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Oscillation Frequencies of Droplets Held Pendant on a Nozzle

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Abstract

Small-amplitude oscillations of liquid droplets held pendant on a nozzle and surrounded by either air or another liquid were investigated experimentally. The oscillations were induced by mechanical means, and the natural frequencies of oscillations were visually determined. Results are compared to existing related theories of Miller and Scriven and of Strani and Sabetta. As may be expected, the presence of the solid support causes measured oscillation frequencies for the lowest oscillation mode to deviate greatly from the Miller and Scriven low viscosity approximation ($n = 2$) for free spherical drops. Experiments are in qualitative agreement with the first ($n = 1$) mode predicted by Strani and Sabetta, with the best correspondence occurring under circumstances where the ratio of nozzle to drop radii is small. The difference between the experimental results and the theory of Strani and Sabetta can be attributed to the restrictive boundary conditions of their analytic model. Thus, better theoretical characterization of pendant droplet oscillations will require numerical computations.

INTRODUCTION

The behavior of droplets and bubbles is of importance in a multitude of applications within the chemical industry as well as such diverse fields as meteorology, printing and paint spraying, and mathematical science. Within the chemical industry the behavior of bubbles and droplets is a major factor in the operation of most multiphase systems such as reactors and separations equipment.

The efficiency of these devices is determined to a large extent by the rate of heat and/or mass transfer between drops or bubbles and the continuous phase. To improve performance, one must provide the means to increase transport rates. Two obvious physical means are to increase the interfacial surface area relative to fluid volume and to increase fluid velocities to enhance convection. This is commonly achieved through the introduction of finely dispersed fluids through nozzles and by bulk fluid agitation. While such an approach achieves the desired results, it represents an inefficient usage of energy.

The next generation of chemical processing equipment will be designed with energy efficiency and waste minimization as primary goals. As such, increased performance, leading to better materials utilization, must be achieved without wasteful use of energy. Major steps toward these goals have been achieved through the application of external fields in chemical processing equipment (*1*, *2*). Two examples recently developed at Oak Ridge National Laboratory are the emulsion-phase contactor (EPC), an advanced solvent extraction device, and the electric dispersion reactor (EDR), an advanced multiphase liquid reactor. Both of these devices take advantage of the fact that applied electrical fields may be used to impart a force at the interface of two fluids with differing electrical properties. This force may be used to deform and disperse droplets or bubbles at a much lower energy consumption rate than by mechanical dispersion. Droplet sizes in these devices are on the order of a micron in diameter, providing large specific interfacial surface area for rapid chemical transport. The EPC has demonstrated performance over one hundred times better than current industrial machines in small-scale operations while requiring only a fraction of the energy to run the process (*1*). The EDR has been operated to produce dense, micron-sized, stoichiometrically homogeneous ceramic precursor materials (*2*).

In order to optimize the performance of devices such as the EPC and the EDR, and to pave the way for other advances, a fundamental understanding of the behavior of droplets and bubbles at the microscopic scale is needed. For instance, in development of the EPC, Scott (*3*) found that the electric field strength required for dispersion of free droplets of water in 2-ethyl-1-hexanol is dependent upon the frequency of applied voltage pulses. The required field strength varied greatly in the vicinity of the natural frequency of oscillation of inviscid fluids determined by Lamb (*4*). A maximum in field strength was required for a pulse frequency very close to the Lamb frequency, while a minimum was achieved for a pulse frequency somewhat lower than the Lamb frequency, near the natural frequency of oscillation for viscous fluids (see below). Reliable information on oscillation frequency would be indispensable both for control of droplet size in a device and for tuning of frequency of applied voltage for minimization of energy consumption.

Further work by Scott (*5*) aimed at determining the effect of frequency upon breakup of an aqueous stream exiting from a nozzle indicated that pulsed electrical fields could achieve dispersion at a lower root-meansquare field strength than dc fields. In addition, the field strength required for effective dispersion by the pulsed fields varied as a function of the viscosities of the fluids. The two pulse frequencies tested displayed opposite trends, with the required field strength increased with increasing viscosity for the 2000 Hz case and decreased with increasing viscosity for the 200 Hz case. The 2000 Hz pulse frequency required less power than the 200 Hz one for the lower viscosities, while the 200 Hz pulse frequency required less power than the 2000 Hz one for the higher viscosities. Scott postulated that application of pulsed electrical fields at the resonance frequency of the liquid drops exiting the nozzle would minimize power requirements.

Thus, a detailed understanding of the oscillations of pendant droplets could lead to improvements in an entire class of chemical processing devices. In this paper, a first step in understanding such processes, we report measurements of oscillation frequencies of droplets held pendant on a nozzle. Relevant theories of drop oscillations are reviewed in the next section, followed by a description of the experimental techniques. The results reported thereafter show that nonspherical shape and the presence of a solid support profoundly affect the oscillations of pendant drops.

SUMMARY OF RELEVANT THEORIES

A droplet will deform in response to an applied stress. If the perturbation is of sufficient magnitude, the droplet will break up, whereas for lesser amplitude perturbations the droplet will deform from its initial shape. From this distorted condition, the droplet will dissipate energy by, depending upon the droplet size and physical properties of the fluids, either aperiodic damped motion or by damped oscillations, leading finally to a return to the initial state. Damped oscillations of a single mode will occur with a characteristic frequency and damping factor.

The oscillation of droplets and bubbles has been studied for over a hundred years (*6*). The most widely known treatment is that of Lamb (*4*), who considered infinitesimally small-amplitude, irrotational oscillations of an inviscid stationary spherical droplet in an inviscid medium. The frequency of oscillation of a droplet of radius R was found to be

$$\omega^* = \left(\frac{\gamma n(n+1)(n-1)(n+2)}{R^3\Gamma}\right)^{1/2} \tag{1}$$

where ω^* is the "Lamb" frequency of oscillation (in radians/second, meaning the period of one oscillation is $2\pi/\omega^*$), γ is the interfacial tension, n is the integer value of the mode of oscillation ($n \geq 2$), and $\Gamma \equiv \rho_o n + \rho_i(n+1)$, where ρ_i is the density of the droplet fluid and ρ_o is that of the surrounding fluid. This inviscid treatment cannot be used to predict the oscillation damping rate.

Miller and Scriven (7) completed a comprehensive analytical treatment of the problem of infinitesimal-amplitude oscillations of stationary spherical fluid droplets of arbitrary viscosity in another viscous fluid. Complete solution of this equation requires finding the determinant of a nonlinear 7×7 matrix. The frequency of oscillations, β, is a complex number, the real part of which, β_R, corresponds to a decay factor, and the imaginary part, β_I, is the angular frequency of oscillation. The authors did not solve the system of equations numerically; rather, they found analytical solutions for nine important limiting cases. The most practical of these solutions is the low-viscosity approximation (LVA), given in corrected form (see Refs. *8–10*) by

$$\beta_R = \frac{(2n+1)^2(\omega^*\mu_i\mu_o\rho_i\rho_o)^{1/2}}{2\sqrt{2}R\Gamma[(\mu_i\rho_i)^{1/2} + (\mu_o\rho_o)^{1/2}]} - \frac{(2n+1)^4\mu_i\mu_o\rho_i\rho_o}{4R^2\Gamma^2[(\mu_i\rho_i)^{1/2} + (\mu_o\rho_o)^{1/2}]^2}$$

$$+ \frac{(2n+1)[2(n^2-1)\mu_i^2\rho_i + 2n(n+2)\mu_o\rho_o + \mu_o^2\mu_i(\rho_i(n+2) - \rho_o(n-1))]}{2R^2\Gamma[(\mu_i\rho_i)^{1/2} + (\mu_o\rho_o)^{1/2}]}$$

$$\beta_I = \omega = \omega^* - \frac{(2n+1)^2(\omega^*\mu_i\mu_o\rho_i\rho_o)^{1/2}}{2\sqrt{2}R\Gamma[(\mu_i\rho_i)^{1/2} + (\mu_o\rho_o)^{1/2}]}, \quad n = 2, 3, 4, \ldots \tag{2}$$

where μ_i is the viscosity of the drop and μ_o is that of the continuous medium. Basaran et al. (*10*) solved the complete set of equations for β numerically, thereby providing solutions that are valid for arbitrary viscosities and densities.

Basaran (*11*) presented a fully numerical solution for nonlinear oscillations of viscous free drops. This treatment, which assumed that perturbations are from spherical shape, with no gravity or other external fields, is not limited as to the viscosity of either fluid, and thus may be seen to fully describe oscillations of isolated spherical drops. Numerical solutions of this type hold promise for solution of any nonspherical case such as pendant droplets; however, application to each case will require adjustments for velocity profiles, applied fields, and/or boundary conditions. In addition, experiments with well-defined operating conditions and boundary conditions will be necessary to verify such calculation methods.

Strani and Sabetta (*12, 13*; hereafter referred to as SS1 and SS2) analyzed the small-amplitude oscillations of spherical drops immersed in a surrounding fluid and in contact with a solid support. In their analytical treatment, the undeformed droplet was limited to a spherical shape, with the solid support shaped as a portion of a spherical cavity of the same radius as the droplet. Both treatments considered axisymmetric oscillations only and neglected gravity.

SS1 considered the case of inviscid fluids. The frequency of oscillation for this case may be calculated from the eigenvalues of a matrix with entries consisting of Green's functions. The presence of the solid support was found to change the oscillation behavior from that of isolated drops in two ways: 1) an extra oscillation mode ($n = 1$), corresponding to the aperiodic translation of an isolated drop, arises, and 2) the frequency of oscillation for the same size droplet is found to increase with increasing area of contact with the support. Oscillation frequencies for this inviscid constrained drop (ICD) treatment were found to correctly approach the Lamb frequency as the support size approaches zero.

SS2 presented a viscous treatment that is similar to that in SS1, but which requires more complicated numerical calculations. The addition of viscous effects was found to decrease the predicted oscillation frequency slightly, and damping constants were predicted. For a system with $\rho_o = 1.002$ g/cm^3, $\rho_i = 1.002$ g/cm^3, $\mu_o = 1.00$ cP, and $\mu_i = 4.41$ cP, values for oscillation frequency were about 6% lower than predicted by ICD of SS1. Experimental results of frequency measurements for such fluids using flat supports were shown to be consistently lower than predictions, with errors of up to 10% for small support size, but reaching 30% for a support of the same radius as the drop.

At present, there exists no theory for oscillations of droplets held pendant on a nozzle. The oscillations of isolated, spherical drops are fully described by Basaran (*11*); the frequency and damping of small amplitude oscillations may be estimated in closed-form by LVA of Miller and Scriven (*7*). The work of Strani and Sabetta has shown that the presence of a solid support will greatly affect the oscillation of spherical droplets; however, the restrictive boundary conditions of their models do not strictly apply to a drop held pendant on a nozzle. The aim of this paper is to measure the oscillation frequencies of pendant drops and compare the results with models which may be most readily used by practitioners, namely the corrected LVA of Miller and Scriven and the ICD model of Strani and Sabetta.

EXPERIMENTAL

A schematic drawing of the experimental setup used is shown in Fig. 1. A stainless steel nozzle was held by a rigid clamp with its tip immersed in a cuvette filled with the surrounding fluid. Flexible silicone tubing connected the nozzle to a syringe filled with water. A portion of the tubing was held by a vibrational transducer (Alpha-M Corporation, Model AV-6). Activation of the transducer by a 25-W solid-state amplifier with a

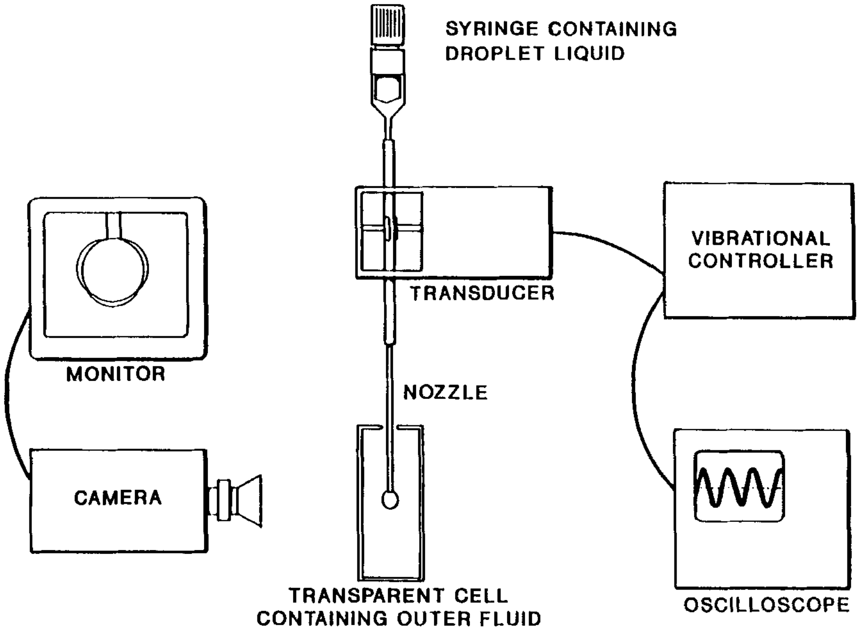

FIG. 1. Experimental setup for studies of pendant drop oscillations.

frequency range of 2 to 20,000 Hz (Alpha-M Corporation, Model OC-25) control unit would cause the tubing to be periodically compressed at a controlled frequency and amplitude. The frequency of oscillation and the amplitude of the input voltage to the transducer were monitored by an oscilloscope (Tektronix Inc., Model 504), while the size and shape of the droplet were observed by using a high-speed camera (Tri-Tronics Inc., Model PCSM-5600), monitor (Panasonic, Model TR124-MA), and a video position analyzer (FOR.A, Model VPA-100).

The size of the droplet was set by manipulating the syringe. The vertical and maximum horizontal dimensions of each drop were measured using the video position analyzer. These measurements were converted to an equivalent radius of the sphere of the same volume through a correlation of profile measurements made on five droplets of various sizes for each fluid pair and nozzle. The transducer frequency was varied at moderate amplitude to find the approximate frequency of oscillation for the droplet, evidenced by motion of the interface. The amplitude was reduced and the frequency was adjusted until the point was reached at which motion of the interface was barely discernable (e.g., 5% radius variation). The frequency(ies) at which interface motion was maximized at the lowest discernable amplitude was recorded as the driven resonance oscillation frequency of the pendant droplet.

RESULTS

Oscillation frequencies of pendant droplets were determined using nozzle supports of two sizes: 1) 0.068 cm outside diameter (OD), 0.045 cm inside diameter (ID), and 2) 0.15 cm OD, 0.074 cm ID. The droplet fluid was water (ρ = 1.0 g/cm^3, μ = 1.0 cP), while surrounding fluids of air (ρ = 0.0012 g/cm^3, μ = 0.182 cP, γ = 71.97 dyn/cm) and cyclohexane (ρ = 0.779 g/cm^3, μ = 0.88 cP, γ = 50.2 dyn/cm) were used.

Figure 2 shows sketches of the first three modes of oscillation of a pendant drop. The first mode appears similar to the prolate-oblate stretching corresponding to the $n = 2$ mode of free drops; however, it is also worth noting that the approximate shape is predicted by the figure for $n = 1$ in SS1. Likewise, the other two modes shown have similarities to the corresponding shapes of both theories.

Results obtained by the experimental means described above for the frequency of oscillation of the lowest mode in the water/air system are shown in Fig. 3. The symbols represent the experimental values of frequency as a function of droplet equivalent radius, while the curves are plots of the lowest mode ($n = 2$) of the low-viscosity approximation (LVA) for isolated drops of Miller and Scriven (7) and the lowest ($n = 1$) mode of the inviscid constrained drop (ICD) theory of SS1 for both nozzle sizes. As may be expected, the presence of the solid support caused the experimental frequencies to deviate greatly from the LVA; the measured frequencies were much lower than those predicted by the LVA for isolated drops. The results for both nozzle sizes are in qualitative agreement with the associated ICD curve. Agreement between the measurements and the theory of SS1 improves as drop size increases. As predicted by Strani and Sabetta, the oscillation frequency for a given drop size increases as the nozzle size increases.

The pendant drop oscillation results for the water/cyclohexane system are compared to the ICD of SS1 in Fig. 4. In this figure the dimensionless frequency, defined as the frequency of oscillation in radians per second multiplied by $(\rho_i r^3/\gamma)^{1/2}$, where r is the support radius, is plotted as a function of the dimensionless drop radius, defined as the ratio of the radius of the drop to that of the support. Results obtained with both nozzles fall upon the same curve in this dimensionless representation. Agreement with the model is quite good for drops whose radii are large relative to the nozzle radius, but as with the water/air system, there is much greater deviation as the dimensionless radius decreases. Moreover, the deviation

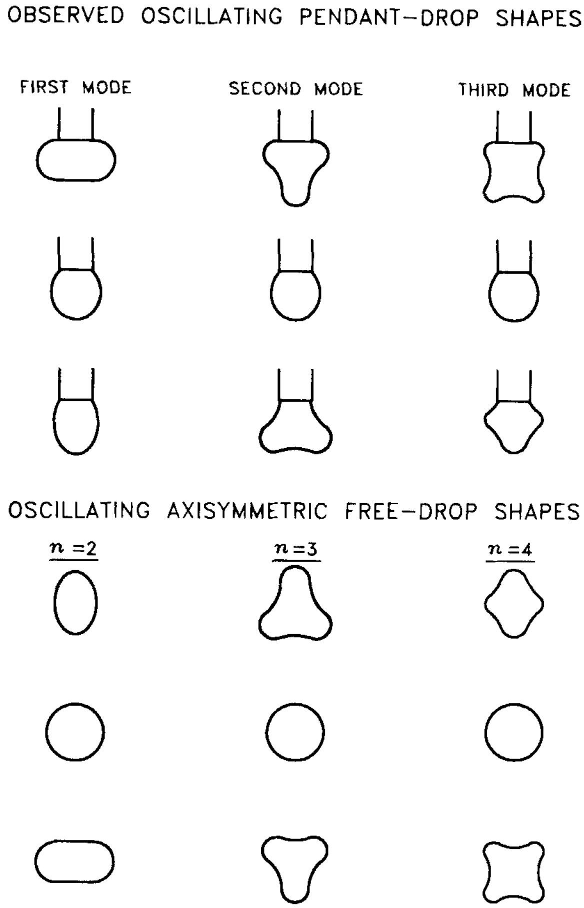

FIG. 2. Comparison of approximate observed shapes of the first three modes of oscillation of pendant drops with shapes of the first three modes of free drops that are neutrally buoyant in the surrounding fluid.

between theory and experiment is greater for cyclohexane than for air; it is probable that the addition of viscous effects described by SS2 would account for part of this difference.

DISCUSSION

The pendant drop experiments and the inviscid theory of SS1 differ in several fundamental ways, including: 1) the support is different (a spherical section matching the drop radius for the theory and a flat tip for the

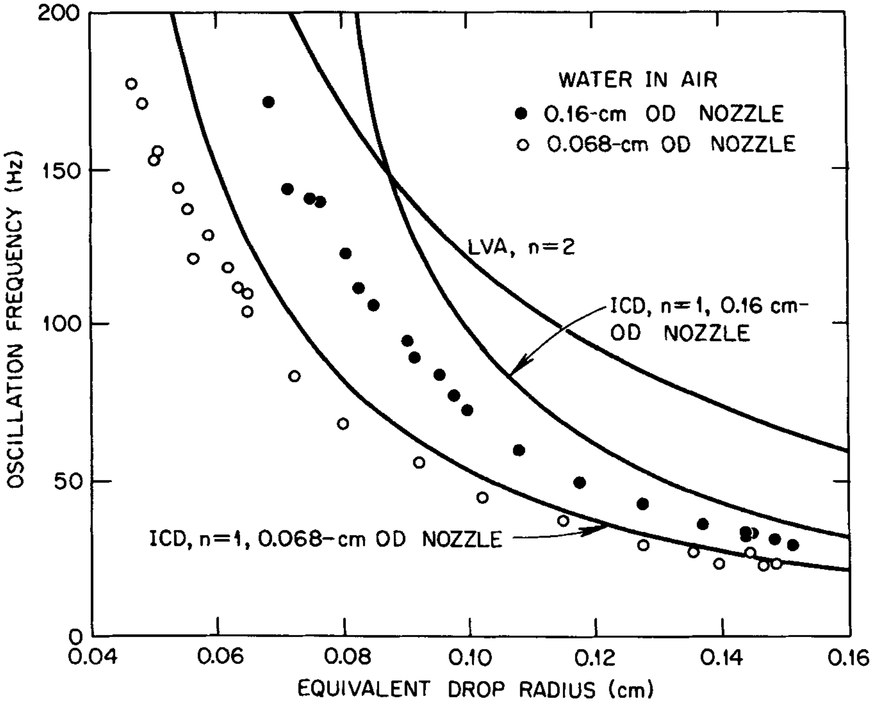

FIG. 3. Comparison of measured lowest-mode oscillation frequency of water drops held pendant on 0.16 cm outside diameter (OD) and 0.068 cm OD nozzles in air with low-viscosity approximation (LVA) for free drops of Miller and Scriven (*1*) and inviscid constrained drop (ICD) theory of Strani and Sabetta (*2*).

experiments), leading to different fluid mechanics where the drop wets the solid and a difficulty in definition and comparison of droplet and support dimensions; 2) the theory neglects gravity, which restricts undeformed drops to be spherical rather than distorted; 3) the theory is for inviscid fluids; and 4) the vertical component of fluid velocity within the nozzle was a small nonzero periodic function in the experiments rather than the imposed zero velocity normal to the spherical base in the model. Other important limitations of the experimental procedure may be noted, such as the fact that oscillations which are visually detected are slightly nonlinear and the mechanical means of excitation caused slight volume perturbation in the drops. Different methods of perturbation and detection, such as electrical excitation with detection similar to that of Trinh et al. (*14*), are planned by the authors for further investigation of the problem.

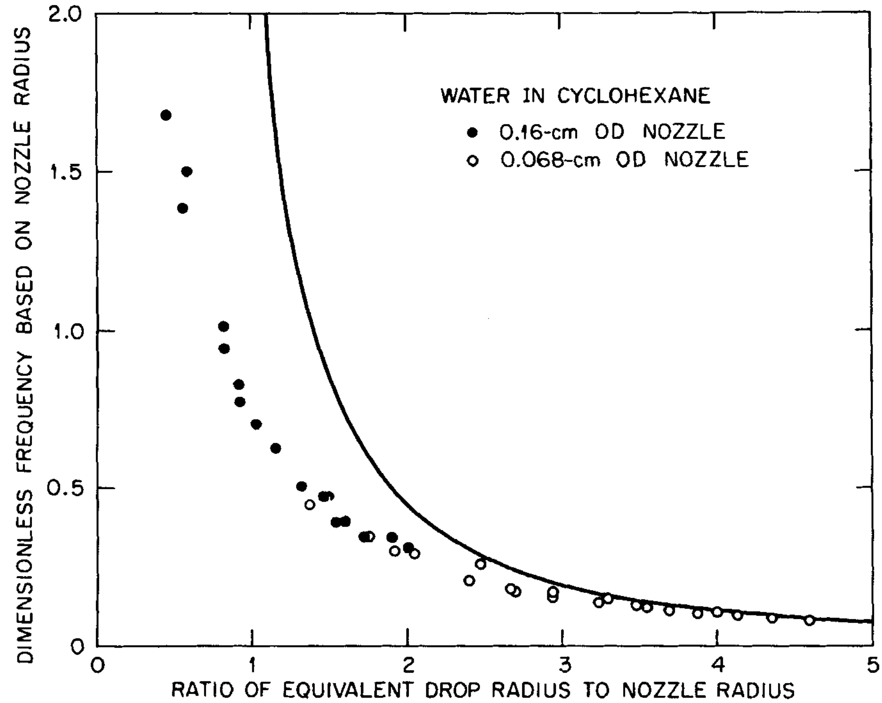

FIG. 4. Variation of the dimensionless frequency of oscillation of the lowest mode of pendant drops of water in cyclohexane with dimensionless radius: comparison of measurements and predictions of ICD.

Despite these important differences, inviscid treatment of SS1 does exhibit several key features of the experimental results. It is apparent that the first oscillation mode ($n = 1$) of the ICD is in better agreement with the results than the first mode ($n = 2$) of Lamb or LVA. The experimental results diverge from the theoretical curves when the radius of the support approaches that of the drop, as might be suspected from the differences in the geometry of the supports and drops in the theory and experiment. Agreement on the whole would become slightly better with the addition of viscous forces, which slightly decrease the frequencies. The qualitative agreement is important, in that Strani and Sabetta predicted the presence of the $n = 1$ mode with a support, the frequency of which falls to zero as the support is eliminated. Indeed, for free drops the $n = 1$ mode is not allowed because it would move the center of mass of the drop. This first oscillation mode is further supported by the agreement between theory and experimental observations of shapes of drops undergoing oscillations in the first three oscillation modes.

CONCLUSIONS

A mechanical oscillation technique has been developed and used to measure the resonance frequency of forced oscillations of pendant drops. The results obtained by this technique are in qualitative agreement with the inviscid constrained drop theory of Strani and Sabetta (*12*). The presence of a solid support, as with a pendant drop, greatly affects oscillation frequency by adding a lower mode of oscillation which is not allowed for nontranslating free drops. The relations of Miller and Scriven (*7*) cannot be applied accurately to the case of a pendant drop because of the different modes of oscillation and vastly different boundary conditions that apply to free drops as compared to supported ones.

Although the inviscid theory of Strani and Sabetta (*12*) produced qualitative agreement with the experimental results and good agreement when nozzle radius is small relative to the droplet radius, differences between the two are still great for smaller droplets. Thus, this theory is only useful as a first approximation to the natural frequency of oscillation of pendant drops. It is likely that better agreement will be achieved through the use of the viscous theory of Strani and Sabetta (*13*); however, the boundary conditions imposed by the theoretical model are too restrictive to apply to drops that are pendant from a nozzle. For full characterization of the dynamics of pendant drops, it will be necessary to develop numerical models which can capture all aspects of the problem, including undeformed shapes that are nonspherical and the presence of external forces.

NOMENCLATURE*

n	mode number of oscillation (dimensionless)
r	scaled radial distance (dimensionless)
r_b	radius of solid support (L)
R	radius of sphere having volume of drop (L)
β	complex frequency of oscillation (T^{-1})
β_I	imaginary part of β, corresponding to oscillation frequency (T^{-1})
β_R	real part of β, corresponding to damping rate of oscillations (T^{-1})
γ	interfacial tension ($M \cdot T^{-2}$)
Γ	$\rho_o n + \rho_i(n + 1)$ ($M \cdot L^{-3}$)
μ_i	dynamic viscosity of drop fluid ($M \cdot L^{-1} \cdot T^{-1}$)
μ_o	dynamic viscosity of surrounding fluid ($M \cdot L^{-1} \cdot T^{-1}$)

*L = length, M = mass, T = time.

ρ_i density of drop fluid ($M \cdot L^{-3}$)
ρ_o density of surrounding fluid ($M \cdot L^{-3}$)
ω oscillation frequency (T^{-1})
ω^* Lamb frequency (T^{-1})

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