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Interfacial dynamics of liquid droplets and thermophysical property measurement

Suryanarayana, Poodipeddi V. R., Ph.D.
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INTERFACIAL DYNAMICS OF LIQUID DROPLETS
AND THERMOPHYSICAL PROPERTY MEASUREMENT

by

POODIPEDDI V. R. SURYANARAYANA

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IN PARTIAL FULFILLMENT OF THE
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APPROVED, THESIS COMMITTEE

Yildiz Bayazitoglu, Professor of
Mechanical Engineering, Director

Alan J. Chapraan, Professor of
Mechanical Engineering

Ruben D. Cohen, Assistant Professor
of Mechanical Engineering

John L. Margrave, Professor of
Chemistry

Houston, Texas
April, 1991
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ABSTRACT

The interfacial dynamics of a viscous droplet immersed in a viscous medium are considered. The characteristic equation is derived using a normal-mode analysis, and solved for arbitrary, finite fluid properties. The cylinder functions in the characteristic equation are solved using a continued fraction algorithm, and the complex decay factor is searched using a modified quasilinearization minimization scheme. Oscillation frequency and damping rate results are presented for various cases of practical interest (liquid-gas and liquid-liquid systems), and the effect of external medium properties are discussed. It is shown that viscosity of the host medium plays an important part in determining the dynamics of the droplet. Numerical results are also compared to exact solutions for limiting cases, and to existing experimental data for both the fundamental and higher-order modes. It is shown that frequency predictions match very well with experiment, and that damping rate predictions underestimate experimental observation in some cases, possibly due to presence of surface impurities. The application of these results to the measurement of surface tension and viscosity of liquid droplets from single-droplet levitation experiments is also discussed. A new inverse method is developed to
determine surface tension and viscosity from a knowledge of the frequency of oscillations, damping rate, droplet radius, droplet mass (or density), and mode of oscillations. Results are presented for various modes of oscillations in nondimensional form. Finally, the effect of static deformation and external forces on the oscillations of a liquid droplet are considered with special reference to levitation. For an arbitrary static shape deformation, the frequency spectrum is shown to split into \((2l - 1)\) peaks for a mode \(l\) oscillation, and this frequency split is calculated to first order for mode 2, 3, and 4 oscillations. The deformation is then assumed to be a consequence of a general external force, and the frequency split and the static deformation are calculated in terms of the external force parameters. Droplets levitated by acoustic, electromagnetic, and combined acoustic-electromagnetic forces are considered in particular, and it is shown that the effects of asphericity adequately explain the splitting of frequency spectra commonly observed in experiments. The interpretation of spectra with regard to accurate surface tension measurement using the oscillations of levitated droplets is discussed, and the results are applied to some previous experimental results. It is shown that the accuracy of surface tension measurements can improve remarkably if the asphericity caused by the levitating force, and the resultant frequency-split, are taken into account.
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TO MY MOTHER, P. LAKSHMI

Maa,
At whose breast humanity is nourished
And on whose lap civilizations are cradled...

(Adapted from a charcoal by Leonardo da Vinci)
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CHAPTER 1
INTRODUCTION

The interfacial dynamics of liquid droplets immersed in another fluid have been of interest for long, and have been studied, in one form or another, from the times of Lord Kelvin [1]. In addition to its intrinsic theoretical value, an understanding of the dynamics of oscillating droplets has application in many spheres of engineering, notably in chemical engineering [2-4], multiphase flows [5], and more recently, in the development of containerless processing and thermophysical property measurement [6-14] using levitation. Recent progress in containerless processing technologies, and the prospect of space-based processing of new materials [15], has prompted a renewed interest in droplet dynamics.

When a liquid droplet is immersed in another fluid, and its interface is disturbed from its equilibrium position, it returns to its original position either aperiodically or by executing damped oscillations, or continues to execute undamped oscillations, depending upon the properties of the droplet and host media. A general analysis of small oscillations of a viscous fluid immersed in a host fluid has been presented by Miller and Scriven [2]. The special case of a viscous droplet oscillating in a vacuum or a low density gas has been considered by Reid [16], and summarized by Chandrasekhar in his treatise [17]. However, in all these works, solutions for damping rate have been discussed in detail only in the limiting cases of high viscosity or low viscosity of droplet and host. Although Miller and Scriven realize the importance of a detailed study of the interfacial
behavior for a general viscous droplet, neither they, nor subsequent workers have addressed this problem in detail. Though Prosperetti [4] considers a more general droplet-host system, the numerical method he uses is prone to severe errors, and some of the basic physical features of droplet dynamics have not been explained him. The hiatus is partly due to the enormous numerical difficulties that arise from solving the large, awkwardly transcendental complex characteristic equations that describe the dynamics of viscous droplets.

With levitation has come the ability to study an uncontaminated, freely suspended droplet, and to examine closely the interfacial dynamics and thermal processes in liquid droplets at elevated temperatures. Levitation also opens up new avenues to the accurate measurement of different thermophysical and transport properties, such as emissivity, heat capacity, optical properties, thermal diffusivity, surface tension, and viscosity. Of the various levitation methods feasible [15], electromagnetic and acoustic levitation have received the maximum attention, owing to their versatility, ease of realization, and commercial viability under microgravity conditions. Thus, a whole new class of thermophysical property measurement techniques and studies that utilize electromagnetic or acoustic levitation have evolved in the recent times, and many properties have been measured under conditions where no data has been hitherto available.

If a levitated droplet executes damped oscillations, and the oscillatory behavior of such a droplet is understood, then an observation of the frequency of oscillations and damping rate may be used to measure the interfacial tension and
viscosity of one of the fluids. In the past, this has been suggested as a means to obtain surface tension and viscosity data for liquids. Marston [18] considers the free decay of oscillations of an acoustically levitated droplet, but only in the limiting case of two fluids of small viscosities. Oscillations of electromagnetically levitated liquids have already been used to measure surface tension [9], and shape oscillations of acoustically levitated droplets have been used to measure surface tension [8], and have been suggested for simultaneous measurement of surface tension and viscosity [7]. Yet, despite many references to this possibility, there appears to be no work which develops such methods, and describes the oscillations of droplets for finite viscosities, or the inverse theory for property measurement, in detail.

In the context of levitated droplets, this analysis is further complicated by the fact that the droplet may assume an aspherical shape, in response to the external levitating and modulating forces, and to gravity. Oscillations, therefore, are about this equilibrium shape, which may have an effect on the resonance characteristics of the droplet. Indeed, experimentally observed frequency spectra for oscillating levitated droplets exhibit triple peaks [8, 12], and this may be due to the static shape deformation of the droplet. Accurate thermophysical property determination is possible only if the aspherical nature of the equilibrium shape is taken into account in the analysis. It must be noted here that most of the previous studies have considered only a spherical droplet.
In this work, the general analysis of the interfacial dynamics of a viscous liquid droplet immersed in a viscous medium has been undertaken. The governing equations for the oscillating droplet are solved using a normal-mode technique, and a numerical method is developed to solve the complete characteristic equation for the general case. The dependence of the oscillation frequency and the damping rate on the fluid properties, and the transition from the aperiodic to oscillatory behavior, are analytically studied, and results are compared to previous experimental observations. In addition, the inverse theory necessary for the measurement of surface tension and viscosity from a knowledge of the oscillation frequency and damping rate is also developed. The application of this theory to thermophysical property evaluation is demonstrated, and graphs and tables are presented to aid such evaluation. The effect of static shape deformation on the resonance characteristics of a liquid droplet is also investigated. Expressions are derived to calculate the static shape deformation and the split in the frequency spectrum that results from an arbitrary external force. These results are then particularized to the case of acoustic, electromagnetic, and combined acoustic-electromagnetic forces. The interpretation of the split spectra with regard to accurate surface tension measurement is discussed, and the results applied to previously observed split spectra. The improvement in the measurement of surface tension when frequency-split is taken into account, is demonstrated.

The work is roughly divided into three parts. In the first part, the complete theory for the dynamics of a viscous droplet in a viscous medium is developed
and compared to experimental observations. In the second part, thermophysical property measurement by using an inverse theory is discussed, and the inverse theory is developed for a viscous droplet in an inviscid medium. Finally, the effect of static shape deformation on the oscillation of levitated droplets is studied, and the interpretation of experimental frequency spectra is discussed.
CHAPTER 2
DAMPED OSCILLATIONS OF A VISCOUS DROPLET
IN A VISCOUS MEDIUM

2.1. Introduction

In this chapter, the dynamics of a viscous droplet in a viscous medium are considered in detail. The characteristic equation describing the damped oscillations is derived, and solved for arbitrary, finite fluid properties of the droplet and medium. The cylinder functions in the characteristic equation are solved using a continued fraction algorithm, and the complex decay factor is searched using a minimization scheme. The dependence of the complex decay factor on the properties of both inner and outer media is analyzed in detail, and results are presented for various cases of practical importance. The results are compared, in the limit, to some previous results, and shown to be accurate. Theoretical predictions are also compared to existing experimental data. The criteria for neglecting the effect of the outer medium are discussed. The results not only provide a detailed study of the effect of fluid properties on the oscillations of the droplet, but will also be of use in obtaining accurate surface tension and viscosity information from levitation experiments on liquid droplets.

2.2. Analysis

Consider a liquid droplet of equilibrium radius $R$ immersed in a viscous host and executing small shape oscillations about its spherical shape. The droplet has
density \( \rho_i \) and kinematic viscosity \( \nu_i \), and the medium has density \( \rho_o \) and kinematic viscosity \( \nu_o \). The interfacial tension is assumed to be \( \gamma \). For simplicity, we assume that there are no external forces acting on the droplet, and that the interface is free of contamination and surface reactants. For very small amplitude oscillations, in the absence of gravity, the perturbation equation governing the flow (in the host and the droplet) is

\[
\frac{\partial \mathbf{v}}{\partial t} = -\frac{1}{\rho} \nabla p + \nu \nabla^2 \mathbf{v},
\]

(2.1)

where \( \mathbf{v} \) is the velocity vector, \( t \) the time, \( p \) the pressure, and \( \nu \) the kinematic viscosity of the medium. The convective terms can be assumed negligible because they are second order in velocity. Taking the curl of Eq. (2.1), the equation governing the vorticity field can be obtained. The radial component of this vorticity equation is

\[
\left( \frac{\partial}{\partial t} - \nu \nabla^2 \right) (r \hat{z}) = 0
\]

(2.2a)

where \( \hat{z} \) is the radial component of vorticity. The radial component of the curl of Eq. (2.2a) gives another equation governing the radial velocity \( w \),

\[
\nabla^2 \left( \frac{\partial}{\partial t} - \nabla^2 \right) r w = 0
\]

(2.2b)

Applying the normal mode analysis to these equations, we suppose that the solutions to the radial velocity and radial vorticity may be expanded in spherical harmonics, with each term in the respective expansions being given by

\[
r w_{lm} = e^{-\beta_m t} W_{lm}(r) Y_{lm}(\theta, \phi)
\]

(2.3a)

and
\[ r \tilde{z}_{lm} = e^{-\beta_{lm} t} Z_{lm}(r) Y_{lm}(\Theta, \Phi) \]  

(2.3b)

with \( r \) as the radial coordinate. Here, \( \beta \) is the complex decay factor,

\[ \beta_{lm} = \pm \tau_{lm} \pm i \omega_{lm} \]

The imaginary part of \( \beta_{lm} \), \( \omega_{lm} \), is the angular frequency of oscillations, and the real part \( \tau_{lm} \) is the damping rate, with positive \( \tau_{lm} \) implying damped oscillations.

The \( Y_{lm} \) are the spherical harmonics. The functions of \( r \) in the expansions are found by substituting Eqs. (2.3) in Eqs. (2.2), and noting that the \( r \) equations that result from simplification are the modified Bessel equations. These are given by

\[ W_i(r) = a_1 r^l + a_2 r^{l-1} + a_3 (\pi/2q)^{1/2} L^{(1)}_{l+1/2}(q) + a_4 (\pi/2q)^{1/2} L^{(2)}_{l+1/2}(q), \quad (2.4a) \]

and

\[ Z_i(r) = b_1 (\pi/2q)^{1/2} L^{(1)}_{l+1/2}(q) + b_2 (\pi/2q)^{1/2} L^{(2)}_{l+1/2}(q) \]  

(2.4b)

Where \( q = \sqrt{\frac{\beta}{\nu}} r \), \( l \) is the mode, and the \( L \)'s are an appropriate pair of half-integral-order Bessel functions. Applying the condition of boundedness of the solutions, Eqs. (2.4) may be particularized for the inner and the outer fluid, with subscript \( i \) standing for the inner fluid (droplet), and subscript \( o \) representing the properties of outer fluid.

In applying the boundary conditions, the effect of the time-dependent oscillations must be included. Therefore, it is assumed that the surface of the sphere is perturbed from its equilibrium radius \( R \) according to

\[ r = R + \zeta(\Theta, \Phi, t) \]  

(2.5)

where the time-dependent perturbations \( \zeta \) are small, and are expanded in spherical harmonics as
\[ \zeta(\Theta, \phi, t) = \sum_{l,m} \zeta_{lm} e^{-\beta_{lm} t} Y_{lm}(\Theta, \phi) \]  

(2.6)

where the summation \( \sum \) is over both \( l \) and \( m \), with

\[ \sum_{l,m} = \sum_{l} \sum_{m = -l}^{l} \]

Finally, application of the kinematic boundary condition, the tangential stress and the normal force balance conditions and simplification (a detailed discussion of the solution method is given in [2] and [17], and is not repeated here) leads to seven linear homogeneous equations in the unknown coefficients and \( \beta \).

In Eqs. (2.4), Eq. (2.6), and in what follows, \( m = 0 \), i.e., the oscillations are supposed to be axisymmetric, about a spherical equilibrium shape. The subscript \( l \) is also dropped, except when there is a possibility of ambiguity.

For a clean, simple interface, the charactereristic equation reduces to the following determinant, which must equal zero for a unique solution to Eq. (2.1).

\[
\begin{vmatrix}
\beta l (l+1) & 0 & 0 & 0 & 0 \\
0 & \beta (l-1) & -\gamma^2 \Gamma & -(l+2) & C(H) \\
-\beta_i^2 \Gamma & B_i - A_i (l+1) & A_i Q_{l+2} - B_i & A_o - B_o (l+2) & -A_o + B_o C(H) \\
0 & 2\mu_i \beta (l^2-1) & \mu_i G(J) & -2\mu_o I (l+2) & \mu_o F(H)
\end{vmatrix}
\]

(2.7)

where \( \rho_i, \mu_i \) are the inner fluid properties, and \( \rho_o, \mu_o \) are the outer fluid properties (\( \mu = \rho \nu \)). The following definitions have been used to simplify the form of the determinant:

\[ z = \sqrt{\frac{\beta}{\nu_i}} R \]
\[
 z' = \sqrt{\frac{v_i}{v_o}} z \\
 \Gamma = \rho_o l + \rho_i (l+1) \\
 A_i = \frac{2\mu_i}{R^2}, \quad A_o = \frac{2\mu_o}{R^2} \\
 B_i = \frac{\beta\rho_i}{l}, \quad B_o = \frac{\beta\rho_o}{l + 1} \\
 Q_{l+\gamma/2}^J (z) = \frac{J_{l+\gamma/2}(z)}{J_{l+1/2}(z)} \\
 Q_{l+\gamma/2}^H (z') = \frac{H_{l+\gamma/2}(z')}{H_{l+1/2}(z')}
\]

\[
 G (J) = -z^2 + 2z \cdot Q_{l+\gamma/2}^J (z) \\
 C (H) = 2l+1 - z' Q_{l+\gamma/2}^H (z') \\
 F (H) = 2(2l+1) + z'^2 - 2z' Q_{l+\gamma/2}^H (z')
\]

and \( \beta_i^* = \pm i \omega_i^* \), where \( \omega_i^* \) is the inviscid natural frequency of the droplet,

\[
 \omega_i^* = \left[ \frac{l(l+1)(l-1)(l+2)\gamma}{\Gamma R^3} \right]^{1/2} \tag{2.8}
\]
as derived by Lamb [19]. The \( J \)'s and the \( H \)'s in the above definitions are the spherical Bessel and Hankel functions of complex argument, respectively. Both Miller and Scriven [2] and Marston [18] have considered the above determinant. Miller and Scriven solve this determinant for the case of a freely decaying droplet in a host fluid under the limiting case of both fluids having high viscosities or low viscosities. Marston solves the determinant for the case where \( z \gg 1 \) and \( z' \gg 1 \), i.e., both fluids having low viscosities. Prosperetti [4] also considers this
determinant, but solves it using a forward-recursion method. This method is prone to numerical errors, and is a classical example of a numerically unstable technique [20]. In this section, we attempt to solve Eq. (2.7) for arbitrary, finite, fluid properties, by using an accurate continued-fraction algorithm.

Using mathematical rules to simplify the determinant of Eq. (2.7), and nondimensionalizing, we obtain

\[
\begin{vmatrix}
  z \ & \ Q_{l+\frac{1}{2}}^l(z) \\
 2zQ_{l+\frac{1}{2}}^l(z) - z^2/l & -2l+1 & 2l+1 - z'Q_{l+\frac{1}{2}}^l(z') \\
-z^2 + 2zQ_{l+\frac{1}{2}}^l(z) & 2(l^2-1) - l(l+2)/\overline{\mu} & A_{33}
\end{vmatrix} = 0 \quad (2.9)
\]

where

\[
A_{22} = \frac{\overline{\rho}z^2}{l(l+1)} \left[ 1 + \frac{\alpha^4}{z^4} \right] - 2(l-1) - \frac{2(l+2)}{\overline{\mu}}
\]

\[
A_{23} = \frac{z^2}{(l+1)\overline{\nu}} + 2\overline{\mu} \left[ 2l + 1 - z'Q_{l+\frac{1}{2}}^l(z') \right]
\]

\[
A_{33} = \frac{1}{\overline{\mu}} \left[ 2(2l+1) + z' - 2z'Q_{l+\frac{1}{2}}^l(z') \right]
\]

and the nondimensional parameters \( \overline{\nu} = \frac{\rho_i}{\rho_o}, \overline{\mu} = \frac{\mu_i}{\mu_o}, \overline{\nu} = \frac{v_i}{v_o} \) have been introduced. The nondimensional parameter \( \alpha \)

\[
\alpha^2 = \frac{\omega_i \cdot R^2}{v} = \frac{\nu^4(l-1)(l+1)(l+2)R}{\Gamma v^2} \quad (2.10)
\]

has also been introduced in Eq. (2.9). The kinematic viscosity in the denominator of Eq. (2.10) can be either the droplet viscosity (\( \alpha = \alpha_i \)), or the host medium viscosity (\( \alpha = \alpha_o \)). In this work, unless otherwise mentioned, \( \alpha \) is \( \alpha_i \). This nondimensional parameter represents the ratio of two competing forces: the surface
tension forces which are restorative, and the viscous forces which are dissipative. It is called \( \alpha^2 \) for historical reasons [17].

The complex \( z \) that solves Eq. (2.9) is the solution required. Once \( z \) is known, the nondimensional damping rate and oscillation frequency can be obtained from

\[
\frac{z^2}{\alpha^2} = \frac{\tau}{\omega_l} \pm i \frac{\omega}{\omega_l} \tag{2.11}
\]

Note that these nondimensional parameters \( \bar{p}, \bar{v}, \bar{M}, \alpha^2, \tau/\omega_l \) and \( \omega/\omega_l \), are the very ones that result from a dimensional analysis of Eq. (2.7).

Clearly, only a numerical solution is possible for Eq. (2.9). The first step in solving for \( z \) is to describe a numerical algorithm for evaluating the Bessel function ratios \( Q_{l+\frac{1}{2}}^l(z) \) and the Hankel function ratios \( Q_{l+\frac{1}{2}}^H(z) \). In this work, the Bessel function ratios are computed using a continued fraction algorithm with error improvement and zero check for the determination of spherical Bessel functions of complex argument [20]. The algorithm uses a novel technique for evaluating continued fractions that eliminates large storage. The algorithm was checked using the trigonometric expansions for spherical Bessel functions of arbitrary argument [21] for \( l = 2 \) and \( 3 \), and found to be very accurate. Details of this algorithm and the corresponding FORTRAN code are given in Appendix A.

The Hankel function ratios \( Q_{l+\frac{1}{2}}^H(z) \), are computed using an algorithm based on this continued fraction algorithm. It utilizes the recursion relationship for cylinder functions of arbitrary argument \( z \) of the form \( f_l(z) = \sqrt{\frac{2\pi}{z}} F_{l+\frac{1}{2}}(z) \),
\[
\frac{f_{l-2}(z)}{f_{l-1}(z)} = \frac{2l-1}{z} - \frac{f_l(z)}{f_{l-1}(z)} \tag{2.12}
\]

and the relationship between \(j_l\) and \(y_l\)

\[
j_{l+1}(z) y_l(z) - y_{l+1}(z) j_l(z) = z^{-2} \tag{2.13}
\]

where \(j_l\) and \(y_l\) are the spherical Bessel functions of the first and second kind, respectively. Once \(Q_{l+\lambda}^I\) is calculated for a given \(l\) from the algorithm described in Appendix A, using Eqs. (2.12) and (2.13), all the spherical Bessel functions can be calculated downward to \(j_0\) and \(y_0\). Then, using the definition for Hankel functions,

\[
h_l(z) = j_l(z) + i \ y_l(z) \tag{2.14}
\]

and the limiting value for the Hankel functions,

\[
h_0(z) = \frac{\sin z}{z} - i \frac{\cos z}{z} \tag{2.15}
\]

the Hankel function ratio \(Q_{l+\lambda}^I\) can be calculated at each step in the same downward process.

For large \(z'\) (i.e., large \(\nu\)), the Hankel ratio becomes indeterminate. In computing the Hankel functions, this problem is solved by taking the limit using L'Hospital's rule, whenever the absolute value of the argument is greater than a preselected maximum. In fact, as the absolute value of the argument approaches \(\infty\), the Hankel ratio approaches \(-i\). L'Hospital's rule is valid because \(f_l(z)\) are continuous and analytic in the domain of interest (which is the outer medium for Hankel functions).
In order to make the calculations faster and simpler, the complex determinant of Eq. (2.9) can be cast into two equations, one for the real and the other for the imaginary parts, by invoking simple determinant laws. Each of these equations involves the calculation of four determinants, all the terms of which are real. Thus, the problem reduces to a minimization problem for a function of two variables $x$ and $y$, $z = x + iy$. In this work, the solutions are searched using a modified quasilinearization algorithm for local extrema. A complete program has been developed based on these algorithms, that takes as inputs the fluid property parameters $\bar{\nu}$, $\bar{\rho}$, $\bar{\varphi}$, and $\alpha$, the mode of oscillations $l$, and the initial guess for $z = x + i y$, and gives the nondimensional damping rate and oscillation frequency.

2.3. Results and discussion

The solution to Eq. (2.9) depends upon several parameters: $\bar{\nu}$, $\bar{\rho}$, $l$, and $\alpha_l$. Although the theory and numerical code developed here can predict the damping rate for any combination of properties and oscillation mode, it is virtually impossible to report a complete parametric study. Therefore, in this section, we chose to report only cases of practical interest in typical engineering applications. Table I lists these cases and the applications where they may be encountered.

a) Viscous droplet in a fluid of negligible viscosity and density

In the limit of outer medium with negligible viscosity and density, i.e., $\bar{\rho} \to \infty$ and $\bar{\nu} \to \infty$, the characteristic equation reduces to
Table I. Droplet-host systems of practical interest

<table>
<thead>
<tr>
<th>Case</th>
<th>$\bar{p}$</th>
<th>$\bar{v}$</th>
<th>Applications</th>
</tr>
</thead>
<tbody>
<tr>
<td>a)</td>
<td>$\rightarrow \infty$</td>
<td>$\gg 1$</td>
<td>Droplet in medium of negligible density and viscosity (vacuum or rare gas).</td>
</tr>
<tr>
<td>b)</td>
<td>$10^3$, $10^4$</td>
<td>$10^{-5} - 10^3$ (arbitrary)</td>
<td>Electromagnetic levitation in gaseous media, sprays, any liquid-gas system.</td>
</tr>
<tr>
<td>c)</td>
<td>1-2</td>
<td>1-2</td>
<td>Acoustic levitation in liquid media, fluid inclusions in biological cells, emulsions, any liquid-liquid system.</td>
</tr>
</tbody>
</table>
\[
\alpha^4(2Q_{l+4\ell\delta}(z) - z) = z^5 - 2z^4Q_{l+4\ell\delta}(z) + 2z^3(1+l-2l^2) + 4Q_{l+4\ell\delta}(z) z^2(l(l^2+2l^2-2l))
\]

This result can be shown to be identical to the characteristic equation derived in [17]. Figure 1 and Table II show the results for \(\tau_l/\omega_l^\star, \omega_l/\omega_l^\star\) from Eq. (2.16). Results for \(\tau\) and \(\omega\) are obtained for \(l = 2, 3, 4,\) and \(5\). In Figure 1, the solid lines represent the nondimensional damping rate \(\tau_l/\omega_l^\star\), and the dotted lines represent the nondimensional oscillation frequency \(\omega_l/\omega_l^\star\) in the mode \(l\). As can be seen, oscillations begin only beyond an \(\alpha_{crit}^2\), which increases with increasing \(l\). This \(\alpha_{crit}^2\) corresponds to the \(\alpha_{max}^2\) that Chandrasekhar [17] derives for aperiodic damping. For \(\alpha^2 < \alpha_{crit}^2\), there are two real roots of Eq. (2.11), i.e., two aperiodic modes of decay exist, one a rapidly decaying mode, and the other a slowly decaying one. Although there are, in reality, an infinity of real solutions (and have to do with the zeros of the Bessel functions of real argument), the first two have been shown in Figure 1. Once \(\alpha^2 > \alpha_{crit}^2\), or, in other words, the viscosity is less than the critical viscosity corresponding to \(\alpha_{crit}^2\), the modes of decay are characterized by complex \(\beta\)'s. As \(v \to 0\) (\(\alpha^2 \to \infty\)), the nondimensional oscillation frequency approaches 1, i.e. the frequency \(\omega_l\) approaches the inviscid frequency for that mode, \(\omega_l^\star\). At the same time, the damping rate reduces and asymptotically approaches \((l+1)(2l+1)\omega_l^\star/\alpha^2\). These limiting results have already been obtained [2], and our numerical results agree with them, as can be deduced from the limiting values of Table I.
Figure 1. Damped free oscillations in a viscous droplet: The dependence of damping rate and frequency of oscillations on properties. The ordinate measures the nondimensional damping rate and frequency of oscillations, with respect to the inviscid frequency in that mode. The abscissa measures $\alpha^2$. 
Table II. The damped free oscillations of a viscous droplet in an inviscid medium

<table>
<thead>
<tr>
<th>$l = 2$</th>
<th>$l = 3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha^2$</td>
<td>$\frac{\tau}{\omega_n^*}$</td>
</tr>
<tr>
<td>$+3.69020$</td>
<td>$0.96286$</td>
</tr>
<tr>
<td>$3.80000$</td>
<td>$0.93980$</td>
</tr>
<tr>
<td>$4.00000$</td>
<td>$0.89330$</td>
</tr>
<tr>
<td>$4.20000$</td>
<td>$0.85125$</td>
</tr>
<tr>
<td>$4.60000$</td>
<td>$0.77818$</td>
</tr>
<tr>
<td>$5.00000$</td>
<td>$0.71687$</td>
</tr>
<tr>
<td>$5.40000$</td>
<td>$0.66471$</td>
</tr>
<tr>
<td>$6.00000$</td>
<td>$0.59961$</td>
</tr>
<tr>
<td>$7.00000$</td>
<td>$0.51614$</td>
</tr>
<tr>
<td>$8.00000$</td>
<td>$0.45375$</td>
</tr>
<tr>
<td>$9.00000$</td>
<td>$0.40541$</td>
</tr>
<tr>
<td>$10.00000$</td>
<td>$0.36687$</td>
</tr>
<tr>
<td>$11.00000$</td>
<td>$0.33544$</td>
</tr>
<tr>
<td>$12.00000$</td>
<td>$0.30932$</td>
</tr>
<tr>
<td>$13.40000$</td>
<td>$0.27939$</td>
</tr>
<tr>
<td>$14.80000$</td>
<td>$0.25518$</td>
</tr>
<tr>
<td>$17.40000$</td>
<td>$0.22059$</td>
</tr>
<tr>
<td>$21.20000$</td>
<td>$0.18516$</td>
</tr>
<tr>
<td>$27.00000$</td>
<td>$0.14976$</td>
</tr>
<tr>
<td>$36.00000$</td>
<td>$0.11624$</td>
</tr>
<tr>
<td>$42.00000$</td>
<td>$0.10131$</td>
</tr>
<tr>
<td>$59.00000$</td>
<td>$0.07440$</td>
</tr>
<tr>
<td>$65.00000$</td>
<td>$0.06805$</td>
</tr>
<tr>
<td>$75.00000$</td>
<td>$0.05958$</td>
</tr>
<tr>
<td>$79.80000$</td>
<td>$0.05623$</td>
</tr>
</tbody>
</table>
Table II (Continued). The damped free oscillations of a viscous droplet in an inviscid medium.

<table>
<thead>
<tr>
<th>$l = 4$</th>
<th>$l = 5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha^2$</td>
<td>$\frac{\tau}{\omega_n}$</td>
</tr>
<tr>
<td>+15.44109</td>
<td>0.88933</td>
</tr>
<tr>
<td>16.00000</td>
<td>0.86057</td>
</tr>
<tr>
<td>16.80000</td>
<td>0.82225</td>
</tr>
<tr>
<td>17.60000</td>
<td>0.78752</td>
</tr>
<tr>
<td>18.40000</td>
<td>0.75590</td>
</tr>
<tr>
<td>19.20000</td>
<td>0.72701</td>
</tr>
<tr>
<td>20.00000</td>
<td>0.70052</td>
</tr>
<tr>
<td>20.80000</td>
<td>0.67615</td>
</tr>
<tr>
<td>21.60000</td>
<td>0.65365</td>
</tr>
<tr>
<td>24.00000</td>
<td>0.59554</td>
</tr>
<tr>
<td>26.40000</td>
<td>0.54845</td>
</tr>
<tr>
<td>31.20000</td>
<td>0.47697</td>
</tr>
<tr>
<td>36.80000</td>
<td>0.41808</td>
</tr>
<tr>
<td>49.60000</td>
<td>0.33399</td>
</tr>
<tr>
<td>67.20000</td>
<td>0.26779</td>
</tr>
<tr>
<td>73.60000</td>
<td>0.25040</td>
</tr>
<tr>
<td>81.60000</td>
<td>0.23175</td>
</tr>
<tr>
<td>90.40000</td>
<td>0.21428</td>
</tr>
<tr>
<td>100.00000</td>
<td>0.19802</td>
</tr>
<tr>
<td>115.00000</td>
<td>0.17705</td>
</tr>
<tr>
<td>134.50000</td>
<td>0.15565</td>
</tr>
<tr>
<td>161.50000</td>
<td>0.13336</td>
</tr>
<tr>
<td>193.00000</td>
<td>0.11430</td>
</tr>
<tr>
<td>199.00000</td>
<td>0.11128</td>
</tr>
</tbody>
</table>

$^+$ Aperiodic damping occurs for coefficients of kinematic viscosity greater than that which corresponds to this entry (i.e. this entry corresponds to $\alpha_{crit}^2$ and $\tau_{crit}$).
From Figure 1, it is apparent that for each value of \( \tau_l \) below a critical damping rate \( \tau_{crit} \), there are two values of \( \alpha^2 \), one below and the other above \( \alpha^2_{crit} \). As we move from the aperiodic damping regime to the damped oscillations regime, at first there are two modes of aperiodic decay, and past \( \alpha^2_{crit} \), damped oscillations occur with slow decay, characterized by \( \tau < \tau_{crit} \). This value of \( \tau_{crit} \) is the highest rate of damping possible for damped oscillations, and like \( \alpha_{crit} \), is constant for a given mode.

b) Viscous droplet in a fluid of small density

In many applications, such as levitation in gaseous media, and particulate flows, a dense, viscous droplet is suspended in a rarer medium, \( (\overline{\rho} \gg 1) \) of arbitrary viscosity. The viscosity ratio \( (\overline{\nu}) \), thus, is arbitrarily large or small, depending, for instance, on the temperature of the gaseous host. For purposes of presentation, we have chosen \( \overline{\rho} = 10^4 \) and \( 10^3 \), with \( \nu \) varying between \( 10^{-5} \) to \( 10^3 \). The larger density ratio will be representative of liquid metal droplets suspended in air or inert gases- for example, electromagnetic levitation-melting of metals. \( \overline{\rho} = 10^3 \) is typical of water, hydrocarbons, and some other room-temperature liquids suspended in gases- a very common engineering situation. For these cases, the determinant (2.9) is solved numerically, and Figures 2 give results for \( \tau/\omega_l^* \) and \( \omega/\omega_l^* \) for various \( \nu \), for the fundamental \( (l = 2) \) mode. Figure 2a. is for \( \overline{\rho} = 10^4 \), and Fig. 2b for \( \overline{\rho} = 10^3 \). In order to facilitate comparison with the benchmark case (Fig. 1), the variation with \( \alpha_l^2 \) is shown, \( \alpha \) being defined with respect to the
Figure 2a. Damped oscillations of a viscous droplet in a viscous medium for $\bar{\rho} = 10^4$ and different $\bar{v}$. Typical examples for this situation are liquid metals levitated in air or inert gases.
viscosity of the droplet. As in the earlier case, for $\alpha < \alpha_{i,\text{crit}}$, there are infinite modes of aperiodic damping. However, these have not been shown in the figures, since damped oscillations are of interest in this work. It must also be noted here that these, and subsequent figures for cases where the viscosity of the host medium is significant, are for $l = 2$. For higher modes of oscillation, the trend is similar to that predicted in Figure 1, and their calculation is only a matter of detail. For the present, our interest lies in investigating the effect of the fluid properties on the droplet dynamics. To this end, it is sufficient to consider $l = 2$. Higher-order modes are therefore considered in a later section [section f]).

For very large viscosity ratios ($V \gg 1$), the viscosity of the host medium is negligible, and the behavior is identical to the earlier case a), and damped oscillations begin after the same $\alpha_{\text{crit}}$. As $V$ decreases, $\alpha_{\text{crit}}$ increases, and the onset of damped oscillations is delayed, though the behavior approaches the inviscid solution ($\nu/\omega^* \to 0$, $\omega/\omega^* \to 1$) as $\alpha \to \infty$. Eventually, as $V \to 0$, the damping is entirely aperiodic i.e., $\alpha_{\text{crit}} \to \infty$. An understanding of this behavior is facilitated by recalling that $\alpha^2$ as the ratio of the surface restorative energy to the viscous dissipative energy. In case a), where the droplet is viscous and the host is inviscid, the only dissipation is in the inner boundary layer and viscous medium. Thus, when $\alpha^2$ is small, i.e., $\Gamma \nu_i^2 \gg \gamma R$, the viscous dissipation in the inner medium dominates the oscillations, and the damping is aperiodic. Oscillatory behavior begins when the surface energy is sufficiently large (i.e., $\alpha = \alpha_{\text{crit}}$). In case b), however, both inner and outer viscosities play a role. Thus in addition to $\alpha_i$, $V$
Figure 2b. Damped oscillations of a viscous droplet in a viscous medium for $\rho = 10^3$ and different $\nu$. Typical examples for this situation are water and hydrocarbons levitated in air.
also influences the onset of oscillations. Note that $\nu = \frac{\alpha_o^2}{\alpha_i^2}$. For very large $\nu$, the droplet viscosity dominates the damping behavior, and the solution is identical to case a). As $\nu$ decreases, the viscous dissipation in the outer medium begins to have an effect. The surface energy must now compete with the dissipation in the droplet, as well the host fluid, and the onset of damped oscillations is further delayed. As $\alpha_i \rightarrow \infty$, the behavior is once again surface tension dominated, and $\nu/\alpha_i^* \rightarrow 0$, with the oscillation frequency approaching the inviscid natural frequency. For small $\nu$, the higher damping rate is due to both the thick boundary layer that is developing at the interface, and due to the dominating viscosity of the bulk host fluid. Figure 3 depicts the ($l = 2$) behavior of $\alpha_i^2_{cri}$ with $\nu$ for various density ratios. As can be observed, the oscillations are entirely aperiodic ($\alpha_i^2_{cri} \rightarrow \infty$), when $\nu_o \rightarrow \infty$, i.e., $\nu \rightarrow 0$. The physical situation that this corresponds to, can be imagined as a solid external medium constraining the oscillations of the droplet.

In most actual applications (such as the levitation of liquid metals or water and hydrocarbons in air or vacuum), it is important to know when the viscous effects of the outer medium are negligible. From an observation of Figs. 2 and 3, for $\nu$ of the order one and above, the results approach that of a droplet oscillating in an inviscid host, for large $\alpha_i^2$. Table IIIa gives the relevant properties, and Table IIIb gives the results for four commonly levitated materials, water, aluminum, copper, and lead. As Table IIIb shows, $\alpha_i^2$ is very large, and the oscillation
Figure 3. Behavior of $\alpha_{i,\text{crit}}^2$ as a function of the viscosity ratio $\nu$, for different $\bar{\rho}$. 

$\bar{\rho} = 10^4$, $\bar{\rho} = 10^3$, $\bar{\rho} = 10^2$, $\bar{\rho} = 10$, $\bar{\rho} = 1.2$, $\bar{\rho} = 1$
Table IIIa. Properties of some liquids suspended in air.

<table>
<thead>
<tr>
<th>Material</th>
<th>( T ) (K)</th>
<th>( \rho ) (Kg m(^{-3}))</th>
<th>( \nu ) (m(^2) s(^{-1}) × 10(^{-6}))</th>
<th>( \gamma ) (N m(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>298</td>
<td>1000</td>
<td>1.002</td>
<td>0.070</td>
</tr>
<tr>
<td>Aluminum</td>
<td>933</td>
<td>2370</td>
<td>1.9</td>
<td>0.915</td>
</tr>
<tr>
<td>Copper</td>
<td>1356</td>
<td>8240</td>
<td>0.55</td>
<td>0.135</td>
</tr>
<tr>
<td>Lead</td>
<td>600</td>
<td>10600</td>
<td>0.25</td>
<td>0.480</td>
</tr>
</tbody>
</table>

Properties given are at or near the temperature indicated in the table. Properties of liquid metals from Sneyd and Moffatt [26] and of water from CPC handbook of Chemistry and Physics [27]. The external medium is supposed to be air, with \( \rho = 1.2 \text{ Kg m}^{-3} \), and \( \nu = 16 \times 10^{-6} \text{m}^2\text{s}^{-1} \).

Table IIIb. Frequency and damping rate results for some liquids suspended in air.

<table>
<thead>
<tr>
<th>Material</th>
<th>( \alpha_i^2 )</th>
<th>( \omega^* ) (s(^{-1}))</th>
<th>( \omega ) (s(^{-1}))</th>
<th>( \tau ) (s(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>1669.31</td>
<td>68.324</td>
<td>68.297</td>
<td>0.22</td>
</tr>
<tr>
<td>Aluminum</td>
<td>2068</td>
<td>157.164</td>
<td>157.142</td>
<td>0.3876</td>
</tr>
<tr>
<td>Copper</td>
<td>1471.8</td>
<td>32.38</td>
<td>32.375</td>
<td>0.1096</td>
</tr>
<tr>
<td>Lead</td>
<td>5383.21</td>
<td>53.832</td>
<td>53.829</td>
<td>0.0515</td>
</tr>
</tbody>
</table>
frequency is almost exactly equal to the inviscid oscillation frequency, and damping is almost negligible. In fact, in most cases of electromagnetic levitation, where \( R \) is typically 5 \( \text{mm} \), \( \alpha_i^2 \) is very large (of the order \( 10^3 \)). Thus, in measurements of surface tension using levitation in air or gaseous media, the effects of the external medium can be neglected, and the results of the previous section may be used directly.

c) Droplet and host medium of comparable densities and viscosities

In many cases of practical interest, such as acoustic levitation, emulsions, and fluid inclusions in biological cells, a liquid droplet is suspended in a medium of comparable density and viscosity. When such a droplet is set into oscillations, the effect of host medium becomes important. Figure 4 gives the \( (l = 2) \) results for the damped oscillations of a droplet immersed in a host of comparable viscosity and density (\( \overline{\rho} = 1, \) and \( \overline{\nu} = 1 \)). Once again, beyond an \( \alpha_{crit} \) damped oscillations begin. However, what is interesting in this case is that as the frequency of oscillations increases with increasing \( \alpha_i^2 \), the damping rate also increases till a maximum, then begins to decrease with increasing \( \alpha \). Unlike in the previous two cases, where \( \alpha_{i, crit} \) corresponds to the maximum \( \tau_{crit} \), here \( \tau_{crit} \) corresponds to an \( \alpha_i > \alpha_{i, crit} \). However, as \( \nu \) increases, oscillations begin after the maximum damping rate is reached. This behavior can be explained by observing that for droplets of comparable viscosities, as \( \alpha_i \) increases beyond \( \alpha_{i, crit} \), though the surface energy (and hence the frequency of damped oscillations) is increasing, the viscos-
Figure 4. Damped oscillations for a viscous droplet in a viscous medium for $\bar{\rho} = 1$ and $\nabla \approx 1$. Typical examples for this situation are water and hydrocarbons levitated in a liquid (liquid-liquid systems).
ity of the outer medium retains its dissipative effect, and the oscillations continue to be damped at an increasing rate, till the surface energy increases beyond a critical value, $\alpha_{\text{crit},2}$. This $\alpha_{\text{crit},2}$ depends on the viscosity ratio $\nu$. From Fig. 4, it can be seen that for increasing $\nu$, $\alpha_{\text{crit},2}$ decreases, which is exactly what is expected.

d) Comparison with previous experimental observations

Trinh et. al. [22] have performed a detailed series of experiments on the damped oscillations of a liquid droplet. They consider droplets of Silicone-CCl$_4$ acoustically levitated in a distilled water medium. Experimental observations for the fundamental mode ($l = 2$) damping rate and oscillation frequency for various viscosity grades and radii of the mixture are reported. In order to compare their results with the predictions of the theory developed in this work, the damping rate and oscillation frequency information is extracted from the graphs presented in [22], with an estimated error of $\pm 5\%$. The density of water is taken to be 998 $Kg/m^3$. The reported surface tension of the droplet mixtures against distilled water varies between 0.035 - 0.04 N/m. In the comparison, the surface tension is taken to be 0.04 N/m. Figure 5 and Table IV present the results of the comparison of the damping rate and frequency of oscillations respectively. From Fig. 5, we observe that the trend of the experimental data agrees very well with the predicted trend ($\tau$ decreasing with increasing radius for given $\nu_i$, and increasing with increasing $\nu_i$ for any given $R$). The data match quite well with theory for $\nu_i$ of 3.2, 36.8, and 68.6 $cst$. However, for $\nu_i$ of 6.5 and 16.5 $cst$, the experimental
Figure 5. Comparison of predicted damping rate results with experimental data from [22]. The line-plots are the theoretical predictions, and the scatter-plots are data from [22]. Results shown are for Silicone–CCl₄ droplets of various viscosity grades in a distilled water medium. The density of the Silicone–CCl₄ droplets is approximately 990 Kg/m³, and interfacial tension is taken to be 0.040 N/m. The density of distilled water is 998 Kg/m³, and the viscosity is 1.002 cst.
observation is significantly (upto 50%) higher than the theoretical prediction. In similar experiments on \( p-xylene \), Marston and Apfel [23] compare their data with the theoretical prediction for two liquids of very small viscosities. In that, the discrepancies between the observed and predicted values are over 100%. In all these experiments, the error in determining the damping rate may itself be fraught with error, which can be upto 50%. In addition, the discrepancies could be due to the extreme sensitivity of the droplet surface to impurities and surfactants, which form a contaminant film at the interface. The apparent lack of dependence of error in Fig. 5 on the viscosity of the droplet is indicative of the likely presence of such impurities in the experiments of Trinh. et. al. In fact, in [22,23], the authors draw attention to such a likelihood. By considering the interface as having an elasticity and viscosity of its own, the effect of this contaminant film may be included. Indeed, Miller and Scriven [2] have theoretically included these interfacial properties in their boundary conditions, and show that they give rise to increased viscous dissipation, i.e., a higher damping rate, as the experiments indicate. Although such an inclusion of surface properties is theoretically possible, the origin of these contaminants and their exact properties must be known before a correction can be applied to theory. In [23], such a correction to theory is attempted, and it is demonstrated that theoretical prediction can be improved to within 50% of the experimentally observed values of damping rate.

Table IV compares the oscillation frequency results from the theory to those observed in experiments. Results are also compared to the frequency prediction
Table IV. Comparison of frequency results with experimental data.

<table>
<thead>
<tr>
<th>$R$ (cm)</th>
<th>$v_i = 3.2$ cst</th>
<th>$v_i = 6.5$ cst</th>
<th>$v_i = 36.8$ cst</th>
<th>$v_i = 68.4$ cst</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$f^2 (19)$</td>
<td>$f^2 (22)$</td>
<td>$f_{th}^2 (22)$</td>
<td>$f_{th}^2 (22)$</td>
</tr>
<tr>
<td>0.49</td>
<td>41.6</td>
<td>37.6</td>
<td>38.63</td>
<td>37.9</td>
</tr>
<tr>
<td>0.52</td>
<td>34.76</td>
<td>31.8</td>
<td>32.36</td>
<td>33.1</td>
</tr>
<tr>
<td>0.55</td>
<td>29.4</td>
<td>27.4</td>
<td>27.38</td>
<td>27.3</td>
</tr>
<tr>
<td>0.60</td>
<td>22.65</td>
<td>21.4</td>
<td>21.12</td>
<td>23.8</td>
</tr>
<tr>
<td>0.64</td>
<td>18.67</td>
<td>17.5</td>
<td>17.42</td>
<td>18.0</td>
</tr>
<tr>
<td>0.69</td>
<td>15.6</td>
<td>16.1</td>
<td>14.54</td>
<td>16.0</td>
</tr>
</tbody>
</table>

$f_{th}^2$ is the theoretically predicted square of the frequency. The natural frequency is calculated using Eq. (2.8). The theoretical prediction is compared with the experimental data from (22) for Silicone–$CCl_4$ droplets of four different viscosity grades: 3.2, 6.5, 36.8, and 68.4 cst. Droplets are levitated acoustically in distilled water. Density of water is 998 $Kg/m^3$, viscosity of water is 1.002 cst and the approximate interfacial tension is 0.040 $N/m$. 

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for inviscid media [19]. It can be seen that the oscillation frequencies match very well with the predicted frequencies. The agreement between experiment and theory suggests that from single-droplet levitation experiments, it is possible to predict surface tension from a measure of the frequency if the theory of this work is used. Indeed, direct application of the inviscid results of [19] yields a significant error, especially when the viscosity ratio is of the order 1, and cannot be used to measure surface tension with acceptable accuracy. A study of the dependence of the square of the frequency on the cube of the droplet radius also indicates that a power law of the form

\[ f \propto R^{-1.5} \]

adequately describes the frequency behavior, as was observed in the experiments. It must also be noted that although the frequency values of [19] are independent of viscosity, our theory predicts a reduction in the frequency values with increasing \( v_i \), a trend that was observed in [22].

e) Critical radius for damped oscillations

In the above discussion, we have seen that for \( \alpha > \alpha_{\text{crit}} \), damped oscillations commence. Often, for a given droplet-host system, it is important to know whether the droplet returns aperiodically to its original shape, or executes damped oscillations. Clearly, for a given droplet-host system, the value of \( \alpha_{\text{crit}}^2 \) is a constant, and determines the critical radius for damped oscillations to occur. Table V gives the computed values for critical radius, for some commonly levitated materi-
Table V. Critical radius for damped oscillations for some droplet-host systems.

<table>
<thead>
<tr>
<th>Droplet</th>
<th>Host</th>
<th>$\alpha_{crit}^2$</th>
<th>$R_{crit} \ (\mu m)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>Air</td>
<td>3.6902</td>
<td>0.023</td>
</tr>
<tr>
<td>Silicone – CCl$_4$ (a)</td>
<td>Water</td>
<td>5.077</td>
<td>1.48</td>
</tr>
<tr>
<td>Silicone – CCl$_4$ (b)</td>
<td>Water</td>
<td>4.266</td>
<td>1475</td>
</tr>
<tr>
<td>Aluminum</td>
<td>Air</td>
<td>3.707</td>
<td>0.016</td>
</tr>
<tr>
<td>Copper</td>
<td>Air</td>
<td>3.709</td>
<td>0.032</td>
</tr>
<tr>
<td>Lead</td>
<td>Air</td>
<td>3.73</td>
<td>0.002</td>
</tr>
<tr>
<td>$P$–xylene</td>
<td>Water</td>
<td>6.683</td>
<td>0.435</td>
</tr>
</tbody>
</table>

(a) $v_i$ for Silicone – CCl$_4$ droplet is 3.2 cst.
(b) $v_i$ for Silicone – CCl$_4$ droplet is 120.4 cst.
als. It can be concluded from the last column of Table V that most levitated droplets, (with radius ~ 2 mm) are large enough to execute damped oscillations. It must be noted here that in [16] and [17], the critical radius for water in air was calculated erroneously as 0.23 mm. The error, it appears, was due to a discrepancy of units, which the authors overlooked.

f) Higher-order modes of resonance frequencies (l = 3, 4 and 5)

The numerical code developed in this work can be directly used to study the damped oscillations of droplets for higher-order modes (l = 3 and above). The behavior is similar to that for mode 2 oscillations, with the only difference being that \( \alpha_{i,\text{crit}} \) is larger for any given \( \varpi \) and \( \nabla \) when higher modes are considered. For a given system, the damping rate and the frequency of oscillations are higher for the higher modes. In Table VI, the frequency results from the present theory are compared with previous theory [19] and the experimental results of Trinh et. al. [22] for acoustically levitated Silicon CCl4 in a distilled water host. The frequency results compare favorably with experimental values, suggesting that surface tension values may be extracted from an observation of the oscillation frequencies, if the mode shapes are correctly recognized.

2.4. Conclusions

The oscillations of a viscous droplet in a viscous medium, with arbitrary fluid properties, have been considered in detail, and the characteristic equation solved for complex decay factors using a numerical scheme. The complete code accepts
Table VI. Comparison of predicted frequency with experimental data for $l = 3, 4$ and $5$.

<table>
<thead>
<tr>
<th>Source</th>
<th>Radius (mm)</th>
<th>$f_3$</th>
<th>$f_4$</th>
<th>$f_5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Expt. [22]</td>
<td>6.49</td>
<td>9.37</td>
<td>13.1</td>
<td></td>
</tr>
<tr>
<td>Theory</td>
<td>7.68</td>
<td>6.01</td>
<td>9.16</td>
<td>12.63</td>
</tr>
<tr>
<td>Lamb</td>
<td>6.21</td>
<td>9.49</td>
<td>13.11</td>
<td></td>
</tr>
<tr>
<td>Expt. [22]</td>
<td>6.81</td>
<td>10.21</td>
<td>13.65</td>
<td></td>
</tr>
<tr>
<td>Theory</td>
<td>7.4</td>
<td>6.35</td>
<td>9.68</td>
<td>13.35</td>
</tr>
<tr>
<td>Lamb</td>
<td>6.56</td>
<td>10.03</td>
<td>13.86</td>
<td></td>
</tr>
<tr>
<td>Expt. [22]</td>
<td>7.21</td>
<td>10.45</td>
<td>---</td>
<td></td>
</tr>
<tr>
<td>Theory</td>
<td>7.1</td>
<td>6.75</td>
<td>10.3</td>
<td>14.2</td>
</tr>
<tr>
<td>Lamb</td>
<td>6.9</td>
<td>10.7</td>
<td>14.75</td>
<td></td>
</tr>
</tbody>
</table>

Experimental data [22] are for Silicone–$CCl_4$ droplets (density approx. 990 $Kg/m^3$) with viscosity 3.2 $cst$, levitated in a distilled water medium. The density of water is 998 $Kg/m^3$, viscosity is 1.002 $cst$ and the approximate interfacial tension is 0.040 $N/m$. 
as inputs the fluid properties, the interfacial tension, the droplet radius and the mode of oscillations, and predicts the damping rate and frequency of oscillations. Using this scheme, some cases of practical interest have been investigated thoroughly. Based on the work reported in this chapter, the following conclusions may be arrived at:

(1) The properties of the host medium influence the damping rate and the frequency of oscillations of liquid droplets. The effect of increasing viscosity of the host medium is to delay the onset of oscillations. Host medium effects may be neglected in liquid-gas systems, but are significant in liquid-liquid systems.

(2) The frequencies match very well with existing experimental data. Thus, surface tension may be measured based on the observation of oscillation frequencies in single-droplet levitation experiments.

(3) The predicted damping rate is, in certain cases, much lower than that observed in experiments. This may be due to the possible presence of surface contamination, which tends to increase viscous dissipation. If the droplet interface is relatively clean and pure, damping rate information may be used to measure the viscosity of one of the liquids when the viscosity of the other is known.

(4) It must be noted, however, that this theory is for a droplet executing oscillations about a spherical shape. This is not always true in experiments, where
the external acoustic and/or electromagnetic forces may cause the droplet to assume a non-spherical shape. It has been shown by earlier workers [24,25] that oscillations about aspherical shapes tend to split the frequency spectrum, and the five-fold degeneracy of the frequency spectrum no longer holds. Thus, in actual experiments, it is reasonable to expect some non-axisymmetric mode \((m \neq 0)\) to be excited. A complete aspherical theory must be developed before more accurate determination of surface tension and viscosity from single-droplet levitation experiments is possible.
CHAPTER 3
SURFACE TENSION AND VISCOSITY FROM
DAMPED OSCILLATIONS OF A VISCOUS DROplet

3.1. Introduction

Levitation methods, in particular electromagnetic and/or acoustic levitation, have often been suggested as a viable option for containerless processing of materials in reduced gravity environments [15]. In addition to possessing many technological advantages, levitation enables closer and better controlled examination of interfacial dynamics and thermal processes in liquid droplets. Levitation methods have become very attractive in the case of high-temperature materials and melts, due to the added complication of crucible contamination, which they effectively eliminate. Electromagnetic and acoustic levitation are the default choices, owing to their simplicity, stability, and high-temperature capabilities. As we have mentioned earlier, previous workers [7,8,12,22] have suggested that from a knowledge of the frequency of oscillations and damping rate of an oscillating levitated liquid droplet, it is possible to determine surface tension and viscosity from a single levitation experiment. Although this is intuitively obvious, no previous work has developed the requisite inverse theory to make this possible. In this chapter, we develop a method to measure simultaneously the viscosity and the surface tension from a knowledge of the frequency of oscillations ($\omega$) and the rate of oscillation damping ($\tau$) of a liquid droplet. The inverse procedure described herein takes $\omega$ and $\tau$ as inputs, and predicts the surface tension $\gamma$ and the
kinematic viscosity \( v \) for any given liquid droplet of known density and geometry, oscillating in a known mode.

3.2. Levitation techniques for thermophysical property measurement

With the increasingly realizable possibility of space-based materials processing, levitation techniques, such as electrostatic levitation [28], aerodynamic levitation [29], electromagnetic levitation [30], and acoustic levitation [23] have been receiving much attention in the recent times. Of these, electromagnetic and acoustic levitation have been most successful in thermophysical property evaluation, and several thermophysical properties have been measured, and new methods to measure properties are being continuously developed [6-14].

**Electromagnetic levitation**

Electromagnetic levitation owes its origin to Muck [31], who first patented the technique. However, it is only after the experiments by Okress et. al. [29] that it has become a technically viable process. Since then, it has been used for several materials and thermophysical property studies, such as calorimetry [11], optical property and temperature measurement [10], surface tension measurements [9], solidification and undercooling studies [33], and casting [34], to name a few.

A typical electromagnetic levitator (see Figure 1) consists of an oppositely wound coil (a copper coil with the upper few windings wound in a direction opposite to the bottom windings), driven by a high frequency current. When an electrically conducting material (such as a metal) is placed in the alternating
Figure 1. A Typical Electromagnetic Levitator
electromagnetic field generated within such a coil, it experiences Lorentz forces and ohmic heating due to the induced currents. When these are of a sufficient magnitude, the specimen may be supported against gravity due to the Lorentz forces, and may be heated due to the ohmic heating, till it melts. This results in a suspended, high temperature, liquid metal droplet, with a surface free of container contact. Since most liquid metals at high temperatures are extremely corrosive, contact with a container causes surface contamination. Electromagnetic levitation is an attractive high temperature materials processing technique, particularly for liquid transition metals and super alloys, for which there are no unreactive containers in the excess of 1500° C, and can be used in vacuum or other protective atmospheres. However, it has its limitations. Firstly, the material must necessarily be electrically conductive, thus limiting its applicability to metals and metal alloys. Secondly, due to the nature of coil design, sample visibility is severely restricted. Thirdly, on earth, due to gravity, there is a restriction on the maximum weight levitable. However, it remains a popular containerless process, and is receiving considerable attention from NASA [35].

**Acoustic Levitation**

Acoustic levitation, on the other hand, uses an ultrasonic acoustic standing wave, generated within a levitating chamber (see Figure 2). When a specimen is placed at the nodes of this standing wave, it levitates. Typically, a piezoelectric transducer is excited using a high intensity, ultrasonic acoustic wave, and a
Figure 2. A Typical Acoustic Levitator
reflector is placed at a suitable position above the transducer (or horn) for a standing wave to be generated within the chamber. The levitating wave can also be modulated by a low frequency acoustic wave, which causes the droplet to oscillate at the modulation frequency. This enables the excitation of predetermined oscillation frequencies. This method was first used by Marston [13], and has since been used to study various room temperature and moderate temperature materials. Unlike in electromagnetic levitation, the material need not be electrically conductive, thus making the technique versatile. However, there is a limitation on the size and weight of the droplet. The size is restricted by the wavelength of the acoustic wave, and the weight is limited by the maximum intensity that can be stably achieved on earth. For example, for a water droplet, an acoustic wave of about 160 dB is required. Beyond this intensity, the system becomes unstable, as cross frequencies are also excited at very high intensities. Also, since the acoustic wave is basically a pressure wave, a host medium is always necessary, and due to instabilities, very high temperatures cannot be handled with the ease possible in an electromagnetic levitator. Some previous works [36] have reported temperatures up to 2000 K for acoustic leviation of metals. Despite these successes, acoustic leviation has remained, in general, a low temperature technique to study levitated droplets.

Regardless of the levitation method employed, it is possible to obtain a freely suspended liquid droplet, executing shape oscillations. Once a droplet begins oscillating, the oscillations are recorded using a high speed camera, or are studied
using some innovative photometric techniques [22,23] in the case of acoustic levitation. The recorded high-speed films are then analyzed. The mode of oscillations is deduced from visual inspection, and using Fast Fourier Transforms (FFT), frequency spectra corresponding to the mode of oscillations are obtained. These are then inspected for the dominant peak, which corresponds to the oscillation frequency. Simultaneously, the damping rate is determined from an oscilloscope trace of the oscillations (which is especially suited to photometric techniques and acoustic levitation), or is extracted from the high speed film assuming an exponential decay. The equilibrium spherical radius of the droplet is calculated either from the pictures, or from its mass and density before levitation commences. These are the experimental data necessary for the determination of surface tension and viscosity. With these data, and an appropriate theory, it is presumably possible to determine surface tension and viscosity of the liquid sample being examined. The development of such a theory is addressed in the next section.

3.3. Surface tension and viscosity from damped oscillations of a levitated droplet: Inverse theory

In Chapter 2, we have seen the development of a theory for the damped oscillations of a viscous droplet. For simplicity, let us consider a viscous droplet oscillating in an inviscid medium, for which Eq. (2.13) is the characteristic equation. For a droplet of known viscosity and density, and a known interfacial tension, Eq. (2.13) has been solved for the damping rate and frequency of oscilla-
tions. However, in this section, it is of interest to calculate the surface tension and viscosity for a droplet whose frequency of oscillations and damping rate are known. Whenever additional dynamic parameters or boundary conditions are known in lieu of properties, it is possible to pose an inverse problem for the unknown properties. Such inverse problems are common in heat conduction [6,37], and are often ill-posed, making their solution often difficult. In [37], a detailed mathematical discussion of such ill-posed inverse problems is given. In our case, the inverse problem is for surface tension and viscosity.

Using the polar form for the complex variable $z$

$$z = x + iy = r(\cos \theta + i \sin \theta),$$

in Eq. (2.13), simplifying, and separating into real and imaginary parts, we obtain

$$\alpha^4[r \cos \theta - 2Q_{RE}(z)] = -r^5 \cos 5\theta + 2Q_{RE}(z) r^4 \cos 4\theta$$
$$- 2Q_{IM}(z) r^4 \sin 4\theta + 2(2l^2 - l - 1) r^3 \cos 3\theta$$
$$- 4l(l-1)(l+2)(r^2 \cos 2\theta Q_{RE} - r^2 \sin 2\theta Q_{IM}) \quad (3.1a)$$

and

$$\alpha^4[r \sin \theta - 2Q_{IM}(z)] = -r^5 \sin 5\theta + 2Q_{RE}(z) r^4 \sin 4\theta$$
$$+ 2Q_{IM}(z) r^4 \cos 4\theta + 2(2l^2 - l - 1) r^3 \sin 3\theta$$
$$- 4l(l-1)(l+2)(r^2 \sin 2\theta Q_{RE} + r^2 \cos 2\theta Q_{IM}) \quad (3.1b)$$

In Eqs. (3.1a) and (3.1b), $\alpha$ is defined as

$$\alpha^2 = \omega \cdot \frac{R^2}{v}, \quad (3.2)$$

where $v$ is the kinematic viscosity of the droplet, and $Q_{RE}$ and $Q_{IM}$ are the real and imaginary parts of the Bessel function ratio $Q^{H}_{l+4\alpha}(z)$ for the complex variable.
Here, and in the rest of this chapter, \( r \) in the polar representation of \( z \) is not to be confused with the radius. As we shall see, expressing the complex variable in the polar form makes the inverse problem easier to solve. As in Chapter 2, the Bessel function ratio is computed using a continued fraction algorithm with error improvement for the determination of spherical Bessel functions of complex arguments [20]. We have already seen, in Figure 1 and Table II of Chapter 2, the damped dynamics of the droplet. However, they cannot directly be used to determine the viscosity and surface tension of the droplet from \( \tau \) and \( \omega \).

In a typical experiment, a droplet suspended in a vacuum or a gas of negligible density is set into damped oscillations. In order for this to happen, the viscosity of the liquid must be small enough (or the radius so chosen) that \( \alpha^2 > \alpha_{\text{crit}}^2 \). For example, for water, this reduces to the requirement that \( R > 0.023 \, \mu m \), which is easily achieved in actual levitation experiments.

From the definition of \( z \) and \( \beta \),

\[
\beta = \frac{v}{R^2} (x + i \, y)^2 = \tau \pm i \omega,
\]

where \( x \) and \( y \) are real. Thus, we have

\[
x = \left[ R^2 / 2v (\tau + \sqrt{\tau^2 + \omega^2}) \right]^{1/2}
\]

(3.3a)

and

\[
y = \left[ R^2 / 2v (\tau + \sqrt{\tau^2 + \omega^2}) \right]^{1/2}
\]

(3.3b)

where, clearly, only the positive root is acceptable for \( x \) and \( y \) to be real. Or, in the polar formulation used to obtain Eqs. (3.1),
\[ r = \left[ R^2 / \sqrt{(\tau^2 + \omega^2)} \right]^{1/2} \] (3.4a)

and

\[ \tan \theta = \left[ \frac{\sqrt{(\tau^2 + \omega^2)} - \tau}{\sqrt{(\tau^2 + \omega^2)} + \tau} \right]^{1/2} \] (3.4b)

Thus, once \( \tau \) and \( \omega \) are known, \( \theta \) is uniquely determined, and \( r = r(\nu) \) in Eqs. (3.1). Thus, Eqs. (3.1) can be solved again, this time for an unknown \( r \) and \( \alpha \). Once \( r \) and \( \alpha \) are known, \( \nu \) and \( \gamma \) can be found. A program that takes \( \tau \) and \( \omega \) as inputs along with \( R \), \( l \), and \( \rho \) or mass and computes the surface tension and viscosity has been developed. If the program is to be used to compute only surface tension values from inviscid oscillations, an artificially low value of \( \tau \) (e.g., \( \tau = 0.01 \)) can be given to the program. The program is very sensitive to initial guesses of \( r \) and \( \alpha^2 \), but this is a characteristic of the modified quasilinearization algorithm. A good guess would be the one derived from the known properties at room temperature.

Results are presented in a nondimensional form for oscillation modes \( l = 2, 3, 4, \) and 5. Table I presents some numerical values of the surface tension to viscosity parameter \( N_{sv} \),

\[ N_{sv} = \frac{\gamma R}{\rho \nu^2} = \frac{\alpha^4}{l(l-1)(l+2)} , \] (3.5)

and the nondimensional viscosity number \( \bar{\lambda} \),

\[ \bar{\lambda} = \tau \frac{R^2}{\nu} , \] (3.6)

for different values of the nondimensional oscillation parameter (or the nondimensional frequency), \( \bar{\omega} \),
Table I. Determination of Surface Tension and Viscosity: Dependence of $N_{sv}$ and $X$ on $\bar{\omega}$ for $l = 2, 3, 4$ and 5.

<table>
<thead>
<tr>
<th>$\bar{\omega}$</th>
<th>$l=2$</th>
<th>$l=3$</th>
<th>$l=4$</th>
<th>$l=5$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$10^{-3}N_{sv}$</td>
<td>$X$</td>
<td>$10^{-3}N_{sv}$</td>
<td>$X$</td>
</tr>
<tr>
<td>0.001000</td>
<td>0.001702</td>
<td>3.570216</td>
<td>0.002594</td>
<td>8.151839</td>
</tr>
<tr>
<td>0.009999</td>
<td>0.001702</td>
<td>3.570216</td>
<td>0.002594</td>
<td>8.151839</td>
</tr>
<tr>
<td>0.287348</td>
<td>0.001857</td>
<td>3.571770</td>
<td>0.002835</td>
<td>8.164824</td>
</tr>
<tr>
<td>0.371391</td>
<td>0.001977</td>
<td>3.572979</td>
<td>0.003023</td>
<td>8.174925</td>
</tr>
<tr>
<td>0.514496</td>
<td>0.002322</td>
<td>3.576426</td>
<td>0.003566</td>
<td>8.203777</td>
</tr>
<tr>
<td>0.894427</td>
<td>0.008794</td>
<td>3.637097</td>
<td>0.014515</td>
<td>8.718832</td>
</tr>
<tr>
<td>0.946863</td>
<td>0.018246</td>
<td>3.713712</td>
<td>0.032528</td>
<td>9.341665</td>
</tr>
<tr>
<td>0.970142</td>
<td>0.032405</td>
<td>3.807543</td>
<td>0.061584</td>
<td>9.981439</td>
</tr>
<tr>
<td>0.980581</td>
<td>0.051841</td>
<td>3.906799</td>
<td>0.102237</td>
<td>10.502523</td>
</tr>
<tr>
<td>0.992278</td>
<td>0.144415</td>
<td>4.157348</td>
<td>0.292799</td>
<td>11.420168</td>
</tr>
<tr>
<td>0.995037</td>
<td>0.234999</td>
<td>4.268935</td>
<td>0.477472</td>
<td>11.756273</td>
</tr>
<tr>
<td>0.998752</td>
<td>1.033843</td>
<td>4.523284</td>
<td>2.111901</td>
<td>12.511064</td>
</tr>
<tr>
<td>0.999445</td>
<td>2.417571</td>
<td>4.622794</td>
<td>4.956428</td>
<td>12.813521</td>
</tr>
<tr>
<td>0.999800</td>
<td>6.966849</td>
<td>4.715748</td>
<td>14.342289</td>
<td>13.100617</td>
</tr>
<tr>
<td>0.999861</td>
<td>10.139791</td>
<td>4.742421</td>
<td>20.901527</td>
<td>13.183807</td>
</tr>
<tr>
<td>0.999898</td>
<td>13.914793</td>
<td>4.762822</td>
<td>28.712662</td>
<td>13.247664</td>
</tr>
<tr>
<td>0.999922</td>
<td>18.293556</td>
<td>4.779069</td>
<td>37.779778</td>
<td>13.298656</td>
</tr>
<tr>
<td>0.999938</td>
<td>23.277454</td>
<td>4.792400</td>
<td>48.106171</td>
<td>13.340581</td>
</tr>
</tbody>
</table>
\[ \bar{\omega} = \frac{\omega}{\sqrt{\omega^2 + \tau^2}} \]  

(3.7)

\( N_s, \bar{X}, \) and \( \bar{\omega} \) are the three nondimensional parameters that define the problem completely.

From Table I, it is apparent that the region \( 0.9 \leq \bar{\omega} \leq 1.0 \) is of greater interest. In order to give as much detail as possible in this region, the results are plotted with \( 1 - \bar{\omega} \) as the abscissa in Figures 3-5. Figures 3 and 4 show two regions, \( 0.9 \leq \bar{\omega} \leq 1.0 \) and \( \bar{\omega} < 0.9 \). It can be observed from these figures that as \( \bar{\omega} \to 0 \), \( N_s \) reaches a minimum which corresponds to the \( \alpha_{\text{crit}}^2 \) for that \( l \). Below this \( N_s \), damping is aperiodic, as discussed earlier. Also, as

\[ \bar{\omega} \to 1, \, \omega \to \omega^* \]  

(3.8)

Or, in terms of \( N_s, \bar{\omega}, \) and \( \bar{X} \), \( \omega \) can be expressed as

\[ \frac{\omega^2}{\omega^*^2} = \frac{\bar{\omega}^2}{1 - \bar{\omega}^2} \frac{\bar{X}^2}{N_s l(l-1)(l+2)} \]  

(3.9)

and Eq. (3.8) implies that the right hand side of Eq. (3.9) approaches one as \( \bar{\omega} \to 1.0 \). For example, for \( \bar{\omega} = 0.999938 \), and \( l = 2 \), from Eq. (3.12), \( \frac{\omega}{\omega^*} = 0.99726 \), and for \( \bar{\omega} = 0.980581 \), \( \frac{\omega}{\omega^*} = 0.959 \), i.e., the inviscid approximation gives an error of less than 5% for \( \bar{\omega} > 0.98 \).

Sometimes, damping rate information is difficult to obtain from the experiment (owing to time constraints on the experiment), or not available at all (for example, in the surface tension measurements from oscillation frequency measurements [9]). In such cases, if viscous effects are considerable, an error may result in the property evaluation. However, for mode 2 oscillations, if the viscosity is
Figure 3. Dependence of surface tension to viscosity parameter $N_{sv}$ on the damping rate and frequency of oscillations: Detail of behavior for $\bar{\omega} > 0.9$. $\bar{\omega}$ is the nondimensional frequency, $\bar{\omega} = \omega^2/\sqrt{\omega^2 + \tau^2}$. 
Figure 4. Dependence of surface tension to viscosity parameter $N_{sv}$ on the damping rate and frequency of oscillations for $\bar{\omega} \leq 0.9$. 
Figure 5. Dependence of nondimensional viscosity number $\tilde{\chi}=\tau R^2/\nu$ on the damping rate and frequency of oscillations.
known, a simple correction can be made to account for the viscous effects when \( \bar{\omega} < 0.98 \), where inviscid approximation may be inaccurate. This is done as follows.

From Figure 5, we observe that for \( l = 2 \), \( \bar{X} \) is virtually constant with \( \bar{\omega} \) (for \( \bar{\omega} < 0.98 \), an average \( \bar{X} \) may be used: \( \bar{X}_{av} = 3.62693 \), with a maximum error of 0.27). Thus, if the viscosity of the droplet is known apriori, the constant value of \( \bar{X} \) is used to find \( \tau \), and this \( \tau \) may be used in the determination of \( \bar{\omega} \). If \( \bar{\omega} \) thus calculated is greater than 0.98, the inviscid approximation may be used with less than 5\% error. If, however, it is less than 0.98, this value of \( \bar{\omega} \) may be used along with Figures 3 and 4 or Table I. to find \( N_{sv} \), from which the surface tension \( \gamma \) can be calculated within the upper and lower bounds of the error in \( \bar{X} \). This rather fortunate state of affairs prevails only in the case of mode 2 oscillations, and that too if \( v \) is already known. Thus, experimental requirements are somewhat eased, and the results of this work may be applied in cases where viscosity corrections are to be investigated, for example in the measurement of surface tension using electromagnetic levitation [9].

In summary, the procedure to find surface tension and viscosity from a single experiment may be described as follows:

1: From experimental observation of an oscillating levitated droplet, the damping rate \( \tau \), the frequency of oscillations \( \omega \), the radius \( R \) and the density \( \rho \) of the droplet are measured. The mode of oscillations \( l \) is also noted.
2: From Table I or Figure 5, the kinematic viscosity $\nu$ is determined from the nondimensional viscosity number $\bar{X}$, $\nu = \tau R^2 / \bar{X}$.

3: Once the viscosity is known, Table I or Figures 3 and 4 are consulted to obtain the surface tension to viscosity parameter $N_{sv}$, and from the known viscosity, the surface tension $\gamma$ is calculated as $\gamma = N_{sv} \nu^2 \rho / R$. For mode 2 oscillations, if damping rate is unavailable, a viscosity correction may also be applied as discussed earlier.

3.4. Conclusions

A numerical investigation of the damped oscillations of a viscous droplet in vacuum or a gas of negligible density has been performed. A continued fraction approach to the evaluation of the spherical Bessel function ratios, and a modified quasilinearization approach to the search for zeros of the characteristic equation has been used. The problem is completely defined by three nondimensional parameters, $\bar{\omega}$, $N_{sv}$, and $\bar{X}$. Using these results, the surface tension and viscosity of liquids can be computed from a knowledge of the damping rate and frequency of oscillations.

It must however, be remembered that the method outlined in this work applies only to free oscillations. It is also applicable to liquid droplets levitated acoustically or electromagnetically, in reduced gravity environments where the influence of the external forces and internal flows is minimal. One way to simulate this on earth is by setting a levitated droplet into oscillations, and dropping
the experiment in a long drop tube. But the effect of levitation forces and internal flows must be accounted for in real applications, before reliable surface tension and viscosity data can be obtained. However, in this chapter, it has been demonstrated that it is possible to determine surface tension and viscosity from a knowledge of damping rate and oscillation frequency.
CHAPTER 4
EFFECT OF STATIC DEFORMATION AND EXTERNAL FORCES
ON THE OSCILLATIONS OF LEVITATED DROPLETS

4.1. Introduction

Thermophysical property measurement methods based on levitation have been suggested in the earlier chapters. It has been noted in Chapters 2 and 3 that observation of damped dynamics of levitated droplets can be used to measure interfacial tension and viscosity. In the measurement of interfacial tension, for example, the droplet is typically levitated using one of the levitation methods, and the frequency of free or forced oscillations noted. An appropriate theory is then used to determine interfacial tension from the recorded frequency of oscillations of the droplet.

However, conventional analysis that accompanies such techniques presupposes oscillations about a spherical shape. Thus, surface tension measurement is based on the expression for the inviscid natural frequency,

\[ \omega_l^* = \frac{\gamma l (l - 1) (l + 2)}{\rho R^3} \]  

Equation (4.1) predicts a \((2l + 1)\)-fold degeneracy in the frequency spectrum. i.e., there is only one distinct peak, at the value of the natural frequency. This is not the case in actual experiments, where one often observes three peaks in the frequency spectrum [7,12] Yet, in an experiment, all but one of the peaks in the frequency spectrum are discarded, and further analysis is based on this peak, and
Eq. (4.1). There are several possible causes for the splitting of the spectrum. It could be a consequence of the inherent asymmetry of the levitation device, or the thermal instability (for high-temperature levitation), or the effect of asphericity and rotation on the droplet oscillations. Here, we are concerned with the effects of asphericity on droplet oscillations, and with determining whether asphericity can account for the splitting of spectra commonly observed in experiments.

Analytical treatment of aspherical droplet oscillations is scant in the literature [24,25]. Most of the previous studies, as noted earlier, are for a droplet executing shape oscillations about a spherical shape. Even when static deformation of the oscillating droplet is included, as in the case of Marston [18], the deformation effects are decoupled from the oscillations, and the influence of the time-independent deformation on the time-dependent oscillations is lost. In Cummings and Blackburn [24] and Suryanarayana and Bayazitoglu [25], the fundamental frequencies have been analytically shown to split. Cummings and Blackburn also study the effect of electromagnetic levitation on the oscillations of a droplet, for the fundamental \((l = 2)\) mode.

The object of this chapter is to develop a general theory for the dynamics of aspherical droplets subjected to external forces. Such a theory is useful in accounting for different external forces that usually act on levitated droplets, and interpreting frequency spectra more accurately. First, the effect of an arbitrary static shape deformation on the time-dependent oscillations of a liquid droplet is considered, without regard to its specific cause. Retaining the first-order effect of
the static deformation on the oscillations, the splitting of the frequency spectra is analytically calculated for the mode 2, 3, and 4 oscillations. The shape deformation is then assumed to be a consequence of an arbitrary external force, and the shifted frequencies are once again calculated. Acoustic, electromagnetic, and combined external levitating forces are considered as specific examples, and the frequencies of oscillation are determined in terms of the external force parameters. Finally, the interpretation of these spectra for surface tension measurements is discussed, and the results are applied to some previous experimental results to demonstrate the possible improvement in surface tension measurement.

4.2. Analysis

Consider an inviscid, incompressible droplet oscillating freely in a medium of negligible density and viscosity. The equation governing the behavior of the droplet is, simply,

$$\frac{\partial \mathbf{u}}{\partial t} = -\frac{1}{\rho} \nabla p ,$$

(4.2)

where the equation is written for the perturbed quantities. In Eq. (4.2), $\mathbf{u}$ is the perturbed velocity, $\rho$ the density, and $p$ the perturbed pressure. When this equation, along with the continuity condition, is solved, the radial velocity, $w$, is obtained as

$$w = \frac{1}{r} \sum_{lm} e^{-\beta t} a_{1r} r^l Y_{lm}(\Theta, \Phi) ,$$

(4.3)

where $r$ is the radial coordinate, and the $Y$'s are spherical harmonics. In addition, the dependence on time is assumed to be of the form $e^{-\beta t}$. The real part of $\beta$ is
the damping or amplification factor, with damping occurring for positive real part. The imaginary part is the frequency of oscillations. When $\beta$ is purely imaginary (such as in the inviscid case), undamped oscillations occur. It must also be noted that in Eq. (4.3), the summation $\sum_{lm}$ is over both $l$ and $m$, with

$$\sum_{lm} = \sum_{l=1}^{\infty} \sum_{m=-l}^{l}.$$  

The pressure may also be determined from the divergence of Eq. (4.2), and its radial component, and is given by

$$p = p_0 + \sum_{lm} \frac{r^l}{l} \beta \rho \ a_1 \ e^{-\beta l} \ Y_{lm}(\theta,\phi).$$  

(4.4)

Here, $p_0$ is the hydrostatic pressure.

In applying the boundary conditions, the effect of static shape deformation as well as time-dependent oscillations must be included. Therefore, it is assumed that the perturbed radius of the sphere may be expressed as an arbitrary change in the equilibrium radius $R$,

$$r = R + X(\theta,\phi) + \zeta(\theta,\phi,t), \quad \text{at the surface},$$  

(4.5)

where the static shape deformation $X(\theta,\phi)$ and the time-dependent distortion $\zeta(\theta,\phi,t)$ can be expanded in terms of independent spherical harmonics as

$$X(\theta,\phi) = \sum_{uv} X_{uv} Y_{uv}(\theta,\phi)$$  

(4.6a)

$$\zeta(\theta,\phi,t) = \sum_{lm} \zeta_{0lm} e^{-\beta t} Y_{lm}(\theta,\phi).$$  

(4.6b)

Thus, the surface of the sphere at any time is given by the equation
\[ \sigma = r - [R + X(\theta, \phi) + \zeta(\theta, \phi, t)] = 0. \]  

(4.7)

In what follows, subscripts \( l,m,u,v \) are dropped unless there is a possibility of ambiguity, and in writing the spherical harmonic functions, their dependence on the angles is understood.

The boundary conditions must now be applied. The kinematic condition simply states that

\[ w = \frac{\partial \zeta}{\partial t}, \quad \text{at the surface}. \]  

(4.8)

or

\[ \frac{1}{R + X + \zeta} \sum_{lm} e^{-\beta l} a_1 (R + X + \zeta) Y_{lm} = \sum_{lm} -\beta e^{-\beta l} Y_{lm}, \]

since Eq. (4.8) refers to the surface, \((R + X + \zeta)\). Expanding using the binomial theorem, neglecting terms of the order \( w \zeta, \zeta^2 \) and \( X^2 \), and employing the orthogonality of spherical harmonics,

\[ \iint Y_{lm} Y_{lm}^* \sin \theta \, d\theta \, d\phi = \delta_{ll'} \delta_{mm'}, \]  

(4.9)

the kinematic condition becomes

\[ \frac{1}{R^2} \sum_{lm} R^l [R + (l - 1)] \sum_{uv} \chi_{uv} \iint Y_{lm} Y_{lm}^* Y_{uv} \, ds \, e^{-\beta l} a_1 = \sum_{lm} -\beta \zeta_0 e^{-\beta l}. \]  

(4.10)

The normal force balance condition at the surface is written as

\[ \Delta p = \gamma \left( \frac{1}{R_1} + \frac{1}{R_2} \right) \]  

(4.11)

where \( \Delta p \), the pressure excess at the surface, is given by Eq. (3), and

\[ \frac{1}{R_1} + \frac{1}{R_2} = \nabla \cdot \hat{n} \]  

(4.12)

\( \hat{n} \) is the unit outward normal to the surface,
\[ \hat{n} = \frac{\nabla \sigma}{|\nabla \sigma|}, \quad (4.13) \]

and hence,

\[ \nabla \cdot \hat{n} = \frac{\nabla^2 \sigma}{|\nabla \sigma|} + \nabla \sigma \cdot \nabla \left( \frac{1}{|\nabla \sigma|} \right). \quad (4.14) \]

Using Eq. (4.14), \( 1/R_1 + 1/R_2 \) can be expressed in terms of \( \chi \) and \( \zeta \). Substituting for \( \sigma \) in Eq. (4.14), simplifying, and neglecting terms of the order \( \chi^2 \) and \( \zeta^2 \), we have,

\[ \nabla \cdot \hat{n} = \frac{2}{r} + \frac{1}{r^2} \hat{L}^2 (X + \zeta) \quad (4.15) \]

where \( \hat{L}^2 \) is the operator

\[ \hat{L}^2 = -\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left[ \sin \theta \frac{\partial}{\partial \theta} \right] - \frac{1}{\sin^2 \theta} \frac{\partial}{\partial \phi^2} \]

For the surface under consideration, Eq. (4.14), after neglecting higher order terms, becomes

\[ \nabla \cdot \hat{n} = \frac{2}{R} + \frac{1}{R^2} \left[ (\hat{L}^2 - 2)X + (\hat{L}^2 - 2)\zeta \right] + \frac{1}{R^3} (4X \zeta - 2X \hat{L}^2 \zeta + 2 \zeta \hat{L}^2 \chi) \quad (4.16) \]

In arriving at Eqs. (4.10) and (4.16), terms of order \( \chi^2 \), \( \zeta^2 \), and higher have been neglected. For small-amplitude oscillations and small static deformation, this is justified. Notice, however, that terms of order \( \chi \zeta \) have been retained. The \( \chi \zeta \) terms are the first-order effect of the static shape deformation on the time-dependent oscillation terms. If \( \chi \zeta \) terms were to be neglected, as has been done, for instance, by Marston [18], the problem would separate into a static part and a time-dependent part, each independent of the other. In such a case, the static deformation and the oscillations can both be calculated, but the solution would fail
to reveal the influence of this static deformation on the oscillations. Since $X \zeta$ represents the interrelation of the static and dynamic parts of the problem, the $X \zeta$ terms must be retained if this effect is of concern, as it is in this work.

Now substituting Eqs. (4.4) and (4.16) into Eq. (4.11), and simplifying, noting that $p_0 = 2\gamma/R$, we have the complete pressure balance equation:

$$\sum_{lm} \frac{R^{l-1}(R + IX)}{l} \beta \rho a_1 e^{-\beta t} Y_{lm} = \frac{\gamma}{R^2} \left[ (l-1)(l+2) \sum_{lm} \zeta_0 e^{-\beta t} Y_{lm} + \right.$$

$$+ (u-1)(u+2) \sum_{uv} \lambda_{uv} Y_{uv} + \frac{4}{R} \sum_{uv} \lambda_{uv} Y_{uv} \sum_{lm} \zeta_0 e^{-\beta t} Y_{lm} -$$

$$- \frac{2}{R} \left[ \sum_{uv} \lambda_{uv} Y_{uv} \left( l(l+1) \sum_{lm} \zeta_0 e^{-\beta t} Y_{lm} + \sum_{lm} \zeta_0 e^{-\beta t} Y_{lm} u(u+1) \sum_{uv} \lambda_{uv} Y_{uv} \right) \right] \right]$$

(4.17)

Once again, employing the orthogonality of spherical harmonics, the time-dependent part of Eq. (4.17) becomes

$$\sum_{lmuv} \frac{\beta \rho a_1 e^{-\beta t}}{l} Y_{lm} R^{-1} \left[ R + l \chi_{uv} \int \int Y_{lm} Y_{l'm}^* Y_{uv} ds \right] = \sum_{lm} \frac{\gamma}{R^2} \left( (l-1)(l-2) + \right.$$

$$+ \sum_{uv} \frac{2}{R} \chi_{uv} \left[ 2 - l(l+1) - u(u+1) \right] \int \int Y_{lm} Y_{l'm}^* Y_{uv} ds \right]$$

(4.18)

In Eqs. (4.10) and (4.18), we have integrals of the products of three spherical harmonics of the form

$$\int \int Y_{lm} Y_{l'm}^* Y_{uv} ds$$

(4.19)
in which integration is over all solid angles. Such integrals are common in quantum mechanics [38,39], and must satisfy the following conditions to be non-zero:

(i) $m' = m + \nu$
(ii) \( |u - l| \leq l' \leq |u + l| \)

(iii) \((l + u + l')\) is even

In this work, these integrals are evaluated using the Gaunt formula \([40]\) terms of the Wigner 3–j symbols \([38]\). The 3–j symbols are commonly used in the matrix representations of the addition of angular momenta in quantum mechanics, and, apart from their group theoretical importance, are very useful in manipulating complicated and cumbersome algebraic expressions. Appendix B has a brief discussion of the Gaunt formula, 3–j symbols and their calculations, and the integrals of the products of three spherical harmonics. Since we consider only axisymmetric deformations, \(v = 0\). From the conditions for the existence of the integrals of products of three spherical harmonics, we have, \(m = m'\), and \(l = l'\). Thus, the integrals depend only on \(l\), \(m\) and \(u\). We will refer to these integrals as \(I_{uln}^{(l)}\). Table I. gives a list of the integrals that are needed for this work.

4.3. Results and Discussion

A. The Frequencies of Oscillation

For an inviscid droplet, \(\beta\) is purely imaginary,

\[ \beta_{lm} = \pm i \omega_{lm} , \]

and Eqs. (4.10) and (4.18) must be solved simultaneously to obtain \(\omega_{lm}\). Eliminating \(a_1\) between the two equations and simplifying, the frequencies of oscillations are obtained as
Table I: Integrals of products of three spherical harmonics

\[ l = 2: \quad I^{(2)}_{wm} = \iint Y_{2m} Y_{u0} Y_{2m}^* \, ds \]

<table>
<thead>
<tr>
<th>( m )</th>
<th>( I^{(2)}_{2m} )</th>
<th>( I^{(2)}_{4m} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.18022375</td>
<td>0.24179554</td>
</tr>
<tr>
<td>± 1</td>
<td>0.09011188</td>
<td>-0.16119702</td>
</tr>
<tr>
<td>± 2</td>
<td>-0.18022375</td>
<td>0.04029926</td>
</tr>
</tbody>
</table>

\[ l = 3: \quad I^{(3)}_{wm} = \iint Y_{3m} Y_{u0} Y_{3m}^* \, ds \]

<table>
<thead>
<tr>
<th>( m )</th>
<th>( I^{(3)}_{3m} )</th>
<th>( I^{(3)}_{4m} )</th>
<th>( I^{(3)}_{6m} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.16820883</td>
<td>0.15386989</td>
<td>0.23708793</td>
</tr>
<tr>
<td>± 1</td>
<td>0.12615663</td>
<td>0.02564498</td>
<td>-0.17781595</td>
</tr>
<tr>
<td>± 2</td>
<td>0</td>
<td>-0.17951487</td>
<td>0.07112638</td>
</tr>
<tr>
<td>± 3</td>
<td>-0.21026104</td>
<td>0.07693494</td>
<td>-0.01185440</td>
</tr>
</tbody>
</table>

\[ l = 4: \quad I^{(4)}_{wm} = \iint Y_{4m} Y_{u0} Y_{4m}^* \, ds \]

<table>
<thead>
<tr>
<th>( m )</th>
<th>( I^{(4)}_{2m} )</th>
<th>( I^{(4)}_{4m} )</th>
<th>( I^{(4)}_{6m} )</th>
<th>( I^{(4)}_{8m} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.16383977</td>
<td>0.13696111</td>
<td>0.14225276</td>
<td>0.23443943</td>
</tr>
<tr>
<td>± 1</td>
<td>0.13926381</td>
<td>0.06848055</td>
<td>-0.00711264</td>
<td>-0.18755154</td>
</tr>
<tr>
<td>± 2</td>
<td>0.06553591</td>
<td>-0.08369846</td>
<td>-0.15647804</td>
<td>0.09377577</td>
</tr>
<tr>
<td>± 3</td>
<td>-0.05734392</td>
<td>-0.15978796</td>
<td>0.12091485</td>
<td>-0.02679308</td>
</tr>
<tr>
<td>± 4</td>
<td>-0.22937568</td>
<td>0.10652531</td>
<td>-0.02845055</td>
<td>0.00334914</td>
</tr>
</tbody>
</table>

\( a \) For values of \( u \) other than those listed in the table, the integral is zero.
\[(\omega_{ln})^2 = \omega_I^2 - \frac{\gamma}{\rho R^3} \left[ 3l(l-1)(l+2) + 2l\mu(l+1) \right] \sum \frac{\chi_{uv}I_{lm}^{(l)}}{R} \] (4.20)

where \(\omega_I^*\) is the natural frequency of an inviscid spherical droplet.

Before attempting to calculate the frequencies for any mode, two limiting conditions are applied to Eq. (4.20). When there is no static shape deformations (the \(\chi\)'s are zero), the solution reduces to Eq. (4.1), as expected. When the deformation is very small, i.e., \(X = \delta R\),

\[-\beta^2 = \frac{\gamma}{\rho R^3} \left[ l(l-1)(l+2)(1 - 3\delta R/R) \right],\]

or

\[\delta \beta^2 = -3l(l-1)(l+2) \delta R \left[ \frac{\gamma}{\rho R^4} \right] \] (4.21)

Eq. (4.21) is the result we get on directly differentiating Eq. (4.1).

As an instructive example, the oscillation frequencies for the fundamental \((l = 2)\) mode are calculated using Eq. (4.20). From Appendix B, for \(l = 2\),

\[\sum_{uv} \chi_{uv} \int y_{lm}^* y_{lm}^* y_{uv} = \chi_{20} I_{2m}^{(2)} + \chi_{4m} I_{4m}^{(2)} \] (4.22)

Using Table I, the values of \(I_{2m}^{(2)}\) and \(I_{4m}^{(2)}\) for \(m = 0, \pm 1, \text{ and } \pm 2\) can now be substituted into Eq. (4.20), and the frequencies are:

\[\omega_{2,0} = \omega_I^* \left[ 1 - \frac{1}{R} (0.5406712\chi_{20} + 1.571670971\chi_{40}) \right] \] (4.23a)

\[\omega_{2,\pm 1} = \omega_I^* \left[ 1 - \frac{1}{R} (0.27035662\chi_{20} - 1.04778186\chi_{40}) \right] \] (4.23b)

\[\omega_{2,\pm 2} = \omega_I^* \left[ 1 - \frac{1}{R} (-0.5406712\chi_{20} + 0.261945125\chi_{40}) \right] \] (4.23c)
We note that there is double degeneracy for \( m = \pm 1 \) and \( m = \pm 2 \). In Appendix C, the frequencies for \( l = 3 \) and \( l = 4 \) have been calculated. We note that in all these cases, the average frequency is the same as the inviscid frequency, for

\[
\frac{1}{2l + 1} \sum_{m=-l}^{l} \omega_{lm} = \omega^*_l
\]

(4.24)

This can be exploited in \( \gamma \) measurements.

To see the actual effect for specific distortions, we consider three examples for Eq. (4.22), \( \chi_{20} = 0.1 \, R \), \( \chi_{40} = 0 \); \( \chi_{20} = 0 \), \( \chi_{40} = 0.1 \, R \); and \( \chi_{20} = 0.05 \, R \), \( \chi_{40} = 0.05 \, R \). Figure 1. shows the deformation that is caused by such an assumed distortion, for a sphere of unit radius. The solid figure is the undistorted sphere. The \( \chi_{20} \) deformation is a prolate-oblative one, with oblate distortion for negative \( \chi_{20} \). This is the quadrupole deformation of a suspended droplet. The \( \chi_{40} \) deformation is a mode-4 type deformation, with the typical four-cornered shape. By suitably choosing the values of the \( \chi \)'s, distortions of different nature can be incorporated. Table II summarizes the frequency results for these distortions.

From Table II, we observe that the spectrum need not split about \( \omega^* \), the inviscid frequency. In fact, the spectrum need not even split into three peaks. It is possible to have distortions for which only two peaks occur in the frequency spectrum (for certain values of \( \chi_{20} \) and \( \chi_{40} \), one of the modes has triple or quadruple degeneracy). However, there are some \( \chi \)'s for which the split is symmetrical, i.e., the bandwidths are equal. For example, for \( \chi_{20} = 0.1R \) and \( \chi_{40} = -0.0344R \), \( \omega_{20} \) coincides with \( \omega^*_2 \), with \( \omega^*_{2\pm1} \) and \( \omega^*_{2\pm2} \) splitting symmetrically about \( \omega^* \).
Figure 1. Distortions of the spherical shape for a) $\chi_{20} = 0.1R$, $\chi_{40} = 0$; b) $\chi_{20} = 0$, $\chi_{40} = 0.1R$; and c) $\chi_{20} = 0.05R$, $\chi_{40} = 0.05R$. Solid lines represent the unit sphere being distorted.
Table II. Frequency Splitting for Different Distortions of the Droplet

<table>
<thead>
<tr>
<th>$\chi_2/R$</th>
<th>$\chi_4/R$</th>
<th>$\beta_{2,0}/\omega^*$</th>
<th>$\beta_{2,\pm1}/\omega^*$</th>
<th>$\beta_{2,\pm2}/\omega^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>0</td>
<td>0.945933</td>
<td>0.9729644</td>
<td>1.054067</td>
</tr>
<tr>
<td>0</td>
<td>0.1</td>
<td>0.8428329</td>
<td>1.1047782</td>
<td>0.973806</td>
</tr>
<tr>
<td>0.05</td>
<td>0.05</td>
<td>0.894383</td>
<td>1.0388713</td>
<td>1.0139363</td>
</tr>
<tr>
<td>0.1</td>
<td>-0.034</td>
<td>1.0000</td>
<td>0.9369207</td>
<td>1.063079</td>
</tr>
</tbody>
</table>
B. Inclusion of Surface Force

Let us assume an arbitrary force acting at the surface of the droplet, and causing a distortion of the droplet static shape. In general, the force can be expanded in terms of spherical harmonics as,

$$ P_{\text{ext}} = \sum C_{l,m}^{n} \left[ \frac{R + X + \zeta}{R} \right]^{n} Y_{lm}(\theta, \phi) \quad (4.25) $$

This is a Taylor-series expansion of an arbitrary function about the origin in spherical coordinates, and $P_{\text{ext}}$ is assumed to be sufficiently continuous. The surface condition,

$$ \delta p = \gamma \left[ \frac{1}{R_1} + \frac{1}{R_2} \right] + P_{\text{ext}} \quad (4.26) $$

can now be applied. The complete surface condition becomes

$$ p_{o} + \sum_{lm} \frac{R^{l-1}(R + IX)}{l} \beta \rho a_{1} e^{-\beta t} Y_{lm} = \frac{2\gamma}{R} + \frac{\gamma}{R^2} \left[ (l-1)(l+2) \sum_{lm} \xi_{0} e^{-\beta t} Y_{lm} + 
$$

$$ + \frac{4}{R} \sum_{uv} \chi_{uv} Y_{uv} \sum_{lm} \xi_{0} e^{-\beta t} Y_{lm} - \frac{2}{R} \left[ \sum_{uv} \chi_{uv} Y_{uv} \right] (l+1) \sum_{lm} \xi_{0} e^{-\beta t} Y_{lm} + 
$$

$$ + \sum_{lm} \xi_{0} e^{-\beta t} Y_{lm} \sum_{uv} \chi_{uv} Y_{uv} \right] + (u-1)(u+2) \sum_{uv} \chi_{uv} Y_{uv} \right] + 
$$

$$ + \sum C_{l,m}^{n} \left[ 1 + \frac{n}{R} X + \frac{n}{R} \zeta + \ldots \right] Y_{lm} \quad (4.27) $$

For the time-independent part, from the zeroth term comparison,

$$ p_{0} = \frac{2\gamma}{R} + \sum_{n} C_{0,0}^{n} \quad (4.28) $$

The $C_{0,0}^{n}$ term is the external force contribution to the static surface energy. The $l=1, m = 0$ terms give $C_{1,0}^{n} = 0$. If gravity were present, the gravitational term
would be balanced by this \( l = 1 \) term, \( \rho_0 g z = \sum_n C^n_{1,0} Y_{10} \).

The remaining time-independent terms, to first order, give a relationship between the external force and the distortion coefficients:

\[
\sum \chi_{uv} = -\frac{R^2}{\gamma} \frac{\sum C^n_{uv}}{(u-1)(u+2)} \quad u > 1. \tag{4.29}
\]

Thus, all the distortion terms in the characteristic equation may now be expressed in terms of the coefficients of the external force, using Eq. (4.29).

The time-dependent terms in Eq. (4.27) give, after simplification,

\[
\omega_{lm}^2 = \frac{\gamma}{\rho R^3} \left[ l(l-1)(l+2) - \frac{3l(l-1)(l+2)}{R} \sum \chi_{uv} I_{um}^{(l)} \right]
- \frac{2l}{R} \frac{u(u+1)}{u} \sum \chi_{uv} I_{um}^{(l)} + \frac{l}{\rho R^2} \sum C^n_{uv} n I_{um}^{(l)} \tag{4.30}
\]

Using Eq. (4.29), Eq. (4.30) can be re-written in terms of the \( C^n_{uv} \)'s of the external force, as

\[
\omega_{lm}^2 = \frac{\gamma}{\rho R^3} l(l-1)(l+2) + \frac{1}{\rho R^2} \sum_n C^n_{uv} \left[ \frac{3l(l-1)(l+2)}{(u-1)(u+2)} I_{um}^{(l)} + \frac{2l}{(u-1)(u+2)} I_{um}^{(l)} \right]
+ \frac{1}{\rho R^2} l \sum n C^n_{uv} I_{um}^{(l)} \tag{4.31}
\]

The \( l = 1 \) frequencies are the translational frequencies. They determine the global position of a droplet inside a levitating chamber, and neither distort the droplet shape, nor cause a time-dependent change in the droplet radius. For \( l = 1 \), \( u = 0 \) and \( u = 2 \) are the only two cases for which the integral exists. Using Eq. (4.31), and noting that \( \sum \int Y_{1m} Y_{2v} Y_{1m}^* ds = 0 \), the translational frequencies are found to be
\[ \omega_i^2 = \sqrt{\frac{1}{4\pi}} \frac{1}{\rho R^2} \sum_n nC_{0,0}^n \]  

(4.32)

The oscillation frequencies can be calculated in terms of the external forces using Eq. (4.31) for any \( l > 1 \). For the fundamental \( (l = 2) \) frequencies, for example, we have

\[ \omega_{2m}^2 = \frac{8\gamma}{\rho R^3} + 2 \sqrt{\frac{1}{4\pi}} \frac{1}{\rho R^2} \sum_n nC_{0,0}^n \quad \sum_n C_{wn}^n \left[ 12I_{2m} + 5.778I_{4m} \right] + \]

\[ + \frac{1}{\rho R^2} \sum_n \left[ C_{20}^n I_{2m} + C_{40}^n I_{4m} \right] \]

(4.33)

or,

\[ \omega_{2m}^2 = \omega_2^{*2} + 2 \omega_i^2 \pm \omega_f^2 \]

(4.34)

Thus, in the presence of external forces, the translational frequencies also influence the oscillation frequencies, unlike in the case of Eq. (4.20), where no external forces are considered, and \( \omega_{2m}^2 = \omega_2^{*2} \pm \omega_f^2 \).

In order to understand exactly the effect of the external forces, the \( C_{uv}^n \) coefficients must be calculated from the external forces. This is also necessary if the translational frequencies, and/or the droplet position are to be determined. This is a formidable task, and is not attempted here, since the main concern of this work is the frequencies of oscillation. To this end, a reasonable first-order solution may be obtained if we consider \( n = 0 \) in the \( C_{uv}^n \), i.e., the first term in the series. This is equivalent to making two approximations: (i) that there are no translational frequencies, and (ii) that the external force is approximated by the force that would be present at the surface if the droplet were perfectly spherical.
These approximations are perfectly valid in our case, where the frequency spectra and not the position or translation of the droplet, are of importance.

With \( n = 0 \), the first order solution becomes,

\[
-\beta_{lm}^2 = \omega_l^2 + \frac{1}{\rho R^2} C_{uv} \left[ \frac{3l(l-1)(l+2)}{(u-1)(u+2)} + \frac{2l}{R} \frac{u(u+1)}{(u-1)(u+2)} \right] I_{lm}^{(l)}
\]

(4.35)

Now, the time independent parts lead to

\[
P_0 = \frac{2\gamma}{R}
\]

(4.36a)

and

\[
\chi_{uv} = -\frac{R^2}{\gamma} \frac{1}{(u-1)(u+2)} C_{uv}
\]

(4.36b)

and the translational frequencies are lost, as was observed earlier.

It now remains to consider specific types of external forces. Here, we consider acoustic, electromagnetic, and combined external forces.

C. Acoustic surface force

When an ultrasonic standing wave is generated in a chamber, a droplet can be levitated at the pressure nodes. By modulating the standing wave, the droplet may be forced into oscillations at the modulation frequency. Levitation, and forced oscillation of the droplet, are a consequence of the modulated radiation stresses at the droplet surface. Acoustic levitation and modulated shape oscillation are well developed techniques, and have received considerable attention [7,8,18,22]. They have been applied to the measurement of various thermophysical properties, including surface tension [8].
The theory of an acoustically levitated droplet was first considered by Marston [18]. In describing the behavior of the droplet, Marston first calculates the radiation pressure by solving the wave equation in the vicinity of the droplet surface, and then uses it in calculating the second-order radiation stresses that appear in the force-balance boundary conditions at the interface. The acoustic waves result in a radiation stress tensor, the radial projection of which is given (at the droplet interface) as

$$\bar{P}_r = \Pi_{rr}^{i} - \Pi_{rr}^{o}$$  \hspace{1cm} (4.37a)

where the superscripts $i$ and $o$ refer to the evaluation of the stress tensor in the inner and the outer medium respectively. Using a spherical harmonic expansion of $\bar{P}_r$, the radial projection of the stress is shown to be dominated by the quadrupole term ($l = 2, m = 0$), which is given by

$$P_{20}^r = (3P_s^2 \beta_0 \sqrt{5\pi}/20) \left[ 1 + \frac{7}{5} (k_{oc} R)^2 + \ldots \right]$$  \hspace{1cm} (4.37b)

for a surrounding medium of negligible density and viscosity, and an unmodulated acoustic wave. In Eq. (4.37b), $P_s$ is the amplitude of the standing wave, $k_{oc}$ is the ratio of the wave frequency to the sound speed in the external medium, and $\beta_0$ is the adiabatic compressibility of the outer medium. It must be noted that in Marston [18] and other workers following him, the R. H. S. of Eq. (4.37b) is negative, due to a difference in sign convention. Whereas in those works, a positive $\bar{P}_r$ represents a radially outward force on the interface, it is clear that in this work, a positive $P_{ca}$ represents a radially inward force, as borne out by Eq. (4.26).
Supposing the radial projection of the acoustic radiation pressure to be the
levitating force, and supposing that it can be expanded in spherical harmonics
according to Eq. (4.25), we have,

$$
\bar{P}_r = \sum_{l \geq 0, m} P^r_{lm} Y_{lm}.
$$

This is the first order representation of the radiation pressure. Or, from Eq. (4.29),

$$
\chi_{uv} = -\frac{R^2}{\gamma} \frac{1}{(u-1)(u+2)} P^r_{uv}
$$

Equation (4.38) is identical with the static deformation calculated by Marston,
although in his case, the deformation is decoupled from the time-dependent
behavior. If $P^r_{20}$ is the dominant term in the radiation pressure, we have, from
Eq. (4.35),

$$
\omega^2_{lm} = \omega^* + \frac{1}{\rho R^2} P^r_{20} \left[ \frac{3}{4} l(l-1)(l+2) + 3l \right] I^l_{2m}
$$

It must be noted that since $P^r_{20}$ is assumed to be the dominant term in the acous-
tic radiation pressure projection, the only distortion coefficient responsible for the
static shape deformation is $\chi_{20}$. As a result, distortions of the form

$$
X = \chi_{20} Y_{20}
$$

are the only ones that can be accommodated. These are prolate-oblate distortions,
(Fig. 1a), and do, in fact, completely represent the flattening at the poles that is
commonly observed in acoustic levitation. The negative $\chi_{20}$ in Eq. (4.38) indi-
cates an oblate distortion for typical acoustic levitation— a fact that has been
observed repeatedly in experiments.
Using Eq. (4.37b) in Eq. (4.39), we have, for the fundamental \((l = 2)\) mode of an acoustically levitated aspherical droplet,

\[
\omega_{2,0}^2 = \omega_2^* + \frac{1.285714288}{\rho R^2} P_s^2 \beta_0 \left[ 1 + \frac{7}{5} (k_{0c} R)^2 \right] \quad (4.40a)
\]

\[
\omega_{2,\pm1}^2 = \omega_2^* + \frac{0.642857144}{\rho R^2} P_s^2 \beta_0 \left[ 1 + \frac{7}{5} (k_{0c} R)^2 \right] \quad (4.40b)
\]

\[
\omega_{2,\pm2}^2 = \omega_2^* - \frac{1.285714288}{\rho R^2} P_s^2 \beta_0 \left[ 1 + \frac{7}{5} (k_{0c} R)^2 \right] \quad (4.40c)
\]

Clearly, the oscillations of an acoustically levitated droplet reveal a triple split in the frequency spectrum for the fundamental \((l = 2)\) mode. Eq. (4.24) is valid, and the average of the five frequencies is \(\omega_l^*\), the inviscid frequency. From Eqs. (4.40), it is also apparent that the \(m = 0\) frequency is shifted upward as a result of the distortion caused by the acoustic forces. Of the three peaks, two lie always above, and the third, always below the inviscid value, with the \(m = \pm1\) mode being closest to the inviscid value.

The positive shift in the axisymmetric \((m = 0)\) frequency due to oblate distortion has been observed before. Trinh et. al. [22] have found experimentally that in immiscible liquid systems, oblate distortion leads to an increase in the axisymmetric frequency. Busse [41] has considered the oscillations of a rotating droplet theoretically, by accounting for the rotational distortion and Coriolis force on the droplet shape. He concludes that in the case of an oblate droplet, a further increase of the frequency of oscillations results, whereas in a prolate droplet, the effect of the distortion is opposite in direction. Similar trends are predicted by Eq.
(4.40a), which represents the axisymmetric (2,0) mode.

D. Electromagnetic surface force

When a metal specimen is placed in the field of an oppositely wound coil (a solenoidal coil with the upper few turns turned in the opposite direction), it levitates due to the Lorentz forces, and melts due to ohmic heating. The Lorentz force is a body force that results from the interaction of the induced current with the magnetic field, and is given by

\[ \mathbf{F} = \mathbf{J} \times \mathbf{B} \]

where \( \mathbf{J} \) is the current density, and \( \mathbf{B} \) the flux density. For large frequencies, the depth of penetration of the induced field (the skin depth) is negligibly small [42], and the droplet is essentially supported by a mean magnetic pressure at the surface,

\[ P_{mag} = \frac{B_s^2}{2\mu} \quad (4.41) \]

where \( B_s \) is the flux density on the droplet surface, to be obtained from a solution of the magnetic problem, and \( \mu \) is the permeability of the droplet material.

A simple model for the electromagnetic field around a levitated magnetic material is to assume that the applied field \( H_z \) varies linearly with axial distance \( z \), and that the force varies linearly with the product of the field and its derivative in the \( z \) direction. For electromagnetic levitation, this is a good assumption [29].

For a magnetic material, the field \( \mathbf{B} \) due to the introduction of a magnetic material into an undistorted applied field \( \mathbf{H} \), is
\[ \mathbf{B} = \mu_0 \mathbf{H} + \mu_0 \mathbf{M} \]  

(4.42)

where \( \mu_0 \) is the permeability of the medium, and \( \mathbf{M} \) is the intensity of magnetization. Now, from Maxwell's equations,

\[ \nabla \cdot \mathbf{B} = 0 \]  

(4.43a)

and

\[ \nabla \times \mathbf{B} = \mu_0 \mathbf{J}_0 \]  

(4.43b)

Assuming the current outside the droplet to be zero, the curl of \( \mathbf{B} \) is also zero, and hence, we can define a scalar magnetic potential \( \Phi \),

\[ \mathbf{B} = \nabla \Phi. \]

From Eq. (4.43a),

\[ \nabla^2 \Phi = 0 \]

and \( \Phi \), in spherical coordinates, is given by

\[ \Phi = \sum_{l=0}^{\infty} f_{lm}(r) Y_{lm}(\theta, \phi) \]  

(4.44a)

where the \( r \) dependent terms are

\[ f_{lm}(r) = \left[ a_{lm} r^l + b_{lm} r^{-l-1} \right] \]  

(4.44b)

From the condition that no current penetrates the surface (\( \mathbf{n} \cdot \mathbf{B} = 0 \)) we have,

\[ a_{lm} \frac{l}{l+1} r^{2l+1} = b_{lm} \]  

(4.45)

From the assumption of a linear field,

\[ \mathbf{H} = -z_0 \frac{\partial H_z}{\partial z} \hat{k} + \frac{1}{2} \frac{\partial H_z}{\partial z} \left( -x \hat{i} - y \hat{j} + 2z \hat{k} \right) \]  

(4.46)

where \( -z_0 \) is the levitation height. Taking the \( r \) component of Eq. (4.46), and comparing it with the \( r \) component of \( \mathbf{B} \), we have
\[ a_{10} = -\sqrt{\frac{4\pi}{3}} \mu_0 \frac{\partial H_z}{\partial z} \]  
(4.47a)

and

\[ a_{20} = \frac{1}{2\sqrt{5}} \frac{4\pi}{\mu_0} \frac{\partial H_z}{\partial z} \]  
(4.47b)

\(\mathbf{B}\) has only a tangential component, and Eq. (4.41) reduces to

\[ P_{mag} = \frac{-1}{2\mu R^2} (\mathbf{\hat{L}} \Phi \cdot \mathbf{\hat{L}} \Phi) \]  
(4.48)

Using the identity

\[ 2 (\mathbf{\hat{L}} f \cdot \mathbf{\hat{L}} g) = \mathbf{\hat{L}}^2 (fg) - f \mathbf{\hat{L}}^2 g - g \mathbf{\hat{L}}^2 f \]

and substituting for \(\Phi\) from Eq. (4.44), and simplifying, we have

\[ P_{mag} = \frac{-1}{2\mu R^2} \sum_{lm} \sum_{jk} \frac{f_{jk} f_{lm}}{2} \left[ u (u+1) Y_{uv} \int \int Y_{uv}^* Y_{lm} Y_{jk} \, ds - \right. \\
- l(l+1) Y_{jk} Y_{lm} - j(j+1) Y_{jk} Y_{lm} \left. \right], \]  
(4.49)

where \(f_{jk}\) and \(f_{lm}\) are the \(r\) dependent terms in \(\Phi\), given by Eq. (4.44b). The external magnetic force can be expanded according to Eq. (4.25), which, to first order, becomes

\[ P_{mag} = \sum_{uv} P_{uv} Y_{uv} \]  
(4.50)

Now, comparing Eqs. (4.49) and (4.50), and using orthogonality, we have

\[ P_{uv} = \frac{f_{jk} f_{lm}}{4\mu R^2} [l(l+1) + j(j+1) - u (u+1)] \int \int Y_{jk} Y_{lm} Y_{uv}^* \, ds. \]  
(4.51)

From Eqs. (4.47), the only conditions under which \(f_{jk}\) and \(f_{lm}\) exist are for \(j = 1,2, k = 0,\) and \(l = 1,2, m = 0\). Thus, from the restrictions on the values of \(u\) and \(v\), five non-zero terms, \(P_{00}, P_{10}, P_{20}, P_{30}, P_{40}\), and \(P_{50}\) are recognized. These are given in Appendix D.
The $P_{00}$ term appears as the additional surface energy in the hydrostatic force balance, and the $P_{10}$ term balances gravitation. These terms do not affect the frequencies or the distortion. In the electromagnetic case, there is an additional distortion term, associated with $\chi_{30}Y_{30}$. This is shown in Fig. 2, and corresponds to the pear-shaped distortion commonly observed in electromagnetically levitated droplets. The complete shape-deformation can now be represented as

$$X = \chi_{20}Y_{20} + \chi_{30}Y_{30} + \chi_{40}Y_{40} \quad (4.52)$$

The frequencies of oscillation are, from Eq. (4.35),

$$\omega_{lm}^2 = \omega^* + \frac{1}{\rho R^2} \left\{ P_{20}I_{2m}^{(l)} \left[ \frac{3l(l-1)(l+2)}{4} + 3l \right] + P_{40}I_{4m}^{(l)} \left[ \frac{3l(l-1)(l+2)}{18} + \frac{20}{9}l \right] \right\}. \quad (4.53)$$

Note that although the $P_{30}$ term influences the distortion through $\chi_{30}$, it does not affect the frequencies. The pear-shaped distortion is a consequence of the balance between the droplet's own weight, and the levitating basket, and is a static phenomenon. Substituting for $P_{20}$ and $P_{40}$ from Appendix D and simplifying, Eq. (4.53) reduces to

$$\omega_{lm}^2 = \omega^* + \frac{\pi}{4\mu R^4} \mu_0 \left\{ \frac{\partial H_z}{\partial z} \right\}^2 \left\{ \left[ -0.37846988z^2 R^2 + 0.1501865R^4 \right] I_{2m}^{(l)} - \left[ 0.2686617R^4 \right] I_{4m}^{(l)} \right\}. \quad (4.54)$$

Eq. (4.54) can now be used to find the oscillation frequencies of an electromagnetically levitated droplet, for any oscillation mode. For the fundamental $(l = 2)$ mode, for instance, the frequencies are
Figure 2. The $\chi_{30}Y_{30}$ distortion for an electromagnetically levitated droplet.
\[
\begin{align*}
\omega_{z,0}^2 &= \omega^* + C_{\text{mag}} (-0.81851132 \ z_0^2 R^2 - 0.0505257 \ R^4) \quad (4.55a) \\
\omega_{z, \pm1}^2 &= \omega^* + C_{\text{mag}} (-0.40925559 \ z_0^2 R^2 + 0.41262397 \ R^4) \quad (4.55b) \\
\omega_{z, \pm2}^2 &= \omega^* + C_{\text{mag}} (0.81851132 \ z_0^2 R^2 - 0.38736133 \ R^4) \quad (4.55c)
\end{align*}
\]
where \(C_{\text{mag}}\) is the magnetic parameter,

\[
C_{\text{mag}} = \frac{\pi}{4\mu_0 R^4} \mu_0^2 \left( \frac{\partial H_z}{\partial z} \right)^2 \quad (4.55d)
\]

Once again, Eq. (4.24) holds, and the average frequency is \(\omega^*\). However, it is no longer certain that there will be a triple split. Depending on the value of \(R\) and \(z_0\), the split may even be double. It is also no longer possible to ascertain the relative placement of the peaks in the frequency spectrum.

E. Interpretation of spectra and surface tension determination

As the above analysis shows, the asphericity of the oscillating droplet adequately explains the splitting of the frequency spectrum into \((2l + 1)\) bands. The problem of analyzing these bands, and describing a method to improve surface tension measurement, is considered in this section.

Given a frequency spectrum, once the mode \(m\) and degeneracy of each peak is recognized, and the mode \(l\) of the oscillations themselves is observed, Eq. (4.24) can be used to evaluate the Rayleigh frequency from the frequency peaks in the spectrum. The Rayleigh frequency is a unique measure of the surface tension of a droplet of known radius and density. Since the droplet is distorted, the radius used is that of a sphere of equal volume. In essence, the problem reduces to an assigning task.
This task of assigning the peaks to the right mode $m$ and degeneracy is simplified in the case of acoustically levitated droplets, irrespective of the oscillation mode $l$. Consider Eqs. (4.40). Since $P_{20}$ is the dominant radial projection, only one additive term affects the frequency, and the peaks can be assigned by observing that

$$\omega_{2,0} \geq \omega_{2,\pm1} \geq \omega_{2,\pm2}.$$ 

Once these peaks are assigned, Eq. (4.24) is applied, and $\omega^*$, and hence $\gamma$ are determined. This procedure is applied to the experimental results of Trinh et. al. [8] to demonstrate the improvement in the surface tension measurements.

In Trinh, only one three-band frequency spectrum is given. This is for an acoustically levitated water droplet, with initial lateral dimension of 1.98 mm and equilibrium axis ratio of 1.25, at a temperature of 24$^o$ C. Three peaks, at 105 Hz, 129 Hz and 158 Hz were observed. However, only the main peak (at 129 Hz) was used in evaluating $\gamma$. The $\gamma$ value thus evaluated had a 10% error.

For this spectrum, using the results described in this section, $\omega_{2,0}/2\pi = 158$, $\omega_{2,\pm1}/2\pi = 129$, $\omega_{2,\pm2}/2\pi = 105$. Using Eq. (24) to calculate $\omega^*$, and hence $\gamma$, we have $\gamma = 71.97$ dyn/cm, which is an error of 1.3% from the expected value of 71 dyn/cm. Thus, in the measurement of surface tension using acoustically levitated droplets, it is possible to improve the accuracy of $\gamma$ remarkably by using the results of this work in interpreting the spectra. Although this single instance does not prove the fact conclusively, it does indicate how this non-axisymmetric analysis can be applied to experimental data, and how spectral data that is
normally discarded can be used in the interpretation of the spectra.

In the case of electromagnetically levitated droplets, however, this task of assigning peaks is impossible, unless information about the magnetic and electrical parameters is available. From Eqs. (4.54) and (4.55), we note that there is no certainty of the placement of the peaks, since, theoretically, any of the peaks can be the doubly-degenerate ones. In fact, as has been observed earlier, the degeneracy of a peak may be three or even four-fold. Thus, Eq. (4.24) can be used only to give upper and lower bounds for the measured surface tension. To order of magnitude, since \( z_0^2 R^2 \sim R^4 \), a surface tension value may also be determined using Eq. (4.24). However, if the accuracy in measuring \( \gamma \) is to be improved, the electromagnetic parameters must be measured (and \( z_0 \) calculated). Further work is necessary before the frequency spectra of electromagnetically levitated droplets may be interpreted better for accurate surface tension determination.

F. Combined acoustic-electromagnetic surface forces

In the recent times, the prospect of an acoustic-electromagnetic hybrid levitator, which combines the advantages of either method, has become increasingly attractive [43]. Such a system would be more versatile, able to handle various materials at a wider range of temperatures, and capable of exciting controlled oscillations. A droplet levitated in such a system must also be distorted, depending on the magnitude of the acoustic and electromagnetic forces. In this section, we calculate the frequencies of oscillation of a droplet levitated in such a system.
When both acoustic and electromagnetic forces are present, it is reasonable to assume that they affect the droplet shape independently. The acoustic force is a pressure wave, and the electromagnetic force is a consequence of the interaction between the induced current and the magnetic field. The only way they interact is in the flow problem, in the force-balance boundary conditions. Thus, in this case, the two forces can be superimposed in the pressure balance boundary condition,

$$\Delta P = \gamma \nabla \cdot \mathbf{f} + P_{mag} + P_{ac}$$ (4.56)

Since both the forces are independent, they are expanded in independent series according to Eq. (4.25), with two different coefficients, $C_{uv}^{n}$, and $D_{jk}^{n}$, corresponding to the acoustic and the electromagnetic terms, respectively. Thus Eq. (4.27) has an additional external force term in the unknown coefficients $D_{jk}$, and the comparison of terms in the complete pressure balance equation simply gives additional terms in the $D_{jk}^{n}$ coefficients.

$$p_0 = \frac{2\gamma}{R} + \sum_n C_{0,0}^{n} + \sum_n D_{0,0}^{n}$$ (4.57)

$$\beta_{x}^2 = \sqrt{\frac{1}{4\pi}} \frac{1}{\rho R^2} \left[ \sum_n n C_{0,0}^{n} + \sum_n n D_{0,0}^{n} \right]$$ (4.58)

and

$$\sum x_{uv} = -\frac{R^2}{\gamma} \frac{\sum C_{uv}^{n} + \sum D_{uv}^{n}}{(u-1)(u+2)}$$ (4.59)

To first order, Eq. (4.57) reduces to Eq. (4.36a), Eq. (4.58) vanishes, and an additional term appears in Eq. (4.36b),
\[ \sum \chi_{uv} = -\frac{R^2}{\gamma} \frac{(C_{uv} + D_{uv})}{(u-1)(u+2)} \] (4.60)

where \( C_{uv} \) and \( D_{uv} \) are substituted from the acoustic and electromagnetic force considerations of the previous sections. Thus, the combined force terms are just superpositions of the acoustic and electromagnetic force terms, respectively. The final frequency results for a droplet in a combined force-field are given by

\[ \omega_{lm}^2 = \omega^*^2 + \frac{1}{\rho R^2} I_{2n}^{(l)} \left[ \frac{3l(l-1)(l+2)}{4} + 3l \right] (P_{20} + P_{20}') + \]
\[ + \frac{1}{\rho R^2} I_{4m}^{(l)} \left[ \frac{3l(l-1)(l+2)}{18} + \frac{20}{9} l \right] P_{40} \] (4.61)

where \( P_{20}' \) is given by Eq. (4.37b), and \( P_{20} \) and \( P_{40} \) are given in appendix D.

Once again, Eq. (4.24) holds, and this theory can be used to include all the peaks in the spectrum, as demonstrated in section E, if all the peaks can be assigned. However, since electromagnetic forces are also present, assignment of peaks becomes difficult, as observed in section D. Further work is necessary to fully understand the implications of the presence of electromagnetic forces in levitating droplets.

4.4. Conclusions

In this chapter, the oscillations of an aspherical inviscid droplet subjected to acoustic, electromagnetic, and combined external forces have been considered in detail. The purpose of this work was to explain the frequency splitting frequently observed in experiments and to interpret these split spectra more accurately in order to improve the surface tension measurements using oscillations of levitated
droplets. We have shown how this aspherical droplet analysis can be applied to real data from experiments, and have demonstrated the possible improvement in the surface tension measurements, in the case of acoustic levitation. Unfortunately, it is not possible to interpret the electromagnetic spectra more accurately, and electromagnetic parameters must be known before the surface tension measurements may be improved. It appears, from analytical considerations at least, that measurement of surface tension using acoustic means is more straightforward than by using electromagnetic techniques. This work also shows that in thermophysical property measurements, the inclusion of asphericity in the analysis is a very inexpensive alternative to maintaining the sphericity of the droplet by means of complex instrumentation.
CHAPTER 5
CONCLUSIONS AND RECOMMENDATIONS

The damped oscillations of a viscous droplet immersed in a viscous medium have been analyzed in detail. The characteristic equation has been solved by using a numerical method, which uses a continued fraction algorithm to solve for the Bessel and Hankel function ratios, and a minimization scheme to search for the solution. In Chapter 2, the effect of the droplet and host medium fluid properties and interfacial tension on the frequency of oscillations and damping rate has been shown, in terms of the nondimensional surface energy to viscous energy parameter, $\alpha^2$. For values of $\alpha$ less than a critical value $\alpha_{\text{crit}}$, it has been shown that the droplet returns aperiodically to its original shape. For $\alpha > \alpha_{\text{crit}}$, damped oscillations commence. The effect of increasing viscosity of the host medium is to delay the onset of this oscillatory behavior. The effect of the host medium may be neglected in liquid-gas systems, but is very important in liquid-liquid systems or in high temperature applications. The results of the theory have been compared to the experimental observations of Trinh et. al. [22], and it has been shown that the theory matches very well with experimental data, especially for oscillation frequency predictions. In some cases of damping rate comparisons, the theoretically calculated damping rate underpredicts the experimental observation. This is most likely due to surface contaminants, which tend to increase the damping rate of an oscillating droplet.
In Chapter 3, the application of levitation methods to the measurement of thermophysical properties has been discussed, with special relevance to surface tension and viscosity measurements. An inverse theory has been developed, which takes as inputs the damping rate, frequency of oscillations, density and radius of the droplet, and mode of oscillations, and predicts the surface tension and viscosity of the droplet. Results, presented in terms of nondimensional parameters, can be used directly to interpret experimental data from levitation experiments. The property measurement procedure has been described in detail, and will be of use in the simultaneous measurement of surface tension and viscosity from a single levitation experiment, at temperatures where no data is currently available.

This procedure has yet to be applied in actual experiments, and such application is recommended. One way to apply this theory is to set a levitated droplet into oscillations, and drop the entire experiment in a long drop tube. Qualification experiments on droplets of known properties are recommended before application to thermophysical property measurement. It must be mentioned here that this inverse theory is for a viscous droplet immersed in an inviscid medium. This limits the application of the theory to liquid-gas systems. One area of future work is the development of the inverse theory for the general case of a viscous droplet in a viscous medium, with arbitrary, finite, fluid properties.

In actual levitation experiments, a droplet is subject to external forces and to the gravitational pull on earth, and must necessarily be distorted. Thus,
oscillations of the droplet are now about this aspherical shape, and the analysis must include the effect of the distortion. The static shape deformation influences the resonance characteristics of the droplet, and the normal \((2l + 1)\)-fold degeneracy for mode \(l\) oscillations no longer holds good. This fact has been analytically shown in Chapter 4, and the non-degenerate modes of oscillation have been calculated for an arbitrary static shape deformation. The static shape deformation has also been calculated for external electromagnetic, acoustic, and combined acoustic-electromagnetic forces, and the effect of this deformation on the frequency spectrum has been shown theoretically. For the acoustic case, it has been shown that the frequency spectrum exhibits \(2l - 1\) peaks for a droplet oscillating in mode \(l\). Irrespective of the droplet and host properties, and the acoustic force present, it has been shown that \(\omega_{2,0} \geq \omega_{2,\pm 1} \geq \omega_{2,\pm 2}\). Thus, in acoustic levitation, without recoursing to the measurement of the acoustic parameters, it is possible to interpret the frequency spectra. The application of this theory to real experiments is demonstrated using the experimental results of Trinh et. al. [8]. By accounting for the different peaks that are normally discarded in the analysis, it has been shown that the surface tension measurements can be improved in accuracy. However, such an assignment of peaks is not so straightforward in the case of electromagnetically levitated droplets, and further information on the electromagnetic parameters is necessary, before the peaks can be assigned, and the split spectra interpreted.
The above aspherical theory was developed for an inviscid droplet. Thus, in terms of thermophysical property measurement, it can be applied only to surface tension measurements. Future work, on the oscillations of aspherical viscous droplet will bring out the essential features of the dynamics of levitated droplets, such as the effect of viscosity on the dynamics, the effect of the aspherical shape on the damping rate, and the effect of the host medium properties. Apart from its theoretical value, such a study will find application in the measurement of viscosity along with surface tension from a levitation experiment, and in any situation where a droplet shares an interface with another viscous fluid. It will also be of interest to study the effect of thermal gradients, and high temperatures on the dynamics and stability of the droplet. These are some of the recommended areas of further research.
APPENDIX A

ALGORITHM TO COMPUTE BESSEL FUNCTION RATIOS

Very often, for example in the Mie scattering calculations in optics [44], or in radiation studies, or in this case, the damped oscillations of a viscous droplet, one comes across ratios of spherical Bessel functions, in the form

\[ \frac{J_{\frac{l-1}{2}}(z)}{J_{\frac{l+1}{2}}(z)}. \] \hspace{1cm} (A.1)

When \( z \) is real, tables exist to give the Bessel functions (for example, see [21]), and the ratios can be calculated using Eq. (A.1). However, for complex \( z \), no such tabulated values are available, and few methods exist to find these ratios. We describe here the method we used, which is based on an algorithm described by Lentz [20]. Another commonly used algorithm is the recursive algorithm described by Ross [45], but the Lentz algorithm is much simpler and faster, and has fewer demands on computer storage.

The method uses a continued fraction approach. In order to simplify the notation, a continued fraction of the form

\[ f = a_1 + \frac{1}{a_2 + \frac{1}{a_3 + \frac{1}{a_4 + \cdots}}}, \] \hspace{1cm} (A.2)

is rewritten as
\[ f = a_1 + \frac{1}{a_2 + \frac{1}{a_3 + \frac{1}{a_4 + \ldots}}} \]  
(A.3)

with the positive sign in the denominator indicating the continuation of the fraction.

The Bessel ratios can be expressed in continued fractions [46], as

\[
\frac{J_{x-1}(z)}{J_x(z)} = 2x \ z^{-1} + \frac{1}{-2(x + 1) \ z^{-1} + \frac{1}{2(x + 2) \ z^{-1} + \frac{1}{(-1)^{n+1} \ 2(x + n - 1) \ z^{-1} + \ldots}}} 
\]  
(A.4)

Once the order and the argument of the function are known, its evaluation reduces to an evaluation of Eq. (A.4). In general, the evaluation has to begin at an unknown \( a_n \) and is worked backwards recursively to \( a_1 \). Needless to say, this is a cumbersome and time-consuming process. In order to circumvent this, we use the following procedure.

Let

\[ f_n(z) = [a_1, a_2, ..., a_n] \]  
(A.5)

be a continued fraction to \( n \). Then,

\[ [a_1] = a_1 \]  
(A.6a)

\[ [a_1, a_2] = a_1 + \frac{1}{a_2} \]  
(A.6b)

\[ [a_1, a_2, a_3] = a_1 + \frac{1}{a_2 + \frac{1}{a_3}} \]  
(A.6c)

and so on. It can be shown that any \( f_n(z) \) can be represented as a product of smaller sets of continued fractions, as
\[
f_n(z) = \frac{[a_1] [a_2, a_1] [a_{n-1}, ..., a_1] [a_n, ..., a_1]}{[a_2] [a_3, a_2] [a_{n-1}, ..., a_2] [a_n, ..., a_2]} \quad (A.7)
\]

Using Eq. (A.7), the evaluation can begin at \(a_1\), and end when the desired accuracy is reached. In other words, the evaluation simply ends when the most recently calculated terms in the numerator and denominator are identical within a specified tolerance, which is the acceptable error in the evaluation of the Bessel function. We consider an illustrative example for the calculation of the Bessel functions.

**Illustrative example:**

In Eq. (A.4), let \(z = 1\), and \(x = 9.5\). Then, in the \(rhs\) of Eq. (A.4), the first term, \(2x \ z^{-1}\), corresponds to \(a_1\) of the continued fraction, the second term, \(1/(2x+1)z^{-1}\), corresponds to \(a_2\) and so on. We recognize that

\[
[a_1] = 19 \quad (A.8a)
\]

\[
[a_2] = -21 \quad (A.8b)
\]

\[
[a_2, a_1] = a_2 + \frac{1}{a_1} = -20.94736842 \quad (A.8c)
\]

\[
[a_3, a_2, a_1] = a_3 + \frac{1}{[a_2, a_1]} \quad (A.8d)
\]

\[
[a_4, a_3, a_2, a_1] = a_4 + \frac{1}{[a_3, a_2, a_1]} \quad (A.8e)
\]

and so on. We notice, from Eqs. (A.8), that the successive terms can be calculated from the most recently calculated terms using a recursive formula. The calculation stops when the desired accuracy is reached. In this example, for a six-digit accuracy, only four terms need be calculated in the numerator, and the fourth
term in the numerator is found to be $-24.95643131$. The corresponding term in
the denominator is the third term, and is found to be $-24.95643154$. These terms
are identical within six decimal places. The solution for the ratio is,

\[ \frac{J_{8.5}(1)}{J_{9.5}(1)} = 18.95228199. \]  \hspace{1cm} (A.9)

This value is identical to the value reported in the tables [21]. If better accuracy
is desired, subsequent terms can be calculated in the numerator and denominator.

It must be noted that, in the representation of Eq. (A.7), a particular term
may be zero within the accuracy of the digital computer. In such cases, the term
must be skipped, along with the corresponding term in the denominator. This is
also incorporated in the algorithm.

The above algorithm was checked for complex argument with the values that
result from applying trigonometric expansions given in [21,46], and was found to
be accurate within the desired limits. The FORTRAN code for the Spherical
Bessel function computation follows.
C
C Program to evaluate the ratios of spherical bessel functions
C With accuracy improvement and zero check incorporated.
C Modified from the paper by Lentz (Appl. Opt, 1976)
C
C AUG 1, 1989, SURI
C
C MAIN PROGRAM

implicit double precision (a-h, o-z)
complex*16 z,tnum,tden,a,an,xnum,xden,dprev,aprev,q

write(*,*)'enter order and argument'
read(*,*)v,z

aprev = a(1,v,z)
dprev = a(2,v,z)
tnum = aprev
tden = dprev
xden = dprev
n = 2
an = a(n,v,z)

C Begin computation of terms in the continued fraction
C representation of the Bessel function ratios

23     xnum = an + 1/aprev
       call checkac(xnum,n,v,z)

C checkac is the zero check subroutine.

tnum = tnum * xnum
q = tden / tnum
aprev = xnum
dprev = xden
temp1 = real(xden) - real(xnum)
temp2 = dimag(xden) - dimag(xnum)
errl = temp1**2 + temp2**2
if (errl .lt. 1.0d-14) then
   write(*,*)'z = ',z,' ord = ',v,
   write (*,*) 'bes-ratio = ',q,' error = ', errl
else
   n = n + 1
   an = a(n,v,z)
xden = an + 1/dprev
   call checkac(xden,n,v,z)
   tden = tden*xden
   go to 23
endif
stop
end

***************************************************************************
C Zero check subroutine

    subroutine checkac(beta, n, v, z)
    implicit double precision (a-h, o-z)
    complex*16 beta, z, a, zeta, znext

    valbeta = (real(beta))**2 + (dimag(beta))**2

    C If not zero, then return.
    C Else skip, and compute the next terms and return.

    if (valbeta .gt. 1.0d-16) then
      return
    else
      m1 = n+1
      zeta = a(m1,v,z)*beta + 1
      m2 = n+2
      znext = a(m2,v,z) + beta / zeta
      beta = znext
      n = n+2
    end if

    return
    end

******************************************************************************

C Function that uses the recursive relationship between
C successive terms to calculate them.

    function a(n, v, z)
    implicit double precision (a-h, o-z)
    complex*16 a, z

    t1 = 2*(v + n - 1)
    n1 = n+1
    a = (-1)**n1*t1/z
    return
    end

******************************************************************************
APPENDIX B

EVALUATION OF INTEGRALS OF PRODUCTS OF THREE SPHERICAL HARMONICS

For the integral

\[ \iint Y_{l''m''}^* Y_{l'm'} Y_{lm} \, ds \]  \hspace{1cm} (B.1)

to exist,

(i) \( m'' = m + m' \)

(ii) \( |l - l'| \leq l'' \leq |l + l'| \)

(iii) \( (l + l' + l'') \) is even

Here, since \( m' = 0, l = l'' \), and \( m = m'' \), the number of these integrals to be evaluated depends on the values of \( l \); i.e., the integrals exist only for even \( l' \), for \( l' \leq 2l \). For \( l = 2 \), for instance, only two integrals exist:

\[ I_{2m}^{(2)} = \iint Y_{2m} Y_{2m}^* Y_{20} \, ds \]  \hspace{1cm} (B.2)

and

\[ I_{4m}^{(2)} = \iint Y_{2m} Y_{2m}^* Y_{40} \, ds \]  \hspace{1cm} (B.3)

Evaluation of the Integrals - The 3-j symbols

The Gaunt formula (in terms of the Wigner 3-j symbols) is [40]

\[ \iint Y_{l''m''}^* Y_{l'm'} Y_{lm} \, ds = (-1)^m'' \left( \frac{(2l''+1)(2l'+1)(2l+1)}{4\pi} \right)^{1/2} \times \]

\[ \times \begin{bmatrix} l'' & l' & l \\ -m'' & m' & m \end{bmatrix} \begin{bmatrix} l'' & l' & l \\ 0 & 0 & 0 \end{bmatrix} \]  \hspace{1cm} (B.4)

where the 3-j symbols are given by [38]
\[
\begin{bmatrix}
  j_1 & j_2 & j_3 \\
  m_1 & m_2 & m_3 \\
\end{bmatrix} = (-1)^{j_1 - j_2 - m_3} \times
\]
\[
\times \left[ (j_1 + j_2 - j_3)! (j_1 - j_2 + j_3)! (-j_1 + j_2 + j_3)! (j_1 + m_1)! (j_1 - m_1)!
\right.
\]
\[
(j_2 + m_2)! (j_2 - m_2)! (j_3 + m_3)! (j_3 - m_3)!
\left. \right]^{-\frac{1}{2}} \left[ (J + 1)! \right]^{-\frac{1}{2}} \times \sum_k (-1)^k \left[ k! 
\right.
\]
\[
(j_1 + j_2 - j_3 - k)! (j_1 - m_1 - k)! (j_2 + m_2 - k)! (j_3 - j_2 + m_1 + k)! (j_3 - j_1 - m_2 + k)!
\left. \right]^{-1}
\]

where, \( J = j_1 + j_2 + j_3 \), and the summation index \( k \) takes all possible integral values for which the arguments of the factorials are non-negative. The number of terms in the summation, in fact, is the smallest of the nine numbers:

\( j_1 \pm m_1, j_2 \pm m_2, j_3 \pm m_3, j_1 + j_2 - j_3, j_2 + j_3 - j_1, j_3 + j_1 - j_2 \).

Eq. (B.5) is known as the symmetric form of Racah.

The 3-j symbols were first introduced by Wigner [38] in the matrix representations of angular momenta, and are also referred to as the Wigner coefficients. They are related to the Clebsch-Gordan coefficients by

\[
\begin{bmatrix}
  j_1 & j_2 & j_3 \\
  m_1 & m_2 & -m_3 \\
\end{bmatrix} = (-1)^{j_1 - j_2 + m_3} (2j_3 + 1)^{-\frac{1}{2}} C_{j_1 j_2 j_3 | m_1 m_2 m_3}
\]

In order for a 3-j symbol to exist, it must meet the following criteria:

(i) \( m_1 + m_2 + m_3 = 0 \)

(ii) \( (j_1 + j_2 - j_3) \geq 0 \)

(iii) \( (j_1 - j_2 + j_3) \geq 0 \)

(iv) \( (-j_1 + j_2 + j_3) \geq 0 \)

(v) \( j_1 + j_2 + j_3 \) integral.

Note that in our case, all these criteria are satisfied.
The 3-j symbols exhibit 72 known symmetries, of which, the most commonly invoked are-(i) Odd permutations of column multiply the symbol by \((-1)^{i+j+k}\), and (ii) Even permutations leave the value of the symbol unchanged.

In this work, the 3-j symbols are calculated using a simple program. It implements the Racah's symmetric form, and exploits the symmetries of the 3-j symbols where necessary. Once the 3-j symbols are evaluated, the integrals \(I_{um}^{(l)}\) can be evaluated, depending on the number of integrals that are non-zero. For \(l = 4\), for instance, the integrals that exist are \(I_{2m}^{(4)}\), \(I_{4m}^{(4)}\), \(I_{6m}^{(4)}\), and \(I_{8m}^{(4)}\). All integrals required in this work are calculated using Eqs. (B.4) and (B.5), and are summarized in Table I.
APPENDIX C

SPLIT FREQUENCIES FOR MODE 3 AND 4 OSCILLATIONS

The frequency values for \( l = 3 \) and 4 for a static shape deformation of the

form \( \sum_{2n} \chi_{2n,0} Y_{2n,0} \), \( n = 1 \cdots l \), are listed below.

\( l = 3 \)

\[ \omega_{3,0} = \omega^* \left( 1 - 0.26913414 \frac{\chi_2}{R} - 0.46160966 \frac{\chi_4}{R} - 1.23285726 \frac{\chi_6}{R} \right) \]

\[ \omega_{3,\pm 1} = \omega^* \left( 1 - 0.20185060 \frac{\chi_2}{R} - 0.07693494 \frac{\chi_4}{R} + 0.92464294 \frac{\chi_6}{R} \right) \]

\[ \omega_{3,\pm 2} = \omega^* \left( 1 + 0.53854460 \frac{\chi_4}{R} - 0.36985718 \frac{\chi_6}{R} \right) \]

\[ \omega_{3,\pm 3} = \omega^* \left( 1 + 0.336417668 \frac{\chi_2}{R} - 0.23080483 \frac{\chi_4}{R} + 0.06164283 \frac{\chi_6}{R} \right) \]

\( l = 4 \)

\[ \omega_{4,0} = \omega^* \left( 1 - 0.30037292 \frac{\chi_2}{R} - 0.35762067 \frac{\chi_4}{R} - 0.54530225 \frac{\chi_6}{R} - 1.2894168 \frac{\chi_8}{R} \right) \]

\[ \omega_{4,\pm 1} = \omega^* \left( 1 - 0.25531698 \frac{\chi_2}{R} - 0.17881034 \frac{\chi_4}{R} + 0.02726511 \frac{\chi_6}{R} + 1.03153348 \frac{\chi_8}{R} \right) \]

\[ \omega_{4,\pm 2} = \omega^* \left( 1 - 0.12014917 \frac{\chi_2}{R} + 0.21854597 \frac{\chi_4}{R} + 0.59983247 \frac{\chi_6}{R} - 0.51576674 \frac{\chi_8}{R} \right) \]

\[ \omega_{4,\pm 3} = \omega^* \left( 1 + 0.10513052 \frac{\chi_2}{R} + 0.41722412 \frac{\chi_4}{R} - 0.46350691 \frac{\chi_6}{R} + 0.14736192 \frac{\chi_8}{R} \right) \]

\[ \omega_{4,\pm 4} = \omega^* \left( 1 + 0.42052209 \frac{\chi_2}{R} - 0.27814941 \frac{\chi_4}{R} + 0.10906045 \frac{\chi_6}{R} - 0.018420242 \frac{\chi_8}{R} \right) \]
These frequencies are calculated using Eq. (4.20), and obtain in the absence of an external force.
APPENDIX D

COEFFICIENTS FOR ELECTROMAGNETIC FORCE EXPANSION

The external magnetic force at the surface, \( P_{\text{mag}} \), is given by

\[
P_{\text{mag}} = \sum_{uv} P_{uv} Y_{uv}
\]

where the \( P_{uv} \) are given by Eq. (4.51),

\[
P_{uv} = \frac{f_{jk} f_{lm}}{4\mu R^2} \left[ (l+1) + j(j+1) - u(u+1) \right] \int \int Y_{jk} Y_{lm} Y_{uv}^* \, ds.
\]

(D.2)

Since \( l = 1, 2 \), \( j = 1, 2 \) and \( k, m = 0 \),

\[
P_{uv} = \frac{1}{4\mu R^2} \left\{ f_{10} \left[ 4-u(u+1) \right] \int \int Y_{10} Y_{10} Y_{uv}^* \, ds +
\right.
\]

\[
+ f_{20} \left[ 12-u(u+1) \right] \int \int Y_{20} Y_{20} Y_{uv}^* \, ds +
\]

\[
+ f_{10} f_{20} \left[ 8-u(u+1) \right] \int \int Y_{10} Y_{10} Y_{uv}^* \, ds \right\}
\]

(D.3)

(D.4)

From Appendix B, it is clear that the integrals are non-zero only for \( v = 0 \), and limited values of \( u \). This means that \( P_{00}, P_{10}, P_{20}, P_{30}, P_{40}, \) and \( P_{50} \) are the only coefficients that exist. On substituting the values of the appropriate integrals, and simplifying, these coefficients are:

\[
P_{00} = \frac{1}{4\mu R^2} \sqrt{\frac{1}{4\pi}} \left[ f_{10}^2 + f_{20}^2 \right]
\]

\[
P_{10} = \frac{1}{4\mu R^2} \sqrt{\frac{1}{4\pi}} \left[ 1.5138795 f_{10} f_{20} \right]
\]

\[
P_{20} = \frac{1}{4\mu R^2} \sqrt{\frac{1}{4\pi}} \left[ -0.5046265 f_{10}^2 + 1.0813425 f_{20}^2 \right]
\]
\[ P_{30} = \frac{1}{4\mu R^2} \sqrt{\frac{1}{4\pi}} \left[ -0.99106678 f_{10} f_{20} \right] \]

\[ P_{40} = \frac{1}{4\mu R^2} \sqrt{\frac{1}{4\pi}} \left[ -1.93436432 f_{20}^2 \right], \]

where

\[ f_{10} = \frac{3}{2} a_{10} R, \]

and

\[ f_{20} = \frac{5}{3} a_{20} R^2, \]

and \( a_{10} \) and \( a_{20} \) are given by Eqs. (4.47).
REFERENCES


