DROPLET IMPACT PHENOMENA

in nanolithography

Sten A. Reijers

Sten A. Reijers - Droplet impact phenomena in nanolithography

Droplet impact phenomena in nanolithography

Sten Arjen Reijers

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Contents

1	Introduction	1
1.1	Extreme ultraviolet nano-lithography	4
1.1.1	Fluid dynamics inside LPP-EUV light sources	5
1.1.2	Laser impact on droplets	6
1.1.3	Droplet impact onto deep liquid pools	9
1.2	Guide through the thesis	11
2	Axisymmetric multiphase Lattice Boltzmann method for generic equations of state	17
2.1	Introduction	17
2.2	Model derivation	19
2.2.1	The lattice Boltzmann method	19
2.2.2	The extension to an axisymmetric method	20
2.2.3	The extension to a multiphase axisymmetric method	22
2.3	Results and Validation	24
2.3.1	Stationary droplet validation	25
2.3.2	Oscillating droplet validation	26
2.3.3	Wave propagation validation	28
2.4	Discussion & Conclusion	29
3	Droplet deformation by short laser-induced pressure pulses	33
3.1	Introduction	33
3.2	Problem formulation & methods	36
3.2.1	Scaling analysis	36
3.2.2	The weakly compressible model	39
3.2.3	The lattice-Boltzmann method	44
3.3	Results	45
3.3.1	Acoustic response of a droplet to the ablation pressure	45
3.3.2	The effect of the pulse duration on the droplet response	48
3.4	Discussion & conclusion	55

4	Laser-to-droplet alignment sensitivity relevant for laser-produced plasma sources of extreme ultraviolet light	61
4.1	Introduction	61
4.2	Experiment	62
4.3	Model	64
4.4	Results	67
4.5	Discussion and Industrial application	71
4.6	Conclusions	73
4.7	Appendix	73
5	Oblique droplet impact onto a deep liquid pool	77
5.1	Introduction	77
5.2	Numerical setup	78
5.3	Results	82
5.3.1	Impact phenomena	83
5.3.2	Impact dynamics	84
5.3.3	Splashing threshold	88
5.3.4	Cavity dynamics	90
5.3.5	Cavity dimensions	92
5.4	Discussion & Conclusion	93
6	A sequence approach to solve the Burgers' equation	101
6.1	Introduction	102
6.2	Preliminaries	103
6.2.1	Banach space and series	103
6.2.2	The differentiation and integration of a series	103
6.2.3	The product of two series	103
6.3	The sequence transformation method	104
6.4	The Burgers' equation	105
6.4.1	The sequence differential equation	105
6.4.2	Solving the sequence differential equation in the complex plane	106
6.5	An initial value problem for the Burgers' equation	107
6.5.1	Solving the initial value problem	108
6.5.2	Proof of convergence	109
6.5.3	The absolute error	111
6.6	Discussion & Conclusion	112
7	Summary & Conclusion	115
	Samenvatting	121
	Acknowledgements	125

1

Introduction

Figure 1.1: (a) Sketches of the impact of a mercury droplet onto a smoked glass substrate observed by Worthington in 1877 [1]. (b) Photographs of the impact stages of a rough marble sphere in a pool of pure glycerine in vacuum by Worthington in 1900 [2].

The impact dynamics of a falling droplet is a broad scientific subject that has been extensively studied over the last centuries. Starting in 1877, Worthington was the first to make detailed drawings of the impact of milk and mercury droplets onto a smoked glass substrate [1], see figure 1.1a. Later he also studied the perpendicular impact of a rough marble sphere onto a liquid pool of pure glycerine in vacuum conditions, where he experimentally observed the ejecta sheet and cavity dynamics of an impacting body onto a pool [2], see figure 1.1b. He started an important branch

of research into droplet impact phenomena, which is still a highly relevant topic in fluid mechanics today.

Apart from scientific interest, droplet impact phenomena plays an important role in many modern industrial applications such as ink-jet printing [20, 21], combustion engines [22], pesticide delivery [23], erosion protection [24] or Extreme Ultraviolet (EUV) light sources [26]. Carefully conducted experiments, analytic modelling and numerical simulations give fundamental insight into each stage of the droplet impact and could therefore lead to printers with higher resolution, more power-efficient combustion engines, reduction in the use of pesticides, less maintenance of erosion sensitive materials and higher output power in EUV light sources as we will discuss in this thesis.

Most of these droplet impact phenomena happen in the blink of an eye and may at first sight not seem very complex, but if you look closer you will find that each impact event consists of many different stages. Many of these different impact stages can be observed while you take a shower. The moment when you turn on the shower, a water jet is produced by the shower head that immediately atomizes away from the nozzle exit [3–5], i.e. the water jet breaks up into a train of droplets. The resulting stream of droplets falls towards the bathtub where the droplets initially impact onto a dry surface [6, 7]. During impact, a rapid ejecta sheet develops along the rim of the impacting droplet resulting in an early splash, which is typically too fast for the human eye to observe. After impact, the droplet spreads radially along the surface and fingers appear on the rim of the expanding sheet that in some cases also lead to a splash [8–11]. Figure 1.2a shows a numerical simulation of this fingering instability. Prior to impact on the surface, droplets could also impact with each other during transit. Figure 1.2c shows a numerical simulation of the head-on collision between two identical water droplets. Directly after impact the two droplets coalesce and deform into a radially expanding thin liquid sheet. Similar to the impact of a droplet onto the dry bathtub surface, fingers appear on the rim of this expanding sheet and lead to the fragmentation of the droplet.

As the bathtub fills, the droplets no longer impact onto a dry surface but instead impact onto a pool of variable depth. Here, an ejecta sheet is created during impact [14–17] that evolves into a crown at later stages [14–17]. Figure 1.2b shows an experimental and numerical example of a perpendicular droplet impact onto a thin liquid film. When a droplet impacts on a deep pool, see figure 1.2d, the impact is accompanied by the formation of a cavity below the pool surface. When the crown collapses capillary waves are created that travel from the pool surface into the cavity. At some point these waves collide and simultaneously with the closure of the cavity a Worthington jet may be created [18, 19].

These fascinating droplet impact phenomena are events that everyone sees on a daily basis and therefore may look easy to understand. Instead, as described above, the dynamics of these phenomena are often highly non-linear and their mechanisms

Figure 1.2: Numerical and experimental examples of droplet impact phenomena. (a) The numerical simulation of a droplet impact onto a dry and solid surface showing a fingering instability. Image taken from [10]. (b) Perpendicular droplet impact onto a thin liquid film, experiments (left) and numerics (right). Image taken from [12]. (c) A numerical simulation of the head-on collision of two water droplets. A thin sheet is formed that fragments into tiny droplets. Image taken from [13]. (d) The perpendicular impact of a droplet onto a deep liquid pool. (from left to right): the formation of the ejecta sheet, the formation of the crown, the onset of the fingering instability above the pool and the formation of a cavity below the pool surface. Image taken from [14].

are still far from being fully understood today.

In this thesis we use both analytic modelling and numerical simulations to understand two types of droplet impact phenomena. In the first part of this thesis we study the impact of a laser pulse onto a falling droplet. Here the laser impact induces a rapid phase change in a thin superficial layer on the illuminated side of the droplet. The resulting recoil pressure deforms and fragments the droplet at later stages. The droplet dynamics shows strong similarities with the sheet expansion and breakup of head-on collisions between two droplets or droplets impacting a dry surface. For high-intensity and sub-nanosecond laser pulses however, the dynamics greatly differs due to the influence of liquid compressibility. In this regime, we aim to analytically and numerically understand the onset of the deformation dynamics. In the second part of this thesis, we investigate the impact of a droplet onto a deep liquid pool under an angle, i.e. obliquely. As a result the droplet can, apart from the classical

Figure 1.3: (a) The wavelength of light used in photolithography machines decreased the last decades by almost a factor 30. (b) A schematic overview of a Laser-Produced Plasma (LPP) EUV light source. A train of tin droplets falls in the focus of a CO₂ laser, where it is hit by a pre-pulse to obtain an extended disk-like target. The disk-like target increases coupling with the next pulse that leads to the creation of a high-density tin plasma. Line emission in the plasma provides the EUV light peaking at 13.5nm which is focused by the multilayer collector to the intermediate focus point. Image taken from [33].

deposition and omni-directional splashing, also splash in a single direction [25]. We study the cavity and sheet dynamics numerically in the regime where the dynamics differ from the perpendicular impact. Both of these phenomena play an important role in the production of EUV light in state-of-the-art nano-lithography machines.

1.1 Extreme ultraviolet nano-lithography

Extreme ultraviolet lithography (EUVL) is the next generation of photolithography technology used by the semiconductor industry for the manufacturing of integrated circuits [26–29]. Over the last decades the semiconductor industry kept up with predictions made by Gordon E. Moore stating that the number of transistors on a dense integrated circuit doubles about every two years [30]. Although the statement was initially just a prediction by Moore in 1965 to get across the idea that electronics is going to get cheap faster [31], it became an empirical law for the semiconductor industry to keep pushing for smaller feature sizes on a silicon wafer. Amazingly Moore’s law has remained applicable for more than 50 years and even today continues to set predictions for the seemingly inexhaustible capacity of exponential growth in number of transistors on dense integrated circuits [32].

The smallest possible angular resolution, or resolving power, of any optical system is set by the Rayleigh criterion

$$R \sim \frac{\lambda}{D}, \quad (1.1)$$

where λ is the wavelength of the light and D is the diameter of the lens' aperture. Therefore in order to pattern smaller features, one must either use a large optical aperture or a short wavelength light source. Prior to 1990, lithography machines used visible g-line 436nm (violet) and i-line 365nm (ultraviolet) light produced by mercury lamps [28]. As feature sizes shrank to sub micron ($< 1\mu\text{m}$), the industry moved to 248nm and 193nm (deep ultraviolet) light sources produced by krypton or argon fluoride lasers, see figure 1.3a. The current state-of-the-art EUVL machines took yet another big step and use Laser-Produced Plasma (LPP) EUV light sources at an operating wavelength of 13.5nm. This step however involved overcoming many technological challenges and a complete redesign of the lithography machine, since light at this wavelength is instantly absorbed by all matter and requires dedicated optics. Consequently the full optical path from the LPP-EUV light source to the wafer stage needed to be housed in a near-vacuum environment and special mirrors, e.g. molybdenumsilicon multilayer mirrors, are used to guide the beam to its destination [33].

One technical challenge that remains unsolved today is the consistent power scaling of the EUV light beyond 250W output power [34, 35]. Fundamental understanding of the fluid and plasma processes inside the LPP-EUV light source is crucial to maximize the EUV output power. In this thesis we therefore focus on two important fluid mechanics processes inside the LPP-EUV light source for which we present both analytic and numerical modelling.

1.1.1 Fluid dynamics inside LPP-EUV light sources

Figure 1.3b shows a schematic overview of a modern LPP-EUV light source [33]. A droplet generator produces a train of liquid tin droplets, several $10\mu\text{m}$ in diameter, that falls down into the source chamber where each individual droplet is hit by a CO_2 laser in two stages. Initially a low energy pre-pulse hits the droplet and induces a rapid phase change in a thin superficial layer on the illuminated side of the droplet. The resulting mass ablation generates a recoil pressure and propels and deforms the tin droplet into a disk-like target [36–38]. The newly shaped target increases the coupling with a second high-energy main-pulse that leads to the formation of a high-density tin plasma in the LPP-EUV source chamber. Line emission from the charged tin ions in the plasma radiate EUV light peaking at 13.5nm [39]. A multilayer collector focuses the resulting EUV radiation at the intermediate focus point, after which multilayer mirrors of the lithography machine guides the beam to the wafer-stage.

The resulting tin debris after plasma formation is captured by a tin catching device at the bottom of the LPP-source where most of the tin debris is headed. Several vanes are added in the source chamber to prevent contamination of the highly sensitive optics. Any tin debris contamination on optics results in a severe loss of EUV radiation power output and must be suppressed.

In order to make ever smaller feature sizes on a silicon wafer, the EUV output power of the LPP-EUV light source must be maximized. It is therefore important to maximize the coupling between the tin droplet and the CO₂ laser impact and to minimize any tin debris that could contaminate sensitive optics in the source chamber. To this end, fundamental understanding of the droplet deformation dynamics after laser impact and the splashing dynamics of a tin droplet onto a pool is required and lead to a better understanding of the conversion efficiency of the CO₂ laser light into the required EUV light. Droplet deformation and splashing can be described in terms of two dimensionless quantities that we will use throughout this thesis. These scales allow us to characterize the fluid-dynamic response in a single and universal way, i.e. independent of laser parameters, liquid properties or droplet sizes and velocities. More importantly, these scales allow us to compare different experiments by simply matching their dimensionless numbers [36–38]. Following the Buckingham- π -theorem [40], the important physical variables in this problem are the liquid density of the droplet ρ , the surface tension σ , the dynamic viscosity μ , the droplet velocity U and the droplet diameter D whereas the three independent physical units are the length, mass and time. Therefore we can identify two dimensionless groups given by the Reynolds number

$$\text{Re} = \frac{\rho DU}{\mu}, \quad (1.2)$$

which relates the fluid inertia forces to the viscous forces, and the Weber number

$$\text{We} = \frac{\rho DU^2}{\sigma}, \quad (1.3)$$

which relates the kinetic energy to the surface energy of the droplet. We note that in this analysis we implicitly assumed that the laser pulse duration τ_p , wavelength λ , intensity I_p and beam profile are captured by a single physical variable: the propulsion speed of the droplet U [38]. Table 1.1 shows the liquid properties and a typical range of dimensionless numbers that are encountered inside the LPP-EUV source.

1.1.2 Laser impact on droplets

Figure 1.4 shows the impact of a Nd:YAG laser pulse on magenta-dyed water droplets and shows the deformation and fragmentation process for three different foci. The top panel shows that laser impact with a tightly focussed beam leads to the formation of a localized water plasma identified by the white glow. The impact is accompanied

Figure 1.4: The impact of a frequency-doubled Nd:YAG laser pulse of wavelength $\lambda = 532 \text{ nm}$ and pulse duration $\tau_p = 10 \text{ ns}$ on magenta-dyed water droplets with initial radii $R_0 = 0.9 \text{ mm}$. (a) A tightly focussed beam leads to the formation of a local water plasma and subsequently violent ablation from the droplet. Furthermore, a shock-wave is generated and visible in air at $t = 30 \mu\text{s}$. The impulsive impact leads to a severe deformation and fragmentation of the droplet on later stages $t = 50 \mu\text{s}$ and $t = 90 \mu\text{s}$. (b) A moderately focussed beam leads to the formation of a mist cloud and a yellow fluorescence emission of the magenta dye. The recoil pressure resulting from the phase change causes the droplet to deform in a strongly curved liquid sheet. (c) The uniform laser irradiation along the droplet surface leads to the formation of a symmetric mist cloud. The recoil pressure in this case causes the droplet to be propelled to the right and deform into an umbrella-shaped target. Image taken from [41].

by a shock-wave in air, which is clearly visible at $t = 30 \mu\text{s}$. Invisible however is the shock-wave that travels inside the droplet that leads to a severe deformation and fragmentation of the droplet on later stages. In the center panel the laser impact leads to the formation of a mist cloud due to localized boiling of water for a moderately focussed beam [41]. The resulting recoil-pressure on the surface deforms the droplet in a curved liquid sheet on a timescale that is larger than the timescale on which pressure waves travel inside the droplet. Finally in the bottom panel, an uniform laser irradiation over the droplet surface leads to a symmetric mist cloud along the laser-impact axis. In this case, the pressure field inside the droplet is always well established during the deformation of the droplet and therefore acoustics do not play a significant role.

A possible way to increase the power output of the LPP-EUV light source is to increase the number of droplets produced by the droplet generator. As a result, we must also speed up the conversion of tin into a plasma to keep up with the increase in throughput of falling droplets. To this end, the laser pre-pulse duration is shortened or the intensity increased such that the deformation into a thin flat sheet is faster. Detailed understanding of this deformation process leads to a better coupling with the second high-energy laser impact. Therefore, in this thesis we analytically and numerically study the laser impact onto a droplet in a regime where the recoil-pressure duration τ_e is short or the recoil-pressure amplitude p_e is large. In this regime Weber number and Reynolds number alone are not sufficient to describe the droplet deformation since acoustics inside the droplet starts to play a crucial role, as seen in figure 1.4a. Two additional dimensionless groups are needed to describe the deformation dynamics: The Strouhal number

$$\text{St} = \frac{c\tau_e}{D}, \quad (1.4)$$

where c is the speed of sound inside the droplet and the acoustic Mach number

$$\text{Ma} = \frac{p_e}{\rho c^2}. \quad (1.5)$$

The Strouhal number is a measure for the distance a pressure wave has travelled inside the droplet compared to the diameter of the droplet. When the Strouhal number is small, the pressure wave inside the droplet after the recoil pressure has not yet travelled a distance D and the front side of the droplet gets compressed. By contrast, when $\text{St} \gg 1$ the pressure field inside the droplet is well established on the timescale of the pulse duration. The acoustic Mach number is a measure for the amplitude of the pressure wave inside the droplet compared to the base pressure of the droplet. When the acoustic Mach number is large the flow inside the droplet is non-linear and shock waves dominate the flow, while when the acoustic Mach number is small the flow can be considered linear. In the regime where the pulse duration is short, but still a significant amount of momentum is transferred to the droplet, the flow could be governed by shock-waves, cavitation phenomena, non-linear viscous damping and rapid interface acceleration. This leads to new flow regimes that are not present in the case when the pre-pulse parameters are such that the flow inside the deforming droplet is incompressible and linear.

Another possible way to increase the LPP-EUV power output is to understand how to reduce the laser-to-droplet alignment sensitivity. Any small laser misalignment from the optimum impact location on the droplet leads to a tilted droplet deformation with a smaller radial expansion and hence to a lower conversion efficiency of the light source. Therefore in this thesis we analytically and numerically study the effects of a relative misalignment between the laser beam and droplet and the resulting droplet deformation. In this work we present a model to calculate the tilt angle

Figure 1.5: An experimental time series of a droplet impacting onto a deep liquid pool with $U = 22\text{m/s}$ under an angle of $\alpha = 28^\circ$. (a) Directly after impact an ejecta sheet forms that splashes on the right hand side of the impact zone. (b) A single sided crown develops whose fingers break and also result in satellite droplets. Below the pool surface a hemispherical cavity develops. (c) The crown collapses and capillary waves are formed inside the cavity, see arrow. (d) The capillary waves focus off-center. (e) A bubble is entrapped into the liquid pool, see arrow, and the cavity starts to retract. (f) A Worthington jet is formed during the closure of the cavity. Image taken from [25].

sensitivity, i.e. the change in droplet tilt angle as function of the misalignment, as function of a single dimensionless parameter: the ratio of the laser spot size to the position of the plasma critical surface. This model can be used to enhance the pre-pulse laser parameters such that the tilt angle sensitivity is minimized in LPP-EUV light sources, which maximizes the radial expansion of the target shape and therefore leads to a better coupling with the second high energy main pulse.

1.1.3 Droplet impact onto deep liquid pools

The tin debris formed after plasma formation is captured by a tin catching device at the bottom and vanes at the walls of the source chamber to prevent the contamination of the sensitive optics in the LPP-EUV light source. While these catchers prevent the debris to directly hit the sensitive optics, they do not prevent secondary contamination

parameter	unit	tin	water
ρ	liquid density (kg/m ³)	6920	998
σ	surface tension (N/m)	0.544	0.072
μ	dynamic viscosity (kg/m s)	1.85×10^{-3}	8.90×10^{-4}
c	speed of sound (m/s)	2.5×10^3	1.41×10^3
Laser impact on droplets			
D	initial droplet diameter (m)	50×10^{-6}	2×10^{-3}
U	typical droplet propulsion velocities (m/s)	10 – 300	2 – 50
We	Weber numbers	$10^2 - 10^5$	
Re	Reynolds numbers	$10^3 - 10^5$	
Ma	Acoustical Mach numbers	$10^{-1} - 10^1$	
St	Strouhal numbers	$10^{-1} - 10^1$	
Droplet impact on pools			
D	initial droplet diameter (m)	6.5×10^{-6}	1×10^{-4}
U	typical droplet impact velocities (m/s)	10 – 100	5 – 30
We	Weber numbers	$10^2 - 10^3$	
Re	Reynolds numbers	$10^2 - 10^3$	

Table 1.1: The liquid properties of tin and water droplets used in this thesis. For the laser impact phenomenon we have included typical propulsion velocities and droplet diameters encountered in experiments with water droplets [36] and tin droplets [38]. For the droplet impact onto a deep liquid pool we used the typical impact velocities and droplet diameters for experiments in water [25] and estimations for the tin debris encountered in the LPP-EUV source chamber.

of the optics originating from, e.g., splashing of impacting debris. It is therefore important to study the splashing behaviour of tin debris depending on the impact angle and the impact velocity. In this thesis we focus on the impact dynamics of oblique droplet impact onto a deep liquid pool. Figure 1.5 shows an experimental time series of a water droplet impacting onto a deep liquid pool under an angle. In figure 1.5a an ejecta sheet is formed directly after impact, but only at the right hand side of the impact zone. The ejecta sheet breaks at the tip and small satellite droplets are formed, as clearly seen in figure 1.5b. In figure 1.5c the ejecta sheet collapses back to the pool and a capillary wave is created that travels along the surface into the cavity and entraps a bubble in figure 1.5d-e. Finally in figure 1.5f a thick jet is created that in some cases could also result in a splash.

In this work we use the adaptive flow solver Basilisk to study the splashing thresh-

old, cavity formation, cavity evolution and the maximum cavity dimensions as a function of the Weber number and the impact angle directly after impact. We validate and compare our numerical results with recent experiments by [25]. The numerical simulations provide a valuable addition to the experimental results, since we have access to the pressure and velocity fields induced in the liquid. This allows for a detailed understanding of the onset of the sheet dynamics and the resulting breakup, allows for a better judgment on the single-sided and omni-directional splashing thresholds. Furthermore we can use the numerical method to explore impact parameters that are hard to reach experimentally, e.g. very large impact angles or high droplet velocities.

Fundamental insight in the splashing behaviour of the tin debris may lead to a better control on the secondary contamination by splashing after impact, which leads to cleaner optics and thus a better conversion efficiency of the LPP-EUV light source.

1.2 Guide through the thesis

In chapter 2 we present a numerical method to study the propagation of pressure waves inside a droplet that has been hit by a laser pulse. The presented axisymmetric multiphase lattice-Boltzmann method is capable of solving liquid-gas density ratios up to 10^3 and gives fundamental insight in the initial deformation dynamics of the droplet. In chapter 3 we derive an acoustic analytic model for the pressure, pressure impulse and velocity fields inside the droplet after laser impact in the limit of small density fluctuations. This model is used to examine how the droplet deformation depends on the pressure pulse duration while the total momentum transferred to the droplet is kept constant. We demonstrate that when the momentum transfer is increased, the flow becomes compressible for short pulse durations, which strongly alters the deformation dynamics compared to long pulse durations. We use the axisymmetric multiphase lattice-Boltzmann model of chapter 2 to support and validate our analytical findings. In chapter 4 we introduce and experimentally validate an analytical model describing the sensitivity of the tilt angle, expansion and propulsion velocity of a tin micro-droplet irradiated by a $1\mu\text{m}$ Nd:YAG laser pulse to its relative laser alignment. We give an analytic expression for the tilt angle sensitivity that is particularly relevant to optimize and understand the disk-like target in LPP-EUV light sources. In chapter 5 we present a numerical study the oblique droplet impact onto deep liquid pools. We quantify the splashing threshold, cavity formation, cavity evolution and the maximum cavity dimensions as function of the Weber number and the impact angle. We compare the numerical results with the experimental results by [25]. Furthermore, we give a detailed overview of the velocity magnitude inside the ejecta sheet directly after impact for different Weber numbers and impact angles. Finally, in chapter 6 we introduce a novel semi-analytic solving method that can potentially be used to get a deeper understanding of the compressible and non-linear

flow regimes inside the droplet after laser impact or after oblique droplet impact onto a deep liquid pool. As a demonstration in this chapter, we have applied the method to the one-dimensional Burgers equation. We present a novel analytic and bounded solution to the Burgers equation for a simple trigonometric initial condition in an infinite domain.

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2

Axisymmetric multiphase Lattice Boltzmann method for generic equations of state *

We present an axisymmetric lattice Boltzmann model based on the Kupershtokh et al. multiphase model that is capable of solving liquid-gas density ratios up to 10^3 . Appropriate source terms are added to the lattice Boltzmann evolution equation to fully recover the axisymmetric multiphase conservation equations. We validate the model by showing that a stationary droplet obeys the Young-Laplace law, comparing the second oscillation mode of a droplet with respect to an analytical solution and showing correct mass conservation of a propagating density wave.

2.1 Introduction

The lattice Boltzmann method (LBM) [1, 2] is an efficient numerical tool to solve the Navier-Stokes equations. This numerical method can be systematically derived from the Boltzmann equations by means of a Hermite expansion approach [3]. In many physically realistic flow problems one has to deal with multiphase flows such as the contact angle hysteresis of a moving droplet on a surface, a capillary rise in a cylindrical tube and droplet impact on solid surfaces. To this end, several extensions have been proposed to support multiphase flows in the LBM. In an early attempt, Gunstensen *et al.* [4] studied a two-component fluid lattice-gas method. Shan *et al.* [5, 6]

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were the first to incorporate intermolecular interactions to achieve phase separation in LBM. A different approach to model a multiphase fluid was developed by Swift *et al.* [7], who associated a free energy functional to the fluid. In their original form, these models lack the ability to achieve high density ratios across fluid interfaces and suffer from spurious currents near the liquid-vapour interface. In many engineering applications density ratios range from $10^1 - 10^3$, posing a serious limitation to the applicability of these lattice Boltzmann models in their original form. Recently, Lee *et al.* [8] showed that the spurious currents are caused by discretization errors in the computation of the multiphase force. These spurious currents can be reduced to machine precision by employing a potential form of the non-ideal pressure and a isotropic central difference approximation scheme for the multiphase force. Kupershtokh *et al.* [9] showed that it is possible to achieve density ratios of 10^6 when the multiphase force is discretized by just a single-neighbour discretization scheme. The ability to achieve high density ratios makes this model applicable to many engineering applications. However, in this scheme the surface tension cannot be varied independently and spurious currents still exist.

Recently the LBM was extended to support axisymmetric multiphase flows. These axisymmetric simulations are effectively 2D simulations in a cylindrical coordinate system. Therefore, the computational cost for axisymmetric 3D flow problems is significantly lower in comparison to the same problem in a full 3D simulation. Halliday *et al.* [10] was the first to implement an axisymmetric LBM for single-phase flows. They introduced additional source and sink terms to the evolution equation and showed that they recover the 2D axisymmetric Navier-Stokes equations. This model was improved by Lee *et al.* [11] who corrected a missing source term related to the radial velocity. In addition, the method of Halliday *et al.* was extended to support non-ideal flows. Premnath *et al.* [12] were the first to implement an axisymmetric multiphase LBM. Their model is able to achieve density ratios up to 10 and was further improved by Mukherjee *et al.* [13] to support density ratios up to 10^3 and perform stable computations at lower viscosities. In this improved model, they use a pressure-evolution based LBM combined with a multiple-relaxation-time (MRT) collision model. Another axisymmetric multiphase LBM model by Srivastava *et al.* [14] is based upon the widely used Shan-Chen model. In this model, they add an extra contribution to the Shan-Chen multiphase force to fully recover three-dimensionality in the system. However, large density ratios (> 30) could not be achieved due to the limits of the original Shan-Chen model.

In this chapter, we introduce a novel and easy-to-implement axisymmetric isothermal multiphase model for high density ratio fluids. The proposed model is based on the axisymmetric LBM of Srivastava *et al.* [14] combined with the multiphase model of Kupershtokh *et al.* [9]. The combined model inherits all advantages and disadvantages of the existing multiphase model by Kupershtokh *et al.*, which we will not discuss in detail here. An extensive study to the accuracy and stability of the Ku-

pershtokh *et al.* multiphase method can be found in [15]. Our implementation is discussed in section II. In section III we present three validation tests. First, we verify that a stationary droplet obeys the Young-Laplace law. Then, we compare the second oscillation mode of an oscillating droplet with an incompressible analytical solution. Finally, we show that the method correctly describes the propagation of a density wave towards and away from the longitudinal z-axis. Our main conclusions and limitations of the method are discussed in section IV.

2.2 Model derivation

We first introduce the standard LBM. In the following sub-sections, we will gradually show the changes necessary to obtain a fully functional axisymmetric isothermal multiphase LBM.

2.2.1 The lattice Boltzmann method

We use the common D2Q9 LBM, based on a two-dimensional Eulerian lattice with nine velocities. For the time evolution of the distribution function f_i , we use the BGK approximation with a single relaxation parameter τ [2]. The time evolution is given by

$$f_i(\mathbf{x} + \mathbf{e}_i \delta t, t + \delta t) = f_i(\mathbf{x}, t) + \frac{\delta t}{\tau} (f_i^{\text{eq}}(\mathbf{x}, t) - f_i(\mathbf{x}, t)) + \delta t S_i(\mathbf{x}, t), \quad (2.1)$$

where $\mathbf{x}$ is the position, t is the time, δt is the time step, τ is the relaxation time, $S_i(\mathbf{x}, t)$ is a source term, f_i^{eq} is the local equilibrium distribution and $\mathbf{e}_i$ is a discrete velocity set given by

$$\mathbf{e}_i = \begin{cases} (0, 0) & i = 0, \\ (1, 0)_{\text{FS}} & i = (1, 2, 3, 4), \\ (\pm 1, \pm 1) & i = (5, 6, 7, 8), \end{cases}$$

where the subscript _{FS} denotes a fully symmetric set of points. The local equilibrium distribution function f_i^{eq} is a second-order Taylor expansion of the Maxwell-Boltzmann distribution [2] and is given by

$$f_i^{\text{eq}}(\mathbf{x}, t) = w_i \rho(\mathbf{x}, t) \left[1 + \frac{1}{c_s^2} (\mathbf{e}_i \cdot \mathbf{u}(\mathbf{x}, t)) + \frac{1}{2c_s^2} \left(\frac{1}{c_s^2} (\mathbf{e}_i \cdot \mathbf{u}(\mathbf{x}, t))^2 - \|\mathbf{u}(\mathbf{x}, t)\|^2 \right) \right], \quad (2.2)$$

where $c_s^2 = \frac{1}{3}$ is the lattice speed of sound in this single-phase model and w_i are the quadrature weights given by

$$w_i = \begin{cases} 4/9 & i = 0, \\ 1/9 & i = (1, 2, 3, 4), \\ 1/36 & i = (5, 6, 7, 8). \end{cases}$$

The hydrodynamic quantities of the fluid, such as density ρ and velocity $\mathbf{u}$ are calculated as weighted sums of the distribution function f_i

$$\rho(\mathbf{x}, t) = \sum_i f_i(\mathbf{x}, t), \quad (2.3)$$

$$\mathbf{u}(\mathbf{x}, t) = \frac{\delta t \mathbf{F}(\mathbf{x}, t)}{2\rho(\mathbf{x}, t)} + \sum_i \frac{\mathbf{e}_i f_i(\mathbf{x}, t)}{\rho(\mathbf{x}, t)}, \quad (2.4)$$

where $\mathbf{u}(\mathbf{x}, t)$ is shifted by means of an internal/external force $\mathbf{F}$. In the past, different implementations of a body force, $\mathbf{F}$, were proposed [16]. Here we use the forcing scheme by Guo *et al.* [17]

$$S_i(\mathbf{x}, t) = w_i \left(1 - \frac{\delta t}{2\tau} \right) \left(\frac{(\mathbf{e}_i - \mathbf{u}) \cdot \mathbf{F}}{c_s^2} + \frac{(\mathbf{e}_i \cdot \mathbf{u})(\mathbf{e}_i \cdot \mathbf{F})}{c_s^4} \right). \quad (2.5)$$

2.2.2 The extension to an axisymmetric method

Figure 2.1: Schematics of the axisymmetric geometry in our LBM. The Cartesian coordinates (x, y) are replaced with the axisymmetric coordinates (z, r) . N_r is the length of the domain in the r -direction and N_z is the length in the z -direction.

In an axisymmetric flow, there is no flow in the azimuthal direction ($u_\theta = 0$) and mass conservation reads

$$\frac{\partial \rho}{\partial t} + \nabla_c \cdot (\rho \mathbf{u}) = -\frac{\rho u_r}{r} \quad (2.6)$$

where $\nabla_c \equiv (\frac{\partial}{\partial z}, \frac{\partial}{\partial r})$ is the gradient operator in a two-dimensional Cartesian coordinate system ($x \rightarrow z, y \rightarrow r$) and $\mathbf{u} = (u_z, u_r)$ is the fluid velocity. The momentum equation reads

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla_c \mathbf{u} \right) = -\nabla_c P + \mu \nabla_c \cdot [\nabla_c \mathbf{u} + \nabla_c \mathbf{u}^T] + \mathbf{C} \quad (2.7)$$

where P is the fluid pressure which in a single-phase LBM is given by $P = c_s^2 \rho$ and $\mathbf{C}$ is given by

$$C_z = \frac{\mu}{r} \left(\frac{\partial u_z}{\partial r} + \frac{\partial u_r}{\partial z} \right), \quad C_r = 2\mu \frac{\partial}{\partial r} \left(\frac{u_r}{r} \right), \quad (2.8)$$

with μ the fluid viscosity. It is clear that equations (2.6,2.7) have additional contributions to the mass and momentum conservation equations in comparison to 2D flow in the (z, r) -plane. These contributions ensure local conservation of mass and momentum when fluid is moving towards or away from the longitudinal z -axis. The single-phase LBM can be supplemented with appropriate source-terms to recover the axisymmetric conservation equations (2.6,2.7) [14]. To this end, the evolution equation (2.1) is rewritten with an additional source term h_i

$$f_i(\mathbf{x} + \mathbf{e}_i \delta t, t + \delta t) = \frac{\delta t}{\tau} (f_i^{\text{eq}}(\mathbf{x}, t) - f_i(\mathbf{x}, t)) + f_i(\mathbf{x}, t) + \delta t S_i(\mathbf{x}, t) + \delta t h_i(\mathbf{x} + \mathbf{e}_i \frac{\delta t}{2}, t + \frac{\delta t}{2}). \quad (2.9)$$

where h_i is evaluated at fractional time steps. Srivastava *et al.* [14] showed by means of a Chapman-Enskog (CE) expansion that when h_i has the following form

$$h_i = w_i \left(-\frac{\rho u_r}{r} + \frac{1}{c_s^2} (e_{iz} H_z + e_{ir} H_r) \right), \quad (2.10)$$

with $\mathbf{e}_i = (e_{iz}, e_{ir})$ and

$$H_z = \frac{e_{iz}}{r} \left(\mu \left(\frac{\partial u_z}{\partial r} + \frac{\partial u_r}{\partial z} \right) - \rho u_r u_z \right), \quad (2.11a)$$

$$H_r = \frac{e_{ir}}{r} \left(2\mu \left(\frac{\partial u_r}{\partial r} + \frac{u_r}{r} \right) - \rho u_r^2 \right), \quad (2.11b)$$

the resulting LBM solves the axisymmetric conservation equations given by (2.6,2.7) in the limit of small Mach number. The velocity derivatives inside (2.11) are approximated by a isotropic fifth-order accurate finite difference scheme that will be given below (2.22). To have consistent boundary conditions, we use a symmetry boundary condition for the derivative evaluation at the axis and we impose a zero derivative for all other boundaries.

Figure 2.2: The co-existence curve relating the equilibrium densities ρ_v and ρ_l to the temperature T for $A = 0$ (black diamonds and blue squares) and $A = -0.152$ (orange diamonds and green squares). The simulation is capable of achieving density ratios up to 10^3 and beyond. Simulation parameters: $N_z \times N_r = 150 \times 100$, $\tau = 1.0$, $\lambda = 0.01$.

2.2.3 The extension to a multiphase axisymmetric method

We employ a temperature-dependent body force to obtain a multiphase fluid [5]. Zhang *et al.* [18] proposed a body force of the form

$$\mathbf{F}(\mathbf{x}, t) = -\nabla U(\mathbf{x}, t), \quad (2.12)$$

for which the overall effective fluid pressure in the system becomes

$$P(\mathbf{x}, t) = c_s^2 \rho(\mathbf{x}, t) + U(\mathbf{x}, t). \quad (2.13)$$

In this notation, it is evident that a particular equation of state (EOS) P_k can be obtained by simply adapting the choice for U accordingly

$$U(\mathbf{x}, t) = P_k(\mathbf{x}, t) - c_s^2 \rho(\mathbf{x}, t). \quad (2.14)$$

In this chapter, we will only consider the dimensionless van der Waals (vdW) EOS

$$P_k = \lambda \left(\frac{8\rho T}{3-\rho} - 3\rho^2 \right), \quad (2.15)$$

$$c_k = \sqrt{\lambda \left(\frac{24T}{(\rho-3)^2} - 6\rho \right)}, \quad (2.16)$$

where $T = T_{\text{vdw}}/T_c$ is an effective temperature and T_c the critical temperature, c_k is the thermodynamic speed of sound at constant entropy, $\rho = \rho_{\text{vdw}}/\rho_c$ is the density and ρ_c the critical density, $P = P_{\text{vdw}}/P_c$ is the pressure and P_c the critical pressure and $\lambda = \frac{P_c}{\rho_c} \left(\frac{\delta t}{\Delta x} \right)^2$ is a scaling parameter. Kupershtokh *et al.* [9] showed that for this vdW-EOS, the theoretical co-existence curve can be fully reconstructed by using the Exact-Difference-Method (EDM) forcing scheme and a special discretization for the body force

$$\mathbf{F}(\mathbf{x}) = \frac{18}{3\delta t} \left(A \sum_i w_i \psi^2(\mathbf{x} + \mathbf{e}_i \delta t) \mathbf{e}_i + (1 - 2A) \psi(\mathbf{x}) \sum_i w_i \psi(\mathbf{x} + \mathbf{e}_i \delta t) \mathbf{e}_i \right), \quad (2.17)$$

$$\psi(\mathbf{x}) = \sqrt{-P_k + \frac{\rho}{3}}. \quad (2.18)$$

where A is a tunable parameter. For $A = 0$ the scheme coincides with a local approximation scheme and for $A = 0.5$ with a mean-value approximation. The vdW co-existence curve is recovered by setting $A = -0.152$ [9].

By means of a Taylor expansion for $\psi(\mathbf{x} + \mathbf{e}_i \delta t)$ one can find a continuum expression for the body force

$$\mathbf{F}(\mathbf{x}) \approx \frac{18c_s^2}{3} \left(\psi(\mathbf{x}) \nabla \psi(\mathbf{x}) + \frac{(\delta t)^2 c_s^2}{2} \left[\psi(\mathbf{x}) \nabla \nabla^2 \psi(\mathbf{x}) + 6A \nabla \psi(\mathbf{x}) \nabla^2 \psi(\mathbf{x}) \right] \right) + \mathcal{O}((\delta t)^4). \quad (2.19)$$

To fully recover axisymmetry in the multiphase force, we need to use correction terms, similar to what has been done for the mass- and momentum conservation equations [14]. In cylindrical coordinates, the Laplace operator is given by $\nabla^2 \equiv \frac{\partial^2}{\partial r^2} + \frac{\partial^2}{\partial z^2} + \frac{1}{r} \frac{\partial}{\partial r} = \nabla_c^2 + \frac{1}{r} \frac{\partial}{\partial r}$. Therefore, we can rewrite (2.19) as

$$\mathbf{F}(\mathbf{x}) \approx \frac{18c_s^2}{3} \left(\psi(\mathbf{x}) \nabla_c \psi(\mathbf{x}) + \frac{(\delta t)^2 c_s^2}{2} \left[\psi(\mathbf{x}) \nabla_c \nabla_c^2 \psi(\mathbf{x}) + 6A \nabla_c \psi(\mathbf{x}) \nabla_c^2 \psi(\mathbf{x}) \right] \right) + \mathbf{F}_{\text{axis}}(\mathbf{x}) + \mathcal{O}((\delta t)^4), \quad (2.20)$$

where $\mathbf{F}_{\text{axis}}$ is given by

$$\mathbf{F}_{\text{axis}}(\mathbf{x}) = 3(\delta t)^2 c_s^4 \left(\psi(\mathbf{x}) \nabla_c \left[\frac{1}{r} \frac{\partial \psi(\mathbf{x})}{\partial r} \right] + \frac{6A}{r} \frac{\partial \psi(\mathbf{x})}{\partial r} \nabla_c \psi(\mathbf{x}) \right), \quad (2.21)$$

and can be identified as the cylindrical contribution to the multiphase force.

The meaning of c_s in the Taylor expansion that leads to (2.21) is ambiguous. When the lattice speed of sound (c_s) is used in (2.21), our validation tests for the surface tension showed a discrepancy in the range of 200-260% in comparison with full 2D simulations. By substituting the thermodynamic speed of sound c_k in (2.21) instead, we observe better consistency between the axisymmetric simulations and fully 2D simulations as is shown in Table 2.1 of the next section. Therefore, we use this substitution in the remainder of the paper. We attribute this ambiguity to the current mathematical formulation of the multiphase LBM. We use a lattice Boltzmann scheme that is derived from a single-phase ideal gas in which the speed of sound is assumed constant. However, by imposing a non-ideal EOS the speed of sound changes with density, which hence invalidates the constant lattice speed of sound. Unfortunately a mathematical proof of the validity of our substitution cannot be constructed at this moment and is left for future work.

The evaluation of $\mathbf{F}_{\text{axis}}$ requires an approximation for the derivatives of ψ accurate up to order $(\delta t)^4$. Therefore, we use the following isotropic fifth-order accurate finite difference approximations given by (2.22) [14]. We use a symmetry boundary condition for the derivative evaluation at the axis and we impose a zero derivative for all other boundaries.

$$\frac{\partial \psi(\mathbf{x})}{\partial r} = \frac{1}{36} \sum_{i=1}^8 \left(8\psi(\mathbf{x} + \mathbf{e}_i \delta t) - \psi(\mathbf{x} + 2\mathbf{e}_i \delta t) \right) c_{ir} + \mathcal{O}((\delta t)^5), \quad (2.22a)$$

$$\frac{\partial^2 \psi(\mathbf{x})}{\partial r^2} = \frac{1}{36} \sum_{i=1}^8 \left(\frac{8\partial \psi(\mathbf{x} + \mathbf{e}_i \delta t)}{\partial r} - \frac{\partial \psi(\mathbf{x} + 2\mathbf{e}_i \delta t)}{\partial r} \right) c_{ir} + \mathcal{O}((\delta t)^5), \quad (2.22b)$$

$$\frac{\partial \psi(\mathbf{x})}{\partial z} = \frac{1}{36} \sum_{i=1}^8 \left(8\psi(\mathbf{x} + \mathbf{e}_i \delta t) - \psi(\mathbf{x} + 2\mathbf{e}_i \delta t) \right) c_{iz} + \mathcal{O}((\delta t)^5), \quad (2.22c)$$

$$\frac{\partial^2 \psi(\mathbf{x})}{\partial r \partial z} = \frac{1}{36} \sum_{i=1}^8 \left(\frac{8\partial \psi(\mathbf{x} + \mathbf{e}_i \delta t)}{\partial z} - \frac{\partial \psi(\mathbf{x} + 2\mathbf{e}_i \delta t)}{\partial z} \right) c_{ir} + \mathcal{O}((\delta t)^5). \quad (2.22d)$$

2.3 Results and Validation

Our model is capable of achieving density ratios up to 10^3 . The co-existence curve relating the equilibrium vapour density ρ_v and liquid ρ_l density to the temperature T is plotted in figure 2.2. In the remainder of this section we validate our axisymmetric isothermal multiphase LBM against three test cases. First, we compare the pressure difference across the liquid-vapour interface of a stationary droplet of radius R_0 for different temperatures T to the Young-Laplace law. Then, we compare the second oscillation mode of an oscillating droplet with its analytical solution. Finally, we show that mass is correctly conserved by simulating a propagating density wave travelling towards and away from the longitudinal z -axis.

T	$A = 0$			$A = -0.152$		
	σ_{lv}^{2D}	σ_{lv}^{axis}	σ_{lv}^{lcs}	σ_{lv}^{2D}	σ_{lv}^{axis}	σ_{lv}^{lcs}
0.9	0.305	0.304	0.606	0.371	0.383	0.965
0.8	0.928	0.928	1.865	1.146	1.158	2.884
0.7	1.983	2.009	3.909	2.285	2.318	5.681

Table 2.1: Surface tension evaluated using (2.23). Here, A denotes the tuning parameter for the multi-phase force, while σ_{lv}^{2D} and σ_{lv}^{axis} denote the surface tensions obtained from full (2D) and axisymmetric simulations. We find that the maximum relative error with 2D simulations is 1.3% for $A = 0$ and 3.2% for $A = -0.152$. An additional surface tension σ_{lv}^{lcs} was simulated by using the lattice speed of sound c_s in (2.21). These results show an maximum relative error of 201% for $A = 0$ and 260% for $A = -0.152$. Simulation parameters: $N_z \times N_r = 500 \times 200$, $\tau = 1.5$, $\lambda = 0.01$.

2.3.1 Stationary droplet validation

The pressure difference across the liquid-gas interface of a stationary 2D droplet with radius R_0 is given by the Young-Laplace equation

$$\Delta P_k = \frac{\sigma_{lv}}{R_0}, \quad (2.23)$$

where $\sigma_{lv} = \sigma_{vdw}/(P_c \Delta x)$ is the surface tension between the liquid-vapour phase, $R_0 = R/\Delta x$ is the stationary droplet radius and $\Delta x = 1$ is the lattice spacing in our LBM. Note that there is only one radius of curvature in a two-dimensional system. In this validation test, we check if the value of the surface tension in the present model σ_{lv}^{axis} is consistent with a full 2D simulation σ_{lv}^{2D} . To this end, we perform simulations of a stationary droplet with radius $R_0 = 150$ (lattice units) for three different temperatures T and two coupling constants A . As boundary conditions for the axisymmetric simulation we use mid-grid specular reflection boundary condition on the longitudinal z -axis, periodic boundary conditions in the r direction and a free-slip boundary condition at $r = N_r$. In the 2D lattice Boltzmann simulation, we used periodic boundary conditions on all sides.

The results of these simulations are summarized in Table 2.1. When comparing the surface tension values between the full 2D and the axisymmetric simulation, we find that the maximum error is 1.3% for $A = 0$ and 3.2% for $A = -0.152$, respectively. Our axisymmetric multiphase LBM is in excellent agreement with the full 2D counterpart.

Figure 2.3: The temporal evolution of the inter-facial position along the longitudinal z -axis for four different initial conditions. We normalized the time by the capillary time $t_c = \sqrt{\frac{\rho R_0^3}{\lambda \sigma_{lv}}}$ and the droplet radius R by its equilibrium radius R_0 . Simulation parameters: $N_z \times N_r = 500 \times 250$, $\tau = 0.7$, $T = 0.8$, $A = 0.0$, $\lambda = 0.01$, $\rho_l/\rho_v \approx 10$.

2.3.2 Oscillating droplet validation

In this validation test we consider the problem of an oscillating axisymmetric droplet immersed in a gas. An analytical solution to the frequency and rate of damping of an oscillating droplet for arbitrary droplet radii, viscosity and surface tension was obtained by Miller and Scriven [19] in the limit of an isothermal, incompressible and Newtonian fluid. Here, we consider only the second mode which is axisymmetric

$$\omega_2 = \omega_2^* - \frac{\alpha(\omega_2^*)^{1/2}}{2} + \frac{\alpha^2}{4}, \quad (2.24)$$

where

$$\omega_2^* = \sqrt{\frac{24\lambda\sigma_{lv}}{R_0^3(2\rho_v + 3\rho_l)}}, \quad (2.25)$$

and

$$\alpha = \frac{25\sqrt{v_l v_v} \rho_l \rho_v}{\sqrt{2} R_0 (2\rho_v + 3\rho_l) (\sqrt{v_l} \rho_l + \sqrt{v_v} \rho_v)}, \quad (2.26)$$

with v_v and v_l the kinematic viscosities of the vapour and liquid phase, respectively.

Figure 2.4: The temporal evolution of the inter-facial position along the longitudinal z -axis for four different initial conditions. We normalized the time by the capillary time $t_c = \sqrt{\frac{\rho R_0^3}{\lambda \sigma_{lv}}}$ and the droplet radius R by its equilibrium radius R_0 . Simulation parameters: $N_z \times N_r = 500 \times 250$, $\tau = 0.72$, $T = 0.655$, $A = -0.152$, $\lambda = 0.005$, $\rho_l/\rho_v \approx 130$.

In this test, we initialized the fluid domain with an axisymmetric ellipsoidal drop, $(r/R_a)^2 + (z/R_z)^2 = 1$, where R_a, R_b are the semi-principal lengths of the ellipsoid. The liquid and gas densities are initialized as the equilibrium densities to the corresponding temperatures. We measure the time dependent radius $R(t)$ of the droplet along the longitudinal z -axis for 4 different initial ellipsoid radii, see figure 2.3 and figure 2.4. Our simulation domain consists of a free-slip boundary condition at the top, periodic boundary conditions along the sides and the mid-grid specular reflection boundary condition on the longitudinal z -axis. The oscillation frequency ω_2 can be calculated by fitting the temporal evolution of the inter-facial position with that of a damped harmonic oscillator $f(t) = a + b \exp(-ct) \sin(\omega_2 t + d)$, where the inter-facial position is picked as the point in space where $\rho = \frac{\rho_v + \rho_l}{2}$. We found that the frequency ω_2 for the four different droplet radii have a minimal relative error of 6% and a maximum relative error of 8% at density ratio $\rho_l/\rho_v \approx 10$ with respect to the incompressible analytical solution (2.24). For density ratio of $\rho_l/\rho_v \approx 130$, we found that the oscillation frequency ω_2 has a minimal relative error of 14.4% and a maximum relative error of 15.8%. Relative frequency errors ranging from 8% to 12% in oscillating droplets are also observed in other multiphase lattice Boltzmann

Figure 2.5: The disturbance density field ρ_1 as function of the position r is plotted at different time stages, where $t_w = \frac{c_k t}{w_d}$ is the dimensionless time. Simulation parameters: $N_z \times N_r = 1 \times 2000$, $\tau = 0.6$, $T = 0.8$, $\rho = 1.8$, $A = 0.0$, $\lambda = 0.01$, $\rho_l/\rho_v \approx 10$.

models [13, 20–23]. In order to show that this frequency error is not related to our axisymmetric implementation, we performed a full 2D oscillating droplet simulation at high density ratio. In this simulation, we used a square domain with 500 lattice units in each direction with periodic boundaries. We placed an ellipsoidal droplet with $R_0 = 150$ (lattice units) in the center of the domain with liquid and gas densities initialized as the equilibrium values to the corresponding temperature. The density ratio for this case was $\rho_l/\rho_v \approx 130$ with parameters $T = 0.655$, $\tau = 0.72$, $A = -0.152$ and $\lambda = 0.005$. The measured oscillation frequency ω_2 showed a relative error of 13.1% with respect to the analytical solution (2.24). We therefore conclude that the discrepancy in the oscillation frequency is not related to our axisymmetric implementation. We also excluded that the discrepancy comes from finite size effects, therefore we suppose it may be due to a discrepancy between the physics modelled in our simulation and the idealized incompressible analytical solution (2.24).

2.3.3 Wave propagation validation

In the limit of small density fluctuations $\rho_1/\rho_0 \ll 1$, the Navier-Stokes equations can be linearized up to first order, where ρ_1 is a density disturbance field and ρ_0 the background density [24]. The resulting cylindrical wave-equation for the disturbance

field $\rho_1(\mathbf{x}, t)$ in the inviscid limit reads

$$\frac{\partial^2 \rho_1}{\partial t^2} - c_k^2 \nabla_c^2 \rho_1 = \frac{c_k^2}{r} \frac{\partial \rho_1}{\partial r}. \quad (2.27)$$

Here, we check if the mass of a propagating density wave towards and away from the axis is conserved and obeys the wave-equation (2.27). The spatial temporal solution for the cylindrical wave-equation (2.27) with an initial condition $\rho(r, 0)$ and a no-slip boundary condition at $r = R$, is given by [25]

$$\rho_1(r, t) = \frac{2}{R^2} \int_0^R \sum_{n=1}^{\infty} \rho_1(r', 0) \frac{J_0(k_n r/R) J_0(k_n r'/R)}{J_1(k_n)^2} \cos\left(\frac{c_k k_n}{R} t\right) r' dr', \quad (2.28)$$

where k_n is the n th zero of $J_0(k) = 0$, R is the channel height and $\rho_1(r', 0)$ is the initial condition.

We initialized the fluid domain with a uniform density ρ_0 supplemented with a small disturbance ρ_1 field of width w_d and amplitude ρ_p in the center of the domain at $t_w = 0$, where $t_w = \frac{c_k t}{w_d}$. In contrast to the oscillating droplet test, here we used the no-slip boundary condition at the top boundary, periodic boundary conditions along the sides and the mid-grid specular reflection boundary condition on the longitudinal z -axis. Furthermore, we set $\tau = 0.6$ to be in the limit of small viscosity. Figure 2.5 shows the disturbance field ρ_1 as function of the position r at different time stages for both the simulation and analytic solutions (2.28). The initial density disturbance causes two propagating waves in the system: one wave travelling towards the longitudinal axis and one travelling away from the longitudinal axis. Figure 2.5 clearly shows that mass is conserved in our simulation: there is a mass build-up in the wave travelling towards the longitudinal axis and mass loss for the wave travelling away from the longitudinal axis. We observe that the perturbed density field in the simulation is in excellent agreement with the analytical solution. There is a small departure from the analytical solution at $t_w = 3.98$ as shown in figure 2.5. This departure is most likely caused by the dissipative nature of the LBM with $\tau = 0.6$, which is not described by (2.27).

2.4 Discussion & Conclusion

We presented an axisymmetric LBM for high density ratio multiphase flows. The method is capable of achieving liquid-to-gas density ratios up to 10^3 and higher. In order to recover the axisymmetric multiphase mass and momentum conservation equations appropriate source terms are introduced in the lattice Boltzmann evolution equation and in the multiphase force. The source terms in the evolution equation ensure the correct modelling for the mass, momentum and viscous tensors, while the source terms in the multiphase force ensure a correct surface tension in the model.

In our axisymmetric formulation of the LBM we used correction terms for the multiphase force that scale with the lattice speed of sound. We substitute the thermodynamic speed of sound for the lattice speed of sound in this correction term. By doing so, we observe better consistency with the fully 2D counterpart. A mathematical proof of the validity of this substitution cannot be provided at this moment and is left for future work.

We validated the model by comparing the Young-Laplace pressure of the axisymmetric simulation with the full 2D simulations. We observe a maximum relative error in the Laplace pressure of 3.2%. Then, we validated the dynamics of an axially symmetric oscillating droplet with a known analytical solution. Here, we observe a maximum relative error of 15.8%. Finally, we validate that a propagating density wave moving towards and away from the longitudinal z-axis correctly conserves mass and found excellent agreement with the inviscid analytical solution.

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3

Droplet deformation by short laser-induced pressure pulses *

When a free-falling liquid droplet is hit by a laser it experiences a strong ablation driven pressure pulse. In this chapter we study the resulting droplet deformation in the regime where the ablation pressure duration is short, i.e. comparable to the time scale on which pressure waves travel through the droplet. To this end an acoustic analytic model for the pressure-, pressure impulse- and velocity fields inside the droplet is developed in the limit of small density fluctuations. This model is used to examine how the droplet deformation depends on the pressure pulse duration while the total momentum to the droplet is kept constant. Within the limits of this analytic model, we demonstrate that when the total momentum transferred to the droplet is small the droplet shape-evolution is indistinguishable from an incompressible droplet deformation. However, when the momentum transfer is increased the droplet response is strongly affected by the pulse duration. In this later regime, compressed flow regimes alter the droplet shape evolution considerably.

3.1 Introduction

The impact of a short laser pulse onto a free-falling absorbing liquid droplet induces a rapid phase change in a thin superficial layer on the illuminated side of the droplet

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Figure 3.1: An illustration of three different impact duration regimes on a droplet. (a) A nanosecond laser pulse impacting from the left on a micron-sized liquid tin droplet leads to plasma formation (white glow in the image) and subsequently plasma-mediated ablation of the droplet (image taken from [2]). The typical ablation pressure duration is comparable to the plasma-decay duration, which is of the order of the acoustic time scale: $\tau_e \lesssim R/c$, where R is the initial droplet radius and c the speed of sound inside the droplet. (b) Impact of a nanosecond laser pulse onto a millimetre-sized dyed water droplet can lead to vaporization and mist cloud formation, the accompanying shock wave in the surrounding air is also visible [1]. The typical vapour-recoil induced ablation pressure duration is much longer than the acoustic time scale, but much shorter than the time scale on which the droplet deforms: $R/c \ll \tau_e \ll R/U$, where U is the propulsion speed of the droplet (image taken $5\mu s$ after laser impact, image courtesy A.L. Klein). (c) For the impact of a droplet onto a solid surface the typical interaction time is equal to the deformation time $\tau_e = R/U$, where U is the impact speed of the droplet (image taken from [6]).

[1, 2]. The resulting vaporization, explosive boiling or even plasma formation gives rise to mass ablation; see figures 3.1a and b. Subsequently, a recoil pressure wave propagates into the droplet and causes a net momentum transfer [1, 3, 4]. As a consequence the droplet is propelled forward and strongly deforms [1, 5]. However, the way in which these pressure waves establish inside the droplet over time, which is in particular relevant for short pulse durations, has so far remained unexplored.

In this chapter we aim to understand the fluid dynamic response of a droplet to a short ablation driven pressure pulse. Next to this ablation pressure, a laser impact could trigger pressure waves inside the droplet through a number of other mechanisms [7]. Electrostriction and radiation pressures are of negligible influence compared to the ablation pressure [7]. However, the local heating of the liquid close to the droplet surface can induce significant thermoelastic waves that result from thermal expansion [3, 8]. Furthermore, for high laser intensities dielectric breakdown on the droplet surface can lead to the generation of shock waves inside the droplet [9–11] or even plasma generation inside a transparent droplet [12–14]. These mechanisms

could have a strong influence on the droplet response. Indeed, cavitation phenomena, shock waves and rapid interface acceleration can give rise to fast jetting, bubble collapse and inter-facial instabilities [10, 14–17]. The study of these violent, highly non-linear response regimes is beyond the scope of the present study. Instead, we examine how an ablation pressure pulse is communicated throughout the droplet and triggers droplet deformation.

An important application of laser-induced droplet deformation is found in Laser Produced Plasma light-sources to generate Extreme Ultra Violet (EUV) light used for nano-lithography [18, 19]. In these sources small tin droplets are converted into a plasma by a two-stage laser impact process [19]. Upon the first impact, the droplet deforms into a thin flat sheet which is thereafter ionized by a second more powerful laser. A key question to improve this source is how the droplet deformation changes when the laser pulse duration is shortened.

Up to now, the response of a droplet due to a laser impact has been studied by using incompressible hydrodynamics to model the droplet deformation [1, 5]. In these models the interaction of the laser with the droplet is described by an ablation pressure p_e acting on the surface of the droplet for a duration τ_e . The impulse $p_e \tau_e$ resulting from this ablation pressure causes a momentum transfer to the droplet $\rho_0 R^3 U$, where ρ_0 is the liquid density, R the initial droplet radius and U the center-of-mass speed, which therefore scales as [5]

$$U \sim \frac{p_e \tau_e}{\rho_0 R}. \quad (3.1)$$

The deformation in these incompressible models is calculated by a pressure impulse approach that is also used for studies on the impact of liquid bodies onto solids [20–22]. As long as the duration of the ablation pressure is long compared to the acoustic time scale R/c , where c is the speed of sound inside the droplet, and the amplitude p_e is such that no shock-waves are created, the droplet response can be considered incompressible [5]. For example, for classical droplet impact onto a solid the deformation time scale $\tau_i = R/U$ is of the same order as the impact duration τ_e which is much longer than R/c (see e.g. [6, 23]), as illustrated in figure 3.1c.

By contrast, the impact of a laser pulse provides a means to shorten the duration of the ablation pressure considerably, and thereby to transfer the same amount of momentum $p_e \tau_e$ to the droplet in a shorter time. The ablation-pressure duration can for example be decreased by increasing the laser pulse energy to move to the plasma-mediated ablation regime, which leads to more violent and shorter lived ablation pressures [2], as illustrated in figure 3.1a. A further decrease of the ablation-pressure duration can be obtained by directly shortening the laser-pulse duration [24]. In these cases τ_e is shortened significantly such that it becomes comparable to or even smaller than R/c such that the droplet response is compressible and incompressible models breakdown. We note that for laser-induced ablation $\tau_e \ll \tau_i$ such that the droplet

remains undeformed during impact [5]. Indeed, in figure 3.1b we observe that the mist cloud resulting from mass ablation acts on the surface of an undeformed droplet.

In this chapter we study the response of a droplet to a short ablation pressure acting on its surface. In particular, we focus on the question how the droplet deformation dynamics depends on the ablation pressure duration at fixed impulse in the regime where $\tau_e \lesssim R/c$. Hence we consider the situation where the pressure field inside the droplet is not yet established during the pressure pulse and the droplet response is no longer incompressible. To this end, we develop a linearly compressible analytic model for the droplet response to short pressure pulses. In §3.2 we introduce the analytic model and discuss the regime in which it applies. In §3.3 we first compare our analytic results to a compressible lattice-Boltzmann simulation. Next we use the analytic model to study the effects of shortening the pulse duration at constant impulse on the pressure-, pressure impulse-, velocity- and deformation fields of the droplet.

3.2 Problem formulation & methods

In this section we derive a model to describe the spatio-temporal response of a droplet to an ablation pressure acting on its surface. In §3.2.1 we provide a scaling analysis to delineate three different regimes in the response of the droplet to this pressure pulse. Analytic expressions for the pressure and velocity fields inside the droplet as function of the pressure pulse are derived in §3.2.2. Finally in §3.2.3 we discuss the lattice-Boltzmann method that we use to support our analytic findings.

3.2.1 Scaling analysis

We consider a spherical droplet with radius R and density ρ_0 that is subjected to an ablation pressure on the illuminated side with an amplitude p_e and a duration τ_e , see figure 3.2. The total impulse received by the droplet is given by $J \sim p_e \tau_e$. In this chapter we explore the effect of decreasing the pulse duration while keeping the total impulse transferred to the droplet constant; i.e. decreasing τ_e at constant J .

During the pulse the pressure disturbance on the surface of the droplet penetrates over a length-scale $\ell_e \sim c\tau_e$, where c is the speed of sound in the droplet. If ℓ_e is short compared to R all momentum is initially concentrated inside a thin layer, as is illustrated in figure 3.2. By contrast, if $\ell_e > R$, all fluid inside the droplet has experienced a change in momentum directly after the pulse. The ratio between ℓ_e and R is quantified by the acoustic Strouhal number and is a dimensionless pressure pulse duration

$$\text{St} = \frac{\ell_e}{R} = \frac{c\tau_e}{R}. \quad (3.2)$$

To investigate the effect of short pulse durations, we are interested in the limit $\text{St} \lesssim 1$.

Figure 3.2: Sketch of the problem: an ablation pressure of amplitude $p_e(\theta, \phi, t)$ and duration τ_e acts on the surface of a droplet with radius R . As a result, a pressure field is induced inside the droplet over a depth $\ell_e \sim c\tau_e$. The colorbar denotes the pressure amplitude (blue is ambient and yellow is peak pressure). The spherical coordinate system (r, θ, ϕ) used is indicated (the azimuthal angle ϕ is not shown but rotates around the z -axis).

When τ_e is decreased at constant J , p_e rises. From momentum conservation in this thin layer it follows that the typical velocity induced inside ℓ_e is given by $u_e \sim p_e/(\rho_0 c)$, where ρ_0 is the density of the liquid droplet. Hence we observe that a large p_e induces large velocities in ℓ_e , which is quantified by the acoustic Mach number and is a dimensionless pressure pulse amplitude

$$\text{Ma} = \frac{u_e}{c} = \frac{p_e}{p_0}, \quad (3.3)$$

where $p_0 = \rho_0 c^2$ is the base pressure of the droplet. When Ma is large the fluid response inside the droplet is non-linear and shock waves dominate the flow. If Ma small, the flow inside the droplet can be considered linear.

The product MaSt sets the total dimensionless impulse received by the droplet

$$\text{StMa} = \frac{p_e \tau_e}{\rho_0 R c} = \frac{U}{c}, \quad (3.4)$$

where (3.1) is used to express the center-of-mass velocity U of the droplet as whole. This product is often referred to as the global Mach number of the droplet.

One can use Ma and St to delineate different regimes in the droplet response, as illustrated in figure 3.3. For lines of constant MaSt (hence constant impulse),

Figure 3.3: A phase diagram showing lines of constant impulse transfer to the droplet. The blue dotted-dashed line is the isoline $StMa = 0.01$, the orange dashed line is the isoline $StMa = 0.1$ and the red dotted line is the isoline $StMa = 0.5$. The plot shows three distinct regimes can be observed at constant impulse: a strongly compressible regime, a weakly compressible regime and an incompressible regime. The weakly compressible regime is the focus of the present work.

we can identify three regimes. Firstly, when St is small and Ma is large, we are in a strongly compressible regime where non-linear advective acceleration and non-linear viscous dampening need to be taken into account to describe the flow. Secondly, for intermediate Ma and St , compressible effects are important but non-linear effects are small, which renders this regime analytically accessible. This regime, which we term the weakly compressible regime, will be the main focus of this paper. Finally, when $St \gg 1$ and $Ma \ll 1$, we enter the incompressible regime that was subject of previous studies where long pulse durations (large St) were considered [5]. This regime is also relevant to droplet impact studies on rigid surfaces, see e.g. [6, 23, 25–28].

3.2.2 The weakly compressible model

The compressible flow equations are given by [20, p164]

$$\frac{\partial \rho}{\partial t} + (\nabla \cdot \rho \mathbf{u}) = 0, \quad (3.5)$$

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \mu \nabla^2 \mathbf{u} + \left(\kappa + \frac{1}{3} \mu \right) \nabla (\nabla \cdot \mathbf{u}), \quad (3.6)$$

where ρ is the density field, $\mathbf{u}$ the velocity field, p the thermodynamic pressure, μ is the shear viscosity coefficient and κ the expansion viscosity coefficient. In this chapter we only focus on the flow inside the droplet and we do not consider the flow of the outer gas phase, since for typical experimental situations (tin droplet in vacuum and water droplet in air): $\rho_g/\rho_l \ll 1$ and $\mu_g/\mu_l \ll 1$ [1, 2]. In order to find a closed analytic expression for the pressure and velocity field inside the droplet, we will first need to simplify (3.5) and (3.6) and finally solve the system obtained with appropriate surface boundary conditions for the droplet.

In the weakly compressible regime, i.e. for intermediate Ma, St (see figure 3.3), we can expand the pressure p , density ρ and velocity $\mathbf{u}$ inside the droplet as a constant plus a small time-dependent part, analogous to [20, p166]

$$\begin{aligned} p(\mathbf{x}, t) &= p_0 + p_1(\mathbf{x}, t), \\ \rho(\mathbf{x}, t) &= \rho_0 + \rho_1(\mathbf{x}, t), \\ \mathbf{u}(\mathbf{x}, t) &= \mathbf{u}_1(\mathbf{x}, t), \end{aligned} \quad (3.7)$$

where t is the time, $\mathbf{x} = (r, \theta, \phi)$ are the spherical coordinates defined in figure 3.2. The thermodynamic pressure can formally be expressed as $p = p(\rho, S, T)$, where S is the entropy and T is the temperature. In this chapter however, we neglect any thermoelastic waves that result from thermal expansion due to the local heating of the surface of the droplet. This simplification is supported by typical experiments found in literature, where both the thermal expansion coefficient and the thermal diffusion coefficient are small [1, 2]. We do note that with increasing laser intensity, thermoelastic waves could become increasingly more important [3, 8]. However, this is typically accompanied by an increase in the acoustic Mach number which is outside the scope of this work. Following these simplifications, we can write a basic equation of state for the fluid inside the droplet

$$p(\mathbf{x}, t) = p_0 + c^2 \rho_1(\mathbf{x}, t), \quad (3.8)$$

where c is the speed of sound and $p_0 = c^2 \rho_0$ is the ideal base pressure of the stationary droplet. Since we are interested in the flow directly after the pulse we introduce the following non-dimensionalisation, following the scaling analysis of the previous

paragraph

$$\mathbf{u} = \frac{p_e}{\rho_0 c} \tilde{\mathbf{u}}, \quad \mathbf{x} = R \tilde{\mathbf{x}}, \quad t = \tau_e \tilde{t}, \quad p = p_e \tilde{p}, \quad \rho = \frac{p_e}{c^2} \tilde{\rho}. \quad (3.9)$$

where the tildes refer to the dimensionless parameters. From now on we drop the tildes and work with the dimensionless parameters. A consistent approximation of (3.5) and (3.6) is given by the system [29, p97]

$$\frac{\partial p_1}{\partial t} + \text{St}(\nabla \cdot \mathbf{u}_1) = 0, \quad (3.10)$$

$$\frac{1}{\text{St}} \frac{\partial \mathbf{u}_1}{\partial t} = -\nabla p_1 + \frac{1}{\text{Re}} \nabla^2 \mathbf{u}_1 + \frac{1}{\text{Re}_v} \nabla(\nabla \cdot \mathbf{u}_1), \quad (3.11)$$

where $\text{Re} = \rho_0 R c / \mu$ is the Reynolds number with μ the dynamic viscosity and $\text{Re}_v = \rho_0 R c / (\frac{1}{3}\mu + \kappa)$ the Reynolds number for volume changes, where κ is the bulk viscosity. Although the Reynolds number in experiments is typically large ($\text{Re} \sim 10^3$) we will see later on that we need to retain the viscous terms in (3.11) to overcome singularities when converging pressure waves superimpose in the center of the droplet. In the high Reynolds regime viscous effects however do not influence the droplet deformation, which was already reported in a previous incompressible and inviscid model by [5]. We note that there is no explicit dependence on the acoustic Mach number in (3.10) and (3.11), since this is a low order Mach expansion of the compressible Navier-Stokes equations and we scaled all our fields accordingly (3.9). By taking the divergence of (3.11) and using (3.10) we obtain a viscous wave equation for the acoustic field inside the droplet

$$\frac{\partial^2 p_1}{\partial t^2} - \text{St}^2 \nabla^2 p_1 = \frac{1}{\text{Re}_a} \left[\nabla^2 \frac{\partial p_1}{\partial t} \right], \quad (3.12)$$

where $\frac{1}{\text{Re}_a} = \text{St} \left(\frac{1}{\text{Re}} + \frac{1}{\text{Re}_v} \right)$ is an effective Reynolds number for the viscous dissipation in the acoustic wave [29, p97]. Below we describe how this equation for $p_1(\mathbf{x}, t)$ is solved for the problem at hand. In §3.2.2 we show how the velocity field $\mathbf{u}_1(\mathbf{x}, t)$ can be computed once $p_1(\mathbf{x}, t)$ is known.

The pressure field

We solve (3.12) subject to a pressure boundary condition on the droplet surface. At the interface of the droplet the stresses between the liquid and the gas phase must be continuous. We assume that the magnitude of the pressure variations in the gas phase are much smaller than those inside the droplet. For typical experiments [1, 2] this assumption is justified since the density and the viscosity of the gas phase are much smaller. As a result, the stress condition on the interface significantly simplifies since

Figure 3.4: Jordan curves used to evaluate the inverse Fourier transform (3.19) in the complex plane where $a \rightarrow \infty$. (a) contour used for $t > t_0$, which includes the poles. (b) contour used for $t < t_0$ to obey the causality condition for the Green's function.

the pressure in the gas phase may be considered constant and equal to p_0 . As a consequence, the time-dependent part of the pressure at the surface of the droplet must satisfy

$$p_1(1, \theta, \phi, t) = 0, \quad (3.13)$$

to ensure that all acoustic energy remains inside the droplet. Furthermore we assumed that the interface remains immobile and therefore the droplet spherical during the pulse. This latter assumption is justified when the pulse duration τ_e is much smaller than the typical interface deformation time scale $\tau_{int} = R/u_e$, or in dimensionless form $\text{StMa} \ll 1$. Typically in experiments $\text{StMa} \sim 10^{-2} - 10^{-1}$. An ablation pressure acting on the surface of the droplet is introduced through a Green's function formalism [30, Chapter 7]. The general solution for the spatio-temporal pressure field inside the droplet is given by

$$\begin{aligned}
 p_1(\mathbf{x}, t) = & \iiint_V \left[\frac{\partial G(\mathbf{x}, t; \mathbf{x}_0, t_0)}{\partial t_0} p_1(\mathbf{x}_0, t_0) - \right. \\
 & \left. \left(\frac{\partial p_1(\mathbf{x}_0, t_0)}{\partial t_0} - \frac{p_1(\mathbf{x}_0, t_0)}{\text{Re}_a} \nabla_{\mathbf{x}_0}^2 \right) G(\mathbf{x}, t; \mathbf{x}_0, t_0) \right] \Big|_{t_0=0}^{t^+} dV_0 + \\
 & \int_0^{t^+} dt_0 \oint_S \left[G(\mathbf{x}, t; \mathbf{x}_0, t_0) \left(\text{St}^2 \nabla_{\mathbf{x}_0} p_1(\mathbf{x}_0, t_0) + \frac{1}{\text{Re}_a} \nabla_{\mathbf{x}_0} \frac{\partial p_1(\mathbf{x}_0, t_0)}{\partial t_0} \right) \right. \\
 & \left. - \nabla_{\mathbf{x}_0} G(\mathbf{x}, t; \mathbf{x}_0, t_0) \left(\text{St}^2 p_1(\mathbf{x}_0, t_0) + \frac{1}{\text{Re}_a} \frac{\partial p_1(\mathbf{x}_0, t_0)}{\partial t_0} \right) \right] \cdot \mathbf{n} dS_0, \quad (3.14)
 \end{aligned}$$

where $G(\mathbf{x}, t; \mathbf{x}_0, t_0)$ is the Green's function satisfying

$$\begin{aligned} & \frac{\partial^2 G(\mathbf{x}, t; \mathbf{x}_0, t_0)}{\partial t^2} - \text{St}^2 \nabla^2 G(\mathbf{x}, t; \mathbf{x}_0, t_0) \\ & - \frac{1}{\text{Re}_a} \left[\nabla^2 \frac{\partial G(\mathbf{x}, t; \mathbf{x}_0, t_0)}{\partial t} \right] = \delta(\mathbf{x} - \mathbf{x}_0) \delta(t - t_0), \end{aligned} \quad (3.15)$$

where we use a spherical coordinate system for $\mathbf{x}$ and $\mathbf{x}_0$. To find the general solution to (3.15), we first define a Fourier transformation

$$\hat{G}(\mathbf{x}, \omega; \mathbf{x}_0, t_0) = \int_{-\infty}^{\infty} G(\mathbf{x}, t; \mathbf{x}_0, t_0) \exp(-i\omega t) dt, \quad (3.16)$$

$$G(\mathbf{x}, t; \mathbf{x}_0, t_0) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \hat{G}(\mathbf{x}, \omega; \mathbf{x}_0, t_0) \exp(i\omega t) d\omega. \quad (3.17)$$

Using (3.16), (3.15) can now be transformed into a Helmholtz equation

$$-\omega^2 \hat{G}(\mathbf{x}, \omega; \mathbf{x}_0, t_0) - \left(\text{St}^2 + \frac{i\omega}{\text{Re}_a} \right) \nabla^2 \hat{G}(\mathbf{x}, \omega; \mathbf{x}_0, t_0) = \delta(\mathbf{x} - \mathbf{x}_0) \exp(-i\omega t_0), \quad (3.18)$$

where i is the imaginary unit. A general solution to this equation can be found by expanding the Green's function into eigenfunctions, resulting in

$$\begin{aligned} G(r, \theta, \phi, t; r_0, \theta_0, \phi_0, t_0) &= \sum_{\text{nlm}} \psi_{\text{nl}}^{\text{m}}(r, \theta, \phi) \psi_{\text{nl}}^{\text{m}}(r_0, \theta_0, \phi_0) \frac{1}{2\pi} \\ & \int_{-\infty}^{\infty} \left(\frac{\exp(i\omega(t - t_0))}{\text{St}^2 \beta_{\text{nl}}^2 + \frac{i\omega \beta_{\text{nl}}^2}{\text{Re}_a} - \omega^2} \right) d\omega, \end{aligned} \quad (3.19)$$

where $\psi_{\text{nl}}^{\text{m}}(r, \theta, \phi)$ are the eigenfunctions of the spherical Helmholtz equation

$$\psi_{\text{nl}}^{\text{m}}(r, \theta, \phi) = \frac{\sqrt{2} j_l(\beta_{\text{nl}} r) Y_l^{\text{m}}(\theta, \phi)}{j_{l+1}(\beta_{\text{nl}})}, \quad (3.20)$$

j_l are the spherical Bessel functions, Y_l^{m} are the spherical harmonics and β_{nl} are the zeros of the spherical Bessel functions. To evaluate the inverse Fourier transform (3.17), we use complex contour integration (see figure 3.4). It can be shown that the contribution of the arc is zero in the limit where the contour radius $a \rightarrow \infty$. Furthermore, there should be no response of an impulse released at t_0 at earlier times $t < t_0$ (causality condition). To this end, we pick the Jordan curve illustrated in figure 3.4a for $t > t_0$ and the curve of figure 3.4b for $t < t_0$, in the limit $a \rightarrow \infty$. A closed form expression of the Green's function is now given by

$$\begin{aligned} G(\mathbf{x}, t; \mathbf{x}_0, t_0) &= \sum_{\text{nlm}} \psi_{\text{nl}}^{\text{m}}(r, \theta, \phi) \psi_{\text{nl}}^{\text{m}}(r_0, \theta_0, \phi_0) \\ & \exp(-\kappa_{\text{nl}}(t - t_0)) \frac{\sin(\eta_{\text{nl}}(t - t_0))}{\eta_{\text{nl}}} \mathcal{H}(t - t_0), \end{aligned} \quad (3.21)$$

where $\kappa_{nl} = \frac{\beta_{nl}^2}{2\text{Re}_a}$ and $\eta_{nl} = \frac{\sqrt{4\text{St}^2\beta_{nl}^2 - \beta_{nl}^4}}{2\text{Re}_a}$. The resulting spatio-temporal pressure field using (3.14) reads (without any initial condition)

$$p_1(r, \theta, \phi, t) = \sum_{nl} \beta_{nl} \frac{j_l(\beta_{nl}r)}{j_{l+1}(\beta_{nl})} \left(1 - \frac{j_{l-1}(\beta_{nl})}{j_{l+1}(\beta_{nl})}\right) \frac{2l+1}{4\pi} \int_0^{2\pi} \int_0^\pi \int_0^t \exp(-\kappa_{nl}(t-t_0)) \frac{\sin(\eta_{nl}(t-t_0))}{\eta_{nl}} \mathcal{H}(t-t_0) P_l(\cos \gamma) \left(\text{St}^2 p_i(1, \theta_0, \phi_0, t_0) + \frac{1}{\text{Re}_a} \frac{\partial p_i(1, \theta_0, \phi_0, t_0)}{\partial t_0}\right) \sin(\theta_0) d\theta_0 d\phi_0 dt_0, \quad (3.22)$$

where $\mathcal{H}$ is the Heaviside theta function, P_l are the Legendre polynomials and $\cos(\gamma) = \cos(\theta)\cos(\theta_0) + \sin(\theta)\sin(\theta_0)\cos(\phi - \phi_0)$. In §3.3, we will use (3.22) using a particular pressure boundary condition specified by $p_i(1, \theta_0, \phi_0, t_0)$. We note that this condition is integrated into the solution at $r = 1^-$, i.e. $r = 1 - \varepsilon$ where $\varepsilon \rightarrow 0$.

The velocity field

The velocity field inside the droplet is given by (3.11). Since there is no initial rotation present in the fluid and there are no rotational forces acting at later times, the velocity field remains irrotational and is given by a scalar potential. A straightforward time integral over the pressure gradient (the first term on the right hand side) based on the spherical Bessel functions (3.22) results in a divergent series. To overcome this problem, we solve the velocity field in a different function basis. To this end, we first define the time integral over the thermodynamic pressure as the pressure impulse

$$J_1(\mathbf{x}, t) = \int_0^t p_1(\mathbf{x}, t') dt'. \quad (3.23)$$

The governing equation for the pressure impulse can be obtained by integration of (3.12) in time

$$\nabla^2 J_1 = \frac{1}{\text{St}^2} \frac{\partial p_1}{\partial t} - \frac{1}{\text{St}^2 \text{Re}_a} \nabla^2 p_1, \quad (3.24)$$

where we used that both the pressure field p_1 and its derivative vanish at $t = 0$. It now becomes apparent that the natural basis functions for the pressure impulse are in fact harmonic functions which results in a convergent series.

The general solution for the pressure impulse inside the droplet is therefore given by

$$J_1(\mathbf{x}, t) = \iiint_V G(\mathbf{x}; \mathbf{x}_0) \left[\frac{1}{\text{St}^2} \frac{\partial p_1(\mathbf{x}_0, t)}{\partial t} - \frac{1}{\text{St}^2 \text{Re}_a} \nabla^2 p_1(\mathbf{x}_0, t) \right] dV_0 - \oint_S [G(\mathbf{x}, \mathbf{x}_0) \nabla_{\mathbf{x}_0} J_1(\mathbf{x}_0, t) - J_1(\mathbf{x}_0, t) \nabla_{\mathbf{x}_0} G(\mathbf{x}, \mathbf{x}_0)] \cdot \mathbf{n} dS_0. \quad (3.25)$$

where the Green's function satisfies the Poisson equation in spherical coordinates

$$\nabla^2 G(\mathbf{x}; \mathbf{x}_0) = \delta(\mathbf{x} - \mathbf{x}_0). \quad (3.26)$$

Completely analogous to the boundary conditions on p_1 , the boundary condition on J_1 is $J_1(1, t) = 0$, which yields

$$G(\mathbf{x}, \mathbf{x}_0) = \frac{1}{4\pi} \left(\frac{1}{\sqrt{r^2 r_0^2 + 1 - 2rr_0 \cos(\gamma)}} - \frac{1}{\sqrt{r^2 + r_0^2 - 2rr_0 \cos(\gamma)}} \right), \quad (3.27)$$

where $\cos(\gamma) = \cos(\theta)\cos(\theta_0) + \sin(\theta)\sin(\theta_0)\cos(\phi - \phi_0)$. We evaluate (3.25) numerically using Mathematica 11.1 to speed up the calculations as compared to evaluating (3.25) analytically [31]. The velocity field now reads

$$\mathbf{u}_1(\mathbf{x}, t) = -\text{St} \nabla J_1(\mathbf{x}, t) - \left(\frac{1}{\text{Re}} + \frac{1}{\text{Re}_v} \right) \nabla p_1, \quad (3.28)$$

where $\mathbf{u}_1(\mathbf{r}, 0) = \mathbf{0}$ and $p_1(\mathbf{r}, 0) = 0$.

3.2.3 The lattice-Boltzmann method

To validate our analytic expression for the pressure field inside the droplet, we employ axisymmetric compressible multiphase lattice-Boltzmann simulations [32]. Instead of solving the compressible Navier-Stokes equations directly, the numerical method solves a probability distribution function for the position and momentum of particles inside a domain. The evolution of the particle distribution in space and time is given by the Boltzmann equation which is solved numerically on a lattice with a reduced set of velocity vectors. It can be systematically shown that this method is able to correctly solve the fully compressible mass (3.5) and momentum conservation (3.6) equations [33].

The pressure in our numerical simulation is given by the van-der-Waals equation of state. This non-ideal equation of state allows us to successfully simulate a liquid-gas system where the interface dynamics is automatically described by the non-ideal pressure. In the vicinity of equilibrium, the van-der-Waals equation of state behaves as an ideal gas. Therefore, we can directly compare the pressure field inside the droplet with our analytic expression (3.22) in the weakly compressible regime.

Our numerical setup consists of an initially stationary liquid droplet surrounded by a gas, with a liquid-gas density ratio of $\rho_l/\rho_g \sim 170$. We identify the position of the interface by tracking the derivative of the density field where spikes reveal the location of the interface. We hit our droplet by applying a force directly on the liquid-gas interface. By tuning the amplitude (Mach number) and duration (Strouhal

number) of this force, we simulate different regimes in the phase space; see figure 3.3. Further details on this numerical method can be found in [34].

Unfortunately, the compressible multiphase lattice-Boltzmann method lacks the advantages of an adaptive grid refinement technique at the moment of writing. Therefore simulations of higher density ratios of $\rho_l/\rho_g > 1000$, simulations in the strongly compressible regime (see figure 3.3) or even long-time simulations in the weakly compressible regime are not possible. Only early time dynamics can be simulated with the computational resources available to us, which we use to validate the analytic expression for the pressure in the next section.

3.3 Results

3.3.1 Acoustic response of a droplet to the ablation pressure

We consider the impact of a uniform laser-beam profile on the left side of the droplet [5]

$$p_i(1, \theta, \phi, t) = \cos(\theta) \mathcal{H}\left(\frac{\pi}{2} - \theta\right) \mathcal{H}(1 - t), \quad (3.29)$$

where $\mathcal{H}$ is the Heaviside function that restricts the pressure profile to the illuminated side of the droplet and limits the duration of the ablation pressure. We note that this boundary condition is only valid for an uniform irradiation along the surface of the droplet, which applies to the experiments described in e.g. [2]. In the case of a focused beam hitting the droplet a Gaussian pressure profile with an appropriate spot size might be preferred, which has been studied extensively in the incompressible limit by [5]. We use (3.29) in all results presented below. The analytic pressure field (3.22) subject to (3.29) is given by

$$\begin{aligned} p_1(r, \theta, \phi, t) = & \text{St}^2 \sum_{\text{nl}} \beta_{\text{nl}} \frac{j_l(\beta_{\text{nl}} r)}{j_{l+1}(\beta_{\text{nl}})} \left(1 - \frac{j_{l-1}(\beta_{\text{nl}})}{j_{l+1}(\beta_{\text{nl}})}\right) \frac{2l+1}{4\pi} \\ & \int_0^{2\pi} \int_0^\pi \int_0^t \exp(-\kappa_{\text{nl}}(t-t_0)) \frac{\sin(\eta_{\text{nl}}(t-t_0))}{\eta_{\text{nl}}} \mathcal{H}(t-t_0) \mathcal{H}(1-t_0) \\ & P_l(\cos \gamma) \cos(\theta_0) \mathcal{H}\left(\frac{\pi}{2} - \theta_0\right) \sin(\theta_0) d\theta_0 d\phi_0 dt_0 \end{aligned} \quad (3.30)$$

for $t < 1$.

In figure 3.5 we show a comparison between (3.30) and the lattice-Boltzmann simulations at different times, for $\text{St} = 6.02$ and $\text{Ma} \ll 1$. In order to obtain the analytic plots we used $\text{Re}_a \sim 200$, $l = 20$ and $n = 500$. The same parameter values will be used for all results in the remainder of this paper. Initially, the pressure disturbance on the surface of the droplet sends out a radially expanding wave for all source points on the boundary inside the droplet (figure 3.5a) which then propagates (figure 3.5b) to the right side (figure 3.5c). During the propagation, the superposition of all

Figure 3.5: The pressure field $p_1(r, \theta)$ inside the droplet at different times for an ablation pressure (3.29) impacting from the left side with a dimensionless duration $St \approx 6.02$. The top row shows the analytic results (plotted up to $r = 0.95$ to restrict the amount of Fourier modes required). The bottom row shows the results of the lattice-Boltzmann simulations, which are in excellent agreement with the analytics. The black dashed line illustrates the centreline axis used to plot the results in figure 3.6.

the waves inside the droplet gives rise to a non-trivial pressure distribution, see figure 3.5b and 3.5c. Note that a negative value for p_1 does not necessarily mean a negative pressure since the total pressure is given by (3.7). The figures show a good qualitative agreement between the analytic model (top row) and the simulated droplet (bottom row).

A quantitative comparison of the pressure profiles along the centreline is given in figure 3.6. Here, we plotted the pressure field as it passes through the center of the droplet (figure 3.6b) and after the reflection on the right interface (figure 3.6c). We observe good quantitative agreement between the analytic results and the simulation, also after wave reflection (figure 3.6c) which confirms the validity of boundary condition (3.13).

Some small discrepancies can be observed in both figure 3.5 and 3.6. In the lattice-Boltzmann simulation, the speed of sound is not constant but depends on the local pressure due to the non-linear nature of the van-der-Waals equation of state.

Figure 3.6: Comparison between the analytic pressure $p_1(r,0)$ (black dotted curve) and the lattice-Boltzmann simulation (blue triangles) along the centreline of the droplet (see figure 3.5) at different times for $St = 6.02$. a) For $St \cdot t = 0.48$ the waves are travelling towards the center of the droplet, b) for $St \cdot t = 1.02$ the waves just passed the center of the droplet, and c) for $St \cdot t = 2.47$ the waves have reflected on the right interface of the droplet and is travelling back towards the left.

Figure 3.7: Three ablation-pressure pulses of different duration but constant impulse ($MaSt = 0.1$). (a) a short pulse with a duration $St = 0.25$ and amplitude $Ma = 0.4$, (b) an intermediate pulse duration $St = 1$ and $Ma = 0.1$ and (c) a long pulse duration $St = 4$ and $Ma = 0.025$.

In the limit where pressure fluctuations are small, the speed of sound is almost constant however when a pressure wave passes through the center the pressure increases considerably. These small changes in speed of sound lead to small discrepancies in comparison to the analytic model where the speed of sound is constant. Furthermore, in the simulation a finite density jump is used. Therefore, a small amount of acoustic energy could be transmitted to the gas phase when a pressure wave hits the interface.

3.3.2 The effect of the pulse duration on the droplet response

We now address how the droplet response depends on the ablation pressure amplitude (Ma) and duration (St), while the total momentum transfer to the droplet remains constant. To this end, we compare the droplet response to the three types of pulses that are illustrated in figure 3.7. In the first case (figure 3.7a) the duration of the ablation pressure is much smaller than the time it takes for a pressure wave to travel through the droplet ($St = 0.25$, $Ma = 0.4$). In the second case (figure 3.7b) the duration of the pulse is exactly equal to the time it takes to travel over a distance of one droplet radius ($St = 1$, $Ma = 0.1$). Finally (figure 3.7c) defines a pulse duration that is much longer than the acoustic time scale of the droplet ($St = 4$, $Ma = 0.025$). In all three cases, the total momentum transfer to the droplet is constant and equal to $StMa = 0.1$. Below, we discuss the differences in the pressure field (§3.3.2), pressure impulse field and velocity field inside the droplet (§3.3.2) and eventually the droplet deformation dynamics (§3.3.2) for these three different pulses.

Figure 3.8: The pressure field p_1 inside the droplet for the three different illustrated in figure 3.7: (a) $St = 0.25$, (b) $St = 1$ and (c) $St = 4$. The results are depicted for different times: at the end of the pulse ($t = 1$), at two times the pulse duration ($t = 2$) and at four times the pulse duration ($t = 4$). We note that the color bar scale is not fixed and that all fields are scaled with p_e . The compression wave is followed by an expansion after the pulse, which is not visible in figure (a) at $t = 2$, since its amplitude is negligibly small at this time instant.

Pressure field

Figure 3.8 shows the spatio-temporal pressure field inside the droplet that is induced by the three pressure pulses discussed in figure 3.7. For a short pulse duration, most of the pressure field is initially (i.e. at $t = 1$) localized in a small compression zone (figure 3.8a). We note that when $t = 1$, all plots are drawn exactly after the pulse in figure 3.8. This zone is the result of the superposition of radial compression waves emitted from source points on the interface. During the propagation ($t = 2$), the superposition of these waves leads to a highly compressed spot in the center, which is clearly visible at $t = 4$. At later times (not shown in the figure) the compression waves reach the right interface of the droplet where they reflect and give rise to an expansion zone. Meanwhile, waves radiated from point sources on the droplet surface continuously reflect on neighbouring surface points leading to expansion waves. The amplitude of these expansion waves however are so small that they only become visible in the plot at $t = 4$, see figure 3.8a. We note that the absolute pressure is not negative, since the absolute pressure is given by (3.7).

The pressure field for intermediate pulse duration is illustrated in figure 3.8b. At the end of the pulse ($t = 1$) the waves have travelled a distance R . Again a compression zone is created in the center, followed by an expansion zone ($t = 2$). At $t = 4$, all waves have at least reflected once on the interface of the droplet which gives rise to another large expansion zone. For a long pulse (figure 3.8c) the pressure field has spread over the entire droplet. The superposition of all compression and expansion waves lead to a non-trivial field that consists of compression and expansion zones.

To summarize, we observe more localized fluctuations in the pressure field directly after a short pulse as compared to longer pulses. As we will demonstrate below, these fluctuations have an important effect on how the impulse is distributed over time and hence on the resulting velocity field inside the droplet.

Pressure impulse and velocity fields

Figure 3.9 shows the spatio-temporal pressure impulse field (3.23) inside the droplet for the three pulse durations. To obtain the plots, we evaluate (3.25) numerically. This scalar field is an important field in our analysis, since it describes the spatio-temporal distribution of momentum inside the droplet and the velocity field (3.28) is derived from it, as discussed in §3.2.2. Note that in all cases the total momentum inside the droplet is constant as soon as the pulse is over, at $t = 1$, while the distribution of the momentum can still change in time.

For a short pulse duration the momentum distribution changes significantly in time, see figure 3.9a. Initially all momentum is concentrated on the left side of the droplet, while it redistributes itself throughout the droplet at later times. As we will show below, this localized momentum distribution results in a stronger interface deformation for short pulses. As the pulse duration increases (figure 3.9b) the time

Figure 3.9: The pressure impulse field J_1 inside the droplet for the three different pulse durations illustrated in figure 3.7: (a) $St = 0.25$, (b) $St = 1$ and (c) $St = 4$. The results are depicted for different times: at the end of the pulse ($t = 1$), at two times the pulse duration ($t = 2$) and at four times the pulse duration ($t = 4$). Note that the color bar scale is not fixed and that all fields are scaled with $p_e \tau_e$.

variation of the momentum distribution becomes smaller. This effect is most prominent for long pulses (figure 3.9c) where the momentum distribution is almost constant after $t = 1$. In the limit $St \rightarrow \infty$ (and consequently $Ma \rightarrow 0$ to keep the impulse finite) the pressure impulse is constant in time which corresponds to an incompressible flow.

The velocity field (3.28) derived from the pressure impulse is plotted in figure 3.10, where we show the θ -component for $r = 0.5$ at different times (solid lines). For comparison, the incompressible velocity field as derived in [5] is plotted as the black dashed line. When the pulse duration is short (figure 3.10a) the velocity field at $r = 0.5$ for $t = 1$ is zero, since the momentum has not yet propagated far enough into the droplet. As time progresses, velocity fluctuations become apparent and even for times long after the pulse ($t \gg 1$, right panel) they are nowhere near the incompressible solution. Figure 3.10b shows the velocity field for an intermediate pulse duration. Here, the velocity field fluctuates around the incompressible solution. However, the amplitude of these fluctuations are large. For the longest pulse the velocity gradually builds up (figure 3.10c, left panel). At later times (right panel) the velocity field shows only tiny fluctuations around the incompressible solution.

Droplet deformation

Finally, we turn to the question how the droplet deformation is affected by the duration of the ablation pressure pulse. To make a prediction for the droplet deformation, we use the velocity field at the droplet surface. Strictly speaking, the analytic solution (3.28) is derived for a constant spherical domain. However, we can obtain a first order approximation of the droplet shape at early times, i.e. when the deviations from a spherical shape are still small, by advecting material points on the interface as described in [5].

We compare the effect of the three different pulse durations illustrated in figure 3.7 on the droplet deformation. The droplet deformation is not only determined by the pulse duration, but also by the pulse amplitude. We therefore additionally consider three different momentum transfers to the droplet: $StMa = 0.01$, $StMa = 0.1$ and $StMa = 0.5$. The acoustical Mach number (or the product $StMa$) now becomes an additional parameter, because we want to quantify the actual differences in deformation. So far, we always scaled out this amplitude dependency (3.9). However, a difference in amplitude now gives a stronger or weaker deformation. In figure 3.11 we show contours of the droplet deformation for the three different momentum transfers and compare the influence of the pulse duration. We note that the unphysical sharp peaks at the poles are due to the singular nature of the pressure boundary condition (3.29), as already discussed by an incompressible inviscid model by [5]. We use a step function to limit the boundary condition to the illuminated side of the droplet. In figure 3.12 we quantify the interface displacement of the droplet at the

Figure 3.10: The θ -component of the velocity field inside the droplet at $r = 0.5$. The left column represents early times ($t \sim 1$) while the right column represents late times ($t \gg 1$). (a) A short pulse duration $St = 0.25$, (b) an intermediate pulse duration $St = 1$ and (c) a long pulse duration $St = 4$. The black dashed line denotes the incompressible velocity field by [5].

Figure 3.11: Contour plots of the droplet deformation for the three different pulse durations: $St = 0.25$ (blue dot dashed), $St = 1$ (orange dashed) and $St = 4$ (red dotted). (a) For $MaSt = 0.01$ all cases give rise to identical deformation behaviour, for (b) $MaSt = 0.1$ we see discrepancies arising between the three pulse durations which aggravate in (c) $MaSt = 0.5$ where we observe a clear influence of the pulse duration on the droplet deformation. Note that for each impulse the contours are sketched at a different absolute time to be able to clearly illustrate the deformations.

axis of impact $\Delta r/R$ in time.

When the momentum transfer to the droplet is small ($MaSt = 0.01$, figures 3.11a and 3.12a), we need to go to late times ($St \cdot t \gg 1$) in order to observe a significant deformation. On the acoustic time scale R/c however, the droplet deformation only shows tiny fluctuations around a static shape. In other words, the fluctuations in the velocity field due to the pressure waves are negligible and the velocity field is to a good approximation incompressible. As a result, on late times the droplet deformation for the three different pulse durations is indistinguishable.

When the amount of momentum transfer to the droplet is increased (figures 3.11b and 3.12b), interface deformations become apparent at earlier times. For the shortest two pulses all momentum was transferred into the droplet at the time of the plot, while for the long pulse the ablation pressure is still acting on the droplet surface. Therefore, the contour of the longest pulse (red dotted curve) is lagging behind compared to the contour of the shorter pulses (yellow dashed and blue dot dashed curves). Furthermore, for shorter pulses the droplet interface compression is followed by an expansion directly after the pulse which is visible in figure 3.12b. Hence, the droplet deformation for short pulses is now clearly a non-monotonous function of time. As $MaSt$ is further increased, the droplet deformation shows even larger fluctuations around a shape that is also globally deforming, see figures 3.11c and 3.12c.

These strong deformations invalidate the assumption that the droplet remain stationary during the pulse. Although figures 3.11c and 3.12c give a first order estimate of the deformations that are to be expected, for a quantitative prediction one has to solve the fields in the deformed geometry. In this regime we therefore anticipate a

Figure 3.12: The advective displacement of the droplet interface $\Delta r/R$ at the axis of impact ($r = 1$, $\theta = 0$) for the three different pulse durations: $St = 0.25$ (blue dot dashed), $St = 1$ (orange dashed) and $St = 4$ (red dotted). (a) $MaSt = 0.01$, (b) $MaSt = 0.1$ (c) $MaSt = 0.5$.

strong influence of the pulse duration on the eventual droplet-shape evolution at later times.

3.4 Discussion & conclusion

The droplet deformation resulting from a laser-induced ablation pressure pulse is studied analytically in the regime where the pulse duration is of the order of the acoustic time scale and the pressure fluctuations are small. The resulting momentum change of the droplet is determined by the pressure pulse amplitude and duration or, in dimensionless form, the acoustic Mach number Ma and the acoustic Strouhal number St . We examined the effect of changing St (i.e. shortening the pulse duration) on the droplet response while keeping the total impulse transferred to the droplet constant.

The pressure, pressure impulse and velocity fields inside the droplet are studied as function of St at constant impulse $StMa$. To keep the analysis simple we used a cosine-shaped ablation pressure profile on the surface of the droplet together with a step function to limit the ablation pressure in time and space. To get a first order estimate of the droplet deformation in time we advected material points on the surface.

In the regime where $StMa \ll 1$, the droplet deformation is independent of St and no significant changes in the deformation were observed for shorter pulses. When St is large, the flow inside the droplet may be considered incompressible since the pressure impulse field is approximately constant in time. By contrast, when $St \ll 1$ the flow inside the droplet is compressible. However, on the deformation time scale the compressible effects average out and the droplet behaves as if it were incompressible.

Therefore, the incompressible model by [5] can be used to describe the deformation dynamics in this regime.

Significant differences in deformation arise when $StMa \lesssim 1$. When $St \gg 1$ the flow is incompressible, but now the droplet deforms significantly during the pulse. When $St \ll 1$ all momentum is localized in a small shell close to the illuminated side of the droplet directly after the pulse. This momentum distribution results in a large acceleration of the interface and consequently a compression of the fluid that leads to a different deformation compared to the case where the pulse duration is long ($St \gg 1$). The droplet deformation in this regime is therefore strongly dependent on the pulse duration. In practice, to study droplet deformation resulting from femto-, pico-, nano-second laser pulses in the plasma-mediated ablation regime (i.e. short ablation pressure pulses $St \lesssim 1$) at high energy (such that $StMa \lesssim 1$), droplet compressibility needs to be taken into account.

In the regime where $Ma \gtrsim 1$, the linear approximation of the proposed analytic model breaks down. In this regime, the flow is governed by shock-waves, cavitation phenomena, non-linear viscous damping and rapid interface acceleration, which result in a highly non-linear droplet response. We argue however that the weakly compressible model can be used as a starting point to identify likely cavitation spots and study first order droplet deformation, since shock fronts first need to develop in time. We do note however that depending on the liquid properties or temperature, cavitation phenomena could be observed in the regime where Ma is small. A more detailed understanding the droplet deformation in these regimes requires numerical simulations and is topic of future work.

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4

Laser-to-droplet alignment sensitivity relevant for laser-produced plasma sources of extreme ultraviolet light ^{*†}

We present and experimentally validate a model describing the sensitivity of the tilt angle, expansion and propulsion velocity of a tin micro-droplet irradiated by a 1 μm Nd:YAG laser pulse to its relative alignment. This sensitivity is particularly relevant in industrial plasma sources of extreme ultraviolet light for nanolithographic applications. Our model has but a single parameter: the dimensionless ratio of the laser spot size to the effective size of the droplet, which is related to the position of the plasma critical density surface. Our model enables the development of straightforward scaling arguments in turn enabling precise control the alignment sensitivity.

4.1 Introduction

Micro-droplets of liquid tin are used to create extreme ultraviolet light (EUV) for next-generation nano-lithography [1–4] that is currently being introduced in high-volume manufacturing. These droplets, several 10 μm in diameter, serve as mass-limited targets [5, 6] for creating a laser-produced plasma (LPP) in EUV light sources.

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In such machines, a pre-pulse laser beam hits a tin droplet to obtain an extended disk-like target [7, 8] that increases coupling with the next pulse. Subsequently, the target is irradiated by a focused nanosecond-pulse laser at intensities that lead the creation of a high-density plasma [2, 6, 9]. Line emission from electron-impact-excited highly charged tin ions in the plasma provides the EUV light peaking at 13.5 nm [2, 6]. Maximizing the conversion efficiency (CE) of laser light into the required EUV light of such sources requires a careful control over the target shape. This shape is very sensitive to the precise alignment of the pre-pulse laser to the initially spherical droplet [10, 11]. Any deviation from the optimum location for laser impact will produce a suboptimal target tilt and decrease the target radial expansion, as shown in Fig. 4.1. Moreover, reflections of the laser light from the incorrectly tilted surface of the target back towards the laser itself may be detrimental to laser operation stability. In spite of its obvious relevance, the precise relation between such laser-to-droplet (L2D) alignment and the resulting target tilt has so far remained poorly explored aside from activities by Tsygvintsev et al. [12] and the recent work by Hudgins et al. [11] who combine modelling with experimental efforts. Due to experimental constraints, however, their model predictions for the tilt sensitivity could not be validated under conditions of controlled misalignment.

Here, we present and experimentally validate an intuitive model describing L2D alignment sensitivity. The model is based on a single parameter: the dimensionless ratio of the laser spot size to the effective size of the droplet target, which we relate to the position of the plasma critical density surface [13–15] and sets the typical length scale for the problem. Our model enables the development of straightforward scaling arguments, which in turn enable the minimization of detrimental alignment sensitivity. We focus our studies on industrially relevant 1- μm -wavelength Nd:YAG laser pulses and experimentally validate our model predictions for two different laser spot sizes over a wide range of laser pulse energies. Furthermore, we apply the validated model to predict sensitivities for several practical cases that are immediately relevant for current state-of-the-art industrial droplet-based EUV light sources.

4.2 Experiment

Our experimental setup has previously been described in detail [8]. For clarity, the most important characteristics are repeated here. A droplet generator was operated in a high-vacuum chamber ($\sim 10^{-7}$ mbar) and held at constant temperature of $\sim 260^\circ\text{C}$. The nozzle produced a 13.4 kHz train of $\sim 43\ \mu\text{m}$ diameter (radius $R_0 \approx 21.5\ \mu\text{m}$) droplets of 99.995% purity tin. The droplets were irradiated by 1064-nm wavelength, 10 ns (full-width-at-half-maximum: FWHM) long pulses of a Nd:YAG laser operated at 10 Hz repetition rate.

The laser pulse energies were varied between ~ 1 and ~ 395 mJ. The laser beam

Figure 4.1: Side-view shadowgraphy images taken $2\mu\text{s}$ after a 40 mJ pulse with $115\mu\text{m}$ diameter (FWHM) spot impinged on a droplet from the left at (a) optimal alignment and at (b) a misalignment of $\Delta x \approx 100\mu\text{m}$ leading to a tilt angle θ , accompanied by a decrease in total propulsion velocity and radial expansion. The white glow visible at the original droplet position is plasma light, where the deformed droplet has moved further downwards and sideways (see main text).

was focused down to a $115\mu\text{m}$ or $60\mu\text{m}$ diameter (FWHM) Gaussian spot on the droplet and was circularly polarized. While significant astigmatism was apparent for a tighter focus, the part of the beam intersecting with the droplet could still be well described by a $60\mu\text{m}$ -diameter Gaussian function. Conversely, the $115\mu\text{m}$ focus produced a circularly symmetric beam spot. In order to capture the dynamics of the expanding droplets two shadowgraphy systems based on 850-nm-wavelength, 15-ns pulsed laser diodes and long-distance microscopes coupled with charge-coupled-device cameras were used. The two systems provide a “front” view and a side view, 30° and 90° with respect to the laser propagation direction. By varying the time delay between the plasma-generating laser pulse and shadowgraphy pulses, a sequence of images was recorded. The images obtained from the side were processed using an image analysis program that tracks the center-of-mass displacement and the size of the expanding droplet as well as the target tilt angle. In order to introduce controlled misalignment, the timing of the Nd:YAG laser pulse was varied around the established time for optimal alignment of laser on droplet, see Fig. 4.1. Since the droplets vertical velocity U_x stayed constant ($\sim 12\text{ m/s}$), this “mistiming” Δt resulted in a laser impact off-centred by a distance Δx , translating the initial spherical droplet into a tilted disk. The final droplet shape is the result of complex force interplay and its study is left for future work. The velocity U_x was obtained by processing the front-view images containing two or more droplets. The target tilt angle, defined as the angle between the target normal and the laser beam propagation direction (see Fig. 4.1b), was mapped as a function of misalignment, with $\Delta x = U_x \Delta t$, for different laser pulse energies. At the same time, we tracked the propulsion velocity u_z along the laser light propagation direction. We obtained an estimate of the droplet expan-

Figure 4.2: Schematic cross-section of the problem: a droplet with radius R_0 is hit by a Gaussian-shaped intensity-profile laser pulse with size σ from the left. The laser hits the droplet off-center with an axial offset Δx creating plasma (light gray area) surrounding the droplet on the illuminated side (dashed line), thereby creating an effective radius R_{eff} .

sion velocity $\dot{R}$ by measuring its radius (i.e. the maximum droplet radius along the tilted axis) shortly ($1 \mu\text{s}$) after the laser pulse impact and assuming a linear expansion on this short timescale, such that $\dot{R} \approx (R(1 \mu\text{s}) - R_0)/(1 \mu\text{s})$.

4.3 Model

In this section, we outline an intuitive model for the tilt angle θ_{tilt} (see Fig. 4.1b) in terms of the axial misalignment and the laser beam width. Figure 4.2 shows a schematic overview of the problem. A droplet with radius R_0 is hit by a Gaussian off-centred laser pulse having an axial offset Δx and beam width σ , which relates to the full-width at half-maximum of the laser beam according to the usual convention $\sigma = \text{FWHM}/(2\sqrt{2\ln 2})$. The target tilt angle can be deduced from the direction of the target's center-of-mass motion. We derive the target's center-of-mass motion from the plasma pressure distribution on the surface of the target where the laser energy is absorbed. This pressure distribution is directly related to the spatial intensity profile of the laser. Below we detail these steps.

During the laser impact, a small liquid layer is ablated and forms a plasma cloud on the illuminated side of the droplet, see Fig. 4.2. The time scale of this plasma generation is typically much shorter than the laser pulse. As a result, most of the laser energy is absorbed just before the plasma critical density surface [13] where the plasma electron frequency equals that of the laser light and no further light penetra-

tion is possible. This gives the system an effective radius $R_{\text{eff}} \geq R_0$, see Fig. 4.2. The laser intensity profile I at R_{eff} is given by a projection of part of the Gaussian laser beam profile onto the laser-facing hemisphere with radius R_{eff} and an offset Δx . It is insightful to formulate this intensity profile in terms of two dimensionless parameters: the dimensionless laser beam width $\alpha = \sigma/R_{\text{eff}}$ and the dimensionless laser offset $\beta = \Delta x/R_{\text{eff}}$, which gives in spherical coordinates

$$I(\theta, \phi) \sim \exp\left(-\left[\frac{\sin(\theta)^2 - 2\beta \sin(\theta) \cos(\phi) + \beta^2}{2\alpha^2}\right]\right) \times H(\pi/2 - \theta) \cos(\theta), \quad (4.1)$$

where θ and ϕ are the polar angle and azimuthal angle respectively, and H is the Heaviside step function to limit the intensity profile to the illuminated side of the droplet. Note that an extra $\cos(\theta)$ term is included in Eq. (4.1) to account for the projection of the beam onto the effective radius, which is assumed to be locally spherical and of constant size.

With the intensity profile at R_{eff} at hand, we now obtain an expression for the plasma ablation pressure p_a acting on this surface. A power-law dependence between this pressure and the impinging laser pulse energy has been established in previous work [8, 11, 16], and experimentally shown to be valid to excellent accuracy over three orders of magnitude in laser energy [8, 16],

$$p_a \propto (E - E_0)^\delta, \quad (4.2)$$

where $E \propto I$ is the fraction of the laser pulse energy that is intercepted by the liquid target. In the remainder of this work, we take $\delta = 0.6$ in accordance to Ref. [13]. Furthermore we neglect the small offset energy E_0 , since typically $E_0 \ll E$ (see Ref. [8]) and use Eqs. 4.1 and (4.2) to find the local pressure $p_a(\theta, \phi)$ acting on the target's effective surface.

The center-of-mass velocity of the target is then obtained from p_a . To this end, we consider the droplet and its surrounding plasma cloud as a single body with radius R_{eff} that is subjected to a local pressure distribution $p_a(\theta, \phi)$. Note that by doing so we neglect the exchange of momentum between this plasma cloud and the droplet. Furthermore, we assume that the liquid body does not deform on the time scale of the pressure pulse, which is justified since the timescale of deformation is typically much longer [7, 8]. The center-of-mass velocity of the body is then given by

$$\begin{aligned}
u_x(\alpha, \beta) &\sim \int_0^\pi \int_0^{2\pi} p_a(\theta, \phi, \alpha, \beta) \sin(\theta) \sin(\theta) \cos(\phi) d\theta d\phi, \\
u_y(\alpha, \beta) &\sim \int_0^\pi \int_0^{2\pi} p_a(\theta, \phi, \alpha, \beta) \sin(\theta) \sin(\theta) \sin(\phi) d\theta d\phi, \\
u_z(\alpha, \beta) &\sim \int_0^\pi \int_0^{2\pi} p_a(\theta, \phi, \alpha, \beta) \sin(\theta) \cos(\theta) d\theta d\phi,
\end{aligned} \tag{4.3}$$

where the coordinates x, y, z are defined in Fig. 4.2.

From the direction of the center-of-mass motion, one can now deduce the target tilt angle. Note that since a pressure always acts perpendicular to the surface it can never induce any rotation of the body. Therefore the target tilt is a result of the preferred expansion direction of the liquid, which by definition is perpendicular to the direction of the center-of-mass motion. As a result, the tilt angle in radians is given by

$$\theta_{\text{tilt}}(\alpha, \beta) = \arctan(u_x(\alpha, \beta)/u_z(\alpha, \beta)), \tag{4.4}$$

which needs to be evaluated numerically. Our approach differs from the one presented in Ref. [11] as we include the full pressure distribution on the surface of the droplet, see Eqs. (4.3). Comparing the two models, our approach consistently yields significantly lower tilt angle sensitivities.

The target tilt angle sensitivity around zero misalignment is of interest for certain industrial applications [17]. To this end we define the tilt angle sensitivity around $\beta = 0$ as

$$f(\alpha) = \left. \frac{\partial \theta_{\text{tilt}}(\alpha, \beta)}{\partial \beta} \right|_{\beta=0}, \tag{4.5}$$

which enables a straightforward inspection of the influence of the dimensionless beam width α . The tilt angle sensitivity as expressed by Eq. (4.5) can be approximated analytically by expanding Eq. (4.1) and (4.4) up to $\mathcal{O}(\beta^2)$ and results in

$$f(\alpha) = \Re \left(\frac{u_x(\alpha, 1)}{u_z(\alpha, 0)} \right). \tag{4.6}$$

The full complex expression is given in its explicit form in the Appendix. The actual tilt angle in this approximation is given by

$$\theta_{\text{tilt}}(\alpha, \beta) \approx \left. \frac{\partial \theta_{\text{tilt}}(\alpha, \beta)}{\partial \beta} \right|_{\beta=0} \beta. \tag{4.7}$$

Another important industrially relevant parameter is the radial expansion velocity $\dot{R}$ as a function of the misalignment. In Fig. 4.1 we observe that laser misalignment

not only induces a target tilt, but also significantly decreases the expansion velocity. We now employ our basic model to obtain a first-order estimate of this reduced expansion. When the laser beam is misaligned with respect to the droplet, the laser intensity absorbed by the droplet decreases. As a consequence, both the center-of-mass speed and the expansion rate of the target decrease. The partitioning of kinetic energy between propulsion (center-of-mass motion) and expansion is set by the laser beam (or pressure) profile acting on the droplet as detailed in Ref. [7]. To obtain an intuitive, first-order estimate of the target expansion velocity, we assume that this energy partitioning remains fixed and is not influenced by the misalignment or laser beam energy, such that

$$\frac{\dot{R}(\Delta x/R_0)}{\dot{R}(0)} \sim \frac{U_{\text{cm}}(\Delta x/R_0)}{U_{\text{cm}}(0)}. \quad (4.8)$$

Here, the left-hand-side is the expansion velocity as function of the misalignment normalized by the expansion velocity at zero misalignment, and the right-hand-side is the center-of-mass velocity as function of the misalignment normalized by the center-of-mass velocity at zero misalignment.

4.4 Results

The experimental results for the tilt angle, the z -component of the center-of-mass velocity u_z and the radial expansion velocity $\dot{R}$ are shown in Fig. 4.3 as a function of misalignment $\Delta x/R_0$. The error bars represent the standard deviation of the measurements where available, otherwise a conservative value of twice the overall average error was used. In Fig. 4.3c the error bars are conservatively set at 20%. We classify three different groups of experimental data. The first data set is obtained for a 115- μm focus with energies of ~ 40 to ~ 395 mJ (green diamonds). The second group represents the experimental data for the same focus spot size, but with laser energies between ~ 5 and ~ 25 mJ (red squares). The third group consists of all data having a 60- μm focus spot size with laser pulse energies between ~ 1 and ~ 95 mJ (blue circles). In the top panel, we observe that the tilt angle monotonically increases with the misalignment. A small but significant influence of the laser pulse energy on the sensitivity is observed in the grouped data of the large focus spot size (green diamonds versus red squares) but this could not be further proven with any measure of significance for the individual (i.e. for single laser pulse energies) data sets. The small focus spot size (blue circles) results in a stronger tilt angle sensitivity to misalignment and show some signs of saturation at large misalignment values. In the center panel, we see that the normalized z -velocity u_z distribution has a typical bell shape, showing no significant influence of the laser pulse energy in the grouped data of the large focus spot size. The smaller spot size results in a more sharply peaked

distribution. In the bottom panel, we observe that the normalized radial expansion velocity $\dot{R}$ also has a typical bell shape, which appears to be of slightly larger width than the u_z distribution. The data also shows no significant influence of the laser pulse energy in the grouped data of the large focus spot size. Again, the smaller spot size results in a more sharply peaked distribution.

To compare the experimental results in Fig. 4.3 to the model described above, we numerically evaluate Eq. (4.4) using a local adaptive solver [18]. For each experimental case we determine α based on the focal spot sizes mentioned above and the effective radius R_{eff} , which we relate to the location of the critical plasma surface. We obtain the location of the critical surface for a Nd:YAG laser pulse on tin droplets from 2D radiation-hydrodynamic simulations [13]. In that work, the distance from a tin droplet (at $R_0 = 15 \mu\text{m}$) to the critical surface is evaluated to be $d_{\text{crit}} \approx 8 \mu\text{m}$ for a Nd:YAG laser pulse. By assuming the same position of the critical surface with respect to the droplet surface in our experimental case with slightly bigger droplets ($R_0 = 21 \mu\text{m}$), we obtain $R_{\text{eff}} = R_0 + d_{\text{crit}} = 29.5 \mu\text{m}$, and hence $\alpha_{115} = 1.7$ (115- μm focus, green diamonds) and $\alpha_{60} = 0.86$ (60- μm , blue circles). The corresponding sensitivity curve is in excellent agreement with the experimental data (see solid lines in fig. 4.3).

For laser energies lower than 40 mJ and a large focus spot size (red squares), the experimental data are found to be well described by the model if we set $\tilde{\alpha}_{115} = 2.3$, i.e., with the effective radius $R_{\text{eff}} \approx R_0$. This observation could be explained by the reasoning that for such low-energy and broad focus pulses the plasma is not fully developed and the critical surface is situated very close to the droplet surface ($d_{\text{crit}} \approx 0$). Unfortunately, no simulation data is available in this regime to support this claim. Furthermore, the difference between α_{115} and $\tilde{\alpha}_{115}$ could also be the result of different plasma pressure distributions [16] or additional dynamical, time-dependent effects that we do not consider here.

Using the above mentioned values for α , we observe that all theoretical curves show good agreement with the experimental data for both the tilt angle and the z -velocity u_z . Following the simple approximation given by Eq. (4.8), we compare the normalized, experimental radial expansion velocity to the theory predictions in Fig. 4.3. We find reasonable agreement between data and our model especially considering the simplifications involved and the experimental uncertainty in determining $\dot{R}$.

Next, we analyse the model prediction for the tilt angle sensitivity around zero misalignment ($\beta = 0$). Figure 4.4 shows the numerical curve (solid line) resulting from Eq. (4.5) with the dimensionless beam widths corresponding to the experimental operating conditions (see Section 4.4). The theoretical curve clearly shows that $f(\alpha)$ first increases, from unity with α peaking near $\alpha \approx 0.3$ after which a monotonic decrease is apparent. The sensitivity rises for larger droplets, keeping a constant beam width. Reversely, a smaller beam for a given effective radius will translate a large

Figure 4.3: (a) Tilt angle of the target at various misalignments. Data for the 115- μm focus with energies below (red squares) and above (green diamonds) 40 mJ have been grouped. Blue circles represent averages of all data for the 60- μm focus case. Red, green, and blue solid lines depict numerical predictions of target tilt with $\alpha = 2.3, 1.7$ and 0.86 respectively. (b) Droplet propulsion velocity u_z in the laser beam propagation direction. The red and green solid lines fully overlap. (c) Normalized droplet expansion velocity $\dot{R}$. In (b) and (c), the velocities have been normalized to the maximum value of each data set.

Figure 4.4: The theoretical tilt angle sensitivity $f(\alpha)$ (in degrees) as function of the dimensionless beam width α around zero misalignment ($\beta = 0$). The black curve is the analytically calculated tilt angle sensitivity from Eq. (4.5), with $\delta_p = 0.6$. The red square, green diamond and blue circle represent the values corresponding to the experimental conditions (see Section 4.4, color coding as in Fig. 4.3).

sensitivity to its alignment. In the limit $\alpha \rightarrow \infty$ one would illuminate the droplet with a flat-top beam of infinite width and would be completely insensitive to misalignment. In the limit $\alpha \rightarrow 0$ (i.e. a delta peak) the sensitivity decreases and eventually saturates to unity. In this limit the center-of-mass velocity u_x and u_z decay to zero equally fast as all energy is used to deform the droplet rather than to move its center-of-mass [7]. Hence the ratio u_x/u_z becomes meaningless and one needs to reconsider the definition of the tilt angle. The maximum in $f(\alpha)$ is caused by a maximum in $u_x(\alpha, 1)$, see Eq. (4.6). As α gets smaller, there is initially an increase in the u_x component since pressure on the surface of the droplet is spread increasingly more onto a surface element that points in the x-direction. However, as α decreases more this surface element gets smaller too and eventually disappears completely as $\alpha \rightarrow 0$. Therefore, there is a competition between the decreasing area in this surface element and the increasing direction of the normal pointing more towards the x-direction. Hence, we find a maximum in the tilt angle sensitivity for small $\alpha \approx 0.3$. However, we note that the model is not applicable for $\alpha \ll 1$, since non-linear plasma and fluid dynamics effects become increasingly more important when all laser energy is focused into a tight spot. In that case, the complete plasma and droplet fluid dynamics must be taken into account. In practice, for the μm -sized droplets considered here such tight focus

Figure 4.5: The calculated tilt angle sensitivity $f(\alpha)/R_{\text{eff}}$ expressed in units $^{\circ}/\mu\text{m}$ as function of beam width σ around zero misalignment ($\beta = 0$). The curves are the analytically calculated tilt angle sensitivities from Eq. (4.5), with $\delta_p = 0.6$ and for several relevant values for the effective droplet size $R_{\text{eff}} = [10, 15, 25, 35, 50] \mu\text{m}$. The red square, green diamond and blue circle represent the values corresponding to the experimental conditions (see Section 4.4, color coding as in Fig. 4.3).

cannot be reached and thus typically $\alpha \gg 0$.

4.5 Discussion and Industrial application

Careful control over the tilt angle sensitivity and target expansion is of crucial importance for the operating stability and CE of EUV light sources [1–4]. In the following, we apply our now validated model to predict sensitivities for several practical cases that are immediately relevant for current state-of-the-art industrial droplet-based EUV light sources. In the industrial context, tilt sensitivity is typically expressed as $\theta_{\text{tilt}}/\Delta x$ (in degrees tilt / μm misalignment). Following Eq. (4.7), $\theta_{\text{tilt}}/\Delta x = f(\alpha)/R_{\text{eff}}$.

In Fig. 4.5 we present our model predictions for the sensitivity $\theta_{\text{tilt}}/\Delta x$ as function of beam width σ around zero misalignment ($\Delta x = 0$) for several values of the effective droplet size $R_{\text{eff}} = [10, 15, 25, 35, 50] \mu\text{m}$. We note that by plotting for several effective droplet sizes R_{eff} , we incorporate both the droplet size R_0 and the distance from the droplet surface to the plasma critical surface d_{crit} for each case, since

$R_{\text{eff}} = R_0 + d_{\text{crit}}$ (see previous section). In the figure we also show the experimental parameters studied in the previous sections, analogous to Fig. 4.4.

Figure 4.5 shows that increasing the laser spot size beyond $\sim 45 \mu\text{m}$ does not significantly change the sensitivity for the given effective droplet sizes and is therefore not useful. However by increasing the laser spot size, the energy required to maintain a similar droplet expansion increases with $1/\sigma^2$ due to finite overlap between the droplet and the laser beam, as discussed in Ref. [8]. Therefore in practical applications, one should find the optimum conditions balancing between a maximum expansion (i.e. minimizing σ) and a minimal tilt sensitivity (i.e. maximizing σ).

Furthermore, from Fig. 4.5 we observe that the tilt angle sensitivity increases sharply with decreasing laser focus spot size, especially when $\sigma < R_{\text{eff}}$. Under such focusing conditions, a change in the effective size of the droplet R_{eff} has a strong effect on the sensitivity. An interesting way to change the tilt sensitivity, apart from adapting the actual droplet radius R_0 , is by changing the laser pulse energy or its wavelength. Shorter wavelengths or lower pulse energies result in a smaller d_{crit} and hence result in a smaller R_{eff} and vice versa.

A particularly interesting industrial application of the model is found in the use of a nanosecond-long CO_2 -laser pre-pulse, at $10.6\text{-}\mu\text{m}$ wavelength. According to radiation-hydrodynamics simulations [13] of the interaction of such energetic laser pulses with tin droplets (at an absorbed intensity of $4 \times 10^9 \text{ W/cm}^2$), the critical surface extends up to about $28 \mu\text{m}$ from the droplet surface. In the particular case of $R_0 = 15 \mu\text{m}$ tin droplets impacted by a laser beam of $\sigma = 25 \mu\text{m}$ studied in Ref. [13], we speculate that the effective system size in our model $R_{\text{eff}} \approx 15 \mu\text{m} + 28 \mu\text{m} \approx 43 \mu\text{m}$. In addition, recent experimental work using CO_2 -lasers impinging on planar solid tin targets showed that the exponent in Eq. (4.2) is significantly larger, $\delta = 0.96$, compared to the case of a Nd-YAG laser [19]. These results allows us to estimate the scaling of the propulsion velocity (and thus, radial expansion velocity [7]) with CO_2 -laser intensity for the $\sigma = 25 \mu\text{m}$ case. Using these values for R_{eff} and δ in Eq. (4.9), we get a sensitivity around zero misalignment of $\theta/\Delta x \approx 0.98^\circ/\mu\text{m}$, which is about 85% larger than the corresponding sensitivity for Nd:YAG $\theta/\Delta x \approx 0.53^\circ/\mu\text{m}$. For the low-energy Nd:YAG cases studied in this chapter, we found that our experiments were well reproduced assuming $R_{\text{eff}} \approx R_0$, which would for the current example lead to a sensitivity of $0.36^\circ/\mu\text{m}$, which differs from the CO_2 case by a factor of 2.7. Of course, the extrapolation of our model to other laser wavelengths requires further experimental validation and is left for future work.

Certain industrial applications may require a finite tilt angle [17]. Our model Eq. (4.4) offers a direct way to predict what misalignment is required to obtain a certain amount of target tilt (see also Fig. 4.3a). The slope of that curve around the required misalignment then gives the new tilt angle sensitivity, which can be calculated numerically by evaluating Eq. (4.5) around this new working point. We note that there are in-fact two planes in which we can induce a finite tilt angle, namely

in the $x - z$ plane (as discussed in this chapter) but also in the $y - z$ plane. Both angles are dependent on misalignments in both x and y directions when the degeneracy is lifted by choosing a finite target tilt through a well-defined, intentional misalignment. Further study is required to see how this fact may be advantageously used to increase source operating stability by minimizing L2D sensitivity along the machine axis with the largest risk of misalignment.

4.6 Conclusions

In tin-droplet-based LPP sources of EUV light, laser-to-droplet alignment plays an important role. A slightly misaligned pre-pulse laser beam can lead to a non-optimal target shape, which causes an inefficient coupling with the main laser pulse and lower conversion efficiency of drive laser light into EUV. Moreover, reflections of the main pulse laser light from the tilted surface may well be detrimental to laser stability.

In this chapter we experimentally validated a simple, intuitive model describing the tilt angle sensitivity of a droplet impacted by a laser pulse with controlled misalignment. Our back-of-the-envelope model for the tilt angle was derived based solely on the direction of the center-of-mass velocity. From this model, we were able to obtain the local tilt angle sensitivity around zero misalignment. We experimentally verified the tilt angle and the tilt angle sensitivity by three different experimental groups with industrially relevant settings of an Nd:YAG laser operating at its fundamental wavelength. We observed an excellent agreement with the model over a broad range of laser pulse energies and two laser focus spot sizes. Further, we applied our validated model to predict sensitivities for several practical cases that are immediately relevant for current state-of-the-art industrial droplet-based EUV light sources.

Our model is a simple first-order approximation of the underlying plasma and fluid physics. Full three-dimensional simulations incorporating the complete plasma dynamics should be carried out to obtain the tilt angle as function of the full parameter space. Nonetheless, the current model already allows to physically understand the target tilt as a function of the key experimental control parameters.

4.7 Appendix

The full solution of the tilt angle sensitivity $f(\alpha, \delta)$ around $\beta = 0$ following (4.5) as used in Fig. 4.4 calculated by Mathematica [18] is given by

$$\begin{aligned}
f(\alpha, \delta) &= 2^{-\frac{\delta}{2}-3} e^{-\frac{1}{2}i\pi\delta} \delta^{\frac{1}{2}-\frac{\delta}{2}} \left(-\frac{\delta}{\alpha^2} \right)^{\delta/2} \Gamma\left(\frac{\delta+1}{2}\right) \\
&\times \left(\alpha^{\delta+1} \left((\delta+3)\Gamma\left(\frac{\delta+3}{2}, -\frac{\delta}{2\alpha^2}\right) - 2\Gamma\left(\frac{\delta+5}{2}\right) \right) \right) \\
&\times \frac{\left(-2\delta^{\frac{\delta+3}{2}} (\delta+3) e^{\frac{1}{2}\left(\frac{1}{\alpha^2}+i\pi\right)\delta} - i2^{\frac{\delta+3}{2}} (\alpha^2(\delta+1)+\delta) \right)}{\alpha^4 \Gamma\left(\frac{\delta+5}{2}\right) \left(\delta \Gamma\left(\frac{\delta}{2}\right) - 2\Gamma\left(\frac{\delta}{2}+1, -\frac{\delta}{2\alpha^2}\right) \right)}, \quad (4.9)
\end{aligned}$$

where i is the imaginary unit, $\Gamma(z)$ is the Euler gamma function and $\Gamma(a, z)$ is the incomplete gamma function.

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5

Oblique droplet impact onto a deep liquid pool *

The oblique impact of a liquid droplet onto a deep liquid pool is studied numerically with the adaptive volume-of-fluid solver Basilisk. The splashing threshold, cavity formation, cavity evolution and the maximum cavity dimensions are quantified as a function of the Weber number and the impact angle. We compare the numerical results with recent experimental work by Gielen et al. [Phys. Rev. Fluids 2, 083602 (2017)]. Similarly to the experimental results, three different impact regimes are observed: deposition of the droplet onto the pool, single-sided splashing in the direction of the impact and splashing in all directions. We show good qualitative and quantitative agreement of the splashing behaviour and cavity formation between the simulations and the experiments. Furthermore, the simulations provide a three dimensional view of the impact phenomenon, give access to velocity and pressure fields, and allow to explore impact parameters that are hard to achieve experimentally.

5.1 Introduction

The impact of a liquid droplet onto a deep liquid pool induces a broad range of fascinating physical phenomena [1]. Droplets impacting with a low velocity coalesce with the pool directly or undergoes a coalescence cascade [2–6]. Higher impact velocities

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induce the ejection of a sheet above the pool surface [7–9], which upon breakup could result in a splash [10–14]. During impact a cavity develops below the pool surface [15–17]. The collapse of this cavity [18–20] in combination with capillary wave dynamics can result in bubble entrapment [21–23], the formation of a fast microjet [24, 25] and a Worthington jet [26].

The vast majority of droplet impact studies in the literature have considered perpendicular impacts. However, in many practical situations droplets do not impact perpendicularly but under an angle, i.e. obliquely. Examples range from rain impact in nature [27, 28] to impact on turbine blades [29], tin catching devices in extreme ultraviolet lithography machines [30] and spray cooling [31] in industry. Precise control of the impact behaviour is key to optimizing these industrial applications in order to prevent damage or contamination.

Recently, the regimes of oblique droplet impact onto deep liquid pools [5, 13, 32], moving films [33, 34] and inclined surfaces [35, 36] have been studied experimentally. Furthermore, over the past decades there has been extensive research into analytic modelling of oblique high-velocity impacts onto a inviscid liquid pool [37]. Although these analytic methods allow for a thorough understanding of the early time evolution of e.g. the ejecta sheet [38–41], numerical studies are required to capture the full impact dynamics.

Most numerical studies so far focused on droplet impact on thin liquid films [42–45], wet walls [46–48], perpendicular impact onto deep liquid pools [11, 49–51] or bubble entrainment by impacting droplets [11, 21, 52]. The coalescence and splashing regimes for droplets impacting a deep and shallow liquid pool under an impingement angle where numerically studied by [53]. As these authors performed two-dimensional simulations, they were only able to capture basic impact features.

In this work we present a three-dimensional (3D) numerical study of oblique droplet impact onto a deep liquid pool using the open-source volume-of-fluid solver Basilisk [54]. The numerical results are compared to recent experimental results by [13] and show that the method is able to capture the impact dynamics with excellent accuracy. Details of the numerical setup are provided in section 5.2. In section 5.3 the simulation results are discussed and compared with the experimental results. In section 5.4 we discuss and conclude this work.

5.2 Numerical setup

To solve the 3D incompressible Navier-Stokes equations together with the Volume of Fluid (VOF) method to support two-phase flow, we employ the parallelized octree-adaptive numerical method Basilisk [54]. Basilisk is the successor of the Gerris flow solver and has been extensively validated on problems regarding two-phase complex

Figure 5.1: Schematic views of the numerical setup. A liquid droplet with diameter D and density ρ_{liq} moves with a velocity U under an angle α through air with density ρ_{air} towards a liquid pool. We define the trailing side as the left and leading side as the right hand side of the droplet. (a) Side-view illustrating the parallel $U_{\parallel}$ and perpendicular $U_{\perp}$ velocity components. (b) Front-view with a symmetry plane at $z = 0$. By simulating only one half of the domain and using a mirror boundary condition on the plane we reduce the simulation time by approximately a factor of two.

flows [55–57]. The momentum and VOF equations solved are

$$\nabla \cdot \mathbf{u} = 0, \quad (5.1)$$

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = \frac{1}{\rho} \left[-\nabla p + \nabla \cdot \mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \rho \mathbf{a} + \sigma \kappa \delta_s \mathbf{n} \right], \quad (5.2)$$

$$\frac{\partial f}{\partial t} + \nabla \cdot (f \mathbf{u}) = 0, \quad (5.3)$$

where $\mathbf{u}$ is the velocity field, ρ the density, p the pressure, μ the viscosity, $\mathbf{a}$ optional body forces, σ the surface tension, κ the surface curvature, δ_s a delta function on the interface between two fluids, $\mathbf{n}$ the surface normal vector on this interface and f the volume fraction field used in the VOF method.

The fluid equations (5.1,5.2) are solved by a finite volume method over an adaptive Eulerian grid with the Bell-Colella-Glaz second-order accurate advection scheme [58, 59]. The volume fraction field (5.3) is solved by a piecewise-linear geometrical scheme by [60] together with a continuum surface force model for the surface tension in (5.2) [55, 61].

A sketch of the numerical setup is given in figure 5.1. A spherical liquid droplet with diameter D and density ρ_{liq} falls with a velocity U under an angle α through air with density ρ_{air} towards a liquid pool with density ρ_{liq} . We define the leading

Figure 5.2: A side-view of three different types of impact behaviour observed above and below the pool surface in the simulations (top panel) and in the experiments (bottom panel) at identical times. From left to right: deposition (first column), single-sided splashing (second column) and omni-directional splashing (last column). Simulation parameters (a) $We = 187$, $\alpha = 26^\circ$, (b) $We = 400$, $\alpha = 30^\circ$ and (c) $We = 600$, $\alpha = 10^\circ$. Experimental parameters (d) $We = 180.5$, $\alpha = 28.1^\circ$, (e) $We = 419$, $\alpha = 27.9^\circ$ and (f) $We = 662$, $\alpha = 0.6^\circ$. Experiments by [13].

side as the right- and trailing side as the left-hand side of the droplet. The pool depth is $8D$, the cubic simulation domain edges are $12D$ and the initial distance from the droplet to the pool is $0.1D$. The density and viscosity ratio between the liquid and gas phase in the simulation are $\rho_{liq}/\rho_{air} = 1000$, $\mu_{liq}/\mu_{air} = 100$, respectively. To ensure stability of the simulation the Courant-Friedrichs-Lewy (CFL) condition is set to $CFL = 0.05$ [54]. Figure 5.1 shows a side view (a) and front view (b) of the simulation. A symmetry plane is enforced in the $y - z$ plane (see figure 5.1b) allowing us to simulate only half the droplet and save simulation time. Note that this constraint may restrict the development of a Rayleigh-Taylor instability at the rim of the ejecta sheet, therefore if one is interested in a detailed analysis of the fingering instability this constraint should be released. At the bottom of the pool no-slip and impermeability conditions are used, while at the top of the domain free-slip and free-outflow conditions are imposed. For the remaining side planes symmetry boundary conditions are used. The impact location is in the center of the domain, such that the domain walls are of little influence on the early-time dynamics of the impact.

The grid is refined adaptively using the wavelet adaptation method, which is a build-in refinement strategy in the Basilisk framework [62, 63]. The maximum refinement level on the grid is bounded to the computational resources and time available, which leads to a maximum refinement level $r_{max} = 12$ and hence a theoretical maximum grid resolution of 4096^3 or about 10^5 cells per droplet cross section when

Figure 5.3: Time series of a single-sided crown splash observed above and below the pool surface in the simulation (two top panels) and in the experiment (bottom panel). Times are non-dimensionalized by $t_i = D/U$, where $t/t_i = 0$ corresponds to the moment of first contact with the pool. Simulation parameters $We = 400$, $\alpha = 30^\circ$, experiment $We = 416.5$, $\alpha = 28.5$. (a) $t/t_i = 0.46$: when the droplet hits the surface a sheet ejects on the leading side of the impact area. (b) $t/t_i = 2.33$: the sheet evolves into a crown leading to several satellite droplets in the simulation that are hard to detect in the experiment. (c) $t/t_i = 8.22$: the crown further expands outwards and the satellite droplets are now clearly visible in both simulations and experiments. (d) $t/t_i = 12.15$: the crown retracts due to capillary forces and a finger appears at the leading side. (e) $t/t_i = 18.00$: the liquid finger collapses onto the pool due to surface tension forces. Experiments by [13].

all volumes are refined. The refinement algorithm is invoked every time-step and refines when the wavelet estimated error exceeds $\mathbf{u}_{\text{err}} = 10^{-2}$ for the velocity field and $f_{\text{err}} = 5 * 10^{-3}$ for the fraction field. Initially we refine the interface of the droplet to r_{max} while the rest of domain stays at $r_{\text{initial}} = 6$. With these settings a typical simulation of a single impact event from the moment of impact until the cavity closes takes about a month using a modern computer cluster in parallel. During the simulation we set fraction field values lower than 10^{-4} to 0 and values higher than $(1 - 10^{-4})$ to 1. This smoothing reduces noise caused by tiny droplets and bubbles in the simulation that do not affect the overall dynamics but are costly to track. This filtering may alter the dynamics of bubble entrapment, which is not the focus of the present work.

By neglecting the effect of the ambient air, oblique impact of a droplet onto a liquid pool can be characterized by four dimensionless parameters: the Weber number $We = \frac{\rho_{\text{liq}} D U^2}{\sigma}$, the Reynolds number $Re = \frac{\rho_{\text{liq}} U D}{\mu_{\text{liq}}}$, the Froude number $Fr^2 = \frac{U^2}{gD}$ and the impact angle α , where g is the gravitational acceleration [13]. We are interested in an

We	$\alpha(^{\circ})$	We	$\alpha(^{\circ})$
187.5	28	400	60
200	15	400	75
200	40	600	10
200	45	600	20
250	35	600	45
250	60	600	75
300	20	674	28
400	12	800	20
400	20	1000	55
400	30	1400	45
400	40	1400	60

Table 5.1: A list of simulations performed with the parameters as described in section 5.2. The Reynolds number is fixed to $Re = 1000$ in all simulations and the gravity is disabled.

impact regime where the influence of Froude and Reynolds is negligible (in the experiments used for comparison [13] $Fr \in [10^2 - 10^3]$ and $Re \in [600 - 2500]$). Hence in the simulations, the gravitational contribution is neglected, the impact velocity is set to $U = 1$ and the liquid viscosity is set such that the Reynolds number is always equal to $Re = 1000$. We then explore a simple two-dimensional phase space (We, α) . Table 5.1 gives a list of the oblique droplet impact simulations performed. We have chosen the simulations parameters (We, α) such that we cover a comprehensive set of splashing phenomena in the phase space. These simulated parameters may therefore not always be a one-to-one match with the experiments performed by [13]. In the analysis of the results it will turn out practical to discriminate between the parallel Weber number $We_{\parallel} = \frac{\rho_{liq} D U_{\parallel}^2}{\sigma}$ and the perpendicular Weber number $We_{\perp} = \frac{\rho_{liq} D U_{\perp}^2}{\sigma}$, where $U_{\parallel} = U \sin(\alpha)$ and $U_{\perp} = U \cos(\alpha)$ are the velocity components parallel and perpendicular to the pool surface, respectively.

5.3 Results

In this section we present our numerical results and we show a qualitative and quantitative comparison to the experiments performed by [13]. In 5.3.1 the different types of impact phenomena are discussed and classified. An overview of the early impact dynamics is given in section 5.3.2. The splashing threshold of the crown above the pool is systematically studied in section 5.3.3. We discuss the cavity formation and evolution as function of time and compare this result to a theoretical model in section 5.3.4. Finally, we show the maximum cavity dimensions below the surface, i.e. the

maximal cavity depth, maximal cavity displacement and collapse angle as function of Weber number and impact angle in section 5.3.5.

5.3.1 Impact phenomena

When a droplet obliquely impacts onto a pool, a cavity forms below the pool surface accompanied by an asymmetric crown above the pool surface. In our simulations we observe three different impact phenomena similar to what has been observed in experiments: deposition, single-sided splashing and omni-directional splashing [13]. Figure 5.2 shows these three phenomena in both the simulation (top panel) and the experiments (bottom panel).

The first column (figure 5.2a and 5.2d) shows a deposition event for $We = 187$ and $\alpha = 26^\circ$ (simulation) and $We = 180.5$ and $\alpha = 28.1^\circ$ (experiment). Directly after impact a thin sheet ejects on the leading side of the impact cavity. The impact energy of the droplet is not sufficient for this ejecta sheet to develop into a crown on all sides. Instead the ejecta sheet retracts back towards the pool due to capillary forces without any breakup. We observe excellent qualitative agreement of the simulation with the experiment.

The second column (figure 5.2b and 5.2e) shows a single-sided splash for $We = 400$ and $\alpha = 30^\circ$ (simulation) and $We = 419$ and $\alpha = 27.9^\circ$ (experiment). Here, the impact energy is large enough for the ejecta sheet to develop several fingers on the leading side of the cavity, which eventually break up into satellite droplets in both the simulation and experiment. The resulting single-sided crown in the simulation compares very well with the overall crown shape of the corresponding experiment.

Finally in the last column (figure 5.2c and 5.2f) an omni-directional splash is observed for $We = 600$ and $\alpha = 10^\circ$ (simulation) and $We = 662$ and $\alpha = 0.6^\circ$ (experiment). In this case the impact energy is large enough to induce a crown with satellite droplets all around the cavity in both the simulation and the experiment.

Figure 5.3 shows the comparison of a detailed time series of a single-sided splash impact event in the simulation ($We = 400$ and $\alpha = 30^\circ$, two top panels) and in the experiment ($We = 416.5$ and $\alpha = 28.5^\circ$, bottom panel). In addition, the simulation provides a top view of the impact event. Time increases from left to right, with $t/t_i = 0$ being the moment of first contact between the droplet and the pool, with $t_i = D/U$ the inertial time. The simulations show that the droplet pushes liquid away on the trailing side (see top panel of figure 5.3a), effectively creating an air layer between the pool surface and the impacting droplet. This air layer marks the start of a cavity forming below the pool surface, as can be observed in the side view (see mid panel in figure 5.3a). The deepest point of this initial cavity starts on the left side and ramps up along the edge of the impacting droplet towards the pool surface, see mid and top view. Unfortunately, at this stage the cavity is too small to be captured in the experiments as can be seen in the bottom panel. In the simulations, the impact

causes the rise of the ejecta sheet on the leading side (see mid panel in figure 5.3a). A similar ejecta sheet is observed in the experiment (bottom panel), but the resolution is too low to clearly distinguish the ejecta sheet from the droplet.

In figure 5.3b, the ejecta sheet rises further above the surface and fingers appear on the peripheral side of the rim in the simulation. The droplet is now fully submerged in the pool and the cavity has grown to cover the complete impact area, as can be seen from both the side-view and the top-view. In the simulation, the fingers on the ejecta sheet break up and result in satellite droplets, which is clearly visible in the top-view (top-panel). This break up is not clearly visible in the experiment (bottom-panel), but the overall shape of the ejecta sheet is similar to the simulation. Figure 5.3c shows the start of a capillary wave on the trailing side of the cavity that gradually deforms the cavity into a hemispherical shape at later times, see figure 5.3d and figure 5.3e. Above the pool, the satellite droplets have clearly separated from the ejecta sheet in both the simulation and experiment, which is now best described as a single-sided crown. The fingers are retracting back to the rim of the crown. As the crown itself is also retracting, this causes the fingers to move towards the leading side of the crown, which is clearly visible in the top view panels.

In figure 5.3d the retraction of the crown continues while there is only one large finger remaining on the leading side of the crown, which is observed in both numerics and experiments. Finally, in figure 5.3e the cavity has reached its maximum expansion. As the remaining finger collapses into the cavity, a capillary wave is triggered. At later times (not shown here) this wave causes fluctuations in the cavity shape.

5.3.2 Impact dynamics

In the simulation we have access to the pressure and velocity fields induced in the liquid, which gives a detailed view of the early impact dynamics. Figure 5.4 shows a cross-section of the velocity magnitude and streamlines during impact at the mirror plane ($z = 0$) for different times t/t_i and angles α at constant $We = 400$. In figure 5.5 we zoom in on the ejecta sheet region on the leading side of the impacting droplet.

Figure 5.4a shows a perpendicular impact event and serves as a reference case for all consecutive oblique impacts. In the first figure of this row, the droplet has not yet fully submerged into the pool and most of the momentum is still concentrated in the impacting droplet. Inside the pool the streamlines show a symmetric velocity field around the point of impact and bend towards the pool surface. We note that there is a small symmetry deviation between the leading and trailing side of the impacting droplet, which is the result of the adaptive grid refinement strategy used in the simulations. An ejecta sheet is formed on both sides of the impacting droplet, which is more clearly visible in figure 5.5a. The maximum velocity is reached where the ejecta sheet is thinnest. At the edge of the ejecta sheet a cylindrical rim forms due to surface tension forces. At later stages, the droplet fully submerges into the pool and

Figure 5.4: A cross-section of the velocity magnitude and streamlines in the liquid phase at the mirror plane ($z = 0$) (see figure 5.1) for $We = 400$ at different angles α and different times t/t_i . The line $y = 0$ denotes the base level of the pool. The velocity magnitudes (color bar) are scaled by the impact velocity U (note that color bar changes for each row). The light blue color indicates the gas phase.

Figure 5.5: A zoom of the cross-section of the velocity magnitude and streamlines of the ejecta sheet on the leading side of the impacting droplet at the mirror plane ($z = 0$) (see figure 5.1) for $We = 400$ at different angles α and different times t/t_i . The line $y = 0$ denotes the base level of the pool. The velocity magnitudes (color bar) are scaled by the impact velocity U (note that color bar changes for each row). The light blue color indicates the gas phase.

a symmetrically shaped cavity forms. The ejecta sheet keeps rising until it retracts at later stages (not shown).

Figure 5.4b shows an oblique impact event at $\alpha = 20^\circ$. The ejecta sheet is no longer symmetric but larger on the leading-side than on the trailing side. Figure 5.5b shows that the initial velocity magnitude inside the ejecta sheet on the leading side is about 50% higher than the original impact speed of the droplet. Furthermore, the maximum velocity in the ejecta sheet exceeds the one achieved during perpendicular impact, see top row. The second figure in this row shows that a satellite droplet is created and as a result the tip now has an oval shape, also seen in the zoomed figure 5.5b. Below the pool surface a cavity starts to form, which grows fastest in the direction of the impacting droplet as shown by the streamlines and velocity magnitudes. Interestingly, the far-field velocity streamlines still resemble the far-field streamlines of the perpendicular case.

The dynamics of an impacting droplet at $\alpha = 40^\circ$ is shown in figure 5.4c. During impact a small air layer is entrapped between the pool and droplet, which leads to a set of bubbles concentrated on the left side along the interface of the coalescing droplet with the pool. These bubbles have a local effect on the velocity field which is clearly visible in the first figure of 5.4c. The ejecta sheet on the trailing side is now better described as a liquid bump and carries little momentum. Instead, most of the momentum is concentrated in the ejecta sheet on the leading side where the velocity magnitude is twice of the original impact speed of the droplet. In contrast to the perpendicular case, the ejecta sheet initially grows in the direction along the pool surface but later on deflects upwards, see figure 5.5c. We note that the evolution and shape of the ejecta sheet in figure 5.5b and 5.5c may have a grid dependence, i.e. depends on the grid refinement history. Unfortunately, increasing the maximal grid resolution to $r = 13$ was not feasible with the computational resources available to us today. The bubbles entrapped in the cavity do not seem to affect the cavity evolution, which similar to previous cases grows faster in the direction of the impacting droplet. Again we note that the far field streamlines resemble those of the perpendicular impact.

Finally, in figure 5.4d we show an impacting droplet at $\alpha = 60^\circ$. Similarly to the previous case bubbles appear on the left-hand side of the interface between the impacting droplet and pool. During impact the droplet partly coalesces with the pool but also partly slides along the surface of the pool, as seen in the first figure of 5.4d. The contour lines of the velocity field confirm this observation, since they form a closely packed layer between the impacting droplet and the pool in which the magnitude and direction rapidly change. The ejecta sheet on the leading side is thicker compared to the previous cases and expands parallel to the surface, as clearly visible in figure 5.5d. Here, the ejecta sheet does not change direction over time and remains parallel to the surface in all plotted stages. The tip of the sheet eventually falls down onto the pool (see second figure of 5.5d). The secondary impact of this sheet onto the pool entraps another air layer on the leading side. Furthermore, the

Figure 5.6: Phase diagram of the impact behaviour as a function of the dimensionless splashing parameter $We^{1/2}Re^{1/4}/K$ and impact angle α for both the experimental results by [13] (a) and the simulation results (b), where K is the critical splashing number. The blue filled squares, black open circles and red closed triangles depict deposition, single-side splashing and omni-directional splashing respectively. The solid and dotted lines are a scaling model that predict the transitions from deposition to single-sided splashing and from single-sided splashing to omni-directional splashing, see main text. In both panels we used $c = 0.44$ for the fit parameter measuring the mass-flow distribution around the crown.

coalescence results in a complex wave pattern on the surface as shown by last figure of 5.5d. The cavity does no longer resemble that of the perpendicular case and even the far-field streamlines (on this scale) start to show asymmetry.

5.3.3 Splashing threshold

The three impact phenomena (deposition, single-sided splashing, omni-directional splashing) observed can be quantified in a phase diagram. Figure 5.6 shows such a splashing phase diagram in terms of the impact parameters $We^{1/2}Re^{1/4}/K$ and α (as discussed by [13]), where K is the critical splashing number. Figure 5.6a shows the experimental results by [13] and figure 5.6b shows the simulation results. The transitions between these different impact phenomena (denoted by the black solid and dotted lines) can be explained by the scaling argument derived by [13]. For completeness, we briefly repeat the argument here.

We assume that the mass flow into the crown is directly proportional to the mass flow into the pool [10]. Since the droplet impacts under an angle α , the leading side of the crown accumulates more mass than the trailing side. The mass balance of the

Figure 5.7: Schematic side-view of the moment when the cavity reaches its maximum depth $d_{\max}$, height $h_{\max}$ and angle α_{cm} .

crown reads [13]

$$\rho_l D^2 U_{\perp} \pm \rho_l c D^2 U_{\parallel} \sim \rho_l e D V, \quad (5.4)$$

where the left-hand side represents the mass flow into the leading (+) and trailing (−) side of the crown, $e \ll D$ is the thickness of the crown at the pool surface, V is the ejection velocity of the crown and c is a fit parameter that accounts for the exact mass-flow distribution over the crown. To define the splashing threshold it is assumed that breakup of the crown occurs when the crown ejection velocity V , scaled by the Taylor-Culick velocity $V_{\text{TC}} \sim \sqrt{\frac{\sigma}{\rho_l e}}$, exceeds a critical value of the splashing number K [10, 13]. By using $e \sim \sqrt{vD/U}$ for the thickness of the crown at its base [8], we find as splashing criterion of the crown [13]

$$\frac{\text{We}^{1/2} \text{Re}^{1/4}}{K} \cos(\alpha)^{5/4} \left[1 \pm c \tan(\alpha) \right] > 1. \quad (5.5)$$

The value of K is determined from a perpendicular impact event ($\alpha = 0$) and used for all other impacts. In the simulation this leads to $K \approx 104$ and $K \approx 130$ for the experiment [13]. We note that the critical splashing number is dependent on the spatial and temporal resolution of the simulation, i.e. sensitive to under resolution, but has converged at the grid resolution used as described in section 5.2. The fitting parameter c is set to $c = 0.44$, following [13].

The splashing criterion (5.5) predicts two transitions in the phase space: (i) a transition from deposition to single-sided splashing when the splashing criterion is met on a single side of the crown (the solid line in figure 5.6) and (ii) a transition from single-sided splashing to omni-directional splashing when the criterion is fulfilled

Figure 5.8: Cavity depth h/D as function of time t/t_i for different Weber numbers and impact angles (symbols). The solid line corresponds to the theoretical model (5.7) as derived by [64].

on both sides of the crown (the dashed line in figure 5.6). We note that (5.5) is a geometric argument and merely gives a dimensionless scale for which we can expect breakup to happen based on the splashing criterion in the perpendicular case.

We find good quantitative agreement between the simulations, the experiments and the scaling model in all splashing regimes. The experiments show a zone where all three impact behaviours overlap for $\frac{We^{1/2}Re^{1/4}}{K} \approx 1$ and $\alpha < 20^\circ$ (see panel a) [13] which is absent in both the model and the simulations (panel b). This impact region is sensitive to small variations in the impact parameters in the experiment and the two-dimensional view makes it difficult to visually distinguish between different impact behaviours. The simulation provides full 3D insight in the impact dynamics and therefore allows for a better judgment on the splashing behaviour in this regime.

5.3.4 Cavity dynamics

We now turn to a quantitative analysis of the cavity dynamics. A schematic view of the cavity shape is given in figure 5.7. We define the maximum cavity dimensions, similar to [13], by the maximum cavity depth $h_{\max}$, maximum cavity displacement $d_{\max}$ and the maximum cavity angle α_{cm} .

The cavity depth h as function of time t/t_i is plotted in figure 5.8 for different Weber numbers and impact angles. It turns out that the cavity dynamics at early

Figure 5.9: (a) Double-logarithmic plot of the maximum cavity depth $h_{\max}$ as function of the Weber number $We_{\perp}$ and (b) the maximum displacement $d_{\max}$ as function of the parallel Weber number $We_{\parallel}$. The solid line in (a) and (b) denotes the scaling law (5.8,5.9) with prefactors of 0.19 and 0.12 taken from experiment and numerical results. (c) Maximum cavity angle α_{cm} as function of the impact angle α . The solid line in (c) is the line with $\alpha_{\text{cm}} = \alpha$. The open black circles correspond to the experiments and the closed red triangles to the simulations. The error bars in the simulation data are a result of the uncertainty in the cavity depth and displacement, which are subject to fluctuations.

times ($t/t_i < 4$) follows the same trend for all impact angles studied. This observation inspired us to describe the cavity depth evolution by a model that was previously derived for perpendicular impacts [64, 65]. Here, the cavity depth is modelled as a hemisphere where the unsteady Bernoulli equation together with a balance of stresses at the cavity interface is used to describe its evolution. In the limiting case of $We \gg 1$ and $Fr \gg 1$ the evolution of the cavity depth reads [64, 65]

$$\frac{d^2 h}{dt^2} = -\frac{3}{2h} \left(\frac{dh}{dt} \right)^2, \quad (5.6)$$

which has the solution

$$h(t/t_i)/D = c_1(5t/t_i - c_2)^{2/5}, \quad (5.7)$$

where $c_1 = 0.47$ and $c_2 = 5.87$ are constants determined by a least-square fit to the data with impact angles $\leq 30^\circ$ (i.e. nearly perpendicular) for $2 < t/t_i < 15$, which is the regime where the model applies. The model captures the simulation results with good accuracy for impact angles smaller than 30° . When the impact angle is larger than 30° the cavity can no longer be modelled as an expanding hemisphere. Instead, due to the large impact angle the cavity is stretched in the direction parallel to the pool surface, leading to a cone-shaped cavity. Indeed, in figure 5.4 we observe that for 40° the streamlines start to deviate from the perpendicular impact case. As a result the cavity depth becomes smaller than the depth predicted by the hemispherical model, as is clear from figure 5.8.

5.3.5 Cavity dimensions

Figure 5.9 gives an overview of the cavity dimensions $h_{\max}$ and $d_{\max}$ as function of $We_{\parallel}$, $We_{\perp}$ and α . In the top two panels we show a double-logarithmic plot of the maximum cavity depth $h_{\max}$ (left) and the maximum cavity displacement $d_{\max}$ (right) as function of respectively the perpendicular and parallel Weber number. The black open circles represent the experimental results by [13] and the red closed triangles are the numerical results. The solid lines corresponds to a scaling argument that relates the kinetic energy of the droplet to the surface energy the cavity acquires, which leads to [13]

$$\frac{h_{\max}}{D} \sim We_{\parallel}^{1/2}, \quad (5.8)$$

$$\frac{d_{\max}}{D} \sim We_{\perp}^{1/2}, \quad (5.9)$$

where it was assumed that $\alpha_{\text{cm}} \approx \alpha$. The simulation results are in agreement with the experimental data. The scaling argument confirms the trend for both the cavity

depth and cavity displacement. There is however a large uncertainty for low Weber numbers in the experimental data for the cavity displacement, since in this regime the displacement is hard to measure [13].

Figure 5.9c shows a double-logarithmic plot of the maximum cavity angle α_{cm} as function of the impact angle α . Similar to the experimental results, we observe that the maximum cavity angle increases linearly with increasing impact angle up to $\alpha \approx 30^\circ$. For larger impact angles, the cavity angle is smaller than the impact angle. This effect was also observed in the experiments by [13], and was attributed to energy dissipation by waves tangential to the surface.

5.4 Discussion & Conclusion

We presented a numerical study of oblique droplet impact onto a deep liquid pool by using the adaptive volume-of-fluid solver Basilisk. The splashing behaviour and cavity dynamics after impact were quantified as function the Weber number and the impact angle. The numerical results were compared to recent experimental work by [13]. We found good qualitative and quantitative agreement between the numerical and experimental results on the dynamics of both the sheet above and the cavity below the pool surface for a broad range of Weber numbers and impact angles. In particular, the splashing threshold, the maximum cavity depth, maximum cavity displacement and cavity angle were found to be in good agreement with the experiments.

The spatial and temporal resolution of the simulations was chosen such that the sheet dynamics up to the moment of breakup and the cavity dynamics up to the moment the maximum cavity depth is reached are converged. These resolution parameters are bound by the time and computational resources available, which limited us to a maximum Weber number $\mathcal{O}(10^3)$. In the higher Weber number regime sharp interfaces and large velocity gradients dominate the dynamics. Insufficient resolution may therefore cause the sheet and cavity dynamics to be captured incorrectly.

To be able to increase the grid resolution while maintaining a reasonable computation time we imposed a symmetry condition in the $y - z$ plane (see 5.1). We found that this symmetry condition improved the convergence of the overall sheet dynamics. This constraint however may restrict the Rayleigh-Taylor instability from developing correctly at the rim of the crown. For a detailed analysis of the fingering instability the constraint should therefore be released.

The numerical simulations provide a valuable addition to the experimental results, because they capture a fully 3D view of the impact phenomenon which allows for a better judgment on the single-sided and omni-directional splashing thresholds. In addition, the method provides access to velocity and pressure fields which are not available in the experiments and allow to obtain more insight into the cavity and ejecta-sheet dynamics. Moreover, the method can be used to explore impact param-

eters that are hard to reach experimentally, e.g. very large impact angles or high droplet velocities.

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6

A sequence approach to solve the Burgers' equation *

The Burgers' equation is a one-dimensional momentum equation for a Newtonian fluid. The Cole-Hopf transformation solves the equation for a given initial and boundary condition. However, in most cases the resulting integral equation can only be solved numerically. In this work a new semi-analytic solving method is introduced for analytic and bounded series solutions of the Burgers' equation. It is demonstrated that a sequence transformation can split the non-linear Burgers' equation into a sequence of linear diffusion equations. Each consecutive sequence element can be solved recursively using the Green's function method. The general solution to the Burgers' equation can therefore be written as a recursive integral equation for any initial and boundary condition. For a complex exponential function as initial condition we derive a new analytic solution of the Burgers' equation in terms of the Bell polynomials. The new solution converges absolutely and uniformly and matches a numerical solution with arbitrary precision. The presented semi-analytic solving method can be generalized to a larger class of non-linear partial differential equations which we leave for future work.

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6.1 Introduction

The Burgers' equation is a momentum equation for a one-dimensional viscous fluid [1]. The equation describes a competition in the velocity field between non-linear convection and linear diffusion. It has an important historical significance in modelling turbulence, plays an important role in non-linear acoustics and has applications in traffic flows [2–6].

The Cole-Hopf transformation converts the Burgers' equation into a linear diffusion equation which can be solved by the Green's function method for any initial and boundary condition [7, 8]. For some initial data the resulting integral equation can be solved explicitly in a closed-form solution or series solution [9, 10]. For many initial and boundary conditions however explicit solutions cannot be obtained and the integral equation has to be solved numerically. As a result, the capability to analytically understand the non-linear dynamics is limited and other semi-analytic solving methods become important.

In this work we solve the Burgers' equation using a sequence approach. We show that the Burgers' equation can be transformed into a sequence of linear diffusion equations for holomorphic and bounded series solutions. Each consecutive sequence element can be solved recursively using the Green's function method. The general solution to any initial value problem of the Burgers' equation can therefore be written as a recursive integral equation. This novel semi-analytic method is termed the sequence transformation method. For some initial and boundary conditions this recursive integral equation can be solved explicitly and written in a closed-form series. We show that we are able to obtain a closed-form series solution for a specific initial condition for which the Cole-Hopf integral solution holds no known closed-form solution.

Other semi-analytical methods such as the homotopy analysis method, Adomian decomposition method, differential transform method and variational iteration method have been successfully used in the past to solve the Burgers' equation [11–18]. The approach in these methods resembles our approach, in the sense that the solution is an infinite series whose terms can be calculated term-wise. However our approach does not require a deformation parameter, an auxiliary convergence parameter, predefined expansion polynomials, an expansion parameter, explicit Taylor series or Lagrange multipliers. Instead, the sequence of linear diffusion equations follows naturally from a sequence transformation applied to the Burgers' equation. Any initial condition, boundary condition or source term can be straightforwardly integrated using the Green's function method.

In §6.2 a Banach space is introduced including a set of sequence operations. The sequence transformation method is presented in §6.3. In §6.4 the complex valued Burgers' differential equation is given. A new and complete analytic solution to the Burgers' equation is presented in §6.5 written in terms of the Bell polynomials for a

complex exponential function as initial condition. Finally in §6.6 we conclude and discuss possible future directions.

6.2 Preliminaries

In §6.2.1 we introduce a Banach space for a class of well behaved and physically relevant functions. In the remainder of this section we discuss four important operations in the Banach space that are used in this work. First in §6.2.2 we show that differentiation and integration for a series in this space can be performed term-wise. Finally in §6.2.3 the Cauchy product is introduced for two series.

6.2.1 Banach space and series

Let $\Omega = \Omega_z \times \Omega_w \subseteq \mathbb{C}^2$ be an open subset and let $\mathcal{F}(\Omega)$ be a Banach space of all bounded holomorphic functions of two variables $f(z, w) : \Omega \rightarrow \mathbb{C}$ with the supremum norm $\|f(z, w)\| := \sup_{(z, w) \in \Omega} |f(z, w)|$. We define the sequence space $\mathcal{F}_s(\Omega)$ as the set of all possible infinite sequences $\{f_n(z, w)\}$ with $f_n(z, w) \in \mathcal{F}(\Omega)$ and $n \in \mathbb{N}$ whose series converges by the Weierstrass M-test. That is, a sequence $\{f_n(z, w)\}$ is only member of $\mathcal{F}_s(\Omega)$ when $\|f_n(z, w)\| \leq M_n$ and $\sum_{n=1}^{\infty} M_n < \infty$. As a result, any sequence $\{f_n(z, w)\} \in \mathcal{F}_s(\Omega)$ has a series that converges absolutely and uniformly on Ω to a member in $\mathcal{F}(\Omega)$, since the Banach space is complete. Our notation for a series is

$$f(z, w) := \sum \{f_n(z, w)\} := \sum_n f_n(z, w), \quad (6.1)$$

where the summation over a sequence results in a series.

6.2.2 The differentiation and integration of a series

A series $f(z, w) \in \mathcal{F}(\Omega)$ of the sequence $\{f_n(z, w)\} \in \mathcal{F}_s(\Omega)$ is infinitely differentiable term-wise with respect to z or w , since $f(z, w)$ converges uniformly in Ω and is analytic in both z and w . Furthermore integration of the series $f(z, w)$ along any continuous path in Ω with respect to the variable z or w can be performed term-wise, since $f(z, w)$ converges uniformly in Ω and is continuous in both z and w .

6.2.3 The product of two series

Two series $f(z, w) \in \mathcal{F}(\Omega)$ of the sequence $\{f_n(z, w)\} \in \mathcal{F}_s(\Omega)$ and $g(z, w) \in \mathcal{F}(\Omega)$ of the sequence $\{g_n(z, w)\} \in \mathcal{F}_s(\Omega)$ may be multiplied together using the Cauchy

product as

$$f(z, w)g(z, w) := \left(\sum_n f_n(z, w) \right) \left(\sum_n g_n(z, w) \right) = \sum_n \left(\sum_{m=1}^{n-1} f_m(z, w)g_{n-m}(z, w) \right). \quad (6.2)$$

The functions $f(z, w)$, $g(z, w)$ and the product $f(z, w)g(z, w)$ are absolutely and uniformly convergent in Ω . Therefore we have that the product $f(z, w)g(z, w)$ is a member of $\mathcal{F}(\Omega)$, since the Banach space is complete.

6.3 The sequence transformation method

Let us define a transformation $\mathcal{T} : \mathcal{F}_s(\Omega) \rightarrow \mathcal{F}(\Omega)$ for a sequence $\{f_n(z, w)\} \in \mathcal{F}_s(\Omega)$ to a tagged series $\hat{f}(z, w, s) \in \mathcal{F}(\Omega)$ by

$$\mathcal{T} \left[\{f_n(z, w)\} \right] := \sum_n f_n(z, w) \exp(ins), \quad (6.3)$$

where i is the imaginary unit and $s \in [-\pi, \pi]$. The tagged series

$$\hat{f}(z, w, s) = \sum_n f_n(z, w) \exp(ins) \quad (6.4)$$

converges absolutely and uniform on $\Omega \times [-\pi, \pi]$, because

$$||f_n(z, w) \exp(ins)|| \leq ||f_n(z, w)|| \leq M_n$$

for all $(z, w, s) \in \Omega \times [-\pi, \pi]$. The inverse transformation $\mathcal{T}^{-1} : \mathcal{F}(\Omega) \rightarrow \mathcal{F}_s(\Omega)$ from a tagged series $\hat{f}(z, w, s) \in \mathcal{F}(\Omega)$ to a sequence $\{f_m(z, w)\} \in \mathcal{F}_s(\Omega)$ is defined as

$$\mathcal{T}^{-1} \left[\hat{f}(z, w, s) \right] = \left\{ \frac{1}{2\pi} \int_{-\pi}^{\pi} \hat{f}(z, w, s) \exp(-ims) ds \right\} = \{f_m(z, w)\}, \quad (6.5)$$

where $m \in \mathbb{N}$ and we are allowed to integrate term-by-term, since the integrand converges uniformly on $\Omega \times [-\pi, \pi]$. The identity transform is given by

$$\begin{aligned} \mathcal{T}^{-1} \mathcal{T} \left[\{f_n(z, w)\} \right] &= \left\{ \frac{1}{2\pi} \int_{-\pi}^{\pi} \left(\sum_n f_n(z, w) \exp(ins) \right) \exp(-ims) ds \right\} \\ &= \left\{ \sum_n \frac{1}{2\pi} \int_{-\pi}^{\pi} f_n(z, w) \exp(i(n-m)s) ds \right\} \\ &= \{f_m(z, w)\}. \end{aligned}$$

The remarkable feature of this transformation is that every sequence in $\mathcal{F}_s(\Omega)$ has a tagged series for which the inverse transformation allows to obtain the original sequence. We can use this property to split a non-linear differential equation into a sequence of linear differential equations which we show in the next section for the Burgers' equation.

6.4 The Burgers' equation

Let $f(z, w) \in \mathcal{F}(\Omega)$ be a series of the sequence $\{f_n(z, w)\} \in \mathcal{F}_s(\Omega)$ and be a solution of the Burgers' differential operator $\mathcal{A} : \mathcal{F}(\Omega) \rightarrow \mathcal{F}(\Omega)$ given by the zero-map

$$\mathcal{A} \left[f(z, w) \right] = \frac{\partial f(z, w)}{\partial w} - \nu \frac{\partial^2 f(z, w)}{\partial z^2} + f(z, w) \frac{\partial f(z, w)}{\partial z} = 0, \quad (6.6)$$

where $\nu \in \mathbb{R} > 0$ is the viscosity.

In §6.4.1 we apply the sequence transformation to the Burgers' equation and show that the non-linear differential equation can be decomposed in a sequence of linear diffusion equations for all holomorphic and bounded series solutions. Finally in §6.4.2 the Green's function method is introduced to solve the resulting sequence of linear diffusion equations for an initial value problem and we present the general solution to the Burgers' equation.

6.4.1 The sequence differential equation

Instead of solving (6.6) directly for a function $f(z, w)$, we first decompose the non-linear equation into a sequence of linear diffusion equations by making use of the sequence transformation, see §6.3. We start by applying the differential operator $\mathcal{A}$ to a tagged series $\hat{f}(z, w, s)$ and calculate the inverse sequence transformation $\mathcal{T}^{-1}$ by

$$\begin{aligned} \mathcal{T}^{-1} \left[\mathcal{A} \left[\hat{f}(z, w, s) \right] \right] &= \mathcal{T}^{-1} \left[\frac{\partial \hat{f}(z, w, s)}{\partial w} - \nu \frac{\partial^2 \hat{f}(z, w, s)}{\partial z^2} + \hat{f}(z, w, s) \frac{\partial \hat{f}(z, w, s)}{\partial z} \right] \\ &= \mathcal{T}^{-1} \left[\sum_n \left(\frac{\partial f_n(z, w) \exp(ins)}{\partial w} - \nu \frac{\partial^2 f_n(z, w) \exp(ins)}{\partial z^2} \right. \right. \\ &\quad \left. \left. + \left(\sum_{l=1}^{n-1} f_l(z, w) \frac{\partial f_{n-l}(z, w)}{\partial z} \right) \exp(ins) \right) \right] \\ &= \left\{ \frac{\partial f_m(z, w)}{\partial w} - \nu \frac{\partial^2 f_m(z, w)}{\partial z^2} + \sum_{l=1}^{m-1} f_l(z, w) \frac{\partial f_{m-l}(z, w)}{\partial z} \right\} \\ &= \{0\}, \end{aligned} \quad (6.7)$$

where $\{0\}$ is the null sequence, $m \in \mathbb{N}$, the product $\hat{f}(z, w, s) \frac{\partial \hat{f}(z, w, s)}{\partial z}$ is written out using the Cauchy product (see section 6.2.3) and we have used that $\hat{f}(z, w, s)$ is a holomorphic function that is absolutely and uniformly convergent. As a result, we see that each sequence element $f_m(z, w)$ is a solution of an in-homogeneous linear diffusion equation.

In the next section we use the Green's function method to solve the sequence of linear diffusion equations.

6.4.2 Solving the sequence differential equation in the complex plane

In order to solve the series of in-homogenous linear differential equations (6.7) using a Green's function, we must first define two open paths in the complex plane along which we will integrate the solution. Let $\gamma_z(t) : [0, 1] \rightarrow \Omega_z$ and $\gamma_w(t) : [0, 1] \rightarrow \Omega_w$ be two continuous and piecewise differentiable open paths, where $\gamma_z(0) = z_a$, $\gamma_z(1) = z_b$, $\gamma_w(0) = w_a$ and $\gamma_w(1) = w_b$. We solve (6.7) by multiplying the differential equation with $G(z, w; z_0, w_0) : \Omega \times \Omega \rightarrow \mathbb{C}$, the Green's function, and integrate over the paths γ_w and γ_z to obtain

$$\int_{\gamma_w} \int_{\gamma_z} G(z, w; z_0, w_0) \left(\frac{\partial f_m(z_0, w_0)}{\partial w_0} - \nu \frac{\partial^2 f_m(z_0, w_0)}{\partial z_0^2} + \sum_{l=1}^{m-1} f_l(z_0, w_0) \frac{\partial f_{m-l}(z_0, w_0)}{\partial z_0} \right) dz_0 dw_0 = 0. \quad (6.8)$$

By using partial integration we can deduce that

$$\begin{aligned} & \int_{\gamma_w} \int_{\gamma_z} f_m(z_0, w_0) \left(-\frac{\partial G(z, w; z_0, w_0)}{\partial w_0} - \nu \frac{\partial^2 G(z, w; z_0, w_0)}{\partial z_0^2} \right) dz_0 dw_0 = \\ & - \int_{\gamma_w} \int_{\gamma_z} G(z, w; z_0, w_0) \left(\sum_{l=1}^{m-1} f_l(z_0, w_0) \frac{\partial f_{m-l}(z_0, w_0)}{\partial z_0} \right) dz_0 dw_0 + \\ & \nu \int_{\gamma_w} \left[G(z, w; z_0, w_0) \frac{\partial f_m(z_0, w_0)}{\partial z_0} - \frac{\partial G(z, w; z_0, w_0)}{\partial z_0} f_m(z_0, w_0) \right]_{z_0=z_a}^{z_0=z_b} dw_0 \\ & - \int_{\gamma_z} [G(z, w; z_0, w_0) f_m(z_0, w_0)]_{w_0=w_a}^{w_0=w_b} dz_0. \end{aligned} \quad (6.9)$$

If the Green's function $G(z, w; z_0, w_0)$ satisfies

$$-\frac{\partial G(z, w; z_0, w_0)}{\partial w_0} - \nu \frac{\partial^2 G(z, w; z_0, w_0)}{\partial z_0^2} = \delta(z - z_0) \delta(w - w_0), \quad (6.10)$$

where δ is the complex Dirac delta function, we can write rewrite (6.9) as

$$\begin{aligned} f_m(z, w) = & - \int_{\gamma_w} \int_{\gamma_z} G(z, w; z_0, w_0) \left(\sum_{l=1}^{m-1} f_l(z_0, w_0) \frac{\partial f_{m-l}(z_0, w_0)}{\partial z_0} \right) dz_0 dw_0 + \\ & \nu \int_{\gamma_w} \left[G(z, w; z_0, w_0) \frac{\partial f_m(z_0, w_0)}{\partial z_0} - \frac{\partial G(z, w; z_0, w_0)}{\partial z_0} f_m(z_0, w_0) \right]_{z_0=z_a}^{z_0=z_b} dw_0 \\ & - \int_{\gamma_z} [G(z, w; z_0, w_0) f_m(z_0, w_0)]_{w_0=w_a}^{w_0=w_b} dz_0, \end{aligned} \quad (6.11)$$

if and only if the paths $\gamma_w(t)$ and $\gamma_z(t)$ cross the Dirac delta function at $z = z_0$ and $w = w_0$. We note that when the integrand is holomorphic on Ω the exact paths $\gamma_w(t)$ and

$\gamma_z(t)$ do not play a role since any closed path in Ω integrates to zero for holomorphic integrands, see Morera's theorem [19].

In order to integrate all boundary and initial conditions only once in the general solution to the Burgers' equation $f(z, w)$, we set

$$\begin{aligned} f_1(z, w) &= \nu \int_{\gamma_w} \left[G(z, w; z_0, w_0) \frac{\partial f(z_0, w_0)}{\partial z_0} - \frac{\partial G(z, w; z_0, w_0)}{\partial z_0} f(z_0, w_0) \right]_{z_0=z_a}^{z_0=z_b} dw_0 \\ &\quad - \int_{\gamma_z} [G(z, w; z_0, w_0) f(z_0, w_0)]_{w_0=w_a}^{w_0=w_b} dz_0, \\ f_{m \geq 2}(z, w) &= - \int_{\gamma_w} \int_{\gamma_z} G(z, w; z_0, w_0) \left(\sum_{l=1}^{m-1} f_l(z_0, w_0) \frac{\partial f_{m-l}(z_0, w_0)}{\partial z_0} \right) dz_0 dw_0. \end{aligned} \quad (6.12)$$

We note that this choice is not unique and other ways are possible to integrate the boundary and initial conditions in the solution which is outside the scope of this work. The general solution of the Burgers' equation (6.6) for any initial and boundary condition can now be written as a recursive integral equation

$$\begin{aligned} f(z, w) &= \sum_{m=1}^{\infty} f_m(z, w) = \\ &\quad - \sum_{m=2}^{\infty} \int_{\gamma_w} \int_{\gamma_z} G(z, w; z_0, w_0) \left(\sum_{l=1}^{m-1} f_l(z_0, w_0) \frac{\partial f_{m-l}(z_0, w_0)}{\partial z_0} \right) dz_0 dw_0 + \\ &\quad \nu \int_{\gamma_w} \left[G(z, w; z_0, w_0) \frac{\partial f(z_0, w_0)}{\partial z_0} - \frac{\partial G(z, w; z_0, w_0)}{\partial z_0} f(z_0, w_0) \right]_{z_0=z_a}^{z_0=z_b} dw_0 \\ &\quad - \int_{\gamma_z} [G(z, w; z_0, w_0) f(z_0, w_0)]_{w_0=w_a}^{w_0=w_b} dz_0. \end{aligned} \quad (6.13)$$

6.5 An initial value problem for the Burgers' equation

Consider the following initial value problem for the Burgers' equation

$$\begin{aligned} \frac{\partial f(z, w)}{\partial w} - \nu \frac{\partial^2 f(z, w)}{\partial z^2} + f(z, w) \frac{\partial f(z, w)}{\partial z} &= 0, \\ f(z, 0) &= \exp(iz) = \cos(z) + i \sin(z) \end{aligned} \quad (6.14)$$

where $\nu \in \mathbb{R} > 0$, $\Omega_w = \{\Re(w) \geq 0, w \in \mathbb{C}\}$ and $\Omega_z = \{\Im(z) \geq 0, z \in \mathbb{C}\}$. To solve the differential equation, we start in §6.5.1 with the ansatz that the solution is a series that can be solved using the sequence transformation method. Later in §6.5.2 we proof that the ansatz is correct and we show that the series indeed converges absolutely and uniform in a domain Ω . Finally in §6.5.3 we give the absolute error of the series solution and compare the solution convergence properties to a numerical solution using the Cole-Hopf transformation.

6.5.1 Solving the initial value problem

We assume that the solution to the initial value problem (6.14) is a series $f(z, w) \in \mathcal{F}(\Omega)$ of the sequence $\{f_n(z, w)\} \in \mathcal{F}_s(\Omega)$. Following (6.12), the solution for the sequence $\{f_m(z, w)\}$ is

$$f_1(z, w) = \int_{\gamma_z} G(z, w; z_0, w_0) \exp(iz_0) dz_0,$$

$$f_{m \geq 2}(z, w) = - \int_{\gamma_w} \int_{\gamma_z} G(z, w; z_0, w_0) \left(\sum_{l=1}^{m-1} f_l(z_0, w_0) \frac{\partial f_{m-l}(z_0, w_0)}{\partial z_0} \right) dz_0 dw_0,$$

where $G(z, w; z_0, w_0)$ is the free-space Green's function

$$G(z, w; z_0, w_0) = \frac{1}{\sqrt{4\pi v(w - w_0)}} \exp\left(\frac{-(z - z_0)^2}{4v(w - w_0)}\right) H(w - w_0), \quad (6.15)$$

i is the imaginary unit, $H(w - w_0)$ is the Heaviside theta function, the path $\gamma_w(t)$ connects $w_0 = 0$ to $w_0 = w^+$ and the path $\gamma_z(t)$ connects $z_0 = -\infty$ to $z_0 = \infty$ [20]. We note that the integrand is holomorphic on the domain of interest and thus the exact paths do not matter as long they cross the Dirac delta function at $z = z_0$ and $w = w_0$. The first three calculated sequence elements of $\{f_m(z, w)\}$ are

$$\begin{cases} f_1(z, w) &= \exp(iz - wv), \\ f_2(z, w) &= \frac{i \exp(-4vw + 2iz) (1 - \exp(2vw))}{2v}, \\ f_3(z, w) &= -\frac{\exp(-9vw + 3iz) (1 - 3 \exp(4vw) + 2 \exp(6vw))}{8v^2}, \\ \dots &= \dots \end{cases}$$

where we have omitted the Heaviside theta functions since $w \geq 0^+$. The sequence can be captured in a closed form as

$$f_m(z, w) = \frac{i^{m-1} \exp(-vm^2w + imz)}{2^{m-1} v^{m-1} (m-1)!} \times \sum_{k=1}^m (-1)^{k-1} (k-1)! B_{m,k}(\mu_1(m, w), \dots, \mu_{m-k+1}(m, w)), \quad (6.16)$$

where $B_{m,k}(\mu_1(m, w), \dots, \mu_{m-k+1}(m, w))$ are the exponential Bell polynomials and

$$\mu_l(m, w) := \exp(lvw(m-l)).$$

Following (6.13), the full solution to our initial value problem is then given by

$$f(z, w) = \sum_{m=1}^{\infty} f_m(z, w), \quad (6.17)$$

Figure 6.1: A plot of the complex-valued solution $f(z, w)$, given by (6.17), for $2\pi < z < 2\pi$ at three different instances $w = 0$, $w = 1$ and $w = 4$ with $\nu = 0.3$. The imaginary part is plotted on the left and the real part is plotted on the right. The series solution has been cut-off at $m = 30$.

where $(z, w) \in \Omega$. In figure 6.1 we show the solution $f(z, w)$ for $2\pi < z < 2\pi$ at different instances with $\nu = 0.3$. The figure shows a competition between non-linear convection and linear diffusion in the velocity field. At $w = 1$ we see the development of a shock wave in both the real and imaginary part of the solution which is dissipated by viscosity on later times.

6.5.2 Proof of convergence

In order to show that our ansatz was correct, we need to proof that (6.17) converges absolutely and uniform in Ω . Instead, here we show that the solution converges on the real line and leave the general proof in the complex plane for future work. We introduce $u(x, t) : \Delta_x \times \Delta_t \rightarrow \mathbb{C}$ defined as $u(x, t) := f(x, t)$, where $\Delta_x = \{x \in \mathbb{R}\}$ and $\Delta_t = \{t \geq 0, t \in \mathbb{R}\}$. We start by

$$\begin{aligned}
 |u_m(x, t)| &\leq \frac{\exp(-\nu m^2 t)}{2^{m-1} \nu^{m-1} (m-1)!} \left| \sum_{k=1}^m (-1)^{k-1} (k-1)! B_{m,k}(\mu_1(m, t), \dots, \mu_{m-k+1}(m, t)) \right|, \\
 &\leq \frac{\exp(-\nu m^2 t)}{2^{m-1} \nu^{m-1} (m-1)!} \sum_{k=1}^m \left| (-1)^{k-1} (k-1)! B_{m,k}(\mu_1(m, t), \dots, \mu_{m-k+1}(m, t)) \right|, \\
 &= \frac{\exp(-\nu m^2 t)}{2^{m-1} \nu^{m-1} (m-1)!} \sum_{k=1}^m (k-1)! B_{m,k}(\mu_1(m, t), \dots, \mu_{m-k+1}(m, t)), \\
 &\leq \frac{\exp(-\nu m^2 t) \exp(\nu(m-1)mt)}{2^{m-1} \nu^{m-1} (m-1)!} \sum_{k=1}^m (k-1)! B_{m,k}(1, 1, \dots, 1), \\
 &= \frac{\exp(-\nu mt)}{2^{m-1} \nu^{m-1} (m-1)!} \sum_{k=1}^m (k-1)! \left\{ \begin{matrix} m \\ k \end{matrix} \right\}, \tag{6.18}
 \end{aligned}$$

Figure 6.2: A log plot of the absolute error between a high-precision numerical solution (30-digits accurate) of (6.22) and the partial series solution (6.21) as function of N for different viscosities in $\Lambda = [-2\pi, 2\pi] \times [0, 3]$, see (6.20).

where $\{^m_k\}$ are the Stirling numbers of the second kind. Now by the ratio test we have that

$$\begin{aligned}
 \lim_{m \rightarrow \infty} \frac{|u_{m+1}(x, t)|}{|u_m(x, t)|} &= \lim_{m \rightarrow \infty} \left(\frac{\frac{\exp(-v(m+1)t)}{2^m v^m (m)!} \sum_{k=1}^{m+1} (k-1)! \{^m_k\}}{\frac{\exp(-vmt)}{2^{m-1} v^{m-1} (m-1)!} \sum_{k=1}^m (k-1)! \{^m_k\}} \right) \\
 &= \lim_{m \rightarrow \infty} \left(\frac{\exp(-vt)}{2vm} \frac{\sum_{k=1}^{m+1} (k-1)! \{^m_k\}}{\sum_{k=1}^m (k-1)! \{^m_k\}} \right), \\
 &= \frac{\exp(-vt)}{2v} \lim_{m \rightarrow \infty} \left(\frac{1}{m} \frac{\sum_{k=1}^{m+1} (k-1)! \{^m_k\}}{\sum_{k=1}^m (k-1)! \{^m_k\}} \right), \\
 &= \frac{r \exp(-vt)}{2v}, \\
 &< 1,
 \end{aligned} \tag{6.19}$$

where $r = \lim_{m \rightarrow \infty} \left(\frac{1}{m} \frac{\sum_{k=1}^{m+1} (k-1)! \{^m_k\}}{\sum_{k=1}^m (k-1)! \{^m_k\}} \right)$ and can be calculated numerically as $r \approx 1.4427$. The convergence condition for the Weierstrass M-test is fulfilled, see section 6.2.1, when $v > r/2$ for all $t \geq 0$. We note that we do not have a more narrow upper bound for the Bell polynomials in (6.18), therefore the series may in fact be convergent for values $v \leq r/2$ as figure 6.1 shows.

Figure 6.3: A log plot of the absolute error between a high-precision numerical solution (30-digits accurate) of (6.22) and the partial series solution (6.21) as function of the viscosity ν for different N in $\Lambda = [-2\pi, 2\pi] \times [0, 3]$, see (6.20).

6.5.3 The absolute error

The series solution (6.17) of the initial value problem (6.14) converges absolutely and uniform in $\Delta = (-\infty, \infty) \times [0, \infty)$ for $\nu > r/2$. This means that we can define an absolute error as

$$\delta U = |U(x, t) - U_N(x, t)|, \quad (6.20)$$

where $(x, t) = \sup_{(x, t) \in \Lambda} |U(x, t)|$, $\Lambda \subseteq \Delta$, $U(x, t)$ is the exact solution of the initial value problem and $U_N(x, t)$ is the partial series solution given by

$$U_N(x, t) = \sum_{m=1}^N u_m(x, t), \quad (6.21)$$

$N \in \mathbb{N}$. The absolute error (6.20) goes to zero as $N \rightarrow \infty$ and can be used to show how fast the partial series solution converges to the exact solution in Λ for increasing values of N .

The exact solution with infinite precision is not known, but we can use the absolute error (6.20) to show how fast the partial series solution (6.21) converges to the numerical solution of the initial value problem (6.14) given by the Cole-Hopf transformation [7]

$$U(x, t) = \frac{\int_{-\infty}^{\infty} \frac{(x-x_0)}{t} \exp\left(\frac{-(x-x_0)^2}{4\nu t} - \frac{1}{2\nu} \int_0^{x_0} \exp(ix') dx'\right) dx_0}{\int_{-\infty}^{\infty} \exp\left(\frac{-(x-x_0)^2}{4\nu t} - \frac{1}{2\nu} \int_0^{x_0} \exp(ix') dx'\right) dx_0}. \quad (6.22)$$

This integral does not have a known closed-form expression, but we can numerically estimate this integral with high-precision (30 digits) using Mathematica [21].

Figure 6.2 shows a log plot of the absolute error between a high-precision numerical solution of (6.22) and the partial series solution (6.21) as function of N for different viscosities in $\Lambda = [-2\pi, 2\pi] \times [0, 3]$. The figure shows that the absolute error decreases exponentially with increasing N . Interestingly, the figure also shows that the rate of convergence decreases for decreasing ν . From a physical point of view we expect this to happen, since in the limit $\nu \rightarrow 0$ the Burgers' equation has a finite time singularity where the solution blows up. Figure 6.3 shows a log plot of the absolute error as function of the viscosity for different N . This figure shows that indeed the viscosity plays an important factor in the convergence of the series solution. A singularity occurs at $\nu \approx 0.239$ for all plotted values N where absolute error starts to grow exponentially. In this case the solution has a finite blow-up time where velocity gradient becomes infinitely steep at some point in $\Lambda = [-2\pi, 2\pi] \times [0, 3]$. Furthermore, the numerical estimation of (6.22) also starts to produce convergence errors around this value for the viscosity. We discussed in §6.5.2 that we need a more narrow upper bound for the Bell polynomials in (6.18) in order to proof that the series is indeed convergent for values $\nu \leq r/2$, which is outside the scope of this work.

6.6 Discussion & Conclusion

In this work we introduced a novel semi-analytic method to solve the Burgers' equation. We have shown that for any holomorphic and bounded series solution, the Burgers' equation can be decomposed into a sequence of linear diffusion equations by means of a sequence transformation. The solution for each individual sequence element can be written as a recursive integral equation using the Green's function method. In some cases, this recursive integral can be explicitly solved and a closed-form expression for the sequence can be obtained.

Using the sequence transformation method, we solved an initial value problem of the Burgers' equation in the complex plane. We showed that, for a complex exponential function as initial condition, the solution can be written as an infinite series using the Bell polynomials. The series solution is analytically complete, bounded and converges absolutely when the viscosity is larger than a threshold value. Furthermore we showed that the series solution converges exponentially to a high-precision numerical solution using the Cole-Hopf transformation for specific values of the viscosity.

In many physical systems one expects the solution of a non-linear differential equation to be analytic or at-least piece-wise analytic in the domain of interest. Therefore, we anticipate the sequence transformation method to be applicable far beyond the Burgers' equation. It would be very interesting to generalize the sequence transformation method to a broader set of non-linear differential equations that admit an-

alytic solutions, e.g. wave equations such as the Sine Gordon equation. Furthermore, it would also be interesting to investigate the possibility of capturing the initial conditions and boundary conditions in a set of basis functions that form an orthonormal set with respect to the domain of interest. This way, the recursive integrals that integrate over this domain simply vanish or can be drastically simplified. Therefore, the general solution could possibly be written without the need of any integrals. This direction could open the doors to more efficient numerical solvers for non-linear differential equations without the need of any spatial or time discretization, which we leave for future work.

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7

Summary & Conclusion

Extreme ultraviolet lithography (EUVL) is the next generation of photolithography technology used by the semiconductor industry for the manufacturing of integrated circuits. In this machine, a Laser-Produced Plasma (LPP) Extreme Ultraviolet (EUV) light source produces light with a wavelength of 13.5nm that is used to create feature sizes of sub-10nm on silicon wafers. One of the major technological challenges that remains unsolved today is the consistent power scaling of the EUV light beyond 250W. This must be solved if the industry wishes to maintain predictions made by Gordon E. Moore stating that the number of transistors on a dense integrated circuit doubles about every two years and results in cheaper, faster and more energy efficient integrated circuits. Fundamental understanding of the fluid and plasma processes inside the LPP-EUV light source is therefore crucial to optimize and stabilize the EUV power output. In this thesis we studied, both numerically and analytically, two types of droplet impact phenomena with applications in the LPP-EUV light source: the deformation dynamics of a droplet after laser impact and the splashing and cavity dynamics after oblique droplet impact onto a deep liquid pool.

In the first part of this thesis we focused on the laser impact onto a falling droplet. In the LPP-EUV light source a tin droplet passes the focus of a CO₂ laser beam where it is hit in two stages by a pre-pulse and main-pulse to create a tin-plasma that radiates the required EUV light. The pre-pulse laser impact results in a rapid phase change of a thin superficial layer on the illuminated side of the droplet. The resulting mass-ablation generates a recoil-pressure on the surface of the droplet and a subsequent pressure wave inside the droplet that leads to its deformation into a thin flat disk. This disk is an optimal target for the second high-intensity main-pulse that efficiently

converts the tin disk into a plasma.

Fundamental understanding of the droplet deformation dynamics after the pre-pulse laser impact may lead to a better coupling with the secondary high-intensity main-pulse impact that generates the EUV light. To this end, in Chapter 2 we presented an axisymmetric multiphase lattice Boltzmann model to study the propagation of pressure waves inside the droplet directly after the pre-pulse laser impact. The numerical method is capable of solving liquid-gas density ratios up to 10^3 , making the method directly applicable in the industrial and experimental regime. We validated this numerical method by showing that a stationary droplet obeys the Young-Laplace law, the second oscillation mode of a perturbed droplet correctly matches the analytical solution and mass conservation of a propagating pressure wave is properly conserved. We used the numerical method to support and validate the analytical findings in Chapter 3.

In Chapter 3 we presented an acoustic analytic model for the pressure, pressure impulse and velocity fields inside the droplet after laser impact in the limit of small pressure fluctuations. With this model at hand, we studied the onset of the droplet deformation as function of the recoil-pressure duration and amplitude at constant momentum transfer. When the momentum transfer to the droplet is increased while the pulse duration remains short compared to the timescale on which pressure-waves propagate through the droplet, the flow inside the droplet becomes compressible and the deformation strongly differs from a long recoil-pressure duration for which the flow can be considered incompressible. In this compressible flow regime the droplet deformation into a flat thin disk could be sped up, which may therefore increase the conversion efficiency of the LPP-EUV light source. Our proposed linear analytic model breaks down in the regime of high-intensity femtosecond laser impact. Here, the flow inside the droplet is governed by shock-waves, cavitation phenomena, non-linear viscous dampening and rapid interface acceleration. We argued however, that our analytic model can still be used as a starting point to identify likely cavitation spots inside the droplet, since shock waves need to develop in time. The numerical model developed in Chapter 2 could then be used to extend upon these analytical results. It would be particularly interesting to employ both the analytical and numerical model to study tightly focused high-intensity laser impacts and contribute to a more thorough understanding of the fragmentation phenomena that occurs after a cavitation event inside the droplet, which we leave for future work.

Both the numerical and analytical method presented in Chapter 2 & 3 give fundamental insight in the early deformation processes of the tin droplet after laser impact and could therefore help to improve the operating regime of the LPP-EUV light source. A possible way to increase the power output of the LPP-EUV light source could for example be to increase the throughput in number of droplets hit by the laser and converted into a plasma in a given time period. Consequently, the process of droplet deformation into a flat disk by the pre-pulse impact must be a significantly

sped up, for example by the use of more violent and/or shorter laser pulses. In this case, the presented numerical and analytical methods provide fundamental insight in the compressible flow regimes and early deformation dynamics as a function of the laser impact parameters. A more detailed understanding the droplet deformation in the highly compressible regime requires further numerical modelling and is left for future work.

A completely different approach to improve the LPP-EUV power output is to reduce the laser-to-droplet alignment sensitivity. When a laser beam impacts a droplet off-center, i.e. the laser impact has a relative misalignment with the center-line of the droplet, the droplet deforms under an angle and the maximum radial expansion of the droplet is decreased. In LPP-EUV light sources laser-to-droplet alignment plays an important role, since a slight misalignment leads to a sub-optimal target shape. In turn, this new target shape leads to an inefficient coupling with the second high-intensity main pulse that generates the EUV light. The deformation dynamics for this case have been addressed in Chapter 4. We derived and experimentally validated a model for the tilt angle sensitivity, expansion and propulsion velocity of a droplet irradiated by a $1\text{ }\mu\text{m}$ Nd:YAG laser pulse as function of its relative alignment. The proposed model has only a single dimensionless parameter: the ratio of the laser spot size to an effective size of the droplet, which is related to the position of the plasma critical density surface. The tilt angle in this model is based on the direction of the center-of-mass velocity of the droplet after impact. It was shown that the dimensionless tilt angle sensitivity is minimized when the laser spot size is large compared to the effective size of the droplet. We experimentally verified the model under three different experimental settings and we observed excellent agreement over a broad range of laser pulse energies and two laser focus spot sizes.

Current LPP-EUV light sources do not use a Nd:YAG pre-pulse laser but a CO_2 pre-pulse laser instead. With the model for the tilt-angle sensitivity at hand, we calculated the theoretical tilt angle sensitivity of a CO_2 pre-pulse onto a tin droplet. The tilt angle sensitivity in this case is about 85% larger than the corresponding tilt angle sensitivity of Nd:YAG laser impact. The extrapolation of our model to other laser wavelengths requires further experimental validation and is left for future work. However, the model provides a clear indication that a LPP-EUV light source using a Nd:YAG pre-pulse laser may be less sensitive to a laser-to-droplet misalignment in comparison to a CO_2 pre-pulse.

In the second part of this thesis we focused on techniques to suppress tin contamination inside the LPP-EUV light source. The tin droplets that are missed by the laser or flying tin debris after laser impact are captured by a tin catching device and dedicated vanes in the source chamber to prevent contamination of the sensitive optics. The primary tin catcher is a container filled with liquid tin located directly below the stream of falling droplets. The depth of the tin inside the container or the depth of the tin coating on the vanes inside the source chamber is much larger than the size

of a single impacting tin droplet. The impact of a tin droplet onto these catching devices typically results in a splash and create smaller debris that is more difficult to be captured. Therefore, fundamental understanding in droplet splashing upon impact is key to a better design of the tin catching device or the vanes inside the LPP-EUV source chamber and the protection of the sensitive optics inside the light source. To this end, we studied the splashing and cavity dynamics after oblique droplet impact onto deep liquid pools.

Oblique droplet impact onto a deep pool leads to two different splashing regimes: an omni-directional or a single-sided crown splash. In Chapter 5 oblique droplet impact on a deep liquid pool was studied numerically as function of the impact angle and the Weber number. We used the adaptive Volume-Of-Fluid (VOF) flow solver Basilisk to study the ejecta sheet formation, splashing threshold, cavity formation, cavity evolution and the maximum cavity dimensions. The numerical results were compared to recent experimental work by Gielen et al. We found excellent qualitative and quantitative agreement between the numerical simulations and experimental results on both the ejecta sheet dynamics and the cavity dynamics for a broad range of Weber numbers and impact angles. We were the first to provide a detailed numerical overview of the ejecta sheet dynamics that includes the velocity field and streamlines until the moment of ejecta sheet breakup as function of the impact angle. We showed that the ejecta sheet and cavity dynamics for impact angles close to perpendicular resemble the dynamics of a perpendicular impact event. However when the impact angle is increased the dynamics start to differ strongly. In this case, the droplet partly slides along the pool surface during impact. The ejecta sheet stretches out parallel to the pool surface instead of perpendicular and eventually falls down on the surface where it entraps multiple air bubbles. The cavity below the pool surface becomes stretched in the direction parallel to the pool, leading to a cone-shaped cavity. The numerical simulations provide a valuable addition to the experimental findings, since they capture a fully 3D view of the impact phenomena and allows for a better judgment on the splashing dynamics of the ejecta sheet. Furthermore, the presented method can be used to explore impact parameters that are hard to reach experimentally, e.g. very large impact angles or high droplet velocities. The simulations provide a toolkit to explore different LPP-EUV source chamber designs more efficiently and may therefore lead to a better design of the tin catching devices inside the source chamber.

Finally, in Chapter 6 we introduced a novel semi-analytical solving method for bounded and analytic series solutions of the Burgers' equation. We demonstrated that a sequence transformation can split the non-linear Burgers' equation into a sequence of linear diffusion equations. Each sequence element can be solved recursively by the use of the Green's function method. The general solution to the Burgers' equation can therefore be written as a recursive integral equation for all initial value problems. We showed that for a complex exponential function in an infinite domain the closed-form

series solution can be written in terms of the Bell polynomials. The solving technique is applicable far beyond the Burgers equation. It would be interesting to generalize the sequence transformation method to a broader set of non-linear differential equations that admit analytic solutions. As such, the novel method could potentially be used to get a deeper understanding of the compressible and non-linear flow regimes inside the droplet after laser impact or after oblique droplet impact onto a deep liquid pool, which we leave for future work.

Samenvatting

Laser-Produced Plasma (LPP) Extreme Ultraviolet (EUV) lichtbronnen spelen een sleutelrol bij de productie van EUV-licht dat wordt gebruikt in de nieuwste generatie lithografie machines. In deze lichtbron vallen tindruppels in het brandpunt van een CO₂ laserstraal, waarna de druppel in twee stappen een vervorming en fase verandering ondergaat om het EUV-licht te genereren. Na de productie van het EUV-licht worden de tinresten opgevangen door een druppelvanger en door verschillende vinnen in de bronkamer om de verontreiniging van de gevoelige optica te voorkomen. Een van de grootste onopgeloste technologische uitdagingen is de stabiele productie van meer dan 250W EUV licht. Fundamenteel onderzoek in de vloeistof- en plasmaprocessen binnen de LPP-EUV lichtbron is cruciaal om deze vermogens in de toekomst te kunnen behalen. In dit proefschrift hebben we zowel numeriek als analytisch naar twee soorten druppelinslag verschijnselen gekeken die een directe toepassing hebben in de lichtbron: de vervormingsdynamiek van een druppel na laserinslag en de uiteenspatting- en holtedynamiek van een druppel na schuine inslag op een diep vloeistofbad.

In het eerste deel van dit proefschrift richtten we ons op de laserinslag op een vallende druppel. De laserinslag op een vallende druppel resulteert in een snelle faseverandering van een klein laagje vloeistof aan de oppervlakte van de druppel. De resulterende massa-ablatie genereert een terugslagdruk op het oppervlak en een lopende drukgolf binnen de druppel die leidt tot een vervormings- en fragmentatieproces. In hoofdstuk 2 werd een assymmetrische Boltzmann-model gepresenteerd dat de voortplanting van drukgolven en de resulterende oppervlakte vervorming numeriek kan beschrijven. Deze methode werd gebruikt om de analytische uitdrukkingen van hoofdstuk 3 te valideren en ondersteunen.

In hoofdstuk 3 presenteerden we een akoestisch analytisch model voor de druk-, drukimpuls- en snelheidsvelden in de druppel na laserinslag in de limiet van kleine drukfluctuaties. Met dit model hebben we de druppelvervorming analytisch bestudeerd als functie van de duur en amplitude van de terugslagdruk bij constante impulsoverdracht. Wanneer de impulsoverdracht naar de druppel wordt verhoogd terwijl de duur van de terugslagdruk kort blijft, wordt de vloeistofstroming in de druppel samendrukbaar en is de vervorming anders dan wanneer de druppel was ingeslagen met een lange terugslagdruk. In dit samendrukbaar regime kan de druppel sneller vervormen

wat kan leiden tot een efficiëntere conversie van de tin druppel naar het vereiste EUV licht. Het gepresenteerde model is niet meer toepasbaar in het regime van een hoge intensiteit en femtoseconde laserinslag. In dit regime wordt de stroming in de druppel namelijk beheerst door niet-lineaire schokgolven, cavitatieverschijnselen, niet-lineaire viskeuze demping. De methode kan echter wel worden gebruikt als startpunt om mogelijke cavitatieplekken in de druppel aan te wijzen. Dit zou kunnen leiden tot een beter begrip van de fragmentatieverschijnselen die optreden wanneer de druppel wordt geraakt door een sterk gefocuste laserpuls met hoge intensiteit. Fundamenteel inzicht in de vroege vervormingsdynamiek en de fragmentatiedynamiek van de druppel zorgt ervoor dat de conversie van het CO₂ laserlicht naar het vereiste EUV-licht beter begrepen wordt.

Wanneer een laserstraal een druppel uit het midden raakt vervormt de druppel onder een hoek en neemt de maximale radiale uitzetting van de druppel af. Bij LPP-EUV lichtbronnen speelt deze uitlijning een belangrijke rol, omdat een kleine fout kan leiden tot een inefficiënte koppeling met de tweede hoge intensiteitspuls die het EUV-licht produceert. In hoofdstuk 4 bestudeerden we daarom theoretisch en experimenteel de gevolgen van een verkeerde uitlijning. We presenteerden een model voor de kantelgevoeligheid, expansie en voortstuwingssnelheid van een druppel die wordt geraakt door een 1 μ m Nd:YAG-laserpuls. Er werd aangetoond dat de dimensieloze kantelgevoeligheid minimaal is wanneer de laserbundel groot is in verhouding tot de positie van het plasma kritische oppervlak van de druppel.

In het tweede deel van dit proefschrift hebben we ons gericht op de uiteenspatting- en holtedynamiek na schuine druppelinslag op een diep vloeistofbad. De schuine druppelinslag leidt tot twee verschillende gedragingen: een uiteenspatting in alle richtingen of een uiteenspatting met de richting van de inslag mee. Fundamenteel inzicht in dit gedrag zou kunnen leiden tot een beter ontwerp van de druppelvanger of de vinnen in de LPP-EUV bronkamer. Daarmee kan de secundaire verontreiniging van de gevoelige optica binnen de lichtbron worden onderdrukt.

In hoofdstuk 5 werd de schuine druppelinslag op een diep vloeistofbad numeriek bestudeerd als functie van de inslaghoek en het Weber getal. We hebben de adaptieve Volume-Of-Fluid (VOF) vloeistofoplosser Basilisk gebruikt om de uiteenspatting, de holtevorming, de holteevolute en de maximale holteafmetingen van de druppelinslag te bestuderen. Hierbij zijn wij de eersten die een gedetailleerd numeriek overzicht gaven van de uiteenspattingsdynamiek onder een hoek. We hebben aangetoond dat de uiteenspatting- en de holtedynamiek voor inslaghoeken in de buurt van loodrecht erg veel lijken op de dynamiek van een loodrechte druppel inslag. Echter, wanneer de inslaghoek wordt verhoogd begint de dynamiek sterk te verschillen. In dit geval glijdt de druppel gedeeltelijk langs het badoppervlak tijdens de inslag. Na inslag strekt een vloeistofuitstulping zich evenwijdig aan het oppervlak uit in plaats van loodrecht en valt uiteindelijk terug naar het oppervlak waar het meerdere luchtbellensluit. De holte breidt zich parallel aan het bad uit, wat leidt tot een kegelvormige holte.

Deze numerieke simulaties bieden een waardevolle aanvulling op de experimentele bevindingen, omdat we hiermee de inslag nog beter kunnen begrijpen en bestuderen.

Ten slotte introduceerden we in hoofdstuk 6 een nieuwe semi-analytische oplossingsmethode voor analytische reeksen van de Burgers-vergelijking. We hebben aangetoond dat een rijtransformatie de niet-lineaire Burgers-vergelijking kan opsplitsen in een verzameling van lineaire diffusievergelijkingen. Elke lineaire vergelijking kan recursief worden opgelost met behulp van de functiemethode van Green. De algemene oplossing voor de Burgers vergelijking kan daardoor worden geschreven als een recursieve integraalvergelijking voor alle initiële randvoorwaarden. We toonden aan dat voor een complexe exponentiële functie als initiële conditie een oneindig domein de reeksoplossing kan worden geschreven in termen van de Bell-polynomen. De oplossingsmethode is ook toepasbaar voor andere niet-lineaire differentiaal-vergelijkingen. We verwachten daarom dat deze methode ook kan worden toegepast voor het beter begrijpen van de niet-lineaire vloeistofstromingen in de druppel na laserinslag of na schuine druppelinslag op een diep vloeistof bad.

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