OSCILLATORY BEHAVIOR OF NANODROPLETS

S. Arcidiacono, Y. Ventikos and D. Poulikakos
Laboratory for Thermodynamics in Emerging Technologies,
Swiss Federal Institute of Technology
Sonneggstrasse 3, CH-8092 Zurich, Switzerland
Tel: + 41 1 632 2738, Fax: + 41 1 632 1176,
E-mail: poulikakos@ltnt.iet.mavt.ethz.ch

ABSTRACT

We are performing molecular dynamics simulations in order to understand the mechanism of the oscillations for an argon nanodroplet. Lord Rayleigh[1], neglecting viscosity effects, was the first to investigate mathematically the nature of such oscillations for a macroscopically describable (based on the principles of continuum mechanics) initially spherical liquid drop in a gaseous environment. He found that the frequency of the oscillations, for a sphere of radius R, surface tension γ, viscosity ν, liquid density ρ_l and vapor density ρ_v are:

$$w^2 = \frac{16gn(n+1)(n-1)(n+2)}{p^2 R^3 [r_l(n+1) + r_v n]}, \quad [1]$$

where the fundamental mode is given by $n=2$.

Subsequently, Lamb[2] studied the effect of small viscosity. He found that the damping depends only on the size of the droplet and on viscosity. The time history of the amplitude of the oscillations reads:

$$b = b_0 e^{-bt} \quad [2]$$

where b_0 is the initial oscillation amplitude, b is the amplitude at time t and β a coefficient given by:

$$b = (n-1)(2n+1)n / R^2 \quad [3]$$

Chandrasekhar[3] obtained later more general results for a viscous spheroid. However, it is eqs. (1-3) that are relevant to the present study and will be compared to results of the molecular dynamics simulations.

In our simulations, the argon droplet is contains 329,245 particles (atoms), interacting with the Lennard-Jones pair potential:

$$\Phi(r) = 4e\left(\left(\frac{s}{r}\right)^{12} - \left(\frac{s}{r}\right)^{6}\right), \quad [4]$$

with a cut-off distance set to 4σ. The computational domain is a box of 52.7nm. The initial temperature is set to 108K and is maintained constant through a rescaling of the velocities, for an equilibration time of 50,000 time steps. The leap-frog algorithm is used to integrate the equation of motion with a time step of 0.01 ps.

For the calculations we use a parallel algorithm that employs simple spatial domain decomposition[4] on a 4 CPU DEC parallel machine.

After the thermodynamic equilibrium is reached, we evaluate the density and the normal component of the pressure tensor profiles in order to calculate the surface tension of the droplet. A comparison is performed with the values found in the literature, in order to validate the approach, and the agreement is deemed to be satisfactory.

The equilibrated droplet is then "squeezed" with an impulsively imposed velocity field to obtain an elliptic shape and it is subsequently allowed to oscillate freely.

We report a comparison between the above-referenced macroscopic analytical results with the ones obtained in this calculation. As we can see from eqs. (2-3), the dumping effect depends on the

surface area of the droplet, i.e. the number of particles present in the simulation has to be large enough to observe at least one period, before the oscillation is damped to magnitudes within our resolution limits. Figure 1 reports the analytical solution (2-3) and the calculated deformation during the simulation. The two results agree well proving that even in nanoscales the macroscopic theory is still valid. Figure 2 shows a graphical depiction of four time steps of the computation, after the droplet has been deformed. A high accuracy interpolation technique was used to convert the Lagrangian description of the atom simulation to an Eulerian-type iso-contour of density that defines the surface of the droplet.

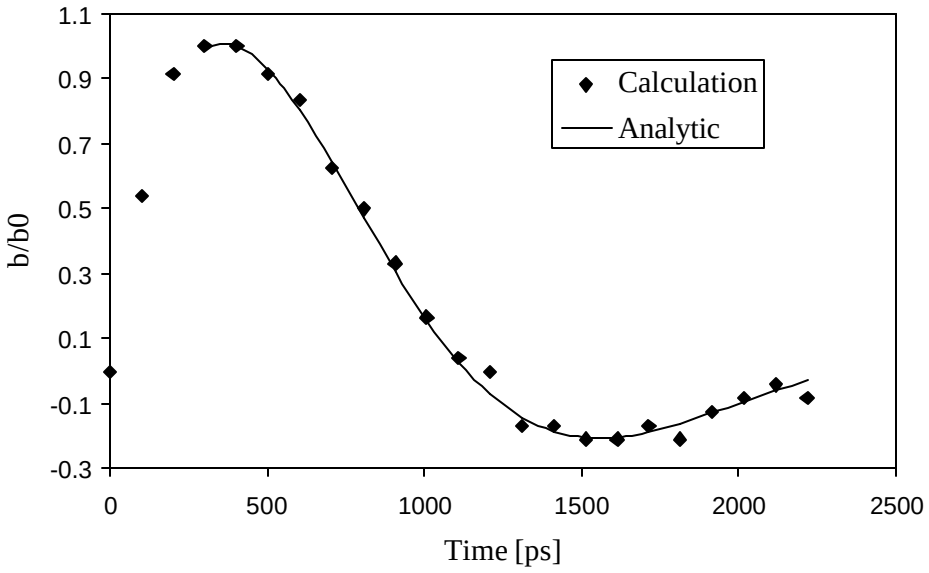

Figure 1: Comparison between eq. (2) and the calculation

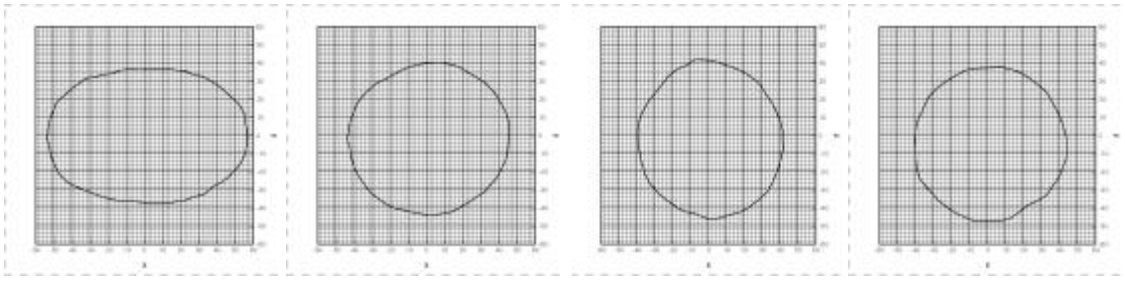

Figure 2: Profiles of the isoline $\rho=(\rho_l-\rho_v)/2$ at four different times steps. Interval between each snapshot is 706.3 ps, the sequence covers about one period.

[1] Lord Raylaigh, On the capillarity phenomena of jets, Proc. Roy. Soc.London 29 (1879), 71
[2] H. Lamb, Hydrodynamics, Dover Publications, New York, 1932
[3] S. Chandrasekhar, The osclillation of a viscous liquid globe, Proc. London Math. Soc. 9 (1970). 68
[4] S. Plimpton, Fast Parallel Algorithms for Short-Range Molecular Dynamics, Journal of Computational Physics, vol 117, p 1-19, 1995