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Surface Tension Measurements on Levitated Aspherical Liquid Nickel Drops

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Abstract

An improved method for measuring the surface tension of liquid metals is presented. Surface oscillations of an electromagnetically levitated liquid nickel droplet are observed by a high speed video camera and digital image processing is used to evaluate the obtained spectra. Identification of the oscillation modes increases the precision of measurements significantly. In our experiments on Nickel we found a very good reproducibility and a decrease of surface tension with increasing temperature.

1. Introduction

The surface tension of liquid metals plays a central role in casting and welding processes. In particular, its dependence on temperature and concentration leads to the well known Marangoni convection. Due to the difficulties in measuring the surface tension, there is a large scatter between available data.
Conventionally, surface tension is determined by measuring the equilibrium shape of sessile or pendant drops. These methods are discussed critically by Passerone and coworkers (1982). An alternative approach is the oscillating drop technique using electromagnetic levitation, in which the oscillations of a levitated droplet about its equilibrium shape are observed. This method avoids any contact to a crucible and thus reduces not only systematic errors due to surface contamination but also allows deep undercooling of the liquid metal (Willnecker and coworkers (1986)). The restoring force for surface oscillations is the surface tension, which therefore can be related to the frequency of the oscillations (Reid (1960)).

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2. Levitation

An inhomogenous, alternating electromagnetic field has two effects on a conducting, diamagnetic body : Firstly, it induces eddy currents within the material, which, due to ohmic losses, heat up the sample (inductive heating) and secondly, it exerts a force on the sample pushing it towards regions of lower field strength (Lorentz force).

The power absorbed by a sample of volume V, averaged over a time τ is given by :

$$P = \frac{1}{\tau} \int_0^{\tau} \int_V j_{ind}^2 \, \rho \, dV \, dt \qquad (1)$$

where ρ is the electrical resistivity and j_{ind} the induced current density in the sample. For a homogeneous magnetic field B , an approximate analytical expression can be given for a spherical sample of radius R (Rony (1964)) :

$$P = \frac{3 \pi R \rho}{\mu_0^2} H(q) \, B^2 \qquad (2)$$

where

$$H(q) = q \, \frac{\sinh(2q) + \sin(2q)}{\cosh(2q) - \cos(2q)} - 1 \quad , \quad q = R/\delta \quad , \quad \delta = \sqrt{\frac{2\rho}{\omega \mu_0}} \qquad (3)$$

The quantity of interest in this expression is δ, the skin depth.

The time averaged force acting on a conducting sample can be calculated from

$$\underline{F_L} = \frac{1}{\tau} \int_0^{\tau} \int_V \underline{j_{ind}} \times \underline{B} \, dV \, dt \qquad (4)$$

For a weakly non-homogeneous magnetic field, the following expression can be derived :

$$\underline{F_L} = - \frac{\pi R^3}{\mu_0} G(q) \, \text{grad} \, B^2 \qquad (5)$$

where :

$$G(q) = 1 - \frac{3}{2q}\,\frac{\sinh(2q) - \sin(2q)}{\cosh(2q) - \cos(2q)} \qquad (6)$$

As can be seen, an inhomogeneous field pushes the sample into regions of low magnetic field strength. A more detailed discussion has been given by Lohöfer (1989,1991).

In figure 1, a schematic drawing of our levitation coil is shown. Due to gravity and the conical shape of the coil, the sample in levitation experiments are is usually not spherical. Figure 2 shows the shape of a nickel sample, which was solidified from high undercooling. The volume change upon freezing is assumed to be isotropic and, therefore, shape preserving, so that this picture gives a good imagination of the distortion of the sample from spherical shape in the liquid state in our levitation facility. The problems, which arise from this asphericity, are treated in the next chapter.

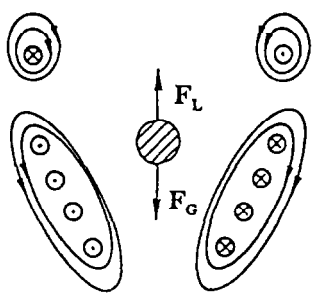

Fig.1 : Levitation coil
F_G is the gravitational force
F_L is the levitation force

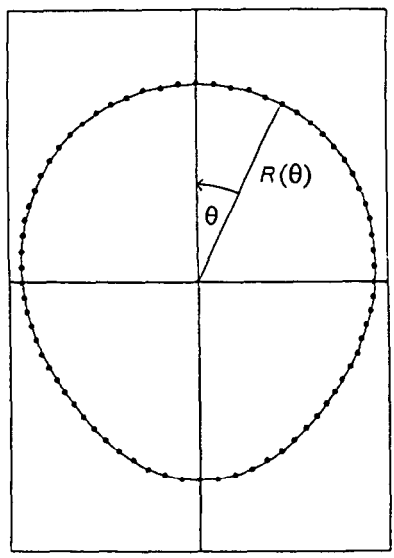

Fig.2 : Shape of liquid sample

3. Surface tension measurements

In the oscillating drop technique, the frequency of the oscillations of levitated droplets about their equilibrium shape is measured. The oscillation frequency ν of a non rotating, spherical droplet is related to the surface tension according to Rayleigh's law (1879) :

$$\sigma = \frac{3\pi}{l(l-1)(l+2)} M \nu^2 \qquad (7)$$

where M is the mass of the droplet and l labels the normal modes of the oscillations. The fundamental mode l=2 yields the Rayleigh frequency ν_R.
For aspherical, non rotating droplets, the l=2 mode is shifted and split into three peaks, ($|m| < 2$), for rotating ones five peaks ($-2 < m < 2$) can be detected (Busse (1984)).
Recently, Cummings and Blackburn (1991) derived the following sum rule to obtain the Rayleigh frequency for split modes :

$$\nu_2^2 = \frac{1}{5} \sum_{m=-2}^{m=+2} \nu_{2,m}^2 \; - \; 2\,\nu_\tau^2 \qquad (8)$$

where ν_τ is the mean translational frequency of the droplet. The subtraction of this term accounts for the apparent increase in surface tension due to the action of the applied magnetic field.

The above formula can only be applied to spectra exhibiting all five peaks. If this is not the case, one needs identification of the peaks to obtain the correct result.
This can be achieved by application of digital image processing in a simple, unambiguous manner as follows.
The oscillating drop is observed by a video camera from the side and from the top and recorded on videotape. The images are analyzed for certain geometrical parameters, which can distinguish between the different modes. A subsequent Fourier Transformation of these time dependent parameters then yields the frequency spectrum.

In a levitation facility, the radius of an axisymmetric sample undergoes oscillations given by

$$R_{l,m} = R_0 \, (1 + f(\theta) + \varepsilon \cos(2\pi\nu_{l,m}\, t)\, P_l^m(\cos\theta) \cos(m\phi)) \qquad (9)$$

where $f(\theta)$ describes the asphericity, $|m|<l$ and P_l^m is an associated Legendre function.

For frequency analysis, we evaluated first the lateral cross section of the sample Q(t). For a side view, one obtains (Egry (1990)) in first order :

$$Q(t) = \pi R_0^2 \left(1 + \frac{2}{\pi} \int_{\theta_1}^{\theta_2} f(\theta)\, d\theta + \varepsilon \cos(2\pi\nu_{l,m}\, t) \frac{2}{\pi} \int_{\theta_1}^{\theta_2} P_l^m(\cos\theta)\, d\theta \right) \qquad (10)$$

where θ_1, θ_2 define the visible area. The Fourier Transform of the resulting signal Q(t) yields the linear frequency spectrum. In figure 3, a spectrum for a nickel sample at T=1520°C is shown. It shows one peak at ν=7 Hz due to translational motion of the sample and some peaks of the split l=2 mode, which can not be identified yet. This is the same situation as in conventional photodetector measurements (Keene, Schade).

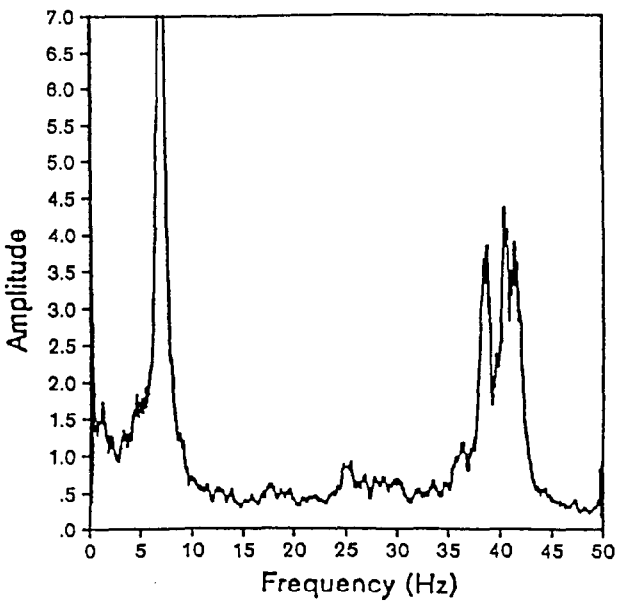

Fig.3 : Fourier Transform of Q(t) for nickel at 1520°C

The modes can be identified most easily by viewing the sample from the top. Defining the quantity

$$S^-_{l,m}(t) = R_{l,m}\left(\frac{\pi}{2}, \phi_0, t\right) - R_{l,m}\left(\frac{\pi}{2}, \phi_0 + \frac{\pi}{2}, t\right) \qquad (11)$$

it follows immediately from eq. (8), that

$$S^{-}_{2,0}(t) = 0 \tag{12}$$

which means, that the m=0 mode is totally supressed in the signal and is not to be observed in the spectrum of S^{-}.
If one defines

$$S^{+}_{1,m}(t) = R_{1,m}(\frac{\pi}{2}, \phi_0, t) + R_{1,m}(\frac{\pi}{2}, \phi_0 + \frac{\pi}{2}, t) \tag{13}$$

it follows, that

$$S^{+}_{2,2}(t) = \text{const} \tag{14}$$

which means, that the m=2 modes should not appear in the spectrum of S^{+}.
The Fourier transforms of the signals $S^{-}(t)$ and $S^{+}(t)$ are shown in figure 4.

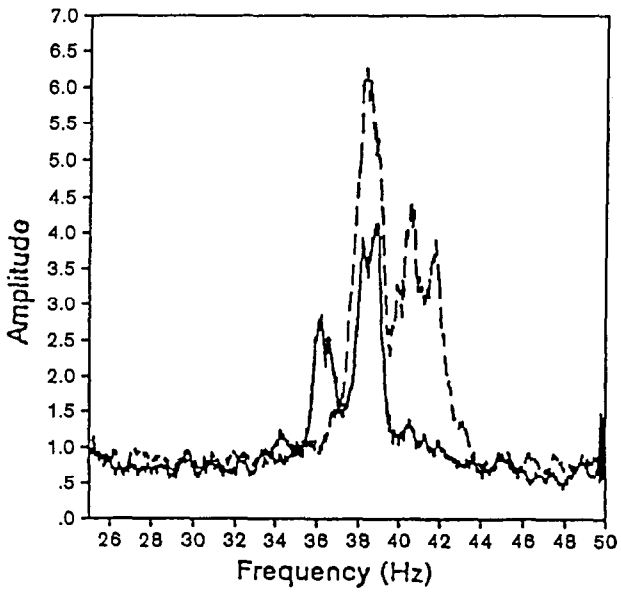

Fig.4 : Fourier transform of S^{-}(dashed line) and S^{+}(full line)
(see text for details)

For S^-, the peak at ν=36 Hz disappears, while for S^+ the two peaks at ν=41 Hz vanish. The latter can be assigned to the |m|=2 modes, whereas the peak at ν=36 Hz can be assigned to the m=0 mode. This gives a complete identification of the l=2 peaks and eq. (8) can now be applied to our spectra.

Results

Using the method described above, we measured the surface tension of pure Ni (99,99%, Johnson Matthey) on different samples and at different temperatures in the range from 1300°C to 1620°C (150 K below to 170 K above the melting point). A detailed description of our levitation facility is given by Herlach et al (1984). As atmosphere, we used a He/H_2 mixture to reduce the oxygen content of our sample. For evaluation of the surface tension, we used eq. (7) for l=2 and ν_2 as obtained from eq (8). The obtained data were fitted to the linear relation

$$\sigma(t) = \sigma(T_m) + \frac{d\sigma}{dT}(T-T_m) \qquad (15)$$

The errors of the measured quantities are assumed as ΔT=10K, $\Delta\nu$=0.25Hz and Δm=1mg, which gives an error for σ in a single measurement of about $\Delta\sigma/\sigma$=1%. Figure 5 shows our results and, for comparison, some previously published data. Please note the two data points at T=1520°C which result from two independent measurements and are indicative for the reproducibility of our measurement. The fit parameters are listed in table 1.

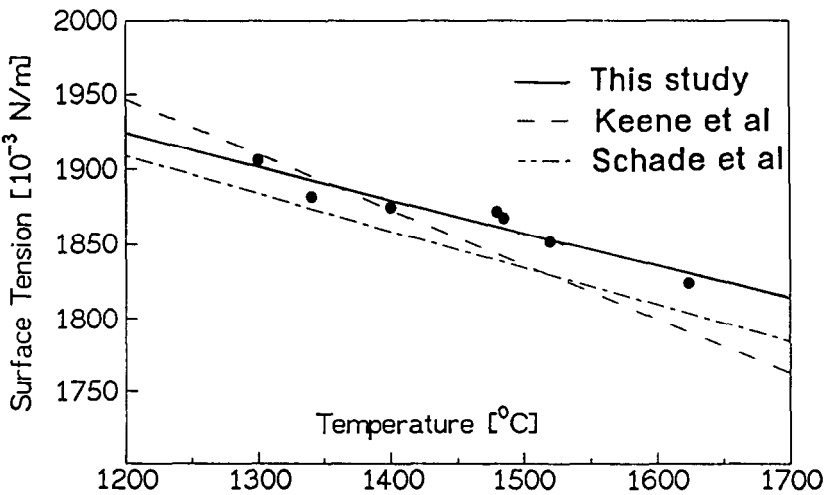

Fig.5 : Temperature dependence of surface tension for nickel

references	$\sigma(T_m)[10^{-3}\frac{N}{m}]$ $T_m = 1455°C$	$\frac{d\sigma}{dT}[10^{-3}\frac{N}{m\,K}]$
This study	1868	-0.22
Keene et al	1854	-0.364
Schade et al	1846	-0.25

Table 1 : Surface tension of nickel

Discussion

Our measurements lie slightly higher than those reported in literature, but show nearly the same temperature dependence. This can be explained with the elimination of systematic errors in the assignment of the modes and a very low oxygen and sulphur content of our samples, which was measured as less than 20ppm before the experiment. The error of each single experiment run is assumed to be less than $\Delta\sigma/\sigma=1\%$, which mainly results from the resolution of the obtained spectra ($\Delta\nu=0.25$Hz), the errors of temperature measurement ($\Delta T=10$K) and determination of mass ($\Delta m=1$mg) can be neglected. Other possible errors are non homogenous temperature distribution in the sample and contamination of the sample during the experiment, which are assumed to be negligible.
In conclusion, this new method eliminates systematic errors due to peak assignment and allows to measure the surface tension on axisymmetrical drops with the same precision as it would be possible on spherical drops.

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