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Ultrafast/nanoscale dynamics studied by X-ray imaging

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Fluid/fluid flow is very commonly encountered in nature such as in rain drops, ink-jet printing, winds surrounding a hurricane or a tornado, whirlpools in the wake of boats, wetting, etc. The flow phenomena are important in understanding physical, chemical, and biological processes occurring at their interfaces. Despite recent advanced science and technology, the *visualization* of interface dynamics, which is an essential gateway to understanding the flow phenomena, is however a challenge because interface dynamics mostly happens in extreme conditions of ultrafast time domains ($\sim \mu s$) and/or nanoscale regimes in bulk systems.

Here we apply *phase contrast x-ray imaging* to visualize the *micro/nano and/or ultrafast* interface dynamics, especially, in '*drop impact*' and '*wetting*'. Phase contrast X-ray imaging in this study is mostly based on refraction enhanced phase contrast.

First, we discover that intense irradiation by hard X-rays decreases the effects of natural *surface tension* of water, as directly evidenced by phase contrast X-ray imaging [1]. A model based on ionization and surface charging explains this so far undetected phenomenon. This is an example of the largely unexplored effects that can be produced by extreme intense x-ray irradiation – an important issue due to current development of x-ray free-electron-lasers with unprecedented brilliance.

When a liquid drop impacts a solid surface, air is generally entrapped underneath. Using ultrafast phasecontrast x-ray imaging, we directly visualize the profile of an extracted air film and its evolution into a bubble during drop impact. We identify a complicated evolution process that consists of three stages: i) inertial retraction of the air film, ii) contraction of the top air surface into a bubble, and iii) pinch-off of a daughter droplet inside the bubble [2]. Energy transfer during retractin drives the contraction and pinch-off of a daughter droplet. The wettability of the solid surface affects the detachment of the bubble, suggesting a method for bubble elimination in many drop-impact applications.

A bubble reaching an air-liquid interface usually bursts and forms a liquid jet. Jetting is relevant to climate and health as it is a source of aerosol droplets from breaking waves. Jetting has been observed for large bubbles ($R >> 100\mu m$). However, few studies have been devoted to small bubbles ($R < 100\mu m$) despite the entrainment of a large number of such bubbles in sea water. Here we show that jet formation is inhibited by bubble size; a jet is not formed during bursting for bubbles smaller than a critical size [3]. Using ultrafast X-ray imaging, we build a phase diagram for jetting and the absence of jetting.

A vortex is a flow phenomenon that is very commonly observed in nature. Despite long studies more than a century, the origin of the vortices and their dynamics remain unclear, mostly due to the lack of appropriate visualization methods. With ultrafast X-ray phase-contrast imaging, we show that the formation of vortex rings originates from the energy transfer by capillary waves generated at the moment of the drop impact [4]. Interestingly, we find a row of vortex rings along the drop wall with different powerlaw dependencies of the angular velocities on the Reynolds number.

One of the most questionable issues in wetting is the force balance that includes the vertical component of liquid surface tension. On soft solids, the vertical component leads to a microscopic protrusion of the contact line, that is, a '*wetting ridge*'. The wetting principle determining the tip geometry of the ridge is at the heart of the issues over the past half century. Here we reveal a universal wetting principle from the ridge tips directly visualized with high spatio-temporal resolution of x-ray imaging. We find that the cusp of the ridge is bent with an asymmetric tip (Fig. 1), whose geometry is invariant during ridge growth or by surface softness [5]. The singular asymmetry is deduced by linking the macroscopic and microscopic contact angles to Young and Neuman laws, respectively.



Figure 1. High-resolution X-ray imaging of a wetting ridge (left) with a bent cusp (middle) and an asymmetric tip (dashed square in middle). Extraction of three interfaces from the ridge tip (right) enables us to measure the macroscopic (θ) and the microscopic (θ_s , θ_v , and θ_L) contact angles. Scale bars, 2 and 1 μm , respectively.

Ultrafast/nanoscale dynamics based on using phasecontrast X-ray imaging will significantly contribute to resolve various unsolved puzzling problems in nature.

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