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Nanoparticle Patterning in a Microfluidic Drop Induced by Surface Acoustic Waves

Leslie Y. Yeo, James R. Friend and Haiyan Li
Micro/Nanophysics Research Laboratory
Department of Mechanical & Aerospace Engineering
Monash University
Clayton, VIC 3800, Australia
Email: leslie.yeo@eng.monash.edu.au

***Abstract*—We demonstrate complex nonlinear pattern formation dynamics associated with standing wave vibrations induced along a piezoelectric substrate on which a small drop of colloidal suspension is placed. Interfacial colloidal islands which self-assemble due to surface acceleration and capillarity are subsequently erased once fluid streaming becomes significant at higher powers. Due to a peculiar instability, the system cycles between colloidal island assembly when streaming ceases and erasure when streaming resumes.**

I. Introduction

Microfluidics offers the potential for facilitating the self-assembly of sub-micron colloidal particles for the patterning of nanostructured materials or for cell sorting [1]. We describe a peculiar phenomenon of colloidal island self-assembly and erasure at the interface of a sub-μl drop comprising a suspension of 500 nm fluorescent particles placed atop a piezoelectric substrate along which a surface acoustic wave (SAW), which is a 10 nm high electroacoustic analogue of an earthquake wave [3], is induced (Fig. 1(a)) [2]. Details on how the SAW is generated and how it leads to the generation of bulk liquid recirculation (acoustic streaming) within the drop are reported elsewhere [4], [5], [6]. Here, it is suffice to mention that the SAW wavelength is λ = 196 μm giving rise to a corresponding resonant frequency of approximately 20 MHz.

II. Linear Concentric Ring Assemblies

Various colloidal patterns are observed at the drop interface depending on the initial drop size and the input power, as shown in Fig. 1(b) [2]. Initially, the colloidal particles assemble into linear concentric rings as shown by the fingerprint-like patterns in Regime A. Curiously, the separation between the rings is roughly 100 μm, corresponding to $\lambda/2$. A frequency scan using Laser Doppler Vibrometry (LDV) indicates that the drop free surface is vibrating at the same frequency (20 MHz) as the SAW vibration induced on the surface of the substrate beneath it, albeit at lower amplitudes. It is thus conceivable that the linear colloidal ring-like assemblies coincide with the nodal lines of these low amplitude 20 MHz standing wave vibrations induced along the drop interface.

A possible mechanism by which the particles drift to form these patterns is due to the capillary force acting on the particles in concert with the surface acceleration of the standing wave vibration, first proposed by Falkovich *et al.* [7] to elucidate the well-known observation that hydrophillic particles assemble onto the nodal lines of a standing wave induced along a liquid free surface whereas hydrophobic particles assemble onto the antinodal lines. Consistent with [7], we observe the inverse particle cluster time to scale with the square of the amplitude of the standing wave (Fig. 1(c)), thus inspiring confidence that the particle drift mechanism above provides a plausible reason for why the particles assemble at the nodes/antinodes of the interfacial vibration.

III. Pointwise Colloidal Island Assemblies

Upon increasing the input power and hence traversing into Regime B, the colloidal particles in the linear ring-like assemblies are observed to cluster to form point-wise colloidal islands, as illustrated in Fig. 1(b). We note that this regime is associated with an increase in the magnitude of the interfacial vibration. A repeat of the LDV frequency sweeps revealed that in addition to the small amplitude 20 MHz standing wave vibrations at the drop interface, large amplitude 1 kHz order vibrations were also significant in this regime; these large amplitude low frequency vibrations are associated with the capillary-viscous resonance of the drop.

The colloidal islands therefore appear to form at the intersection between the nodal lines of the low amplitude 20 MHz standing wave vibration and the circular nodal ring of the large amplitude 1 kHz vibration associated with the capillary-viscous resonance of the drop (Fig. 1(d)). As in Regime A, the inverse of the time it takes for the particles to cluster into these point-wise assemblies again scales with the square of the amplitude of the vibration, therefore suggesting the same particle drift mechanism as before.

The number and position of the colloidal island assemblies depends on the size of the drop. If the input power is kept constant and the drop is allowed to evaporate, the number of islands decreases successively, as seen in Fig. 2. This peculiar phenomenon can be explained with reference to the Rayleigh-Lamb dispersion relationship. As the drop evaporates and hence its size decreases, the capillary-viscous resonant frequency at which the drop vibrates increases, resulting in a decrease in the corresponding wavelength. Consequently, the size of the circular nodal ring shrinks. If the wavelength of

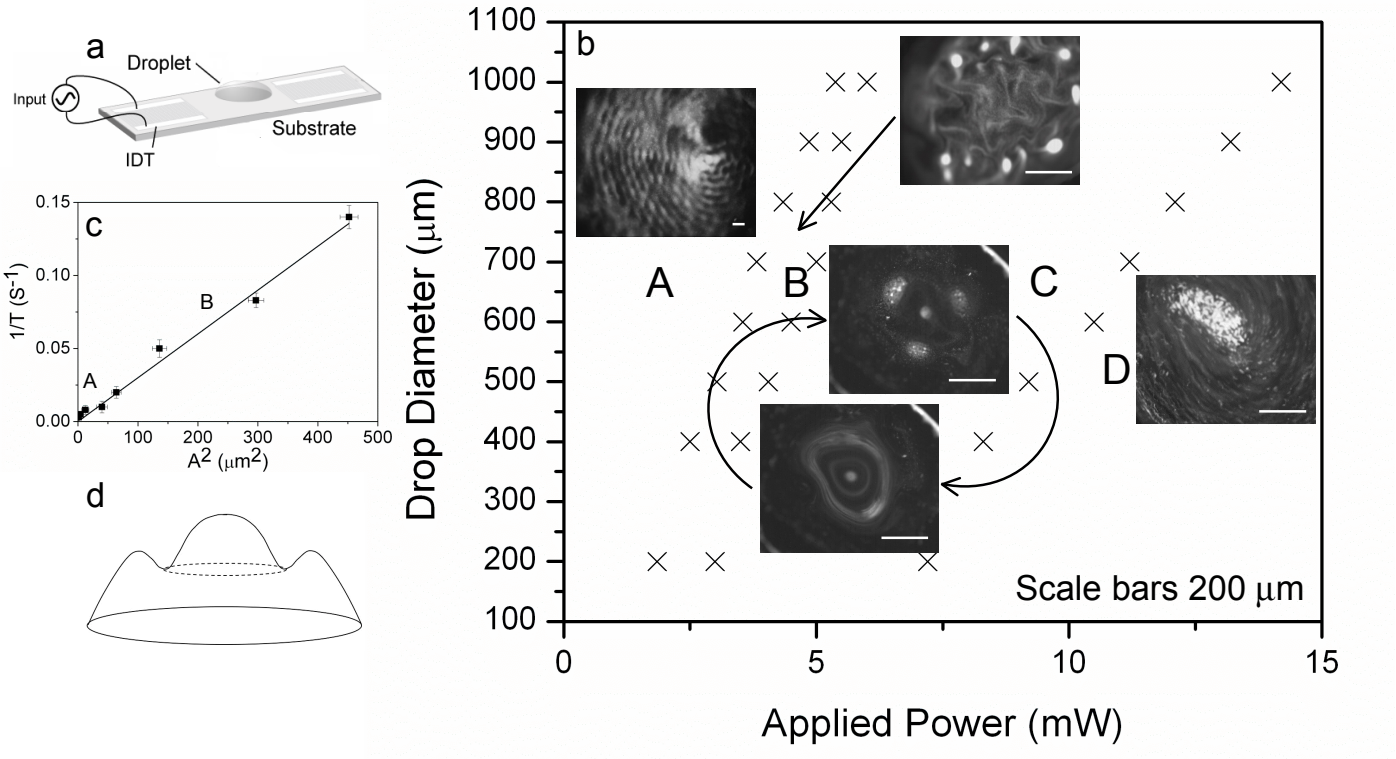

Fig. 1. Colloidal patterning on the free surface of a microfluidic drop [2]. (a) Schematic depiction of the experimental setup. (b) Interfacial colloidal patterns as a function of the initial drop diameter and input power. (c) Inverse particle clustering time as a function of the square of the amplitude of the standing wave. (d) Circular nodal ring arising from large amplitude vibration of the drop interface at the capillary-viscous resonant frequency.

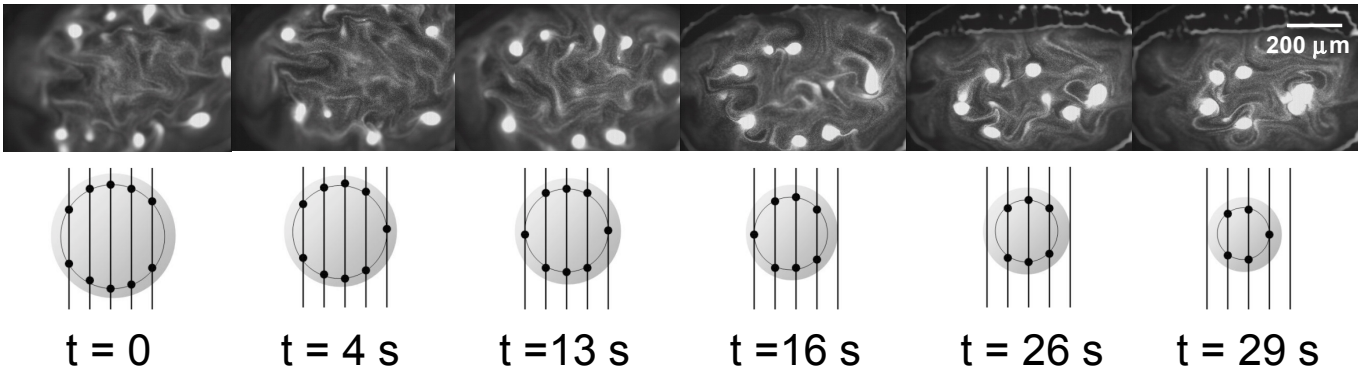

Fig. 2. The number of colloidal islands formed depends on the drop size which decreases due to evaporation in time [2]. The bottom schematic shows the successive decrease in the number of intersection points between the 20 MHz nodal lines and the 1 kHz circular nodal ring as the drop and hence the nodal ring shrinks.

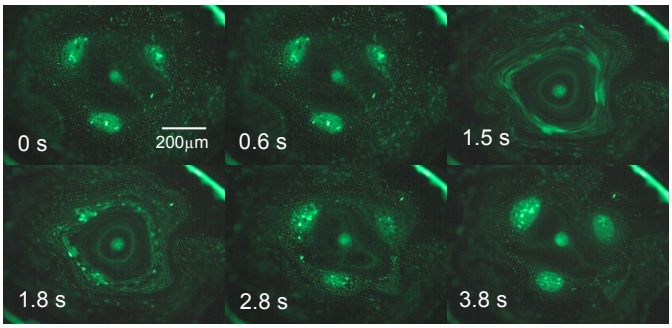

Fig. 3. Metastable transient state in which the system cycles randomly between colloidal island formation when there is no streaming and colloidal island erasure when streaming commences [2].

the low amplitude 20 MHz interfacial vibration and hence the separation between the linear nodal lines is assumed constant, then an intersection point (and hence a colloidal island) is sequentially lost as the nodal ring decreases in size.

IV. Cyclic Erasure and Reformation of the Colloidal Island Assemblies

Further increases in the input power into Regime C leads to the onset of significant fluid streaming within the drop. When streaming commences, the particles are dispersed and hence the colloidal island assemblies are destroyed (Fig. 1(b)). However, after a short transient, the streaming ceases and the colloidal islands are observed to reform until the streaming recommences and erases them again, as depicted in Fig. 3. This cyclic phenomenon occurs aperiodically and the direction of the streaming (clockwise/anticlockwise) is noted to be reasonably random, suggesting that this regime is a transient metastable state and that the commencement and cessation of the streaming is triggered by a peculiar instability arising from the highly nonlinear coupling between the acoustic, hydrodynamic and capillary forces. As the input power is increased (Regime D), however, the streaming becomes stronger and more consistent, leading to permanent dispersion of the particles (Fig. 1(b)). Once this occurs, the interfacial colloidal patterns are no longer evident.

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