



LIQUID MICROJETS -
 A USEFUL TOOL FOR THE MEASUREMENT OF MATERIAL PROPERTIES



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ABSTRACT

A nonintrusive optical method for the measurement of surface tension and viscosity of liquids has been developed. It is based on the fact that oscillations of small liquid drops are governed by surface tension and viscosity of the medium. The applicability of this dynamic method to the measurement of surface tension and viscosity of liquid metals will be discussed.

INTRODUCTION

Surface tension and viscosity are significant properties for the description of fluid flows with free surfaces, like dispersion of liquids or Marangoni convection. The latter, for instance, is encountered during the processing of silicon rods by zonal melting. Both of the fluid parameters, and in particular surface tension, are very sensitive to the experimental conditions and the purity of the sample, and are difficult to measure by standard methods if the conditions change quickly in time. In the present paper, we give a short introduction to the method which has been published previously [1], report on the progress of analyzing and describing droplet oscillations from experimental observations, and finally propose an experiment for the investigation of high temperature liquid jets.

THE OSCILLATING DROPLET

According to Lamb [2] the oscillatory motion of a droplet can be developed into modes if the amplitude is small and the viscosities of both the droplet and the surrounding medium are negligible. For the frequency of the n^{th} mode of oscillation one obtains:

$$\Omega_n^2 = \frac{(n-1) \cdot (n+1) \cdot (n+2) \cdot n \cdot \sigma}{[(n+1) \cdot \rho + n \cdot \rho_e] \cdot R^3} \quad (1)$$

where σ is the interfacial tension between the droplet medium of density ρ and the surrounding medium of density ρ_e , and R is the radius of the droplet. Since the droplet medium is an incompressible fluid and there are no periodic external forces acting on the particle, the first mode of interest is $n=2$.

The influence of viscosity on the oscillations were considered by Chandrasekhar [3], Reid [4], and Valentine & al. [5]. The general solution for a viscous droplet oscillating in a viscous medium was derived by Miller & Scriven [6] and Prosperetti [7]. From their considerations, it follows that for the case of a not too small liquid droplet oscillating in a gas the oscillation frequency remains nearly unaffected. The am-

plitude of the oscillation, however, is damped exponentially. From [6,7] the decay of the amplitude A_n of the n^{th} mode is described by:

$$A_n = A_{n0} \exp(-t/\tau_n) \quad (2)$$

with:

$$\tau_n = \frac{\rho \cdot R^2}{\mu \cdot (n-1) \cdot (2n+1)} \quad (3)$$

A_{n0} is the dimensionless amplitude at time $t=0$ with respect to the radius of the particle and μ is the dynamic viscosity. As an example, we display in Fig.1 the dependence of the oscillation frequency Ω_L for the second mode R of the droplet and its deviation $\delta = 1 - \Omega_V/\Omega_L$ from the "viscous" frequency Ω_V as calculated by Brosa [8] and Prosperetti [7], respectively. From Fig.1 it becomes immediately obvious that the influence of the viscosity on the frequency shift for droplets of $R \approx 0.1$ mm, as we apply them in our experiments, becomes negligible. The number of oscillations N during which the amplitude decays to $1/e$ is proportional to $\sqrt{(\rho \cdot R \cdot \sigma)}/\mu$. For low viscosity liquids like water, light oils and alcohols, and liquid metals, N will be sufficiently high as to permit the observation of a series of oscillations from which we can extract the frequencies and the logarithmic decrement of the corresponding modes.

EXPERIMENTAL

An overall view of the of the setup is displayed in Fig.2. It consists essentially of a droplet generator and a high-speed imaging device connected to a computer. The droplets are generated by the break-up of a laminar jet which discharges from a convergent nozzle. The diameter of the nozzle can be chosen in the range from 40 to 200 μm so that droplets of app. 80 to 400 μm can be generated. The pressure inside the plenum chamber

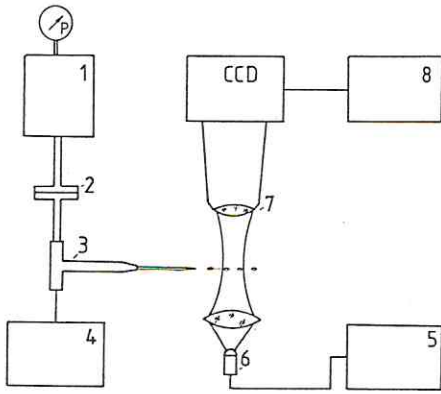


Figure 1. Schematic view of the experimental set-up. (1) pressurized reservoir; (2) teflon filter; (3) drop generator; (4) piezo-driver; (5) LED pulse driver; (6) LED; (7) microscope with CCD camera; (8) image processor and computer.

of the nozzle is modulated by a piezoceramic device [1]. Depending on the magnitude of the pressure oscillations, the break-up point of the jet can be controlled over a wide range, and by proper choice of the frequency, one achieves nearly monodispersed droplets which are oscillating in axially symmetrical modes. The droplets are observed through a microscope in a bright field illumination, and thus the projections of the droplets into the plane of observation appear as dark spots. The temporal change of the shape of these spots is recorded by a CCD imaging device in such a way, that a series of 15 to 20 subsequent pictures is superimposed on a single frame. The information contained in the frame is digitized and stored in a computer for further processing. A series covers approximately one period of oscillation. A detailed description of this technique and of the high speed light source used for illumination is found in [1,9]. Fig.3 illustrates this method for the case of a single droplet which has been exposed 5 times at a frequency of 25 kHz on one frame.

RESULTS

Evaluation and Analysis of the Modes of Oscillation. With help of computer-aided image analysis the boundaries of the droplet at corresponding moments of exposure are traced with 100 to 400 points. In Fig. 3, these points are already inserted into the photo of the droplet as black dots. In a second step, these points are matched to a series of Legendre functions $P_n(\cos\theta)$ up to 5th order, as they describe axially symmetrical oscillations correctly for the case of vanishing amplitudes A_n . Then, in a last step, the oscillation frequency Ω_n , the phase shift ϕ_n at time $t=0$ and the logarithmic decrement τ_n are evaluated for each mode by a six parameter least-square error minimization procedure. Thus, we end up with the following expression for the oscillating droplet :

$$r(t, \theta) = R_0 \{ \delta(t) + \sum A_n \sin(\Omega_n t + \phi_n) P_n(\cos\theta) \} \quad (4)$$

In this expression, δ is a volume integral of the Legendre function to fulfill

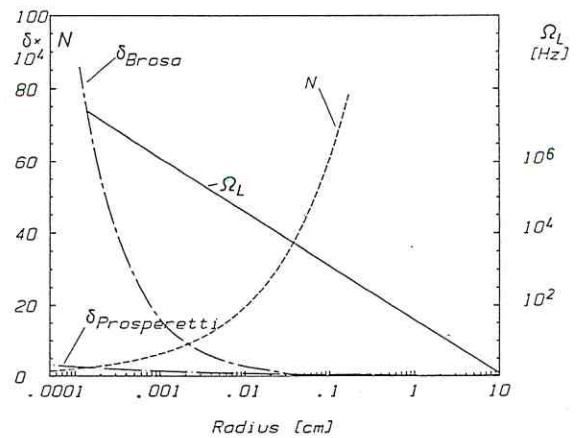


Figure 2. The second mode oscillation frequency of liquid iron, Ω_L ; the normalized deviation, $\delta = 1 - \Omega/\Omega_L$, of the Lamb's frequency, Ω_L , from the "viscous" frequency, Ω , given by Brosa and Prosperetti; and the number, N , of oscillations during which the amplitude of the 2nd mode decays to $1/e$ - shown as a function of the droplet radius, R .

the normalization condition. The value of δ depends very weakly on time and, even for large oscillation amplitudes, deviates insignificantly from 1.

Using Eq.(1 & 3) we get the surface tension and the viscosity of the medium. The results for the surface tension of water and ethanol droplets have been reported in [1]. They agree within 0.5% with values that we have obtained by the static ring method.

Observation and Influence of Nonlinear Effects. As has been already mentioned, Eq.(1, 3 & 4) are derived under the assumption that the droplets execute small-amplitude oscillations. A first prediction on the influence of nonlinearity on the duration of the period of oscillation for the case when viscosity can be neglected has been advanced by Brosa [10], who claims a frequency shift to lower values for large amplitudes. In order to quantify these effects from the experimental findings, ethanol droplets have been observed by stroboscopic illumination along their path from the origin where they separate from the jet until the region of almost final spherical shape. A single run consists of apr. 200 frames, which are taken with high temporal resolution compared to the period of oscillation. From each of these pictures the boundary contour has been extracted and developed into a series of Legendre functions. Figs. 4 and 5 display a time sequence of the dimensionless amplitude A_n for the modes $n=2$ and $n=3$, respectively. By introducing into Eq. (4) two additional parameters: α_n which takes into account the dependence of the oscillation frequency Ω_n on the magnitude of the amplitude, and β_n which allows for a non-symmetry of the reaction force between prolate and oblate deformation of the droplet, one arrives at the following expression for the amplitude A_n^* of the n^{th} mode:

$$A_n^* = A_n^2 \beta_n + A_n \sin\{\Omega_n [(1 - (\tau_n \Omega_n)^{-2})^{1/2} + A_n^2 \alpha_n] t + \phi_n \} \quad (5)$$

The lines in Figs. 4a and 4b display the decay of the amplitudes for an optimized set of the six fitting parameters: α , β , τ , Ω , ϕ , A_0 . Two conclusions can be drawn immediately from this analysis:

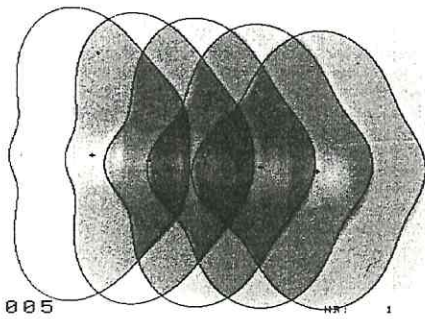


Figure 3. Multiple exposure of a strongly oscillating droplet.

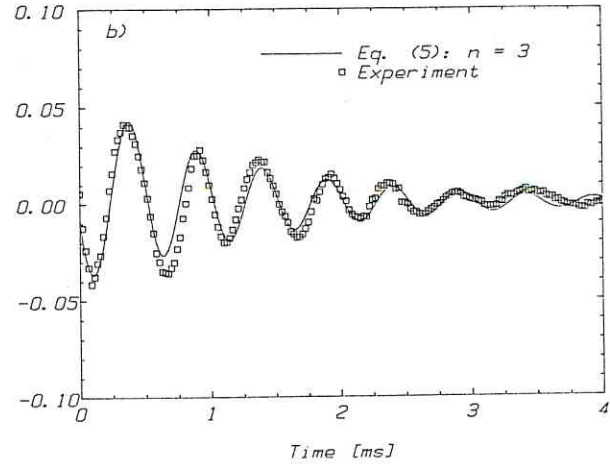
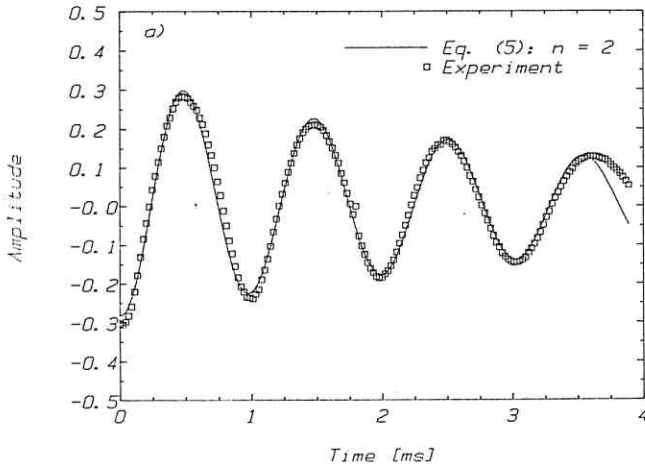


Figure 4. The amplitude of the second (a) and third (b) mode for an oscillating ethyl alcohol droplet. Droplet radius $R_0 = 177\mu\text{m}$.

1. There is a weak dependence of the frequency on the amplitude. For $A_2 = 0.4$ which is about the highest value we observe in our experiments, the frequency deviation of the 2nd mode amounts to about 7%. It falls down to below 0.5% for $A_2 = 0.1$ and results into an error of approx. 0.7% of the surface tension if the uncorrected relation Eq. 1 is employed.

2. In most cases observed up to now, the oscillations speed up at higher amplitudes. Sometimes, however, the frequency goes down.

3. There seems to be no significant excitation of higher modes from the base mode $n = 2$ if its amplitude does not exceed 0.3. This is inferred from the observation that the dynamic viscosity calculated from the measured decay time τ , yields a value very close to that found from tables. If there would be a strong excitation of higher modes which from the point of linear theory are more strongly damped (see Eq. 3), one should expect a larger damping decrement for the base mode than we have measured. For a short time lapse, immediately after the droplet is formed one should not expect a too good description of the particle oscillation by Eq. (5) [7].

4. As for the third mode, which is displayed in Fig. 4b, surface tension is still within 5% of the value obtained from the second mode, though the amplitudes very quickly disappear in the noise of the instrumentation. This noise, from our point of view, is probably the main cause for the more pronounced deviation of the viscosity.

5. From the experiments performed up to date, it seems possible to make corrections for the nonlinear effects and thereby keep

their influence small enough that this dynamic method can be developed into an operative tool for nonintrusive measurements.

MEASUREMENT OF HIGH TEMPERATURE LIQUIDS - A PROPOSAL

One of the aims of our investigations is the measurement in high temperature fluids like liquid iron or silicon. For this purpose we plan to perform an experiment which is shown schematically in Fig. 5. The material to be investigated is melted in a crucible. A high pressure inert gas which is supplied to forcing the liquid out of the containment

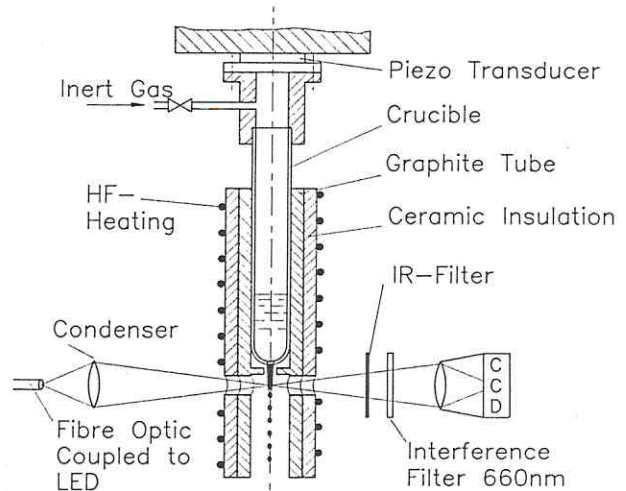


Figure 5. Experimental set-up for the measurement in high temperature liquids.

through a nozzle integrated into the closed lower end of the crucible. Both, crucible and part of the jet are placed inside a graphite cylinder which is heated by a high frequency induction generator. This technique of forming small liquid jets is well established. To force axially symmetric oscillation, it is best to excite the jet at its capillary eigenfrequency which for an iron jet of 0.1 mm of diameter is about 20 kHz. For this purpose the upper cold end of the crucible will be connected to a piezo-transducer. The jet and

the droplets are to be observed through a small hole drilled into the graphite cylinder. By adjusting the strength of the oscillations the break-up point of the jet can be shifted over a wide range. As a light source we can either use the pulsed LEDs which emit at 660 nm at a bandwidth of 30nm or the light produced by the jet itself. As for the first case, the radiation of the glowing jet and the graphite cylinder has to be held blocked by means of an infra-red filter and a narrow band interference filter. In the second case a gateable image intensifier matched to the CCD-chip could be applied.

By these measurements which are planned for the future, we hope not only to obtain precise values of surface tension and viscosity which are needed for the calculation of Marangoni flow, but also to be able to observe surface aging due to oxidization or solidification.

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