Abstract

A sessile droplet forms a fundamental building block in analyzing particle distribution and deposition in both industrial applications and lab on a chip research such as inkjet printing, surface patterning, photonic crystals, microarray deposition of DNA, mixing of biological fluids and many others. Thus, study of the fundamental dynamics of sessile droplets of both pure and particle laden (complex) fluid becomes important. Dynamics of such droplets can broadly be classified into evaporation and interface oscillations. Asessile droplet is bounded on one side by a material support. The presence of the support introduces the three-phase contact line (solid/liquid/gas) at the droplet edge. This contact line brings about interesting dynamics in both evaporation and oscillations of a sessile droplet. Evaporation is more basic of the two dynamics in sessile droplets. So, in the simplest case the contact line behavior and evaporation govern the droplet shape evolution and internal flow. In complex fluid droplets the particle deposition is thus dependent on both. For this reason, the body of work in this thesis starts off with the particle deposition in complex fluid sessile droplets induced by natural evaporation and contact line dynamics.

From there oscillations are introduced on the drying of such droplets. Different mode shapes are observed at different oscillation frequencies. It has been observed that for the stationary droplets and for droplets whose oscillations are initiated at resonance of the lowest allowable oscillation mode, the structures are similar having larger spread over height, while for higher frequencies the structures are dome-like with more uniform outer dimensions. The structures are investigated using experimental techniques such as high-speed imaging of droplet oscillations, internal flow visualization and SEM imaging. The possible reasons behind the contrast in particle deposit morphologies at higher and lower frequencies include the higher oscillation modes which cause segregation of the internal flow (observed through flow visualizations) and sectorial particle distribution. The overall particle content is homogenized throughout the droplet. Now an interesting feature is observed as the oscillating droplet evaporates that is the oscillation mode changes. The higher frequencies have a cascade of progressively lower modes showing up while the lower frequencies only have the lowest

allowable mode. Since the internal flow and particle mixing is primarily dependent on the mode order higher degree of mixing occurs for the cases of higher frequencies (even though lower modes appear towards the end of evaporation). Naturally theseinteresting inter-dynamics between evaporation and interface oscillations (oscillation mode transition) is taken up as the next line of study.

Being a purely fluid dynamical problem, such inter-dynamics are studied in a pure fluid droplet to simplify the theoretical analysis. As stated earlier a sessile droplet excited at a constant frequency exhibits a temporal sequence of interface modes when allowed to evaporate. Evaporation tunes the droplet to resonate at different modes in a descending order. The life of each mode spans over a transition phase from the onset of its own resonance to that of the next lower order mode. Mode lifetime during the evaporation period of any droplet has been found to decrease in a power law fashion both theoretically and experimentally when higher driving frequencies are considered. Such dynamics are driven by both evaporation and interface oscillations, the latter being governed by the dispersion relation of one dimensional capillary stationary wave. The natural excitation of sequential mode resonances in the present case is governed by a tuning parameter. Variation of this parameter by evaporation is analogous to external frequency sweep in non-evaporating droplets to detect similar mode resonances. The dynamics of transition phase corresponding to each mode is characterized by a second parameter denoted as mode transition parameter. The transition phase (higher to lower order) is initially fast but slows down close to the resonance of lower order mode. Theoretical expressions of mode lifetimes and the tuning parameter have been derived based on reasonably valid assumption of constant static contact angle (a measure of the contact angle of the unperturbed hypothetical droplet shape in an oscillation cycle). An approximate linear relation has been established between detuning (complement of the tuning parameter) and mode transition parameters which governs the dynamics of the transition phase. The experimental data corresponding to the detuning and mode transition parameters show universal merging for all excitation frequencies. The experimental data also follows the proposed theoretical linear trend but deviates towards the later part of the transition phase. Such deviations have been attributed to neglecting higher order terms and possible viscous damping from boundary layer at the substrate. Even then the universality of mode transition across all frequencies is maintained throughout the evaporative lifetime.

Flow visualizations in the previous study of complex fluid oscillating sessile droplets revealed features which evolved with evaporation similar to oscillation mode transition. Since oscillation generated internal flow is instrumental in governing particle mixing, study of the flow features and their evolution forms the subsequent line of study. 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. As stated previously 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 to provide further insights regarding manipulation of nano-particle deposition patterns in colloidal droplets using controlled oscillations.