THERMOCAPILLARITY AND RADIATIVE HEAT FLUX OSCILLATIONS

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Abstract
We present a detailed experimental study of the thermocapillary motion of an aniline drop in a stably stratified fluid system driven by a laser beam. The thermocapillary motion of drops is the result of the temperature dependence of the interfacial tension. If the surface of the drop is subject to thermal gradients, then non-equilibrium surface tension effects appear, which in some cases can move the drop. We measure some of the velocity induced fields, vorticity, oscillations and intermittency of this complex flow. The source of the non uniformity of the temperature of the surface can be, as is in this experiment, the non uniform heating of the floating drop by a laser beam. In recent years, the thermocapillary movement of bubbles and drops under the influence of laser radiation has received more experimental attention thanks to the improvement in the flow visualization techniques.

Keywords: Thermocapilarity, Laser flow, PIV

1 Introduction
The thermocapillary motion of a drop in a neutral buoyant position is initiated by a strongly non homogeneous heating by the flux of the laser radiation coming to its surface. As the process has been studied [1,2] experimentally with a horizontal laser source while previous experimental and theoretical configurations were often vertical [3-6], the effect of buoyancy produces also strong oscillations, which may be analysed in detail following Extended self similarity techniques, such as those indicated in [7-11]. In the experiment, different degrees of stable stratification may be used in order to balance drops of different substances and irradiances. Drop sizes and compositions are discussed together with the radiation conditions. The stable conditions and the experimental procedure is discussed in [1]. The analysis presented here was done for several different stabilities, drop sizes, laser power and irradiance. Diode array spectrophotometers are capable of acquiring complete UV/Visible absorbance spectra. The key is that the grating of these instruments is fixed, and rather than moving to measure the spectra, hundreds of detectors are placed at the exit of the monochromator. The detectors are all integrated on a single silicon chip called a photodiode array. The diodes act as capacitors that discharge in proportion to the incident light flux. The system is a single beam instrument, so a first scan on a cuvette containing just the solvent to determine the intensity of the lamp at each wavelength, I0(l). Then the sample is placed in the same cuvette and the spectrum measured again. The absorbance is then calculated from the ratio of the two spectra. Figure box 1 shows the configuration and value of the velocity induced by the thermocapilarity process, references [3-6] describe the theoretical interaction between the spherical distribution of the local heat flux and the bubble or drop surface tension.
Equation (1) shows the velocity of the drop in the configuration shown in figure 1. The notation is as follows: index 1 corresponds to salt solutions and index 2 correspond to drop. The drop velocity $U$ ($U_{Th}$) is expressed in terms of drop diameter per unit time (second), $J$ is the intensity of the laser radiation flux (cal/cm²s), $\lambda_{1,2}$ is the thermal conductivity of the two fluids (cal/cm.s.oC), $\mu_{1,2}$ is the shear viscosity of the two fluids (g/cm.s), $\sigma$ is the surface tension (g/s²), $T$ is the temperature (oC), and $a$ is the radius of the drop (cm).

The expression (1) for the velocity is based on the use of some important simplifications of the problem [2]. The movement of the droplet is steady and it moves in an infinite space which is filled with a homogeneous fluid of constant temperature and zero velocity at infinity. The flux of the laser radiation is totally absorbed by the droplet surface, and the reflection and refraction of the rays on its surface are considered negligible. The non homogeneous distribution of the laser intensity in the cross section of the beam is supposed to be negligible. Equation (2) shows how the absorbance is evaluated from the comparison of the two spectra mentioned above.

$$U = \frac{1}{6} \frac{\sigma_J}{2(2\lambda_1 + \lambda_2)(2\mu_1 + 3\mu_2)}$$

$$A(\lambda) = \log \left( \frac{I_o(\lambda)}{I(\lambda)} \right).$$

Figure 1: Configuration of the Thermocapillary motion of a droplet heated by radiation (Oliver et al 1988), Rednikov and Ryazantsev 1989). The different aniline drops float at a density interface.
Table 1: Some of the measured absorbances used in the experiments [1,2]

<table>
<thead>
<tr>
<th>Substance</th>
<th>Absorbance (cm(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pure aniline (cuvette 1 cm width)</td>
<td>4.79 (10^{-2})</td>
</tr>
<tr>
<td>Nigrosine-Aniline (2.5 g/l, cuvette 1 cm width)</td>
<td>&gt;=3.786</td>
</tr>
<tr>
<td>Nigrosine-Aniline (0.5 g/l, cuvette 0.1 cm width)</td>
<td>5.514</td>
</tr>
<tr>
<td>Water (cuvette 1 cm width)</td>
<td>7.63 (10^{-5})</td>
</tr>
</tbody>
</table>

Figure 2: Three different aniline drops on a stable density interface (the coloured fluid is the denser layer) produced by brine. The aniline drops have different diameter (from 4 mm to 2 mm) and colour (from orange to black)

2. Design Conditions and Results

Once the aniline drop was carefully placed at the center of the density interface (either sharp or created with a linear vertical density gradient using the two tank method [8,9], the thermocapillary motion of the drop was initiated by its strongly non homogeneous heating by the flux of the laser radiation coming to its surface. As a source of radiation we used the coherent Verdi V5 Laser system with a wavelength of 532 nm (green colour), an output power up to 2 W and a beam divergence less than 0.5 mrad. The initial diameter of the laser beam is equal to 2.25 mm (±10%) and it can be increased with the use of suitable optics.

As mentioned before, the roughest estimation is the assumption of the full absorption of the radiation by the drop surface. To obtain the optimal conditions for the observation of the thermocapillary motion, the absorption of laser radiation by the liquid of the drop should be as much as possible to ensure a strongly non homogeneous distribution of temperature on the surface drop. The working liquid aniline appears to be a suitable liquid for use in the proposed experiment because is almost immiscible in water (and salt solutions) and its absorption coefficient can be modified by adding some chemical substance. First, we obtained the aniline absorbance spectra. A Hewlett Packard 8452A Diode Array Spectrophotometer was used which is capable of acquiring complete UV/Visible absorbance spectra. The system is a single beam instrument which permits to obtain the dependence of the absorption coefficient on wave length in standard form, in [2] more details may be found, in figure 3 an example of the motion of the drop,
towards the right can be clearly seen in the presented sequence. In figure box 1, the motion is towards the left. In both cases the horizontal motion takes place towards the warmer point in the spherical drop, where the laser plane beam interacts the drop. The recirculation patterns in the flow near the drop from [3-5] is shown in box figure 4. The parameter $A$ describes the streamlines of the inside and outside flow in a non-stratified flow. In the present sharp interface configuration, we define the buoyancy or Brunt-Vaisalla frequency, $N$, in terms of the density gradient $\rho$, gravity $g$, as:

$$N = -\sqrt{\frac{g \partial \rho}{\rho \partial z}}$$  \hspace{1cm} (3)

Figure 3: Time sequence with several frames. The aniline droplet, located on a stable density interface, moves towards the green laser beam, that is, in the direction of the temperature different situations.

Figure 4: Sketches of the stream functions for the aniline droplets, for different conditions and substances, (both for fluid drops and solid particles), $A$ is a geometrical parameter.
Figure 3 shows a time sequence of frames corresponding to the experiment carried out. In the beginning the droplet has been placed on a level of neutral buoyancy (figure 3a) and began to move after a short relaxation time due to the inclusion of the laser beam (figure 3b). The thermocapillary migration of the aniline droplet is evident in the time sequence of frames. During the movement the droplet keeps its spherical form and remains very close to this zero buoyancy plane in spite of its temperature change (figures 3c to 3g). Simple approximate estimations show that the density of the drop will decreased due to heating to 1.02 g/cm³. However, vertical displacement of the drop during the time interval of the order of several minutes will not be more than 0.4 mm. Figure 5. shows another experiment where the slight asymmetry of the thermal induce surface tension produces strong vertical oscillations.

Figure 5: Time sequence of an experiment showing the vertical oscillations of the bubble.

In the models describing the flow, buoyancy has not been fully modeled and furthermore contain empirical constants determined for particular conditions and calculated only at first order. Video analysis shows in figure 6, both the rectilinear movement of the aniline droplet, located on a stable density interface as it moves towards the green laser beam, that is, in the direction of the temperature gradient (Left).

Figure 6: (left) Horizontal position obtained from a time sequence with several frames of the centre of the aniline droplet, located on a stable density (right) Vertical position.

3. Experimental Conclusions

The description of model (1) does not solve the problem, since this model is not closed, which makes it necessary to use various approximations and estimates. This situation makes topical any attempt to expand the limits of modeling of local mixing and motion of the bubbles, which is possible only within the framework of two-point turbulence models. We perform a correlation particle image velocimetry (CPIV) on a sequence of frames such as that shown in figure 5 which makes it possible to map the plane velocity and vorticity flow near the drop and interface. Some results are shown in figure 7 relating to the determination of the strong stability. This is an indication of non-linear internal gravity waves as variations in N are measured.
The main result of this experiment is that we observed the thermocapillary movement of a single droplet of nigrosine–aniline solution caused by a laser beam heating. This experimental configuration seems to be the first one [1,2] in which the observation of the thermocapillary motion, which is initiated by a laser beam motion, are observed and measured clearly because the phenomenon is not complicated by either natural convection, gravity, the presence of other droplets, the drop vaporization or the presence of limiting walls. The average experimental velocity of the drop was about 0.02 - 0.5 cm/s. Strong vertical oscillations of average frequency $N$ decrease in time.

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References