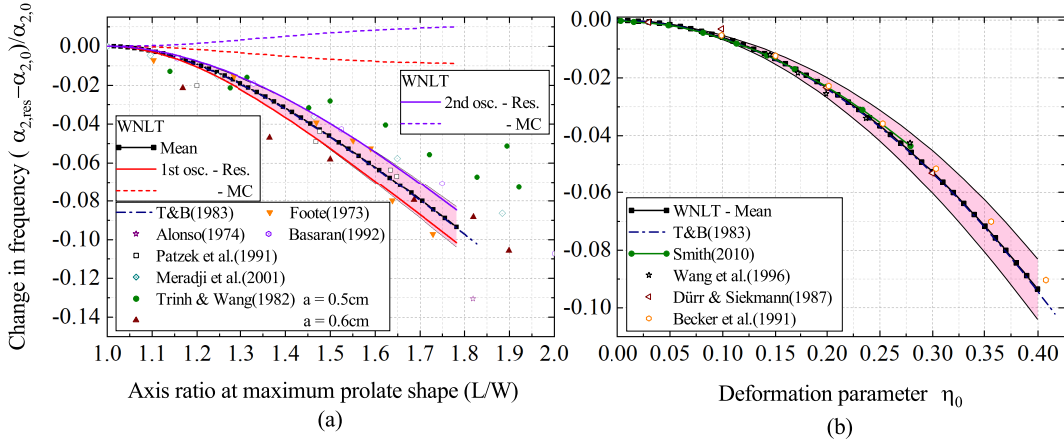


Figure 4.9.: Decrease of the angular frequency (a) as a function of the drop aspect ratio L/W , and (b) as a function of the deformation parameter η_0 compared with the results from different literature. The results presented are for the fundamental initial mode. The standard deviations are presented with shaded areas. The figures originate from Zrnić and Brenn (2021b).

The decrease of the oscillation frequency with increasing drop deformation is another nonlinear effect in drop shape oscillations, which the WNLT represents. For the initial deformation mode $m = 2$, figure 4.9 (a) presents the relative decrease of the oscillation frequency below the Rayleigh value $\alpha_{2,0}$ as a function of the maximum aspect ratio L/W of the drop shape reached in the oscillations. For comparison with other literature, figure 4.9 (b) shows the data as a function of the deformation parameter η_0 . In figures 4.9 (a) and

(b), the oscillation frequencies depicted by the WNLT curves are determined from traces of the position of the drop north pole ($\theta = 0$) as a function of time, as in figure 4.5 (a). In these data, the period length is extracted determining the resultant angular oscillation frequency termed $\alpha_{2,res}$ on the ordinate axis of figure 4.9. The period lengths are measured for a set of ten oscillations on the trace for every deformation parameter. Alternatively, the surface deformation was plotted as a function of the stretched time coordinate τ , which was introduced to suppress secular terms in the solutions of a differential equation arising in the theoretical analysis. Determining the period length from these traces, a frequency decrease or increase is found due to mode coupling. The mode coupling phenomenon arises in the various time dependencies of the nonlinear terms in the governing equations and boundary conditions for the second- and third-order approximations. In order to visualize the dependencies, three different WNLT results are presented in figure 4.9 (a). The solid lines depict the resultant values of the frequency decrease for the first and second oscillations of each trace, while the dashed lines depict the contributions from mode coupling only. The resultant variation of the oscillation frequency is quantified by the standard deviation, represented by the shaded areas. This variation of the period in every oscillation makes oscillations quasi-periodic. The effect is due to the mode coupling. When one subtracts the mode coupling result from the resultant values, the results coincide with the data by Tsamopoulos and Brown (1983). The solid lines with square symbols show the third set of results from the WNLT, which represent the mean values from each set of ten oscillations analysed. The concave shape of the resultant frequency decrease for $m = 2$ is due to the quadratic form of the strained time coordinate. For the same mode, one can conclude that the appearance of a narrow shaded area is due to weak mode coupling, and we can see that the mean results coincide perfectly with the data by Tsamopoulos and Brown (ibid.). The data by Foote (1973) and by Patzek et al. (1991) fall in the region of the WNLT results when the standard deviation is included and follow the same quadratic shape.

The inviscid numerical results by Dürr and Siekmann (1987), Becker, Hiller, and Kowalewski (1991), Wang, Anilkumar, and Lee (1996), Smith (2010), and

the classical results from Tsamopoulos and Brown (1983) transcribed into the dependency on the deformation parameter by using their analytical solutions are shown in figure 4.9 (b). Smith uses different time scales and a formal perturbation scheme to determine original modulation equations. The author derives expressions for the drop surface deformation by using two ordinary differential equations for the balance of energy and the averaged projection of the Navier-Stokes equations onto the vorticity vector. An analytical work that deals with the motion of an inviscid, incompressible droplet in a vacuum is due to Becker, Hiller, and Kowalewski (1991), while Dürr and Siekmann (1987) carried out numerical simulations using the boundary-integral technique. All the three investigations confirmed a decrease of the oscillation frequency with increasing deformation amplitude as a nonlinear characteristic of the $m = 2$ oscillations. Including the variations due to mode coupling, the present WNLT data coincides with all the compared data.

The analysis of excess time and frequency decrease is extended to the higher modes of initial deformation $m = 3$ and $m = 4$. The percentage of the oscillation period the drop north pole ($\theta = 0$) spends at a larger distance from the drop center than in the spherical state is shown in figure 4.10 for the modes $m = 2, 3$ and 4 . These north pole positions indicate an elongation of the drop in the direction of the symmetry axis, and the shape corresponds to the prolate shape for $m = 2$. The mean values and standard deviations of the percentage of the oscillation period spent in elongated shape are shown for the second and third order of approximation in figures 4.10(a) and 4.10(b), respectively. For every m , the lines represent the mean values, and the shaded areas the standard deviations as functions of the deformation parameter η_0 . As in the data for $m = 2$, the corresponding figures show the WNLT results deduced from the first ten, eight and six oscillations for the initial deformation modes $m = 2, 3$ and 4 , respectively. At the third order of approximation, the surface deformation trace with time becomes more complicated with increasing η_0 . To make the identification of period lengths possible, the number of oscillations in the set is smaller for the higher initial deformation modes. For the same reason, the data for the initial mode $m = 4$ runs to $\eta_0 = 0.3$ only. Figures 4.10 (a) and 4.10 (b) present the increase of the excess time with the mode of initial deformation,

with the deformation parameter η_0 , and with the order of approximation. At more pronounced deformation, an important difference between the present WNLT and Tsamopoulos and Brown (1983) is the quasi-periodic motion. On the other hand the oscillations of the fundamental mode exhibit a weak mode coupling and, consequently, smaller variations. In figure 4.10(a) the data from both theories at second-order approximation does not show quasi-periodicity for the fundamental initial deformation mode. From this finding one can conclude that quasi-periodicity is a second-order effect for the higher modes of initial deformation, but a third-order effect for $m = 2$.

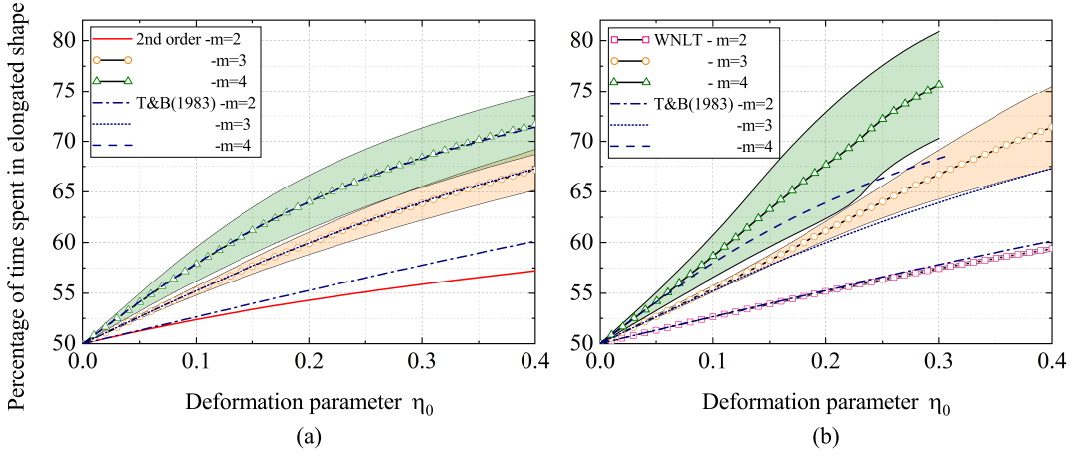


Figure 4.10.: Percentage of the oscillation period the drop spends in a shape elongated along the symmetry axis for oscillations with $m = 2, 3$ and 4 as a function of the deformation parameter η_0 . Mean value and standard deviation including (a) the second-order and (b) the third-order approximation, compared with the results from Tsamopoulos and Brown (1983). The figures originate from Zrnić and Brenn (2021b).

The last part of this section focuses on the results for the frequency decrease for the higher modes of initial deformation. Figure 4.11 shows a comparison of the frequency decrease as a function of η_0 between the modes $m = 2, 3$ and 4 . For the WNLT results, the mean values and standard deviations of the relative frequency change for second and third order of approximation are shown in figures 4.11 (a) and 4.11 (b), respectively. Similar to the results from the previous figure, the data are deduced from the first ten, eight and six

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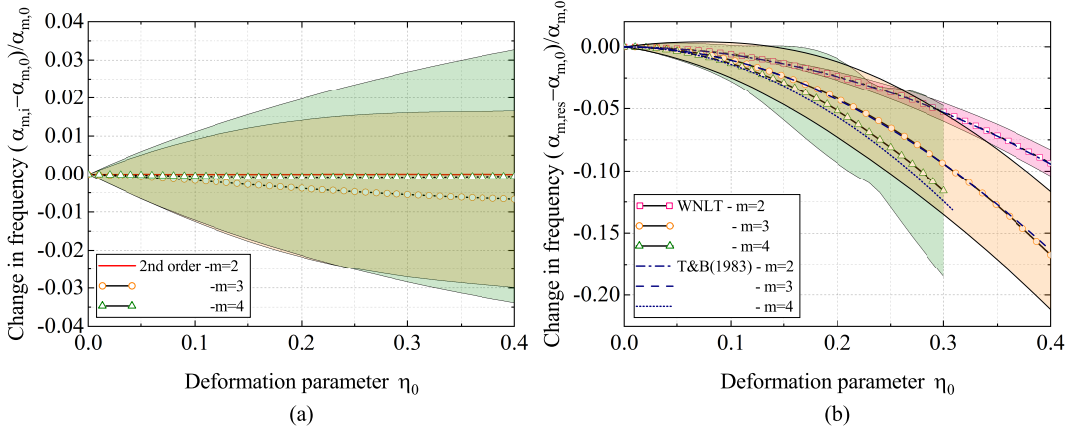


Figure 4.11.: The angular frequency decrease as a function of the deformation parameter η_0 for oscillations with initial mode $m = 2, 3$ and 4. Mean value and standard deviation of the relative decrease of (a) $\alpha_{m,i}$ at second, and (b) $\alpha_{m,res}$ at third order of approximation, compared with the results from Tsamopoulos and Brown (1983). The figures originate from Zrnić and Brenn (2021b).

oscillations for $m = 2, 3$ and 4, respectively. The reasons for doing this are described in the text for the previous figure. In figure 4.11 (b), the results by Tsamopoulos and Brown (1983) are also presented for comparison. For the fundamental initial deformation mode, the frequency does not change with the deformation parameter in the second-order approximation, thus requiring the theory to be taken to third order to see this effect for this mode. The mean relative frequency change for $m \geq 3$ at second order (figure 4.11 (a)) is also close to zero. As represented by the shaded areas, the frequency fluctuates in time. The fluctuations increase with the mode of initial deformation and with the deformation parameter. We explain this behavior with the modes excited already in the second-order approximation, which are absent at $m = 2$. As shown in table 3.1, the number of excited modes increases with increasing initial deformation mode, and therefore the frequency fluctuations increase with the mode. Consequently, the quasi-periodicity is enhanced with increasing mode of initial deformation. It produces stronger frequency fluctuations, which for $m = 4$ start to oscillate for varying η_0 . There is very good agreement of the mean relative frequency decrease with the results by Tsamopoulos and Brown (ibid.).

4.4. Conclusions

The current chapter presents the weakly nonlinear analysis of shape oscillations of an inviscid, axisymmetric liquid drop in a vacuum. The method expands the components of the velocity vector field, the pressure and the drop shape in power series of a small deformation parameter η_0 and yields the set of equations of motion with their boundary and initial conditions that correspond to different orders of the approximation. The analysis is taken to third order in order to describe the nonlinear effects for all the modes m studied. During the derivations, secular solutions for the surface deformation appear at second and third orders of approximation. Steered by the work of Yuen (1968), they are eliminated using a strained time coordinate. An additional pair of secular terms arises for the special case of the initial fundamental mode $m = 2$. This pair of terms is suppressed by setting the related coefficients in the series expansions to zero.

The method yields the linear, first-order solution known from Rayleigh (1879), while the second- and third-order solutions represent the nonlinear influences from the first order and from the first and second orders, respectively. Since the method represents the drop shape as the truncated power series, the non-dimensional drop volume deviates from the correct value of $4\pi/3$. The relative deviation at large deformations up to $\eta_0 = 0.4$ is around 1% and 2.5% for the two-lobed $m = 2$ and three-lobed deformation modes $m = 3$, respectively. The main benefit of the weakly nonlinear approach is its ability to represent quasi-periodic motions induced by mode coupling for increasing deformation amplitude. The quasi-periodical behaviour is found by other authors in numerical simulations, but was never broken down into details for better understanding. Due to the mode coupling, various frequencies appear in the Fourier power spectra. As the deviation from the spherical shape increases, the drop surface area also increases, which is more pronounced for higher deformation parameters. If one fixes $\eta_0 = 0.4$, the surface area increases by around 6%, 10% and 14% for the initial deformation modes $m = 2, 3$ and 4, respectively. An excess time in the prolate deformed state, and a decrease of the oscillation frequency, are known nonlinear effects for the mode $m = 2$. These effects are presented correctly and are in agree-

ment with existing experimental and theoretical literature. The results are extended to the multi-lobed deformation modes $m = 3$ and $m = 4$. For the fundamental initial mode, the method yields the correct frequency decrease only if the method is expanded to the third order. The frequency decrease is seen in the second-order approximation for all higher-order initial modes of deformation. Both the time asymmetry and the frequency vary in time due to the quasi-periodic oscillatory motion, which is a direct cause of mode coupling.

The detailed time behaviour of the drops is revealed for the nonlinear shape oscillations. The study represents the influence of the coupling of modes, excited by the mode of initial deformation, on the time behaviour of the oscillations and therefore goes beyond the classical literature. Due to expansion of the analysis up to third order, the quasi-periodic nature of the oscillations is presented and is caused by the interaction between the various modes. Thus, the analytically determined drop surface area shows that drops in nonlinear oscillations, when strongly deformed, never reach the spherical state, thus exhibiting a resultant surface area increase above the value of the sphere. The nonlinear effects observed on the surface are closely related to the velocity field in the drop, which influences the transport of momentum, heat and mass across the drop surface. The transfer processes are influenced by the motion of the drop surface via the motion of the gas phase. Therefore, the fields of study that investigate transport processes across the oscillating drop surface can greatly benefit from the results presented. Application examples are containerless materials processing on the basis of single drops and in-air microfluidics.

5. Conclusions and outlook

This study presents a weakly nonlinear analysis of viscoelastic, viscous and inviscid liquid drops, which are axisymmetric and placed in a vacuum. It establishes the governing equations, as well as the boundary and initial conditions, which represent the problem of drop shape oscillations. The weakly nonlinear analysis used is also termed the method of successive approximations. To the best of my knowledge, this is the first weakly nonlinear analysis of viscoelastic and Newtonian drop shape oscillations. In this method, the quantities involved are expanded in power series of a small deformation parameter η_0 . The quantities are the components of the velocity and pressure fields, the drop shape, extra stress and rate of deformation tensor. The variables expanded are substituted into the governing equations, boundary and initial conditions, and collecting all the terms with equal powers of the deformation parameter results in equations and conditions with various orders of approximation. For the inviscid, viscous and viscoelastic cases, the method is extended to the third order. The solutions are derived to the third order, except for the viscoelastic case, where we present only the first-order solutions. At the boundary, the fields are expanded in Taylor series, making it possible to satisfy boundary conditions on the deformed boundary, but still allowing the functions to be evaluated at $r = 1$. A Legendre polynomial defines the initial deformation of the drop surface against the spherical state. The surface is initially at rest.

The method yields the linear, first-order solutions known from Brenn and Teichtmeister (2013), Lamb (1881) and Rayleigh (1879) for viscoelastic, viscous and inviscid liquid drops, respectively. The characteristic equation for the complex angular frequency of the drop is obtained agreeing with Brenn and Teichtmeister (2013) for the viscoelastic, with Chandrasekhar (1959) and Prosperetti (1977) for the viscous, and with Rayleigh (1879) for the inviscid

case. The second- and third-order solutions represent the nonlinear influences from the first order, and from the first and second order, respectively. The method represents the field variables including the drop shape as truncated power series. As a consequence, the nondimensional drop volume deviates from the correct value of $4\pi/3$. For the inviscid drop, the deviations are not more than 2.5% for the worst case, and for $Oh < 1$ they are not more than 2%.

The main benefit of the weakly nonlinear analysis is to present the detailed time behaviour of drops due to nonlinear drop shape oscillations. The study presents the effects and quantifies the influence of mode coupling, excited by the mode of initial deformation. The quasi-periodic motion obtained is due to mode coupling. The solutions of the analysis reveal the modes arising from the motion, and the analysis quantifies their contribution to the resultant motion. The shapes in both the inviscid and the viscous cases are compared with the related literature, and the comparison shows good agreement. The nonlinear effects, such as excess time in the prolate shape compared with the oblate, and a frequency decrease, were also positively validated with existing literature. In the case of the latter effect, the present results coincide with the classical work by Tsamopoulos and Brown (1983). The effects are presented for the higher-order modes also, which proves that the theory has no limit regarding the initial modes of deformation. In the inviscid case, and for the higher-order initial modes of deformation, the frequency decrease is seen in the second-order approximation, unlike the viscous and viscoelastic cases. Since the drop at stronger deformation exhibits quasi-periodic oscillatory motion, both the time asymmetry and frequency vary in time. The variation weakens with increasing viscosity of the liquid, due to enhanced damping.

The mode coupling revealed in the higher-order solutions has an effect on the behaviour of the drop shape oscillations for supercritical Ohnesorge numbers. In motions with Newtonian liquid at higher-order initial deformation mode, the linear theory fails to predict an oscillatory motion. Even though the initial mode behaves as aperiodic, lower-order modes, which are excited, respond oscillation-like, and therefore the resultant motions are also oscillatory. To the best of our knowledge, this effect was never revealed before.

The drop surface area increases as the deviations from the spherical shape increase. The surface motion of the drop influences the transport of momentum, heat and mass across the drop surface. Since the nonlinear effects observed on the surface are closely related to the drop shape oscillations, the nonlinearities influence the transfer process across the surface. Thus, applications involving liquid drops and their transport processes, will benefit from the current study.

In an extension of this work one can take transport processes into account, such as drop evaporation. Another attractive field would be the inclusion of oscillating gas bubbles in an ambient liquid. This case is known to include complicated deformed shapes of the interface during the rise of the bubbles in the liquid. In view of bubble columns in chemical engineering, this option appears very attractive, since the state of development of simulation techniques which consider bubbles has only little potential to predict bubble flow behaviour on industrial scales.

Appendix

Appendix A.

Derivation details for Newtonian drop shape oscillations

The present chapter of the appendix details the determination of the velocity and pressure fields and surface deformation amplitudes of an oscillating Newtonian drop. The details pertain to chapter 3 of the thesis. In order to improve the readability, the present details of the analysis were removed from the bulk of the thesis, but the structures of the various solutions are retained. The aim of the present representation is to provide all the details of the derivations missing from chapter 3 and is intended as a collection of equations for coefficients and deformation amplitudes in the solutions for the various orders of approximation.

A. First-order solutions

The angular velocity component and pressure of first order, $u_{\theta 1}(r, \theta, t)$ and $p_1(r, \theta, t)$, read

$$\begin{aligned}
 u_{\theta 1}(r, \theta, t) = & -\frac{\hat{\eta}_1^{(p)} \alpha_m^{(p)}}{m} P_m^1(\cos \theta) e^{-\alpha_m^{(p)} t} \left[\left(1 + \frac{2(m^2 - 1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right) r^{m-1} \right. \\
 & - \frac{2(m-1)q^{(p)}}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \left((m+1) \frac{j_m(q^{(p)} r)}{j_m(q^{(p)})} \frac{1}{q^{(p)} r} - \frac{j_{m+1}(q^{(p)} r)}{j_m(q^{(p)})} \right) \Big] \\
 & - \frac{\hat{\eta}_1^{(n)} \alpha_m^{(n)}}{m} P_m^1(\cos \theta) e^{-\alpha_m^{(n)} t} \left[\left(1 + \frac{2(m^2 - 1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right) r^{m-1} \right. \\
 & - \frac{2(m-1)q^{(n)}}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \left((m+1) \frac{j_m(q^{(n)} r)}{j_m(q^{(n)})} \frac{1}{q^{(n)} r} - \frac{j_{m+1}(q^{(n)} r)}{j_m(q^{(n)})} \right) \Big] \\
 = & - \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)} e^{-\alpha_m^{(p)} t} f_{\theta}^{(p)}(r) + \hat{\eta}_1^{(n)} \alpha_m^{(n)} e^{-\alpha_m^{(n)} t} f_{\theta}^{(n)}(r) \right] P_m^1(\cos \theta) ,
 \end{aligned} \tag{A.1}$$

$$\begin{aligned}
 p_1(r, \theta, t) = & -\frac{r^m}{m} \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)2} e^{-\alpha_m^{(p)} t} \left(1 + \frac{2(m^2 - 1)}{2q^{(p)} Q_m^{(p)} - q^{(p)2}} \right) \right. \\
 & + \hat{\eta}_1^{(n)} \alpha_m^{(n)2} e^{-\alpha_m^{(n)} t} \left(1 + \frac{2(m^2 - 1)}{2q^{(n)} Q_m^{(n)} - q^{(n)2}} \right) \Big] P_m(\cos \theta) \\
 = & - \left[\hat{\eta}_1^{(p)} \alpha_m^{(p)2} e^{-\alpha_m^{(p)} t} f_p^{(p)}(r) + \hat{\eta}_1^{(n)} \alpha_m^{(n)2} e^{-\alpha_m^{(n)} t} f_p^{(n)}(r) \right] P_m(\cos \theta) ,
 \end{aligned} \tag{A.2}$$

where $P_m^1(\cos \theta)$ denotes a Legendre function of degree m and order 1.

B. Solutions for the modified pressure $\mathcal{P}_{21}$

The right-hand side of equation (3.44) is simplified introducing the series expansions of the function $qr j_m(qr)$, of its square, and of the square of its

derivative with respect to r , each for the eigenvalue (p) or (n) . Using the work by Dong and Lemus (2002), the squares of the Legendre polynomial $P_m(x)$ and of the Legendre function $P_m^1(x)$ are represented as series expansions in Legendre polynomials. Therefore, we re-write (3.44) as

$$\begin{aligned} \Delta \mathcal{P}_{21} = & -q^6 C_{2m}^2 \left[\left(\sum_{k=0}^N c_{km}(qr)^{2k+2m-2} - \sum_{k=0}^N b_{km}(qr)^{2k+2m} \right. \right. \\ & + \left. \frac{C_{1m}}{C_{2m}} \frac{m+1}{q^{m+1}} \sum_{k=0}^N (2k+m+1) a_{km}(qr)^{2k+2m-2} \right) \times \sum_{l=0}^{2m} C_{3l} P_l(x) \\ & + \left(\sum_{k=0}^N b_{km}(qr)^{2k+2m-2} + \frac{C_{1m}}{C_{2m}} \frac{1}{q^{m+1}} \sum_{k=0}^N a_{km}(qr)^{2k+2m-2} \right) \\ & \times m^2(m+1)^2 \sum_{l=0}^{2m} C_{4l} P_l(x) \Big] e^{-2\alpha_m t}, \end{aligned} \quad (\text{A.3})$$

where this form is with the eigenvalues (p) or (n) . The approximated right-hand side of the Poisson equation for the time dependency according to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$, denoted (pn) , reads

$$\begin{aligned} \Delta \mathcal{P}_{21}^{(pn)} = & -C_{2m}^{(p)} C_{2m}^{(n)} \left\{ \left[\sum_{k=0}^N \left(q^{(p)^2} + q^{(n)^2} \right) q^{(p)^2} q^{(n)^2} f_{km} r^{2k+2m-2} \right. \right. \\ & - \sum_{k=0}^N 2q^{(p)^3} q^{(n)^3} t_{km} r^{2k+2m} + \sum_{k=0}^N \left(q^{(p)^6} \frac{C_{1m}^{(n)}}{C_{2m}^{(n)}} \frac{m+1}{q^{(p)^{m+1}}} q^{(p)^{2k+2m-2}} \right. \\ & + \left. \left. q^{(n)^6} \frac{C_{1m}^{(p)}}{C_{2m}^{(p)}} \frac{m+1}{q^{(n)^{m+1}}} q^{(n)^{2k+2m-2}} \right) (2k+m+1) a_{km} r^{2k+2m-2} \right] \\ & \times \sum_{l=0}^{2m} C_{3l} P_l(x) + \left[\sum_{k=0}^N \left(q^{(p)^2} + q^{(n)^2} \right) q^{(p)^2} q^{(n)^2} t_{km}^* r^{2k+2m-2} \right. \\ & + \left. \sum_{k=0}^N \left(q^{(p)^6} \frac{C_{1m}^{(n)}}{C_{2m}^{(n)}} \frac{1}{q^{(p)^{m+1}}} q^{(p)^{2k+2m-2}} \right. \right. \end{aligned}$$

$$\begin{aligned}
 & + q^{(n)6} \frac{C_{1m}^{(p)}}{C_{2m}^{(p)}} \frac{1}{q^{(n)m+1}} q^{(n)} r^{2k+2m-2} \Bigg) a_{km} r^{2k+2m-2} \Bigg] \\
 & \times m^2 (m+1)^2 \sum_{l=0}^{2m} C_{4l} P_l(x) \Bigg\} e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t} .
 \end{aligned} \tag{A.4}$$

The coefficients are

$$a_{km} = (-1)^k \frac{2^m (k+m)!}{k! (2k+2m+1)!} , \quad b_{km} = \sum_{l=0}^k a_{lm} a_{(k-l)m} , \tag{A.5}$$

$$d_{lkm} = (2l+m+1)(2(k-l)+m+1), \quad c_{km} = \sum_{l=0}^k d_{lkm} a_{lm} a_{(k-l)m} ,$$

$$t_{km} = \sum_{l=0}^k a_{lm} a_{(k-l)m} q^{(p)2l+m} q^{(n)2(k-l)+m} , \quad t_{km}^* = q^{(p)-1} q^{(n)-1} t_{km} ,$$

$$f_{km} = \sum_{l=0}^k d_{lkm} a_{lm} a_{(k-l)m} q^{(p)2l+m-1} q^{(n)2(k-l)+m-1} ,$$

$$C_{3l} = - \left(m^2 + m \right) (2l+1) \begin{pmatrix} m & m & l \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} m & m & l \\ -1 & 1 & 0 \end{pmatrix} ,$$

$$C_{4l} = (2l+1) \begin{pmatrix} m & m & l \\ 0 & 0 & 0 \end{pmatrix}^2 .$$

The coefficients C_{3l} and C_{4l} defined for given values of l and m contain 3-j symbols. The right-hand sides of the Poisson equations contain two sums; one of them extends to N , which is the parameter of approximation and defines the maximum index of summation. To produce the results, the value of N is always greater than 50 and ensures that maximum relative deviations of products of the functions with their derivatives from the exact expressions are $O(10^{-17})$. The other sum in the series expansion extends to $2m$ and affects the degrees of the Legendre polynomials. The terms on the right-hand sides of the Poisson equations are grouped according to the powers of qr and, due to the linearity, the particular solutions are found for every term

independently. One can write equation (A.3) as

$$\begin{aligned} \frac{\partial}{\partial r} \left(r^2 \frac{\partial \mathcal{P}_{21}}{\partial r} \right) + \frac{\partial}{\partial x} \left((1-x^2) \frac{\partial \mathcal{P}_{21}}{\partial x} \right) = & \left[\sum_{k=0}^N \sum_{l=0}^{2m} C_{2km} C_{3l} (qr)^{2k+2m+2} P_l(x) \right. \\ & - \sum_{k=0}^N \sum_{l=0}^{2m-1} (C_{1km} C_{3l} + C_{3km} C_{4l}) (qr)^{2k+2m} P_l(x) \\ & - \sum_{k=1}^N \left(C_{1km} C_{3(2m)} + C_{3km} C_{4(2m)} \right) (qr)^{2k+2m} P_{(2m)}(x) \\ & \left. - \left(C_{10m} C_{3(2m)} + C_{30m} C_{4(2m)} \right) (qr)^{2m} P_{(2m)}(x) \right] e^{-2\alpha_m t}, \end{aligned} \quad (\text{A.6})$$

where the coefficients are defined as

$$C_{1km} = C_{2m}^2 \left(q^4 c_{km} + \frac{C_{1m}}{C_{2m}} \frac{(m+1)}{q^{m-3}} (2k+m+1) a_{km} \right), \quad (\text{A.7})$$

$$C_{2km} = q^4 C_{2m}^2 b_{km}, \quad (\text{A.8})$$

$$C_{3km} = m^2 (m+1)^2 C_{2m}^2 \left(q^4 b_{km} + \frac{C_{1m}}{C_{2m}} \frac{a_{km}}{q^{m-3}} \right). \quad (\text{A.9})$$

The particular solution of the inhomogeneous equation (A.6) for the positive (p) or negative (n) time dependency is written as

$$\begin{aligned} \mathcal{P}_{21,I} = e^{-2\alpha_m t} & \left[\sum_{k=0}^N \sum_{l=0}^{2m} G_{2kl} (qr)^{2k+2m+2} P_l(x) + \sum_{k=0}^N \sum_{l=0}^{2m-1} \delta_{kl} (qr)^{2k+2m} P_l(x) \right. \\ & \left. + \sum_{k=1}^N \delta_{k(2m)} (qr)^{2k+2m} P_{(2m)}(x) + \delta_0 (qr)^{2m} \ln(r) P_{(2m)}(x) \right]. \end{aligned} \quad (\text{A.10})$$

For the whole domain of calculation, the last term in the square brackets is zero and therefore it is discarded from the further analysis. In equation

(A.10), the coefficients are defined as

$$\delta_0 = -\frac{C_{10m}C_{3(2m)} + C_{30m}C_{4(2m)}}{4m+1}, \quad (\text{A.11})$$

$$\delta_{kl} = -\frac{C_{1km}C_{3l} + C_{3km}C_{4l}}{(2k+2m)^2 + (2k+2m) - l(l+1)}, \quad (\text{A.12})$$

$$G_{2kl} = \frac{C_{2km}C_{3l}}{(2k+2m+2)^2 + (2k+2m+2) - l(l+1)}. \quad (\text{A.13})$$

Following the same approach as for the time dependencies (p) and (n), equation (A.4) for the time dependency according to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$ can be represented in the form

$$\begin{aligned} \frac{\partial}{\partial r} \left(r^2 \frac{\partial \mathcal{P}_{21}^{(pn)}}{\partial r} \right) + \frac{\partial}{\partial x} \left((1-x^2) \frac{\partial \mathcal{P}_{21}^{(pn)}}{\partial x} \right) = & \left[\sum_{k=0}^N \sum_{l=0}^{2m} C_{2km}^{(pn)} C_{3l} r^{2k+2m+2} P_l(x) \right. \\ & - \sum_{k=0}^N \sum_{l=0}^{2m-1} \left(C_{1km}^{(pn)} C_{3l} + C_{3km}^{(pn)} C_{4l} \right) r^{2k+2m} P_l(x) \\ & - \sum_{k=1}^N \left(C_{1km}^{(pn)} C_{3(2m)} + C_{3km}^{(pn)} C_{4(2m)} \right) r^{2k+2m} P_{(2m)}(x) \\ & \left. - \left(C_{10m}^{(pn)} C_{3(2m)} + C_{30m}^{(pn)} C_{4(2m)} \right) r^{2m} P_{(2m)}(x) \right] e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t}, \end{aligned} \quad (\text{A.14})$$

where the coefficients in this form of the equation are

$$\begin{aligned} C_{1km}^{(pn)} = & C_{2m}^{(p)} C_{2m}^{(n)} \left[\left(q^{(p)^2} + q^{(n)^2} \right) q^{(p)^2} q^{(n)^2} f_{km} \right. \\ & + \left(q^{(p)^6} \frac{C_{1m}^{(n)}}{C_{2m}^{(n)}} \frac{m+1}{q^{(p)^{m+1}}} q^{(p)^{2k+2m-2}} + q^{(n)^6} \frac{C_{1m}^{(p)}}{C_{2m}^{(p)}} \frac{m+1}{q^{(n)^{m+1}}} q^{(n)^{2k+2m-2}} \right) \\ & \left. \times (2k+m+1) a_{km} \right], \end{aligned} \quad (\text{A.15})$$

$$C_{2km}^{(pn)} = 2C_{2m}^{(p)} C_{2m}^{(n)} q^{(p)3} q^{(n)3} t_{km}, \quad (\text{A.16})$$

$$\begin{aligned} C_{3km}^{(pn)} = & C_{2m}^{(p)} C_{2m}^{(n)} m^2 (m+1)^2 \left[\left(q^{(p)2} + q^{(n)2} \right) q^{(p)2} q^{(n)2} t_{km}^* \right. \\ & + \left(q^{(p)6} \frac{C_{1m}^{(n)}}{C_{2m}^{(n)}} \frac{1}{q^{(p)m+1}} q^{(p)2k+2m-2} \right. \\ & \left. \left. + q^{(n)6} \frac{C_{1m}^{(p)}}{C_{2m}^{(p)}} \frac{1}{q^{(n)m+1}} q^{(n)2k+2m-2} \right) a_{km} \right]. \end{aligned} \quad (\text{A.17})$$

The particular solution of the inhomogeneous equation (A.14) reads

$$\begin{aligned} \mathcal{P}_{21,I}^{(pn)} = & \left[\sum_{k=0}^N \sum_{l=0}^{2m} G_{2kl}^{(pn)} r^{2k+2m+2} P_l(x) + \sum_{k=0}^N \sum_{l=0}^{2m-1} \delta_{kl}^{(pn)} r^{2k+2m} P_l(x) \right. \\ & \left. + \sum_{k=1}^N \delta_{k(2m)}^{(pn)} r^{2k+2m} P_{(2m)}(x) + \delta_0^{(pn)} r^{2m} \log(r) P_{(2m)}(x) \right] e^{-\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)t}, \end{aligned} \quad (\text{A.18})$$

with the coefficients

$$\delta_0^{(pn)} = -\frac{C_{10m}^{(pn)} C_{3(2m)} + C_{30m}^{(pn)} C_{4(2m)}}{4m+1}, \quad (\text{A.19})$$

$$\delta_{kl}^{(pn)} = -\frac{C_{1km}^{(pn)} C_{3l} + C_{3km}^{(pn)} C_{4l}}{(2k+2m)^2 + (2k+2m) - l(l+1)}, \quad (\text{A.20})$$

$$G_{2kl}^{(pn)} = \frac{C_{2km}^{(pn)} C_{3l}}{(2k+2m+2)^2 + (2k+2m+2) - l(l+1)}. \quad (\text{A.21})$$

Throughout the domain of calculation the last term of equation (A.18) is zero. It is therefore discarded from further analysis.

The pressure contributions $p_{21}^{(n)}$ and $p_{21}^{(pn)}$ found by this analysis read

$$\begin{aligned}
 p_{21}^{(n)}(r, \theta, t) = & -e^{-2\alpha_m^{(n)}t} \left\{ \sum_{l=0}^L D_{21l}^{(n)} r^l P_l(x) - \left[\sum_{k=0}^N \sum_{l=0}^{2m} G_{2kl}^{(n)} \left(q^{(n)} r \right)^{2k+2m+2} P_l(x) \right. \right. \\
 & + \left. \sum_{k=0}^N \sum_{l=0}^{2m-1} \delta_{kl}^{(n)} \left(q^{(n)} r \right)^{2k+2m} P_l(x) + \sum_{k=1}^N \delta_{k(2m)}^{(n)} \left(q^{(n)} r \right)^{2k+2m} P_{(2m)}(x) \right] \Big\} \\
 & - \frac{1}{2} \left(\hat{\eta}_1^{(n)} \right)^2 \alpha_m^{(n)2} \left[f_r^{(n)2}(r) P_m(\cos \theta)^2 + f_\theta^{(n)2}(r) P_m^1(\cos \theta)^2 \right] e^{-2\alpha_m^{(n)}t},
 \end{aligned} \tag{A.22}$$

$$\begin{aligned}
 p_{21}^{(pn)}(r, \theta, t) = & - \left\{ \sum_{l=0}^L D_{21l}^{(pn)} r^l P_l(x) - \left[\sum_{k=0}^N \sum_{l=0}^{2m} G_{2kl}^{(pn)} r^{2k+2m+2} P_l(x) \right. \right. \\
 & + \left. \sum_{k=0}^N \sum_{l=0}^{2m-1} \delta_{kl}^{(pn)} r^{2k+2m} P_l(x) + \sum_{k=1}^N \delta_{k(2m)}^{(pn)} r^{2k+2m} P_{(2m)}(x) \right] \Big\} e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t} \\
 & - \hat{\eta}_1^{(p)} \hat{\eta}_1^{(n)} \alpha_m^{(p)} \alpha_m^{(n)} \left[f_r^{(p)}(r) f_r^{(n)}(r) P_m(\cos \theta)^2 \right. \\
 & + \left. f_\theta^{(p)}(r) f_\theta^{(n)}(r) P_m^1(\cos \theta)^2 \right] e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t}.
 \end{aligned} \tag{A.23}$$

C. Solutions for the radial velocity u_{r21}

The radial momentum equations for all the time dependencies read

$$\begin{aligned}
 r^2 \frac{\partial^2 u_{r21}^{(p)}}{\partial r^2} + 4r \frac{\partial u_{r21}^{(p)}}{\partial r} + 2 \left(1 + q^{(p)2} r^2 \right) u_{r21}^{(p)} + \frac{\partial}{\partial x} \left(\left(1 - x^2 \right) \frac{\partial u_{r21}^{(p)}}{\partial x} \right) = \\
 = \frac{r^2}{Oh} \frac{\partial \mathcal{P}_{21}^{(p)}}{\partial r} + \frac{q^{(p)2} C_{2m}^{(p)2}}{Oh} \left[\frac{C_{1m}^{(p)}}{C_{2m}^{(p)}} (m+1) (q^{(p)} r j_m^{(p)}) r^m \right. \\
 \left. + (q^{(p)} r j_m^{(p)}) \frac{\partial (q^{(p)} r j_m^{(p)})}{\partial r} \right] \times \left(\frac{dP_m(x)}{dx} \right)^2 (1 - x^2) e^{-2\alpha_m^{(p)}t},
 \end{aligned} \tag{A.24}$$

$$\begin{aligned}
 r^2 \frac{\partial^2 u_{r21}^{(n)}}{\partial r^2} + 4r \frac{\partial u_{r21}^{(n)}}{\partial r} + 2 \left(1 + q^{(n)2} r^2\right) u_{r21}^{(n)} + \frac{\partial}{\partial x} \left(\left(1 - x^2\right) \frac{\partial u_{r21}^{(n)}}{\partial x} \right) = \\
 = \frac{r^2}{Oh} \frac{\partial \mathcal{P}_{21}^{(n)}}{\partial r} + \frac{q^{(n)2} C_{2m}^{(n)2}}{Oh} \left[\frac{C_{1m}^{(n)}}{C_{2m}^{(n)}} (m+1) (q^{(n)} r j_m^{(n)}) r^m \right. \\
 \left. + (q^{(n)} r j_m^{(n)}) \frac{\partial (q^{(n)} r j_m^{(n)})}{\partial r} \right] \times \left(\frac{dP_m(x)}{dx} \right)^2 (1 - x^2) e^{-2\alpha_m^{(n)} t}, \quad (A.25)
 \end{aligned}$$

$$\begin{aligned}
 r^2 \frac{\partial^2 u_{r21}^{(pn)}}{\partial r^2} + 4r \frac{\partial u_{r21}^{(pn)}}{\partial r} + \left(2 + q^{(pn)2} r^2\right) u_{r21}^{(pn)} + \frac{\partial}{\partial x} \left(\left(1 - x^2\right) \frac{\partial u_{r21}^{(pn)}}{\partial x} \right) = \\
 = \frac{r^2}{Oh} \frac{\partial \mathcal{P}_{21}^{(pn)}}{\partial r} + \left\{ \frac{q^{(p)2} C_{2m}^{(p)}}{Oh} \left[C_{1m}^{(n)} (m+1) (q^{(p)} r j_m^{(p)}) r^m \right. \right. \\
 \left. + C_{2m}^{(n)} (q^{(p)} r j_m^{(p)}) \frac{\partial (q^{(p)} r j_m^{(p)})}{\partial r} \right] + \frac{q^{(n)2} C_{2m}^{(n)}}{Oh} \left[C_{1m}^{(p)} (m+1) (q^{(n)} r j_m^{(n)}) r^m \right. \\
 \left. + C_{2m}^{(p)} (q^{(n)} r j_m^{(n)}) \frac{\partial (q^{(n)} r j_m^{(n)})}{\partial r} \right] \left. \right\} \left(\frac{dP_m(x)}{dx} \right)^2 (1 - x^2) e^{-(\alpha_m^{(p)} + \alpha_m^{(n)}) t}, \quad (A.26)
 \end{aligned}$$

where we define $q^{(pn)2} = q^{(p)2} + q^{(n)2}$. The series expansions approximate the terms with products of spherical Bessel functions and their derivatives on the right-hand sides. The series expansions in Legendre polynomials approximate the products of Legendre polynomials under the derivations following the work by Dong and Lemus (2002). For every time dependency, we introduce the definitions for the modified pressure. The approximated right-hand side of the radial momentum equation for the positive (p) or negative (n) time dependencies reads

$$\begin{aligned}
 -\frac{r^2}{Oh} \frac{\partial u_{r21}}{\partial t} + \frac{\partial^2}{\partial r^2} \left(r^2 u_{r21} \right) + \frac{\partial}{\partial x} \left(\left(1 - x^2\right) \frac{\partial u_{r21}}{\partial x} \right) = - \left\{ \sum_{l=0}^L \frac{D_{21l} l}{Oh} r^{l+1} P_l(x) \right. \\
 \left. + \sum_{k=0}^N \sum_{l=0}^{2m} \left[\epsilon_{km} C_{3l} (qr)^{2k+2m+1} + \frac{(2k+2m+2) G_{2kl}}{q^{(p)} Oh} (qr)^{2k+2m+3} \right] P_l(x) \right\}
 \end{aligned}$$

$$\begin{aligned}
 & + \sum_{k=0}^N \sum_{l=0}^{2m-1} \frac{(2k+2m)\delta_{kl}}{qOh} (qr)^{2k+2m+1} P_l(x) \\
 & + \sum_{k=1}^N \frac{(2k+2m)\delta_{k(2m)}}{qOh} (qr)^{2k+2m+1} P_{(2m)}(x) \Big\} e^{-2\alpha_m t} ,
 \end{aligned} \tag{A.27}$$

where for the positive (p) or negative (n) time dependencies the new coefficients ϵ_{km} are introduced as

$$\epsilon_{km} = C_{4km} + C_{5km} , \quad C_{4km} = \frac{q^{2-m}}{Oh} C_{1m} C_{2m} (m+1) a_{km} , \tag{A.28}$$

$$C_{5km} = \frac{q^3}{Oh} C_{2m}^2 \sum_{l=0}^k (2(k-l) + m + 1) a_{lm} a_{(k-l)m} . \tag{A.29}$$

For the time dependency according to $\exp[(\alpha_m^{(p)} + \alpha_m^{(n)})t]$ one can define the approximated right-hand side of the radial momentum equation as

$$\begin{aligned}
 & r^2 \frac{\partial^2 u_{r21}^{(pn)}}{\partial r^2} + 4r \frac{\partial u_{r21}^{(pn)}}{\partial r} + u_{r21}^{(pn)} \left(2 + q^{(pn)^2} r^2 \right) + \frac{\partial}{\partial x} \left((1-x^2) \frac{\partial u_{r21}^{(pn)}}{\partial x} \right) = \\
 & = - \sum_{l=0}^L \frac{ID_{21l}^{(pn)}}{Oh} r^{l+1} P_l(x) e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t} \\
 & + \left[\sum_{k=0}^N \sum_{l=0}^{2m} \left(\epsilon_{km}^{(pn)} C_{3l} r^{2k+2m+1} + \frac{(2k+2m+2) G_{2kl}^{(pn)}}{Oh} r^{2k+2m+3} \right) P_l(x) \right. \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m-1} \frac{(2k+2m) \delta_{kl}^{(pn)}}{Oh} r^{2k+2m+1} P_l(x) \\
 & \left. + \sum_{k=1}^N \frac{(2k+2m) \delta_{k(2m)}^{(pn)}}{Oh} r^{2k+2m+1} P_{(2m)}(x) \right] e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t} ,
 \end{aligned} \tag{A.30}$$

with the new coefficients $\epsilon_{km}^{(pn)}$ as

$$\epsilon_{km}^{(pn)} = C_{4km}^{(pn)} + C_{5km}^{(pn)} , \tag{A.31}$$

$$C_{4km}^{(pn)} = a_{km} (m+1) \left(\frac{q^{(p)2-m}}{Oh} C_{1m}^{(n)} C_{2m}^{(p)} (q^{(p)})^{2k+2m+1} + \frac{q^{(n)2-m}}{Oh} C_{1m}^{(p)} C_{2m}^{(n)} (q^{(n)})^{2k+2m+1} \right), \quad (\text{A.32})$$

$$C_{5km}^{(pn)} = \frac{C_{2m}^{(p)} C_{2m}^{(n)}}{Oh} \sum_{l=0}^k (2(k-l) + m + 1) a_{lm} a_{(k-l)m} \times \left(q^{(p)2l+m+3} q^{(n)2(k-l)+m+1} + q^{(p)2(k-l)+m+1} q^{(n)2l+m+3} \right). \quad (\text{A.33})$$

The solutions u_{r21} for the time dependencies (n) and (pn) have the forms

$$\begin{aligned} u_{r21}^{(n)}(r, \theta, t) &= - \sum_{l=0}^L \left[A_{21l}^{(n)} r^{l-1} + B_{21l}^{(n)} q^{(n)2} \frac{j_l(\sqrt{2}q^{(n)}r)}{q^{(n)}r} \right] l(l+1) P_l(x) e^{-2\alpha_m^{(n)}t} \\ &+ \left\{ \sum_{k=0}^N \sum_{l=0}^{2m} \left[K_{1kl}^{(n)} (q^{(n)}r)^{2k+2m+1} + K_{2kl}^{(n)} (q^{(n)}r)^{2k+2m+3} \right] P_l(x) \right. \\ &+ \sum_{k=0}^N \sum_{l=0}^{2m-1} K_{3kl}^{(n)} (q^{(n)}r)^{2k+2m+1} P_l(x) \\ &\left. + \sum_{k=1}^N K_{3k(2m)}^{(n)} (q^{(n)}r)^{2k+2m+1} P_{(2m)}(x) \right\} e^{-2\alpha_m^{(n)}t} \\ &= - \sum_{l=0}^L \left(A_{21l}^{(n)} f_{ar}^{(n)} + B_{21l}^{(n)} f_{br}^{(n)} \right) P_l(\cos \theta) e^{-2\alpha_m^{(n)}t} + A p_{r21}^{(n)}(r, \theta, t), \end{aligned} \quad (\text{A.34})$$

$$\begin{aligned} u_{r21}^{(pn)}(r, \theta, t) &= - \sum_{l=0}^L \left[A_{21l}^{(pn)} r^{l-1} + B_{21l}^{(pn)} \frac{j_l(Q^{(pn)}r)}{r} \right] l(l+1) P_l(x) e^{-\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)t} \\ &+ e^{-\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)t} \left\{ \sum_{k=0}^N \sum_{l=0}^{2m} \left[K_{1kl}^{(pn)} r^{2k+2m+1} + K_{2kl}^{(pn)} r^{2k+2m+3} \right] P_l(x) \right. \\ &\left. + \sum_{k=0}^N \sum_{l=0}^{2m-1} K_{3kl}^{(pn)} r^{2k+2m+1} P_l(x) + \sum_{k=1}^N K_{3k(2m)}^{(pn)} r^{2k+2m+1} P_{(2m)}(x) \right\} \end{aligned}$$

$$\begin{aligned}
 &= - \sum_{l=0}^L \left(A_{21l}^{(pn)} f_{ar}^{(pn)} + B_{21l}^{(pn)} f_{br}^{(pn)} \right) P_l(\cos \theta) e^{-\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)t} \\
 &+ A p_{r21}^{(pn)}(r, \theta, t).
 \end{aligned} \tag{A.35}$$

For every time dependency, the solutions contain two terms with the unknown coefficients A_{21l} and B_{21l} in a sum with respect to the index l . The general solutions of the homogeneous forms of equations (A.24)-(A.26) are represented with the first term that contains a sum, while $A p_{r21}^{(pn)}(r, \theta, t)$ symbolizes the corresponding particular solutions of the inhomogeneous equations. The relations between the coefficients A_{21l} and the coefficients D_{21l} in equations (3.49), (A.22) and (A.23) are

$$A_{21l}^{(p)} = \frac{D_{21l}^{(p)}}{2\alpha_m^{(p)}(l+1)}, \quad A_{21l}^{(n)} = \frac{D_{21l}^{(n)}}{2\alpha_m^{(n)}(l+1)}, \quad A_{21l}^{(pn)} = \frac{D_{21l}^{(pn)}}{\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)(l+1)}. \tag{A.36}$$

It is necessary to determine unknown coefficients for other particular solutions of the radial velocity. We substitute u_{r21} in the Poisson equation for the radial velocity and compare the unknown coefficients on the left-hand side with the known coefficients on the right-hand side of the equation. For the positive or negative time dependency, the comparison yields recursive relations in the coefficients, which are written

$$\begin{aligned}
 K_{10l} &= \frac{\epsilon_{0m} C_{3l}}{2m(2m+1) + 4(2m+1) + 2 - l(l+1)}, \\
 K_{1kl} &= \frac{\epsilon_{km} C_{3l} - 2K_{1(k-1)l}}{(2k+2m)(2k+2m+1) + 4(2k+2m+1) + 2 - l(l+1)}, \\
 K_{20l} &= \frac{(2m+2)G_{20l}}{qOh[(2m+2)(2m+3) + 4(2m+3) + 2 - l(l+1)]}, \\
 K_{2kl} &= \frac{(2k+2m+2)G_{2kl} - 2qOhK_{2(k-1)l}}{qOh[(2k+2m+2)(2k+2m+3) + 4(2k+2m+3) + 2 - l(l+1)]}, \\
 K_{30l} &= \frac{2m\delta_{0l}}{qOh[2m(2m+1) + 4(2m+1) + 2 - l(l+1)]}, \\
 K_{3kl} &= \frac{(2k+2m)\delta_{kl} - 2qOhK_{3(k-1)l}}{qOh[(2k+2m)(2k+2m+1) + 4(2k+2m+1) + 2 - l(l+1)]},
 \end{aligned}$$

$$\begin{aligned}
 K_{31(2m)} &= \frac{2m\delta_{1(2m)}}{qOh [4(2m+1)+2]} , \\
 K_{3k(2m)} &= \frac{(2k+2m)\delta_{k(2m)} - 2qOh K_{3(k-1)(2m)}}{qOh [2k(2k+2m+1) + 4mk + 4(2k+2m+1) + 2]} .
 \end{aligned} \tag{A.37}$$

For the time dependency according to $\exp[(\alpha_m^{(p)} + \alpha_m^{(n)})t]$ the comparison yields

$$\begin{aligned}
 K_{10l}^{(pn)} &= \frac{\epsilon_{0m}^{(pn)} C_{3l}}{2m(2m+1) + 4(2m+1) + 2 - l(l+1)} , \\
 K_{1kl}^{(pn)} &= \frac{\epsilon_{km}^{(pn)} C_{3l} - Q^{(pn)^2} K_{1(k-1)l}^{(p)}}{(2k+2m)(2k+2m+1) + 4(2k+2m+1) + 2 - l(l+1)} , \\
 K_{20l}^{(pn)} &= \frac{(2m+2)G_{20l}^{(pn)}}{Oh [(2m+2)(2m+3) + 4(2m+3) + 2 - l(l+1)]} , \\
 K_{2kl}^{(pn)} &= \frac{(2k+2m+2)G_{2kl}^{(pn)} - 2Oh K_{2(k-1)l}^{(pn)}}{Oh [(2k+2m+2)(2k+2m+3) + 4(2k+2m+3) + 2 - l(l+1)]} , \\
 K_{30l}^{(pn)} &= \frac{2m\delta_{0l}^{(pn)}}{Oh [2m(2m+1) + 4(2m+1) + 2 - l(l+1)]} , \\
 K_{3kl}^{(pn)} &= \frac{(2k+2m)\delta_{kl}^{(p)} - 2Oh K_{3(k-1)l}^{(p)}}{Oh [(2k+2m)(2k+2m+1) + 4(2k+2m+1) + 2 - l(l+1)]} , \\
 K_{31(2m)}^{(p)} &= \frac{2m\delta_{1(2m)}^{(pn)}}{Oh [4(2m+1) + 2]} , \\
 K_{3k(2m)}^{(pn)} &= \frac{(2k+2m)\delta_{k(2m)}^{(pn)} - 2Oh K_{3(k-1)(2m)}^{(pn)}}{Oh [2k(2k+2m+1) + 4mk + 4(2k+2m+1) + 2]} .
 \end{aligned} \tag{A.38}$$

D. Solutions for the polar velocity $u_{\theta 21}$

The solutions $u_{\theta 21}$ for the negative time dependency (n) and the time dependency (pn) are

$$\begin{aligned}
 u_{\theta 21}^{(n)}(r, \theta, t) = & - \left\{ \sum_{l=0}^L \left[A_{21l}^{(n)} (l+1) r^{l-1} \right. \right. \\
 & + B_{21l}^{(n)} q^{(n)2} \left((l+1) \frac{j_l(\sqrt{2}q^{(n)}r)}{q^{(n)}r} - \sqrt{2}j_{l+1}(\sqrt{2}q^{(n)}r) \right) \left. \right] P_l^1(x) \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m} \left[L_{1kl}^{(n)} (q^{(n)}r)^{2k+2m+1} + L_{2kl}^{(n)} (q^{(n)}r)^{2k+2m+3} \right] \frac{P_{l+1}(x) - P_{l-1}(x)}{\sqrt{1-x^2}} \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m-1} L_{3kl}^{(n)} (q^{(n)}r)^{2k+2m+1} \frac{P_{l+1}(x) - P_{l-1}(x)}{\sqrt{1-x^2}} \\
 & + \sum_{k=1}^N L_{3k(2m)}^{(n)} (q^{(n)}r)^{2k+2m+1} \frac{P_{(2m+1)}(x) - P_{(2m-1)}(x)}{\sqrt{1-x^2}} \left. \right\} e^{-2\alpha_m^{(n)}t} \\
 = & - \sum_{l=0}^L \left(A_{21l}^{(n)} f_{a\theta}^{(n)} + B_{21l}^{(n)} f_{b\theta}^{(n)} \right) P_l^1(\cos \theta) e^{-2\alpha_m^{(n)}t} + A p_{\theta 21}^{(n)}(r, \theta, t),
 \end{aligned} \tag{A.39}$$

$$\begin{aligned}
 u_{\theta 21}^{(pn)}(r, \theta, t) = & - \left\{ \sum_{l=0}^L \left[A_{21l}^{(pn)} (l+1) r^{l-1} \right. \right. \\
 & + B_{21l}^{(pn)} \left((l+1) \frac{j_l(Q^{(pn)}r)}{r} - Q^{(pn)} j_{l+1}(Q^{(pn)}r) \right) \left. \right] P_l^1(x) \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m} \left[L_{1kl}^{(pn)} r^{2k+2m+1} + L_{2kl}^{(pn)} r^{2k+2m+3} \right] \frac{P_{l+1}(x) - P_{l-1}(x)}{\sqrt{1-x^2}} \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m-1} L_{3kl}^{(pn)} r^{2k+2m+1} \frac{P_{l+1}(x) - P_{l-1}(x)}{\sqrt{1-x^2}} \\
 & + \sum_{k=1}^N L_{3k(2m)}^{(pn)} r^{2k+2m+1} \frac{P_{(2m+1)}(x) - P_{(2m-1)}(x)}{\sqrt{1-x^2}} \left. \right\} e^{-(\alpha_m^{(p)} + \alpha_m^{(n)})t}
 \end{aligned}$$

$$\begin{aligned}
 &= - \sum_{l=0}^L \left(A_{21l}^{(pn)} f_{a\theta}^{(pn)} + B_{21l}^{(pn)} f_{b\theta}^{(pn)} \right) P_l^1(\cos \theta) e^{-\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)t} \\
 &+ A p_{\theta 21}^{(pn)}(r, \theta, t) .
 \end{aligned} \tag{A.40}$$

Defined for each oscillation frequency, the functions f_{ar} , f_{br} , $f_{a\theta}$ and $f_{b\theta}$ are introduced to keep the expressions for the flow quantities compact. $A p_r$ and $A p_\theta$ for all time dependencies denote the particular solutions defined as such to facilitate the expressions in the section of the coefficients for the second-order solutions. For the positive (p) or negative (n) time dependency, the new coefficients in the particular solutions for the polar velocity component are

$$L_{1kl} = \frac{K_{1kl} (2k + 2m + 3)}{1 - 2l} , \quad L_{2kl} = \frac{K_{2kl} (2k + 2m + 5)}{1 - 2l} , \tag{A.41}$$

$$L_{3kl} = \frac{K_{3kl} (2k + 2m + 3)}{1 - 2l} , \quad L_{3k(2m)} = \frac{K_{3k(2m)} (2k + 2m + 3)}{1 - 4m} , \tag{A.42}$$

and for the time dependency related to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$

$$L_{1kl}^{(pn)} = \frac{K_{1kl}^{(pn)} (2k + 2m + 3)}{1 - 2l} , \quad L_{2kl}^{(pn)} = \frac{K_{2kl}^{(pn)} (2k + 2m + 5)}{1 - 2l} , \tag{A.43}$$

$$L_{3kl}^{(pn)} = \frac{K_{3kl}^{(pn)} (2k + 2m + 3)}{1 - 2l} , \quad L_{3k(2m)}^{(pn)} = \frac{K_{3k(2m)}^{(pn)} (2k + 2m + 3)}{1 - 4m} . \tag{A.44}$$

E. Solutions for the polar velocity $u_{\theta 22}$ and pressure p_{22}

The solutions for the pressure p_{22} and the angular velocity component $u_{\theta 22}$ read

$$\begin{aligned}
 u_{\theta 22}(r, \theta, t) = & - \sum_{k=0}^K \frac{\hat{\eta}_{22k}^{(p)} \alpha_{2k}^{(p)}}{k} e^{-\alpha_{2k}^{(p)} t} \left[\left(1 + \frac{2(k^2 - 1)}{2q_{2k}^{(p)} Q_k^{(p)} - q_{2k}^{(p)2}} \right) r^{k-1} \right. \\
 & - \frac{2(k-1)q_{k2}^{(p)}}{2q_{k2}^{(p)} Q_k^{(p)} - q_{k2}^{(p)2}} \left((k+1) \frac{j_k(q_{k2}^{(p)} r)}{j_k(q_{k2}^{(p)})} \frac{1}{q_{k2}^{(p)} r} - \frac{j_{k+1}(q_{k2}^{(p)} r)}{j_k(q_{k2}^{(p)})} \right) \Big] P_k^1(\cos \theta) \\
 & - \sum_{k=0}^K \frac{\hat{\eta}_{22k}^{(n)} \alpha_{2k}^{(n)}}{k} e^{-\alpha_{2k}^{(n)} t} \left[\left(1 + \frac{2(k^2 - 1)}{2q_{2k}^{(n)} Q_k^{(n)} - q_{2k}^{(n)2}} \right) r^{k-1} \right. \\
 & - \frac{2(k-1)q_{2k}^{(n)}}{2q_{2k}^{(n)} Q_k^{(n)} - q_{2k}^{(n)2}} \left((k+1) \frac{j_k(q_{2k}^{(n)} r)}{j_k(q_{2k}^{(n)})} \frac{1}{q_{2k}^{(n)} r} - \frac{j_{k+1}(q_{2k}^{(n)} r)}{j_k(q_{2k}^{(n)})} \right) \Big] P_k^1(\cos \theta) \\
 & = - \sum_{k=0}^K \left[\hat{\eta}_{22k}^{(p)} \alpha_{2k}^{(p)} e^{-\alpha_{2k}^{(p)} t} f_{2\theta}^{(p)}(r) + \hat{\eta}_{22k}^{(n)} \alpha_{2k}^{(n)} e^{-\alpha_{2k}^{(n)} t} f_{2\theta}^{(n)}(r) \right] P_k^1(\cos \theta) ,
 \end{aligned} \tag{A.45}$$

$$\begin{aligned}
 p_{22}(r, \theta, t) = & - \sum_{k=0}^K \frac{r^k}{k} \left[\hat{\eta}_{22k}^{(p)} \alpha_{2k}^{(p)2} e^{-\alpha_{2k}^{(p)} t} \left(1 + \frac{2(k^2 - 1)}{2q_{2k}^{(p)} Q_k^{(p)} - q_{2k}^{(p)2}} \right) \right. \\
 & \left. + \hat{\eta}_{22k}^{(n)} \alpha_{2k}^{(n)2} e^{-\alpha_{2k}^{(n)} t} \left(1 + \frac{2(k^2 - 1)}{2q_{2k}^{(n)} Q_k^{(n)} - q_{2k}^{(n)2}} \right) \right] P_k(\cos \theta) .
 \end{aligned} \tag{A.46}$$

Defined for each oscillation frequency, the function $f_{2\theta}$ is introduced to keep the expressions of the flow field quantities compact.

F. The second-order coefficients

For every l and each time dependency, three unknown coefficients A_{21l} , B_{21l} , H_{21l} have to be deduced to fully determine the contribution "21" of the

second-order solutions. In order to do this we consider the three boundary conditions (3.13)-(3.15), and we substitute the first-order solutions and the second-order solutions with subscript 21 with the forms of the eigenvalues (p) or (n) into the kinematic boundary condition. It yields

$$\begin{aligned} & \sum_{l=0}^L (A_{21l} f_{ar} + B_{21l} f_{br} + 2\alpha_m H_{21l}) \int_{-1}^1 P_l(x) P_j(x) dx = \\ & = \hat{\eta}_1^2 \alpha_m \left[\frac{f_\theta}{r} \int_{-1}^1 P_j(x) P_m^1(x)^2 dx - f_r' \int_{-1}^1 P_j(x) P_m(x)^2 dx \right] \\ & - \int_{-1}^1 A p_{r21} P_j(x) dx, \end{aligned} \quad (\text{A.47})$$

where the derivatives with respect to the radial coordinate are denoted with primes. For the time dependency related to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$, the corresponding equation reads

$$\begin{aligned} & \sum_{l=0}^L \left[A_{21l}^{(pn)} f_{ar}^{(pn)} + B_{21l}^{(pn)} f_{br}^{(pn)} + (\alpha_m^{(p)} + \alpha_m^{(n)}) H_{21l}^{(pn)} \right] \int_{-1}^1 P_l(x) P_j(x) dx = \\ & \hat{\eta}_1^{(p)} \hat{\eta}_1^{(n)} \left(\alpha_m^{(p)} \frac{f_\theta^{(p)}}{r} + \alpha_m^{(n)} \frac{f_\theta^{(n)}}{r} \right) \int_{-1}^1 P_j(x) P_m^1(x)^2 dx \\ & - \hat{\eta}_1^{(p)} \hat{\eta}_1^{(n)} \left(\alpha_m^{(p)} f_r^{(p)'} + \alpha_m^{(n)} f_r^{(n)'} \right) \int_{-1}^1 P_j(x) P_m(x)^2 dx \\ & - \int_{-1}^1 A p_{r21}^{(pn)} P_j(x) dx. \end{aligned} \quad (\text{A.48})$$

The kinematic boundary condition for the three different time dependencies is defined with these two equations. For the eigenvalue (p) or (n), the second-order zero shear stress boundary condition reads

$$\begin{aligned} & \sum_{l=0}^L r \left[A_{21l} \left(\frac{f_{a\theta}}{r} \right)' + B_{21l} \left(\frac{f_{b\theta}}{r} \right)' + A_{21l} \frac{f_{ar}}{r^2} + B_{21l} \frac{f_{br}}{r^2} \right] \int_{-1}^1 P_l^1(x) P_j^1(x) dx \\ & = -\hat{\eta}_1^2 \alpha_m \left[\left(r \left(\frac{f_\theta}{r} \right)' \right)' + 3 \left(\frac{f_r}{r} \right)' + 2m(m+1) \frac{f_\theta}{r^2} \right] \int_{-1}^1 P_j^1(x) P_m(x) P_m^1(x) dx \end{aligned}$$

$$\begin{aligned}
 & + \hat{\eta}_1^2 \alpha_m \frac{f_\theta}{r^2} \left(\int_{-1}^1 P_j^1(x) P_{m-1}(x)^2 P_m^1(x) P_1(x) dx \right. \\
 & + m(m+1) \int_{-1}^1 P_j^1(x) P_m^1(x) P_{m-1}(x) P_1(x) dx \Big) \\
 & - \int_{-1}^1 \left(r \left(\frac{A p_{\theta 21}}{r} \right)' + \frac{B p_{r 21}}{r} \right) P_j^1(x) dx ,
 \end{aligned} \tag{A.49}$$

while the time dependency related to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$ is written

$$\begin{aligned}
 & \sum_{l=0}^L r \left[A_{21l}^{(pn)} \left(\frac{f_{a\theta}^{(pn)}}{r} \right)' + B_{21l}^{(pn)} \left(\frac{f_{b\theta}^{(pn)}}{r} \right)' + A_{21l}^{(pn)} \frac{f_{ar}^{(pn)}}{r^2} + B_{21l}^{(pn)} \frac{f_{br}^{(pn)}}{r^2} \right] \\
 & \times \int_{-1}^1 P_l^1(x) P_j^1(x) dx = -\hat{\eta}_1^{(p)} \hat{\eta}_1^{(n)} \left\{ \left[\alpha_m^{(p)} \left(r \left(\frac{f_\theta^{(p)}}{r} \right)' \right)' \right. \right. \\
 & + \alpha_m^{(n)} \left(r \left(\frac{f_\theta^{(n)}}{r} \right)' \right)' \Big] + 3 \left[\alpha_m^{(p)} \left(\frac{f_r^{(p)}}{r} \right)' + \alpha_m^{(n)} \left(\frac{f_r^{(n)}}{r} \right)' \right] \\
 & + 2m(m+1) \left[\alpha_m^{(p)} \frac{f_\theta^{(p)}}{r^2} + \alpha_m^{(n)} \frac{f_\theta^{(n)}}{r^2} \right] \Big\} \int_{-1}^1 P_j^1(x) P_m(x) P_m^1(x) dx \\
 & + \hat{\eta}_1^{(p)} \hat{\eta}_1^{(n)} \left[\alpha_m^{(p)} \frac{f_\theta^{(p)}}{r^2} + \alpha_m^{(n)} \frac{f_\theta^{(n)}}{r^2} \right] \left(\int_{-1}^1 P_j^1(x) P_{m-1}^2(x) P_m^1(x) P_1(x) dx \right. \\
 & + m(m+1) \int_{-1}^1 P_j^1(x) P_m^1(x) P_{m-1}(x) P_1(x) dx \Big) \\
 & - \int_{-1}^1 \left(r \left(\frac{A p_{\theta 21}^{(pn)}}{r} \right)' + \frac{B p_{r 21}^{(pn)}}{r} \right) P_j^1(x) dx ,
 \end{aligned} \tag{A.50}$$

where the definition of the $B p_{r 21}$ and $B p_{r 21}^{(pn)}$ is

$$\begin{aligned}
 B p_{r 21} = e^{-2\alpha_m t} & \left\{ \sum_{k=0}^N \sum_{l=0}^{2m} \left[K_{1kl}(qr)^{2k+2m+1} + K_{2kl}(qr)^{2k+2m+3} \right] P_l^1(x) \right. \\
 & + \sum_{k=0}^N \sum_{l=0}^{2m-1} K_{3kl}(qr)^{2k+2m+1} P_l^1(x) + \sum_{k=1}^N K_{3k(2m)}(qr)^{2k+2m+1} P_{(2m)}^1(x) \Big\} ,
 \end{aligned} \tag{A.51}$$

$$\begin{aligned}
 Bp_{r21}^{(pn)} = e^{-\left(\alpha_m^{(p)} + \alpha_m^{(n)}\right)t} & \left\{ \sum_{k=0}^N \sum_{l=0}^{2m} \left[K_{1kl}^{(pn)} r^{2k+2m+1} + K_{2kl}^{(pn)} r^{2k+2m+3} \right] P_l^1(x) \right. \\
 & \left. + \sum_{k=0}^N \sum_{l=0}^{2m-1} K_{3kl}^{(pn)} r^{2k+2m+1} P_l^1(x) + \sum_{k=1}^N K_{3k(2m)}^{(pn)} r^{2k+2m+1} P_{(2m)}^1(x) \right\}.
 \end{aligned} \tag{A.52}$$

The second-order zero normal stress boundary condition for the eigenvalue (p) or (n) is defined as

$$\begin{aligned}
 & \sum_{l=0}^L \left[2A_{21l} \alpha_m(l+1)r^l + 2Oh \left(A_{21l} f'_{ar} + B_{21l} f'_{br} \right) + (l(l+1) - 2) H_{21l} \right] \\
 & \times \int_{-1}^1 P_l(x) P_j(x) dx = \hat{\eta}_1^2 \left[\alpha_m^2 f'_p - 2\alpha_m Oh f''_r + 2(m(m+1) - 1) \right. \\
 & \left. - \frac{1}{2} \alpha_m f_r^2 \right] \int_{-1}^1 P_j(x) P_m^1(x)^2 dx + \hat{\eta}_1^2 \left[2Oh \alpha_m \left(\left(\frac{f_\theta}{r} \right)' + \frac{f_r}{r^2} \right) \right. \\
 & \left. - \frac{1}{2} \alpha_m^2 f_\theta^2 \right] \int_{-1}^1 P_j(x) P_m(x)^2 dx + \int_{-1}^1 (\mathcal{P}_{21,l} - 2Oh A p_{r21}') P_j(x) dx,
 \end{aligned} \tag{A.53}$$

where for the time dependency according to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$ it reads

$$\begin{aligned}
 & \sum_{l=0}^L \left[A_{21l}^{(pn)} (\alpha_m^{(p)} + \alpha_m^{(n)}) (l+1) r^l + 2Oh \left(A_{21l}^{(pn)} f_{ar}^{(pn)'} + B_{21l}^{(pn)} f_{br}^{(pn)'} \right) \right. \\
 & \left. + (l(l+1) - 2) H_{21l}^{(pn)} \right] \int_{-1}^1 P_l(x) P_j(x) dx = \hat{\eta}_1^{(p)} \hat{\eta}_1^{(n)} \left\{ 4(m(m+1) - 1) \right. \\
 & \left[\left(\alpha_m^{(p)} \right)^2 f_p^{(p)'} + \left(\alpha_m^{(n)} \right)^2 f_p^{(n)'} \right] - 2Oh \left(\alpha_m^{(p)} f_r^{(p)''} + \alpha_m^{(n)} f_r^{(n)''} \right) \\
 & \left. - \alpha_m^{(p)} \alpha_m^{(n)} f_r^{(p)} f_r^{(n)} \right\} \int_{-1}^1 P_j(x) P_m(x)^2 dx \\
 & + \hat{\eta}_1^{(p)} \hat{\eta}_1^{(n)} \left\{ 2Oh \left[\alpha_m^{(p)} \left(\left(\frac{f_\theta^{(p)}}{r} \right)' + \frac{f_r^{(p)}}{r^2} \right) + \alpha_m^{(n)} \left(\left(\frac{f_\theta^{(n)}}{r} \right)' + \frac{f_r^{(n)}}{r^2} \right) \right] \right\}
 \end{aligned}$$

$$\begin{aligned}
 & -\alpha_m^{(p)} \alpha_m^{(n)} f_\theta^{(p)} f_\theta^{(n)} \Big\} \int_{-1}^1 P_j(x) P_m^1(x)^2 dx \\
 & + \int_{-1}^1 \left(\mathcal{P}_{21,I}^{(pn)} - 2OhA p_{r21}^{(pn)'} \right) P_j(x) dx .
 \end{aligned} \tag{A.54}$$

Further details on the products of Legendre functions and their evaluation are presented in appendix B.

The right-hand sides of the equations (A.47), (A.49) and (A.53) with evaluated integrals for certain j are named R_1 , R_2 and R_3 , respectively. Using these forms, we multiply equation (A.47) with $(j(j+1)-2)/2\alpha_m^{(p)}$, and subtracting equation (A.53), we obtain

$$\begin{aligned}
 A_{21j} \left[\frac{j(j+1)-2}{2\alpha_m} f_{ar} - 2\alpha_m(j+1)r^j - 2Ohf'_{ar} \right] \\
 + B_{21j} \left[\frac{j(j+1)-2}{2\alpha_m} f_{br} - 2Ohf'_{br} \right] = \frac{j(j+1)-2}{2\alpha_m} R_1 - R_3 .
 \end{aligned} \tag{A.55}$$

With an introduction of new variables a_1 and b_1 , this is written as

$$A_{21j}a_1 + B_{21j}b_1 = \frac{j(j+1)-2}{2\alpha_m} R_1 - R_3 . \tag{A.56}$$

Equation (A.49) with evaluated integral on the left-hand side for certain j with rule (B.1) reads

$$A_{21j} \left[r \left(\frac{f_{a\theta}}{r} \right)' + \frac{f_{ar}}{r} \right] + B_{21j} \left[r \left(\frac{f_{b\theta}}{r} \right)' + \frac{f_{br}}{r} \right] = \frac{R_2}{j(j+1)} . \tag{A.57}$$

Introducing new variables a_2 and b_2 yields

$$A_{21j}a_2 + B_{21j}b_2 = \frac{R_2}{j(j+1)} . \tag{A.58}$$

The unknown coefficients are deduced using the equations (A.56) and (A.58) for the time dependency including the eigenvalues (p) or (n) . They are

written as

$$A_{21j} = \frac{b_2}{a_1 b_2 - a_2 b_1} \left(\frac{j(j+1) - 2}{2\alpha_m} R_1 - R_3 - \frac{R_2}{j(j+1)} \frac{b_1}{b_2} \right), \quad (\text{A.59})$$

$$B_{21j} = \frac{1}{b_2} \left(\frac{R_2}{j(j+1)} - A_{21j} a_2 \right), \quad (\text{A.60})$$

$$H_{21j} = \frac{1}{2\alpha_m} (R_1 - B_{21j} f_{br} - A_{21j} f_{ar}). \quad (\text{A.61})$$

Functions $f_r, f_\theta, f_{ar}, f_{br}$ and their derivatives have to be evaluated at $r = 1$ due to the formulation of the boundary conditions. Using equation (B.2), we calculate the integrals on the right-hand side of all three boundary conditions.

The same procedure follows for deducing the $A_{21j}^{(pn)}, B_{21j}^{(pn)}$ and $H_{21j}^{(pn)}$. The coefficients for the time dependency according to $\exp[-(\alpha_m^{(p)} + \alpha_m^{(n)})t]$ read

$$\begin{aligned} A_{21j}^{(pn)} &= \frac{b_2^{(pn)}}{a_1^{(pn)} b_2^{(pn)} - a_2^{(pn)} b_1^{(pn)}} \left(\frac{j(j+1) - 2}{\alpha_m^{(p)} + \alpha_m^{(n)}} R_1^{(pn)} - R_3^{(pn)} - \frac{R_2^{(pn)}}{j(j+1)} \frac{b_1^{(pn)}}{b_2^{(pn)}} \right), \\ B_{21j}^{(pn)} &= \frac{1}{b_2^{(pn)}} \left(\frac{R_2^{(pn)}}{j(j+1)} - A_{21j}^{(pn)} a_2^{(pn)} \right), \\ H_{21j}^{(pn)} &= \frac{1}{\alpha_m^{(p)} + \alpha_m^{(n)}} \left(R_1^{(pn)} - B_{21j}^{(pn)} f_{br}^{(pn)} - A_{21j}^{(pn)} f_{ar}^{(pn)} \right). \end{aligned} \quad (\text{A.62})$$

Having put together these details for the second-order solutions, we now proceed to the details of the third-order solutions.

G. Solutions for the third-order radial velocity u_{r31}

For the time dependencies not detailed in the thesis, the contributions to the third-order radial velocity component u_{r31} read

$$u_{r31}^{(n)}(r, \theta, t) = - \sum_{h=0}^H \left[A_{31h}^{(n)} r^{h-1} + B_{31h}^{(n)} q^{(n)2} \frac{j_h(\sqrt{3}q^{(n)}r)}{q^{(n)}r} \right] \times h(h+1)P_h(x)e^{-3\alpha_m^{(n)}t} + Ap_{r31}^{(n)}(r, \theta, t), \quad (\text{A.63})$$

$$u_{r31}^{(ppn)}(r, \theta, t) = - \sum_{h=0}^H \left[A_{31h}^{(ppn)} r^{h-1} + B_{31h}^{(ppn)} \frac{j_h(q^{(ppn)}r)}{r} \right] \times h(h+1)P_h(x)e^{-(2\alpha_m^{(p)} + \alpha_m^{(n)})t} + Ap_{r31}^{(ppn)}(r, \theta, t), \quad (\text{A.64})$$

$$u_{r31}^{(pnn)}(r, \theta, t) = - \sum_{h=0}^H \left[A_{31h}^{(pnn)} r^{h-1} + B_{31h}^{(pnn)} \frac{j_h(q^{(pnn)}r)}{r} \right] \times h(h+1)P_h(x)e^{-(\alpha_m^{(p)} + 2\alpha_m^{(n)})t} + Ap_{r31}^{(pnn)}(r, \theta, t), \quad (\text{A.65})$$

$$u_{r31}^{(nn)}(r, \theta, t) = - \sum_{h=0}^H \sum_{k=0}^K \left[A_{31hk}^{(nn)} r^{h-1} + B_{31hk}^{(nn)} \frac{j_h(q_k^{(nn)}r)}{r} \right] \times h(h+1)P_h(x)e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(n)})t} + Ap_{r31}^{(nn)}(r, \theta, t), \quad (\text{A.66})$$

$$u_{r31}^{(pn)}(r, \theta, t) = - \sum_{h=0}^H \sum_{k=0}^K \left[A_{31hk}^{(pn)} r^{h-1} + B_{31hk}^{(pn)} \frac{j_h(q_k^{(pn)}r)}{r} \right] \times h(h+1)P_h(x)e^{-(\alpha_m^{(p)} + \alpha_{2k}^{(n)})t} + Ap_{r31}^{(pn)}(r, \theta, t), \quad (\text{A.67})$$

$$u_{r31}^{(np)}(r, \theta, t) = - \sum_{h=0}^H \sum_{k=0}^K \left[A_{31hk}^{(np)} r^{h-1} + B_{31hk}^{(np)} \frac{j_h(q_k^{(np)}r)}{r} \right] \times h(h+1)P_h(x)e^{-(\alpha_m^{(n)} + \alpha_{2k}^{(p)})t} + Ap_{r31}^{(np)}(r, \theta, t). \quad (\text{A.68})$$

The coefficients in the arguments of the spherical Bessel functions are

$$q^{(ppn)2} = 2q^{(p)2} + q^{(n)2}, \quad q^{(pnn)2} = q^{(p)2} + 2q^{(n)2}, \quad q_k^{(pp)2} = q^{(p)2} + q_{2k}^{(p)2}, \\ q_k^{(nn)2} = q^{(n)2} + q_{2k}^{(n)2}, \quad q_k^{(pn)2} = q^{(p)2} + q_{2k}^{(n)2}, \quad q_k^{(np)2} = q^{(n)2} + q_{2k}^{(p)2}. \quad (\text{A.69})$$

The solutions for the radial velocity consist of pairs of terms with the unknown coefficients A_{31h} and B_{31h} , as well as A_{31hk} and B_{31hk} , under sums with respect to the summation indexes h and k . Equations (3.70) and (A.63)-(A.65) exhibit the coefficients A_{31h} and D_{31h} , and equations (3.71) and (A.66)-(A.68) exhibit the coefficients A_{31hk} and D_{31hk} . The relations are

$$\begin{aligned}
 A_{31h}^{(p)} &= \frac{D_{31h}^{(p)}}{3\alpha_m^{(p)}(h+1)}, & A_{31h}^{(n)} &= \frac{D_{31h}^{(n)}}{3\alpha_m^{(n)}(h+1)}, \\
 A_{31h}^{(ppn)} &= \frac{D_{31h}^{(ppn)}}{(2\alpha_m^{(p)} + \alpha_m^{(n)})(h+1)}, & A_{31h}^{(pmn)} &= \frac{D_{31h}^{(p)}}{(\alpha_m^{(p)} + 2\alpha_m^{(n)})(h+1)}, \\
 A_{31hk}^{(pp)} &= \frac{D_{31hk}^{(pp)}}{(\alpha_{2k}^{(p)} + \alpha_m^{(p)})(h+1)}, & A_{31hk}^{(nn)} &= \frac{D_{31hk}^{(nn)}}{(\alpha_{2k}^{(n)} + \alpha_m^{(n)})(h+1)}, \\
 A_{31hk}^{(pn)} &= \frac{D_{31hk}^{(pn)}}{(\alpha_{2k}^{(p)} + \alpha_m^{(n)})(h+1)}, & A_{31hk}^{(np)} &= \frac{D_{31hk}^{(np)}}{(\alpha_{2k}^{(n)} + \alpha_m^{(p)})(h+1)}. \quad (\text{A.70})
 \end{aligned}$$

H. Solutions for the third-order polar velocity $u_{\theta 31}$

For the time dependencies not detailed in the thesis, the contributions to the third-order angular velocity component $u_{\theta 31}$ read

$$\begin{aligned}
 u_{\theta 31}^{(n)}(r, \theta, t) &= Ap_{\theta 31}^{(n)}(r, \theta, t) - \sum_{h=0}^H (h+1) P_h^1(x) \left[A_{31h}^{(n)} r^{h-1} \right. \\
 &\quad \left. + B_{31h}^{(n)} q^{(n)2} \left(\frac{j_h(\sqrt{3}q^{(n)}r)}{q^{(n)}r} - \sqrt{3} \frac{j_{h+1}(\sqrt{3}q^{(n)}r)}{(h+1)} \right) \right] e^{-3\alpha_m^{(n)}t}, \quad (\text{A.71})
 \end{aligned}$$

$$\begin{aligned}
 u_{\theta 31}^{(ppn)}(r, \theta, t) &= Ap_{\theta 31}^{(ppn)}(r, \theta, t) - \sum_{h=0}^H (h+1) P_h^1(x) \left[A_{31h}^{(ppn)} r^{h-1} \right. \\
 &\quad \left. + B_{31h}^{(ppn)} \left(\frac{j_h(Q^{(ppn)}r)}{r} - Q^{(ppn)} \frac{j_{h+1}(Q^{(ppn)}r)}{h+1} \right) \right] e^{-(2\alpha_m^{(p)} + \alpha_m^{(n)})t}, \quad (\text{A.72})
 \end{aligned}$$

$$u_{\theta 31}^{(pnn)}(r, \theta, t) = Ap_{\theta 31}^{(pnn)}(r, \theta, t) - \sum_{h=0}^H (h+1) P_h^1(x) \left[A_{31h}^{(pnn)} r^{h-1} + B_{31h}^{(pnn)} \left(\frac{j_h(Q^{(pnn)} r)}{r} - Q^{(pnn)} \frac{j_{h+1}(Q^{(pnn)} r)}{(h+1)} \right) \right] e^{-\left(\alpha_m^{(p)} + 2\alpha_m^{(n)}\right)t}, \quad (\text{A.73})$$

$$u_{\theta 31}^{(nn)}(r, \theta, t) = Ap_{\theta 31}^{(nn)}(r, \theta, t) - \sum_{h=0}^H \sum_{k=0}^K (h+1) P_h^1(x) \left[A_{31hk}^{(nn)} r^{h-1} + B_{31hk}^{(nn)} \left(\frac{j_h(Q_k^{(nn)} r)}{r} - Q_k^{(nn)} \frac{j_{h+1}(Q_k^{(nn)} r)}{(h+1)} \right) \right] e^{-\left(\alpha_m^{(n)} + \alpha_{2k}^{(n)}\right)t}, \quad (\text{A.74})$$

$$u_{\theta 31}^{(pn)}(r, \theta, t) = Ap_{\theta 31}^{(pn)}(r, \theta, t) - \sum_{h=0}^H \sum_{k=0}^K (h+1) P_h^1(x) \left[A_{31hk}^{(pn)} r^{h-1} + B_{31hk}^{(pn)} \left(\frac{j_h(Q_k^{(pn)} r)}{r} - Q_k^{(pn)} \frac{j_{h+1}(Q_k^{(pn)} r)}{(h+1)} \right) \right] e^{-\left(\alpha_m^{(p)} + \alpha_{2k}^{(n)}\right)t}, \quad (\text{A.75})$$

$$u_{\theta 31}^{(np)}(r, \theta, t) = Ap_{\theta 31}^{(np)}(r, \theta, t) - \sum_{h=0}^H \sum_{k=0}^K (h+1) P_h^1(x) \left[A_{31hk}^{(np)} r^{h-1} + B_{31hk}^{(np)} \left(\frac{j_h(Q_k^{(np)} r)}{r} - Q_k^{(np)} \frac{j_{h+1}(Q_k^{(np)} r)}{(h+1)} \right) \right] e^{-\left(\alpha_m^{(n)} + \alpha_{2k}^{(p)}\right)t}, \quad (\text{A.76})$$

where for every time dependency $Ap_{\theta 31}(r, \theta, t)$ represents the corresponding particular solution of the non homogeneous radial momentum equation.

I. Solutions for the polar velocity $u_{\theta 32}$, pressure p_{32} and deformation amplitude $\hat{\eta}_{32h}^{(n)}$

The solutions for the angular velocity component $u_{\theta 32}$, the pressure p_{32} and deformation amplitude $\hat{\eta}_{32h}^{(n)}$ read

$$\begin{aligned}
 u_{\theta 32}(r, \theta, t) = & - \sum_{h=0}^H \frac{\hat{\eta}_{32h}^{(p)} \alpha_{3h}^{(p)}}{h} e^{-\alpha_{3h}^{(p)} t} \left[\left(1 + \frac{2(h^2 - 1)}{2q_{3h}^{(p)} Q_h^{(p)} - q_{3h}^{(p)2}} \right) r^{h-1} \right. \\
 & - \frac{2(h-1)q_{3h}^{(p)}}{2q_{3h}^{(p)} Q_h^{(p)} - q_{3h}^{(p)2}} \left((h+1) \frac{j_h(q_{3h}^{(p)} r)}{j_h(q_{3h}^{(p)})} \frac{1}{q_{3h}^{(p)} r} - \frac{j_{h+1}(q_{3h}^{(p)} r)}{j_h(q_{3h}^{(p)})} \right) \Big] P_h^1(\cos \theta) \\
 & - \sum_{h=0}^H \frac{\hat{\eta}_{32h}^{(n)} \alpha_{3h}^{(n)}}{h} e^{-\alpha_{3h}^{(n)} t} \left[\left(1 + \frac{2(h^2 - 1)}{2q_{3h}^{(n)} Q_h^{(n)} - q_{3h}^{(n)2}} \right) r^{h-1} \right. \\
 & - \frac{2(h-1)q_{3h}^{(n)}}{2q_{3h}^{(n)} Q_h^{(n)} - q_{3h}^{(n)2}} \left((h+1) \frac{j_h(q_{3h}^{(n)} r)}{j_h(q_{3h}^{(n)})} \frac{1}{q_{3h}^{(n)} r} - \frac{j_{h+1}(q_{3h}^{(n)} r)}{j_h(q_{3h}^{(n)})} \right) \Big] P_h^1(\cos \theta) ,
 \end{aligned} \tag{A.77}$$

$$\begin{aligned}
 p_{32}(r, \theta, t) = & - \sum_{h=0}^H \frac{r^h}{h} \left[\hat{\eta}_{32h}^{(p)} \alpha_{3h}^{(p)2} e^{-\alpha_{3h}^{(p)} t} \left(1 + \frac{2(h^2 - 1)}{2q_{3h}^{(p)} Q_h^{(p)} - q_{3h}^{(p)2}} \right) \right. \\
 & \left. + \hat{\eta}_{32h}^{(n)} \alpha_{3h}^{(n)2} e^{-\alpha_{3h}^{(n)} t} \left(1 + \frac{2(h^2 - 1)}{2q_{3h}^{(n)} Q_h^{(n)} - q_{3h}^{(n)2}} \right) \right] P_h(\cos \theta) ,
 \end{aligned} \tag{A.78}$$

$$\begin{aligned}
 \hat{\eta}_{32h}^{(n)} = & \frac{1}{\alpha_{3h}^{(n)} - \alpha_{3h}^{(p)}} \left[\alpha_{3h}^{(p)} \left(H_{31h}^{(p)} + H_{31h}^{(n)} \right) \right. \\
 & + \sum_{k=0}^K \left(H_{31hk}^{(pp)} + H_{31hk}^{(nn)} + H_{31hk}^{(pn)} + H_{31hk}^{(np)} \right) + H_{31h}^{(ppn)} + H_{31h}^{(pnn)} \Big) \\
 & + \left(3\alpha_m^{(p)} H_{31h}^{(p)} + 3\alpha_m^{(n)} H_{31h}^{(n)} + (2\alpha_m^{(p)} + \alpha_m^{(n)}) H_{31h}^{(ppn)} \right)
 \end{aligned}$$

$$\begin{aligned}
 & + (\alpha_m^{(p)} + 2\alpha_m^{(n)})H_{31h}^{(pnn)} + \sum_{k=0}^K (\alpha_m^{(p)} + 2\alpha_{2k}^{(p)})H_{31hk}^{(pp)} + \sum_{k=0}^K (\alpha_m^{(n)} + 2\alpha_{2k}^{(n)})H_{31hk}^{(nn)} \\
 & + \sum_{k=0}^K (\alpha_m^{(p)} + \alpha_{2k}^{(n)})H_{31hk}^{(pn)} + \sum_{k=0}^K (\alpha_m^{(n)} + \alpha_{2k}^{(p)})H_{31hk}^{(np)} \Bigg] . \tag{A.79}
 \end{aligned}$$

Appendix B.

Orthogonality of the Legendre polynomials

In the coefficient calculation, integrals of products of Legendre functions $P_l^n(x)$ appear from the boundary conditions. Most of the work presented in this chapter is from Dong and Lemus (2002). The degree l assumes all natural numbers, and the order n is either zero or unity. Taking into account the orthogonality of the Legendre functions, the integrals are evaluated over the interval $[-1, 1]$ of the independent variable $x = \cos \theta$. Integrals of products of two Legendre functions are

$$\int_{-1}^1 P_{l_1}^n(x) P_{l_2}^n(x) dx = \frac{2(l_1 + n)!}{(2l_1 + 1)(l_1 - n)!} \delta_{l_1 l_2} , \quad (\text{B.1})$$

where $\delta_{l_1 l_2}$ is the Kronecker delta.

Multiplication of the boundary conditions with a Legendre function and integration produces integrals of products of three or four Legendre functions of different degrees. The integrals of the form of three Legendre polynomials ($n = 0$) read

$$\int_{-1}^1 P_{l_1}(x) P_{l_2}(x) P_{l_3}(x) dx = 2 \begin{pmatrix} l_1 & l_2 & l_3 \\ 0 & 0 & 0 \end{pmatrix}^2 . \quad (\text{B.2})$$

On the right of this equation, the $3-j$ symbols can be computed using the

Racah formula

$$\begin{aligned} \begin{pmatrix} a & b & c \\ \alpha & \beta & \gamma \end{pmatrix} &= (-1)^{a-b-\gamma} \sqrt{\Delta(a, b, c)} \\ &\times \sqrt{(a+\alpha)!(a-\alpha)!(b+\beta)!(b-\beta)!(c+\gamma)!(c-\gamma)!} \sum_t \frac{(-1)^t}{x}, \end{aligned} \quad (\text{B.3})$$

where $\Delta(a, b, c)$ is a triangle coefficient defined by

$$\Delta(a, b, c) = \frac{(a+b-c)!(a-b+c)!(-a+b+c)!}{(a+b+c+1)!}, \quad (\text{B.4})$$

(Shore and Menzel (1968), p. 273), and

$$x = t!(c-b+t+\alpha)!(c-a+t-\beta)!(a+b-c-t)!(a-t-\alpha)!(b-t+\beta)!, \quad (\text{B.5})$$

with an integer t , and the sum in (B.3) is over all integers t for which the factorials in x have non-negative arguments (Messiah (1962), p. 1058; Shore and Menzel (1968), p. 273). In particular, the number of terms is equal to $\nu + 1$, where ν is the smallest of the nine numbers

$$\begin{aligned} a \pm \alpha, \quad b \pm \beta, \quad c \pm \gamma, \\ a + b - c, \quad b + c - a, \quad c + a - b, \end{aligned}$$

(Messiah (1962), p. 1058).

Upon multiplication with a Legendre function and integration, the right-hand sides of the boundary conditions may also exhibit overlap integrals of three or four Legendre functions. In this analysis, the general solutions by Dong and Lemus (2002) include all the possible combinations of degrees and orders. For three Legendre functions, the overlap integral is

$$\int_{-1}^1 P_{l_1}^{m_1}(x) P_{l_2}^{m_2}(x) P_{l_3}^{m_3}(x) dx = \sqrt{\frac{(l_1 + m_1)! (l_2 + m_2)! (l_3 + m_3)!}{(l_1 - m_1)! (l_2 - m_2)! (l_3 - m_3)!}} \quad (\text{B.6})$$

$$\times \sum_{l_{12}} \sum_{l_{123}} G_{12} G_{123} \times \sqrt{\frac{(l_{123} - m_{123})!}{(l_{123} + m_{123})!}} I(l_{123}, m_{123}), \quad (\text{B.7})$$

while for four Legendre functions it is written as

$$\begin{aligned}
& \int_{-1}^1 P_{l_1}^{m_1}(x) P_{l_2}^{m_2}(x) P_{l_3}^{m_3}(x) P_{l_4}^{m_4}(x) dx = \\
& = \sqrt{\frac{(l_1 + m_1)! (l_2 + m_2)! (l_3 + m_3)! (l_4 + m_4)!}{(l_1 - m_1)! (l_2 - m_2)! (l_3 - m_3)! (l_4 - m_4)!}} \\
& \times \sum_{l_{12}} \sum_{l_{23}} \sum_{l_{1234}} G_{12} G_{123} G_{1234} \sqrt{\frac{(l_{1234} - m_{1234})!}{(l_{1234} + m_{1234})!}} I(l_{1234}, m_{1234}) . \quad (B.8)
\end{aligned}$$

The unknown coefficients G are defined as

$$G_{12} = (-1)^{m_{12}} (2l_{12} + 1) \begin{pmatrix} l_1 & l_2 & l_{12} \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_1 & l_2 & l_{12} \\ m_1 & m_2 & -m_{12} \end{pmatrix} , \quad (B.9)$$

where the summation indexes $|l_1 - l_2| \leq l_{12} \leq l_1 + l_2$ and $l_{12} \geq m_{12}$, defining $m_{12} = \sum_{i=1}^2 m_i$,

$$G_{123} = (-1)^{m_{123}} (2l_{123} + 1) \begin{pmatrix} l_{12} & l_3 & l_{123} \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_{12} & l_3 & l_{123} \\ m_{12} & m_3 & -m_{123} \end{pmatrix} , \quad (B.10)$$

where $|l_{12} - l_3| \leq l_{123} \leq l_{12} + l_3$ and $l_{123} \geq m_{123}$, defining $m_{123} = \sum_{i=1}^3 m_i$, and

$$G_{1234} = (-1)^{m_{1234}} (2l_{1234} + 1) \begin{pmatrix} l_{123} & l_4 & l_{1234} \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_{123} & l_4 & l_{1234} \\ m_{123} & m_4 & -m_{1234} \end{pmatrix} , \quad (B.11)$$

where $|l_{123} - l_4| \leq l_{1234} \leq l_{123} + l_4$ and $l_{1234} \geq m_{1234}$, defining $m_{1234} = \sum_{i=1}^4 m_i$. The coefficient $I(l_{123}, m_{123})$ in equation (B.7) is defined as

$$I(l_{123}, m_{123}) = \frac{\left((-1)^{m_{123}} + (-1)^{l_{123}} \right) \Gamma(l_{123}/2) \Gamma((l_{123} + m_{123} + 1)/2)}{2^{2-m_{123}} ((l_{123} - m_{123})/2)! \Gamma((l_{123} + 3)/2) m_{123}^{-1}} , \quad (B.12)$$

where Γ is the gamma function. Switching the l_{123} to l_{1234} and m_{123} to m_{1234} , the same definition applies for $I(l_{1234}, m_{1234})$ in equation (B.8). Using

the $3-j$ symbols, we calculate the integrals of the Legendre polynomials. They appear in the coefficients $G_{12...i}$ and in relation (B.2). They are zero whenever the triangle inequality between the integers of the first row is not satisfied. This property terminates the expansion of the coefficients in all the boundary conditions. Following the same example, the zero value occurs when $m_1 + m_2 + m_3 \neq 0$, or when $a = b = c = 0$ and simultaneously $l_1 + l_2 + l_3 = 2\lambda + 1$ appears, where $\lambda = 0, 1, 2, \dots$. This property eliminates the odd-order Legendre polynomials from the second-order approximation (Tsamopoulos and Brown, 1983).

Appendix C.

Derivation details for inviscid drop shape oscillations

The present chapter details the determination of the velocity and pressure fields, and the surface deformation amplitudes for oscillating inviscid drops. The appendix pertains to chapter 4 of the thesis and is intended as a collection of equations for coefficients and deformation amplitudes in the solutions for the various orders of approximation. The motivation for details removed from the bulk of the thesis text is to improve the readability of the latter. Still, the thesis has retained the structures of the various solutions.

A. Second-order solutions

In the second-order solutions "21", the coefficients C_{21l} are calculated using the second-order zero normal stress boundary condition (2.25). The pressure solution p_{21} and the known η_{21} are substituted from (3.22) and (3.28), respectively, into the second-order zero normal stress boundary condition. The first-order solutions are also substituted into (2.25), setting r to 1. The coefficients C_{21l} , applying the orthogonality of the Legendre polynomials, for every l between zero and L , and for the time dependencies according to $\exp[-2\alpha_m^{(p)}\tau]$ and $\exp[-2\alpha_m^{(n)}\tau]$, read

$$C_{21l} = \frac{2l+1}{2} \left[\left(\frac{m(m+1)-1}{2} - \frac{3}{8}\alpha_{m,0}^2 \right) \int_{-1}^1 P_m(x)^2 P_l(x) dx \right]$$

$$\left. - \frac{\alpha_{m,0}^2}{8m^2} \int_{-1}^1 P_m^1(x)^2 P_l(x) dx \right] - H_{21l}(l-1)(l+2). \quad (C.1)$$

The method presented in Dong and Lemus (2002) is used for solving the integrals appearing in this equation. The method is detailed in chapter B below. From the equations for the second-order coefficients it follows that $C_{21l}^{(p)} = C_{21l}^{(n)}$. The coefficient $C_{21l}^{(pn)}$ was set to zero for every l , as stated in the earlier derivation of the solutions.

The coefficients C_{22k} , for the contributions "22" to the second-order solutions, are deduced from the homogeneous kinematic boundary condition for each time dependency and each summation index k

$$C_{22k}^{(p)} = \frac{\hat{\eta}_{22k}^{(p)} \alpha_{2k}^{(p)}}{k(k+1)}; \quad C_{22k}^{(n)} = \frac{\hat{\eta}_{22k}^{(n)} \alpha_{2k}^{(n)}}{k(k+1)}. \quad (C.2)$$

The second-order initial conditions determine the two amplitudes $\hat{\eta}_{22k}^{(p)}$ and $\hat{\eta}_{22k}^{(n)}$. For values $k \geq 1$ they read (with H_{21k} taken from (4.50))

$$\hat{\eta}_{22k}^{(p)} = \frac{\alpha_{2k}^{(n)} \left(H_{21k}^{(p)} + H_{21k}^{(n)} + H_{21k}^{(pn)} \right) - \left(2\alpha_m^{(p)} H_{21k}^{(p)} + 2\alpha_m^{(n)} H_{21k}^{(n)} \right)}{\alpha_{2k}^{(p)} - \alpha_{2k}^{(n)}}, \quad (C.3)$$

$$\hat{\eta}_{22k}^{(n)} = - \frac{\alpha_{2k}^{(p)} \left(H_{21k}^{(p)} + H_{21k}^{(n)} + H_{21k}^{(pn)} \right) - \left(2\alpha_m^{(p)} H_{21k}^{(p)} + 2\alpha_m^{(n)} H_{21k}^{(n)} \right)}{\alpha_{2k}^{(p)} - \alpha_{2k}^{(n)}}, \quad (C.4)$$

while for $k = 0$ we have

$$\hat{\eta}_{220}^{(p)} = \hat{\eta}_{220}^{(n)} = -\frac{1}{2} \left(H_{210}^{(p)} + H_{210}^{(n)} + H_{210}^{(pn)} + \frac{1}{2m+1} \right). \quad (C.5)$$

The contributions "22" to the second-order solutions can be formulated in a simple form because $\hat{\eta}_{22k}^{(p)} = \hat{\eta}_{22k}^{(n)} =: \hat{\eta}_{22k}$, and because $\alpha_{2k}^{(p)}$ and $\alpha_{2k}^{(n)}$ are complex conjugate frequencies.

B. Coefficients for third-order solutions "31"

In the simplified forms of the third-order solutions, the relations between the coefficients are

$$C_{31h}^{(p)} = C_{31h}^{(n)} =: C_{31h}^{(1)} \quad C_{31h}^{(ppn)} = C_{31h}^{(pnn)} =: C_{31h}^{(2)} \quad (C.6)$$

$$C_{31hk}^{(pp)} = C_{31hk}^{(nn)} =: C_{31hk}^{(3)} \quad C_{31hk}^{(pn)} = C_{31hk}^{(np)} =: C_{31hk}^{(4)} \quad (C.7)$$

$$H_{31h}^{(p)} = H_{31h}^{(n)} =: H_{31h}^{(1)} \quad H_{31h}^{(ppn)} = H_{31h}^{(pnn)} =: H_{31h}^{(2)} \quad (C.8)$$

$$H_{31hk}^{(pp)} = H_{31hk}^{(nn)} =: H_{31hk}^{(3)} \quad H_{31hk}^{(pn)} = H_{31hk}^{(np)} =: H_{31hk}^{(4)}. \quad (C.9)$$

where superscripts (1), (2), (3) and (4) are used to distinguish different time dependencies.

C. Coefficients for the third-order solutions "32"

The coefficients C_{32h} in the contributions "32" to the third-order solutions are deduced from the homogeneous kinematic boundary condition for each time dependency and every summation index h in the general solution as

$$C_{32h}^{(p)} = \frac{\hat{\eta}_{32h}^{(p)} \alpha_{3h}^{(p)}}{h(h+1)}, \quad C_{32h}^{(n)} = \frac{\hat{\eta}_{32h}^{(n)} \alpha_{3h}^{(n)}}{h(h+1)}. \quad (C.10)$$

The coefficients $\hat{\eta}_{32h}^{(p)}$ and $\hat{\eta}_{32h}^{(n)}$ in the solutions remain to be determined. Using the third-order initial conditions, we can deduce the deformation amplitude of the drop surface for each time dependency and summation index h . The surface deformation amplitudes of contributions "31" and "32" are mutually related through the initial conditions. The amplitudes read

$$\begin{aligned} \hat{\eta}_{32h}^{(p)} = & \frac{1}{\alpha_{3h}^{(p)} - \alpha_{3h}^{(n)}} \left[\alpha_{3h}^{(n)} \left(H_{31h}^{(p)} + H_{31h}^{(n)} \right. \right. \\ & + \sum_{k=0}^K \left(H_{31hk}^{(pp)} + H_{31hk}^{(nn)} + H_{31hk}^{(pn)} + H_{31hk}^{(np)} \right) + H_{31h}^{(ppn)} + H_{31h}^{(pnn)} \Big) \\ & \left. - \left(3\alpha_m^{(p)} H_{31h}^{(p)} + 3\alpha_m^{(n)} H_{31h}^{(n)} + (2\alpha_m^{(p)} + \alpha_m^{(n)}) H_{31h}^{(pn)} + (\alpha_m^{(p)} + 2\alpha_m^{(n)}) H_{31h}^{(pnn)} \right) \right] \end{aligned}$$

$$\begin{aligned}
 & + \sum_{k=0}^K (\alpha_m^{(p)} + \alpha_{2k}^{(p)}) H_{31hk}^{(pp)} + \sum_{k=0}^K (\alpha_m^{(n)} + \alpha_{2k}^{(n)}) H_{31hk}^{(nn)} \\
 & + \sum_{k=0}^K (\alpha_m^{(p)} + \alpha_{2k}^{(n)}) H_{31hk}^{(pn)} + \sum_{k=0}^K (\alpha_m^{(n)} + \alpha_{2k}^{(p)}) H_{31hk}^{(np)} \Bigg) \Bigg] , \tag{C.11}
 \end{aligned}$$

$$\begin{aligned}
 \hat{\eta}_{32h}^{(n)} = & \frac{1}{\alpha_{3h}^{(n)} - \alpha_{3h}^{(p)}} \Bigg[\alpha_{3h}^{(p)} \left(H_{31h}^{(p)} + H_{31h}^{(n)} \right. \\
 & + \sum_{k=0}^K \left(H_{31hk}^{(pp)} + H_{31hk}^{(nn)} + H_{31hk}^{(pn)} + H_{31hk}^{(np)} \right) + H_{31h}^{(ppn)} + H_{31h}^{(pnn)} \Bigg) \\
 & + \left(3\alpha_m^{(p)} H_{31h}^{(p)} + 3\alpha_m^{(n)} H_{31h}^{(n)} + (2\alpha_m^{(p)} + \alpha_m^{(n)}) H_{31h}^{(pn)} + (\alpha_m^{(p)} + 2\alpha_m^{(n)}) H_{31h}^{(pnn)} \right. \\
 & + \sum_{k=0}^K (\alpha_m^{(p)} + \alpha_{2k}^{(p)}) H_{31hk}^{(pp)} + \sum_{k=0}^K (\alpha_m^{(n)} + \alpha_{2k}^{(n)}) H_{31hk}^{(nn)} \\
 & \left. \left. + \sum_{k=0}^K (\alpha_m^{(p)} + \alpha_{2k}^{(n)}) H_{31hk}^{(pn)} + \sum_{k=0}^K (\alpha_m^{(n)} + \alpha_{2k}^{(p)}) H_{31hk}^{(np)} \right) \right] . \tag{C.12}
 \end{aligned}$$

These equations show that $\hat{\eta}_{32h}^{(p)} = \hat{\eta}_{32h}^{(n)}$, which we denote $\hat{\eta}_{32h}$. Furthermore, since $\alpha_{3h}^{(p)}$ and $\alpha_{3h}^{(n)}$ are complex conjugate frequencies, this allows for a simple form of the formulation of the contributions "32" to the third-order solutions.

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