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Evolution of internal flows in mechanically oscillating sessile droplets undergoing evaporation

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HIGHLIGHTS

- Time averaged flow features are observed – upward drift, interface vortex pairs.
- Seemingly horizontal segregations planes are observed.
- Evaporation of the droplet leads to resonances of different modes.
- Upward drift is dominant at instances away from any mode resonances.
- With evaporation segregation planes show downward shift.

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ABSTRACT

Sessile water droplets on hydrophobic substrates have been subjected to mechanical oscillations to excite mean streaming flows in the liquid phase. The driving frequencies are below 1 kHz. The flow features have been imaged at a frame rate much lower than the driving frequencies. Counter-rotating vortices are observed along the oscillating liquid-vapor interface (droplet free surface) while an upward drift originating from the substrate exists in the interior. The mean flow features arise out of steady streaming from the substrate and the oscillating liquid-vapor interface. The upward drift is segregated along the droplet height by seemingly horizontal planes. These planes are a characteristic of the time periodic velocity rather than the mean flow. Furthermore when the oscillating droplet is allowed to evaporate under stationary ambient conditions and constant driving frequency, these flow features evolve in a spatio-temporal fashion. In one of our previous studies, we have shown that the oscillation mode of the droplet changes when allowed to evaporate. Mode transition therefore also leads to evolution of the mean streaming flow. The aim is to provide a physical understanding of the evolution of the time averaged flow. This work is motivated by studies related to manipulation of nano-particle deposition patterns in colloidal droplets using controlled oscillations.

1. Introduction

Internal mixing in droplets and evaporation of particle laden droplets is vital to a number of specialized areas such as biological and micro fluidic applications (Song et al., 2003; Ma et al., 2011; Giuffrida et al., 2015; Tangen et al., 2015). It also forms the building block in industrial applications such as inkjet printing (De Gans and Schubert, 2004), patterning through dip-coating (Brinker et al., 1999; Darhuber et al., 2000) to name a few. In most of these applications, evaporation leads to internal flow structures that preferentially transport particles or enhance mixing within the droplet at different spatial locations. For droplets with high contact angle (as in the present case) this flow is buoyancy driven (Kang et al., 2004). Now modulating the internal flow offers the possibility of controlled spatio-temporal distribution of particles (since particles are carried by the flow field) enabling newer engineering designs. Acoustic excitation (Brunet et al., 2010), electrowetting (Ko et al., 2008; Lee et al., 2009; Mugele et al., 2011; Malk et al., 2011; Mampallil et al., 2011), and mechanical oscillations (Whitehill et al., 2010; Sanyal et al., 2014, 2016; Kim and Lim, 2015) are some of the prevalent methods to excite flows inside such droplets. As will be discussed subsequently in details, oscillations generate both periodic and time averaged flow. The latter is instrumental in governing particle transport in droplets. Primarily for this reason visualization of such internal flows is required to gain insight into the mixing dynamics induced by oscillations. In recent years, increasing number of studies has focused on

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electrowetting induced internal mixing. Mainly two types of flows exist in such a scenario – low and high frequency flows, the origins of which are completely different. At low frequencies (<10 kHz) flows are induced through interface oscillations (Ko et al., 2008) while for high frequencies they are of electro thermal nature (Joule's heating by the electric field) (Lee et al., 2009; García-Sánchez et al., 2010). For low frequency oscillations, a non-zero time averaged flow exists because of non-linear effects. Two different explanations have been provided for the origin of such flows. The first one involves steady streaming similar to acoustic streaming (Ko et al., 2008). This flow may originate at the substrate boundary layer in the normal direction (Batchelor, 2000). The other mechanism reported is Stokes's drift (Mugele et al., 2011; Oh et al., 2012) for droplets with oscillating contact lines. In such cases, asymmetry in shape during the advancing and receding phase of the contact line motion induces a net drift along the oscillating free surface. However the directions of the time averaged flow explained by these two mechanisms are opposite to each other. The steady streaming is directed upwards along the symmetry axis. The Stokes's drift on the other hand is directed upwards along the free surface but downwards along the symmetry axis. For brevity this oscillating free surface will be referred to as the interface in any subsequent mentioning.

For low frequencies, an alternate method for exciting internal flows is mechanical oscillations of droplets through substrate (support) vibration. The substrate can be set to periodic oscillations which at high Reynolds number induce internal mixing. Although origin of these flows is similar to that of low frequency electrowetting, there are some inherent variations in the way they are setup. Electrowetted flows are generally sustained by contact line oscillations. For mechanical oscillations, if the substrate acceleration is small, the contact line does not oscillate. Data on such internal flows is surprisingly scarce. Therefore to bridge this gap, it is important to investigate the origin of the internal flow features in a mechanically oscillated sessile droplet. These features include counter-rotating vortices along the interface, an upward drift from the substrate and horizontal segregation planes along the symmetry axis. It should be noted that the velocity magnitudes are modified because of the optical distortion of the oscillating interface. Now for a static droplet, a correction method has been developed (Kang et al., 2004) which has been previously applied for small oscillations of the droplet (Lee et al., 2009). For a dynamically oscillating interface (as in the present case) with substantial amplitude, no existing correction method is applicable (to the best of our knowledge). This however does not affect our conclusions regarding either the origins of the observed flow features or the evolution of the drifts along the symmetry axis (presented at the end of the paper). However actual magnitudes of the drifts can be different due to optical distortion.

In addition to the above, we have investigated how these flow features evolve when the droplet is allowed to evaporate in a controlled environment. The effect of "modified" diffusion driven evaporation on the oscillation dynamics of a sessile droplet has been previously reported by us (Sanyal and Basu, 2016). There we have shown experimentally how the mode of oscillation changes with controlled evaporation i.e. the droplet exhibits different oscillation modes at different time instances during the entire evaporation lifecycle. We have also explained the pathways of mode transitions. In the current paper, we will make use of such reported pathways to explain the evolving flow features. The significance of considering evaporation lies in the evolution of the different flow features which affect particle/colloid transport and mixing in functional droplets (Ko et al., 2008; Mugele et al., 2011; Oh et al., 2012).

Recent studies show that particle deposition in colloidal droplets can be controlled using externally imposed oscillations (Sanyal et al., 2014, 2016; Eral et al., 2011; Mampallil et al., 2012, 2013; Zhang et al., 2016). As mentioned earlier in the section the oscillations generate mean flows which lead to changes in particle deposition patterns by otherwise stationary evaporation conditions (Deegan et al., 1997; Brutin, 2013; Chen and Evans, 2010). Thus for effective control of surface pattering, it is mandatory to have sufficient insights into the internal flows generated by mechanical oscillations and how they vary with evaporation.

2. Materials and methods

In the current work, we have used sessile water droplets on hydrophobic substrate as our test cases. The hydrophobic substrates are prepared by ultrasonically cleaning glass slides with 2-propanol for 30 min. These clean slides are dried and coated with PDMS 10:1 (Sygard 184, Dow Corning) and cured in a furnace for 6 h at 90 °C. Each slide is attached to the platform of an electrodynamic shaker (TMS Inc., model- K2075E040). For each experimental run involving flow visualization, a single droplet of de-ionized water seeded with polystyrene spheres of 1 μm diameter (Alfa Aeser) at concentrations of 5×10^{-4} to 25×10^{-4} wt.% is deployed onto the PDMS coated slide. The volume deployed is 5.4 ± 0.4 μl. The equatorial diameter and the initial static contact angle are found to be 2.4 ± 0.04 mm and 110 ± 1°. A transparent enclosure is positioned over the top of the platform housing the substrate to shield against convection effects. A small gap is maintained at the bottom of the enclosure to ensure outside ambient conditions are also maintained inside the housing. The temperature and relative humidity inside the enclosure are found to vary in the ranges 27 ± 1 °C and 40.5 ± 1.5% respectively (monitored using a Thorlabs temperature and humidity sensor (TSP01)). For each run, the driving frequency (f) and amplitude of the shaker is kept constant. The frequencies chosen are $f = 120$ Hz and 400–800 Hz in steps of 100 Hz. The amplitude of the shaker/substrate is maintained in the range 12 ± 1.5 μm across all the runs. A CW laser (Cobolt Samba™ 300 mW 532 nm) coupled with a combination of spherical and cylindrical lenses is used to produce a vertical laser sheet of thickness around 170 μm. This laser sheet is made to pass through the vertical mid-plane of the droplet. The visualization plane is imaged using a Photron FASTCAM SA5 camera fitted with a Navitar zoom lens. Low frame rate (50 fps) is chosen to capture both the superposed oscillatory motion (particle streaks) and time averaged streaming flows. Separate videos of 300 frames each have been captured at intervals of 1 min to cover the entire evaporation process. 2–3 runs for each experimental setting have been performed to ensure repeatability in the observed flows. For comparison, backlit images of pure (de-ionized) water droplets of same volume are captured to quantify the shape oscillations as described in Sanyal and Basu (2016). The pure water droplet exhibit same contact angle as the micro-particle laden droplet.

3. Results

The interface oscillations are stationary capillary waves forming nodes and antinodes (Fig. 1) along the droplet surface. For axisymmetric oscillations about z axis (Fig. 1), the nodes lie along latitudes or circles. These nodal circles are denoted as solid red lines in Fig. 1. The contact line does not oscillate at any time during droplet evaporation. Thus it naturally forms a nodal circle. The oscillations are generated along the interface from the contact line due to the vertical motion of the substrate. The crest of the generated wave (GW) can be visualized as an annular lobe travelling across the droplet interface and closing in on itself at the apex. The wave is then reflected in the opposite direction (reflected wave RW) and interferes with the GW leading to the formation of a stationary wave