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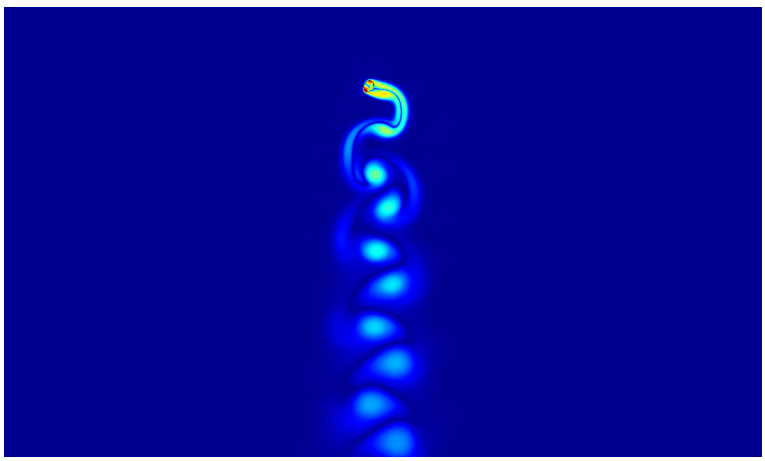

Numerical frameworks for small-scale bubble dynamics

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Department of Mechanics and Maritime Sciences
CHALMERS UNIVERSITY OF TECHNOLOGY
Göteborg, Sweden 2020

THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING IN THERMO AND FLUID DYNAMICS

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Contours of the absolute vorticity field for a zigzagging rising bubble in a quiescent liquid.

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Abstract

Designing efficient bubbly flow systems requires the prediction of the dynamics of the bubbles, the liquid and how the gas and liquid phases interact. Currently, the complex dynamics in bubbly flows are not yet fully explained, and we rely on incomplete models in our numerical methods. A major concern when investigating bubbly flows using numerical methods is the large range of length and time scales. The length scales may vary from nanometers, for the formation of vapor bubbles, to tens of meters, when system-size bubbly flow structures are induced. To predict the dynamics of the entire system, it is important to understand the phenomena at other scales, such as the evolution of vapor bubbles or the dynamics of individual bubbles.

In this PhD project, we aim at increasing our knowledge about the bubbly flow dynamics and developing numerical methods for investigation bubbly flows across all relevant length scales. So far into the project, we have focused on the small scale bubble dynamics where small scales refer to length scales of individual bubbles and below. We start by studying the evolution process of vapor bubbles by developing a multiphase DNS framework, and a less computationally expensive 1D framework, that resolve the conditions in both phases and takes into account phase change and thermal effects. These frameworks can be used to study both boiling and cavitation processes, and we use it to investigate the challenging case of laser-induced thermocavitation bubbles. These bubbles are studied as a promising tool to achieve good control in the process of crystallization. We simulate such bubbles and identify plausible mechanisms behind experimentally observed crystallization events and provide guidelines for appropriate setups to attain conditions favorable for crystallization.

Then, we shift the focus to investigate rising bubble dynamics at small scales. For this purpose, we develop an efficient multiphase DNS framework with a moving reference frame (MRF) technique that follows the bubbles. This method significantly reduces the size of the computational domain and eliminate the need for a priori estimations of sufficient domain sizes to capture the bubble dynamics. With the MRF method, we aim at obtaining the closures for bubble dynamics at small scales and use them to investigate bubbly flows up to industrial scales in the continuation of the project.

Keywords: Bubbly flows, DNS, multiphase, phase change, moving reference frame, laser-induced, cavitation

To my family

Acknowledgements

I would like to begin by thanking my supervisor Gaetano Sardina and my examiner Srdjan Sasic for granting me the very possibility to pursue my PhD in this complex and fascinating subject. Without their excellent supervision and support this thesis would not be possible. I would also like to thank my co-supervisors Henrik Ström and Dario Maggiolo for their valuable input and assistance throughout the different turns in the project. I have learned so much from you all and look forward to continue on that path in the continuation of the project.

I would also like to thank my wonderful co-workers at the division. You make all the time we spend here a sheer pleasure. A special thank to all the members of the multi-phase group and many more that ensures interesting and entertaining lunches every day and are always there for discussions and support when simulations crash or mistakes have been made.

Finally, I would like to give my gratitude to Malin and the rest of my family for your invaluable support and for patiently enduring me through this endeavor.

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ABBREVIATIONS

DNS	–	Direct Numerical Simulations
VOF	–	Volume of Fluid
MRF	–	Moving Reference Frame
PID	–	Proportional-Integral-Derivative
PLIC	–	Piecewise Linear Interface Reconstruction

NOMENCLATURE

Dimensionless numbers

$Eo = \dfrac{\rho_l g D^2}{\sigma}$	Eötvös number. Ratio of buoyancy to surface tension.	$-$
$Ga = \dfrac{\rho_l \sqrt{gD} D}{\mu_l}$	Galilei number. Ratio of buoyancy to viscosity.	$-$
$Sr = \dfrac{\omega D}{\sqrt{gD}}$	Dimensionless shear rate.	$-$

Greek Letters

μ_f	Dynamic fluid viscosity	$\mathrm{kg\,m^{-1}\,s^{-1}}$
ν_f	Kinematic fluid viscosity	$\mathrm{m^2\,s^{-1}}$
ω	Shear rate	$\mathrm{s^{-1}}$
ρ_f	Fluid density	$\mathrm{kg\,m^{-3}}$
σ	Surface tension	$\mathrm{N\,m^{-1}}$

Roman Letters

D	Spherical equivalent bubble diamter	m
g	Gravitational acceleration	$\mathrm{m\,s^{-2}}$
p	Pressure	Pa
R	Bubble Radius	m
r	Radial coordinate	m
T	Temperature	K
t	Time	s

Superscripts and Subscripts

$_\infty$	Surrounding
$_B$	Bubble
$_l$	Liquid
$_r$	Ratio
$_v$	Vapor

List of publications

This thesis consists of an extended summary and the following appended papers:

Paper A N. Hidman, G. Sardina, D. Maggiolo, H. Ström, and S. Sasic. Laser-induced vapour bubble as a means for crystal nucleation in supersaturated solutions - Formulation of a numerical framework. *Experimental and Computational Multiphase Flow* **1**.4 (2019), 242–254. DOI: 10.1007/s42757-019-0024-z

Paper B N. Hidman, G. Sardina, D. Maggiolo, H. Ström, and S. Sasic. Numerical Frameworks for Laser-Induced Cavitation: Is Interface Supersaturation a Plausible Primary Nucleation Mechanism? *Crystal Growth & Design* (2020). DOI: 10.1021/acs.cgd.0c00942

Paper C N. Hidman and G. Sardina. "Bubble dynamics in a moving reference frame - Development of an efficient multiphase DNS framework". *To be submitted to a scientific journal.* (2020)

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Part I

Extended Summary

1 Introduction

Bubbly flows is a multiphase fluid flow regime characterized by a dispersed gaseous phase in a liquid phase continuum. The gaseous phase is distributed as individual bubbles within the heavier liquid phase and, due to gravity, a buoyant force acts on the bubbles in the opposite gravitational direction. Bubbly flows are an integral part in a range of industrial and natural processes such as; bubble columns reactors, froth flotation tanks, nuclear reactors, heat exchangers, bubble drag reduction on marine vessels, oil and gas transport, atmosphere-ocean exchanges, cavitation etc [4]. To understand or design such systems, the ability to predict the dynamics of a single or a group of bubbles, and how the bubbles affect the liquid phase, is crucial. However, those dynamics are not yet fully understood, and reliable models for the dynamics are still needed [5]. One of the main challenges in the development of such models is the large range of time and length scales [6]. Bubbles may form at the scale of nanometers and, through complex interactions with other bubbles and the liquid phase, form structures in the order of tens of meters [7].

In this thesis work, we define three length scales for bubbly flow phenomena as small, intermediate and large scales. The small length scales are defined as the scales of individual bubbles or below. Intermediate scales refers to the length scales of bubble swarms where the bubbles exhibit a collective behavior, and the large scales refers to flow features that are at the length scales of the entire bubbly flow system. These scales are all above the molecular length scales that are not investigated in this work. Nonetheless, at the molecular scales, important processes such as the nucleation of vapor bubbles occur and are briefly discussed in Section 2.1.1.

To get an appreciation of multi-scale features in bubbly flows, consider a case of saturated nucleate boiling as illustrated in Figure 1.1. At molecular scales, the vapor bubbles are nucleated at the heating surface due to superheating of the adjacent liquid. The vapor bubbles grow across the small length scales by evaporation of the superheated liquid at the vapor-liquid interface. After reaching a critical size, the bubbles detach from the surface and rise due to buoyancy. At the small scales, the bubbles may coalesce with other bubbles, or, due to interaction with the liquid phase, breakup into smaller bubbles. These processes occur continuously within the system resulting in a poly-disperse bubbly flow with a large variation of bubble behaviors. Around the rising bubbles, the liquid is perturbed, and small-scale velocity fluctuations can be generated, something that is called bubble-induced (or pseudo-) turbulence [8]. In addition, small-scale viscous boundary layers are formed at the rising bubble interface and in the case of chemical reactions or phase change, also thermal and mass transfer boundary layers appear in the liquid around the bubbles. At intermediate scales, the hydrodynamics interaction between the rising bubbles can lead to the formation of bubble swarms that rise in a seemingly collective manner. At large scales, the buoyant forces from the bubble swarms can produce motions in the two-phase system such as re-circulation zones in the order of the largest length scales in the system. The dynamics at these large scales affect the processes at smaller scales resulting in a system that is coupled across the scales.

In this context, a fundamental understanding of the processes involved at one scale, such as the growth and dynamics of individual bubbles, is essential to predict the complex dynamics of the entire system. In the entire PhD project, we aim at increasing our knowledge about these processes and on developing numerical methods for investigating the bubbly flows across all relevant length scales. Our focus so far into the project has been on the small-scale bubble dynamics where we start by studying the evolution of nucleated vapor bubbles using two different numerical frameworks, one formulated in Paper A and the other developed in Paper B. In these papers we use the developed tools to investigate the fast and complex dynamics of laser-induced thermocavitation events. Then, we shift the focus to developing numerical frameworks for studying the dynamics of rising bubbles in Paper C. Here, we develop a framework with a moving reference frame that is suitable for studying the rising bubble dynamics and show the frameworks ability to capture relevant phenomena such as coalescence, breakup and unsteady rise behaviors.

This thesis is composed of five themed chapters. We begin with an overview of relevant phenomena and numerical challenges that are characteristic for the different length scales in Chapter 2. In Section 2.1, we present the phenomena and modelling challenges involved in the nucleation and growth of small vapor bubbles. We proceed to review the dynamics and modelling issues of intermediate scale bubbles rising in a liquid in Section 2.2 and then give an overview of the dynamics in industrial-scale bubbly flows in Section 2.3. To model this range of phenomena and length scales, we outline a commonly used multi-scale modelling approach for bubbly flows in Section 2.4. Based on this background, we continue by giving a summary of the developed numerical frameworks (in Paper A, B and C) and outline the numerical investigations we perform in Chapter 3. In Chapter 4, we give a summary of our main contributions and findings, and in Chapter 5, we discuss how we aim to investigate bubbly flows across the intermediate and large scales in the continuation of the project.

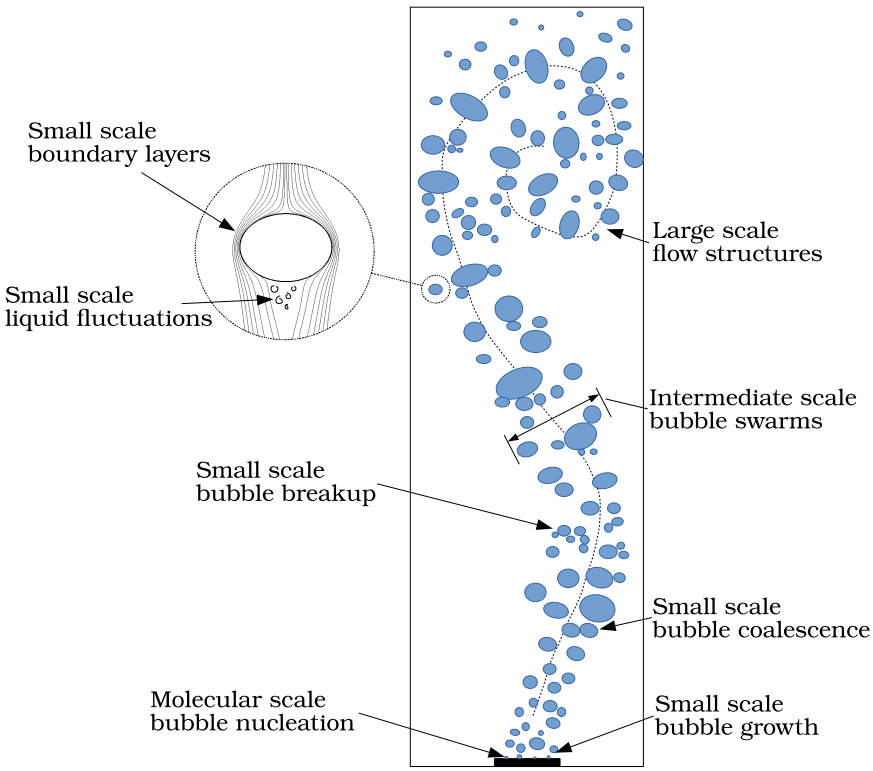


Figure 1.1: *Illustration of bubbly flow processes occurring at a range of length scales during saturated nucleate pool boiling. Vapor bubbles may nucleate at* $O(10^{-9}\ m)$ *and through complex interactions with other bubbles and the liquid, form flow structures in* $O(10\ m)$.

2 Background

2.1 Formation and evolution of small vapor bubbles

The formation and evolution of a vapor bubble can be divided into two phases, the nucleation of the bubble itself and the evolution of that bubble. First, we give a brief overview of the nucleation process and then focus on the small-scale bubble evolution process.

2.1.1 Vapour bubble nucleation

Vapor bubble nucleation is a stochastic, molecular, process that is governed by the liquids offset from saturation conditions and the surface tension between the phases. Because of the stochastic nature and the small scales of the problem, the nucleation process itself is not entirely understood [9]. We will not analyze this process in detail but instead, give some general concepts and explain the terminology.

The nucleation process can be divided into two main types, homogeneous and heterogeneous. In the homogeneous case, the nucleation occurs within the pure liquid phase due to thermal motions within the liquid. The motions produce temporary voids that may grow to bubbles if the former are sufficiently large. In the heterogeneous case, the nucleation takes place at a solid wall or particle in the liquid. The presence of a foreign particle or wall significantly increases the likelihood of nucleation, and therefore the heterogeneous nucleation is the most common type in engineering applications [10].

Regardless of the type of nucleation, the vapor bubble formation process is commonly separated into two categories, cavitation and boiling. These categories can be differentiated by defining cavitation as the process of nucleation when the liquid pressure falls below the liquid/vapor saturation pressure and boiling as the nucleation that occurs when the liquid temperature is raised above the saturation temperature.

Essentially, the two bubble formation processes are very similar, but in most practical situations, the dynamics of the resulting vapor bubbles differ considerably. In the cavitation case, it is the liquid pressure around the bubble that governs the bubble evolution process. The liquid pressure can change both rapidly and uniformly in the liquid due to local flow phenomena or changes of the system pressure. The rapid change of liquid pressure lead to fast growth and collapse dynamics in many cavitation cases. On the other hand, in the boiling case, it is the phase change and the temperature of the surrounding liquid that governs the bubble evolution process. Compared to the possibly rapid change of the liquid pressure, the liquid temperature around the bubble usually varies much more slowly. Because of this difference, the bubble evolution dynamics are usually slower in

boiling situations than in the cavitation cases. There are, however, processes for which also the temperature is rapidly varying, and the difference between cavitation and boiling dynamics is not so obvious. One such process is termed laser-induced thermocavitation and will be discussed further in Section 3.1.1 and in Paper A and B. Next, we examine the phenomena that govern the evolution of the nucleated vapor bubble.

2.1.2 Evolution of small vapor bubbles

Small bubbles are usually spherical due to the surface tension that results in a net force per unit area in the radially inward direction. The net force increase inversely with the radius of the bubble as $2\sigma/R$, where σ is the surface tension and R is the bubble interface radius. Since the radius of the nucleated bubble is small, the surface tension force is relatively large compared to inertial or viscous forces. These force ratios result in a spherical bubble with a minimum of surface energy.

The pressure of the gas inside the spherical bubble, p_B, is, at equilibrium conditions and for pure vapor, the saturation pressure, p_{sat}, corresponding to the vapor temperature, T_B. The pressure in the surrounding liquid, p_∞, and the surface tension force compress the gas inside the bubble. At equilibrium conditions, the pressures must balance across the bubble interface according to the Young-Laplace equation [11, 12]

$$p_B = p_\infty + 2\sigma/R\,. \tag{2.1}$$

At non-equilibrium conditions, the pressures may not balance and the bubble interface either grows or shrinks according to the generalized Rayleigh-Plesset equation [13–15]

$$\frac{p_B(t) - p_\infty(t)}{\rho_l} = R\frac{d^2R}{dt^2} + \frac{3}{2}\left(\frac{dR}{dt}\right)^2 + \frac{4\nu_l}{R}\frac{dR}{dt} + \frac{2\sigma}{\rho_l R}\,. \tag{2.2}$$

Here, ρ_l and ν_l are the liquid density and kinematic viscosity, respectively and the effects of phase change are not included. What differentiates the bubble growth processes in cavitation and boiling situations are the phenomena that govern the pressure difference on the left-hand side of Equation 2.2. In cavitation cases, the surrounding liquid pressure $p_\infty(t)$ is reduced, whereas, in boiling situations, the liquid is evaporated into the bubble causing the bubble pressure $p_B(t)$ to increase. Both phenomena result in bubble growth, but since the physical processes are different, the resulting bubble dynamics differ as well.

For most cavitation bubbles, the effects of phase change are relatively small, and the growth rate of the bubble is controlled by the inertial effects of the surrounding liquid, described by the two first terms on the right-hand side of Equation 2.2 [10]. With rapid changes in the surrounding liquid pressure, the cavitation bubble can display fast growth and collapse dynamics. When the liquid pressure falls, the bubble rapidly expands, and if the bubble moves into a high-pressure liquid region, the bubble may collapse implosively. During the collapse phase, the liquid inertia and the surface tension force can compress the bubble to a size of the order of microns. Such implosive collapses can produce

maximum gas temperatures over 15 000K and the emission of shock waves as fast as 4000 m/s [7, 16]. These gas temperatures are sufficient to produce visible light, a phenomenon called cavitation luminescence, and, if the collapse occurs in the proximity of a wall, the emitted shock waves and possible formation of liquid jets can cause significant erosion of the solid material.

In boiling cases, the growth rate is mainly governed by the evaporation rate of the interfacial liquid. Because of latent heat of evaporation, the liquid at the interface is cooled towards the saturation temperature at which point the evaporation rate becomes controlled by the ability of the phases to transport heat to the interface. Since the thermal conductivity is usually much higher in the liquid phase, the growth rate is thus governed by the heat transport in the liquid surrounding the bubble. The cooling of the interfacial liquid produces a thermal boundary layer around the bubble with a thickness that can be of the order of nanometers. Such small scales significantly complicate simulations of the growing bubble, since a well resolved thermal boundary layer is necessary to accurately capture the liquid heat transport that governs the evaporation rate.

To predict the evolution of boiling or cavitation vapor bubbles, the model must take into account both fluid- and thermodynamic effects and include the effects of surface tension and phase change at the interface. Because of the fast dynamics, compressibility effects can be significant and, at the moving bubble interface, large transient gradients of the fluid conditions need to be accurately estimated. The number of physical phenomena and the complex interactions between them constitute a unique challenge. Indeed, research on this topic has been conducted for more than a century and is still ongoing. Rayleigh (1917) [13] developed an equation for the growth of an inertia-controlled spherical vapor bubble. This formulation was further developed by, among others, Plesset and Zwick (1954) [14], Scriven (1959) [17], Mikic et al. (1970) [18], Dalle Donne and Ferranti (1975) [19], Prosperetti and Plesset (1978) [20] and Lee (1993) [21] to consider growth regimes governed by thermal and surface tension effects. Although the mathematical models have become increasingly accurate, they are only valid under certain conditions and growth regimes, and universal models are still lacking.

2.2 Dynamics of small-scale rising bubbles

The rising motion of gas bubbles in a liquid has intrigued researchers for a long time. In a quiescent liquid, the rising behavior of a bubble can be rectilinear, zigzagging, spiraling or chaotic depending on certain two-phase flow parameters [22]. Interestingly, Leonardo da Vinci documented that this dynamics is indeed three-dimensional already in the 1500s and Prosperetti (2004) termed the path instability phenomenon as Leonardo's Paradox since it was not known why an axisymmetric bubble would move in a zigzagging, spiraling or chaotic path [23]. More recent numerical investigations of this phenomenon have shown that the shape and path of a rising bubble are closely associated and depend upon specific force ratios in the two-phase flow [24, 25].

For a single rising bubble, the problem is completely described by the following four dimensionless parameters [26]; the Galilei number $Ga = \frac{\rho_l \sqrt{gD} D}{\mu_l}$ that relates buoyancy to viscous forces, the Eötvös number $Eo = \frac{\rho_l g D^2}{\sigma}$ that relates buoyancy to surface tension forces, the density ratio $\rho_r = \frac{\rho_l}{\rho_g}$ and the dynamic viscosity ratio $\mu_r = \frac{\mu_l}{\mu_g}$. Here, g is the gravitational acceleration, D is the spherical equivalent bubble diameter, σ is the surface tension and l and g denotes the liquid and the gaseous phases. If the bubble is rising in a shear liquid flow, also the dimensionless shear rate is introduced $Sr = \frac{\omega D}{\sqrt{gD}}$ where ω is the shear rate of the surrounding liquid flow.

The interfacial forces acting on the bubble govern the rising path dynamics. Consequently, for a bubble rising in a zigzag or spiral path, the interfacial forces are continuously varying but with some periodicity. However, in the rectilinear rise regime, the forces are constant, and in the chaotic rise regime, they vary without any regularity. To develop models that can predict the interfacial forces for all rising behaviors is a formidable task. Thus, most interfacial force models focus on predicting the forces for a given set of problem parameters and often with a quasi-steady approach to avoid describing the complex transient behaviors. For rising bubble dynamics at this small scale, universally applicable models are incomplete.

If bubbles come in contact with each other, they may coalesce into larger bubbles that display entirely different dynamics. Contrarily, if the surface tension force is not sufficient to keep the integrity of the bubble interface, external flow forces may cause the bubble to breakup into smaller bubbles. In certain bubbly flow systems, these two processes govern the global bubble size distribution and can therefore alter the dynamics and characteristics of the entire system. To accurately predict the behavior of large-scale gas-liquid systems, models are needed to predict these small-scale dynamics. Without such models, the simulations of large-scale systems need to accurately resolve all small-scale dynamics and the computational cost becomes unfeasible. However, rigorous models for the breakup and coalescence dynamics are not yet available.

2.3 Dynamics of intermediate and large-scale bubbly flows

The rising of a single bubble disturbs the motion of the surrounding liquid and, therefore, affects the dynamics of other bubbles nearby. Consequently, the interfacial forces that govern the motion of the single bubbles are altered by the presence of nearby bubbles. So, even if models can predict the single bubble interfacial forces, these models need to be modified to predict the motion of the bubbles in bubble swarms. However, how the interfacial forces are altered is not yet clear, and different bubble swarm behaviors have been observed depending on the problem parameters.

The gravitational forces induce the buoyancy force acting on the bubbles and cause the

rise of individual bubbles. At large scales, spatial nonuniformities of the bubble distribution, i.e. bubble swarms, result in an inhomogeneous gravitational force distribution. These inhomogeneous forces cause large-structure, gravity-induced, flows that enhance the inhomogeneity of the bubble distribution and can produce turbulent flows [4]. In some bubbly flows applications, such as bubble columns, the turbulence characteristics are fundamental aspects to design efficient processes.

The transient motions of many rising bubbles can both induce and dampen the turbulent fluctuations in the liquid phase [8]. These fluctuations have different characteristics compared to single-phase flows and result in different behavior of the turbulent energy spectrum [27]. Because of varying flow behavior at different gas volume fractions, the strong coupling between the phases and the complex dynamics and interactions at single bubble scales, it is very challenging to model the turbulence in bubbly flows. Existing turbulence models are still inadequate to describe all types of bubbly flows.

2.4 Multi-scale modelling strategies

Given the background of the critical phenomena and dynamics of bubbly flows at different scales, it can now be appropriate to give a brief overview of the available numerical techniques that are used to study such flows. To handle the wide range of time and length scales, it is common to use a multi-scale simulation strategy [28] as illustrated in Figure 2.1. In this approach, different numerical techniques are used to investigate phenomena at one scale and to derive closures that can be used to simulate the bubbly flow at larger scales.

If we disregard molecular phenomena, such as phase change and nucleation, the smallest scales in the flow can be resolved by multiphase direct numerical simulations (DNS). The DNS approach fully resolves the bubble shape and the flow field inside and outside the bubble. From these simulations, it is possible to derive closures, e.g. the interfacial forces, bubble-induced turbulence and bubble coalescence and breakup processes. Currently, due to the excessive computational cost, DNS simulations are only practical for studying $O(100)$ bubbles if closure laws valid for a broad parameter range are sought for.

The Eulerian-Lagrangian approach can be used to study larger bubble swarms ($O(10^6)$). Here, the detailed flow field around the bubbles and the bubble shapes are not resolved, and the motion of the bubbles are determined using interfacial force closures. If also closures for the coalescence and breakup processes are included, it is possible to obtain the bubble size distribution from this type of simulation.

To model industrial-scale systems, an Eulerian-Eulerian continuum approach is used where both the gas and liquid phases are treated as interpenetrating continua. Now, the same interfacial force closures and bubble coalescence and breakup models can be used but need to be supplemented with a bubble population balance. Since the simulation

techniques at the large scales depend on a high number of closures, their accuracy is governed by the quality of those closure models. Moreover, as stated before, reliable and accurate closure models are still needed.

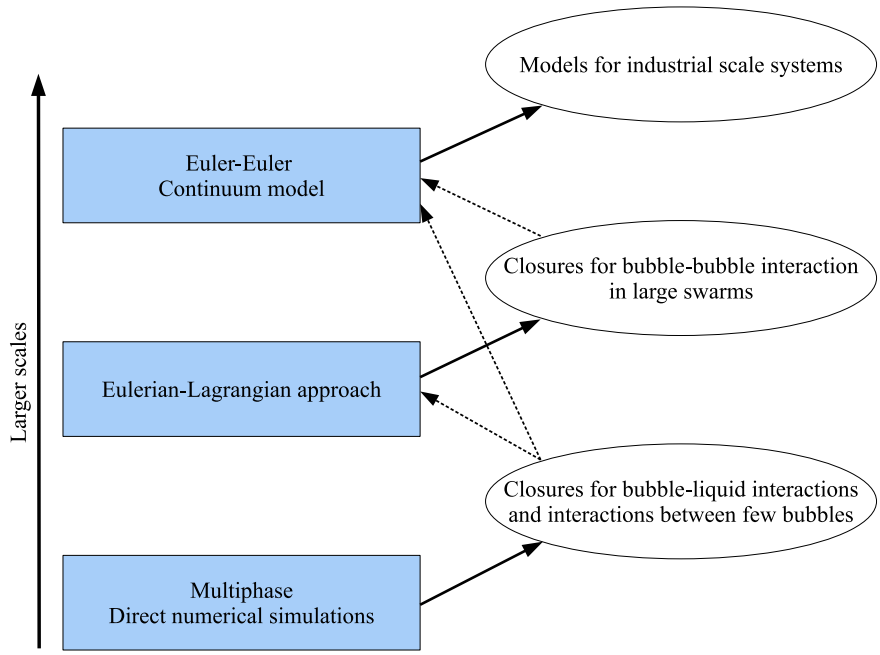


Figure 2.1: *Illustration of the multi-scale modelling approach in bubbly flows.*

3 Numerical investigations

As discussed in the previous chapter, there is still a significant need for further development and improvement of the closures and numerical techniques used in the study of bubbly flows. In this thesis work, we have chosen to focus on the numerical investigation of phenomena at small scales but aim at contributing to the numerical methods across all scales in the future.

First, we present a summary of our investigations on the evolution of small vapor bubbles (Paper A and B). Then, we shift the focus to show our attempt at improving the numerical methods for studying small-scale rising bubble dynamics (Paper C).

3.1 Evolution of small vapor bubbles

To capture any type of bubble evolution process, a numerical framework needs to take into account relevant phenomena such as fluid- and thermodynamics effects, phase change and surface tension effects. As discussed in Section 2.1.2, several mathematical models are developed for this kind of bubble dynamics, but those models are derived for bubble evolution under specific conditions and evolution regimes.

We aim at providing numerical frameworks that can handle general bubble evolution processes. For this purpose, we avoid common modelling assumptions such as homogeneous vapor conditions, constant physical properties and negligible viscous- inertia- or thermal effects under specific bubble evolution periods. Instead, we consider the relevant effects during the entire evolution process, resolve the fluid conditions in both space and time for both phases and take into account the variation of the physical properties. This approach adds complexity and requires a higher computational cost compared to simplified mathematical models but provides a general methodology for predicting the evolution process of any bubble. By resolving the fluid conditions also gives detailed information about the variations of those conditions, something that is essential information in certain applications such as laser-induced thermocavitation bubbles used for crystallization. This application is further discussed in Section 3.1.3.

The laser-induced thermocavitation method is a suitable example that exemplifies challenges and complexities from both the boiling and cavitation processes. Here, the bubble exhibits the fast dynamics seen in cavitation bubbles, but the bubble growth rate is governed by rapid phase change and thermal effects. These types of bubbles constitute a major numerical challenge and are therefore useful cases to study when formulating a general numerical framework.

3.1.1 Laser-induced thermocavitation

In the laser-induced thermocavitation method, a short (often nano-second) laser pulse superheats a region of the liquid phase. Within this region, a vapor bubble is nucleated and starts to grow due to rapid evaporation of the superheated liquid at the bubble interface. The evaporation leads to high pressure in the bubble that results in bubble growth. Once all the superheated liquid is evaporated or cooled, the vapor starts to condense due to heat loss to the surrounding liquid and the bubble begin to collapse. In a typical laser-induced thermocavitation event, the entire bubble lifetime is $O(100\mu s)$, and the maximum bubble radius is $O(100\mu m)$.

3.1.2 Multiphase DNS framework

To investigate the laser-induced thermocavitation bubbles numerically, we use a multiphase DNS approach that, apart from the fluid dynamics, also consider thermal effects in both phases, surface tension and includes the effects of the interfacial phase change. Because of the rapid dynamics, also compressibility effects are considered.

One of the main numerical challenges in multiphase DNS of bubbly flows is the presence of the interface between the different fluid phases. Since the interface is a 2D phenomenon, i.e. it does not have a thickness, it is not possible to resolve the interface with the standard finite volume method. Therefore, special numerical methods have been developed to handle the discontinuity between the phases and the position of the interface within the finite volume approach.

We use in this work the Volume of Fluid (VOF) method to handle the two-phase flow[29]. Here, the volume fraction field of the phases is tracked, and the interface between the phases is identified as the position where the volume fraction field is between 1 and 0. With this approach, the interface can be treated in the same finite volume approach as the other governing equations, which significantly reduces the complexity of the numerical method.

In VOF, the transition from one fluid phase to the other occurs over the length scale of at least one computational cell. Because of this, it is not straightforward to define the exact location and orientation of the interface. Also, for a curved interface, the surface tension results in a net force in the interface normal direction, and, if the surface tension varies along the interface, a tangential force is induced at the interface, a phenomenon called the Marangoni effect. Since these forces act at the interface, it is not trivial to include them in the finite volume method either.

In the case of a laser-induced thermocavitation bubble, the bubble is small and the interface highly curved. This lead to high forces in the interface normal direction. These forces are implemented as volume forces in the computational cells containing the interface. For the highly curved, deformable and rapidly changing interfaces of our application,

the numerical implementation of the interfacial forces needs careful consideration. To accurately resolve such interfaces requires high spatial and temporal resolutions, and to maintain a relatively sharp interface, we use the PLIC interface reconstruction method that estimates the location and orientation of the interface within the computational cells at each computational time step. This method reduces the amount of smearing of the interface that may occur due to numerically diffusive volume fraction advection schemes. The interfacial forces are implemented using the method proposed by Brackbill et al. [30] that depend on an accurate representation of the interface to predict the correct forces.

It is the phase change, i.e. evaporation and condensation, that governs the evolution of the bubble in the laser-induced thermocavitation case. The phase change takes place across the interface and is a complex process that involves the transport of both heat and mass across the interface and absorption or release of thermal energy due to latent heat. To include these phenomena in VOF, we implement the approach proposed by Hardt & Wondra [31] and extended it to increase accuracy and reduce numerical instabilities at the interface. In this approach, the effects of phase change are included by a phase change model and mass and energy source terms in the vicinity of the interface. The source terms ensure that the correct amount of mass and energy that is added at one side of the interface is removed on the other side. The implementation of the phase change approach is explained in detail in Paper A and B.

In this numerical framework, we considering both phases as compressible and solve for the entire system of continuity, volume fraction, momentum and energy governing equations. The numerical framework is developed, validated and described in detail in Paper A and in Paper B, we further develop it to improve the accuracy of the interfacial energy transfer. In Paper A, the validation shows that the framework predicts bubble dynamics in agreement with analytical models for boiling cases. We also simulate an experimentally observed laser-induced thermocavitation event and predict the dynamics of the bubble in reasonable agreement with the experimental measurements. These results indicate that our multiphase DNS framework can capture the relevant phenomena in the extreme case of a laser-induced thermocavitation event. The numerical framework should, therefore be able to handle most types of bubble evolution processes.

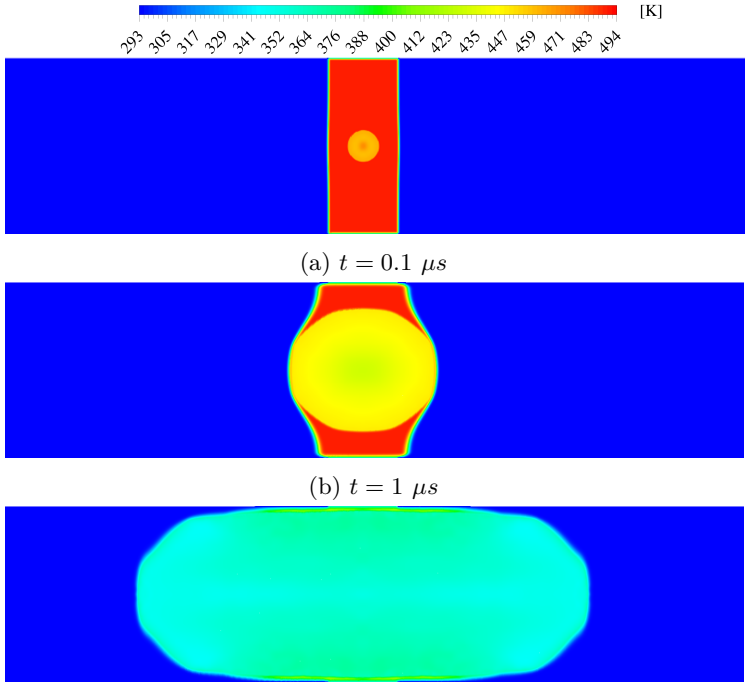
3.1.3 Numerical investigations of laser-induced thermocavitation for crystallization

The laser-induced cavities are increasingly studied as a promising new tool to achieve good spatiotemporal control in the process of crystallization [32]. Crystallization has been observed in experiments around the laser-induced cavities in saturated solutions, but the mechanisms behind the crystallization are not entirely clear [33–36]. One crystal nucleation hypothesis is that the evaporation of the solvent at the bubble interface produces a high solution supersaturation around the bubble [37]. The evaporation increases the concentration of solute in the solution around the bubble and at the same time, cools the solution. These effects lead to an increase of the solution supersaturation,

and with sufficient duration and degree of supersaturation, crystals may nucleate within the solution around the bubble.

Because of the small spatiotemporal scales and fast dynamics of the cavitation event, it is very difficult to measure the degree of supersaturation experimentally. With fully resolved DNS simulations it is, however, possible to obtain such estimates. In general, the supersaturation level is dependent on the concentration of the dissolved solute and the saturation concentration at the solution temperature and pressure. The temperature and pressure in the liquid around the bubble are resolved in the DNS framework but also the evolution of the solute concentration is needed.

In Paper B, our DNS framework is extended with considering the solute transport in the liquid around the vapor bubble and also with an improved formulation of the interfacial energy transfer. In this study, we test if the crystal nucleation hypothesis about high supersaturation in the solution around the bubble is plausible by simulating a thermo-cavitation event with experimentally observed crystallization. In Figure 3.1, we show the temperature contours from three instants during this simulation. Here, a solution at 293 K is placed between an upper and lower wall, 50 μm apart, and with outlets to the sides. Initially, a 2 μm vapor bubble is placed in the center of the domain, and during the 9 ns laser pulse, a cylinder of liquid is heated to almost 500 K. The superheated liquid evaporates into the vapor bubble that rapidly expands between the walls and reaches a diameter of over 130 μm in only 6 μs. By extracting the temperature and pressure of the liquid around the bubble, and by computing the evolution of the solute concentration in this liquid, an estimate of the solution supersaturation is obtained. The simulation results show bubble growth rates in fair agreement with the experimental measurements and a significant peak of supersaturation that is not possible to achieve in evaporative or cooling crystallization under normal conditions [37]. These results indicate that the crystal nucleation hypothesis is plausible and deserves to be investigated further.



(a) $t = 0.1\ \mu s$

(b) $t = 1\ \mu s$

(c) $t = 6\ \mu s$

Figure 3.1: *Temperature contours at three instants from the simulation of a laser-induced thermocavitation bubble performed in Paper B.*

3.1.4 Formulation of a 1D model and parameter investigation

From experimental studies, it has been observed that crystallization is only obtained under certain conditions and laser pulse parameters [33, 34]. If the high supersaturation in the solution around the bubble is indeed the mechanism behind the observed crystal nucleation, it raises questions about the conditions that are necessary for obtaining the high supersaturation.

To investigate these conditions, we first identify the important parameters that affect the supersaturation level and then investigate those parameters using numerical simulations. We reason in Paper B that it is the evaporation of the solvent that produces the increase of the solute concentration in the solution around the bubble. The laser pulse energy and the spatial distribution of that energy govern the rate and duration of the evaporation, and the diffusivity and solubility of the solute affect the solute concentration and supersaturation level in the solution. This gives us at least four important parameters to investigate under a range of relevant values.

Unfortunately, there is an enormous computational cost associated with the multiphase

DNS simulations that makes such parameter investigations practically impossible to perform. However, as discussed in Section 2.1.2, the small scale of the laser-induced cavities makes them approximately spherical, and by assuming the bubble evolution occurs far from an external boundary, the entire process can be approximated as spherical-symmetric. This approximation allows us to formulate the entire problem in a 1D framework, in the radial direction. This approach reduces the computational cost to an almost negligible fraction of the DNS framework.

The 1D framework is developed, validated and used to examine the important parameters in Paper B. The validation cases consist of experimentally observed laser-induced thermocavitation bubbles and the 1D framework predict the bubble radius evolutions in good agreement with the experiments, from bubble growth to the collapse phase. In the parameter investigations, the 1D framework shows results that clearly indicate the high supersaturation is only possible under specific ranges of the investigated parameters. This conclusion is in line with the aforementioned experimental observations, and the presented parameter studies can be used as guidelines for any user to find appropriate setups to reach conditions favorable for crystallization.

3.2 Rising bubble dynamics

Here, we outline the second part of our work that focuses on numerical methods for investigating rising bubble dynamics at small scales. So far into the PhD project, the main focus has been on the evolution of small vapor bubbles discussed in Section 3.1. Consequently, the work on rising bubble dynamics is not yet as thorough, but we aim at focusing on this topic at small, intermediate and large scales in the continuation of the project.

As discussed in Section 2.4, there is still a need of reliable closures for the interfacial forces, breakup, coalescence and bubble-induced turbulence effects in numerical modelling approaches at intermediate and large scales in bubbly flows. These closures could be developed or improved by numerical investigations of the bubble dynamics using multiphase DNS methods in systems with few bubbles.

In many of the closures used in intermediate and large-scale modelling approaches, it is the quasi-steady bubble behavior or forces that is needed. A major challenge with investigating the small-scale bubble dynamics is that the dynamics may develop over relatively large spatial and temporal scales. So, to develop or improve the closures for quasi-steady behavior, the DNS simulations may require large computational domains and run for long simulation times.

The excessive computational cost associated with the multiphase DNS methods is prohibiting detailed numerical investigations across the relevant parameter ranges. For this reason, it is essential that the efficiency of the DNS simulations is improved and that the

computational cost is decreased.

3.2.1 Non-inertial moving reference frame

It is the flow field in the vicinity of the bubbles that governs the bubble dynamics. Therefore, it can be convenient to make a change of coordinates to a reference system moving with the bubbles. By following the bubbles, the computational domain can be significantly reduced, and non-important flow features far from the bubbles are disregarded. Also, with a reference frame moving with the bubbles, there is no need to estimate a sufficient domain size a priori, in order to capture the quasi-steady behavior.

In Paper C, we develop, validate and demonstrate a multiphase DNS framework with a reference frame moving with the bubbles. Since the bubbles move in a non-inertial manner, also the moving reference frame becomes non-inertial. In such reference frames, the governing equations for the flow need to be modified by including the acceleration of the reference frame itself. The motion of the reference frame is continuously adjusted to keep the bubbles in the center of the computational domain. This adjustment can be determined in many ways, and in Paper C, we have chosen to update the velocity of the reference frame using a Proportional-Integral-Derivative (PID)-control approach. This approach gives a general implementation that can handle both single and multiple bubble systems and ensures that the center of mass of the bubble is kept in the center of the domain even for longer simulation times.

With this framework, the computational cost of the DNS simulations can be significantly reduced, and the setup of the simulations, e.g. specifying the domain size, is simplified. We aim to use the framework to perform numerical investigations of the bubble dynamics and to develop or improve the closures for these dynamics in the continuation of the project.

4 Conclusions

The aim of the PhD project is to develop numerical methods for investigating and understanding bubbly flow phenomena across many relevant length scales. So far, we have focused on studying the bubble dynamics at small scales but plan to investigate processes at intermediate and large scales in the remaining phase of the PhD project.

We started by studying the evolution of vapor bubbles. For this purpose, we developed and validated a multiphase DNS framework in Paper A. This framework takes into account the governing phenomena in both cavitation and vapor bubble evolution processes. In Paper A, we used the framework to simulate the fast and complex dynamics of a laser-induced thermocavitation bubble with growth rates governed by rapid phase change and thermal effects at the bubble interface. The predicted growth rates are in fair agreement with experimental results and show that the framework can capture the correct physics of the vapor bubble evolution process.

In Paper B, we extended the multiphase DNS framework with an improved formulation of the energy transfer at the bubble interface and by considering the transport of a dissolved solute in the liquid phase. Here, we used the DNS tool to investigate plausible mechanisms behind the experimentally observed crystallization in the solution around a laser-induced thermocavitation bubbles. We showed that the rapid solvent evaporation during the early bubble growth phase produces a peak of solution supersaturation at the bubble interface that is not possible to obtain using conventional crystallization techniques under normal conditions. Since crystals have a higher probability of nucleating at increased supersaturation levels, our results indicate that the predicted peak due to rapid solvent evaporation may be the mechanism behind the observed crystallization.

In addition, we developed a 1D numerical framework for the bubble dynamics to study the conditions that are necessary for obtaining the high supersaturation. Because of the high computational cost of the multiphase DNS framework, the DNS approach is not feasible to use for an extensive investigation of relevant parameters. With the 1D framework, we examined the effects of the laser pulse energy, the spatial distribution of that energy, the solute diffusivity and the solute solubility on the maximum supersaturation level that is reached during the bubble evolution process. The results showed that high supersaturation peaks are only attained under specific ranges of the studied parameters. From our results, guidelines were provided to identify suitable sets of parameters that produce conditions favorable for crystallization.

After investigating the evolution process of vapor bubbles, we shifted our focus to the small-scale rising bubble dynamics. Here, our final aim is to investigate and develop reliable closures for phenomena such as bubble breakup/coalescence and interfacial forces. We developed in Paper C an efficient multiphase DNS method with a moving reference frame that follows the center of mass of the rising bubbles. The motion of the moving reference frame is determined using a PID-control approach that inherently handles both

single and multiple bubble systems. With this approach, the computational domain, and cost, of the DNS simulations can be significantly reduced, and the setup of the simulations is simplified by eliminating the need for a priori estimations of sufficient domain sizes to capture accurate statistics of the bubble dynamics.

In the next phase of the PhD project, we intend to use the method outlined in Paper C to perform extensive numerical investigations of the small to intermediate-scale bubble dynamics. These investigations will provide insights and closures that will be used to develop novel numerical approaches and investigate bubble dynamics across larger length scales.

5 Future work

In the next phase of the PhD project, we will use the numerical framework developed in Paper C to investigate interfacial forces and breakup dynamics for single bubbles and localized bubble swarms. The interfacial force we will focus on is the shear-induced lift force. This force arises due to velocity gradients in the liquid phase that interact with the bubble and the bubble wake. The interaction leads to a force acting on the bubble in the transversal direction, towards the low-speed flow regions. Because of the complex interaction between the bubble shape, wake and shear flow field, the resulting lift force is a highly nonlinear function of the problem parameters. Under specific conditions, the lift force even changes sign to the opposite direction (towards the high-speed flow regions). Moreover, surfactants in the liquid phase can accumulate at the bubble interface and significantly affect the lift force coefficient. These aspects make reliable models difficult to obtain, and currently, the available models for the lift force are only applicable for specific ranges of the problem parameters. Since the lift force acts in the transversal direction, the direction and magnitude of the force governs the spatial bubble distribution in many important bubbly flow systems, such as bubble column reactors and bubbly pipe flow, and it is therefore essential to model the lift force accurately.

We will start by investigating the lift force on a single bubble rising in a linear shear flow and then examine how the lift force is altered in a localized bubble swarm. These investigations will be performed for a wide range of the problem parameters and lead to improved closures for the lift force. Using the framework developed in Paper C, we will also investigate the bubble breakup dynamics and develop improved closures for this phenomenon. The novel sub-grid models extracted from these investigations can be used in Eulerian-Lagrangian and Eulerian-Eulerian numerical frameworks to study the dynamics in systems with a large number of bubbles or industrial-scale bubbly flow systems.

Another objective is to investigate bubble-induced turbulence. In particular, we want to extract differences and similarities between the statistics of single-phase homogeneous turbulence and bubble-induced turbulence and derive improved closures for the interphase momentum transfer terms and subgrid stresses used in the simulation of industrial-scale systems.

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