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Holographic microscopy of micron-sized droplets at liquid-liquid interfaces

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Abstract

When a millimetre-scale or larger bubble or droplet reaches a fluid-fluid interface, and after the liquid film between it and the interface has drained, it should form a three-phase contact line and change shape according to the surface tension and the viscosity of the fluids. Studies have mainly looked at the behaviour of these large drops using conventional imaging methods such as bright-field video microscopy. These conventional methods, however, are not suitable for smaller, micrometre-sized droplets. We describe how by using digital holography microscopy, we are able to obtain high precision, three dimensional information about the position and behaviour of droplets and bubbles. We also show an example demonstrating how this method can be used to look at the dynamic behaviour of a droplet before and after reaching a fluid-fluid interface.

Introduction

The interaction of a droplet or bubble with a fluid interface is important to understanding natural phenomena and improving the performance of biomedical and industrial applications [1, 2]. In particular, the dynamics of a micrometre-scale bubble or droplet upon reaching an interface are not well understood. In some cases, bubbles have been observed to “bounce” at the interface [3-7], and models of the bouncing have been developed [4, 8]. Other studies have examined the interaction and formation of daughter bubbles from bubbles that burst at interfaces [9, 10]. Some smaller bubbles were shown to be trapped when crossing a horizontal interface or to show transitions in shape from spherical to toroidal [11].

For droplets, the spreading behaviour at the interface depends on the surface tensions. After the liquid film between the drop and the interface drains, the contact line is formed between the three fluids, and the droplet behaviour is governed by the viscosities and surface tensions of the three fluids [12]. Studies examining this phenomenon have used fast speed cameras to directly look at the behaviour of large millimetric drops. However, the interaction and spreading of a smaller droplet might differ due to less dominant gravitational forces. Studying such behaviour with conventional imaging techniques is difficult. We demonstrate how digital holography can be used as a tool to precisely study the precise of small droplets and their interaction with the interface.

Experimental Method

We use digital holography, a fast three-dimensional imaging technique, to measure the position and shape of micrometre-scale droplets and bubbles. Light from a laser is collimated, so that it is approximately a plane wave incident on the sample. The scattered and undiffracted waves interfere to form a two-dimensional pattern, or hologram, which contains phase information that can be used to recover three-dimensional information about the droplet. Because the droplets are spherical before they breach the interface, we model their scattering using the

Lorenz-Mie solution to Maxwell's equations. We then numerically interfere the scattered light with a planar reference wave, and we fit the resulting calculated holograms to the measured ones to obtain the position of the droplet. This method allows us to recover the position of the particle with 10 nm precision on timescales of milliseconds [13, 14].

Experimental Setup

We generate droplets of mineral oil by agitating a mixture of mineral oil and water back and forth between two linked syringes. A range of polydisperse droplet sizes are formed, depending on the rate of agitation. These droplets are then diluted in deionised water and 10 µL of the droplet solution put into the bottom of custom made polyetheretherketone (PEEK) troughs [14]. This makes a water-air interface with a water phase containing the oil droplets. The troughs are placed on a Nikon inverted microscope [14]. To generate holograms, we use a 660 nm laser diode, coupled into a single mode fibre and collimated through the condenser. We also use a counter-propagating beam, generated by an 830 nm diode coupled into a single mode fibre and focused by the objective, as an optical trap. A 60x Nikon, 1.2 NA water immersion objective collects the light and focuses the trap beam, while a fast camera (Photon Focus MVD-1024E-160) records holograms at 50 frames per second. We use the optical trap to move the droplets within a few micrometres of the interface. We then release the droplets so that they can freely rise to the interface. We record background images with no droplets in the field of view, which we use to correct for scattering from dust and imperfections in the optical train.

We fit the background-divided holograms using an open-source Python-based library (HoloPy), available at https://launchpad.net/holopy. The best-fit refractive index, size, and centre-position for each frame are used as initial guesses for the next frame. Through this procedure we obtain the trajectory of the droplet before reaching the interface.

When the shape of the droplet deviates from a sphere, as for example when it breaches the interface, fitting the holograms to a sphere model is no longer appropriate. Instead, we numerically propagate light back through the hologram to an arbitrary axial distance [15]. This reveals the three-dimensional scattered field at that axial distance, which is related to the shape of the original scattering object. For instance, when a hologram of a point scatterer is reconstructed to the distance from which the hologram was originally taken, the reconstruction looks like the original point scatterer. Similarly, reconstructions of a spherical droplet hologram at different axial distances show a droplet coming into focus. We can therefore view reconstructions of the breached droplet at the axial distance where the pre-breach spherical droplet appeared in focus to glean information about the shape of the breached droplet at the interface.

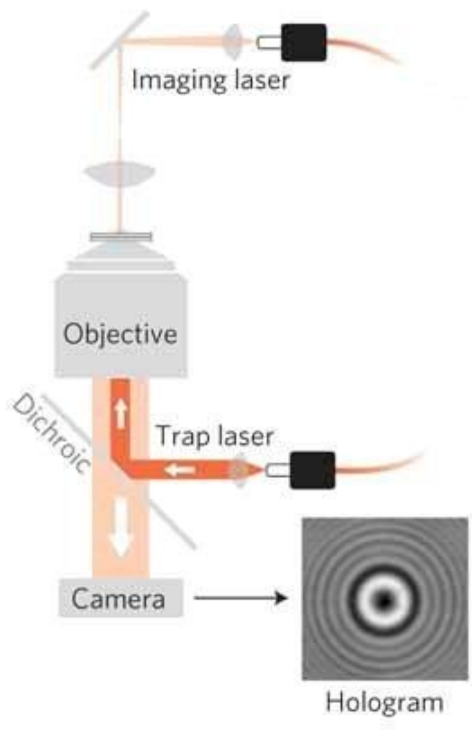


Figure 1. Experimental set up of the digital holography microscopy with the trap laser. [14]

Results

First we show that we are able to precisely fit holograms of mineral oil droplets in water to extract their 3D positions. From the fits we find the diameter of the droplet to be 3.2 µm, which agrees qualitatively with the size of the droplet from reconstruction. The fitted axial position of the droplet, shown in figure 2, shows that the droplet slows down as it nears the interface. This is consistent with enhanced hydrodynamic drag near the interface, which is associated with drainage of the film between the particle and the interface. A similar slowing of dynamics has been observed with solid particles [14]. This demonstrates the effectiveness of holography as a tracking tool.

The contrast and fringe spacing of the hologram then abruptly change within one frame (20 ms), as shown in figure 3. We associate this change with the droplet breaching the interface. To understand what is happening to the droplet, we numerically reconstruct the holograms using back-propagation (figure 4). The reconstructed images show that the droplet becomes a larger and hence flatter shape once it reaches the interface, in contrast to the smaller spherical droplet immediately beforehand.

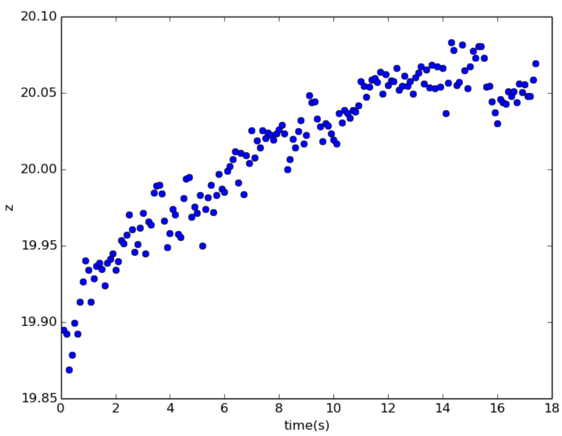
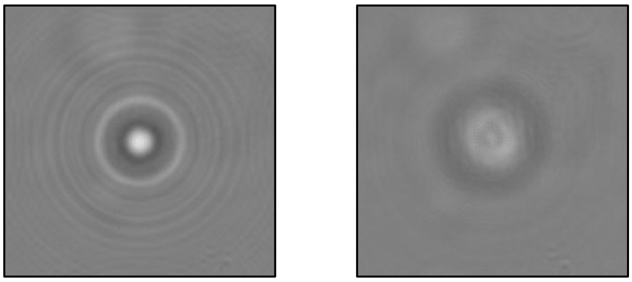
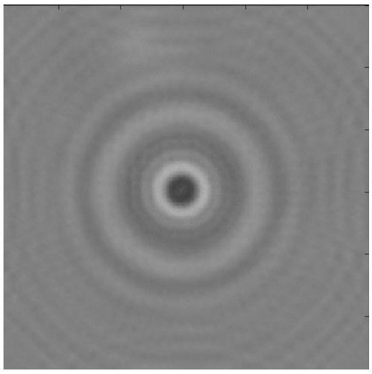


Figure 2. The height z (µm) of the droplet, as measured by fitting the holograms, just before the droplet reaches the interface

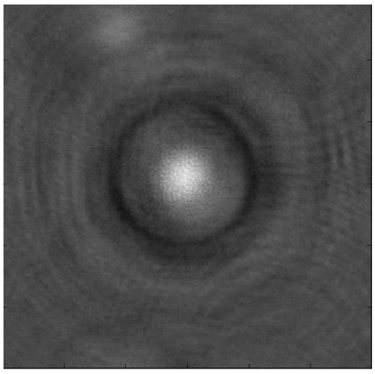


(a) (b)

Figure 3. Holograms of the droplet (a) before spreading in spherical shape (b) At one time frame (20 milliseconds) after



(a)



(b)

Figure 4. Reconstruction amplitude plot of the droplet to the focus plane (a) Before reaching the interface- Spherical shape (b) right after reaching the interface- flat film shape

From the reconstructions we are able to find the size of the final flat droplet. For this particular droplet the diameter of the final shape was found to be 14 µm in diameter.

We have shown that using the reconstruction and fitting tools in HoloPy we are able to obtain detailed information about the droplet size and position before and after reaching the interface. Because of the 3D nature of such phenomena we were not able to see such detail using bright field microscopy or other conventional methods. We are interested in the dynamic behaviour of the droplets before reaching the interface as well as the flat droplet shape spreading after it breaches the interface. In future studies, digital holography will allow us to look into these dynamics.

Conclusion

Digital holography enables the detailed study of the behaviour of small micrometre-sized fluid particles close to an air-water interface. In contrast to previous studies that used digital holography to investigate solid particles, our study suggests that the position, shape, and dynamics (such as the spreading behaviour) can all be extracted and investigated.

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