Nonlinear Oscillations of Viscous Droplets

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Abstract

This paper deals with the problem of non-linear droplet oscillations, whereas effects of liquid viscosity are of our special interest. In the experimental part finite amplitude, axial-symmetric oscillations of small liquid droplets in a gaseous environment are studied. Experimental results are compared with available theoretical models. It was found that for a wide range of viscous damping the numerical model, based on applying a least squares approximation to the boundary conditions and the vorticity equation [1], provides results which are in a good agreement with the experimental findings. At low and medium viscosities also a simple 'mechanical' model [2] of droplet dynamics offers adequate description of the numerical and experimental data. The linear irrotational model of Lamb [3] suffers serious discrepancies between predicted and observed oscillation patterns.

1 INTRODUCTION

The problem of oscillating droplets has long been the focus of many theoretical studies. Every such analysis has to start from the statements of mass and momentum conservation in form of the equation of continuity $\nabla \cdot \vec{v} = 0$ and the Navier-Stokes equation:

$$\mathrm{A}\frac{\partial \vec{v}}{\partial t} + \mathrm{A}^2(\vec{v}\cdot\nabla\vec{v}) = -\nabla p + \mathrm{A}\,\mathrm{Re}^{-1}\nabla^2\vec{v} \qquad (1)$$

Reynolds number is defined as

$$Re = \frac{1}{\nu}\sqrt{\sigma R_0/\rho}, \qquad (2)$$

where σ, ρ and ν are the surface tension, density and viscosity of the droplet medium, respectively. The equation (1) is non-dimensionalized using length scaled with undeformed droplet radius R_0, velocity with maximum deformation amplitude A, pressure with σ/R_0, and time with $\sqrt{\rho R_0^3/\sigma}$.

Limiting our considerations to axisymmetric deformations, droplet surface is given as function $R(t,\theta)$ of time t and polar angle θ. Motion of the droplet surface and flow velocity are coupled by the kinematic boundary condition

$$\frac{d}{dt}\left(R(\theta,t) - r\right) = 0\,\Big|_{r=R}, \qquad (3)$$

the tangential stress condition

$$(\mathbf{T}\,\vec{n})\cdot\vec{t} = 0\,\Big|_{r=R}, \qquad (4)$$

and dynamic boundary condition

$$(\mathbf{T}\,\vec{n})\cdot\vec{n} = 2\sigma H\,\Big|_{r=R}, \qquad (5)$$

where $\mathbf{T}$ is the Newtonian stress tensor and H the mean curvature of the droplet surface.

Due to nonlinearities arising from inertia, capillarity and coupling of the surface kinematics to the velocity field, solution of the above equations is a nontrivial free-boundary problem. Assuming small amplitudes ($A \to 0$) and neglecting viscous term ($Re \to \infty$), the problem becomes linear and can be solved analytically.

The widely known linear, irrotational approximation given by Lamb [3] describes the instantaneous deformation of the droplet shape by an infinite series of the surface spherical harmonics:

$$R(t,\theta) = R_0\left(1 + \sum_{l=2}^{\infty} a_l(t) P_l(\cos\theta)\right). \qquad (6)$$

Each term l of the Legendre series $P_l(\cos\theta)$ describes one mode of droplet oscillation, characterized by its amplitude $a_l(t)$. The oscillation frequency Ω_l of each mode l and the decay time τ_l are simply given as:

$$\Omega_l^2 = \frac{\sigma l(l-1)(l+2)}{\rho R_0^3}, \quad \tau_l = \frac{R_0^2}{\nu(l-1)(2l+1)} \qquad (7)$$

The equation (7) shows that damping increases very quickly for higher oscillation modes. Therefore, in practice description of the 'natural' droplet oscillation can be limited to the first few modes.

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Diffusion of vorticity from the surface to the bulk of the droplet becomes essential for oscillating viscous droplet. Prosperetti [4] has shown that even in the linear limit, the droplet motion consists of modulated oscillations, with frequency and damping factor varying in time. Two limiting time scales can be defined. Initially ($t \rightarrow 0$) motion is that executed by damped harmonic oscillator of natural frequency Ω_l and damping τ_l (irrotational approximation). Normal mode analysis, including viscosity (Reid, Chandrasekhar) applies for $t \rightarrow \infty$. In between these two asymptotic regimes the correct solution is significantly different from either case. The characteristic time scale for vorticity diffusion $t_D = R_0^2/\nu$, in non-dimensional form is a reciprocal of Reynolds number (2).

For a large Re irrotational approximation will dominate during several periods of oscillations, whereas for $Re \rightarrow 1$ only during a short initial time of droplet motion vorticity diffusion is negligibly small. For decreasing Reynolds number, droplet motion approaches its aperiodic limit, which was found to be at $Re = 1.3$.

It is obvious that linear description has only very limited application to 'real' droplet oscillations. Indeed, experiments with oscillating droplets [5, 6], indicated that nonlinear effects are already observed for the fundamental mode if its amplitude exceeds 0.1. Even weak nonlinearity of this mode is accompanied by a strong nonlinear excitation of the higher modes [6]. The linear models completely fail for these modes.

One of the first nonlinear analysis of droplet oscillations was given for the case of inviscid liquids by Tsamopoulos & Brown [7]. Looking for strictly periodic oscillations they have found that the oscillation frequency of the fundamental mode decreases with increasing amplitude. The effect of small viscosity was incorporated into a nonlinear numerical study by Lundgren & Mansour [8]. They found that viscosity may have a relatively large effect on the behaviour of the higher oscillation modes, changing their near-harmonic resonance coupling with the fundamental mode. This result was confirmed when recently full viscous and nonlinear analysis of the droplet motion was given by Basaran [9] and Becker et al. [1]. Basaran has shown, using Galerkin-finite-element method, that both oscillation frequency and viscous damping are time/amplitude depended functions. This analysis confirms experimental findings [6] that effects of viscosity on mode interaction are more significant than it was anticipated in 'low viscosity' computations. However, aside from complexity, pure numerical approaches have limited practical applicability when direct comparison with experiments have to be performed.

In the second nonlinear and viscous approach [1] equations of motion are solved by deriving an appropriate system of ordinary differential equations by the use of the standard variational principle of Gauß. This seems to be well suited to the analysis of nonlinear droplet oscillations, since it offers the straightforward possibility of treating the boundary conditions as additional constraints of the Navier–Stokes equation. Since the method offers description of nonlinear droplet oscillations in terms of natural degrees of freedom, the straightforward comparison with the experimental data is immediate. In practice computational 'expensive' calculations of the viscous motion inside the whole droplet volume are not always necessary. It has been found [2] that further simplifications, limiting effects of viscosity to the surface boundary layer, are allowed at higher Reynolds numbers. In the case of free boundary conditions the vorticity of this viscous surface layer remains finite even while its thickness goes to zero with $Re \rightarrow \infty$. Lamb [3] made use of this fact in his calculation of the linear damping constants (7). Application of the irrotational approximation to the nonlinear equations has been found appropriate for the interpretation of large amplitude oscillations observed in the experiments performed with small ethanol and water droplets [2].

Another approach to the problem is offered by the formalism of classical mechanics. Describing droplet dynamics in terms of effective masses, kinetic and surface energy, and a dissipation tensor one can construct an ordinary differential equation for damped mechanical oscillator [2]. By evaluating nonlinear terms of the equation it has been possible to construct a simple, fast numerical tool to model droplet mechanics. It was found, that for typical (natural) oscillations of low viscosity droplets, results obtained using this 'reduced' model are equivalent to the irrotational nonlinear approximation.

Limits of applicability of the numerical models are usually difficult to estimate *a priori*. Hence, beside the model development efforts, their validation is not the least problem. In case of droplet oscillations nonlinearity of equation of motion (1) cannot be simply described by a single parameter, i.e. Reynolds number Re. At finite oscillation amplitude ($A > 0$) both, the relative value of nonlinear terms in (1) and coupling through the boundary conditions (3)-(5), dominate droplet dynamics. Therefore, usual estimates of 'strong' or 'weak' nonlinear effects are misleading. For example, nonlinearity of higher oscillation modes remains even at infinitesimal amplitudes. Hence, in the following we consider applicability of three theoretical models, namely M1 - a full viscous nonlinear model given in [1], M2 - irrotational nonlinear approximation and its 'reduced' form described in [2], and M3 - linear model of Lamb. Our primary aim is experimental validation of model M1, especially in the low Reynolds number range, where viscous effects become decisive. Then, applying M1 as numerically trusted model, we can also verify limits of the irrotational approximations.

2 EXPERIMENTAL

Details of the experimental set-up can be found in [6]. The test liquid is injected vertically through the nozzle to atmosphere. The droplets are generated by the controlled break-up of a laminar liquid jet. By properly adjusting the oscillation frequency of the piezoceramic driver in the nozzle the jet can be forced to break up into a row of practically monodispersed droplets, oscillating in axis-symmetric modes. The typical radii of the droplets used are in the range from 50 to $400\mu m$. Since the droplet velocity is relatively small (below 10 m/s) the influence of aerodynamic forces on the droplet shape is negligible. Water, ethyl alcohol and their mixtures with glycerin have been used as fluid medium. It allowed to cover a wide range of the Reynolds number (1.3 - 90) from the aperiodic limit to weakly damped oscillations.

The oscillations of the droplet are observed through a microscope using a CCD-camera. Bright field illumination is applied, i.e. the projection of the droplets appears as dark shadows in front of a bright background. To visualize the droplet oscillations a beat-frequency stroboscopic technique is applied. For further evaluation the images are acquired by an 8- bit image processor and saved on a computer hard disk.

As only the projections of the droplets can be observed, the primary condition for an efficient measurement is that droplets are formed axi-symmetrically, with their axis of symmetry parallel to the plane of observation, i.e. to the sensor area of the CCD-camera. Then the three-dimensional form of a droplet is completely defined by its two-dimensional projection. The contour of the droplet projection is obtained using specially developed computer code which automatically analyses droplet images. The detected points of the droplet boundaries are fitted to theoretically given contour function comprising a limited series of Legendre polynomials (6). For typical droplet deformations the number of terms can be limited to $lmax = 5$. Subsequently, temporal variation of the oscillation amplitude a_l for a sequence of 200-300 single images of the droplet is used to compare with the corresponding time series predicted by the theoretical models.

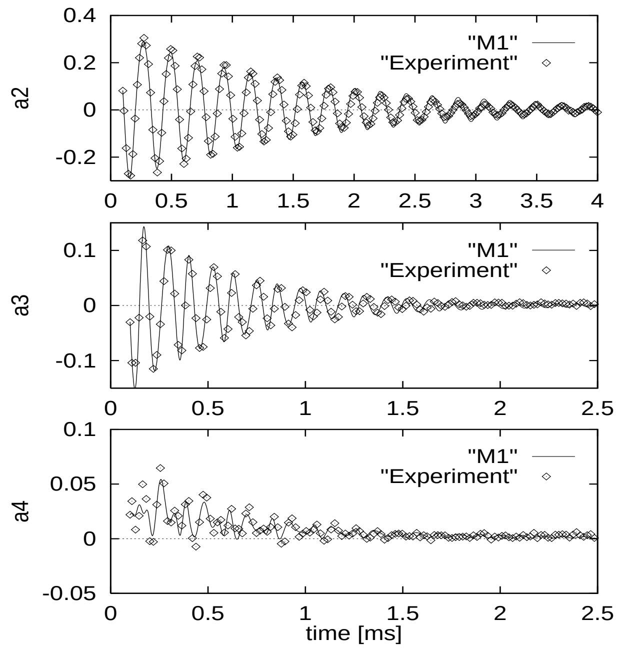

Figure 1: *Measured oscillation amplitudes $a_2 \ldots a_4$ of a water droplet (points); $R_0 = 87\mu$, $Re = 79$. Comparison with nonlinear model M1 (solid line).*

3 RESULTS

3.1 Validation of model M1

At 'high' end of Reynolds number damping effects are relatively low, what allows observation of several oscillation periods before their amplitude completely ceased. A typical time series of $a_2 \ldots a_4$ observed for the water droplet is shown in fig 1. Initial oscillation amplitude is relatively low ($a_2 \approx 0.3$). The nonlinearity of the fundamental mode appears mainly in form of a frequency drift and asymmetry of prolate and oblate surface deformations. More evident are the nonlinear effects of higher oscillation modes ($l > 2$). The nonlinear behaviour of these modes remains even when their amplitudes became very small. It is worth to notice that a_4 is practically always positive for the whole analysed period. To simulate numerically observed time series we start calculation at an arbitrary selected time t, using interpolated values of $a_2 \ldots a_{lmax}$ and their velocities $\dot{a}_2 \ldots \dot{a}_{lmax}$ as initial conditions. The comparison (Fig. 1) between experimental points and results generated using 'full ' model M1 shows very good agreement. Some differences visible for higher modes are mainly due to the limits of the experimental resolution of the very small deformation amplitudes.

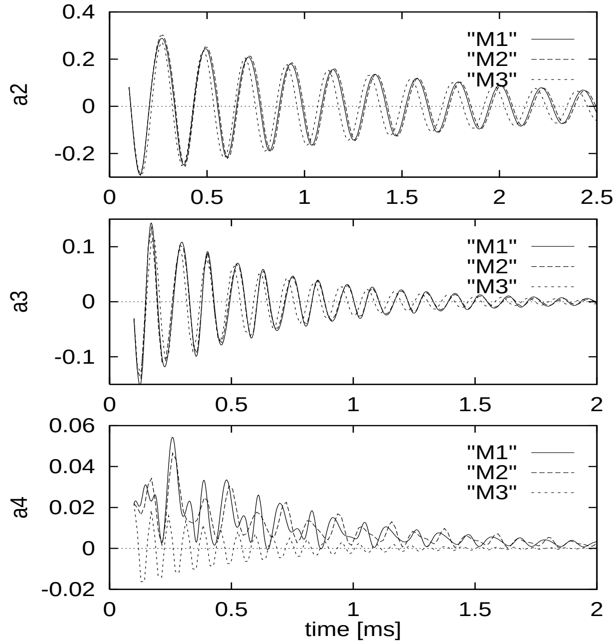

Figure 2: *Comparison of non-linear full viscous model M1, irrotational model M2, and linear model M3 for the case of previous figure.*

Figure 2 illustrates results generated by nonlinear and linear models for the case from figure 1. There is practically no difference between both nonlinear approaches. Linear model predicts behaviour of the fundamental mode only qualitatively.

An experimental result obtained for liquid of medium viscosity (Re=11) is shown in Fig. 3. The nonlinearities remain both for fundamental mode and higher modes and are fairly good followed by model M1. The linear description systematically overestimates oscillation frequency and damping of the fundamental mode, and completely fails for a_4.

Figures 4 and 5 show the effect of increasing viscous forces relative to inertial forces on large amplitude initial droplet deformation. The nature of the oscillations changes to critically damped regime for $Re = 1.8$. Simulation using the nonlinear model M1 remains in good agreement with experimental data. However, due to 'artificial' excitation of droplets[1], not all of the experimental runs could be perfectly reproduced by the model. These occurs, when initial velocity-vorticity field in the droplet significantly differs from its simplified description based on the initial droplet deformations a_l and their time derivatives.

[1] For viscous liquids large amplitude oscillations of droplets have been forced by applying non-sinus excitation and collisions of droplet.

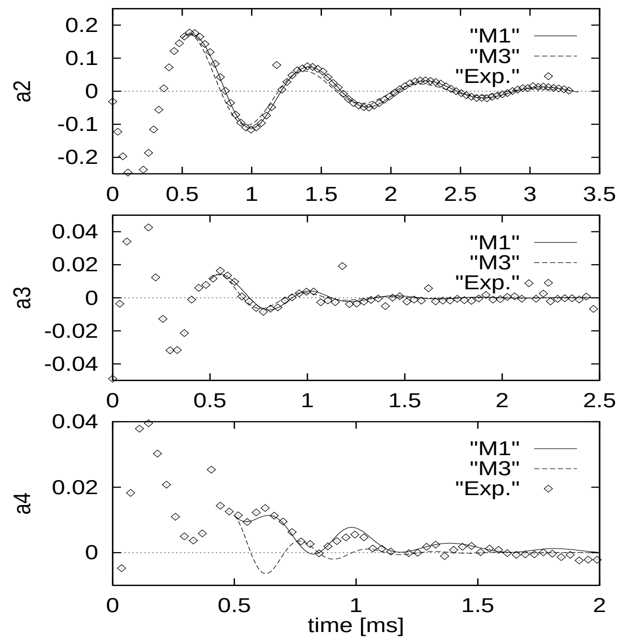

Figure 3: *Oscillation amplitudes* $a_2 \dots a_4$ *measured for a glycerin-ethanol mixture (points);* $R_0 = 155\mu$, $Re = 11$. *Comparison with nonlinear model M1 (solid line) and linear model M3.*

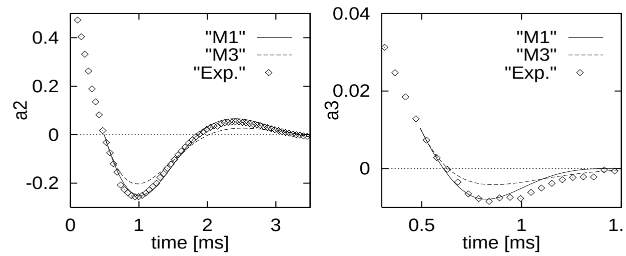

Figure 4: *Oscillations amplitude* a_2 *and* a_3 *measured for a glycerin-water mixture (points);* $R_0 = 417\mu$, $Re = 3.3$. *Comparison with nonlinear model M1 (solid line) and linear model M3.*

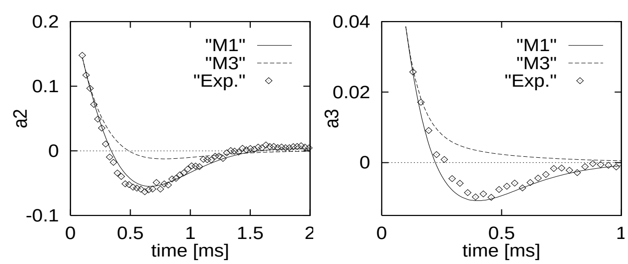

Figure 5: *Aperiodic limit of droplet oscillations measured for a glycerin-ethanol mixture (points);* $R_0 = 264\mu$, $Re = 1.8$. *Comparison with nonlinear model M1 (solid line) and linear model M3.*

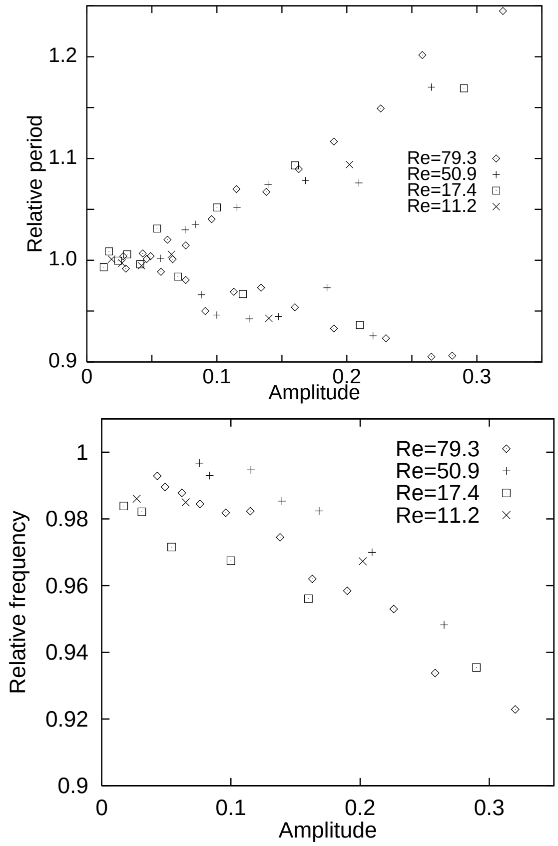

Figure 6: *Observed nonlinear characteristics of droplet oscillations, effect of oscillation amplitude and Reynolds number; (top) - asymmetry of periods of prolate and oblate deformation, (bottom) - relative oscillation frequency. The ordinates normalized with linear-model values.*

3.2 Effects of Reynolds number and deformation amplitude

Two main nonlinear characteristics of the fundamental mode appearing in experiments, i.e. asymmetry of periods for prolate and oblate deformations and amplitude dependent frequency shift have been displayed in fig 6. Apparently in considered range of parameters this nonlinear 'behaviour' of droplets does not show dependence on Reynolds number. Also another nonlinear significance, frequency modulation $a_4(a_2)$, described already in [2] , have been observed to remain down to the low Reynolds number limit (comp. fig 7).

A model calculation performed for $Re = 33$ and three initial droplet deformations, shown in Fig.8 (right), illustrates well amplitude dependent frequency shift for the fundamental mode. Nonlinear mode cou-

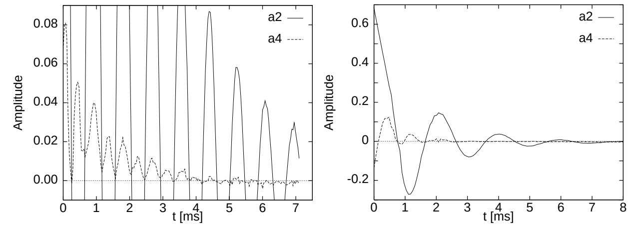

Figure 7: *Observed $a_4(a_2)$ modulation for oscillating droplets; left - $Re = 45.6$,right - $Re = 7.3$.*

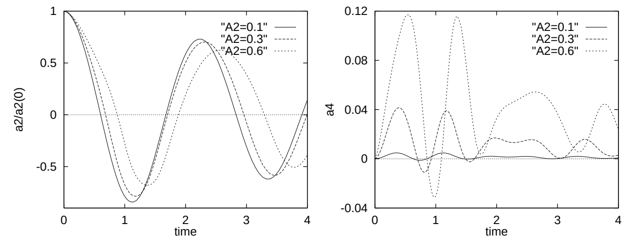

Figure 8: *Effect of deformation amplitude. Numerical simulation using model M1, $Re = 33$, initial conditions: $a_2 = 0.1, 0.3, 0.6$, $\dot{a}_2 = 0$, and all remaining modes set to zero; (right) - normalized amplitude a_2, (left) - excited amplitude a_4.*

pling immediately initiates also excitation of higher modes Fig. 8(left).

One may expect that in the limit of small initial deformation amplitude, the linear model of Lamb could give correct description of motion. It is only partly true. In fact, experimental and numerical runs performed for initial deformations below 0.1 have shown that at large enough Reynolds number ($Re > 50$) fundamental mode executes oscillations according to the linear predictions. But even for such low amplitudes linear description fails if we monitor higher modes. Their nonlinear characteristics remain. It becomes obvious, if we take into account, that not only deformation amplitude A, but also the Reynolds number Re must be small to justify linearization of (1). One may try to approach linear limit decreasing Reynolds number. Linear model of Lamb exceeds its limits for low Reynolds number. Hence, for comparison we take results given by Prosperetti (Fig.3 & Fig.4 in [4][2]), who used linear but fully viscous model. In Fig. 9 numerical results obtained with nonlinear model M1 are collected, starting calculations at three initial amplitudes, i.e. $a_2 = 0.1$, $a_2 = 0.3$ and $a_2 = 0.5(0.4)$ ($a_{l>2} = 0$,$\dot{a}_l = 0$). For comparison also results generated using linear Lamb's model are shown. If we com-

[2]Fig.4 contains probably misprint; time scale changed, and $\varepsilon = 0.3$ in our comparison.

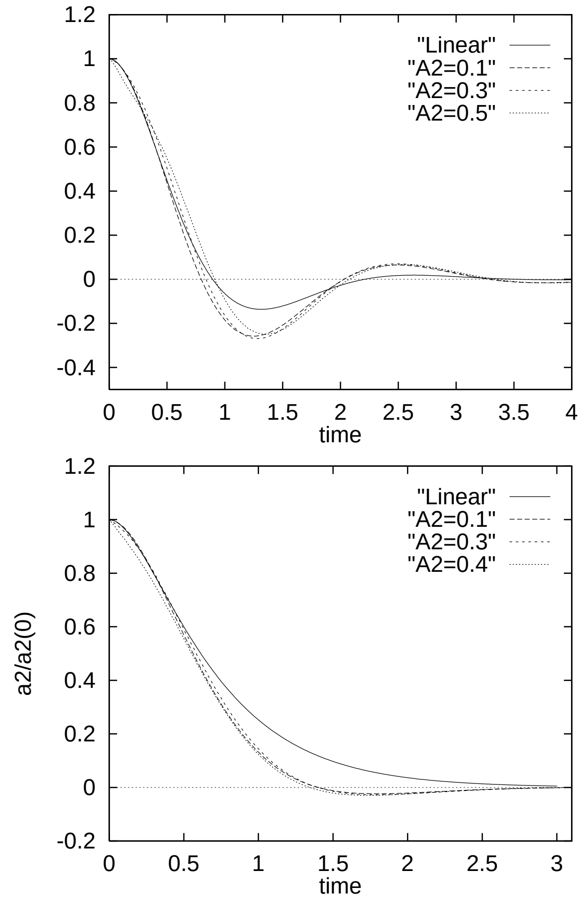

Figure 9: *Effect of deformation amplitude for near aperiodic limit. Linear model M3 and simulation using model M1 at three different initial deformations; (top)- Re = 3.3 - case to be compared with Fig.4 in [4], (bottom)- Re = 1.67 - case to be compared with Fig.3 in [4].*

pare Figs.9 with results of Prosperetti, we find that for small amplitudes ($a_2 = 0.1$) our nonlinear model M1 overlaps both his results entirely. Hence, model M1 fully comprise all key viscous effects present at the near aperiodic limit. However, when initial deformation amplitude exceeds $a_2 > 0.3$ nonlinearity starts to play a role. Also transfer of energy to the higher modes, becomes sufficiently high to perceivably excite amplitudes a_4. All considered irrotational models failed in this highly viscous limit.

6 CONCLUSIONS

Typical nonlinear properties, like the dependence of the oscillation frequency on the amplitude, the asymmetry of the oscillation amplitude, and mode interaction are observed even for relatively small initial droplet deformations. Viscosity of droplet medium in the investigated range has little effect on 'linearization' of motion. Nonlinear model, proposed previously [1], gives reliable description of droplet dynamics up to its aperiodic limit. Both, the location of amplitude extremes and the general form of the temporary amplitude variation (modulation and damping) coincide quite well with the experimental results. Irrotational, nonlinear description is practically equivalent for less viscous liquids. For typical 'natural' droplet deformations its applicability is limited to $Re > 30$.

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