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Nguyen, Dang Minh

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The Physics of Oscillate Boiling

Nguyen Dang Minh

SCHOOL OF PHYSICAL AND MATHEMATICAL
SCIENCES

2019

The Physics of Oscillate Boiling

Nguyen Dang Minh

SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES

A thesis submitted to the Nanyang Technological University
in partial fulfillment of the requirements for the degree of
Doctor of Philosophy

2019

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Assoc. Prof. Massimo Pica Ciamarra

Authorship Attribution Statement

The thesis contains materials from 2 papers published in the following peer-reviewed journals where I was the first and/or corresponding author.

Chapter 2 was published as [Li, F., Gonzalez-Avila, S. R., Nguyen, D. M., Ohl, C. D. \(2017\). Oscillate boiling from microheaters. *Physical Review Fluids*, 2\(1\), 014007.](#)

The contributions of the authors are as follows:

- Dr. Li Fenfang designed and setup the experiment, collected and analyzed the initial result, i.e. Fig. 1-2 of the main text, and wrote the first-half of the manuscript.
- Dr. Roberto Gonzalez-Avila designed, setup the experiment and helped review the manuscript.
- I conducted the numerical simulation, provided physical explanation and performed additional experiments, i.e. Fig. 3-5 of the main text. I wrote the second-half of the manuscript and the supplementary materials.
- Prof. Claus-Dieter Ohl supervised the project, provided physical insights of the phenomenon and revised the manuscript.

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The contributions of the authors are as follows:

- I designed the sample and the experimental setup, conducted the experiments, analyzed the results and wrote the manuscript.
- Dr. Hu Liangxing fabricated the samples and helped review the manuscript.
- Prof. Miao Jianmin gave advice on the sample design and supervised the fabrication process.

- Prof. Claus-Dieter Ohl supervised the project, provided guidance, contributed to the writing and revised the manuscript.

Chapter 4 was published as [Nguyen, D. M., Supponen, O., Miao, J., Farhat, M., Ohl, C. D. \(2019\). Gravity-Independent Oscillate Boiling. *Microgravity Science and Technology*, 1-7.](#)

The contributions of the authors are as follows:

- I designed and conducted the experiments, analyzed the results and wrote the manuscript.
- Dr. Outi Supponen contributed to the experimental setup, took care of safety, regulations and coordinated the campaign. Later she helped reviewed the manuscript.
- Prof. Miao Jianmin supervised the sample fabrication process.
- Prof. Mohammed Farhat initiated the project and wrote the proposal that enabled the team to join the parabolic flight campaign.
- Prof. Claus-Dieter Ohl collected parts of the data, supervised the project, and revised the manuscript.

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The contributions of the authors are as follows:

- I designed the sample and the experimental setup, conducted the experiments, analyzed the results, performed numerical simulation and wrote the manuscript.
- Dr. Muttikulangara Swaminathan Sanathanan fabricated the samples and helped review the manuscript.
- Prof. Miao Jianmin supervised the sample fabrication.

- Prof. David Fernandez Rivas gave advice on the sample design and reviewed the manuscript.
- Prof. Claus-Dieter Ohl supervised the project, provided guidance, contributed to the writing and revised the manuscript.

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Abstract

The boiling crisis is widely known as the greatest challenge that limits the working capacity of nucleate boiling. This phenomenon takes place when excessive heat supply creates a vapor film, separating the boiling liquid from the heating substrate. This results in the drop of heat transfer efficiency and a significant rise of the device's temperature. Our work introduces a new regime called oscillate boiling, in which by confining the heat source, the boiling bubble oscillates stably for millions of cycles instead of growing. This mechanism eliminates the possible formation of the vapor film and might be the solution for the boiling crisis.

The thesis begins with the first experimental observations where the phenomenon is characterized with high speed imaging, acoustic emission and thermal readings of the heating substrate. We then explain the oscillatory behavior of the boiling bubble with two mechanisms: the liquid jet created by the bubble's non-spherical collapse, and the thermal kick upon the jet's impact onto the heating substrate. The explanation is supported with numerical simulation on the bubble dynamics and the flow profile of the surrounding liquid.

The following works focus on bringing oscillate boiling towards application, which begins with moving from a laser-based heater to an electrical heater, which allows easier setup, higher energy efficiency and much better controls. Along this way we discover two different modes of oscillate boiling, namely unstable oscillate boiling, associated with low heating power, where the bubble oscillates shortly for only a few milliseconds and starts growing, and stable oscillate boiling, associated with high heating power, where the bubble oscillates stably with a fixed maximum radius.

Finally, we present two potential applications of oscillate boiling. Firstly, by performing experiments on parabolic flights, we show that unlike nucleate boiling

which depends heavily on gravity, oscillate boiling's functionality remains intact regardless of the gravity condition. Secondly, we study the interaction of two closely-placed oscillate boiling bubbles and discover that depending on their distance and heating power, the two bubbles can either quickly merge, oscillate independently, or most interestingly, synchronize. This results provide the design criteria to number-up the oscillate boiling phenomenon to commercializable scale.

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Nguyen Dang Minh, July 2019

“If you are good at something, never do it for free.”

—The Joker (The Dark Knight, 2008)

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Acronyms

CFD Computational Fluids Dynamics

CHF Critical Heat Flux

ODE Ordinary Differential Equation

Chapter 1

Introduction

1.1 The Boiling of Liquid and the Boiling Crisis

Boiling is a phase-transition process where liquid turns into vapor after receiving sufficient thermal energy. Then, vapor bubbles nucleate and grow on the heated surface, detach, and rise into the bulk liquid due to buoyancy. The large amount of thermal energy involved in this phase-change process allow boiling to transport heat very efficiently. As a result, it is widely exploited in many energy conversion systems. These include heating applications such as cooking [1], as a reactor blanket in nuclear power plant [2], or cooling applications for example in heat pipes for high-performance computer [3] and engines [4], etc.

The energy efficiency of a boiling system may be described by the boiling heat transfer rate as a function of the wall superheat, i.e. the heating substrate temperature minus the boiling point of liquid. The relationship between these two quantities results in the boiling curve, as shown in Fig. 1.1. At the beginning of the process, the substrate is heated up from room temperature and energy is transported from the substrate to the lower liquid by heat conduction, further away through convection. When the substrate reaches the liquid's boiling temperature (point A), nucleate boiling is initiated. In this boiling regime, individual vapor

FIGURE 1.1: Graph of the relationship between boiling heat transfer rate and the wall superheat when nucleate boiling transits into film boiling regime. This figure is adapted and colorized from Ref. [5] with permission

bubbles form at isolated nucleation sites along the surface. The bubbles grow until being departed into the bulk liquid due to buoyancy. New bubbles are then nucleated at the same nucleation site and the cycle repeats. Nucleate boiling transports energy into the liquid by two main mechanisms: the first is the transient conduction during and right after the bubble detachment. In this process, the departing bubble carries the superheated liquid layer as it rise, allowing colder bulk liquid to re-wet the heating surface (step 2 and 3 in Fig. 1.2). The superheated layer is then renewed via heat conduction. This mechanism contributes more than 75% of total energy transferred. The second mechanism of heat transfer is attributed to the evaporation liquid meniscus at the three-phase contact line during the bubble's growth [6].

The higher the temperature, the more nucleation sites becoming active and more

FIGURE 1.2: One full nucleate boiling cycle where the suggested heat transfer mechanism is marked in red. The figure is reprinted from Ref. [6] with permission.

bubbles are formed. This increase in bubbles nucleation rate result into their coalesces and bigger bubbles are formed. The liquid flow near the substrate is stalled and the bubble's detachment is obstructed, which gradually reduces the heat transfer efficiency. When the relative reduction of the heat transfer coefficient balances with the relative increase in temperature difference (point B), the maximum heat transfer rate is achieved. This point has been termed the critical heat flux (CHF). Beyond the CHF, considerable vapor is formed, preventing the liquid from continuously wetting the surface. As heat transfer drops sharply, thermal energy is trapped within the substrate and boosts its temperature. The system is said to enter the boiling crisis [7]. The heat flux then continues to fall until the liquid is fully separated from the substrate by a vapor film (point C). This phenomenon is called the Leidenfrost effect [8, 9]. From here on, the thin film boiling regime completely replaces nucleate boiling. Thermal energy is transferred to the liquid mostly by radiation from the substrate and by conduction across the vapor layer. The heat transfer rate then recovers, but at the cost of significantly overheating the substrate.

The boiling crisis is a major issue in all thermal management devices. For example, a hypothetical boiling-based cooling device that follows the boiling curve in Fig. 1.1 will function properly until point B. However, once the thermal load is pushed beyond the CHF, the system will transit from point B to point D, causing the device's temperature to rise by approximately 1000 °C. This will most likely result in thermal failure and breakdown of the device. Thus, the boiling crisis becomes the operational limit of generating and cooling systems.

1.2 Overcoming the Boiling Crisis

1.2.1 The physics behind the boiling crisis

Many research on the boiling crisis has been conducted in the last fifty years, yet it is still remained as one of the most challenging problem among thermal and fluidic instabilities [10]. Five different mechanisms were proposed to explain and estimate the CHF of the boiling crisis, namely: bubble interference [11], hydrodynamic instability [12], macrolayer dryout [13], hot/dry spots [14, 15] and interfacial lift-off [4]. An extensive review of these studies can be found in Ref. [16]. Among the proposed mechanisms, Zuber's models using hydrodynamic instability [12] has attracted the most attention.

A schematic of Zuber's proposed model is presented in Fig. 1.3. This model describes the heating substrate prior to CHF as a set of multiple square cells, each having a single vapor jet column at the center and is surrounded by the boiling liquid. The vapor columns originate from the nucleation sites on the substrate and are replenished by the liquid flowing between them. The assumption is that all heat taken from the substrate is used to vaporize the incoming liquid to sustain the vapor jet. Due to the differences in velocity between the vapor jet and the liquid, the Helmholtz instability may occur at their interfaces, causing the expansion and eventually merging of adjacent jet columns. The CHF is reached when the jet

FIGURE 1.3: Zuber's hydrodynamic instability model: (a) The formation of vapor jet columns prior to CHF (b) Proposed substrate's geometry consisting of multiple unit cells, each having a single jet at the center and liquid at the side (c) The formation of the vapor blanket due to Helmholtz instability. This figure is reprinted from Ref. [16] with permission

columns completely merge to form a vapor blanket separating the liquid and the substrate. Following this argument, the value of the CHF can be obtained as [17]:

$$q''_{CHF} = \rho_g L [\sigma g (\rho_f - \rho_g) / \rho_g^2]^{1/4} K \quad (1.1)$$

where ρ_g and ρ_f are the density of the vapor state and liquid state, respectively, L is the latent heat of evaporation, g is the gravity constant, σ is the surface tension and K is the dimensionless critical heat flux constant, which is estimated to be 0.131. This model assumes a perfectly flat surface and uniform distribution of nucleation sites, thus CHF only depends on the properties of the liquid. For water, the CHF can be calculated following Eqn. (1.1) to be $\approx 1.14 \text{ MW/m}^2$. Later on, the characteristics of the heating surface such as materials, roughness, wettability, orientation and more were identified as important factors, however their detailed contributions are still under debates [16, 18–20]. Nevertheless, one important, generally accepted result is that when water serves as the liquid and metals (copper, aluminum, stainless steel) serve as the substrate, the critical temperature where the boiling crisis takes place is between 150–250 °C and the CHF is of the order of 1 MW/m^2 [20–22].

1.2.2 Previous approaches to overcome the boiling crisis

Along with fundamental studies on the mechanism behind the boiling crisis, many studies were conducted in order to push the CHF-limit. They can be categorized into two main approaches. The first approach involves coating the heating surface with micro/nano structures such as micro/nano pillars, porous mesh, nano wires, micro tunnels or reentrant cavities (Fig. 1.4a). Such modifications extended the surface area, increase the nucleation site density or surface's wettability and enhance the liquid flow through capillary pumping. The techniques and results of these studies are extensively reviewed in many articles. Some examples are found in the review Scott *et al.* [18], which summarizes boiling enhancement technique

FIGURE 1.4: (a) SEM images of microstructured heating substrate. (b) SEM images of NiCr wires in water diluted with Carbon nanotubes at various concentrations after pool boiling experiment. Figure a is reprinted with permission from Ref. [32], Figure b is reprinted with permission from Ref. [33].

before the year 2001. Later, Mc Carthy *et al.* [23] and Kim *et al.* [24] review the fabrication techniques and effectiveness of microstructured heating surfaces. Lu and Kandlikar [25] focuses on nanoscale surface modification, followed by Patil and Kandlikar [26], which addresses the manufacturing of porous surfaces for pool boiling applications. The most recent reviews of surface modification approaches are Refs. [19, 27, 28]. The second approach is to dilute the liquid with nano particles or nano rods (nanofluid), which significantly enhances heat transfer within the liquid (Fig. 1.4b). Once they sediment to the heated bottom, these nano-particles increase thermal conduction and temporary modify the surface just like textured coating mentioned above. The results of this approach are reviewed in Ref. [27, 29–31]. Both surface modification and nanofluid approaches can increase the CHF by several times, yet two main issues remain. Firstly, their reported performances are inconsistent when comparing different studies. The collected data is scattered widely and no physical model has been proposed for quantification of the effect. This may be attributed to the stochastic nature of the onset and instabilities occurring during the boiling process, amplifying minor differences in the coating of the substrates or the properties of the liquid to large differences in the performances. The second and more fundamental issue is that these methods only delay the limit of the nucleate boiling regime but do not prevent the formation of the vapor film. Eventually, they all succumb to the inevitable boiling crisis.

FIGURE 1.5: Schematic plot of the oscillate boiling mechanism

1.3 Introducing a New Boiling Regime: Oscillate Boiling

Our work also aims at overcoming the boiling crisis. However, rather than trying to extend the limit of nucleate boiling like previous approaches, we develop and study a new boiling regime termed *oscillate boiling*. The functionality of this boiling regime does not involve the growth and detachment of boiling bubble. Instead, by localizing the heating region, the boiling bubbles undergo high frequency volume oscillation with maximum radius of $\approx 15 \mu\text{m}$ while remaining pinned at the heating spot (Fig. 1.5). This eliminates the possibility of them merging to form a vapor layer and effectively prevent the boiling crisis. Moreover, oscillate boiling bubble induces a Marangoni and a streaming flow, which enhances thermal transport, thus speeding up the heating of the bulk liquid and cooling of the heating substrate.

The occurrence of an oscillatory behaviour of a bubble when heated with a constant power source might have been first reported by Sukhodolsky [34, 35]. They focused a continuous laser beam onto a transparent wall of a cell containing light absorbing liquid, and thus create a so-called “thermocavitation” bubble. The bubble, after

FIGURE 1.6: Previous reported observations that resemble oscillate boiling. (a) The formation of the jet-resembling “structure” that is reported by Peng [40] (b) The bubble formed on thin strip heater, reported by Gureca *et al.* [41] (c) The “plasmonic bubble” reported by Lohse *et al* [42]. The figures are reprinted with permission.

expansion, collapses and detaches from the wall before the cycle repeats with a new bubble. The distinguishing feature is that in “thermocavitation”, the energy is absorbed at the liquid region, leading to a relatively large bubbles with low frequencies (≈ 20 kHz). While in oscillate boiling the energy is absorbed at the heater surface, leading to a surface-attached bubble oscillating at much higher frequencies of several 100 kHz). In fact, the idea of using miniature surface-attached heaters to create boiling bubble has been explored. Some previous studies have employed singular heating element to study the formation and thermal profile of a single boiling bubble [36–39]. However, being larger than $500 \mu\text{m}$ in size, these devices can only create nucleate boiling and not reaching the stable oscillate boiling regime.

The observation that most closely resembles oscillate boiling might have been reported by Peng [40]. There thin-wire heaters are immersed into a subcooled liquid, leading to the formation of boiling bubbles that eject a jet-resembling “structure” (Fig. 1.6a). This structure is similar to the bubble jet emitted by oscillate boiling bubble. The author explains this so-called “jet flow phenomena” as the results of thermocapillary flows induced by the boiling bubbles on the heating wire, which is driving the liquid away. However, neither the bubble’s dynamics nor the jet-resembling “structure” are resolved and thoroughly studied. Guereca *et al.* [41]

might have also observed the unstable oscillate boiling. Using a single thin film strip heater, they create a boiling bubble and observe the heater's temperature oscillating for several cycles during the process (Fig. 1.6b). Even though the oscillation frequency matches that of oscillate boiling bubble (≈ 1 MHz), the spatial and temporal bubble's dynamics is not resolved and the phenomenon is left unexplored. Most recently, after our work on oscillate boiling have been published, Lohse *et al.* [42] reports a similar observation. Using the same technique, i.e. focusing a continuous wave laser beam onto a gold-coated substrate, the authors observe an initial bubble explosion, followed by a few cycles of bubble oscillation (Fig. 1.6c). We believe this so-called "plasmonic bubble" is, in fact, the initial step of a long-lasting oscillate boiling process that might follow. The later process however was not reported. This study is also limited to reporting the observation without providing in-depth physical explanation leading to the oscillations.

In summary, besides great potential application, oscillate boiling is rather unexplored. This sparks great interest to conduct fundamental studies to understand the physics behind this phenomenon. This thesis explores the fluid dynamics and limits of the oscillate boiling regime. For this we report on the experimental observations, numerical simulation and theories to describe the phenomenon, then study different approaches to create, manipulate and scale up this regime for later applications.

1.4 Guide to the Thesis

Chapter 2 presents the first experimental observations of the oscillate boiling regime created by focusing a continuous laser beam onto a gold-coated substrate. The regime is characterized with high-speed imaging and the pressure emitted into the surrounding liquid. Its driving mechanism is explained with the formation of a liquid jet during the bubble's collapse and its impact onto the heating substrate. The theory is supported by a potential flow numerical simulation. Visions on the potential application of the regime are also provided.

Laser-based heating is not feasible to be implemented in a commercial devices. Therefore, **Chapter 3** presents another approach to create the oscillate boiling regime using an electrical microheater. The microheater is integrated with a temperature sensor, which provides thermal readings of the substrate during the boiling process. This additional data strengthens the theory proposed in Chapter 2. With the electrical heater, two different modes of the oscillate boiling regime are now resolved: the stable mode and the unstable mode. The design criteria and input power that lead to these modes are also reported.

Unlike nucleate boiling which relies heavily on buoyancy [6], oscillate boiling does not involve the detachment and rise of the boiling bubbles and thus is speculated to be independent of gravity. **Chapter 4** proves this claim by presenting the dynamics of oscillate boiling bubble in different gravity condition on parabolic flights. The results suggest the potential application of oscillate boiling for outer-space thermal application.

Chapter 5 studies the dynamics of two-closely placed oscillate boiling bubbles. This is a fundamental work towards the goal of scaling up the device where heating is done by a substrate covered with many oscillate boiling bubbles simultaneously. Depending on their distance and driving power, these bubbles can either oscillate independently, merge and grow, or most interestingly, synchronize. The mechanisms that lead to these different behaviors are studied, backed with a potential flow model. The findings provides the design criteria to scale up the phenomenon for applications.

Chapter 6 summarizes and concludes the findings on the oscillate boiling regime. The thesis ends with an outlook on future research directions that may allow to enable the commercialization of this new boiling regime.

Chapter 2

The Physics of Oscillate Boiling

2.1 Introduction

Oscillate boiling regime offers a path to avoid the film boiling regime in future heater designs, which may lead to substantial energy savings and reductions in carbon emission. This regime can be easily observed with a simple setup by focusing a continuous wave laser beam onto a flat substrate coated with a layer of light absorber. After some transient dynamics a single bubble remains at the heated spot which undergoes large clock-like volume oscillations at unexpected high frequencies. These oscillations together with thermocapillary stresses advect heat and stabilize the thermal condition at the heater.

In this chapter, we resolve the dynamics of the oscillate boiling regime with high speed imaging and simultaneously monitor of the acoustic emission. Then we try to understand the unexpected phenomena with simplified modeling. We also demonstrate the potential of this regime for application. The results presented in this chapter was published as “[Li, F., Gonzalez-Avila, S. R., Nguyen, D. M., Ohl,](#)

C. D. (2017). Oscillate boiling from microheaters. *Physical Review Fluids*, 2(1), 014007.” [43] ¹.

2.2 Materials and Methods

2.2.1 Test section preparation

A pre-cleaned microscope glass slide (8 mm x 8 mm, 1 mm thickness) is deposited with gold nanoparticles by sputtering (JFC-1600, Auto fine coater, JEOL) at 10 mA for 300 s to achieve a coating thickness of around 165 nm. The thickness of the gold layer was chosen to be sufficiently thick to absorb incoming laser beam, yet thin enough to allow approximately 50 % transparency for bottom view observation. The gold-coated substrate is enclosed with an acrylic container (10 mm x 10 mm x 12 mm) that has two opposite glass windows for better quality of illumination and imaging, and is attached to a microscope slide, thus forming the test section. The test container is filled with deionized (DI) water (fresh distilled water from Milli-Q (Millipore Corporation) with resistivity below 18.2 MΩcm at 25 °C.) to a height of about 7 mm above the submerged gold-coated substrate.

2.2.2 Experimental setup

A schematic of the experimental setup is shown in Fig. 2.1. The test section is coupled to a three-axis stage (minimum resolution 0.01 mm) for positioning accuracy and placed on a custom-made microscope stage of an inverted microscope (Olympus IX-71, Japan). A 1 W continuous wave laser ($\lambda = 532$ nm, 1W, Becen Optoelectronics, Shenzhen, China) is used to heat the liquid in the test container. The laser beam is attenuated with a rotating neutral density filter ($0 \leq ND \leq 2$) in such

¹L.F designed, setup the experiment, analyzed the data and wrote the first half of the manuscript. S.R.G designed, setup the experiment and reviewed the manuscript. The author, D.M.N conducted the numerical simulation and additional experiments, wrote the second-half of the manuscript and prepared supplementary materials. C.D.O supervised the project.

FIGURE 2.1: A schematic diagram of the experimental setup (not drawn to scale)

a way that the energy added to the system can be varied before it is expanded and collimated with a telescope. An electrically-controlled mechanical shutter (10 Hz) is used to control the on period of the laser. The laser beam is focused onto the gold coating on the substrate through a water-immersed objective (20X/0.50NA, Olympus, Japan). The gold coating enhances the absorption of the laser energy and nucleation of cavitation bubbles. A photodiode (DET10A/M, Thorlab) placed in the optical path monitors the on-time of the laser.

The bubble dynamics is recorded from the side-view with a high speed camera (SA-X2), between 100,000 and 1 million frames per second (fps) illuminated with a metal halide lamp (SUMITA 250). Two types of objectives are used throughout the experiments: 20X and 10X long working distance objective (20x PTEM M Plan Apo, 10x EO Plan Apo). The resolution obtained is $1\ \mu\text{m}$ and $2\ \mu\text{m}$ per pixel, respectively. A hydrophone (ONDA, Model HNR-1000) is placed in the liquid next to the laser focus to simultaneously measure the acoustic signal from the bubble oscillations. Both hydrophone and photodiode are connected to an oscilloscope (Lecroy, Wave Runner 64 Xi-A, 2 GHz maximum sampling rate) to monitor the pressure signal and the laser beam profile. The experiment begins when a pulse

delay generator (BNC model 575) triggers the mechanical shutter (CW laser), the high-speed camera, and the oscilloscope simultaneously.

2.2.3 Laser power measurement

The absorption of the gold coated substrate is measured by subtracting the reflected and transmitted laser power from the total laser power. A powermeter (PM100D, Thorlabs) is used to measure the laser power after it passes through the microscope objective. Each laser power setting has been tested with 3 experimental runs to get the average value. Then by multiplying the absorption coefficient, the laser power supplied to the system is calculated. The absorbed laser power by the gold coated substrate is measured to be from 29.5 to 106.1 mW.

2.3 Experimental Results

2.3.1 Generation of surface attached oscillating bubble from a constant power input

After opening the shutter, the gold substrate is illuminated with the laser beam (Fig. 2.2), we observe a rapid vaporous cavitation event where one bubble is explosively expanding to a few hundred micrometer in diameter, rapidly shrinking, fragmenting, and dissolving. Yet a small bubble remains at the laser focus, grows in size and oscillates while its triple contact line seems like remaining pinned. The diameter of the contact area is around $15\ \mu\text{m}$ and its maximum height during the oscillation is about $13\ \mu\text{m}$. Interestingly, the unaided eye observes a dark streak on top of the bubble resembling a smoke screen as shown in Fig. 2.2b; yet using a shorter exposure time it becomes evident that the dark screen is composed of microbubbles leaving the apex of the surface attached oscillating bubble as shown in the inset of Fig. 2.2b.

FIGURE 2.2: Laser powered microheater: (a) The 532 nm CW laser (29.5 mW) is focused onto a thin layer of gold (around 165 nm thickness). (b) A bubble forms on the gold layer and sheds off microbubbles into the bulk (see inset). (c) Details of the bubble oscillation leading to the microbubble pinch-off at the bubble's apex. The selected frames show one typical period of the bubble dynamics. The red-marked frame represents the moment of liquid jet impact (discussed later). The oscillation frequency is around 90 kHz as detected by the hydrophone and the images are taken from a recording at 100,000 frames/s. (d) Captured images of a more stable, repeatable oscillate cycle. The oscillation frequency is higher than the recording frequency, thus selected frames are used. The oscillation frequency is 305 KHz and scale bar is 10 μm .

Selected frames from a high-speed recording taken at short exposure times reveal the bubble oscillation in greater details, see Fig. 2.2c. The bubble expands into a spherical cap, reaches a maximum volume and shrinks while the contact line seems like remaining pinned. This leads to a microbubble pinch-off from the bubble's apex while at minimum volume the bubble obtains a pancake shape. The bubble then grows back into an elongated shape before it reaches again an approximately spherical cap shape. For clarity we show in Fig. 2.2c the sequence of events during one oscillation period for a relatively large bubble of $R_{max} = 21 \mu\text{m}$. A similar but more typical oscillate boiling case of a smaller bubble is shown in Fig. 2.2d. The stream of microbubbles in the liquid observed in Fig. 2.2b is formed by a steady bubble pinch-off from a surface attached bubble oscillating on a rigid boundary. The rise velocities of the microbubbles close to the oscillating bubble are of the order of 2 cm/s while further away they decrease to 5.5 mm/s, indicating a background flow generated by the oscillating bubble.

2.3.2 Characterization of the oscillation frequency

The clock-like repeatability of the bubble oscillation becomes evident from the recording of the acoustic emission. Signals from a hydrophone located close to the bubble (see Fig. 2.2a) are presented in Fig. 2.3. Figure 2.3a correlates the acoustic emission with the laser power monitored with a photodiode. After opening the laser shutter at time $t = 0$ a large hydrophone signal is observed as caused by the vaporous explosion; it is followed by a smaller and steady signal which stops at the moment when the laser is switched off. Zooming into the hydrophone signal, Fig. 2.3a inset, we find a sinusoidal signal at a frequency of about 320 kHz emitted from the bubble. With time dependent Fourier analysis, the so-called spectrogram of the hydrophone signal is shown in Fig. 2.3b. It reveals that after a brief broadband signal at $t \approx 0$ ², a strong fundamental frequency is observed which lasts until the laser is switched off at $t = 250$ ms. Figure 2.3b also reveals, besides the fundamental frequency, sharp bands of higher harmonics, which are expected

²It takes about 1 ms for the cavitation event to occur at the laser power of 29.5 mW.

FIGURE 2.3: Measurement of the bubble oscillation through acoustic emission. (a) The signal picked up by a hydrophone at nucleation and during continuous illumination with the laser beam. The inset showing the sinusoidal signal oscillating with 320 kHz. (b) The spectrogram depicts the fundamental frequency and higher harmonics at nearly constant frequency over the duration of the laser heating of 250 ms.

for large amplitude bubble oscillations [44]. The oscillation can be sustained for more than 20s (not shown here) given that laser illumination is provided.

2.3.3 Explaining the bubble oscillation

What stabilizes this mechanical oscillator for nearly 10 million cycles while driven by a continuous and constant power input? A famous result reported by Crum [45] and Blake [46] is that when a bubble collapses close to a solid surface, a liquid jet forms, propagates towards and eventually impacts onto the surface. We speculate that the same happens in our oscillate boiling bubble where a jet impinges onto the heated surface during the bubble's collapsing phase (i.e. red-highlighted frame in

FIGURE 2.4: (a) Experimental setup to observe simultaneously the side-view and the bottom-view of an oscillate boiling bubble. (b) Schematic diagram showing how the jet formation is indicated by a central dark spot of the bottom-view observation. Synchronously captured (c) side-view and (d) bottom view of an oscillate boiling bubble. The images with liquid jet is highlighted with the red rectangle. The frame rate is 250 kfps.

Fig. 2.2c). Upon this impact, the jet is instantly vaporized, disrupting the bubble's collapse and drive it into the next expansion phase. This event acts like a “kick” that provides the energy needed to sustain the bubble oscillation. We attribute the liquid jet impact as the main heat transfer mechanism of the oscillate boiling regime.

To support this hypothesis, a stable bubble oscillation is captured simultaneously from both side view and bottom view (see Fig. 2.4a). The formation of the liquid jet, should it exist, will create a concave-like shape on the bubble's interface. As a result, this interface refracts incoming light, leaving the center bottom unilluminated, as shown in Fig. 2.4b. This is what reflected in the experimental results (Fig. 2.4c-d), which shows a central dark spot at the bubble's bottom during the collapsing phase. This observation support our hypothesis regarding the 'kick' mechanism that accounts for the energy transfer of the bubble oscillation.

2.4 Model for The Bubble Oscillation

2.4.1 The kicked oscillator model

The bubble dynamics is simplified by assuming a hemispherical bubble containing gas and vapor which is undergoing oscillations at a boundary with only a radial velocity. This neglects the pinned triple phase contact line, unsteady boundary layers, and bubble pinch-off, yet includes the liquid inertia and the restoring forces from the bubble content. With this assumption the momentum conservation equation for the fluid can be formulated as a 2nd order, non-linear ODE. Here we choose the Keller-Miksis model that accounts for the bubble's contaminant gas, surface tension, viscosity and acoustic radiation from the oscillation. We assume a potential flow problem, whereas our hemispherical bubble staying on top of a solid boundary can be treated as a fully spherical bubble using the method of mirror images. The potential flow assumption is more valid at high Reynold number, where viscosity is negligible, which prevents formation of radial flow. The same technique has proven sufficiently accurate to simulate the oscillation of surface attached bubbles [47]. The high Reynolds number in our case supports this approach, $Re = 2UR_{max}/\nu \approx 100$ where U is the bubble wall velocity and ν is the kinematic viscosity.

At every time-step, the bubble's state is determined by two variables: its radius R and its temperature T . In order to solve for these two unknowns, two equations are required. The first equation, as previously mentioned, is the momentum equation following Keller-Miksis model:

$$\left(1 - \frac{\dot{R}}{c}\right) R\ddot{R} + \frac{3}{2}\dot{R}^2 \left(1 - \frac{\dot{R}}{3c}\right) = \left(1 + \frac{\dot{R}}{c}\right) \frac{p_l}{\rho} + \frac{R}{\rho c} \frac{dp_l}{dt} \quad (2.1)$$

where c is the sound velocity, p_l is the liquid pressure and ρ is the density of the gas-vapour mixture. The pressure p_l can be calculated as:

$$p_l = p_b(T) - p_\infty - \frac{2\sigma}{R} - \frac{4\mu_L \dot{R}}{R} \quad (2.2)$$

Here, p_∞ , σ , μ_L , $p_b(T)$ represents the pressure at infinity, surface tension, liquid viscosity and pressure inside bubble, respectively. The inner bubble pressure $p_b(T)$ is contributed by water vapour p_v and contaminant gas p_g :

$$p_b(T) = p_v(T) + p_g(T) \quad (2.3)$$

Assuming the bubble has an equilibrium radius R_0 at temperature T_0 , the stable gas pressure can be calculated as:

$$p_{g0} = p_\infty + \frac{2\sigma}{R_0} - p_v(T_0) \quad (2.4)$$

This stable pressure can subsequently be used to calculate the gas pressure at any later time given a determined temperature T and volume V of the bubble:

$$\frac{p_g(T)V}{T} = \frac{p_{g0}V_0}{T_0} \quad (2.5)$$

Substituting equation (2.2), (2.3) and (2.4) into (2.1) gives us the first equation describing the dynamics of the oscillating bubble. The second equation is the energy conservation equation. To describe the energy behavior in the system, we apply the first law of thermodynamics for the gas/vapour mixture inside the bubble:

$$pdV + dU = dQ \quad (2.6)$$

where V , U and Q is the bubble's volume, potential energy and heat supply, respectively. The first term on the LHS is the work done by the gas/vapour mixture:

$$pdV = p_b(T)4\pi R^2 dR \quad (2.7)$$

The second term on the LHS is the change in potential energy:

$$dU = dn_v c_V T + (n_v + n_g) c_V dT \quad (2.8)$$

where the change rate of water vapour dn_v is associated with the change of bubble's radius and temperature, the gas molecular number n_g can be deduced from the current gas pressure and temperature, and c_V is the isobaric molecular heat capacity of the mixture. The RHS in equation (2.6) indicates the heat supplied to the bubble, which equals the heat provided by laser power subtracting the heat lost from heat conduction and water evaporation:

$$dQ = P dt - 4\pi R^2 k \frac{T - T_\infty}{l_{th}} - dn_v M L \quad (2.9)$$

In this equation, P is the power input from laser illumination, k , M , L are thermal conductivity, molecular weight, and latent heat of water, respectively. The parameter l_{th} is the thermal boundary layer thickness, calculated as [48]:

$$l_{th} = \min \left(\sqrt{\frac{Rk}{|\dot{R}|}}; \frac{R}{\pi} \right) \quad (2.10)$$

Equation (2.9) is where the 'kick' mechanism comes in. During the expansion phase of the oscillation, a thick layer of gas and vapour mixture with low heat conductivity prevents the energy from the laser to be fed to the system. This mechanism is represented by a small value of P compared to the original laser power. Only during a brief moment when the bubble collapses, the liquid jet is formed and impinges onto the heated surface. Then energy transfer is significantly enhanced, which is represented by an increase of the P value. In short, the heating power P , is chosen so that whenever bubble's effective radius is larger than a critical radius, R_{crit} , the input power is zero (due to poor heat conduction of the bubble content), and when the effective radius is smaller than the critical radius, the input power is equal to the power of the laser. At the time of the work, we chose arbitrarily $R_{crit} = 1 \mu\text{m}$ as it is clear that $R_{crit} \ll R_{max}^{ss}$ (steady-state maximum

radius), and in the experiments we find a similar minimum bubble size. After the work of Supponen et al. [49] comes online, the value of R_{crit} can be estimated following Eqn. (6.1) in that paper.

Substituting Eqn. (2.6)-(2.10) into Eqn. (2.5) yields the second equation to solve for the dynamics of the oscillating bubble.

2.4.2 The simulation result

Figure 2.5a depicts the dynamics of the bubble oscillator for typical experimental parameters of water at $T_l = 40^\circ \text{C}$ and two initial conditions. We find that both radius-time curves show the same final dynamics, i.e. an inertial-driven bubble oscillation with strong collapses and a constant steady-state maximum radius, R_{max}^{ss} . A bubble starting with a radius $R(t=0) > R_{max}^{ss}$ decreases its amplitude within a few cycles while a bubble starting with a smaller radius $R(t=0) < R_{max}^{ss}$ increases its amplitude. This supports the robustness of the bubble oscillator observed in experiments. The inset of Fig. 2.5a is a phase-space plot ($U = dR/dt, R$) for these two initial conditions; it clearly demonstrates that both trajectories eventually converge to the same limit cycle.

The simple ordinary differential equations (ODE) model allows now to study the resonance frequency of the system as a function of the laser power or R_{max}^{ss} . For this plot the laser power is varied between 3 mW and 120 mW and the resulting steady state maximum radius and the fundamental frequency of the bubble oscillation (resonance frequency) is measured. It is well known that the oscillation period of an inertial bubble near a boundary is prolonged. For hemispherical bubbles an increase of $k = 1.2$ has been measured [50, 51] and is multiplied to the calculated radius R_{max}^{ss} .

We compare the prediction of the model with the experiment by plotting simulations and experimental data of the resonance frequency as a function of the steady state R_{max}^{ss} . Also, we compare the predicted and measured R_{max}^{ss} as a function of

FIGURE 2.5: Modeling results of bubble oscillation and comparison to experiments. (a) Solution of the kicked bubble oscillator model (see text) for two initial conditions $R(t = 0)$. One starting at a bubble radius larger than the steady state maximum radius $R_{\text{max}}^{\text{ss}}$ and one at a smaller radius. The phase-space inset demonstrates that both trajectories approach the same attractor. The bubble's effective radius is calculated by assuming a hemispherical cap of the same volume. (b) Dependence of the resonance frequency (solid line) and laser power (dashed line) on the maximum bubble radius $R_{\text{max}}^{\text{ss}}$. The experimental data points are plotted as filled circles and squares.

the laser energy. With increasing laser power we see a monotonically decreasing resonance frequency $420 \text{ kHz} \geq f_{res} \geq 200 \text{ kHz}$, all having period-one solutions of the non-linear oscillator. The predicted bubble radius R_{max}^{ss} increases from $8.7 \mu\text{m}$ to $20 \mu\text{m}$. The model captures the experiments quantitatively for the smaller bubbles, while some difference is observed for largest bubbles which are about 30% smaller than predicted. We also validate the model by comparing the measured laser power absorbed within the gold film with the power input of the source used in the simulations, see dashed curve in Fig. 2.5b. Again qualitative agreement with the simple ODE model is achieved supporting that the important fluid and thermodynamics is accounted for.

2.5 Model for the Thermocapillary Flow

2.5.1 The simulation setup

The small size of the heater suggests that large thermal gradients may lead to a thermocapillary flow. Therefore, we model the thermal field to obtain the Marangoni stresses on the bubble interface and the resulting flow pattern. As we have been measuring only the time-averaged flow we simplify the problem by neglecting the bubble oscillation; the bubble has then the time averaged shape of a spherical cap.

Figure 2.6 presents the computational domain, which consists of two parts: the bubble domain and the liquid domain. The bubble domain has a constant temperature following Clausius-Clapeyron equation:

$$T = T_s + \frac{2\sigma T_s}{L\rho_V R_c} \quad (2.11)$$

where T_s is the saturation temperature and R_c is the gas constant. For the liquid domain, we used Boussinesq approximation to approximate liquid density at different temperature. In this approximation, variations in fluid properties other

FIGURE 2.6: The simulation domain for the flow profile of the liquid around the oscillate boiling bubble

than density ρ are ignored. Furthermore, the variation in ρ is only considered when multiplied with gravitational acceleration g . The density dependency of liquid on temperature is stated as:

$$\rho_l = \rho_{l0} - \alpha \rho_{l0} (T - T_0) \quad (2.12)$$

where α is the coefficient of thermal expansion. For mass conservation, the continuity equation reads:

$$\frac{\partial \rho_l}{\partial t} + \nabla \cdot (\rho_l \mathbf{u}) = 0 \quad (2.13)$$

where $\mathbf{u}$ is the liquid velocity. If we ignore variations in density, Eqn. 2.13 becomes:

$$\nabla \cdot \mathbf{u} = 0 \quad (2.14)$$

For momentum conservation, we use the Navier-Stokes equation for incompressible fluid:

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{1}{\rho_l} \nabla p + \nu \nabla^2 \mathbf{u} + \frac{1}{\rho_l} \mathbf{F} \quad (2.15)$$

whereas $\mathbf{F} = \Delta \rho \mathbf{g}$ is the gravitational force. Substituting Eqn. (2.12) into Eqn. (2.15) gives:

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{1}{\rho_l} \nabla p + \nu \nabla^2 \mathbf{u} + \mathbf{g} \alpha (T - T_0) \quad (2.16)$$

The equation for thermal flow is stated as:

$$\frac{\partial T_l}{\partial t} = D \nabla^2 T_l - \nabla \cdot (\mathbf{u} T_l) \quad (2.17)$$

where D is the thermal diffusivity of the liquid. Equations (2.14), (2.16) and (2.17) are to be solved for the liquid domain.

Open boundary condition is applied to the outer boundary of the liquid domain. For the vapour-liquid boundary two constraints are applied: the first one is the energy equation for heat transfer at the interface:

$$dQ = h_i (T - T_i) \quad , \quad (2.18)$$

with h_i being the interfacial heat transfer coefficient, calculated following Ref. [52]. Equation (2.16) describes the Marangoni effect along the bubble surface. The driving mechanism of this effect is the temperature gradient along the vapor-liquid boundary, which results in a surface tension gradient, causing a viscous stress component:

$$\tau_{R\theta} = -\frac{1}{R} \frac{d\sigma}{dT_i} \left(\frac{\partial T_i}{\partial \theta} \right) \quad (2.19)$$

where θ is the angular coordinate relative to the center of the bubble (see Fig. 2.6). This thermocapillary stress accelerates the liquid from high temperature regions near the contact line to the bubble's apex. The effect of laser illumination is

represented by a heat source boundary condition on the gold surface. The equations are solved with the finite element method (COMSOL Multiphysics).

2.5.2 The simulation results

FIGURE 2.7: Modelling results of the thermocapillary flow and comparison to experiments. (a) Simulation of the heat transfer (right) and the thermocapillary effect induced flow (left) of a steady bubble under laser irradiation of 40 mW for 10 milliseconds. (b) Flow field obtained from particle image velocimetry with $2\text{ }\mu\text{m}$ diameter particles. Due to illumination constraints we could not identify particles in the bottom part of the image. Note the different image scales on the left and right figure.

The simulation results are shown in Fig.2.7a with a bubble diameter of $17\text{ }\mu\text{m}$ and laser power of 40 mW. The flow field profile is presented on the left and the temperature distribution is presented on the right. The flow field profile shows a Marangoni flow at the gas-liquid interface. This flow drives liquid upward at the top and toward the bubble at the side. The upward flow (around 40 cm/s close to the bubble's top surface, and decreasing further away) contributes to the rise of pinched-off bubble and significantly enhances heat transfer from the hot substate to the cold liquid in the upper region. Meanwhile, the flow at the side drives liquid

towards the bubble and causes a cooling of the substrate, i.e. stabilises the thermal gradients near the bubble. Thereby, the boiling region remains localised. Although the present simulations don't account for this, we understand that the recirculation of cold liquid is essential for the stability of the bubble oscillation.

The general features of the flow field of Fig. 2.7a (left) are found in experiments using particle image velocimetry (PIV), Fig. 2.7b. There we see a central flow pointing upward and a cut in the (r, z) -plane through a vortex ring as predicted in the simulations. The maximum velocities are of the order of 10 mm/s approximately 200 μm above the oscillating bubble while simulations predict a value of 2 mm/s at the same location. It may be that the nonlinear advection term in the Navier Stokes equation results in a streaming flow solution responsible for the transport, see Mannaseh [53].

In principle, if the PIV data could be acquired at different phases of the bubble dynamics, it would add important fluid dynamics information to each individual phase. An excellent example of such job was done by Sankin et. al [54] where μPIV was used to capture interactions between laser-generated bubbles and biological cell. Here, we have captured the velocity field throughout all of bubble's phases such as forming, oscillating, microbubble pinching and transiting to nucleate boiling. However, due to equipment limitation, we trade off temporal resolution for a bigger capture window so that a complete picture of the surrounding liquid can be observed. Yet, we believe the average velocity field is informative and supportive to our numerical result.

Conventional nucleate boiling relies solely on buoyancy to transport hot liquid from the heated surface to the upper liquid region. In contrast, heat transfer in oscillate boiling has the additional support from the thermocapillary flow. As a result, the rate of temperature increase, dT/dt , in the liquid with the presence of the oscillate boiling bubble can be much faster than that from a non-oscillating but growing bubble. This feature is partially shown in the simulated liquid thermal profile (Fig. 2.7a right). To further support this claim, another experiment is conducted.

FIGURE 2.8: (a) The diagram of the temperature measurement two cases: oscillate boiling and nucleate boiling. (b) The liquid temperature picked up by the thermocouples in two cases.

Here we use a $50\mu\text{m}$ -diameter thermocouple type K to probe liquid temperature at $200\mu\text{m}$ directly above the bubble in two cases. The first case is where the bubble is in the aforementioned oscillate boiling regime. In the second case, by using the same energy input, but slightly increase the lasing area, we create a growing bubble as in the nucleate boiling regime (Fig. 2.8a). As expected, the thermal readings show liquid temperature increases 3.4 times faster in the presence of oscillate boiling bubble (Fig. 2.8b). This indicates that the oscillate boiling regime could be suitable for general heat transfer applications.

2.6 Discussions

The critical ingredient of the oscillate boiling phenomenon is the transport of minute amounts of liquids onto the heater during the collapse of the bubble. This initiates the repetitive start of the new oscillation cycle. Crucial here is the hypothesized contact line pinning, which stabilizes the lateral bubble position and therefore the repeatability of the phenomenon. The heat transfer simulations demonstrate that the thermocapillary flow qualitatively explains the observed flow

field. Besides thermocapillary flow, bubbles in contact with a surface and undergoing volume oscillations can generate a streaming flow and show a rather similar flow pattern [55]. Here we have not investigated the importance of the acoustic streaming flow as the small amplitude approximation for the bubble oscillation is not valid for the inertial collapse. Pinch-off of microbubbles has been previously observed for resonantly driven bubbles in hydrophobic pits [56, 57]. It is observed that the pinched-off microbubbles do not shrink or condense back into the liquid while rising upward. This indicates that these microbubbles contain mainly non-condensable gas, thus contributing to degassing of the main oscillating bubble. It is well known that an oscillating bubble, even in an undersaturated liquid, will accumulate some non-condensable gas due to rectified diffusion and keeps growing [58]. Our oscillate boiling bubbles are no exception, even though initially they contain mostly vapour. The pinch-off mechanism is therefore necessary for the bubble to obtain a stable size and frequency despite the accumulated gas through the rectified diffusion.

A hot stream (jet) of liquid from the top of a bubble has also been reported for nucleate boiling on thin wires and termed *jet flow* phenomena. There, the bubbles are generally moving along the wire and reveal complex dynamics and interaction. For one particular case of these jet flows termed the “bubble bunch jet flow”, it was speculated to consist of a stream of vapour bubbles [59]. Interestingly, this observation was made close to the critical heat flux that may relate to the loss of the liquid microlayer [60] which resulted into a pinning of the three phase contact line.

A second manifestation of the oscillate boiling regime may have been reported by MacDonald *et al.* [61]. They inferred from a high frequency oscillations picked up with a photodiode that a bubble oscillates at the tip of an optical fiber submerged into liquid nitrogen. The bubble may have been generated with $1\ \mu\text{s}$ long laser pulses absorbed at the tip of a metallic film-coated fiber ($9\ \mu\text{m}$ core diameter). They observed bubble oscillations lasting for $20\ \mu\text{s}$ with a decaying frequency starting from 17 MHz.

2.7 Conclusion

In summary, this chapter shows the existence of an novel oscillate boiling regime on sufficiently small heaters with constant power input in subcooled water. Using high speed imaging and acoustic detection, we resolve stable, clock-like oscillations of a surface attached bubble repeating on a cycle of microseconds. The self-driven oscillation can be explained by the non-spherical bubble collapses and a large scale recirculating flow, as captured both in experiments and by simplified modeling of a thermally kicked oscillator and Marangoni flow. Temperature measurements and modeling indicate the oscillate boiling regime may enhance heat transfer efficiency as compared to conventional boiling.

Chapter 3

Oscillate Boiling from Electrical Microheater

3.1 Introduction

Chapter 2 reported the *oscillate boiling regime* which might allow to overcome the boiling crisis through stable localized heating. The finding was that bubbles at the heater undergo stable high-frequency oscillations while remaining pinned. In particular, the bubble does not grow over time. This is achieved by shedding excess amount of gas and vapour by pinching off a microbubble during each oscillation. Additionally, a large scale flow is generated that cools the substrate thereby keeps the temperature distribution localized to the heater. The key feature to generate the oscillate boiling regime is a miniature and localized heat source. A typical size of the heater to operate in water is about $10\text{ }\mu\text{m}$ in diameter. As presented in Chapter 2 and Ref. [43], this was achieved by tightly focusing a continuous wave laser beam onto an absorbing substrate made of gold nanoparticles. Yet, using laser light as a heat source carries two drawbacks: Firstly, due to optical loss from the focusing optics and through reflections at the gold layer, only about 20 % of the laser power remains available for heating. Secondly, the optical setup to generate

more than one bubble becomes rather complex and rules out practical scaling up of the phenomenon.

In this chapter, we present our approach to overcome these two restrictions (efficiency and scalability) by creating oscillate boiling with an electric heater. The additional benefit is the ability to measure the instantaneous heater’s temperature to elucidate the complex fluid mechanics in the novel regime. The results presented in this chapter was published as “Nguyen, D. M., Hu, L., Miao, J., Ohl, C. D. (2018). Oscillate boiling from electrical microheaters. *Physical Review Applied*, 10(4), 044064.” [62] ¹.

Previously, electric miniature heaters have attracted attentions, e.g [36–39, 41, 63]. They were used to create and study the dynamics of the conventional nucleate boiling, where the liquid is heated rapidly above the spinodal limit leading to an explosive vaporization. We are not aware of reports demonstrating a stably oscillating bubble characterizing the oscillate boiling regime.

3.2 Materials and Methods

3.2.1 Electrical microheater

We have designed and fabricated a miniature heater to achieve the above mentioned temperatures on a sufficiently small scale. The heater consists of a 50 nm thick platinum (Pt) layer, deposited on a 200 μm thick glass substrate. The heater is contacted with a 600 nm thick gold layer deposited with magnetron sputtering (Fig. 3.1a). The gold electrodes have a high electrical conductance and large cross-sectional area to ensure a current supply with little resistive loss. In contrast, the high resistance from the thin 15 $\mu\text{m} \times 15 \mu\text{m}$ Pt heater creates a very localized heating – approaching the conditions of the laser focus based heater [43]. Prior

¹The author, D.M.N, designed the heater, conducted the experiment, analyzed the results and wrote the manuscript, H.L fabricated the sample and reviewed the manuscript, M.J supervised the fabrication process, C.D.O supervised the project and revised the manuscript.

FIGURE 3.1: Design of the electrical microheater. (a) The fabrication process. The platinum heating layer is deposited on top of glass substrate, followed by the gold electrode layer. (b) The four probe structure used to measure the heater's resistance. (c) Captured image of the microheater. The scale bar shows 20 μm

to the experiment, a numerical simulation was carried out to compare the thermal profile of an electrical heater with the prior laser based heater. For the former, the electric current and Joule's equation are solved simultaneously with the heat transfer equation. In the latter, the laser-focused point is represented by a solid heat source at the gold surface and only the heat transfer equation is solved. The equations are solved with finite element methods using the commercial COMSOL Multiphysics software package. The temperature distribution presented in Fig. 3.2 reveals close similarity between the two heating methods. This gave us confidence that the electrical microheater could perform very similar to its optical counterpart.

To measure the instantaneous temperature of the heater the temperature dependent electrical resistivity of platinum is utilized. Besides its relatively high resistivity it also has a nearly constant thermoresistance coefficient up to 500°C, see [64]. The temperature-resistance dependency of the heater reads:

FIGURE 3.2: Thermal profile of (a) an electrical heater and (b) a laser-induced heater with the same size and power.

$$R = R_0[1 + \alpha(T - T_0)] \quad , \quad (3.1)$$

where R_0 and R are the initial and current resistance; T_0 and T are the initial and current temperature, respectively; α is the temperature coefficient of resistance. The resistance R to determine the temperature T is obtained from two additional probes measuring the voltage drop across the heater as shown in Fig. 3.1b. To convert from R to T , we went through a calibration step to find α . In this calibration, the test section with the microheater at its bottom is filled with externally pre-boiled water. Then as the water cools down, we measure the heater's resistance as a function of temperature obtained with a thermocouple type K submerged in the water. The data is then fitted to a straight line to obtain α . The results of the calibration is presented in Fig. 3.3, from which α can be calculated to be $1.023 \times 10^{-3} \text{ K}^{-1}$.

Figure 3.4 describes in detail the fabrication process of the electrical microheater. Initially, a 4-inch diameter glass wafer with a thickness of $200 \text{ } \mu\text{m}$ is immersed into sulfuric acid (H_2SO_4) at 120°C for 30 minutes for cleaning. Next, the wafer is coated with hexamethyldisilazane (HMDS) for 2 minutes in a prime oven in order to improve photoresist adhesion onto the glass surface. Then a layer of $1.5 \text{ } \mu\text{m}$ thick photoresist 7217 (AZ Electronics Materials) is spin-coated onto the substrate

FIGURE 3.3: Calibration curve of the heater's temperature as a function of temperature.

at 5000 revolutions per minute (rpm) for 30 s. Thereafter, the structure pattern on a prepared glass photomask is exposed onto the coated wafer under 365 nm wavelength ultraviolet (UV) light for 5 s. This pattern is subsequently developed in an AZ 300MIF developer solution (AZ Electronics Materials) for 1 minute (step 1). Afterwards, the patterned wafer is coated with a layer of 20/50 nm thick Cr/Pt by magnetron sputter for 4 minutes (step 2). The wafer is then immersed into acetone for etching off the photoresist, i.e. lift-off (step 3) which finishes the deposition of the Pt heating layer. The photolithography and sputtering processes are done one more time to deposit the heater's electrodes composed of a layer of 20/600 nm thick Cr/Au (step 4 and 5). After the last lift-off step (step 6), the device's fabrication process is completed.

FIGURE 3.4: Step-by-step fabrication process of the electrical microheater.

3.2.2 Experimental setup

The substrate with the heater is at the bottom of an rectangular acrylic container (10 mm $\times$ 10 mm $\times$ 12 mm). Two of the faces of container are replaced with microscope cover glass to ensure good imaging of the microscopic dynamics. The container is then filled with gas saturated DI water to the height of approximately 7 mm.

The microheater is operated in a constant current mode using a current controller for laser diodes (Thorlabs LDC 220C). The instantaneous current is measured from the voltage drop across a 10 Ω resistor connected in series with the heater. The heater's current and its resistance are recorded with a 12-bit oscilloscope (Lecroy HRO 64Zi).

The small size of the bubble and its rapid dynamics demand for a high-speed camera. We use a SA-X2 (Photron) which is operated between 300,000 and 400,000

frames/s using a 20x magnification lens (20x PTEM M Plan Apo, Edmund Optics). This provides a resolution of $1\ \mu\text{m}$ per pixel. Additionally, a hydrophone (ONDA, model HNR1000) is placed near the heater to capture the acoustic signal emitted from the bubble oscillation. The hydrophone is connected to a third channel of the oscilloscope.

Figure 3.5a illustrates the setup of the experiment. All electronic components (heater, power source, camera, oscilloscope, etc.) are connected to a common ground to reduce electrical noise. For the experiment the power source, camera and oscilloscope are simultaneously triggered with a digital pulse generator (BNC 575, Berkeley Nucleonics).

3.3 Experimental Results

3.3.1 Bubble oscillations

After the power source is triggered, an initial vaporous explosion is observed, leaving behind a small bubble in contact with the microheater (Fig. 3.5b). Within a millisecond, this bubble enters an oscillation state at a fixed frequency. During the collapsing phase of each oscillation, a microbubble is ejected from the apex of the main bubble, which are then driven upward due to thermocapillary flow and form a stream of microbubbles. The main bubble then re-expands and a new cycle starts. Figure 3.6a shows frame-by-frame images of one oscillation period of a relatively large bubble for better visualization while Fig. 3.6b shows a more stable and repeatable one. The overall dynamics can also be obtained from the acoustic emission of the bubble. Figure 3.7a depicts short-time power spectra of the acoustic emission recorded with hydrophone signal. The spectra is plotted as a function of time (spectrogram). The amplitude of the frequency components is indicated with the color; brighter colors depict larger amplitudes. For the hydrophone signal, Fig. 3.7a, the fundamental oscillation frequency is about 160 kHz and remains constant over the covered measurement observation. Additionally, a

FIGURE 3.5: (a) Sketch of the experimental setup to study the oscillate boiling heat transfer. (b) Zoomed-in image of the test section where the bubble is formed on top of the platinum heater.

second harmonic at 320 kHz is evident, which is well known for large amplitude bubble oscillations. In general, the fundamental frequency varies between 90 kHz and 300 kHz as a function of the bubble's size; it increases for smaller bubbles.

3.3.2 Temperature measurements

The obvious advantages of an electrical system are its simpler control of the power input and the possibility to monitor the instantaneous heater temperature during

FIGURE 3.6: Consecutive images from one period of the oscillation cycle. (a) Images of a relatively large (yet unstable) oscillating bubble to visualize the release of microbubbles in greater detail. (b) Images of a more stable and repeatable oscillating boiling bubble. The scale bar shows $20\ \mu\text{m}$ and the frame rate is 400,000 fps.

FIGURE 3.7: (a) Spectrogram from a typical bubble's emitted acoustic signal recorded with a hydrophone. It reveals a strong fundamental frequency and higher harmonics. (b) The spectrogram of the heater's resistance is recorded simultaneously and shows a very similar power spectrum. The heating power used in this experiment is 86 mW.

the bubble oscillation. Figure 3.8a depicts such a recording. The signal consists of a series of cycles with a fast downward spike connected with a positive sloped signal of increasing temperatures in-between. Figure 3.8b depicts a zoomed-in view of the thermal signal (see dashed line in Fig. 3.8a) together with frames from a synchronous high-speed recording. The frames are recorded at the center of each interval indicated with the vertical lines, i.e. frame C showing a re-expanding bubble was taken at $t = 11.25 \mu\text{s}$. The drop of the heater temperature occurred shortly before this frame was recorded. The slope of the temperature change is remarkable: during the interval C in Fig. 3.8b the heater temperature drops within approximately 50 ns by 30 K. Thus the heater experiences a cooling rate of $6 \times 10^8 \text{ K/s}$! That is also the time when the microbubble is pinched off. The temperature recovery is slower, typically within about 500 ns. After this fast recovery the temperature continues to rise at a constant slope, i.e. from $\approx 270^\circ \text{C}$ to about 280°C before the next cycle starts.

Next we compare the thermal signal with the acoustic signal as shown in Fig. 3.7b. There the spectrogram of the thermal signal reveals close similarity with the acoustic signal. The larger noise floor of the thermal signal is due to the smallness of the electric signal and some noise pick-up.

In Section 2.3, it was argued that during the bubble collapse, e.g. between frame B and C of Fig. 3.8b, a liquid jet impinges onto the substrate. Upon its contact with the hot surface, the heat transfer is strongly enhanced, leading to an instantaneous vaporization of a minute amount of liquid and the subsequent vapor explosion is driving the re-expansion of the bubble. During this process the heater loses energy, resulting in a drop in temperature. Soon after, the heater recovers back to its initial temperature through Joule's heating and the cycle starts all over.

This observation suggests that a sufficiently small electric heater is actively cooled by the oscillate boiling phenomenon. Applications which may profit from the cooling are integrated circuits or single high-power transistors operating above the

FIGURE 3.8: (a) Heater's temperature during the oscillate boiling events using an input power of 86 mW. (b) Zoomed-in temperature of the region marked with a dashed line in figure (a) and the corresponding bubble image. The scale bar is 20 μm .

Leidenfrost temperature. Here, oscillate boiling provides means to retain the device at a constant low temperature. It does so by utilizing the heat energy to drive to transport cooled liquid to the device.

Compared with the estimated Leidenfrost temperature of 227°C from Baumeister *et al.* [21] for water as the fluid and platinum as the substrate, the heater's temperatures shown in Fig.3.8 are higher. Thus oscillate boiling remains operational above the Leidenfrost temperature which is in stark contrast to nucleate boiling. Thus oscillate boiling may offer a route to overcome the boiling crisis.

There have been reports where electrical heaters are used to study the thermal profile of a boiling bubble [37, 41]. Interestingly, Romera *et al.* [41] have also reported temperature oscillation from an electric microheater following the nucleate boiling event. They explained the bubble oscillations cycles with vaporization and

condensation due to the thermal gradient in the surrounding subcooled liquid. We in contrast explain the cooling with liquid jetting; the bubble oscillation is essentially inertial cavitation (for a model see Section 2.4.4 and Ref. [43]). The most striking difference is that Romera *et al.* observed a few tens of thermal oscillations before they died out and the heater temperature rose. In contrast we see oscillations lasting for millions of cycles. This difference can be explained that their heater is a thin strip which may allow the bubble to grow in the less confined direction destabilizing the oscillations.

3.4 The Stable and Unstable Regimes

3.4.1 Observations of the unstable regime

The key advantage of an electrical heater over the previous laser heat source is the ability to have a good control of the heating power by adjusting the electric current into the heater. Figure 3.9a plots the maximum bubble radius and the heater's mean temperature of a square heater ($15\mu\text{m}$ width) as a function of the electrical power. Here the bubble's effective radius is defined as the radius of a hemispherical with equivalent volume. The error bar is derived by combining the relative error from multiple measurements and the absolute error coming from the resolution of the camera. With increasing power, 3 distinct parameter regimes are identified: for electrical powers below 35 mW the vapor bubble explosion is absent and convective heating prevails. Once the electrical power is above 35 mW, a vapor explosion occurs followed by a small bubble undergoing volume oscillation (phase 1). However, unlike the one shown in Fig. 3.8, this oscillation shows neither the periodic microbubble pinch-off nor the temperature spikes from the liquid jet impact (phase 2). It lasts for several milliseconds and then decays as the bubble's size increases (phase 3). The oscillation eventually stops and the bubble continues to grow due to heating (phase 4). During this transition, the heater's temperature increases from about 120°C to more than 200°C . Figure 3.9b-d depicts the acoustic

FIGURE 3.9: (a) Properties of oscillate boiling bubble with different input power. In the unstable regime, the data is captured during the oscillations before the bubble grows thermally. (b) Spectrogram of the acoustic signal emitted from an unstable oscillate boiling bubble. (c) Heater's temperature during the transition from oscillate boiling to growing mode. The inset is the enlargement of the red-dashed rectangle. (d) Bubble's effective radius derived from the captured recording. The inset is the enlargement of the red-dashed rectangle.

signal, the heater's temperature, and the effective radius of the bubble during the course of the phases 2 to 4 respectively. We termed this regime unstable oscillate boiling to distinguish it from the stable oscillate boiling (shown in Fig. 3.6-3.8) where the oscillation has a constant frequency and lasts for hundreds of milliseconds. Likely this unstable behavior is the result of gas accumulation from the liquid, i.e. through rectified diffusion [58] and from residual gas of the vaporized water. We speculate that the smaller bubble in this regime does not collapse as violently. Then the jet is not reaching the hot substrate which stabilizes the bubble shape during re-expansion and avoids the microbubble pinch-off. This hypothesis is supported by the absence of thermal spikes in the heater's temperatures, see inset in Fig. 3.8c. We believe that the observation of Romera *et al.* [41] falls into this unstable regime.

Bubbles in the unstable oscillate boiling regime have 4 phases in common: The start through an explosive nucleation (phase 1) followed by an initial oscillation (phase 2). The frequency may or may not be constant. Phase 2 is followed by a decrease in oscillation amplitude and frequency while the bubble increases in size (phase 3). The oscillation then ceases and the bubble grows thermally (phase 4). The duration of each phase varies greatly and is not able to attribute reproducible trends. For example, Fig. 3.10 shows a second example of an unstable oscillate boiling bubble. Again the bubble reveals the 4-phases. However, here the oscillation phase (phase 2) lasts for ~ 2 ms - a relatively short time as compared to the decay phase (phase 3) that lasts for ~ 8 ms. This is different to the case presented in Fig. 3.9b-d. Overall we do not find a simple relation between the heating power and the duration of each phase nor the frequency of the unstable oscillation. The stable and unstable mode can be easily distinguished and a summary of their features is presented in Table 3.1. Yet, we acknowledge the lack of quantitative description of the unstable regime, which will be overcome in future works.

The regime of *stable oscillate boiling* can only be reached if the gas uptake of the bubble is balanced by the gas release through microbubble pinch-off. For the present geometry the electric power must be above 50 mW to achieve stable

FIGURE 3.10: Unstable oscillate boiling revealing only a short interval of oscillate boiling. (a) The spectrogram. (b) The bubble's effective radius taken from individual frames from the high-speed recording. The inset show the zoomed-in data of the marked region. (c) Captured images of the bubble. The time between two consecutive frames is $3.33 \mu\text{s}$. The scale bar represents $20 \mu\text{m}$.

oscillate boiling. As the bubble enters this regime, its maximum effective radius increases, see Fig 3.9a, leading to a stronger bubble collapse which favors liquid jet impact. The size of the bubble increases and the oscillation frequency decreases with increasing power. The maximum power of this electrical design is 90 mW. Above this value the platinum thin film resistor is destroyed, likely due to melting.

Figure 3.9c reveals another thermal effect of oscillate boiling. While the bubble oscillations decay (phase 3) and the thermal growth sets in (phase 4) the heater's temperature increases by about 100 K. We explain this temperature rise with the absence of advective cooling of the heater once the flow generated by the bubble oscillations ceases. The oscillate boiling regime provides means to sustain the

Oscillate Boiling	Unstable	Stable
Heating power range	35mW - 50mW	50mW - 90mW
Periodic microbubble pinch-off	no	yes
Rapid cooling during each cycle	no	yes
Phase 1	Explosive nucleation results in a small oscillating bubble.	
Phase 2	Either varying oscillation frequency or adjusts to a fixed frequency.	The bubble quickly adjusts to a fixed size and frequency.
Phase 3	Bubble increases in size while frequency drops.	N.A.
Phase 4	Oscillations decays and bubble grows thermally.	N.A.

TABLE 3.1: Distinguishing stable from unstable oscillate boiling regimes on a $15\ \mu\text{m} \times 15\ \mu\text{m}$ size heater.

heater at a lower operating temperature by advective cooling.

3.4.2 Model for the unstable regime

In Section 2.4, a model for the dynamics of oscillate boiling bubble was proposed. There the Keller-Miksis [65] equation is coupled with the energy equation for the inner gas/vapor:

$$dQ = pdV + dU \quad (3.2)$$

to solve for the the bubble's instantaneous radius and temperature. Here dQ is the total thermal energy received, p is the bubble's pressure, V is the bubble's volume and U is the bubble's potential energy. In the case of stable oscillate boiling, to mimic the liquid jet impact, the heating power supplied to the bubble (which appears in the term dQ) is approximated to a step function. When the radius smaller then a threshold value, indicating the point of collapse with the jet touching the heating substrate, the bubble receives full power from the heater,

FIGURE 3.11: (a) The power profile used in two cases: with liquid jet impact (stable oscillate boiling) and without liquid jet impact (unstable oscillate boiling). (b) The corresponding calculated bubble dynamics.

and zero otherwise. The value of this threshold can be derived from the power law Eqn. (6.1) in Ref. [49]. With this power profile, the bubbles show a stable oscillation with fixed maximum radius and frequency after a few cycles (blue lines in Fig. 3.11).

In unstable oscillation where there is no jet impact, the smoother heat transfer is approximated to be inversely proportional to the bubble radius. The reason for this approximation is that without the jet impact, heat is transferred to the bubble via conduction through the inner gas/vapor. As the radius increases, the thermal conductance reduces and the bubble receives less power. With this power profile, there is a decay in the oscillation amplitude. The bubble's radius is bigger and its frequency is lower compared to stable oscillate boiling (green lines in Fig. 3.11).

FIGURE 3.12: Parameter study of oscillate boiling bubble with different input power where the size of the heater is $10\mu\text{m} \times 10\mu\text{m}$. Here the bubble's radius is defined as the radius of a hemisphere having the same volume.

These behaviors and the values of bubble's radius see a good agreement with the experimental results.

3.5 Further Discussions

3.5.1 Different sizes of the electrical microheater

Experiments were carried out with square heaters of different sizes, i.e. $10\mu\text{m}$, $15\mu\text{m}$, and $20\mu\text{m}$ side length. The $10\mu\text{m}$ and $15\mu\text{m}$ -sized heaters show similar behaviors, namely with increasing electrical power a transition from convective heating to unstable and then to stable oscillate boiling. As expected the electrical power thresholds are lower for the $10\mu\text{m}$ -sized heater as compared to the $15\mu\text{m}$ heater. These properties are presented in Fig. 3.12. Interestingly the $20\mu\text{m}$ -sized heater can't produce stable oscillate boiling. Up to the point of electrical failure, only unstable oscillate boiling was found. This regime quickly transits into the thermal growth (phase 4). Again, this observation agrees with previously reported results with laser heating. There the focus had to be sufficiently small to obtain stable oscillate boiling.

FIGURE 3.13: The relationship between the bubble's maximum effective radius and its resonance frequency. Also results from laser-based heating are shown,

3.5.2 Comparison between electrical heating and laser-based heating

Figure 3.13 shows that oscillate boiling bubbles generated by electrical heater have qualitative and even quantitative similar oscillation frequencies in the stable regime with the ones obtained from laser-based heat source, which has been presented in Section 2.4 and Ref. [43]. Yet, one needs to take into account that the two approaches lead to rather distinct thermal distributions. The electrical heater has a mostly constant temperature over the surface, which also restricts the size of the bubble. The laser heater, on the other hand, provides a Gaussian intensity distribution, which allows the bubble to choose its size. The previously proposed model which assumes that the bubble's size can be independently adjusted is therefore less accurate in the case of electrical heater.

Another observation is that the maximum bubble radius achievable in the case of electrical heater is greater than in laser heater. We attribute this observation as the electrical heater, having better manufacturing process, is of better quality and can sustain greater amount of heat as compared to laser-based heating.

FIGURE 3.14: Captured images and spectrogram of the emitted acoustic signal of oscillate boiling bubble in (a) normal DI water and (b) degassed water. The scale bare represents $20\ \mu\text{m}$. The power used for both cases are 50 mW.

3.5.3 Effect of gas concentration in the liquid

Additional experiments were conducted using degassed DI water. For that the water has been placed within a vacuum chamber at 3 mbar residual pressure for 60 minutes. A comparison of the bubble dynamics in saturated and degassed water is shown in Fig. 3.14. We find stable oscillate boiling in the degassed water, too. The bubbles demonstrate at the same heating power very similar size and

oscillation frequencies. Bubbles in standard and degassed water emit a stream of microbubbles. At first thought one may argue that the bubbles therefore consist mostly of vapor. However, due to the high temperatures near the bubble, the liquid even at low absolute concentrations from the degassing procedure may still be oversaturated and therefore available for diffusion into the bubble. While the overall gas concentration does not make a strong effect on the bubble dynamics, we can't rule out the contributions from non-condensable gas within the bubble. More careful experiments are necessary to answer the question of what comprises the bubble. Our prediction is that in the ideal case where contaminant gas are completely depleted, oscillate boiling will still be presented. The bubble however will oscillate at higher frequency as the absence of contaminant gas has lower its inertial.

3.6 Conclusion

In conclusion, the oscillate boiling regime can be achieved not only through optical energy as shown previously but also with electrical Joule's heating. This chapter demonstrates a suitable design to achieve precise and simple electrical control with the advantage that the heater's temperature can be monitored. The fast temporal response of the thermal sensors reveal enormous cooling rates of 6×10^8 K/s during a brief moment when the jet impacts. The temperature recordings of the heater for the three identified regimes of convective heating, unstable and stable oscillate boiling are reported. In general, at lower electric power oscillate boiling is unstable; the average bubble volume increases over time due to accumulation of gas inside the bubble. At a higher power stable oscillate boiling is obtained. In this regime the heater's temperature is controlled and limited by the effective heat transfer of jet impact caused by the non-spherical bubble collapse. Thus the oscillate boiling phenomenon may not only be a promising candidate for heating applications it also allows the effective removal of heat from electrical devices plagued by Ohmic heating.

Chapter 4

Oscillate Boiling in Microgravity

4.1 Introduction

The development of space technology involves light-weight, compact and high-power devices, while at the same time requires strict temperature control to maintain proper operation. In ground applications, i.e. terrestrial condition, such tasks are managed by boiling heat transfer, in particular nucleate boiling, as it ensures high thermal efficiency and low temperatures of the dissipating devices [66]. Here, the heat transfer mechanism is attributed to the detachment and rise of the nucleate boiling bubbles [6], in which gravity plays an important role. However, in space with greatly reduced gravity, buoyancy-based boiling encounters serious limits. Earliest observation of pool boiling in microgravity reported by Abe et al. [67], Ohta et al. [68] using sounding rockets, and Oka et al. [69] on parabolic flights show that without buoyancy, boiling bubbles fail to rise. Instead, they grow, coalesce with their neighboring bubbles and hover close to the substrate's proximity (see Fig. 4.1). Later on, using a set of multiple temperature-controlled heating elements, Kim et al. [70] managed to capture the boiling curve and the substrate's temperature during the boiling process. The results reveal that as the bubbles fail to rise, they cause the dryout of the substrate and trap thermal energy within. Consequently, the heater experiences a significant rise in temperature together

FIGURE 4.1: Captured images of pool boiling process in earth gravity and microgravity. The figure is copied from Ref. [81]

with a drop in the critical heat flux (CHF). The same observations have been reported in numerous studies on pool boiling with different heater's geometries, test liquids [71–76], and microgravity platforms [77–80]. These works are extensively reviewed by Mudawar et al. [81] and Colin et al. [82]. Thermocapillary stress has been speculated to be capable of transporting heat from the substrate into the bulk liquid in the absence of gravity. However, its efficiency is still far inferior compared to convection in terrestrial condition [70, 83–85]. Thus, utilizing pool boiling for outer-space thermal management remains a challenge.

Chapter 2 and Chapter 3 have introduced the oscillate boiling regime, which may provided a solution for heat transfer in micro-gravity environment. Unlike nucleate boiling, oscillate boiling is speculated to be independent of gravity as its operation does not involve the detachment and rise of the boiling bubble. Instead, the bubble remains pinned on the heating substrate and undergoes volume oscillation with a stable maximum radius. The heat is carried by a flow driven by thermocapillary stress (Marangoni flow [86]) and a streaming flow set up by the bubble oscillations [43], both of which are also independent on buoyancy. To test this concept, we have conducted experiments with substrates tilted at 30° and 90° degree relative to the horizontal direction. The results presented in Fig. 4.2 reveal that oscillate boiling retain normal operation, i.e the direction of the bubble oscillation and the flow of the pinched off microbubbles are both perpendicular to the heater surface and not affected by the direction of gravity. This is a promising result,

FIGURE 4.2: (a) Oscillate boiling on a 30° degree tilted substrate with 89 mW heating power. (b) Oscillate boiling on a 90° degree tilted substrate with 62 mW heating power. The scale bars in each figures represents 20 μm .

however, there has not been any study on the behavior of oscillate boiling in actual non-conventional gravity environment, i.e. low- g and high- g .

In this chapter, we report on the experimental observation of oscillate boiling in water both in low- g and high- g on-board parabolic flights. We then compare its behavior with results collected in terrestrial condition to draw conclusion on the effect of gravity on oscillate boiling. The experiments utilize a compact, simple, yet effective setup that can inspire future high-speed imaging experiments in low-gravity conditions. The results presented in this chapter was published as “[Nguyen, D. M., Supponen, O., Miao, J., Farhat, M., Ohl, C. D. \(2019\). Gravity-Independent Oscillate Boiling. *Microgravity Science and Technology*, 1-7.](#)” [87]¹.

¹The author, D.M.N, designed and conducted the experiments, analyzed the results and wrote the manuscript, O.S took care of safety and regulation of the campaign and reviewed the manuscript, M.J supervised the sample fabrication steps, M.F initiated the project and wrote the proposal for the campaign, C.D.O collected parts of the data, supervised the project and revised the manuscript.

FIGURE 4.3: (a) Sketch of the experimental setup where red lines represent power connections and black lines represent flow of electrical signals. (b) 3D-view of the devices used.

4.2 Materials and Methods

4.2.1 Experimental setup

Figure 4.3 describes the complete experimental setup. The test section base was a glass wafer that contained a microfabricated $15 \times 15 \mu\text{m}^2$ electrical heater made of platinum with gold electrodes wiring. The heater's design and fabrication steps were the same as described in Chapter 3 and Ref [62]. Microscope cover glasses formed the side walls of the $1 \times 1 \times 1 \text{ cm}^3$ liquid container. The container was filled

with de-ionized water with bulk temperature of $\approx 17^\circ\text{C}$. The top of the container was covered with a layer of foam to prevent any leakage of the liquid while equilibrating with the cabin pressure (≈ 0.8 bar). The electrical microheater was powered with a laser diode controller (Thorlabs LDC220) operating at constant current mode. The heater's instantaneous temperature was obtained from its resistance, which was recorded at a sampling rate of 10 MHz with 12 bits accuracy with a data acquisition (DAQ) board (model USB 2020 from MCDAC). The periodic drop of the temperature during each cycle (see below) allowed determining the bubble's oscillation frequency from the resistance signal.

The bubble oscillations were imaged with a sensitive CMOS camera (global shutter, model 1920-155um from Basler) equipped with a 5x-magnification lens (Makro lens MO-E65mm from Canon). This configuration provided a maximum temporal resolution of 1 ms and a spatial resolution of $1\ \mu\text{m}$. A typical period of the bubble oscillation is $4\ \mu\text{s}$, thus it demanded a short exposure time to capture the bubble dynamics free of motion blur. Because the minimum shutter duration of the camera was $100\ \mu\text{s}$, a strobed light source was synchronized with the camera. In particular, the camera exposure-out signal triggered a current amplifier [88] that generated a $1\ \mu\text{s}$ -long pulse for the illuminating LED (Cree CXB1304). The 1 kHz triggering signal for the camera, the current controller, and the DAQ device were controlled with a microcontroller (Teensy 3.6). It was configured with a laptop through a USB port that also stored the images from the camera and waveforms from the DAQ board. An accelerometer (Adafruit LIS3DH) connected to the laptop measured the effective gravity and provided the signal to start the experiment. A software was built by the author, written in C++ and Visual Basic to control the setup such that it can either run automatically based on gravity signal or manually by the user's interaction. The complete code is available on Github: <https://github.com/dmnguyen92/Automation-in-parabola-flights>.

Figure 4.3a shows the electrical diagram and Fig. 4.3b shows the full mechanical design of the setup. The use of a data acquisition board and a USB-connected camera instead of an oscilloscope and a high speed camera saved weight, size and

FIGURE 4.4: Real photo of the experimental setup when it is assembled on-board the parabolic air plane.

FIGURE 4.5: A typical profile of the aircraft altitude and effective gravity inside the cabin during one parabolic maneuver.

cost of the setup. The experiments enclosing dimension reduced to only $50 \times 30 \times 25 \text{ cm}^3$ with a total weight of 5 kg. The setup may be used individually or, as we operated it, attached to a separate enclosed experiment (see Fig. 4.4). Its components can be easily assembled/disassembled during transportation.

4.2.2 Experimental procedure

The experiments were conducted in November 2017 during the European Space Agency (ESA) 67th parabolic flight campaign totalling 3 flights on board the Airbus A310 Zero-G. Each flight contained 31 parabolic maneuvers, each starting with an initial high- g ($1.8 g_e$) pull-up, followed by a low- g ($0.01 g_e$) period of 22 seconds, and ending with a high- g ($1.8 g_e$) pullout. Data were obtained during normal- g , high- g , and low- g phases (Fig. 4.5). In particular, once the gravity stabilized at a certain preset level for over 3 seconds, the microcontroller initialized simultaneously the camera, LED, current controller, and data acquisition board. The electrical and imaging data were stored locally inside the acquisition board and the camera. After a run time of 0.5 s the experiment ended and the data were transmitted to the computer. All equipment were then returned to the stand-by state waiting for the next run. Besides gravity, the electrical heating power was varied between 0 mW and 120 mW to probe the parameter space.

4.3 Results and Discussions

4.3.1 Captured dynamics of oscillate boiling bubbles

Figures 4.6a and 4.6d presents the dynamics of an oscillate boiling bubble at low- g and high- g driven at similar electrical powers of 63.8 mW and 64.1 mW, respectively. The resulting maximum radius at low- g is $13.2 \mu\text{m}$ and at high- g is $13.6 \mu\text{m}$. The bubble's dynamics are the same as in terrestrial condition (see Fig. 2.2 and Fig. 3.6). During their oscillations, both bubbles release smaller microbubbles at their apex, which are transported in a direction perpendicular to the substrate independent on the strength of gravity. Their oscillation is controlled only by the power supply and both stop once the electrical current is switched off after 0.5 s. The recorded microheater's temperature is presented in Fig. 4.6b and 4.6e. When the bubble collapses, a liquid jet impacts onto the substrate and rapidly cools the heater:

FIGURE 4.6: Captured bubble dynamics of oscillate boiling bubbles, the temperature of the heating substrate and its frequency spectrogram in: (a-c) low- g condition and (d-f) high- g condition. Images from subfigure a and d are taken from Video 1 and Video 2. The scale bar in subfigure a,d represents $20\ \mu\text{m}$, the time difference between two consecutive frames is $1\ \text{ms}$ and the arrow indicate the orientation of the effective gravity inside the cabin. The high frequency noises that appears every $1\ \text{ms}$ in subfigure c and f is induced by the current driving the LED at $1\ \text{kHz}$.

within less than $0.5\ \mu\text{s}$, the heater's temperature drops from $\approx 225\ ^\circ\text{C}$ to $\approx 185\ ^\circ\text{C}$, which is a cooling rate of $\approx 10^8\ \text{K/s}$. Having received thermal energy from the vaporization of the liquid, the bubble re-expands and creates a dry spot below on the heater. The heater thus recovers to its peak temperature and the cycle repeats. A Fourier analysis of the temperature signal reveals the oscillation frequencies of the bubbles in the two gravity conditions, namely $305\ \text{kHz}$ and $310\ \text{kHz}$ (Fig. 4.6c and 4.6f).

4.3.2 Parameters study

By changing the electrical power, three regimes of oscillate boiling can be achieved. Below a threshold power the heater is not able to nucleate a bubble. Above $40\ \text{mW}$ a bubble starts forming at the heating spot and undergoes unstable oscillation. In this unstable mode, the bubbles show neither microbubble pinch-off nor periodic

FIGURE 4.7: Three oscillate boiling regimes at different input power and gravity conditions. The height of the column represents the mean power and the error bar represents the standard deviation of the sample. Each columns contained at least 8 data points.

cooling of the heater. After ≈ 10 ms, the oscillations halt and the bubble grows similar to a conventional nucleate boiling bubble (as discussed in Section 3.4). Once the input power surpasses 60 mW, stable oscillation featuring microbubble pinch off and microbubble streaming together with the periodic cooling of the heater is observed, see Fig. 2.2. Figure 4.7 categorizes the observed oscillate boiling regimes and plots their electrical current as a function of gravity. Overall we find very similar behavior with no clear dependency on gravity.

In the stable oscillate boiling mode, the oscillation frequency decreases monotonically with increasing maximum bubble radius, which has been predicted with the model proposed in Section 2.4 and 3.4. As shown in Fig. 2.5, each input power results in a stable's maximum radius and resonant frequency. Here the same model is applied to derive the radius-frequency relationship when the input power varies in different gravity conditions. The results presented in Fig. 4.8 show that while there is some scatter, we find overall quantitative agreement with the model (black line in Fig. 4.8) and its independence of gravity.

FIGURE 4.8: The frequency-radius characteristics of oscillate boiling bubble in different gravity conditions. The simulation data follows the calculation reported in Section 2.4 and reference [43] with a change of ambient pressure to 0.8 bar, matching the cabin’s pressure. All data points share the same experimental error and is indicated by the error bar plotted in the bottom-left corner.

4.3.3 Discussion

The analysis of the bubble’s dynamics and its oscillation frequency shown in Figs. 4.7 and 4.8 demonstrate that its quantitative and qualitative characteristics are independent of the effective gravity conditions: high- g , low- g , and normal g . These bubble’s behaviors might indicate the independence of the oscillate boiling regime on gravity within the ranges tested. We attribute this characteristics to the governing physics: the oscillation is driven by the bubble’s inertia and the liquid jet impact, the rise of the microbubble stream is driven by the Marangoni flow along the bubble’s interface and likely by a streaming flow caused by the bubble oscillation. These driving factors are independent of buoyancy. The experimental verification not only demonstrates the independence on gravity but also confirm that the previously proposed explanations and model in Chapter 2 and Ref. [43] are sound.

Within our system, the working capacity of oscillate boiling is limited by the heater. Above a certain input power, the heater breaks, likely because of thermal stress and mechanical stress induced by the bubble’s collapse. The maximum power achieved

in this experiment is 120 mW with a $15 \times 15 \mu\text{m}^2$ heater size, corresponding to a heat flux of 10^8 W/m^2 . This number is significantly higher as compared to previously reported critical heat fluxes of 10^5 W/m^2 for water [68], 10^6 W/m^2 for FC-72 [70], and nPFH [79, 89]. This may be partly explained that oscillate boiling operates above the critical Leidenfrost temperature (see Fig. 3.8).

One limiting factor of oscillate boiling is that the size of the heat source must be smaller than $20 \mu\text{m}$ [43, 62]. Currently only miniature devices such as high power transistors or microfluidic platforms may benefit from this boiling mode. The next challenge is to scale-up the regime, thus to widen the range of application.

4.4 Conclusion

Experiments in various gravity conditions on-board parabolic flights confirm the independence of the oscillate boiling bubble on gravity. In low- g and high- g conditions, oscillate boiling bubble retains its characteristic behavior and operation limits. The gravity-free feature together with its high thermal efficiency encourage the development of oscillate-boiling-based heat transfer devices for applications in terrestrial and outer space environments. To achieve this, the driving, packing, and the interaction of many heaters and oscillate boiling bubbles should be studied.

Chapter 5

Interactions Between Two Oscillate Boiling Bubbles

5.1 Introduction

Chapter 2 presents the first observations and physical mechanism of oscillate boiling, Chapter 3 brings the phenomenon one step closer towards application by introducing the electrical microheater and Chapter 4 presents the potential of oscillate boiling for outer space thermal application. Yet, in those studies, only one single, microsize bubble was created. This is not beneficial for practical use where boiling takes place at a much greater scale. Thus, towards application, one needs to scale-up the system and study the interactions between multiple oscillate boiling bubbles.

In fact, the study of many-bubble system has received a lot of research interest [90–104], yet, the specific interaction between two oscillating bubbles remains an experimental challenge [90, 94, 98, 101–103]. Besides experimental complexity, a more fundamental problem is that the acoustic interaction of two bubbles is clouded by the external sound field driving the bubbles into oscillations. So far all studies on this interaction and synchronization between nonlinear bubble oscillators are

obscured by the presence of the external forcing, which prescribes a phase and frequency for both oscillators [105]. Oscillate boiling, however, can avoid this issue as it is not driven by acoustic wave but instead by the constant heat sources. By putting them next to each other, they become nonlinear oscillators that only through their mutual interaction obtain an in-phase synchronization [106] at their self-selected frequency.

In this chapter, we present the study on the mutual interaction of two oscillate boiling bubbles placed in close proximity. Here the frequency and phase of oscillations are not prescribed by an external source, but the auto-oscillator, i.e. each bubble, is free to choose. The experimental control parameters are the distance and the electrical power supplied to the heaters. The coupled oscillations are characterized with high-speed photography, recording of the acoustic emissions, and instantaneous temperature measurements. The results presented in this chapter was published as “Nguyen, D. M., Sanathanan, M. S., Miao, J., Rivas, D. F., Ohl, C. D. (2019). In-phase synchronization between two auto-oscillating bubbles. *Physical Review Fluids*, 4(4), 043601.” [107] ¹.

5.2 Materials and Methods

The electrical heater design for two bubbles follows Chapter 3 and Ref. [108], i.e. with a 50 nm thin film of platinum deposited on a glass substrate and connected to a 600 nm thick gold layer supplying the electrical current (see Fig. 5.1a). The electrical connections and locations of the generated bubbles are sketched in Fig. 5.1b. The two heaters have an area of $10 \times 10 \mu\text{m}^2$ each and are connected in series. The electrical power is provided from a constant electrical current supply (Thorlabs LDC 220). While the power delivered to each heater should theoretically be the same, we observe small deviations as their individual resistance are not strictly

¹The author, D.M.N designed and conducted the experiment, analyzed the results, did the numerical simulation and wrote the manuscript, M.S.S fabricated the sample and reviewed the manuscript, M.J supervised the sample fabrication steps, D.F.R advised on the sample design and reviewed the manuscript, C.D.O supervised the project and revised the manuscript.

FIGURE 5.1: (a) Top view of the electrical microheater. The scale bars represents $20\ \mu\text{m}$. (b) Sketch of the experiment to generate and monitor two auto oscillating bubbles on two closely-placed electrical micro-heaters.

equal. During the experiment the voltages across each heater and the current I are monitored with a 12-bit oscilloscope (Lecroy HRO 64Zi). The memory of the oscilloscope allows to monitor the instantaneous temperature and power of each heater for several 100,000 oscillations.

The glass chip with the heaters is attached to the bottom of an acrylic container ($10 \times 10 \times 10\ \text{mm}^3$) forming the test section. Two faces of the container are replaced with microscopic glass slides for better imaging with the high-speed camera (Photron SA-X2) running at 300,000 frames per second (fps). A long distant objective lens (20x PTEM M Plan Apo, Edmund Optics) provides a final resolution of $1\ \mu\text{m}$ per camera pixel. Besides the electrical signals, also the acoustic emission from the bubble oscillation is recorded with a hydrophone (ONDA HNR1000). The timing of the experimental devices is synchronized with a digital pulse generator (BNC 575, Berkeley Nucleonics). The detailed setup as depicted in Fig. 3.5a.

In Section 3.4, it was found that an individual oscillate boiling bubble may either oscillate stable for many million cycles or stops oscillating within tens of milliseconds [108]. The *stable oscillate boiling* mode is only found above a certain power threshold which in turn depends on the size of the heater, i.e. $20\ \text{mW}$ for $10 \times 10\ \mu\text{m}^2$ heater. In each cycle the bubble pinches off a microbubble from its apex and rapidly cools the heater with a liquid jet impact during the bubble collapse. The other mode termed *unstable oscillate boiling* lacks the pinch off and

therefore due to rectified diffusion the bubble grows steadily. With time, the frequency of oscillation decreases and eventually the oscillation ceases. Due to the missing jet impact, the temperature of the heater remains approximately constant over a cycle. In this work, all experiments are conducted at an electrical power of more than 20 mW. For an individual bubble this would always result in the stable oscillating mode.

To limit thermal stress and damage to the heaters, we run the experiment for a short time of up to only 0.5 s.

5.3 Experimental Results

5.3.1 In-phase synchronization

We report first the bubble dynamics at an intermediate distance of $40\ \mu\text{m}$ and at a relative high electrical power of approximately 35 mW for each heater. The findings are summarized in Fig. 5.2. On the top, Fig. 5.2a, selected frames in the upper row depict the oscillations of the two bubbles recorded shortly after the start of the experiment and in the row below a few milliseconds later. Due to small differences in the heaters the two bubbles have a slightly different size and nucleation time, resulting into different oscillation periods and phases of oscillations. Over time, however, the two bubbles synchronize their oscillations and they both reach in each cycle a maximum radius of approximately $10\ \mu\text{m}$. The temporal development leading to this synchronization is revealed in the spectrogram of their acoustic emission below. There in Fig. 5.2b around $t = 1.5\ \text{ms}$, two distinct frequencies and their higher harmonics are noticeable. The initially (on average) smaller left bubble oscillates at a higher frequency (above 200 kHz), while the larger right bubble oscillates at the lower frequency (below 200 kHz). From $t = 3.5\ \text{ms}$, as indicated with the vertical dashed line, both bubbles synchronize into a single frequency. Thereafter the frequency remains about constant before it steadily decreases over time. The equivalent radius (to that of a hemisphere) of both bubbles is plotted

FIGURE 5.2: Two synchronized oscillate boiling bubbles. Here the distance between heaters is $40\mu\text{m}$. The power of the left and right heater are 34.9mW and 36.0mW respectively. (a) Captured images before and during the time when two bubbles synchronize. A digital filter was applied to remove dirt in the background. Please refer to Video 1 for the raw data. The time between two consecutive frames is $3.33\mu\text{s}$. The scale bar shows $20\mu\text{m}$. (b) The spectrogram of the acoustic signal emitted from two synchronized oscillate boiling bubbles. (c) The distance of the bubble's center of mass and their effective radius retrieved from the recording. (d) The correlation coefficient between the effective radius of the two bubbles, calculated with a window time of $100\mu\text{s}$, i.e. 30 frames of the recordings.

FIGURE 5.3: The spectrograms of: (a) emitted acoustic signal (b) temperature of the left heater and (c) temperature of the right heater when two bubbles synchronize. These results belong to the same experiment with the one presented in Fig. 5.2.

in Fig. 5.2c in green and blue. Initially, at time $t = 1$ ms, their radii differ in phase, but from $t = 3.5$ ms the smaller bubble (blue line) follows the phase of the larger bubble and their time series become nearly indistinguishable (see inset in Fig. 5.2c). In other experiments, we observe some differences between the bubble's radii, yet their phases are still perfectly synchronized. Both bubbles then increase slowly in size, which is consistent with the decrease in frequency seen in Fig. 5.2b. Due to this growth, the liquid contact lines expand. Yet instead of a symmetrical expansion they move closer towards each other and thereby decreasing their center of mass distance (red line in Fig. 5.2c). This movement can be explained by the attractive secondary Bjerknes force for in-phase oscillating bubbles [93] and by the spreading of the heating region (see Discussions). The in-phase synchronization of the two bubbles is a remarkable dynamic feature, which is nicely confirmed by plotting the correlation coefficient of their instantaneous radius, see Fig. 5.2d.

FIGURE 5.4: Two stable oscillate boiling bubbles. Here the distance between two heaters is $50\mu\text{m}$. The power of the left and right heater are 25.4mW and 25.1mW respectively. (a) Captured images of the bubble dynamics. The sizes of the pinch-off bubbles are close to the optical resolution so they are barely noticeable. The time between two consecutive frames is $3.33\mu\text{s}$. The scale bar represents $20\mu\text{m}$. (b) The spectrogram of the acoustic signal emitted from the two bubbles. (c) The distance of the bubble's center of mass and their effective radius retrieved from the recording during the period marked in Fig.b. The inset shows the zoom-in data of the black rectangle.

One important observations is that in the synchronization case, both bubbles oscillate in the *unstable mode*. This is indicated by the absence of both the microbubble pinch-off, see Fig. 5.2a, and the periodic temperature oscillation of the heater, which is visible from the spectrogram of the heater's thermal readings (see Fig. 5.3). In a later discussion, it will be revealed that the oscillation mode of each individual bubble is directly related to its interactive behavior.

5.3.2 Other different interactive behaviors

By changing the distance and electric power, qualitative different behavior of the two-bubble system can be observed. We start with bubbles separated further apart

FIGURE 5.5: The spectrograms of: (a) emitted acoustic signal (b) temperature of the left heater and (c) temperature of the right heater when two bubbles oscillate independently. These results belong to the same experiment with the one presented in Fig. 5.4.

at a distance of $50\ \mu\text{m}$, see Fig. 5.4. Unlike the synchronization case, here both bubbles operate in the stable oscillating mode, i.e. with microbubble pinch-off. With the start of the electrical current, the two vapor bubbles start oscillating and each reaches a stable maximum radius at a constant frequency. During each cycle they release a microbubble from their apexes and induce a rapid cooling on their corresponding heater. Both the acoustic and thermal readings spectrogram reveals a superposition of two distinct frequencies (see Fig. 5.5), an indication that the two bubbles do not synchronize. As a result, the time-series of their bubble radii, presented in Fig. 5.4c, are uncorrelated.

By increasing the heating power, the system reveals a third regime: the bubbles oscillate only for a few cycles while their average radius grows with time before they eventually merge. Here, the oscillation is unstable with no microbubble pinch-off, no periodic cooling of the heater, and no synchronization. This radically different regime is reflected in the images and the spectrogram of the acoustic signal shown in Fig. 5.6.

FIGURE 5.6: Two bubbles grow and merge. Here the distance between two heaters is $40\mu\text{m}$. The power of the left and right heater are 48.0mW and 47.7mW respectively. (a) Captured images when two bubbles grow and merge. The time between two consecutive frames is $33.3\mu\text{s}$. The scale bar shows $20\mu\text{m}$. (b) The spectrogram of the acoustic signal emitted from two growing bubbles. (c) The distance of the bubble's center of mass and their effective radius retrieved from the recording.

5.3.3 Parameters space

As previously mentioned, we find that the dynamics of two neighboring oscillating bubbles fall in one of three distinct regimes. Bubbles sufficiently close together and driven at an intermediate power synchronize perfectly, bubbles further apart oscillate independently, while further increasing the power at short distance results in the merging of the bubbles. Figure 5.7 reveals the regions in the parameter plot where in-phase synchronization is achieved, i.e. distances between $40\mu\text{m}$ and $50\mu\text{m}$ and powers between 32mW and 42mW . At larger distance most bubbles fall into the independent oscillating regime, while they merge at closer distance and at high driving.

FIGURE 5.7: Parameter space of two nearby oscillate boiling bubble as a function of different heating power and distance between heaters. All data points share the same error bar, which is plotted separately. The classification region is constructed using k-nearest neighbors algorithm [109] with $k=5$.

The classification region in Fig. 5.7 is calculated using k-nearest neighbors algorithm [109] with $k=5$. Prior to the calculation, each attribute of the data set is normalized. For example, each heating power data P_i is normalized to P'_i as:

$$P'_i = \frac{P_i - P_{mean}}{\sigma_P} \quad (5.1)$$

where P_{mean} and σ_P are the mean and standard deviation of the heating power data set. The same procedure is applied for each heater's distance data point d_i . As a results, two sets of normalized, non-dimensional data P'_i and d'_i are obtained. The distance between two arbitrary points A and B on Fig. 5.7 is then calculated as:

$$D_{AB} = \sqrt{(P'_A - P'_B)^2 + (d'_A - d'_B)^2} \quad (5.2)$$

This distance is then used to find the 'nearest' data points. The value $k = 5$ is chosen as it is both the smallest value that leads to the formation of three distinct regions in the parameters space and also gives the highest accuracy.

5.4 Explanation of different synchronization modes

5.4.1 Why only unstable oscillate boiling bubbles synchronize?

The two bubbles couple through the flow field and the acoustic emissions. To understand the physics of the process we use a simple model based on potential flow which ignores the boundary layers and finite sound speed of the two bubble system [93, 110]. The solution of the problem leads to a pressure interaction term p_i between two neighboring (hemi)spherical bubbles in the Keller-Miksis equation:

$$p_i = \rho d^{-1} (R_i^2 \ddot{R}_i + 2R_i \dot{R}_i^2) \quad (5.3)$$

where R is the bubble radius, d the distance between two bubbles and ρ is the liquid density. The detailed derivation of Eqn. 5.3 is presented in Ref. [93]. In stable oscillate boiling the liquid jet impact prematurely disrupt the bubble's natural collapse, causing rapid vaporization and rapid re-expansion of the bubble driven by the expanding vapor (see Chapter 2, 3 and Ref. [43, 108]). The pressure interaction from the neighboring bubble is therefore not sufficient to overcome this violent mechanism. In contrast, the unstable mode is driven by a more gentle bubble's inertia and can be affected by the acoustic emission.

To support this hypothesis, we extend the model presented in Section 2.4 and Section 3.4 with the interaction term p_i . Equation 5.3 serves as an additional term on the Keller-Miksis equation for each individual bubble. Equation 2.1 for bubble i thus becomes:

$$\left(1 - \frac{\dot{R}_i}{c}\right) R_i \ddot{R}_i + \frac{3}{2} \dot{R}_i^2 \left(1 - \frac{\dot{R}_i}{3c}\right) = \left(1 + \frac{\dot{R}_i}{c}\right) \frac{p_l}{\rho} + \frac{R_i}{\rho c} \frac{dp_l}{dt} - \frac{1}{d} (R_j^2 \ddot{R}_j + 2R_j \dot{R}_j^2) \quad (5.4)$$

FIGURE 5.8: Numerical calculation of two closely-placed bubble which both of them operating in (a) stable oscillate boiling mode with liquid jet impact and (b) unstable oscillate boiling mode without liquid jet impact. Here the power supplied to bubble 1 is 34 mW and to bubble 2 is 30 mW and their distance is 40 μm .

For the unknowns R_i and temperature T_i with $i = 1, 2$ numbering the bubble, we solve the coupled Keller-Miksis equation (Eqn. 5.4) and energy equation for the gas/vapor inside the bubbles. To mimic the thermal kick received during the jet impact in stable oscillate boiling, the input power is approximated with a step function. More specifically, the bubble receives the full power from the heater if its radius is smaller than a certain threshold (during the jet impact) and otherwise this term is zero. The value of this threshold can be derived from the power law Eqn. (6.1) in Ref. [49]. For the unstable mode without the jet, a slower energy transport is represented with a smoothed function where the input power is inversely proportional to the instantaneous bubble's radius.

Figure 5.8 presents the radii versus time curves for the strongly collapsing (stable) and mildly oscillating (unstable) bubbles when the interaction term is switched

on. In the strong oscillation regime, see Fig. 5.8a, the two bubbles oscillate each at a different frequency and therefore their phases drift with time. When both bubbles oscillate at a smaller amplitude, Fig. 5.8b, they oscillate synchronously with the same frequency after a brief transient. The model provides qualitative similar features of independent and synchronous oscillations as in the experiments described above. The model also predicts that the bubble oscillation frequency in the coupled mode is lower than in the independent mode. Yet for a more complete understanding, the mass transport into the bubble through rectified diffusion should be accounted for.

5.4.2 The connections between bubble's distance, heating power and synchronization mode

Placing two heaters in close proximity results in the spreading of some of the heat between the heaters in the substrate. Thereby the effective microheater size is enlarged as compared to the isolated microheater. In Section 3.5 and Ref. [108] we reported that the regime of stable oscillate-boiling requires a higher power when the heater size is increased, i.e. from $10 \times 10 \mu\text{m}^2$ to $15 \times 15 \mu\text{m}^2$. Thus, while at a certain power an individual heater operates in the stable oscillate-boiling regime, it shifts to the unstable regime when accompanied by a second microheater, which then leads to synchronization. At even higher electrical power we also observed the merging of two bubbles. Besides the secondary Bjerknes force, a more subtle explanation may be that this thermal spreading forms a heat bridge between the heaters and merge them into one big heating spot.

To support this claim, we conducted a numerical simulation on the thermal profile of the heating substrate. Figure 5.9 show the geometry of the simulation domain. To reduce computational cost, this domain only includes a small area ($500 \mu\text{m} \times 0.5 \mu\text{m}$) surrounding the heaters. The thickness of the water column is $100 \mu\text{m}$ and of the glass substrate is $30 \mu\text{m}$. To solve for the thermal profile, first the current density $\mathbf{J}$ in the circuit is calculated:

FIGURE 5.9: Geometry of the simulation for the thermal profile of the heating substrates

$$\mathbf{J} = \sigma_e \mathbf{E} = -\sigma_e \nabla V \quad (5.5)$$

Here σ_e is the electrical conductivity of the material, $\mathbf{E}$ is the vector electric field, which is obtained from the electrical potential V . The current density is then used to calculate the Joule's heating per unit volume:

$$P_{Joule} = \mathbf{J} \cdot \mathbf{J} / \sigma \quad (5.6)$$

which is latter used as the heat source in the thermal energy equation to solve for temperature:

$$\rho C \frac{\partial T}{\partial t} - \nabla \cdot (k \nabla T) = P_{Joule} \quad (5.7)$$

Here ρ , C and k is the material's density, heat capacity and thermal conductivity, respectively. Equation 5.5, 5.6, 5.7 is solved at each time step using finite elements method with commercial software COMSOL Multiphysics. The temperature at

FIGURE 5.10: Temperature distribution of the substrate at 1ms after the current is trigger. Here each heater is operated at 35mW and placed at four different distance following the experiment. (a)-(b) Top-view of the the thermal profile of the substrate with the longest and shortest distance. (c)-(f) Temperature distribution along the white line with different distance of the heaters.

the boundary is set at room temperature. The electrical potential at one side of the gold electrode is set to fit the experimental values, and at the other side is set to ground.

Figure. 5.10 shows the simulated thermal profile of the heater at 1 ms after the current is triggered. This is the typical time scale for the bubbles to settle at either unstable or stable oscillation. Here we approximate the boiling point of water, i.e. 100°C to be a threshold above which the heater becomes a nucleation spot that favors the formation of bubbles. It can be seen that as the heaters are

closer to each other, the thermal spreading effect becomes more significant and enlarges the effective nucleation size. The unstable regime is therefore more likely to occur. When the distance reduces to $d = 30\mu\text{m}$, the heating regions connect, giving the bubbles a tendency to merge. This thermal spreading effect may explain why the parameter region of synchronization is limited to intermediate distances and intermediate heating powers (see Fig. 5.7).

5.4.3 The benefits of non-synchronicity for thermal application

Chapter 2 and Chapter 3 have shown that oscillate boiling has improved heat transfer efficiency as compared to nucleate boiling. Towards application, the oscillate boiling phenomenon needs to be scaled up/numbered up to multi-oscillate-boiling bubbles covering the whole heating floor. Our results show that if placed sufficiently closed, those bubbles might synchronize, but then shortly after they will grow, merge and become just nucleate boiling bubbles. This severely limits the intended outcome. To maintain a system of longer operation time, oscillate boiling should be in the stable mode (with microbubble streaming, periodic cooling and no synchronization). The results presented in Fig. 5.7 provides the design criteria for the scaled-up heating device, i.e. the suitable distance between individual heating elements, maximum operating power, etc.

5.5 Conclusion

In summary, the self-driven nature of oscillate boiling allowed for the first time to resolve the mutual interactions of two oscillating bubbles in absence of an external field. Thus without imposing a frequency or phase the bubbles obtain in-phase synchronization. Besides we have identified two more interaction regimes: independent oscillations at far distances and merging for small distances and strong heating. Our main finding is that only mildly oscillating bubbles synchronize, while large

amplitude oscillations result in independent oscillations. The former are driven by a lower peak power, while the latter are the result of higher power impulsive kicks. If we extrapolate our results to acoustic driven bubbles, it would suggest that bubbles at moderate driving may synchronize mutually, while at higher amplitude they follow the external sound field at their specific phase. If that holds for continuous driving remains to be shown. The results also provides design criteria in term of heating power and distance for scaling up this phenomenon for thermal application, in which the heating floor will be covered by many independent, non-synchronized oscillate boiling bubbles.

Chapter 6

Conclusions and Outlooks

6.1 Conclusions

In this thesis I present a possible route to overcome one of the biggest challenges in heat transfer – the boiling crisis – with a novel approach termed *oscillate boiling*. The thesis starts with the very first experimental observation of the phenomenon, followed by models of the underlying physics, to searching routes to bring the phenomenon closer to some later applications.

Chapter 2 shows the first experimental observations of the oscillate boiling regime. By focusing a continuous laser beam with a power range of 50-100 mW onto a $10 \times 10 \mu\text{m}^2$ region of a gold-coated substrate, we create a surface-attached bubble that experiences clock-like volume oscillation at several hundreds kHz and maximum radius of $\approx 15 \mu\text{m}$. In each cycle, a small microbubble is pinched off from the main bubble. We explain this behavior with the cyclical non-spherical bubble collapse that creates a liquid jet impacting onto the heated substrate. Direct contact between the liquid jet and the heater supply sufficient heat for the bubble to remain oscillating. This dynamics is explained with a simplified model of a thermally kicked-oscillator, and in the later with the cyclic spiking of the heater temperature (see Fig. 3.8). The temperature measurement of the surrounding

liquid and the model on the recirculating Marangoni flow indicates an improved heat transfer efficiency of oscillate boiling as compared to nucleate boiling.

In **Chapter 3**, we replaced laser based heater with an electrical heater. This step is crucial for potential application of the phenomenon. With the electrical heater, we resolve two different modes of the oscillate boiling regime. The first mode named *unstable oscillate boiling* takes place at lower heating power. In this mode, the main bubble does not release the microbubbles, there is no liquid jet impact onto the heater and the bubble's average volume increases over time due to accumulation of non-condensable gas. After a few millisecond the oscillation stops and the bubble grows thermally. The second mode, named *stable oscillate boiling*, is achieved at higher heating power. In this mode, frequent microbubble pinch-off and liquid jet impact occur and lead to an oscillation with a constant amplitude and frequency similar to the laser based heating.

Chapter 4 test a potential advantage of oscillate boiling as compared to nucleate boiling. While nucleate boiling relies on gravity to facilitate the detachment and rise of the boiling bubble, oscillate boiling depends solely on the Marangoni flow and is therefore independent of gravity. To confirm this hypothesis, we conduct experiment on-board parabolic flights and test oscillate boiling in different gravity condition. The results show that the oscillate boiling bubble retains its characteristic behavior and operation limits regardless of the direction and magnitude of gravity.

In the last chapter, **Chapter 5**, techniques to scale-up the phenomenon are presented by studying the interaction between two oscillate boiling bubbles. This is also the first study that resolves the dynamics of closely-placed oscillating bubbles without imposing external acoustic driving. Depending on the distance between two bubbles and their heating power, three regimes can be observed: growth and merge, independently oscillation, and most interestingly, synchronization. The important finding is that synchronization only takes place at intermediate distance

FIGURE 6.1: (a) One single oscillate boiling bubble, which is the object of the current study. (b) The ultimate goal: a heating floor fully covered with oscillate boiling bubbles

and power and both bubbles must oscillate in the unstable mode. Only in the unstable mode that the acoustic interaction between oscillating bubbles can overcome their individual thermal kicks. At higher power, the thermal kick is more violent, causing both bubbles to be in the stable mode and as a result, they oscillate independently. These findings provide the design criteria to scale up the phenomenon for thermal application.

6.2 Outlooks

Oscillate boiling is a newly found regime with many interesting features left unexplored. Our studies presented in this thesis have established a fundamental foundation for the regime. Based on these results, there can be many directions for development, especially towards commercializing this phenomenon.

6.2.1 Scaling up the oscillate boiling regime

The object in our studies are mostly single or double oscillate boiling bubbles, covering a heating area of approximately $20 \times 60 \mu\text{m}^2$ (see Fig. 6.1a). While

FIGURE 6.2: (a,b) Schematic design of the (a) 1×3 heaters and (b) 3×3 heaters. (c) Side-view recordings of the 1×3 heater. (d) Top-view recordings of the 3×3 heater. In (c) and (d), the frames are captured at 250,000 fps and the scale bar represents $20 \mu\text{m}$.

this can be beneficial for cooling of high-power miniature chips, this scale is not beneficial for conventional heating application whose sizes are ranging from $10 \times 10 \text{ cm}^2$ to $10 \times 10 \text{ m}^2$. Thus, our ultimate goal is being able to cover the extended heating floor with oscillate boiling bubbles, as presented in Fig. 6.1b.

The results presented in Chapter 5 provide some design criteria in term of heater's dimension and heating power to produce multiple long-lasting oscillate boiling bubbles. Based on this criteria, we have attempted different heater configuration-matrices, namely 1×3 and 3×3 and the preliminary results are presented in Fig. 6.2. Here, the inter-row and inter-column distances are $60 \mu\text{m}$ and the average power per heater is 40 mW. Figure 5.7 predicts that this setting should produce independent oscillate boiling bubbles and it is indeed the case as we see no correlation among the bubble dynamics; they all oscillate in the stable mode. Yet, the effects of their interactions are clearly visible as the radius of the central bubbles are almost twice as large as the side bubbles and the microbubble stream has the

tendency to move towards the center. A more rigorous analysis is required to clarify if other findings from the two-bubbles system remain the same for multi-bubble system.

Many interesting ideas can be tested in this experiment, which will also come with technical challenges. The most simple one is to use a hydrophone to gain insight on the frequencies of these oscillations. However, this can be challenging since with so many bubble involved, their individual frequencies might be difficult to resolve from just one single acoustic signal. One possible way out is to redesign the circuit so that each heater can be individually controlled and thermally probed. The frequency of each bubble can then be inferred from their corresponding temperature signal. This, however, will require a smart and rigorous design of the heater and the electrical control system. The electrical circuit can be very complex as each individual heater will require a separate line, yet it also needs to be highly packed to be compatible with the small scale of the devices. The electrical source will pose another problem. Currently the heater is powered by an laser-diode controller, which provides a very stable current with precise control. However, this devices, similar to many other high-precision instruments, come at high cost and low power capacity. For scaling up purpose, we will need to seek for a replacement that can balance quality, capacity and cost. Once these problems are resolved, one can further increase the array dimension to 4×4 or 5×5 to see a clearer effect of scaling up.

6.2.2 Recovery from system failure

One critical functions of a commercial device is the ability to recover from a system failure. While this may seem trivial to nucleate boiling, it is not the case for oscillate boiling. The reason is that once the electrical power drop, a stable oscillating bubble can becomes unstable and grow on top of the initial hot heater. Yet, there is not a simple way back, i.e. once a bubble start growing, it cannot go back to stable oscillation even if the heating returns. Figure 6.3 describes this process.

FIGURE 6.3: (a) Recordings of oscillate boiling bubble before the power disruption (the grey dash line). (b) Recordings when the bubble starts growing after the disruption (the black dash line). (c) The heater's power profile. Above the red line, the bubble is in stable oscillation. Below the green line, there is no bubble. In between, the bubble is in unstable oscillation. (d) The spectrogram of the emitted acoustical signal. (e) The bubble's effective radius that is derived from the recordings.

When the power is dropped from the stable oscillation regime to below-bubble-formation threshold for 2 ms, the bubble stops oscillating and continue to grow even after the power comes back.

Interestingly, we repetitively observe a possible route out of this recovery problem. When the power is disrupted for 1.5 ms, the bubble transits into unstable oscillation, accumulates additional gas such that its volume grows. However, when the power returns, the main bubble, for currently-unknown reasons, manages to releases all the extra accumulated gas and recovers to the stable oscillation state. This extra gas appears in the form of an exceptionally large microbubble hovering above the main bubble (see Fig. 6.4a). Affected by the emitted acoustic wave, the released bubble also starts oscillating synchronously with the original bubble. After approximately 1 ms, the oscillation stops and this large bubble is driven away, likely by the Marangoni flow. This observation suggests that the recovery from a power loss may depend on its pattern and duration. What patterns might that be, and what is the underlying physics behind such behaviors are surely important questions that should be followed by research.

6.2.3 Full CFD simulation

This thesis has presented two major numerical models: The first model makes use of Keller-Miksis equation to simulate the thermal kick that explains the bubble dynamics. The second model simulate the thermocapillary stress to calculate liquid flow profile surrounding the bubble. While these models manage to capture the characteristic behavior of the system, they are based on rather limiting simplification. To obtain better insight, one needs to model the fluid mechanics more carefully. A complete model should take into account heat transfer of the bubble's inner (gas/vapor) and interface, mass transfer, surface tension gradients, complex free boundaries changing topology. A very nice framework that one can adopt to tackle this problem is OpenFOAM, a CFD open-source libraries written in C++

FIGURE 6.4: (a) Recordings of oscillate boiling bubble before the power disruption (the grey dash line). (b) Recordings when the bubble starts growing after the disruption (the black dash line). (c) The heater's power profile. Above the red line, the bubble is in stable oscillation. Below the green line, there is no bubble. In between, the bubble is in unstable oscillation. (d) The spectrogram of the emitted acoustical signal. (e) The bubble's effective radius that is derived from the recordings.

[111]. This framework makes use of the volume of fluid method to solve for the compressible Navier-Stokes equation. It takes into account surface tension, viscosity and phase-change and has been widely used and recognized in academic research for its high accuracy and ease of usage (for example, see [112–115]). Thus, creating a better CFD model to simulate oscillate boiling bubble would be an exciting route to go.

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