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Droplet behaviour under high intensity acoustic vibration

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ABSTRACT

The reduction in scale of fluidic based chemical and biological processes offers significant analytical and sensitivity improvements as well as reduced reagent usage, increased automation and reduced manufacture costs. Droplets deposited on a planar surface offer a convenient way of investigating very small sample sizes. We investigate the effect of vibration of droplets in the direction normal to the surface on which they sit. When the contact line of the droplet is constrained by use of a very shallow well and suitable frequencies of vibration (order 100s Hz) are selected such that a resonant standing surface wave is established, collection of particles in predictable patterns can be achieved. When the droplet contact line is unconstrained high amplitude acoustic vibration (again order of 100s Hz) causes spreading of the droplet to occur. This effect can be so pronounced that during actuation the contact angle falls below that of the receding angle. We demonstrate the use of this effect by the merging of two droplets which are deposited a small distance away from each other. Once merged, a process which occurs due to surface energy minimisation as soon as the droplets spread such that they touch at one location, further vibration causes rapid mixing of the fluids through acoustic streaming.

INTRODUCTION

Interest in "lab-on-a-chip" or micro total analysis systems (μTAS) [1] involves a reduction in scale of fluidic based chemical and biological processes this offers significant analytical and sensitivity improvements as well as reduced reagent usage, increased automation and reduced manufacture costs [2]. An issue arising from the use of enclosed fluidic channels, which constitute the majority of micro fluidic systems, is that whether made by etching in silicon and sealed with glass, hot embossed in plastics, or moulding in PDMS, they must be filled if the samples need to be handled as otherwise pumping mechanisms fail [3]. When attention is turned to very small volumes of sample, this requires separation in an immiscible buffer solution [4], or the switching from using enclosed volumes to the use of droplets deposited on plane surfaces [5]. In this switch, some of the key technological building blocks required remain the same including the manipulation of suspended matter, sample mixing and fluid motion (whether pumping or droplet movement).

Whilst the use of ultrasound has become relatively widespread in micro fluidics, performing mixing [6], pumping and particle manipulation [7], there is less work applied to this field on lower frequency excitation (here used to refer to 100s Hz range). Low frequency fluid oscillations [8] and direct vibration [9] have been shown to cause fluid mixing in enclosed micro fluidic channels. Droplets have been moved on flat surfaces by asymmetric lateral vibration [10, 11], up an inclined surface with vertical vibration [12] and across a surface by a combination of both [13]. The mechanisms behind droplet movement relate to the contact angle and the associated hysteresis which occurs due to the difference in advancing and receding angles which occur on real surfaces and result from energy related to the contact line. When a puddle (a larger volume than a droplet, in which gravity plays a significant role in the shape of the fluid interface) is vibrated vertically, regardless of if it has been deposited such that the initial contact angle is advancing or receding, the resulting angle will be that at which the surface tensions balance, the hysteresis having been effectively removed, this arises as the contact line de-pins under vibration so negating the effect of the contact line energy [14]. When examining droplets, vibration at resonant frequencies cause well described surface waves [15].

In this work we demonstrate the use of low frequency vibration in the key microfluidic tasks of particle manipulation[16] and fluid mixing. The former is achieved when the droplet is constrained by a shallow well, the latter involves two droplets deposited next to each other, first being caused to spread, and then once contacting to mix rapidly.

EXPERIMENTAL SET-UP

The experimental apparatus, depicted in Fig. 1, consists of a droplet on a glass slide. The slide was arranged horizontally and vibrated in the vertical direction by an electromagnetic shaker (LDS, model V201) driven by a signal generator (Stanford Research SDR 345) via a power amplifier (LDS, model PA 25E). Low speed video image recordings were made using a CCD camera (Hitachi, KP-D20AU) coupled with a magnification lens (InfiniVar Video Microscope, Infinity Photo-Optical Company). The images were recorded at 25 frames per second directly onto a standard DVD recorder. Images obtained by playback from the DVD were transferred to a PC via a frame grabber driven by imaging software (Alliance Vision, Vision Stage). High speed video recordings were made using a camera (Fastec Imaging, Troubleshooter TS1000ME) at 500 frames per second. These were registered directly onto the internal hard drive of the camera, producing an uncompressed video file, which was then transferred to the PC by splitting into 250 individual images. In both cases lighting was provide by a cold source gooseneck lamp (Olympus, LG-PS2).

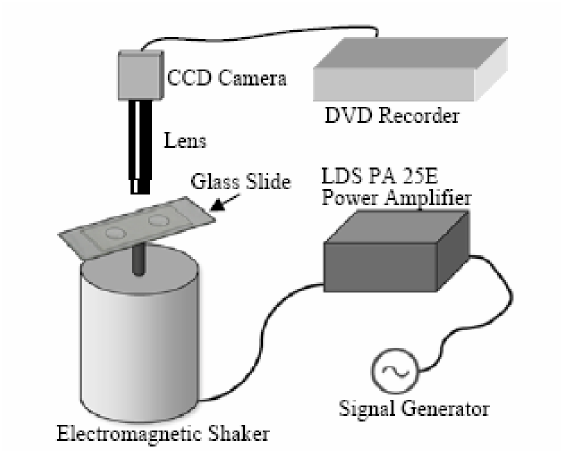

Figure 1. A depiction of the key apparatus used

PARTICLE MANIPULATION

A 50 µl deposited within a shallow (250 µm deep) well on a glass slide has been used to demonstrate particle manipulation possibilities using vibration frequencies in the range of 100 Hz. The well is used to contain the droplet under vibration to counter effects such as translation [12] or spreading (as will be seen later).

The nearest comparison to the use of acoustic vibration for manipulation is the use of ultrasound. At frequencies typically in the hundreds of kHz range, two main mechanisms are induced. These are acoustic radiation forces and acoustic streaming, both of which arise from non-linear momentum terms in the Navier-Stokes Equation. The acoustic radiation force acts directly on the particle, it can be calculated by integrating of the momentum terms around the surface of the sphere, and tends to cause particle which are stiffer and denser tan the surrounding medium to congregate in pressure nodes in a resonant ultrasonic field. Acoustic streaming arises from a body force which acts on the fluid, again this body force is due to the momentum terms, and the result is a swirling pattern of fluid motion. When particles are present this fluid motion induces drag forces on the particles. Hence there is an interplay between the two effects, the one seeking to bring particles to certain fixed (nodal) locations, the other seeking to sweep particles along streamlines. The balance of the result of the ensuing motion lies in the relative amplitudes of the forces, the acoustic radiation force is proportional to the radius of the particle cubed, whilst the drag force is linearly related to radius, hence for larger particle the former is dominant, whilst below a critical level the latter prevails. These ultrasonic effects will be compared and contrasted to what is seen for acoustic vibration.

Droplets of deionised (DI) water, containing suspensions of 10-30, 42, 60 and 116 µm glass microsphere particles (Whitehouse Scientific) were actuated at two frequencies. The frequencies, 60.59 and 111 Hz, were chosen such that resonance of the droplet would occur, that is the surface waves become standing waves. A depiction of the droplet and a typical resonant modal shape are shown in Fig. 2, with the particles in different clusters depicted in different colours.

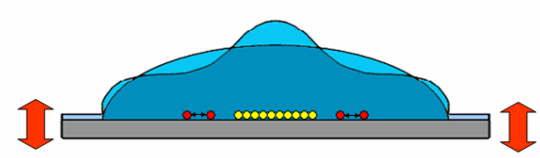

Figure 2. A depiction of the modal shape of a vibrating droplet with particle collected on the solid surface [16]

Of the particle sizes tested, it was found that larger particles (42 µm upwards) form regular ring patterns at the solid/fluid interface (Fig. 3b-d). The locations of these rings corresponded with the locations of the surface waves anti-nodes in the resonating droplet. The smaller particles (a mixture of 10 to 30 µm) however do not settle into such rings, but rather follow the swirling motion of the fluid, as depicted by arrows in Fig. 3a. This demonstrates that the dominant mechanisms at play are particle size sensitive, just as is the case for ultrasonic actuation.

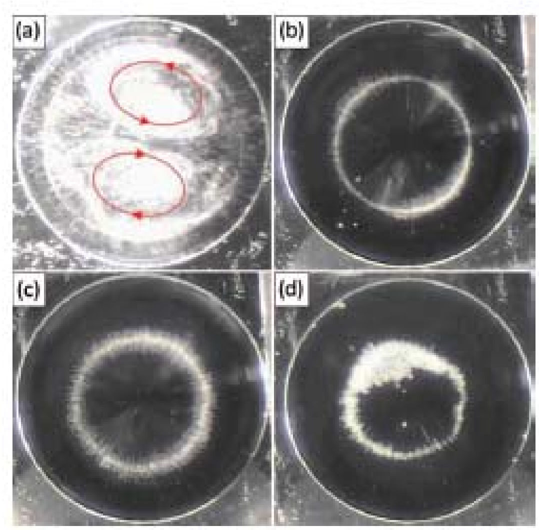

Figure 3. Droplets containing suspensions of (a) 10-30, (b) 42, (c) 60 and (d) 116 µm particles undergoing actuation at 60.59 Hz [16]

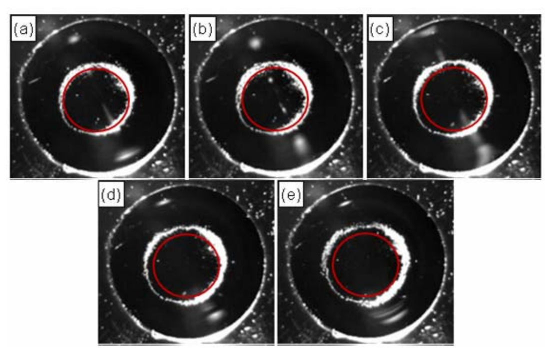

Figure 4. Images taken over half a cycle of 42 µm particles undergoing actuation at 60.59 Hz, as can be seen by comparison from the red circle, the radius of the ring of particles increases across the series of images [16]

It is clear from Fig. 3a that acoustic streaming occurs in the droplet, hence the swirling pattern followed by the particles. The larger particles however react to forces which tend to bring them to a certain location. In stark contrast to the ultrasonic case this location moves over the cycle of the vibration, this can be detected from images Fig 3 b and c where the particles appear elongated, this blurring occurs as the shutter speed of the camera is considerable lower than the actuation frequency. Fig 4 shows the movement of 42 µm the particles over the duration of half a cycle, using high speed photography. It can be seen that the radius of the ring formed increases over the course of the five images. So rather than a time averaged effect as is the case for acoustic radiation forces, this low frequency induced force field is altering during the course of each cycle. It is possible this arises due to a form of hydrodynamic focusing, in which the particles move in response to each flow arising from each cycle, and each time are progressively moved towards their end location, a similar effect has been predicted mathematically for an enclosed channel [17].

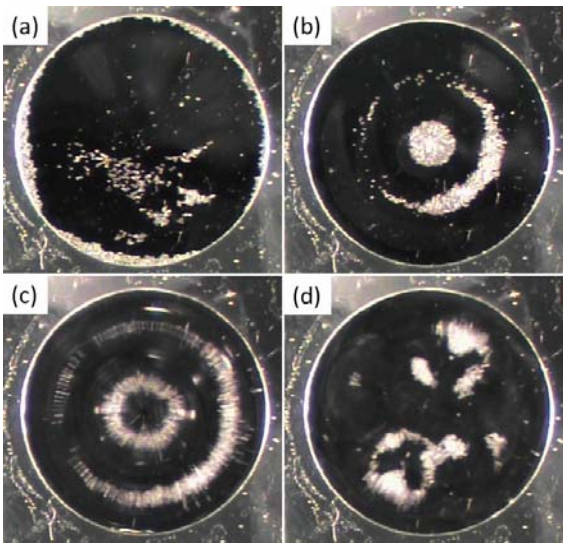

Figure 5. The collection of 116 µm particles under different amplitudes of oscillation: (a) 0 amplitude, (b) 360 µm, (c) 440 µm and (d) 500 µm (peak to peak) [16]

In many resonant wave manipulation methodologies, such as ultrasonic standing waves, the amplitude of oscillation is related to the forces generated and not to the patterns formed by the particles. However, in the scheme of applying low frequency vibration to a droplet, here we see a change in the pattern of particles with increasing amplitude, as shown in Fig 5. The free surface of the droplet can be expected to move significantly at higher amplitudes, causing two effects. Firstly, we believe that the motion of the upper liquid-gas interface at the centre of the droplet can be so great at high amplitudes that the minima of the cycle touches the glass slide, causing the central disk (Fig 4b) of particles to disperse into a ring (Fig 4c). Secondly, as the fluid surface is oscillating about a sloped surface at the outer rings, the amplitude tends to increase the radius of the ring formed as the fluid media translates. At even higher amplitudes (Fig 4d) no predictable patterns are formed as a spatiotemporal regime occurs. This final disintegration of the modal shape at high amplitude is in line with studies on sessile droplets. Finally, when the amplitude is reduced to zero, a useful feature with the approach lies with the propensity of the particles to remain at their collected positions. This is attributed largely to the fact that the collection process was developed at the solid-liquid interface, lending further evidence that this is where accumulation took place.

At moderately high amplitude Fig 5c the central disk becomes a ring, as stated previously this is believed to be due to the proximity of the central antinode to the glass surface at its' lowest point in the cycle. Further evidence for this is provided by examining the vibration of a fluid volume contained in a square container (30 x 30mm), with a much higher depth than the droplet. Fig 6. shows that at the surface of such a fluid volume, floating (42 µm) particles are collected together in clusters [18], whilst the particles at the lower surface of the chamber are randomly distributed (seen as a milky cloud).

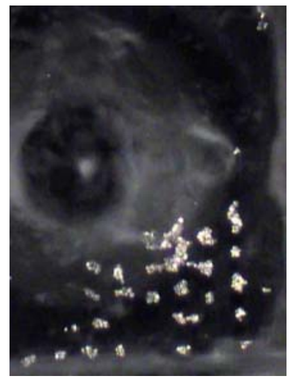

Figure 6. The collection of 42 µm particles floating on the surface of a fluid volume held in a square chamber, whilst the suspended particles lie unaligned on the lower surface of the chamber. Indicating the alignment achieved in the droplets results due close proximity with the surface waves.

DROPLET SPREADING

Particle manipulation has been demonstrated in constrained droplets. Now we examine the effect of low frequency vibration on unconstrained droplets, that is droplet which are placed on nominally flat surfaces, the shape of the droplet then being defined by the contact angle and line energy of the contact line. The former is the angle at which the air fluid interface meets the solid surface, it can be found by way of a force balance between the various surface tensions (fluid/gas, fluid/solid, and gas/solid). However, the actual angle usually takes a range of values from the lowest, the receding angle, to the highest the advancing angle. This range can be linked to the energy required to move the contact line, this being the line energy. In other words the force balance must be overstepped by a certain amount so that the contact line actually moves, whether receding as fluid is extracted, or advancing when fluid is added.

When excited by low frequency vibration, droplets are found to spread; hence, a change is affected in the energy balance defining the location of the contact line. To investigate this further, a droplet was placed on the surface and subjected to varied vibration frequency and amplitude. When placed under vibration, droplets spread to a maximum radius for the given excitation. Of course, the nature of the spread is subject to some stochastic effects, as the contact line motion is related to the degree of localised roughness.

A droplet of constant volume, 25 µl, was subjected to a frequency of 200Hz under a range of amplitudes; 0.08mm to 0.14mm.

Under excitation of 200Hz with an amplitude of 0.08mm, the droplet expanded approximately radially, once actuation ceased no retraction occurred. The result of the force imbalance created due to the excitation was for the three phase boundary interface to shift outward, resulting in a larger wetted area radius and so a smaller contact angle. The fact that the droplet did not retract after excitation ceased, indicates that the resulting angle after spreading still lies above the receding angle, and hence the spread droplet is stable. The before and after states can be seen in Fig 7.

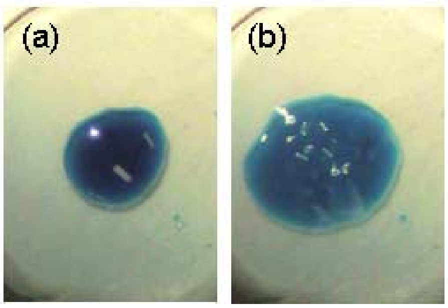

Figure 7. Completely stable spreading of a 25microlitre droplet. Part a) before vibration is applied. Part b) shows the droplet after it has finished spreading due to vibration excitation of 200Hz and 0.08mm. In b) the actuation is still occurring, as can be seen by the surface waves on the droplet.

It is clear that due to the change of size and shape of the droplet that the resonant frequency of the droplet will alter during actuation. As such the initial frequency was not selected to coincide with a resonant value for the initial droplet shape.

Under higher values of excitation, the spreading becomes more random in nature, and the resulting contact line can not be considered as being circular. An example of this is, shown in Fig 8, in which a 20 µl droplet has been vibrated at 200 Hz, with an amplitude of 0.12mm. Again, at this value of amplitude, after actuation ceased the droplet did not retract, showing that the contact line energy can not be overcome by the force balance, which implies the receding angle has not be passed.

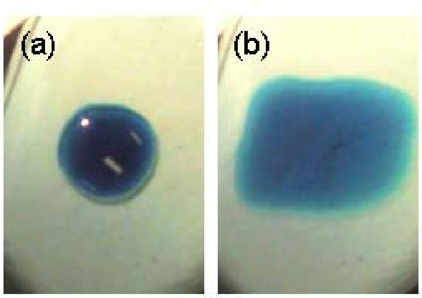

Figure 8. Non-uniform spreading of a 25microlitre droplet, a) before vibration is applied, and b) after the droplet after it has finished spreading due to vibration excitation of 200Hz and 0.08mm. In b) the actuation has been turned off, hence the lack of surface waves.

As the amplitude of excitation is increased further the spreading of the droplet becomes rapid and extensive. Once the excitation is removed, the much of the fluid recedes to form more regular shapes. This indicates that during the spreading process it is possible to overstep the receding angle. This is shown in Fig 9 in which the same size droplet has been excited by a 200Hz vibration with an amplitude of 0.14mm.

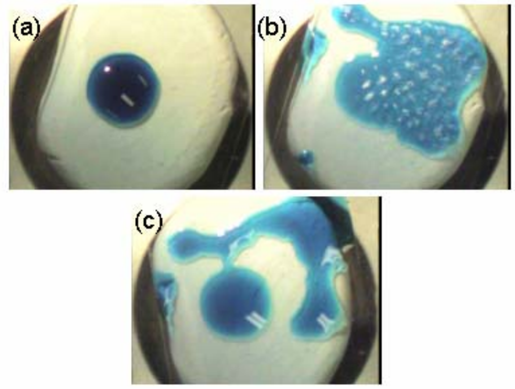

Figure 9. Extensive spreading of a 25microlitre droplet is shown, caused by actuation at 200 Hz at an amplitude of 0.14 mm. Image a) shows the droplet before applied vibration, b) during actuation, the large surface waves can be clearly seen, and c) once actuation has ceased, much of the fluid can be seen to retract to a uniform circular area, indicating that the receding angle had been exceeded, hence the spread droplet was no longer stable once excitation ceased.

The spreading of the droplet past the receding angle allows the formation of films which could not be achieved without vibration; this can be made stable if the film spreads over an area bounded by walls. Such applications are currently under investigation. Furthermore, the spreading can be used to cause adjacent droplets to merge, as will be shown in the following section.

DROPLET MIXING

In the work on particle manipulation it is clear that acoustic streaming plays a role, demonstrated by the inability to concentrate the smallest particles used, instead they followed to swirling streamlines so characteristic of acoustic streaming. Here we examine the use of low frequency excitation for mixing of droplets, however we do not use a resonant frequency, as when the two drops are combined this will in any case alter. To examine the degree of mixing between two droplets we first perform an experiment in which no actuation takes place and so mixing is mainly diffusive in nature. In this experiment, the results of which are shown in Fig 10, a droplet of blue dye is deposited onto a droplet of pure water which sits on a flat surface. It can be seen that 120s pass before the droplet is fully mixed, based on visual perception of the colour achieved. The initial swirl which is visible in Fig 10 (b) is due to the flow which arises when the two droplets merge, rather then any streaming, hence this swirl quickly dies out. Even after 120s the mixing can not be guaranteed, depending on the size of the initial swirl, the effect of which is to extend the interface between the two fluids, promoting diffusion.

In a second experiment, the water droplet deposited on the surface is excited by 200Hz actuation, whilst the dye droplet is added, in this scenario the steaming in the water droplet, causes rapid and complete mixing, as shown in Fig 11, over a period of a second. Whilst further work must be completed to quantify the degree and speed of mixing, this method clearly has significant potential. Furthermore, it was observed that mixing is enhanced by randomly changing the excitation frequency so further complicating the streaming patterns established.

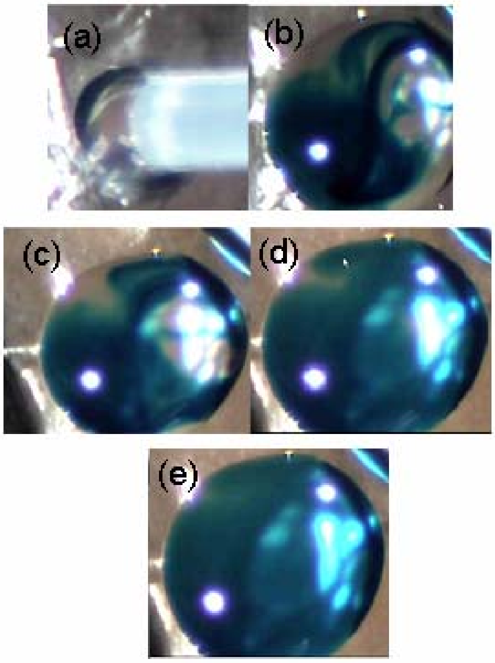

Figure 10 Droplet on Droplet mixing under no excitation. Fixing times a) t=0sec, b) t=.1sec, c) t=12sec, d) t=65sec, e) t = 120sec

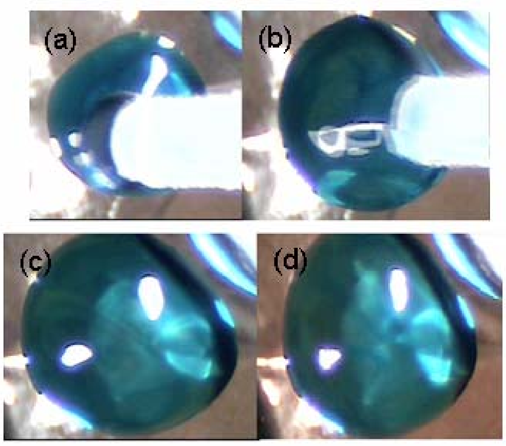

Figure 11 Droplet on Droplet mixing under low frequency vibration. Mixing times a) t=0sec, b) t=.1sec, c) t=1sec, d) t=20

Finally, by combining both the spreading and streaming effects, droplets which are close proximity can be first merged and then mixed, indeed if the vibration amplitude is high enough they can then be spread into a thin film. The advantage of this method over pipetting one droplet into another is that the fluid dispensing is more repeatable, splashing does not occur during dispensing and the mixing can be started at a time of choice.

In figures 12 a) to d), the water droplet shows clear signs of deformation and movement towards the blue dye droplet as a result of the applied vibration. The amplitude used to excite the droplets was 0.02mm. Figures 7 d) and e) show the very rapid development of a large contact surface area, consistent with the minimisation of surface energies. The continued vibration, Figures 7f) and g), causes streaming in the resultant shape resulting in near complete mixing 43 s after contact was made.

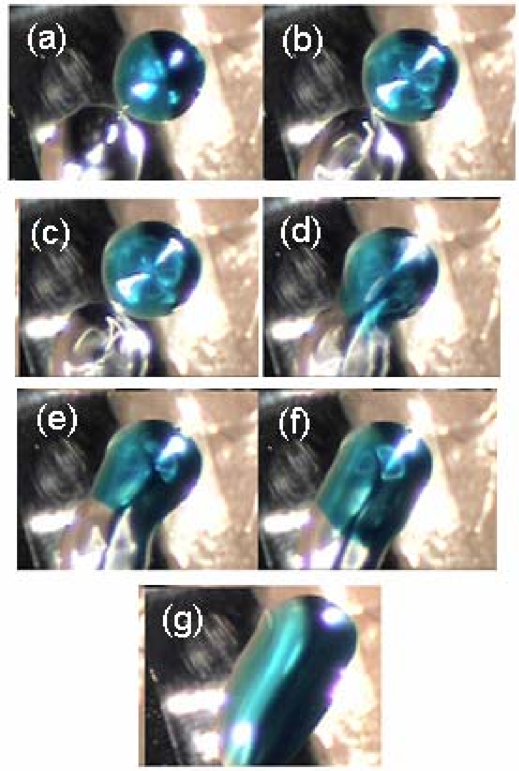

Figure 12 Droplet next to droplet, contact manipulated under variable low frequency vibration a) t=0sec, b) t=5sec, c) t=7sec, d) t=7.1sec, e) t=7.2sec, f) t = 10sec, g) t = 50sec

CONCLUSIONS

Low frequency actuation has been demonstrated to cause a range of effects on droplets. In constrained droplets suspended particles can be manipulated into rings or disks, with both frequency and amplitude playing a role in the geometry of the patterns formed. Unconstrained droplets can be spread extensively under vibration, even beyond the receding contact angle; this could find applications in creating thin films of fluids on surfaces which do not need to be highly wetting. Furthermore, mixing can be seen in droplets despite not using resonant frequencies. Indeed by first causing spreading then streaming, two droplets adjacent to each other on a plate can be merged and mixed.

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