Droplets

Drag, Coalescence and Impact

Pascal Sleutel

DROPLETS: DRAG, COALESCENCE AND IMPACT

Pascal Sleutel

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1

Introduction

Seldom a water surface observed in nature is completely flat. On a windless day, a pond acts like a mirror to the sky above and the surroundings; when this occurs we tend to quickly take a picture with a smartphone to capture its beauty. A small breeze or a fish attacking its prey dramatically changes this situation, causing gravity and surface tension to counteract this disturbance [1–3]. This action results in the formation of waves travelling over the water surface where we can understand that the large scale (typically on the size of a meter) results in waves dominated by gravity and the small scale (typically on the size of a millimeter) by surface tension.

Figure 1.1: Waterstrider or "Schaatsenrijder" in dutch. In French, this animal is know as "Gerris" inspiring the name of the numerical simulation package for solving fluid flow [4]. Image from: [5]

Gravity is known as one of the four fundamental forces in nature [6]. Surface tension derives its action from cohesive forces among liquid molecules. More specifically, surface tension depends on the difference in electrical attraction between molecules of two intersecting liquids or surfaces. An example of the working of surface tension can be observed in Fig. 1.1 where a water strider uses the surface tension of water to stay afloat. The legs are water repellent (hydrophobic) and the mass of the insect is small enough so that the water can support it. The water surface behaves as an elastic membrane, supporting the strider [7]. The unit of surface tension is force per unit length or energy per unit area, the associated length is called the radius of curvature, visible as the indentations of the water surface under the leg of the water strider. Surface tension acts on every curved liquid surface and minimizes the amount of surface energy; the result is a force that keeps the water strider afloat or a rain drop (nearly) spherical [8].

It is remarkable that molecular attraction on the length scale of a molecule (10^{-10} m) [9] can play a role on the size of rain or a waterstrider (length scale 10^{-2} m). This linkage of scales is something typical for physics in general and fluid dynamics in particular and it is partly where the huge predicting powers in many engineering problems of fluid dynamics is based on. By dimensional analysis, the physical parameters of a problem can be put in dimensionless ratios, making it independent of material properties, size or timescales. These ratios are called dimensionless numbers which play an important role in this thesis so they will be introduced immediately. Their names are mostly connected to the names of famous engineers and scientists.

1.1 Dimensionless Numbers

A widely used dimensionless number is the Reynolds number (Re) which is the ratio of inertial forces to viscous forces.

$$Re = \frac{\rho UR}{\mu} \quad (1.1)$$

where ρ is the liquid density, U is the typical velocity, R is the typical length scale and μ is the liquid viscosity. Typically it is associated with the transition from stable laminar flow to turbulent flow but its applicability is much wider. In this thesis the Weber number (We) which is important for jetting and impact will play an important role as well; it is the ratio between inertia and surface tension σ ,

$$We = \frac{\rho RU^2}{\sigma}. \quad (1.2)$$

The next dimensionless number is the Bond number (Bo)

$$Bo = \frac{\rho a R^2}{\sigma}, \quad (1.3)$$

where a is a local acceleration which usually refers to the gravitational acceleration. In that form, this number is also known as the Eötvös number [10]. The Bond number is a balance between acceleration and surface tension. It sets for example the transition from dripping to jetting [11]. Ohnesorge is a dimensionless number that balances viscosity with inertia and surface tension:

$$Oh = \frac{\mu}{\sqrt{\rho\sigma R}} = \frac{\sqrt{We}}{Re}. \quad (1.4)$$

The Ohnesorge number is used in determining for inkjet printing stability [12].

The similarity which is central in this thesis, is the one which connects liquid tin with water since they have an almost identical surface tension to density ratio, as can be observed in table 1.1, while at the same time the viscosities are almost the same as well. The particular relevance of liquid tin to this thesis will be explained in the following section.

Parameter	Unit	tin	water
T	Drop temperature (°C)	250	20
ρ_l	Liquid density (kg/m ³)	6968	1000
μ_l	Liquid dynamic viscosity (mPa·s)	1.85	1.0
σ	Surface tension (mN/m)	544	70
V	Impact velocity (m/s)	70	17
R	Drop radius (μm)	25	396
We	Weber number	1569	1569
Re _l	Reynolds number of the liquid	6591	6591

Table 1.1: Parameter values of tin droplets in the ASML EUV source. Values taken from [13], updated with recent data [14].

To do experiments with molten tin is difficult [15–17] since a normal working temperature is 250 degrees Celsius. Moreover it oxidizes as soon as it comes in contact with oxygen and tin oxides are ceramic materials with a melting temperature much higher than 250 degrees Celsius. All this makes experimenting with molten tin and the investigation of free surface flows challenging. To avoid oxidation, experiments must be carried out under a nitrogen or argon purge or in vacuum, resulting in a limited optical and mechanical access to the measurement site.

1.2 Industrial Partners and Societal Relevance

Nowadays, personal computers, smartphones, tablets and electronics are crucial elements in daily life. A car management system demands for extensive computing

power to control combustion, monitoring a large variety of sensors, planning of maintenance periods, route planning and the like. Today's hype is about smart devices, about the wireless connection of all kind of electric and electronic equipment, from washing machines, refrigerators, household robots and coffee machines to audio/video equipment and theft protection means. All these applications are enabled by semiconductor technology with an associated global semiconductor industry generating a revenue of approximately 325 billion dollars [18] in 2016.

In order to cope with the requirements regarding speed, memory use, and reliability, semiconductor companies try to produce integrated circuits (ICs) with as many features integrated as possible. Although such advanced ICs are more expensive, their increased functionality makes that the number of ICs and external (passive) components needed per apparatus is reduced and, in the end, the price of the application. The race to maximize functionality and profit in the semiconductor industry has resulted in a gigantic technological effort to decrease the average size of a transistor on an IC. The systematic decrease in transistor size or the increase in the amount of transistors per unit area is called Moores law [19] (after one of the founders of Intel, the largest semiconductor company) which is depicted in Fig. 1.2. Moore's law states that the number of transistors integrated per unit area doubles every two years.

Figure 1.2: a) The amount of transistors per integrated circuit. The blue dots refer to Intel products and the red squares to those of Motorola [20]. b) The exponential world of the semiconductor industry. The amount of transistors made grows and at the same time the price decreases exponentially [21].

The online material on the 50th birthday of Moore's law in 2015 and 30 years ASML in 2014 are highly recommended, including interviews with Gordon Moore himself. Two passages from the interview are quoted here, the first speaks for itself: "By making things smaller, everything get's better at the same time. The transistors get faster, the reliability goes up, the cost goes down A unique violation of Murphy's law.". The second quote represents the aspect that turned Moore's law into a self fulfilling prophecy, because the original paper only predicted exponential

growth until 1975: "Whatever has been done, can be outdone.". From a fit of data in the beginning of the sixties, Moore's law turned into an industry defining purpose being successful for more than 50 years.

Figure 1.3: Semiconductor production process, schematically shown. Lithography is done in the Exposure step, marked in red.

The semiconductor equipment industry represents around 10% in revenue of the total semiconductor industry. The leading company in the world for lithographic equipment is ASML, headquartered in the Netherlands. Lithography is a crucial step in the production of semiconductors, as the schematic view in Fig. 1.3 shows. With a beam of light, the designed pattern (mask) is projected onto a wafer in order to create the transistors later. In order to fulfil Moores law, the smallest sizes on the chip, also known as the critical dimension (CD) has to shrink continuously. The CD depends on properties of the optical system and can be calculated using the Rayleigh criterion:

$$CD = k_1 * \frac{\lambda}{NA} \quad (1.5)$$

where k_1 is a process dependent pre-factor with a theoretical maximum value of 0.25, λ is the wavelength of light used for projection and NA is the numerical aperture of the optics. Schematically, this is shown in Fig. 1.4 where $NA = n \sin(\theta_{max})$ expresses NA in terms of the index of refraction n and the maximum angle at which the system can transmit light θ_{max} . The technological goal of ASML is to decrease CD, which can be done by increasing NA or to use light with a smaller wavelength. Typically in ASML, every two years there is a step in NA and every decade a step in wavelength. The process of improvement in NA has been going on with 193 nm ArF DUV light but now a giant step in wavelength reduction is taken, to 13.5 nm or EUV light. This giant step in wavelength seems large and not in line with historical developments but actually it is really well argued. A crucial element in (photo)lithography is a material with sufficiently high optical transparency and refractive index which is used for lenses. No materials can be found that have the

required properties at wavelengths below 150 nm. Most promising candidates to be the next-generation lithography technology to replace the current technology have been: electron beam lithography, focused ion beam lithography, X-ray lithography, nanoimprint lithography and EUV [22, 23]. All candidates were in time subject to heavy fluctuations in popularity with throughput capability and costs of operation and implementation as main drivers. Currently, EUV is winning and big steps have been and will be made in order to improve on throughput and reliability [24].

Figure 1.4: Schematic diagram of the optics of a lithography system. The numerical aperture can be calculated from the index of refraction n and the maximum angle at which the system can transmit light θ_{max} , $NA = n \sin(\theta_{max})$

For creating light with 13.5 nm, only a few options are available [23, 25, 26]. One of the challenges with light of this wavelength is that it gets absorbed by almost all materials, so instead of transmissive optics, reflective optics need to be used. The reflective optics that are used for EUV are the so called Bragg-reflectors or dielectric mirrors. The source and optics system performance is measured by the amount of photons per unit time at the wafer level, because it defines the amount of wafers (and in the end IC's) a machine can produce per unit time. Calculating back the output performance on wafer level determines the power of the EUV source. This is where the tin becomes important, because a tin plasma appears to be the most effective way to produce EUV photons at the power level needed (200-1000 W). Other options would be synchrotron radiation or a free electron laser.

From the few options for creating EUV photons by means of a tin plasma, ASML has taken the route of making EUV by means of a laser-produced plasma (LPP), shown schematically in Fig. 1.5. In a large vacuum vessel, small high speed droplets of tin are made by means of a droplet generator; a thin high-speed jet is formed through a small sized nozzle. By modulating the velocity of the jet, it breaks up into small droplets that in course of their flight toward the focus of

Figure 1.5: A schematic drawing of an EUV source [14]. A stream of droplets, produced in the droplet generator, is protected from hydrogen streams by the shroud during coalescence and moves into the primary focus of the elliptical collector. This point coincides with the focal point of the CO₂ laser optics (laser not shown here). Most of the EUV radiation shines onto the collector and is then collected in the intermediate focus, which is a small hole between the source vessel and the projection box. Droplets not hit by the laser are collected in the tin catcher. The pre-pulse is not depicted here.

the collector coalesce to droplets of the specified size. With a laser pre-pulse the droplet is deformed into a pancake-like shape which is then hit by a 10 μm CO₂ IR laser pulse (0.7 Joule per pulse at 50 kHz) to convert the tin into tin plasma.

In the plasma, tin molecules lose up to 14 of its electrons, upon restoring its electron shield the tin atom radiates EUV. The EUV is generated in the primary focus of an elliptical collector (mirror). The collector reflects the light towards the secondary focus of the elliptical mirror. The secondary focus is a small hole that connects the vacuum vessel of the light source with the box containing the reflective optics, reticle stage and wafer stage. Apart from EUV, energy is radiated until deep in the infrared, comparable to the long tail of black body radiation. To protect the collector from high intensity radiation, high energy ions and free flying tin particles, hydrogen gas streams are maintained that direct the debris away from the collector, primarily as SnH₄. Small droplets are produced when the tin droplet is not fully evaporated [27–29]. The injected gas streams are guided away through big ducts towards vacuum pumps, such that no contamination enters the projection box. The output power of the source scales with the injection frequency of the droplets, typical working conditions are droplet size $\sim 25 \mu\text{m}$, droplet velocity 70 m/s, droplet frequency 50 kHz [14].

The development of an EUV-lithography machine, that is be able to keep up

with Moore's law, in a cost effective way takes an enormous research, development and manufacturing effort. The Dutch government, representing the citizens of the Netherlands is supporting companies willing to make this investment to be able to run a profitable and sustainable business and and to create high-valued jobs in the future. One of these initiatives is called NanoNextNL, the organisation that provided funding for this PhD research project. NanoNextNL is a consortium of more than one hundred companies, universities, knowledge institutes and university medical centres, which is aimed at research into micro and nanotechnology [30]. Other companies related to this project via NanonextNL are e.g. Océ Technologies which is in the business of industrial printing [31] and Medspray, a spray nozzle technology company [32]. All companies in the project have their interest in micro nozzles in common. The companies explicitly mentioned here work with micro-droplet generation, their interaction with the surrounding medium and splashing after impact.

1.3 Guide Through the Thesis

For this thesis, the tin droplets of ASML were inspiration for a more fundamental study into droplet coalescence and impact. The capillary waves on submillimeter scale as described before, are crucial in understanding both phenomena. In chapter 2, droplet formation and coalescence is studied with experiments and numerical simulations. The introduction of the chapter contains a historical account of the understanding of jet breakup and droplet formation which is therefore omitted here. The observed influences of air drag led to the work presented in chapter 3 where drag on a train of droplets and coalescence is studied experimentally. In chapter 4 we study drop impact, cavity formation and bubble entrainment for perpendicular droplet impact. This is continued in chapter 5 where the angle of impact is varied, to study splashing and cavity formation for oblique drop impact. In chapter 6, a droplet bouncing on an pool of the same liquid is evaluated as method for teaching quantum mechanics.

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2

Controlling jet breakup by a superposition of two Rayleigh-Plateau-unstable modes ^{*} [†]

We experimentally, numerically, and theoretically demonstrate a novel method of producing a stream of widely spaced high-velocity droplets by imposing a superposition of two Rayleigh-Plateau-unstable modes on a liquid jet. The wavelengths of the two modes are chosen close to the wavelength of the most unstable mode. The interference pattern of the two superimposed modes causes local asymmetries in the capillary tension. The velocity of the initial droplets depends on these local asymmetries. Due to their different velocities, the droplets coalesce to produce a stream of larger droplets spaced at a much larger distance than the initial droplets. We analytically derive the perturbations that robustly induce this process and investigate the influence of the non-linear interactions of the two Rayleigh-Plateau-unstable modes on the coalescence process. Experiments and numerical simulations demonstrate that the jet breakup and the subsequent droplet merging are fully governed by the selected modes.

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[†]The experimental work in this chapter is part of the present thesis. The numerical simulations are performed by Theo Driessen

2.1 Introduction

Droplets spawn and perish continuously. The splashing of rain or the generation of aerosols during the breaking of waves are examples of drop formation in nature. In industry, droplets serve the need for controlled delivery of material. Drop formation is used in the production of powders, where the liquid fraction of the droplets of a suspension is evaporated to obtain a powder [1]. Droplet generators are also used in the dispersion of respiratory medicines, where the droplet size determines where in the lungs the medicines are deposited [2]. In inkjet printing droplets are deposited for graphical purposes or as functional materials [3–5]. These three examples have in common that the underlying processes can be optimized when the droplet size and spacing are controlled. In this chapter we investigate a method to produce a continuous stream of droplets from a periodically perturbed continuous jet, where we control the size and spacing of the resulting droplets.

Drop formation has been studied for a long time. Savart [6] was the first to report that a continuous liquid jet breaks up into a stream of droplets. Later, Plateau [7] observed that a varicose perturbation of the jet grows if its wavelength is longer than the circumference of the jet. In the literature, a distinction is made between an infinite jet, and a jet that is ejected from a nozzle. The infinite jet with a periodic varicose perturbation is an academic case, used to study the stability of a liquid jet. A jet that is ejected from a nozzle is finite, and ages with the distance it has traveled from the nozzle. Growing perturbations on the ejected jet are not periodic in space. Rayleigh [8] derived the dispersion relation for an infinite jet with a periodic perturbation. Later, he extended the theory by adding viscous effects [9]. In his paper on the instability of a liquid jet ejected from a nozzle, Weber [10] reported a linear approximation for the theory by Rayleigh which has a solution that is almost identical to the original one. In the same paper Weber discussed the influence of air on a laminar jet. Chandrasekhar [11] examined the effect of finite viscosity on the Rayleigh-Plateau instability. Keller [12] continued the study of the spatial instability of a liquid jet. Keller derived the complex eigenvalues of the dispersion relation as a function of the Weber number of the jet. He found that when the jet is ejected at high Weber number, the spatial asymmetry of the perturbations that grow on the jet is small. This implies that the growing perturbations on a fast jet move along with the jet, as is the case for growing perturbations on an infinite jet with periodic perturbations. Hence, the spatial growth-rate of a Rayleigh-Plateau-unstable mode on a fast jet is related to the temporal growth-rate by the jet velocity. Mathematically speaking, this is reflected in the feature that the imaginary part of the wavenumber that describes the spatial instability is small in the case of a high Weber number.

Experiments have shown that the system indeed behaves according to the Rayleigh-Plateau instability analysis. When a periodic perturbation is applied at

the nozzle, the jet breaks up according to the theory. A Rayleigh-Plateau-unstable mode can be triggered by different means. Two common perturbation methods are a pressure perturbation inside the nozzle [13], and an undulating electric field around the liquid jet [14]. For the perturbation to be triggered by an electric field, the liquid needs to be conductive. With the pressure perturbation, only the velocity is perturbed. With the electric field on the other hand, only the radius of the jet is perturbed. A perturbation of either the velocity or the radius not immediately results in a single Rayleigh-Plateau-unstable mode, since a single Rayleigh-Plateau-unstable mode consists of a perturbation both in velocity and in radius. A Rayleigh-Plateau-unstable mode has both a growing and a decaying mode at the same wavenumber. When the velocity is directed away from the troughs in the varicose perturbation, the mode is growing. In the decaying mode, the velocity is directed towards the troughs. By simultaneously applying both the growing and decaying mode of the same Rayleigh-Plateau-unstable mode, the resulting jet can have a perturbation either in the radius, or in the velocity alone, depending on the phase difference between the two modes. Garcia [15] investigated how the different perturbation mechanisms affect the onset of the Rayleigh-Plateau instability. When the perturbation amplitude at the nozzle is small, the decaying mode has a negligible influence on the Rayleigh-Plateau instability.

When the perturbation amplitude approaches the jet radius, non-linear interactions in the perturbation cause satellite droplet formation [16, 17]. The relative size of the satellite droplets increases when the perturbation wavelength increases [13]; experiments show that below a dimensionless wavenumber $kR_0 \approx 0.2$ the volume contained in the satellite droplet is even larger than the main droplet. By adding higher harmonics to the actuation signal, the size and speed of the satellite droplet can be influenced [18, 19]. Another way to reduce or eliminate the satellite droplets is to induce coalescence between multiple main droplets. Orme [20] pioneered this field by using an amplitude modulated (AM) pressure perturbation at the nozzle [21, 22]. The spectrum of the AM signal consists of a carrier frequency and two sidebands. The sidebands are mirrored into the carrier frequency. The fastest growing Rayleigh-Plateau-unstable mode is chosen as the carrier frequency. The interference of the side bands causes the modulation. The modulation frequency determines the wavelength of the modulation envelope. The droplets in one perturbation envelope merge into one large droplet per modulation envelope.

In this chapter, we present an efficient and robust method to generate a periodic stream of droplets from a continuous jet, in such a way that we are able to precisely control the droplet size and inter-droplet distance. The novel feature of the method is the selection of the perturbation wavenumbers, such that a fully modulated beat signal can be applied at the nozzle, which maintains its shape during the growth of the two perturbations. The basic idea is introduced in the next section, and in the

Figure 2.1: The dispersion relation of the Rayleigh-Plateau instability for an infinite inviscid jet, which is valid for $We \gg 1$, and $Oh \ll 1$. The dimensionless wavenumbers χ_1 and χ_2 represent the different perturbation wavenumbers that we induce on the jet. The contours show their individual radius perturbations. The superposition of mode χ_1 and mode χ_2 is also shown, at the wavenumber that corresponds to the shortest common period of χ_1 and χ_2 . Since χ_1 and χ_2 grow equally fast, the shape of the superposition is conserved in the regime where the perturbation amplitude is small compared to the jet radius.

analysis section (section 2.3) the selection criteria are explained in more detail. Furthermore, section 2.3 contains an investigation of the influence of nonlinear interactions on the initial droplet formation and the final coalescence process. With the theoretical concept known, we then introduce our experimental setup (section 2.4). The technical details of the operation of the setup are given in Appendix 2.8. The numerical scheme is explained in section 2.5. By comparing the experimental results with the results from the numerical analysis, we show that the evolution of the jet into a stream of widely spaced droplets is fully determined by the perturbation signal composed of two sinusoidal perturbations (section 4.4). The chapter ends with conclusions and an outlook (section 2.7).

2.2 The Concept

Put in a nutshell, the idea is as follows: We simultaneously impose two sinusoidal perturbations of the pressure drop across the nozzle. These two perturbation modes are chosen such that they are both Rayleigh-Plateau-unstable modes, see figure 2.1, where $Oh = \mu / \sqrt{\rho R_0 \sigma}$ gives the ratio between the viscosity and the surface tension, in which μ is the dynamic viscosity, ρ the density, R_0 the unperturbed

jet radius, and σ the surface tension. We $= \rho u_0^2 R_0 / \sigma$ gives the ratio between inertial and capillary effects; here u_0 is the unperturbed jet velocity. Furthermore, $\chi = kR_0$ is the dimensionless wavenumber of the perturbation and ω is the growth-rate of the perturbation. The time scale of the growth of the perturbations is given by the capillary time $t_{cap} = \sqrt{\rho R_0^3 / \sigma}$. The interference of the two modes causes the droplets to merge into one droplet per shortest common period of the two perturbations as will be demonstrated in Fig. 2.2. By means of two small pressure perturbations at the nozzle that trigger the two separate Rayleigh-Plateau instabilities [15], we thus control the droplet size and the spacing between the droplets.

The breakup and coalescence resulting from the superposition of the two Rayleigh-Plateau-unstable modes is shown in Fig. 2.2. This figure shows the breakup and coalescence for three differently chosen combinations of perturbations. To demonstrate the basic idea, we show the numerical results for the perturbed infinite jet. This allows us to use periodic boundary conditions, with the same periodicity as the perturbation. The periodic boundary conditions are set at the anti-nodes of the beat signal. Note the satellite droplets that form at the anti-nodes.

The superposition of the two Rayleigh-Plateau-unstable modes results in a beat signal of the radius perturbation along the jet, and thus also in the difference $R(x, t) - R_0$ between the perturbed jet radius $R(x, t)$ and the unperturbed radius R_0 . The perturbation wavelengths are chosen such that the growth-rate of the two modes is equal, hence the beat signal shape is maintained until the non-linear interactions become significant. As the perturbation amplitude grows towards the magnitude of the jet radius, the first pinchoff occurs close to the anti-node of the beat signal. The beat signal causes a difference in radius between the front and the back of a pinching droplet. Since the tension applied by capillarity decreases with the jet radius at the pinch-off location, this difference causes a net force acting on the pinching droplet. Over time, this force accelerates the pinching droplets away from the thinnest point, which is at the anti-node of the beat signal. After pinch-off, all droplets formed between two adjacent anti-nodes move towards the node of the beat envelope. This convergent motion results in coalescence of the small droplets into one large droplet per shortest common wavelength of the perturbations. We call the difference between the breakup time of the jet and the time at which the last small droplet merges the merge time t_{merge} .

With the superposition of two fast growing modes, we have access to small (dimensionless) wavenumbers that otherwise suffer from noise [23] and large satellite droplets [13]. We use the properties of the Rayleigh-Plateau instability to select the wavenumbers for the beat signal. By selecting the phases and frequencies of the perturbations carefully, the spacing of the resulting droplets can be controlled with great accuracy, as we show in the result section of this chapter. In the next

Figure 2.2: A small initial perturbation triggers jet-breakup and subsequent coalescence. The final droplet spacing is given by the shortest common wavelength of the two perturbations. The small perturbation consists of the superposition of two Rayleigh-Plateau-unstable modes. From left to right, the shortest common wavelength of the two perturbations increases. The method used to choose the wavenumbers of the two Rayleigh-Plateau-unstable modes is described in section 2.3.2. From top to bottom we show the different stages in the evolution of one shortest common period of the two Rayleigh-Plateau-unstable modes. The time between the consecutive contours is respectively 5, 20 and 55 times t_{cap} , where $t_{cap} = \sqrt{\rho R_0^3 / \sigma}$. The beat signal wavelength is 22.9, 31.8, and 40.8 times R_0 for $n = 2, n = 3$, and $n = 4$. Until the breakup, the tips of the ligaments are accelerated by surface tension away from the point where the jet breaks first. The results shown in Fig. 2.2 have been generated using a numerical model, see section 2.5. For this demonstration, periodic boundary conditions were chosen. At $t = 0$ we impose the two Rayleigh-Plateau-unstable modes. The initial amplitudes of both radial perturbations is $0.005 R_0$. The velocity perturbations are set accordingly, see equations (2.5), (2.6), and (2.7).

section we will explain the wavenumber selection in more detail.

2.3 Analysis

2.3.1 Linear theory

A laminar liquid jet is subject to the Rayleigh-Plateau instability. We demonstrate how the superposition of two Rayleigh-Plateau-unstable modes is employed to achieve a robust drop formation method with a controllable droplet size. The Rayleigh-Plateau instability is approximated in the slender jet approximation [24, 25], which is a systematic reduction of the Navier-Stokes equation for an

axisymmetric jet, and which turned out to be extremely valuable in inkjet printing, cf. [26], [27], [28], and [29]. There are also cases in inkjet printing where a more advanced model is necessary, such as the case when the meniscus retracts into the nozzle, see e.g. [30]. The slender jet approximation in conservation form is given by (cf. [31])

$$\partial_t A + \partial_x(Au) = 0, \quad (2.1)$$

$$\partial_t(\rho Au) + \partial_x(\rho Au) = \partial_x(\tau_\sigma + \tau_\mu), \quad (2.2)$$

where $A = A(x, t)$ is the time dependent cross sectional area along the jet, $u = u(x, t)$ is the time dependent axial velocity along the jet, and $\tau_\sigma = \tau_\sigma(x, t)$ and $\tau_\mu = \tau_\mu(x, t)$ are respectively the capillary and the viscous tension. For an axi-symmetric liquid jet, the capillary tension τ_σ acting on the jet is given by [32]

$$\tau_\sigma = -\pi\sigma \left(\frac{R}{\sqrt{1 + (\partial_x R)^2}} + \frac{R^2 \partial_{xx} R}{(1 + (\partial_x R)^2)^{3/2}} \right), \quad (2.3)$$

where $R = R(x, t)$ is the time dependent radius along the jet. The viscous tension τ_μ is given by the extensional viscosity in an axisymmetric jet (cf. [24]),

$$\tau_\mu = 3\mu A \partial_x u. \quad (2.4)$$

The ratio between the viscous and capillary forces is called the Ohnesorge number $\text{Oh} = \mu / \sqrt{\rho R_0 \sigma}$. In our experiments $\text{Oh} = 0.03$, hence for the stability of our jet the capillary effects are dominant.

After coalescence however, viscosity plays an important role. When two droplets merge, the excess surface energy causes oscillatory modes on the resulting droplet. Each time another droplet merges with the oscillating droplet, more excess surface energy is added to the oscillating droplet. Due to the presence of viscosity, this excess energy is dissipated over time. Neglecting viscosity means that the droplet oscillations do not dampen out, which may or may not cause unnatural droplet breakup after coalescence [33, 34]. Therefore, we included τ_μ in our slender jet approximation model.

In the analysis of the onset of the Rayleigh-Plateau instability, the following ansatz for the perturbation is used:

$$R(x, t) = R_0 + R_a \cos(kx) \exp(\omega t), \quad (2.5)$$

$$u(x, t) = u_0 - u_a \sin(kx) \exp(\omega t), \quad (2.6)$$

where u_0 is the unperturbed velocity, R_a and u_a are the initial perturbation amplitudes, k is the perturbation wavenumber, and ω is the growth-rate of the perturbation. The perturbation amplitudes R_a and u_a are much smaller than the unperturbed

values u_0 and R_0 . From equations (2.1)-(2.2) it follows that the value of u_a is related to R_a by

$$u_a = \frac{2\omega}{k} \frac{R_a}{R_0}. \quad (2.7)$$

As the unstable modes have a wavelength that is longer than the circumference of the jet, the long wavelength approximation is sufficiently accurate for describing the jet evolution [10]. The dispersion relation for the Rayleigh-Plateau instability in the long wavelength approximation is found by substituting the ansatz into the slender jet approximation, equations (2.1)-(2.2), giving

$$\omega = \frac{1}{t_{cap}} \left(\sqrt{\frac{1}{2}(\chi^2 - \chi^4) + \frac{9}{4}\text{Oh}^2\chi^4 - \frac{3}{2}\text{Oh}\chi^2} \right). \quad (2.8)$$

A continuous jet ejected from a nozzle is obviously not an infinite jet. On a continuous jet with a periodic perturbation, both the perturbation amplitude at the nozzle and the breakup length are constant. This implies that the amplitude of the Rayleigh-Plateau instability on a continuous jet grows in space. The spatial growth-rate is governed by the Weber number, which gives the balance between inertia and surface tension, $We = \rho u_0^2 R_0 / \sigma$. [12] showed that the wavenumber is a complex number at low Weber numbers. This implies that when the spatial asymmetry of the Rayleigh-Plateau instability is large, a growing perturbation also travels over the surface of the jet. In our experiments the Weber number is above 100, hence we can assume that the perturbation grows like on an infinite jet. Mathematically speaking, this means that both the temporal growth-rate and the spatial wave number of the growing instabilities are real valued numbers: The growing perturbations have no velocity relative to the fluid jet.

The amplitude of a Rayleigh-Plateau-unstable mode on a finite jet only grows in space, and the phase travels along with the jet. We rewrite the ansatz for a Rayleigh-Plateau-unstable mode on an infinite cylinder (equations (2.5)-(2.6)) to a Rayleigh-Plateau-unstable mode on a jet ejected by a nozzle:

$$R(x, t) = R_0 + R_a \cos(k(x - u_0 t)) \exp\left(\omega \frac{x}{u_0}\right), \quad (2.9)$$

$$u(x, t) = u_0 - u_a \sin(k(x - u_0 t)) \exp\left(\omega \frac{x}{u_0}\right), \quad (2.10)$$

where R_a and u_a are the radius and velocity perturbation at the nozzle, where $x = 0$. At high Weber number the perturbations move along with the jet, and their amplitudes grow with the distance the jet has propagated from the nozzle. As long as the amplitudes of the two Rayleigh-Plateau-unstable modes are small, linear superposition of the two perturbations is justified. The superposition of two Rayleigh-Plateau-unstable modes with different wavelengths leads to an interference pattern with a growing amplitude.

2.3.2 Wavenumber selection

The goal of the method is to obtain a robust stream of droplets, using only a small amplitude perturbation on the pressure drop over the nozzle. In principle, any combination of two different growing modes leads to a beat signal in the radius perturbation, but when the growth-rate of the noise is much higher than that of the applied perturbations, it induces a random fluctuation in the droplet velocities [23]. Therefore, for the selection of the wavenumbers we take the properties of the dispersion relation of the Rayleigh-Plateau instability into account. For a robust stream of droplets, it is important that the signal to noise ratio remains large throughout the entire droplet formation process. When we carefully select two wavenumbers with equal growth-rates, close to the maximum growth-rate, we can use a very small perturbation amplitude, as we will demonstrate quantitatively in section 4.4.

Using two selection criteria we select the fastest growing combination of wavenumbers for a given shortest common period. The first criterion is that the growth-rate of both modes is equal. In this case, a beat signal applied at the nozzle will grow in amplitude, but maintain its shape until the non-linear interactions emerge. The perturbation amplitudes of both modes will still be the same near pinch-off. When the two perturbations have an equal growth-rate, we can apply them at nearly the same small amplitude to obtain a fully modulated beat signal.

For the stream of droplets to be periodic, the shortest common wavelength of the two superimposed perturbations should be finite. A finite shortest common wavelength is guaranteed when both dimensionless wavenumbers are an integer multiple of the dimensionless base wavenumber, χ_0 . This leads to the second criterion, given by $(n+1)\chi_1 = n\chi_2$, where n is an integer. The shortest common wavelength increases with n . Note that n does not stand for the number of merging droplets, n is only the integer value that gives the relation between χ_1 and χ_2 . For any combination of two growing modes with a given shortest common wavelength, this is the combination that has the highest growth-rate for the given shortest common wavelength.

The two selection criteria lead to a unique combination of wavenumbers. For the case of $Oh \ll 1$ this combination is given by:

$$\chi_1 = \frac{\left(\frac{n}{n+1}\right)}{1 + \left(\frac{n}{n+1}\right)^2} \quad (2.11)$$

$$\chi_2 = \frac{1}{1 + \left(\frac{n}{n+1}\right)^2} \quad (2.12)$$

Since the dispersion relation of the Rayleigh-Plateau instability is concave, the wavenumbers of the two chosen Rayleigh-Plateau-unstable modes approach the

Figure 2.3: Superposition of two Rayleigh-Plateau-unstable modes on a liquid jet. The linear approximation (dashed line) and the numerical simulation (solid line) agree very well, until the satellite droplets start to form. The Weber number of this jet is 300, the value of $n = 2$. This figure shows that linear superposition of the two perturbations gives a good approximation when the perturbation amplitude is small.

wavenumber with the highest growth-rate as n increases.

2.3.3 Non-linear interactions

When the amplitude of the Rayleigh-Plateau-unstable modes approaches the jet radius, the non-linear terms emerge in the perturbation spectrum [16]. [35] demonstrated experimentally that the non-linear analysis on an infinite cylinder is valid for a continuous jet with a high Weber number, as expected by [12]. To illustrate the validity range of the linear approximation, we show the linear ansatz for the spatial instability equations (2.9)-(2.10) versus a numerical simulation that contains the non-linear terms in Fig. 2.3. The dimensionless wavenumbers for this comparison are selected for $n = 2$. We obtained the non-linear evolution of the perturbed jet numerically by solving the slender jet approximation in the implementation as explained in [31]. The linear approximation is valid until satellite droplets start to form.

The growth of the beat signal ends with the jet-breakup. As explained in the conceptional section (sec. 2.2) the jet breaks first at the anti-nodes of the beat signal. One by one, the transient droplets break up from the jet, see Fig. 2.2. Each transient droplet pinches first at the anti-node side, and later at the node side. [36] proposed that this asymmetry results in a net capillary force on the pinching transient droplet, and that the relative velocity of these droplets can be calculated by integrating the capillary forces in the linear approximation during the breakup process. The success of the method depends on the modulation of the beat signal near breakup. The asymmetry that causes the capillary forcing is largest just before pinch-off. We now compare the results, from the full nonlinear model and the linear approximation thereof. This comparison will show that the non-linear interactions that cause the satellite formation have a significant quantitative influence on the relative velocities of the small droplets.

For this comparison the net capillary forces on the pinching droplets within the full nonlinear model and the (analytical) linear approximation thereof are

Figure 2.4: Time evolution of the relative velocity u/u_{cap} of a pinching transient droplet, during the final stages of the pinch-off. The Weber number for this jet is 151, i.e. the same as the jet in the later experimental Fig. 2.8. Also $Oh = 0.03$ is the same as in experiment. The pinching transient droplet is shown as the shaded part of the jet in the contour in the upper right corner of the graph. The velocity induced by the surface tension (this velocity is relative to the jet velocity) is found by integrating the acceleration caused by the net capillary tension on the pinching transient droplet. For the linear result (dashed line), the ansatz of equations (2.9)-(2.10) is used. For the numerical result (solid line) a simulation is performed with the same initial parameters as for the linear ansatz. In the linear approximation, the integral on the capillary forces results in a relative velocity that is 36% higher (modulo-wise) than the numerical result of the full nonlinear model.

calculated. For both cases we used the same initial conditions, namely the ansatz for small amplitude perturbations, equations (2.9)-(2.10). The numerical results are obtained with a numerical implementation of equations (2.1)-(2.4), cf. [31]. From equation (2.3) the capillary tension in the axisymmetric liquid jet and also in the formed droplets is known until pinch-off. The last droplet to merge is the droplet next to the anti-node of the beat signal. The merge time of this last droplet with the rest of the droplets scales inversely with its relative velocity. In Fig. 2.4 we show the velocity change of the last droplet to merge for both the full nonlinear solution and the linear approximation thereof. The part of volume that will form this droplet is shown as the shaded part of the jet shape. The velocity change of the pinching droplet is determined by integrating the accelerations $a = F/m$ over time, where m is the mass of the pinching droplet, and F is the force due to the capillary asymmetry around the pinching droplet. We assured that we can ignore viscous stresses and the momentum flux through the edges of the pinching droplet. The result of this integral (Fig. 2.4) shows that the relative velocity is 36% larger (modulo-wise) within the linear approximation. We thus conclude that the non-linear terms have a significant influence on the final relative droplet velocity. For the comparison with experiments we thus use the full nonlinear (and therefore only numerical) model, rather than the analytical result which we can obtain within the linear approximation.

2.4 Experimental Setup

The continuous liquid jet is ejected from a Microdrop AD-K-501 micropipette. Such a pipette consists of a glass capillary of which the end is shaped like a nozzle. The nozzle radius is $15\mu\text{m}$. Part of the capillary is surrounded by a tubular piezoelectric actuator. Note that the jet radius R_0 is smaller than the nozzle radius. There are two mechanisms that increase the average velocity after the fluid leaves the nozzle. The main effect is the relaxation of the Poiseuille profile inside the nozzle into a plug flow profile of the jet. A second reason for the contraction is the sinusoidal velocity perturbation, which causes the average momentum-flux to be higher than the momentum-flux of an unperturbed jet [37]. A calibrated Shimadzu LC-20AD HPLC pump supplied degassed micropore filtered water at a constant flow rate. The fluid properties used in the calculations are density $\rho = 1000\text{kgm}^{-3}$, surface tension $\sigma = 0.072\text{N/m}$, and dynamic viscosity $\mu = 0.001\text{Pas}$. All measurements were done in a conditioned room at $20 \pm 0.5^\circ\text{C}$. For the experiments the flow rate was $Q = 1.00\text{ml/min}$ in Fig. 2.8 and 0.90ml/min in figures 2.6, 2.7, 2.9, 2.10, and 2.11. With the piezoelectric element of the micropipette we applied periodic pressure perturbations to the fluid. The electric signals for these perturbations are supplied by a 40 MHz Wavetek Waveform

Figure 2.5: Schematic representation of the experimental setup. A continuous jet is emitted through a nozzle at the end of a glass capillary. A pressure perturbation is applied by a piezo electric actuator mounted on the glass capillary. The drop formation is recorded using a camera and a pulsed LED. The drive signal to the LED is coupled electrically to the piezoelectric actuator. By changing the delay time between the signal to the piezoelectric actuator and the LED, the complete evolution of a periodically perturbed jet is recorded.

Generator (Model 195), amplified 50 times with a Falco Systems DC-5MHz High Voltage Amplifier WMA-500. The two sinusoidal signals are generated separately, using an integer number of clock steps to ensure precise setting of the frequencies and the common period, such that no unwanted phase changes occur. The two signals are then summed on a third channel. The technical implementation of the method is discussed in detail in Appendix 2.8.

Since the jet perturbation is periodic, a single flash stroboscopic setup suffices to record the complete jet evolution, see Fig. 5.2. We used the JetXpert, the drop-in-flight visualization and analysis system from ImageXpert, which is equipped with a Stingray F-080 camera and a blue pulsed LED. The measurement setup is driven such that one image is made at the time. By sweeping the delay between the waveform and the LED trigger over one period of the beat signal, we record the jet evolution at a high temporal resolution.

2.5 Numerical Simulations

We use the full nonlinear slender jet approximation (equations (2.1)–(2.4)), to verify that the jet-breakup is indeed fully governed by the two imposed sinusoidal perturbations. The slender jet approximation has been used for the study of the stability of the axisymmetric liquid jet before [38–40]. For the case of the Rayleigh-Plateau-unstable jet, the slender jet model has been proven to be a great predictive tool, see e.g. [14, 24, 29, 31]. In other cases, such as end pinching of a liquid jet [41], a more advanced model is necessary to predict the dynamics of the system [42, 43].

For the presented results we used a previously developed numerical implementation of the slender jet approximation [31]. In this implementation, the singularity that occurs at pinchoff in the continuum approximation does not occur, due to a regularization of the surface tension term. When the jet radius becomes smaller than a critical radius, the surface tension approaches a constant value. This way, a thin filament is present between the separate liquid bodies, and a change in topology is prevented. The model is thoroughly validated [31] and used [29, 44].

The initial conditions at the inlet of the simulations have to be determined from the experimental data. However, we cannot use the data directly at the nozzle. First of all, the distortions directly behind the nozzle are so small that they are hard to experimentally detect. Secondly, close to the nozzle the flow is still influenced by the Poiseuille profile inside the nozzle, but in the slender jet approximation plug flow was assumed. Instead of starting the simulations directly at the nozzle, we start the simulations at a location where we can optically detect the perturbations. We call this distance sufficiently far from the nozzle x_{dis} . The work of Garcia [15] shows that the transient effects of the nozzle conditions vanish rapidly when the perturbation amplitude is small. At x_{dis} the perturbation amplitude is still much smaller than the jet radius R_0 . Ambravaneswaran [28] investigated the influence of the Poiseuille profile on the drop formation for the case of a dripping jet. From that location we can integrate our dynamical model equations (2.1)–(2.4) backwards in space and time, to deduce the initial distortion at the nozzle. We will use this approach in subsection 2.6.1.

In this section, we integrate the dynamical model equations (2.1)–(2.4) forward in space and time, starting from the position x_{dis} , and assuming that the radial and velocity perturbation are linear, according to equations (2.9)–(2.10). These two perturbations are applied simultaneously, both with their own amplitude and phase:

$$\begin{aligned} R(x_{dis}, t) &= R_0 - R_{a,1} \cos(u_0 k_1 t - k_1 x_{dis} - \phi_1) - R_{a,2} \cos(u_0 k_2 t - k_2 x_{dis} - \phi_2), \\ u(x_{dis}, t) &= u_0 + u_{a,1} \sin(u_0 k_1 t - k_1 x_{dis} - \phi_1) + u_{a,2} \sin(u_0 k_2 t - k_2 x_{dis} - \phi_2). \end{aligned}$$

Here $R_{a,1}$ and $R_{a,2}$ are the radial perturbation amplitudes, $u_{a,1}$ and $u_{a,2}$ are the velocity perturbation amplitudes, and ϕ_1 and ϕ_2 are the phases of the two separate

Figure 2.6: The radius perturbation along the jet, obtained from the stroboscopic recordings (black dots). The Weber number of this jet was 107, and $x_{dis} = 1.46$ mm. The amplitude and phase of the two perturbations are found with a linear fit (solid red line) to equation (2.13). This linear fit of the complete perturbations is possible in the regime where the amplitude is small compared to the jet radius R_0 and, as seen, describes the experimental data very well.

Rayleigh-Plateau-unstable modes, all taken at the distance x_{dis} from the nozzle, where the four perturbation amplitudes are already much larger than at the nozzle.

To experimentally determine these parameters, we first determine the jet contour of the recorded jet with sub-pixel accuracy [27]. Note that the contour that we determine in this way, has an a priori unknown offset normal to the real liquid interface. Since the offset is constant in space, we can subtract the average jet radius from the contour, to obtain the beat signal (see Fig. 2.6). This beat signal is linearized,

$$R(x, t=0) - R_0 = R_{a,1} \cos(k_1 x + \phi_1) \exp\left(\omega_1 \frac{x}{u_0}\right) + R_{a,2} \cos(k_2 x + \phi_2) \exp\left(\omega_2 \frac{x}{u_0}\right), \quad (2.13)$$

and the fitting parameters u_0 , $R_{a,1}$, $R_{a,2}$, ϕ_1 , and ϕ_2 are adjusted to well describe the measured beat signal. The fit is basically indistinguishable from the data itself, see Fig. 2.6. The remaining parameters R_0 , ω_1 , ω_2 , k_1 en k_2 , are dependent on u_0 , Q , and the distortion frequencies. Note that in this way R_0 is determined with a much higher precision than optically, simply by employing the relation

$$Q = \pi R_0^2 u_0, \quad (2.14)$$

as we know the volume flux Q with a very high precision. The whole procedure obviously only makes sense after temporal transients have died out, which we assured.

Figure 2.7: Experimental (top) and numerical (bottom) results for a jet ($We = 107$) perturbed with two Rayleigh-Plateau-unstable modes for $n = 2$. The perturbation frequencies were 133 kHz for mode χ_1 and 200 kHz for χ_2 . For each shortest common period, three main droplets are generated and one satellite droplet. All droplets merge into one big droplet after a short time. The phase and amplitude of the two Rayleigh-Plateau-unstable modes fully determine the breakup of the liquid jet and the coalescence of the resulting small droplets.

2.6 Results

We now present our main results, namely that the perturbation of a continuous jet with the two distortion modes chosen according to our scheme (Fig. 2.1) leads to widely spaced droplets, with a radius much larger than the jet radius. We start this section by first demonstrating that the method indeed works, namely by comparing the experimental and numerical results for $n = 2$. The comparison is performed for two different cases. The first case is characterized by $We = 107$, the second by $We = 151$. The differences between these two cases are discussed in the appendix. In section 2.6.2 we use the case for $We = 107$ for further investigation of the presented method and demonstrate how the droplet size and inter-droplet spacing can be controlled using the presented method. In section 2.6.3 we show, also for the case $We = 107$, how the phase difference between the two modes influences the coalescence process.

2.6.1 Breakup and coalescence for $n=2$

First we show the comparison between the experiment and the numerical result of the drop formation for a Weber number of 107. In Fig. 2.7 the experimentally observed jet is shown together with the numerical simulation thereof. The jet is ejected from the nozzle, and after traveling a distance of 3 mm the jet breaks up in a stream of small droplets. After traveling another 2 mm, the small droplets coalesce into larger droplets, widely spaced by the shortest common wavelength λ_0 . The three main droplets merge within $5 \lambda_0$ after the breakup distance. In the numerical simulation the three main droplets merge slightly later than in the experiment. From Fig. 2.7 we conclude that indeed we succeed to produce large widely spaced droplets with our method, and that experiment and numerical results

reasonably agree.

It is interesting to deduce how large the radial and velocity distortions, which lead to the observed distortions at x_{dis} (Fig. 2.6 and 2.7), were at the nozzle $x = 0$. When we assume that at x_{dis} we are still in the linear regime, we can simply apply equation (2.9) and from our measured values $R_{a,1}(x = x_{dis}) = 0.21 \mu\text{m}$ and $R_{a,2}(x = x_{dis}) = 0.15 \mu\text{m}$ we obtain the initial distortion $R_{a,1}(x = 0) = 10 \text{ nm}$ for mode 1, and $R_{a,2}(x = 0) = 7.4 \text{ nm}$ for mode 2. The radial distortions at the nozzle are indeed very small compared to the jet radius of $R_0 = 14.5 \mu\text{m}$, and clearly not detectable using visible light. The corresponding velocity perturbations at the nozzle follow from equation (2.7) and are $u_{a,1}(x = 0) = 1.8 \text{ mm/s}$ and $u_{a,2}(x = 0) = 0.87 \text{ mm/s}$ for mode 1 and 2 respectively, again small, now compared to the jet velocity $u_0 = 22.8 \text{ m/s}$.

In Fig. 2.8, we show the results for the case with $We = 151$. The data has been shown as a space time plot, moving along with the shortest common period of the perturbations on the jet. A space time plot allows us to compare the results at a high spatial and temporal resolution. This way of plotting was used before in [45, 46]. In the figure we get a detailed view on the growth of the perturbation, the jet-breakup and on the coalescence process. The periodicity of the experiment allowed us to use many different recordings of the jet for the construction of this plot. The experimental data for this plot are processed only at a pixel-level accuracy, therefore the color does not match in the initial stages of the comparison. Since the initial conditions for the numerical model are determined at sub-pixel accuracy [27], this does not influence the later stages of the comparison.

Directly after breakup, the position and size of the droplets match very well. The droplet velocities are slightly different. This results in minor differences in the coalescence process. The three large droplets in the experiment merge simultaneously, whereas the two droplets in the front merge slightly earlier in the numerical result. We believe that the minor differences between the droplet velocities are caused by a small error in the determination of the phases of the different modes. In section 2.6.3 we will show that the droplet velocities are very sensitive to the phase difference between the two signals that form the initial conditions of the simulations.

Summarising this subsection, we conclude that the breakup and merging dynamics of the experiment and the numerical simulation match for both analysed cases $We = 107$ and $We = 151$. From this comparison we can conclude that the drop formation is indeed solely governed by the two imposed distortion modes.

2.6.2 Drop size modulation for higher n

The volume of the droplets in the final droplet stream scales with the shortest common wavelength of the two perturbations. When the two wavenumbers ap-

Figure 2.8: Comparison of the experimental (left) and numerical (right) results with the experimental results for $n=2$. The Weber number for this jet is 151, and the perturbation frequencies for the two perturbations were 162.6 kHz and 243.9 kHz, respectively. The periods of the applied perturbations are integer multiples of the time step of the 40 MHz waveform generator. The values for the phases ϕ_1 and ϕ_2 were 0.23 and -1.44 radians respectively. The data for $R(x,t)$ are shown as a space-time plot, moving along with one common period of the two perturbations. The color shows the radius. In these graphs, the evolution of one common period of the two perturbations is shown, moving along with the jet. First the jet breaks up into main and satellite droplets, later the droplets merge into one droplet per common period. Until after breakup, the experimental and numerical results agree within experimental precision.

proach each other in the k -space, the shortest common wavelength increases. The wavenumbers required for optimal performance of the method were already derived in section 2.3.2. In Fig. 2.9, we show the jet-breakup and coalescence for the modes $n = 3$, $n = 4$, and $n = 5$. The amplitude and phase of all perturbations are set as explained in appendix 2.8.1. For moderate values of n , the aspect ratio, being the ratio between the merging distance and the nozzle radius, becomes large. This means that it becomes impossible to capture the whole process in one image. We constructed one image of the complete breakup and coalescence process by stitching together recordings at different heights. We have chosen to show the jet breakup and coalescence in the frame of reference of the jet velocity. From top to bottom, the camera shifts with λ_0 away from the nozzle for each next row, while the phase of the perturbations remains fixed. The time between two frames is λ_0/u_0 . The jet breakup and coalescence process can be observed in great detail in this way. Note that due to the increase of λ_0 with n , the time between two lines in the figure increases with n .

The fluid motion displayed in Fig. 2.9 is shown relative to the jet velocity. As the merge time increases, the influence of air drag becomes significant for the droplet motion. Indeed, for $n = 4$ and $n=5$, the curved droplet trajectories in space and time show that the droplet velocities clearly decrease in time, and we attribute this deceleration to the air drag. The curvature of the trajectories of the small droplets is higher, since they are more sensitive to air drag. For all three cases we first observe the jet breakup into main and satellite droplets. Over time, the droplets coalesce into one big droplet per shortest common period of the perturbations. As n increases, the merge time increases. In the experiments presented in Fig. 2.9 the merge time for $n = 3, 4$, and 5 is $331 \mu\text{s}$, $689 \mu\text{s}$, and $969 \mu\text{s}$, respectively. There are two reasons for this increase: First, as the common wavelength becomes longer as n increases, the transient droplets have to travel larger distances. Second, the relative amplitude difference between the local minima near the anti-node of the beat function is smaller. The smaller asymmetry results in a smaller net capillary force on the pinching droplet. Hence these droplets, which have to travel from the anti-node to the node, do this at a smaller velocity for higher n . In theory, there is no upper limit for the value of n . In practice, however, the method is limited to moderate values of n , $n < 10$. The limitation is mainly caused by the increase of the merge time, which in combination with the high Weber number results in a large merging distance. To achieve a stable droplet stream at the higher values of n , the droplet generator should be placed in a large vacuum container as in [36] to avoid interaction with air.

Figure 2.9: Experimental results for the coalescence dynamics of the transient droplets for modes $n = 3, 4$, and 5 . The Weber number for all three jets was 107 . The perturbation periods for mode $n = 3, 4$, and 5 were: 144.93 kHz & 193.24 kHz, 105.94 kHz & 188.68 kHz, and 155.04 kHz & 186.05 kHz respectively. Note that the periods of all applied perturbations are integer multiples of the time step of the 40 MHz waveform generator. To show the evolution of the breakup and merging, the camera moves along with the jet, at the velocity of the relaxed jet u_0 . The time between the consecutive recordings is λ_0/u_0 . In order to get this image the phase locked droplet streams were captured at different distances from the nozzle, and then pasted below each other. Small corrections in height were applied, all within the error margin of the linear motor which controlled the nozzle position. The superposition of two Rayleigh-Plateau modes provides a robust method to control the size and the inter-droplet spacing of continuous streams of droplets.

Figure 2.10: The merge time as a function of the phase shift between the two sinusoidal perturbations, determined numerically (line) and experimentally (diamonds) for mode $n = 2$. The Weber number was 107. The merge time has two local maxima as a function of the phase difference between the two modes. At $\phi_1 = 0$ the two modes are in phase at a local minimum. In this case, a satellite forms at the anti-node of the beat signal, which travels at the same velocity as the coalesced droplets (see Fig. 2.2). At $\phi_1 = \pm\pi/3$ the two modes are in phase at a local maximum. In this case, the beat signal has two equal minima per common period. This results in two large droplets that travel at the same velocity. The merge time exceeds 10 000 capillary times close to these phase values. In the experiment, interaction with air inhibits these extremely long merge times. This graph demonstrates that the phase shift can have a large influence on the merge time of the droplets.

2.6.3 Phase dependence

The droplet formation depends strongly on the phase difference between the two perturbations. In Fig. 2.10 we show the experimental and numerical results of a phase shift between the two modes. We added schematic contours to explain the different phases. Before discussing the results, we first explain how the phase differences are defined. As a result from the wavenumber selection (see section 2.3.2), λ_1 fits n times into λ_0 and λ_2 fits $n + 1$ times into λ_0 . Therefore, by shifting χ_2 over χ_1 there are $n(n + 1)$ possible phases in λ_0 for which the two modes are in phase. The shown phase scan was done for the case $n = 2$, hence there are 6 different phase shifts for ϕ_1 that give the same beat signal. The minimal sector for the phase shift, that contains all possible phase combinations, is $2\pi/6$ from the period of the beat signal. The equivalent in the periodicity of mode χ_1 is two times larger, since χ_1 fits two times in the beat wavelength at $n = 2$. In the experiments of Fig. 2.10 the phase shift was implemented by setting ϕ_2 to an arbitrary fixed value, and shifting ϕ_1 over a domain of $[0 \dots 2\pi/3]$. The actual phases were determined with the linear fit afterwards. The resulting values of ϕ_1 were shifted with $-\phi_2$, so that the phase difference is given by the value of ϕ_1 alone (ϕ_2 is zero after this shift). For the numerical result, we fixed $\phi_2 = 0$ and shifted ϕ_1 over the domain $\phi_1 = [-\pi/3 \dots \pi/3]$. To compare both results, we show the merge time in the domain of $\phi_1 = [-\pi/3 \dots \pi/3]$.

The merge time in the numerical result becomes very large for some phase differences. This leads to a huge aspect ratio for which we need to calculate the droplet motions. To shorten the calculations, we assumed that the droplets become spherical after the surface oscillations have dampened out, and that they travel at constant velocity until they coalesce with another droplet. This assumption is allowed since the relative velocity of the droplets is of the order of the capillary velocity (see Fig. 2.4), rendering the Weber number of the droplet collision of order $We = o(1)$. Since the collision is head on, we can safely assume that coalescence will take place, based on the parameter scan by [47]. By assuming coalescence at every droplet collision, we only need to solve the nonlinear slender jet equations until all droplets in one λ_0 have broken up from the jet. The merge time was then found using an event-driven hard-sphere coalescence model, where the droplets coalesce when they collide.

The phase scan in Fig. 2.10 shows that the merge time varies strongly for different phases. There are two phase differences at which the merge time peaks, namely near $\phi_1 = 0$ and near $\phi_1 = \pm\pi/3$. We immediately observe that these peaks are not symmetric around $\phi_1 = 0$. Note that this also holds for the peak around $\phi_1 = \pm\pi/3$, which is less evident from this graph. These asymmetries are caused by the finite Weber number of the jet. As the jet moves away from the nozzle, the perturbation amplitude increases, hence the amplitude of the beat function

grows in space. This puts an extra asymmetry over the pinching droplets. In the case of an infinite Weber number, the perturbation amplitude would be constant in space, and growing only in time. In Fig. 2.2 we have already shown the case of an infinite Weber number, with $\phi_1 = 0$. In the next two paragraphs we explain the physics behind the two peaks in the phase scan. For readability, we ignore the extra asymmetry caused by the finite Weber number for this explanation; it does not play a role to understand the mechanism.

At $\phi_1 = \pm\pi/3$ the two perturbations are in phase at a local maximum. The contours under the graph show that in the case of $\phi_1 = \pm\pi/3$, two large droplets of different size are formed simultaneously, since there are two equal local minima per λ_0 . Due to this equal amplitude on both sides of these pinching droplets, there is no net capillary force on the pinching droplets. Their velocities after pinch-off are equal.

At $\phi_1 = 0$, the two perturbations are in phase at a local minimum. This means that there is one global minimum per λ_0 , hence from the linear growth we expect all droplets to move away from this global minimum after pinch-off. Due to non-linear interactions however, a satellite droplet forms at this location. When the two perturbations are exactly in phase, the symmetry around this satellite is not broken, and it does not gain any net velocity from the surface tension. The initial velocity of the merged main droplets and the satellite droplet is the same. From Fig. 2.2 we can see that the satellite droplet moves at the same velocity as the merged main droplets combined at $\phi_1 = 0$.

An identical initial velocity would mean that the droplets never collide in the absence of air. Hence in the numerical results there are two values for ϕ_1 for which the merge time diverges. The exact values for these two phases depend on the Weber number; they converge to $\phi_1 = 0$ and $\phi_1 = \pm\pi/3$ as $We \rightarrow \infty$. In the experiment however, the merge time is finite due to the interaction with air.

2.7 Conclusion and Outlook

The superposition of two Rayleigh-Plateau-unstable modes is an efficient and robust method to generate a periodic stream of droplets, that allows for control of the droplet sizes and distances. The method is efficient because it uses the available surface energy of the continuous jet to induce the coalescence. The method is robust because the wavenumbers of the employed Rayleigh-Plateau-unstable modes are close to the fastest growing mode. By extracting the amplitudes and phases of the two Rayleigh-Plateau-unstable modes from the detailed experimental data and using them as the initial conditions at the inlet of the numerical simulation, we showed that the drop formation is governed by only these two frequencies. The acoustic properties of the device for jet generation and modulation are not needed

for the analysis when the images of the drop formation are used to tune the settings of the perturbation signals. By tuning the phase difference between the two modes, the coalescence pattern is controlled. The phase difference with the shortest merge time is not the one for which the two perturbations are in phase at a minimum, due to the presence of satellite droplets. The final droplet size, spacing and merge time increase with the beat number n .

2.7.1 Dropsizes modulation for high-speed droplet streams

That it is difficult to generate a steady stream of widely-spaced monodisperse droplets with only *one* Rayleigh-Plateau unstable mode, has two main reasons: Firstly, the growth rate of the long wavelengths is so low that the drop formation can be dominated by noise [23], and, secondly, the relative size of the satellite droplets increases rapidly for increasing perturbation wavelengths [13, 48]. These satellite droplets form an extra nuisance at high Weber numbers, since they tend to travel at approximately the same velocity as the main droplets [49], causing a bi-disperse drop size distribution. By altering the actuation signal, satellite droplets can be prevented, for example by using high amplitude perturbations, or by exciting higher order modes on top of the unstable mode of choice [18].

With the presented droplet size modulation method, it is simple and energetically cheap to prevent satellite droplets. In section 2.6.3 we demonstrate that the phase difference between the two modes strongly influences the velocity of the satellite. This makes it easy to let the satellite quickly merge with the larger droplets. Since the complete drop formation is governed by the two chosen unstable modes, and since the energy needed for the coalescence of the droplets comes from the available surface energy, the presented method is very suitable for droplet size modulation in high-speed laminar fluid jets.

2.7.2 Interaction with air

When comparing the experimental and numerical results, we observed that the interaction with air has an influence on the coalescence pattern of the small droplets. For applications under ambient pressure, it is relevant to quantitatively investigate the interaction between the droplets and the surrounding air. For the ever decreasing droplet size and ever increasing droplet velocity the effect of air drag will become increasingly important.

2.8 Appendix: Experimental Implementation of the Optimal Perturbations

In this chapter we used the results of two different experiments. These two experiments are characterized by their Weber numbers, namely $We = 107$ and $We = 151$. We employed different procedures for these experiments. For the experiment with $We = 107$, the wavenumber selection is optimized for merging time. The experiment with $We = 151$ has an easier implementation, but gives a longer merging time.

2.8.1 Experiment with $We = 107$

The success of the method depends on the modulation of the beat signal near breakup. When the two signals have an equal amplitude near breakup, the beat signal is fully modulated there. In that case, the asymmetries that allow the surface tension to pull the droplets away from the anti-nodes of the beat signal are maximized. Since the radius and velocity of the jet are determined with a small experimental error, we do not a priori know if we applied two perturbations with exactly the same growth-rate. Furthermore, we do not know the acoustic and electrical transfer function of the micropipette. These frequency dependent transfer functions may induce extra phase shifts and amplitude differences between the two electrically imposed signals. When the jet velocity and the jet radius are known within experimental precision, we choose the perturbation frequencies according to the procedure explained in section 2.3.2. Using our setup we then tune the amplitude and phase of the two modes to ensure that the amplitudes of the two perturbations are equal near pinch-off, which minimizes the merge time when the two modes are combined.

First we tune the amplitudes of the two perturbations such that both modes have an equal breakup length, as shown in Fig. 2.11. In the experiments presented in this chapter, the difference in breakup length before tuning was generally around one beat signal wavelength λ_0 . With the amplitudes matched, the phase of one of the perturbations is changed until the merge time is minimal. A good criterion to find the minimal merge time is when the two outer droplets, coming from the nodes of the beat signal, merge with the large droplet at the anti-node simultaneously.

2.8.2 Experiment with $We = 151$

In the procedure for the experiment with $We = 151$, we can choose the perturbation frequencies without knowing the jet velocity. This simplifies the implementation, at the cost of a larger merge time. We start by setting the amplitude of the electrical signal for driving the different modes to an equal voltage. Then we scanned the

Figure 2.11: The evolution of the perturbed continuous jet ($We = 107$), for $n = 2$. From top to bottom we show the jet perturbed with the χ_1 mode, the jet perturbed with the χ_2 mode, and a jet perturbed with the superposition thereof. The breakup lengths of the two modes are matched by tuning the actuation voltage of one of the distortions. The phase difference between χ_1 and χ_2 in the superposition of the distortion, is tuned to minimize the merge time. In these recordings, we show perturbed jets after the tuning, hence the separate perturbations from the upper two recordings have the same phase in the lower recording, where the two perturbations are applied simultaneously.

frequency domain to find two perturbations with a roughly equal breakup length, while obeying the $(n + 1)\chi_1 = n\chi_2$ condition exactly. With the frequencies set, the phase of one of the perturbations is changed until the merge time is minimal. A good criterion to find the minimal merge time is when the two outer droplets, coming from the nodes of the beat signal, merge with the large droplet at the anti-node simultaneously.

We determined the values of R_0 and u_0 to find the perturbation wavenumbers a posteriori. With the dispersion relation (equation (2.8)) we then found the growth rates. The growth rate of χ_2 is 10% higher than the growth rate of χ_1 in the experiment with $We = 151$. The different growth rates imply that the beat signal is not fully modulated during its growth, resulting in a larger merge time than in the case of equal growth rates.

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3

Destabilization of a microscopic monodisperse droplet train due to air drag *

Air drag on droplets in a droplet train can lead to axial and lateral spreading of the droplets eventually leading to coalescence and loss of monodispersity. A popular study by H.C. Lee (IBM J.Res. Dev. 21 (1),1977) has successfully modeled drag within a droplet train, however, the presented theory does not contain a physical description of the destabilization. Here we present experimental data to show a mechanisms responsible for destabilization. We use ultrafast high-speed imaging and advanced image processing to capture and track individual monodisperse droplets created by piezo-acoustic actuation of a microjet. Our results show that the change in droplet spacing within the droplet train is a direct result of air drag. The measured drag coefficients critically depend on the spacing between droplets, where the closely spaced droplets have a significantly lower drag coefficient, as expected. Surprisingly, despite this shielding effect and intricate details of the flow, the drag on the ensemble-averaged system is very well captured by Lees theory, until coalescence breaks down the description. Our results apply directly to inkjet printing, medical spray formation and 3D printing.

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3.1 Introduction

Droplet trains are used in inkjet printing, medical spray formation and spray painting [1], as well as 3D printing [2, 3]. In these processes, the droplets move collectively through air to reach their target [4, 5]. For optimum print quality, a thin deposition layer is crucial which implies that droplets should not coalesce in flight. However, air drag, at some point, inevitably leads to coalescence. For pulmonary drug delivery with medical sprays, prevention of coalescence is crucial since only a narrow size distribution of drug-loaded droplets are transported into the deepest alveoli of the lungs upon inhalation [6]. Drag-induced breakup of monodispersity leading to coalescence within sprays of electrically charged droplets in applications such as promising designs for green energy conversion [7, 8], paper fabrication [9] and diesel engines [10] may severely impact on the achieved efficiency of the devices. On the other hand, in the downscaling of novel lithography techniques, the formation of liquid tin microdroplets for the generation of laser-excited extreme ultraviolet (EUV) radiation [11, 12] rapid coalescence is crucial and air drag can promote this process. Thus, a quantitative understanding of the destabilization of a droplet train would make it possible to control the processes leading to coalescence, or how to prevent it.

Experiments on a small pack of four droplets, electrically separated from a droplets stream [13] show that the first droplet of the train, experiences the most drag as expected and is insensitive to trailing droplets until coalescence. The trailing droplets experience a drag reduction between 25 and 30 % and the authors speculate that the drag reduction depends on the droplet-to-droplet spacing, however they were unable to quantify this effect due to experimental limitations. Furthermore, in literature [14, 15], data is presented showing that drag on droplets in decelerating flows is different from steady-state drag, and it is observed that an increase in relative deceleration increases the departure from steady-state drag. The authors hypothesize that this effect is due to a change in the size of the recirculation zone in the flow trailing the droplet. Inertia of the surrounding gas may deform droplets [16, 17] but in our experiments on microdroplets, this effect is negligible below 11 m/s. Droplets come to a complete stop due to air drag and coalesce all together in turbulent flow [18]. The typical length scale ($X = r \frac{\rho_l}{\rho_g}$) is the stopping distance, at which the air drag starts to slow down the droplets.

The typical size of the droplets in the aforementioned applications is in the range of 10 to 100 μm and they travel with a speed of up to 10 m/s. Optically, these small length scales are only accessible with a high-resolution microscope and the corresponding timescales require the use of ultrafast high-speed imaging equipment [19]. Figure 3.1 shows high-speed imaging frames of a microscopic droplet train at different downstream distances. Figure 1A shows the breakup of a microjet in air. The pinch-off leads to the formation of droplets, including

Figure 3.1: The transition of a monodisperse and equidistant dropttrain to a coalesced droplet train. Panel A shows the jet, breaking up into two droplets that rapidly coalesce. This leads to panel B, where a monodisperse droplet train is displayed with a droplet diameter of $115 \pm 0.4 \mu\text{m}$. In panel C the droplets are not monodisperse anymore since coalescence has set in, the c marks coalesced droplets.

satellite droplets that subsequently merge with the preceding droplet to form a monodisperse droplet train, see Fig. 3.1B, where the droplets are captured 100 droplet diameters downstream of the nozzle. It is seen that all droplets have the same size and that they have equal droplet spacing. Another 100 droplet diameters downstream from the nozzle, Fig. 3.1C, the uniform droplet spacing is lost and breakup of monodispersity due to coalescence is observed. How the microscopic droplet train is destabilized is the subject of the present study. First, we will review the popular theory of Lee [20] to describe the ensemble-averaged behavior of the droplet train. We will then look into the motion of individual droplets within the droplet train to come to a description of how air drag affects the internal dynamics of the droplets in the train. We discuss the role of air drag for the potential applications and end with the conclusions.

3.2 Lee's theory

3.2.1 Boundary layer description

The effect of drag on a liquid jet is elegantly described in a paper by H.C. Lee [20], whose theory applies very well to droplet trains. The flow system is modeled assuming monodisperse droplets with equal spacing, which is then assumed to be a solid cylinder with the corresponding average density. The system is slowed down by the build up of a flow boundary layer around the cylinder, and mass conservation dictates that the cylinder should widen. Coalescence is not included in this model and lateral spreading out from the mean does not occur.

A schematic view of the model is illustrated in Fig.3.2. The density of the droplet train $\bar{\rho}$, is assumed to be the volume averaged density of the liquid (ρ_l) and

Figure 3.2: Schematic view of the approach of Lee.

the inter droplet gas (ρ_g), taking into account the difference between the nozzle or jet diameter a_{l0} and droplet radius R :

$$\bar{\rho} = \frac{\rho_g}{\rho_l} \frac{R^2}{a_{l0}^2}. \quad (3.1)$$

Lee derives an equation for the velocity at dimensionless length $\bar{z} = \frac{4}{Re} \frac{z}{a_{l0}}$, with the Reynolds number $Re = \frac{2\rho_g UR}{\mu_g}$, containing μ , the dynamic viscosity of the medium, U the droplet velocity and R the radius. For a typical water droplet in air ($U = 10$ m/s, $R = 50$ μm , $\mu_g = 1.8 \cdot 10^{-5}$ Pa·s, $\rho_g = 1.29$ kg/m³) $Re \approx 60$. In the model of Lee, the initial velocity v_{l0} is decreased with a factor $\bar{v}_l(\bar{z})$ depending on the dimensionless density and the outer limit of the boundary layer $\beta(\bar{z})$;

$$\bar{v}_l(\bar{z}) = \frac{v_l(\bar{z})}{v_{l0}} = \frac{1}{1 - \bar{\rho} \left[1 + \frac{1}{\beta(\bar{z})} - \frac{1}{2\beta^2(\bar{z})} (e^{2\beta(\bar{z})} - 1) \right]} \quad (3.2)$$

A value crucial in the work of Lee is the rate of momentum loss to the boundary layer, this is captured in the cumulative momentum loss $\beta(\bar{z})$ and this quantity is independent of the initial velocity or radius. Lee derives β by assuming no

slip flow between the droplets and the surrounding boundary layer which grows logarithmically. The value for β can be calculated by solving an integral defined by Lee numerically, (for example using the integral function in Matlab). The initial conditions of the droplet stream to compare experiments with Lee's theory, are the droplet velocity and the radius. In $\bar{p}$ the droplet radius, $R = \frac{3}{4\pi} \frac{Q}{f}$ is used which is set by the frequency f and the flow rate Q . In principle, the droplet radius also depends on the nozzle diameter but since we kept it constant, the flow rate and frequency is sufficient to calculate the droplet radius.

Figure 3.3: A: Drag coefficient on a single sphere compared to the drag coefficient derived from Lee. The initial velocity is 9.25 m/s , $D = 130 \text{ }\mu\text{m}$ and $Re=32$. In order to calculate the C_D for a single sphere, the velocity calculated with Lee at that distance is used, in order to keep the comparison clear. B: Drag reduction relative to single sphere drag for two spheres in Stokes flow as a function of droplet spacing [21].

Since Lee's theory describes the decrease in velocity by an analytical expression, the droplet deceleration (a) as a function of distance can be calculated directly from the velocity change. To do this, the time difference as a function of position needs to be calculated since it is not explicitly included in Lee's theory. Here we integrate cumulatively the reciprocal velocity over distance and then calculate the time difference. Knowing the deceleration and the velocity of the droplets, the drag coefficient C_D can be calculated (see Fig. 3.3) using the rewritten force balance of inertia ($\frac{4}{3}\pi R^3 \rho_l a$) and the drag force ($\frac{1}{2}\rho_g C_D A U^2$),

$$C_D = \frac{8R\rho_l a}{3\rho_g U^2}, \quad (3.3)$$

with A the cross section of the droplet. For single droplets in a low Re flow, the drag coefficient can be expressed analytically by $C_D = \frac{24}{Re}$ [22]. Based on this so-called Stokes drag, the drag reduction can be calculated for 2 spheres with a given spacing (s) [21]. The theoretical maximum lies around a 30 % drag reduction

for the second sphere. The result for Stokes flow and the possible drag reduction in Stokes flow's theory is plotted in Fig. 3.3, together with the drag coefficient derived from Lee's theory. The dimensionless spacing s/D is the distance between the centers of mass of two droplets, normalized by the droplet diameter.

3.3 Experimental Setup and Data Analysis

Figure 3.4 shows the setup used for our droplet train experiments. Distilled water is squeezed through a Microdrop AD-K-501 printhead with a nozzle diameter of $50\text{ }\mu\text{m}$. The jet is actuated by a piezo-acoustic element inside the printhead, triggered by a function generator. This setup is designed to study the full length of the jet near the nozzle at $(15D)$ from jet breakup onto the coalescence region at $(2000D)$.

Figure 3.4: Experimental setup suitable for measuring $100\text{ }\mu\text{m}$ diameter droplets in a train. The linear motor has a vertical range of 750 mm .

The method used here to transform a jet into a monodisperse train of droplets is through Rayleigh breakup, induced by piezo acoustic actuation. The actuation is done sinusoidally with a driving frequency f and an amplitude U_f and the frequency translates into a wavelength $\lambda = \frac{v_{j0}}{f}$, with v_{j0} the jet speed, as before. The jet is decaying due to instabilities and this process is described in terms of the dimensionless wavenumber $\chi = \frac{2\pi a_{j0}}{\lambda}$ where due to geometry, only frequencies corresponding to $\chi < 1$ lead to a growing instability [23]. The maximum growth rate occurs at resonance or at the 'Rayleigh frequency'. The dispersion relation for a liquid jet in the long wave description [24–26] ($\omega^2 = \frac{\sigma}{R^2\rho} (\frac{1}{2}\chi^4 - \chi^2)^{1/2}$) can be used to calculate the growth rate for every χ , where in this case viscosity is set to zero and gravity is neglected. Due to the unstable nature of jet breakup [27], not only the actuated wavelength influences the breakup, also the natural present in the system induces a spontaneous breakup once it exceeds the breakup

threshold. Experimentally, this is shown by Orme [28] with experiments in vacuum. In literature, mostly the actuation frequency is discussed but also the actuation amplitude has an influence on velocity of the droplets in the breakup. [29] So in order to study the coalescence and compare with the unactuated or spontaneous 'Rayleigh' breakup the actuation amplitude is kept as small as possible.

The droplet train is imaged with a Navitar 12 $\times$ zoom lens with a Navitar 2 $\times$ F-mount. This lens system allows for a good balance between zoom and field of view and is mounted in front of a Photron SA-X2 high speed camera. The frame rate of the camera was typically chosen to be a factor 2 higher than the frequency of the function generator. The frame size depends on the frame rate of the camera, which is typically 600 $\times$ 128 pixels. The camera is triggered by a pulse-delay generator. The droplets are illuminated by a high-intensity light source (Olympus ILP-2) projected through collimating optical elements to evenly illuminate the droplet train. The laboratory has climate control and is held constant at 20°C. The jet is shielded with covers to prevent any external air flow to influence the droplet train. The entire setup is placed on a vibration-free table.

Figure 3.5: The droplet edge is defined as the location of the inflection point of the intensity. The blue dot in the middle is the center of mass of the droplet. $R=50\text{ }\mu\text{m}$

From the image frames we deduce the position and the size of the droplets. For a typical measurement, we process 10,000 frames filled with droplets sized between 90 and 120 μm . As explained in [30] the optimal way to perform a high-speed imaging experiment and obtain precise information of droplets requires

three main considerations. First, the magnification must be sufficient to avoid spatial undersampling of the image while maintaining a large enough field of view to image as many droplets as possible in a single recording. Secondly, the image contrast should be maximized by adjusting the illumination intensity to cover the full dynamic range of the camera. The third criterion constitutes the minimization of motion blur by adjusting the exposure time. In all experiments, we use the lowest time possible with the camera (293 ns). We established that a droplet needs to cover at least 10 pixels to provide sufficient resolution and to have negligible motion blur at the highest velocities.

Since the area of interest lies not only in the center of the image where paraxial rays are detected but also near the edge of the image, where spherical aberrations can lead to errors, we use a distortion correction on all images [31]. An inflection point method [32] was applied as shown in to find the boundary of each droplet and to obtain its size and position, see Fig. 3.5.

Figure 3.6: (a) Large droplet spacing, intensity profiles do not overlap. (b) Small dot spacing, droplet intensities do overlap around $x = 0$. Overlapping intensities cause the inflection point to shift towards the droplet. In (d) for example; this means that the inflection point of the red droplet edge is found closer to its core than it really is. There is no inflection point shift with the largely spaced droplets as (c) shows.

From the droplet size and position, the following quantities are calculated: spacing, velocity and deceleration. While the inflection point method is built to accurately find the edges of single droplets, once droplets are close to each other interference of the diffraction patterns can potentially lead to an error in droplet

spacing, see Fig. 3.6. In order to reduce this effect, we evaluate the algorithm by varying the spacing of two droplets with a known spacing, see Fig. 3.7. It shows that we accurately measure drop spacing until 7 pixels distance. For smaller spacings, the inflection point method was only applied on the other half-circles with a slight decrease in the accuracy, in particular for the determination of the center of mass.

Figure 3.7: The difference between the real droplet spacing and the spacing according to the inflection point method, as a function of the spacing for an arbitrary droplet. For spacings larger than seven pixels, the difference is negligible.

The velocity is calculated based on the imaging frame rate and the displacement of the center of mass of the droplet. The droplet stays in the field of view for about 15 frames and individual droplets are tracked by accounting for the droplet size, eccentricity and intensity. The calculated velocity is then an average of these 15 measurements with an accuracy of 0.1%. The deceleration can be calculated through a fit to all velocity datapoints for each single droplet and the accuracy is 9%.

In an ideal situation, the droplets have a single size and a uniform spacing. In experiment, this can only be true until a certain level of accuracy, first, because of the noise present in the breakup process, and secondly, because of the pressure fluctuations due to the dispensing system, and finally, because of the acoustic characteristics of the printhead which induces a very small, but observable noise to the system. In order to characterize our system, we measured the velocity of approximately 7,500 droplets and we calculated for several wavenumbers the standard deviation of the initial velocity as shown in Fig. 3.8. We observe a small increase in the magnitude of the variations for higher velocities, as expected, but the standard deviation in the initial velocity is always smaller than 1% which is the cut-off value that is the accuracy level of the experiment. To assure convergence, we also plotted the standard deviation for subsets of the data (not shown here) and the results were identical.

Figure 3.8: The standard deviation of the initial velocity of the droplet train, as a function of the wavenumber

3.4 Results

3.4.1 Velocity

After jet breakup, the droplets interact with the surrounding air leading to a velocity decrease. Measurements of the velocity as a function of the downstream distance z were performed by translating the linear motor in the streamwise direction, see Fig. 3.9. Five datasets taken at different frequencies are shown and the experimental data agrees very well with Lee's theory, Eq. 3.2.

Figure 3.9: Mean droplet velocity as function of distance to printhead compared to Lee's prediction.

The experimental inputs are the initial velocity of the droplet train and the droplet size, used to calculate $\bar{\rho}$ from Eq. 3.1. By definition, the theory of Lee predicts a decreasing spacing for a stream of droplets, since the droplets slow down but the interdroplet time remains unchanged. The graphs terminate at the point where the first droplet coalescence is observed in experiment.

3.4.2 Droplet spacing

In order to understand the onset of coalescence, we measure the spacing of the droplets as a function of distance from the printhead. This results in a probability density function of the spacings shown in Fig. 3.10. It is clear that the spacing distribution dramatically widens, and that as the average velocity decreases as a function of distance, also the average spacing decreases.

Figure 3.10: Probability density function of the spacings for a 50 kHz actuated droplet train for different distances.

In line with what was observed in section 3.4.1, Fig. 3.11 shows how the spacing evolves of all droplets at different downstream distances. The result is plotted in a velocity-spacing parameter space where every color represents a different downstream distance from the printhead. Figure 3.11 clearly displays the change in spacing and velocity when the droplets move away from the printhead. With the simultaneous detection of droplet size, we determine whether or not droplets have coalesced and that allows us to filter these droplets from the data. Figure 3.11 therefore shows only the monodisperse single size droplet data for a clean comparison. This explains the cut-off for data at $s/D=1$. For large spacings ($z=70$ mm and $z=80$ mm) data is found for droplets even larger than 2 times the initial spacing.

This way of presenting the data gives a different perspective on the theory of Lee and it now becomes clear how well the theory is followed. Again, we observe the average spacing to decrease as the droplet train slows down. However the range of spacings grows rapidly until coalescence starts. This means the theory of Lee applies very well to the average droplet distribution but certainly not to an individual droplet. A model including a stochastic approach to the droplet velocity and spacing, still obeying Lee's theory for the averages would solve part of the

problem.

Figure 3.11: Distribution of spacings at different distances from the printhead. Coalesced droplets are not included in this plot, every dot represents a single uncoalesced droplet. $v_0=9.2$ m/s. The standard deviation of the velocity is σ_v .

Fig. 3.12 shows the standard deviation of the spacing and velocity of the droplet for different frequencies. Independent of the wavenumber, both increase as a function of downstream distance. The increase in σ_v is faster than linear with distance, which one expects when the initial velocity distribution is sustained. Thus the initial velocity distribution is not sustained and the deceleration is distributed non-uniformly over the individual droplets. We look more into detail to the deceleration of the droplets as a function of the spacing.

3.4.3 Drag coefficient

From a simple force balance in Eq. 3.3 we can calculate the drag coefficient (C_D) on the droplets from the measured deceleration. Figure 3.13 shows C_D for droplets just before coalescence sets in. The spread in C_D is a direct result of the error in the deceleration. To compensate for this spread, a large number of droplets is used. We observe a direct relationship between C_D and the spacing, revealing that a shielding effect exists for droplets at smaller spacing. For small spacings, the center of mass is more difficult to determine, as explained in section 3.3, leading to increased spread.

Figure 3.12: Left: The dimensionless spread in spacings as a function of the dimensionless distance to the printhead. The flowrate is 1 ml/min. Right: Spreading of droplet velocities as a function of the distance to the printhead.

Figure 3.13: Droplet drag coefficients. Compare with an individual droplet with $Re=50$, $C_D=0.48$. The increase in spread for spacings below 1.75 is due to a switch in method for measuring spacing, described in section 3.3.

Interpretation of the data is simplified by a smoothing function, see the red line in Fig. 3.13, and the result for many experiments is plotted in Fig. 3.14. All experiments show the same trend of the spacing dependent drag coefficient and they are grouped nicely for increasing frequency or wavenumber. For the smallest frequencies, the initial spacing is already large so this allows for a larger range of measurement in s/D before reaching coalescence at $s/D=1$. All these graphs follow a trend comparable to the result of Fig. 3.3B and we will use that relation to model a droplet stream in section 3.5.

3.4.4 Lateral dispersion

In Fig. 3.15 the spreading of the droplets in the lateral direction is plotted as a function of the downstream distance. It is obvious that the spreading in the lateral direction is very small compared to the total traveling distance, it's less than $0.1D$ while the droplets have travelled already $100D$, as the data acquired closest to the

Figure 3.14: Drag coefficient as a function of droplet spacing for different flowrates and wavenumbers. The curves are smoothed splines, fitted through the datasets.

nozzle at 10 mm shows. The plot in Fig. 3.15 shows the lateral spreading for a range of frequencies. For most frequencies, it takes more than 2000D to reach a σ_x larger than D. That lateral spreading is compared to Rayleigh breakup, which does not stand out among the results from actuated data. A reason for this could be that the lateral spreading is a consequence of the deceleration of the droplet train. The deceleration reduces the spacing, causing droplets to squeeze out a part of the air in between and to spread simultaneously. Lee accounts for this with the growth of $a_l(z)$ as shown in Fig. 3.2. In 250 mm, the expansion due to deceleration is approximately 30% of the diameter for our conditions, which is a significant part of the total lateral spreading. The results confirm that lateral spreading is not driving any of the effects observed in previous sections, because the effects are too small and partly a consequence of deceleration.

Figure 3.15: Lateral spreading of the droplets for different frequencies in the lateral direction for a larger span in downstream distances, compared with Rayleigh Breakup.

3.4.5 Coalescence

The detection of individual droplet sizes also allows for the analysis of coalescence behavior. By detecting the size of the droplets, it's possible to measure how many droplets have coalesced. Once coalescence starts, our algorithm only analyses the droplets which are nearly spherical, leaving out the rest. Figure 3.16 shows how the

Figure 3.16: Coalescence as a function of downstream distance for a 50 kHz actuated droplet train.

coalescence develops as a function of downstream distance. For small distances, a monodisperse peak is observed but once coalescence is initiated, almost all droplets coalesce, the droplet train grows increasingly polydisperse and the average droplet size keeps growing as shown in Fig. 3.17. We expect that the mechanism underlying this growth is a combination of the initial velocity distribution, the change in size, and the spacing-dependent drag. The latter lowers C_D for small spacings and increases C_D for larger spacing. Therefore coalescence occurs earlier and at a higher rate. Figure 3.17 shows that coalescence occurs later for actuated jets compared to Rayleigh Breakup, as expected. However, the slope is much larger for the actuated case.

3.5 Simulation

To capture our observations in a physical picture, we model a droplet train travelling through air with a simple dynamics model. The trajectory of droplets with an initial velocity distribution with a spread $\sigma = 0.5\%$ is calculated. The deceleration of the droplet train (a_{Lee}) is calculated with Lee's theory, as described in section 3.2.1. The droplets position x on time $t_{n+1} = t_n + \Delta t$ with time step $\Delta t = 0.01/f$ using

Figure 3.17: Averaged droplet size for multiple frequencies. A flow rate of 1 ml/min and a voltage of 50 V were used. Rayleigh breakup is done without any actuation, as a base for comparison.

the following equation:

$$x_{t+1}^i = \frac{1}{2} a_{Lee}(t_n) C_D^* \Delta t^2 + v_t^i \Delta t + x_t^i. \quad (3.4)$$

The crucial parameter here is the relative drag coefficient C_D^* . We include the spacing dependency of C_D using a theoretically derived function for the Stokes drag [21] on two rigid spheres as explained in section 3.2. The definition of C_D^* is:

$$C_D^* = \frac{C_{D(s)}}{C_{D(\langle s \rangle)}}, \quad (3.5)$$

where $C_{D(\langle s \rangle)}$ is the value of C_D^* corresponding to Lee. This definition allows for a direct comparison between a simulation with and without spacing dependent drag, by setting $C_D^*=1$ or let it vary. The parameter taking every individual droplet in the droplet train into account is $C_{D(s)}$, which depends on s and has a value drawn from Fig. 3.3B. C_D^* is thus a parameter allowing to take into account the effects observed in Fig. 3.13 without deviating from the Lee velocity profile shown in Fig. 3.9.

The results are shown in Fig. 3.18, where the top three images (A,B and C) are simulated with $C_D^*=1$ and the bottom three images (D,E and F) with C_D^* varying, with the spacing. Starting from the left with A and D, the average velocity for both simulations is identical, as expected from the model. Comparing B and E in the middle panels, the spacings as a function of velocity for different distances from the printhead are plotted, to reproduce Fig. 3.11. The bottom result with the spacing-dependent drag coefficient clearly resembles the experimental result better. This reconfirms that the initial velocity fluctuations alone cannot be responsible for the coalescence and drag effects observed in Fig. 3.11. Finally, the right panels in Fig. 3.18 show the spread in droplet spacing. The difference between both

graphs appears to be the difference between a linear increase and exponential increase. Comparison with Fig. 3.12 shows again that the introduction of a spacing-dependent C_D leads to numerical results with satisfactory agreement with the experiment.

Figure 3.18: Top three images: Simulation results of a droplet train with drag coefficient calculated with only Lee. Bottom three images: Simulation results of droplet train with drag coefficient calculated with Lee, including a spacing dependence or relative drag coefficient.

3.6 Discussion and Conclusions

We present an experimental study on drag in a monodisperse droplet train. The results show quantitative agreement with H.C. Lee's theory [20] a volume averaged droplet distribution for a range of droplet actuation wavenumbers. When we zoom in on the level of single droplets, a lateral dispersion in drag depending on the spacing towards the preceding droplet is observed. We interpret these results as a shielding effect where the droplet which has moved closer towards the preceding droplet, loses less kinetic energy to drag, as expected.

The dispersion of the droplet spacing, measured in the standard deviation, increases exponentially as a function of the downstream distance to the printhead.

The relative drag reduction of droplets in a droplet train is independent of the flow rate in our parameter range. The exponential behaviour of lateral dispersion can only be reproduced by introducing a spacing-dependent drag. Numerical simulation where C_D is based on the average velocity from Lee's theory and initial velocity variations simply translates into linearly (and therefore much slower) increasing standard deviation of the spacings. However, the average velocity decrease for a droplettrain is identical with or without a spacing-dependent drag description despite the growing spread in spacing.

No systematic pattern was observed in the spreading of droplets in the lateral direction, after $100D$, the maximum lateral spreading was $0.1D$ and the lateral dispersion σ_x reaches D after travelling approximately $2000D$. We hypothesize the spreading in lateral direction is caused by air flowing out from between the droplets, caused by the deceleration which reduces spacing.

Studying the exact moment of coalescence is a challenge due to the relative slow time scale of the process compared to the droplet velocity and the randomness of the event. It could provide some extra insight in the apparent off-axis movement of droplets, just before coalescence, as observed in Fig 3.1. This can be caused by a lubrication layer between the droplets but a small surface charge could also be responsible for this. Unfortunately, the inflection point method is unsuitable for analysis of droplets within seven pixels, making it unsuitable to study the coalescence in detail. A combination of high magnification and a controlled coalescence process, as presented in [12] could be a solution. Also the theoretical description of C_D^* can be improved. Numerical simulations, fully resolving the flow field in the wake of the droplets could be a first step in this direction. This will then also test the hypothesis of Temkin and Mehtaf [15] that a change in the recirculation zone behind the droplet is responsible for a change in the drag coefficient.

Thus; in addition to the work of Lee, we propose to include a spacing-dependent relative drag coefficient for individual droplets which adds to the understanding of droplet flight behaviour in many industrial applications like inkjet printing, 3D printing and medical spray formation. Also it refines coalescence models currently in use, since our results highlight the crucial role played by spacing-dependent drag in the forcing of the prevention of coalescence.

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4

On the capillary wave propagation in a cavity formed by high-speed impact of a microdroplet on a liquid pool *

Droplets impacting on a liquid pool and the subsequent bubble entrainment has been extensively studied for mm-sized droplets, such as rain droplets. Here we study droplet impact and bubble entrainment in an entirely new parameter regime. We continue the pioneering work of Oguz & Prosperetti, for microdroplets with a velocity beyond their terminal velocity. Cavity dynamics were found to be surface tension dominated, in analogy to the hydrostatically dominated cavity in the case of mm-sized droplets. We model the cavity with a 1D Rayleigh-Plesset type model and decouple the capillary wave travelling down. A capillary wave is formed at the crown tip, therefore we account for the formation time and height of the crown in predicting the cavity collapse. Analysis of numerical simulations in Gerris showed that the capillary wave has a wavelength corresponding to half the circumference of the impacting droplet. This sets the velocity of the capillary wave and combining it with the Rayleigh-Plesset type model, the collapse time and depth of the cavity can be estimated. Both high-speed imaging experiments and numerical simulations validate the simple empirical model.

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4.1 Introduction

Droplet impact on a liquid pool is ubiquitous both in nature and in technology. It has been studied both for fundamental insight and for applications ranging from inkjet printing, diesel engines and painting to agriculture, soil erosion and meteorology, starting with the pioneering work of Worthington [1]. Considerable progress has been made both in experimental observations and in numerical modelling the various impact phenomena; [2–6].

A striking element of drop impact is bubble entrainment [7–10]. One of the mechanisms to entrain a bubble is to first form a cavity upon impact, then to pinch off a bubble at the bottom of the cavity as it closes. This mechanism is schematically drawn in Fig. 4.1a-d and it is termed regular bubble entrainment. In nature it is known for the characteristic noise produced by impacting rain droplets caused by the bubble oscillation after the pinch-off [11]. For mm-sized droplets, bubble entrainment has been studied experimentally by Pumphrey and Elmore [12]. They measured a distinct region, shown in Fig. 4.1e, in the droplet radius (R) versus droplet velocity (U) parameter space where regular bubbles were entrained. Oguz and Prosperetti [7] used their data to derive a scaling law, for the upper and lower boundaries in terms of the Weber number $We = \frac{2\rho RU^2}{\sigma}$ and the Froude number $Fr = \frac{U^2}{2gR}$, where ρ is the liquid density, σ the liquid surface tension and g the gravitational acceleration. The Fr vs We parameter space maps directly onto the R versus U parameter space if a single liquid is considered.

Oguz and Prosperetti [7] model the cavity dynamics using hydrostatic pressure as the driving force and assuming a hemispherical shape. For the lower entrainment boundary, two time scales are compared to determine if entrainment takes place. First, a calculation of the time it takes for the axisymmetric wave, travelling with speed $c_{\min} = \left(\frac{4g\sigma}{\rho}\right)^{1/4}$, see Fig.4.1c, close the cavity at the bottom is performed. This travel time is then compared to the time at which the bottom of the cavity has reached its maximum depth and starts to retract. As long as the cavity bottom is moving downward at the time the wave hits itself, a bubble can be entrained. The upper bound is reached at a Weber number for which the bottom of the cavity is so flat, that the wave cannot entrain a bubble anymore. The flatness of the cavity bottom is assumed to be related to the spreading of the impacting droplet, calculated from balancing droplet inertia and surface tension. When the inertia of the droplets is so high that the spreading radius reaches the radius of the hydrostatic cavity, the cavity is too large and therefore too flat for bubbles to be entrained. The scaling laws of Oguz and Prosperetti [7] are supported by boundary integral simulations.

Figure 4.1: a) Droplet with velocity U and radius R impacting a deep pool. b) Formation of the crown with height h_{crown} and the cavity with depth h , bottom velocity $\dot{h}$ and radius r . c) Wave travelling downward over the cavity wall with speed c d) Regular bubble entrainment with a bubble radius R_{bubble} . e) Data of Pumphrey and Elmore [12] plotted with the bubble entrainment region predicted by Oguz and Prosperetti [7].

Further details on bubble entrainment have been elucidated in recent years. It has been shown that bubbles are also entrained during the initial contact of the droplet with the pool [8]. A small oblate-to-prolate oscillation of the impacting droplet can generate entrainment of a bubble even larger than the original droplet, an entirely different mechanism from regular bubble entrainment [13, 14]. Elmore [15] repeated the experiments of Pumphrey and Elmore [12] after a decade of improvements in recording technology and compared them directly to simulations. Details of their BI simulation showed that the movement of the cavity bottom is reversed at a different time compared to the experiment. The mismatch was hypothesized to be due to the absence of vorticity in the simulation. Chen [16] extended the work of Oguz and Prosperetti by including viscous dissipation on the wave propagation, while Deng [17] studied bubble entrainment for various liquids and showed that the maximum bubble size decreases with increasing viscosity. Other than drop impact, following bubble coalescence, capillary waves travelling over the bubble surface are also known to entrap a bubble [18, 19].

Closure of the cavity was modeled by Gekle et al. [20, 21] and Bergmann et al. [22, 23] using a cylindrical 2D version of the Rayleigh-Plesset equation. The cavity depth at the bubble pinch-off and time were estimated and validated with experiments and numerical simulations. The large cavity size helped to describe the systems by hydrostatic pressure alone, in the absence of surface tension. Ray et al. [24, 25] modeled drop impact and bubble entrainment with the cylindrical Rayleigh-Plesset equation on of drop impact. Another attempt to model the cavity

combines potential flow and the unsteady Bernoulli equation for cavity formation at sufficiently high We and Fr numbers [26–28]. The authors found that the time to reach the maximum cavity size scales as $t \sim R^{5/2}$. This result was obtained by neglecting both the start of the cavity formation the surface tension effects on the cavity. An identical result was obtained before based on an energy balance [29–31].

4.1.1 Outline

The present work is on the impact of fast microdroplets. Specifically, it deals with water droplets of a size much smaller than a millimeter and velocities above the terminal velocity of falling rain drops. While the physical arguments for closure of a hydrostatically driven cavity are based on inertia and gravity, at smaller length scales, the closure of the cavity is driven by capillarity. We show using numerical simulations and experiments that regular bubble entrainment also occurs in our parameter range. Using the scaling laws different than those of Oguz and Prosperetti [7]. We model the cavity dynamics and the wave travelling down to close the cavity. This allowed for a prediction of the cavity closing time and its depth upon closure. After explaining the setup and the numerical approach, experimental results are presented in section 4.4 which are compared with numerical simulations in Gerris. The simulations are used to find entrainment and we measure the depth of the cavity and the collapse time. We model the system by decomposing the cavity and the wave closing the cavity in section 4.5. Bubble entrainment is then studied in section 4.6 and the entrainment region found in the numerical simulations is compared to the experimental results. The chapter ends with conclusions and an outlook.

4.2 Experiments

Single microdroplets with a velocity beyond the terminal velocity under ambient pressure are created by employing an electrical droplet deflector to extract single droplets from a droplet stream every few milliseconds [32]. The setup is sketched in Fig. 5.2. A continuous droplet train is created by actuating a liquid jet emerging from a glass micropipette (Microdrop AD K-501) by piezo acoustic actuation driven by a 40 MHz Wavetek waveform generator (Model 195) and amplified 50 times with a Falco Systems DC-5 Mhz high-voltage amplifier WMA-500. The liquid is pumped through the nozzle with a calibrated Shimadzu LC-20AD HPLC pump. The microdroplets impact onto a deep liquid pool, formed by a glass container. Imaging of the droplet impact (not shown here) was performed with a Photron SA-X2 camera and the region of interest was illuminated with an

Figure 4.2: Sketch of the experimental setup. A liquid jet is actuated piezo-acoustically to generate a monodisperse stream of droplets. Every few milliseconds a droplet is not charged to pass undisturbed through the electric field generated by the deflection plates into the liquid pool.

Olympus ILP-2 Xenon light source. The typical recording frame rate is 100 kfps with an exposure time of 293 ns.

The nozzle size sets the droplet size and velocity. Our nozzle diameters were 25, 50 and 70 μm and by using a superposition of two waves from chapter 2 [33] we were able to create droplets with a diameter ranging from 47 to 334 μm and a velocity ranging from 6 m/s to 27 m/s. For even smaller droplet sizes (and lower velocities), we relied on a droplet on demand (DOD) droplet generation technique. With this technique, single droplets with a velocity between 2 m/s and 12 m/s and a diameter between 25 μm and 70 μm can be created at a frequency of 100 Hz. Before each impact experiment the droplet size and velocity was measured from the high-speed recordings.

4.3 Numerical Simulations

The parameter space is limited by a finite size of the experimental dataset and we therefore also perform numerical simulations to complement our experiments. Numerical simulations were performed in Gerris, a volume of fluid (VOF) code developed by Popinet [34]. This code is known as a reliable multiphase flow solver with an adaptable grid size. It has been validated with various theoretical standard

cases and applied to many multiphase problems. Examples include droplet impact [9, 35], atomization [36] and wave generation after a landslide [37]. Many more examples exist on the source code website of Gerris [38]. We use an axisymmetric version of Gerris for our simulations of droplet impact on a deep pool. The refinement level of the simulations is 11 which means that the maximum amount of grid points for the simulation box is 2^{11} . About 30 percent of the simulations were performed at a refinement level of 10 in order to reduce the computation time. The width of the simulation box width is 20 times the droplet diameter and the depth of the pool is 12 times the diameter to exclude effects of a finite pool depth. The boundary condition in air and in the liquid is a constant pressure. At the water-air interface, the maximum grid refinement is enforced to minimize errors in surface reconstruction. In all simulations, gravity was included. A total 121 simulations of droplets impacting onto a pool were performed.

4.4 Results

We now compare the results of two experiments with the numerical simulations. Figure 4.3 shows how an impacting droplet ($U=20.2$ m/s and $D=113\text{ }\mu\text{m}$) forms a cavity below the surface. In Fig 4.3b the cavity has grown and a wave is clearly visible. The wave travels down the cavity wall and in Fig 4.3c we observe wave dispersion due to differences in wave speed and wavelength. The cavity has now reached its maximum size. Panels d), e) and f) show how the waves interfere at the bottom, which is followed by the collapse of the cavity without entraining a bubble. The agreement between numerical simulations and experiment, in the formation and growth of the cavity, the wave formation and wave propagation and in the interference at the bottom of the cavity was found to be excellent.

Figure 4.4 shows another impact ($U=6.6$ m/s, and $D=321\text{ }\mu\text{m}$). The cavity dynamics is comparable to the previous experiments; we observe a hemispherical cavity which grows at a decreasing growth rate. Here, the interference of the waves at the bottom of the cavity leads to a different phenomenon. The coupling between the wave and the dynamics of the cavity is such that a bubble is pinched off. The match between the numerical simulations and experiments is moderately good. The cavity angle is smaller in the experiment, which can be a result of eccentricity of the impacting droplet or another small mismatch in the initial conditions at impact.

Figure 4.5 shows the results from our numerical simulations and we show in green whether we observed regular bubble entrainment. The experimental data is also plotted in Fig. 4.5. The results show that there is a clear region where regular bubble entrainment occurs. Regular bubble entrainment for mm-sized drops is modeled with only hydrostatic pressure considered and the minimum phase speed

Figure 4.3: Captured images of a formed cavity, compared with numerical simulation results. $U=20.2$ m/s and $D=113$ μm .

Figure 4.4: Captured images of a formed cavity and the pinch off of a bubble, compared with numerical simulation results. $U=6.6$ m/s and $D=321$ μm

Figure 4.5: Parameterspace with all numerical and experimental data obtained for this work, compared with data of Pumphrey and Elmore [12] and scaling from Oguz and Prosperetti [7] for rain drops. Numerical simulations results reveal the bubble entrainment region highlighted in green. The plot also shows the bubble entrainment by the second to last wave in purple.

for gravity-capillary waves $c_{\min} = \left(\frac{4g\sigma}{\rho}\right)^{1/4}$ [7], but in this case, the waves will have a capillary wave speed.

4.5 Model of Cavity Formation and Closure

We model the experimental and numerical results of cavity closure by decoupling the cavity and the wave dynamics. Figure 4.1a schematically shows the droplet before impact resulting in a cavity at depth $h=R$ as shown in Fig. 4.1b. In the same figure we also define the other elements of the response of the system to the impact, namely the height of the crown h_{crown} , the cavity width r and the vertical velocity $\dot{h}$. Figure 4.6 shows the time dependence of the depth $h(t)$ and width $r(t)$ of the cavity for a numerical simulation ($D=80 \mu\text{m}$ and $U=20 \text{ m/s}$). Fig. 4.6A confirms the phenomenological description from section 4.4 where we saw that the depth of the cavity grows at a reduced growth rate where in the end the bottom of the cavity is driven by capillary waves and their interferences. The wave is clearly visible at $t=90 \mu\text{s}$, (Fig. 4.6B) propagating to the cavity bottom where it arrives at $t=250 \mu\text{s}$. The capillary wave period is also indicated in Fig. 4.6A.

Figure 4.6: A: Depth of the cavity, extracted from numerical simulations. The period T of the last wave before the cavity closes is indicated. B: Radius of the cavity measured at $h=0$, extracted from numerical simulations. Both are extracted for $R=40 \mu\text{m}$ and $U=20 \text{ m/s}$

4.5.1 Dynamics of the cavity

The cavity dynamics is modeled with a Rayleigh-Plesset type equation for spherical bubbles [39] containing inertia and surface tension of the cavity.

$$h \frac{d^2 h}{dt^2} + \frac{3}{2} \left(\frac{dh}{dt} \right)^2 + 2 \frac{\sigma}{\rho h} = 0 \quad (4.1)$$

The rationale behind this is that the cavity behaves as a growing spherical bubble. In order to use the model with initial conditions fitting the numerical simulations, we neglect the initial formation of the cavity and the crown, comparable to the approach of Bisighini et al. [26]. We start to follow the cavity at the point where $h = R$ at $t = t_{RP}$, the situation that is depicted in Fig 4.1b. The difference with earlier models in the literature, is the influence of surface tension on the formation of the cavity, which will play a role due to the small size of the cavity. In Fig. 4.7, both the initial velocity and the initial dimensionless time are plotted. Fig. 4.7A shows that the cavity reaches $h=R$ at approximately the same dimensionless time, $\frac{U t_{RP}}{R} = 3.5$. We now set the initial conditions for wave propagation with an offset t_{RP} after impact. Fig. 4.7B shows the velocity of the cavity bottom at $h=R$ where we find a linear correspondence with slope 0.56. For lower velocities there is some deviation from the straight line but for most experiments, the velocity of the cavity bottom nicely follows the linear fit. This means that we can now use this as an initial condition in the Rayleigh-Plesset model (Eq. 4.1). This leads to the definition of a Weber number for the cavity: $We_{cav} = \frac{\rho 2R (0.56U)^2}{\sigma}$. We_{cav} is connected to the formation of the cavity, when the initial phase of droplet impact with deformation of the droplet and rapid deceleration has passed. These simple

Figure 4.7: A: Time at which $h=R$, at which we initiate the Rayleigh-Plesset model. We need this time to synchronise the starting time of the capillary wave with the starting of the cavity. B: Velocity of the cavity bottom at $h=R$, We fit this with $\dot{h}=0.56 \cdot U$ to find the initial cavity velocity.

relations were the rationale behind modelling the cavity from the arbitrary point $h=R$. The initial phase of droplet deformation and momentum transfer from the droplet to the pool is highly dissipative. From a kinetic energy point of view, approximately 69 % is dissipated in droplet deformation and crown formation, the remaining energy (31%) goes into the formation of the cavity. This is in line with the results from literature, where Leng [28] states that 28% of the kinetic energy is used for the formation of the cavity and with Bishighini et al. [26] that uses a relation of $\dot{h}=0.44 \cdot U$ (or 19%) extracted from experimental results for rain drops.

4.5.2 Capillary wave starting position

The initialization of the wave is a crucial condition in modelling the cavity collapse. We initiate the wave in our model at the moment when the crown reaches its maximum height. Figure 4.8A shows the contours of a numerical simulation. In the contours, the wave can be discerned and the reversal of the travelling direction coincides (± 0.05 ms) with the crown height reaching a maximum. This point is emphasised in Fig. 4.8B, where we plot the crown height as a function of time, together with a sketch of the wavefront. Before reaching the maximum height, the wave is already travelling downward on the crown but with a speed lower than the velocity of the crown tip, resulting in a net upward speed. At some point during the deceleration of the crown, the downward speed of the wave will be higher than the upward velocity of the tip, meaning that the net movement of the wave becomes downward. In first order, we approximate this process by initiating the wave at the point when the crown reaches its maximum and giving it the full capillary wave speed from the start. In Fig. 4.9, the maximum crown height and the time at which this maximum height occurs is plotted versus the Weber number.

Figure 4.8: A: A set of contours after impact. The wave starts to travel downward at the moment the crown reaches its maximum height. $R=161 \mu\text{m}$, $U=12.6 \text{ m/s}$ and Δt between the contours is $27.8 \mu\text{s}$. B: Crown height and a sketch of the wave front as a function of time. The crown reaches its maximum at $t=0.32 \text{ ms}$.

Figure 4.9: A: Maximum height of the crown plotted as a function of We . The fit is $\frac{h_{\text{crown}}}{R}=0.69+0.0038We$. B: Time of maximum height of the crown as a function of We . The fit is $t_{\text{crown}} \frac{U}{R}=0.011We+1.1$

A linear fit accurately describes both the height ($\frac{h_{\text{crown}}}{R}=0.0038We+0.69$) and the time ($t_{\text{crown}} \frac{U}{R}=0.011We+1.1$) at which the crown reaches this maximum. In both cases, for low Weber numbers (<100), the crown does not reach sufficient height to be discernible from waves on the surface of the pool, causing a deviation from the trend.

4.5.3 Capillary wave speed

Now that the cavity dynamics and the resulting distance to be traveled by the wave is known, the next objective is to find the speed of the wave arriving. In order to characterize the wave propagation, we measure the period T of the last wave before the cavity closes from $h(t)$, see Fig. 4.6. The period we take the time between

Figure 4.10: The wave starts to travel downward at the moment the crown reaches its maximum height. The frequency is measured from all numerical simulations where this was possible and converted into a wavelength, applying the dispersion relation for capillary-gravity waves. There are multiple datapoints for every radius, representing different velocities.

the minimum in h and the local minimum before the upward motion of the cavity. Not all simulations resulted in accurate determination of the period of the wave, which limits the amount of data. From the period of this wave (or its frequency ω) we calculate the wavelength λ using the dispersion relation for capillary-gravity waves [40].

$$\frac{2\pi}{T} = \omega = \left(gk + \frac{\sigma k^3}{\rho} \right)^{1/2} \quad (4.2)$$

Equation 4.2 relates the period T to the wavenumber $k = \frac{2\pi}{\lambda}$ which allows for a calculation of the phase speed when either the wavenumber or the wavelength is known. This calculation gives the wavelength as a function of the droplet radius, plotted in Fig. 4.10. The graph shows a clear relation between the droplet radius and the wavelength, namely $\lambda \sim \pi R$ which corresponds to a wavelength of half the circumference of the droplet,

$$k = \frac{2\pi}{\lambda} = \frac{2\pi}{\pi R} = \frac{2}{R}. \quad (4.3)$$

With this result, we can describe the speed of the slowest capillary wave in the cavity as a function of the droplet radius [40],

$$c = \frac{\omega}{k} = \left(\frac{g}{k} + \frac{\sigma k}{\rho} \right)^{1/2} = \left(\frac{gR}{2} + \frac{2\sigma}{\rho R} \right)^{1/2}. \quad (4.4)$$

We can rationalize this result by realizing that the wavelength corresponds to the largest length scale in the problem and this length serves as a cut-off in the

Figure 4.11: A: The speed of the capillary wave propagating along the cavity wall, for a droplet with $R=160 \mu\text{m}$ and $U=8 \text{ m/s}$. B: The average wave speed, measured for droplet radii 20, 40, 80, 160 and 300 μm with impact velocities of 33, 15, 13, 8 and 5 m/s respectively.

wavelength. Since the speed is inversely proportional to the wavelength in a capillary wave ($c \approx \left(\frac{\sigma k}{\rho}\right)^{1/2}$ when gravity is neglected), this corresponds again to the slowest wave exited in the system. Note the clear distinction with rain drops, for which the slowest wave speed in the system, always corresponds to $c_{\min} = \left(\frac{4g\sigma}{\rho}\right)^{1/4}$ or the corresponding wavelength $\lambda = 2\pi \left(\frac{\sigma}{\rho g}\right)^{1/2}$, which is independent of the droplet size.

We can reconfirm these results by tracking the wave in the numerical simulations and measuring its speed. Figure 4.11A shows the speed of the capillary wave down the cavity wall, normalized to the capillary wave speed from Eq. (4.4). We track the wave using the algorithm presented by Ding [41] and Zhang [18] in an adapted version to allow for the cavity growth. The tracking of the wave starts when it is clearly discernible at the crown tip. It ends when the wave starts to interfere with itself. The initial speed is larger than the capillary speed. This is caused by the limited thickness of the crown since Eq. (4.4) is valid under the assumption of a deep pool. For a liquid sheet with thickness e , the Taylor-Culick velocity ($V_{TC} = \frac{2\sigma}{\rho e}$) [42, 43] is larger than the capillary speed when the sheet has a thickness $e < R$. Figure 4.11B shows the wave speed measured in five individual simulations for different droplet diameters. Thus, these results reconfirm our earlier conclusion that the wave speed can be directly calculated from the droplet size alone, independent of the impact velocity.

Figure 4.12: Comparison of numerical results with the Rayleigh-Plesset description for four different Weber number. The dashed line indicates the cavity and the dotted line indicates the distance traveled by the capillary wave. When the two meet, the dot represents the point at which the cavity collapses according to the model. These points are used for the next two graphs.

4.5.4 Comparison of the model with numerical simulations

We compare the model, with numerical simulations in Fig. 4.12. Capillary wave propagation is modeled as $x_{\text{cap}}(t) = c \cdot (t + 3.5 \frac{R}{U} - t_{\text{crown}}) - h_{\text{crown}}$. The depth of the cavity is calculated with Eq. (4.1) using an ODE-solver in Matlab. For all cases plotted, the Rayleigh-Plesset description follows the cavity expansion very well. Only for later times, when the wave reaches the bottom of the cavity, the agreement is less satisfactory due to wavesuperposition. The steep upward motion of the cavity bottom, present in all numerical simulations results, is a jet caused by the focussing of the capillary wave. We find qualitative agreement for the time and depth where and when focussing of the waves occurs. Figure 4.13 compares the results measured from numerical simulations with the solution from our simple empirical model for all simulations and the results agree very well. A prediction of the collapse depth seems to be difficult, as seen in Fig. 4.12 and this is attributed to the amplitude of the capillary waves which is not included in the model.

Figure 4.13: A: Maximal height from the Rayleigh-Plesset description calculated for a set of Weber-numbers, compared with results from numerical simulations. B: Time of the maximum height from the Rayleigh-Plesset description calculated for a set of Weber-numbers, compared with results from numerical simulations.

Figure 4.14: A: The bubble size scaled with the droplet size plotted as a function of the Weber number for a single droplet radius. B: The bubble size scaled with the droplet size plotted as a function of the Bond number for a range of droplet diameters. The experimental data for millimeter sized droplets is taken from [13].

4.6 Bubble Entrapment

Now that we understand the dynamics of the cavity and propagation of the capillary wave, we turn back to the problem of bubble entrapment. Figure 4.14A shows the bubble size for numerical simulations with the same droplet radius ($R=40 \mu\text{m}$) plotted as a function of We . We observe that the bubble size (R_{bubble}) for constant R varies with the impact velocity and takes a maximum in the middle of the entrainment region. This observation, combined with the observation from Fig. 4.5 that the entrainment region has approximately the same width independent of the droplet size suggests that a wave phase mechanism is active, that is independent of the droplet size and the resulting cavity size.

Figure 4.14B shows the relative bubble size as a function of the Bond number

Figure 4.15: Experimental image of bubble, just before pinch off by the second to last wave and just after, $\Delta t = 10 \mu s$. $U = 26.1$ m/s and $R = 62 \mu m$

$Bo = \frac{\rho g R^2}{\sigma}$. The difference in the relative size of the entrained bubble between microdroplets and millimeter-sized droplets is evident and a direct consequence of size alone.

We combine both experimental and numerical data on bubble entrainment in Fig. 4.5. It turns out that the results from numerical simulations and experiment match very well. The empirical model that was developed for the cavity and wave dynamics, should in principle be able to predict bubble entrainment when the following ingredients are included. First, the amplitude of the capillary wave, which is related to the remaining energy not dissipated in the wave. Secondly the direction of motion of the cavity bottom which in principle we know from the Rayleigh-Plesset description. And finally the phase of the wave at the moment of cavity collapse.

A few remarks can be made on the basis of the results presented in Fig. 4.5. The experimental bubble entrainment region for radii around $150 \mu m$ is larger than the numerical one, which is probably caused by oblate-to-prolate deformations of the droplet, as a result of air drag. Secondly, for the set of datapoints around $25 \mu m$ in the bubble entrainment region, the velocity is calculated from the flow rate. The actual velocity can be up to 10% lower, due to airdrag.

In some experiments a bubble entrainment phenomenon was encountered that was different from the regular bubble entrainment mechanism. Figure 4.15 plots the image of the cavity, just before pinch-off and just after. We attribute this to phenomenon to the second to last wave bubble entrainment mechanism which we confirm both in numerical simulations and in experiment, see Fig. 4.5. Typically these phenomena were observed for droplets with impact velocities higher than regular bubble entrainment for droplets with the same size. The amplitude of the second to last wave should be large enough to entrain a bubble. This phenomenon is close related to bubble entrainment is bubble bursting [44]. Both problems consist of a cavity which closes due to capillary waves travelling downward and resulting in the subsequently entrainment of a bubble. The authors find two distinct bubble entrainment regions in their numerical simulations. One where the wave

which closes the cavity entraps a bubble, c.f. in regular bubble entrainment and a second region, where the bubble is entrapped with the second to last wave.

4.7 Conclusion and Outlook

In this chapter we have presented our experimental and numerical modelling results of microdroplet impact on a deep pool. We have extended the work of Oguz and Prosperetti to the regime beyond the terminal velocity under ambient conditions for microdroplets. The results show that a region exists in the velocity-radius parameter space where the movement of the cavity bottom and the incoming capillary wave constructively entrain a bubble.

The dynamics of the cavity is well described by a spherical 1D Rayleigh-Plesset model including surface tension of the cavity. Without the surface tension term in the Rayleigh-Plesset model, all curves in Fig. 4.12 will collapse on a single curve, describing purely inertial cavity dynamics. The model is initiated at a depth $h=R$, after the initial phase of the impact. The cavity is closed by a capillary wave which has a speed set only by the droplet size alone. In order to predict the closure of the cavity, the crown formation duration and the resulting crown height need to be taken into account. Both vary approximately linearly as a function of the Weber number allowing for simple implementation in our empirical wave model. The model predicts the closing time of the cavity and is in good agreement with our numerical simulations. The agreement for the closing depth is moderately well and is the result primarily due to the absence of the unknown capillary wave amplitude in our model. Next to regular bubble entrainment, we also found a region in which the second to last wave is leading to entrainment of a bubble with smaller size, this is also confirmed numerically.

The open question is how the lower and upper bounds of the bubble entrainment region above the terminal velocity scales. Clearly, the scaling of Oguz and Prosperetti is not valid anymore because gravity is negligible. The physics behind the boundaries may still apply. The lower boundary is set by equating the time scale of the cavity with the time it takes for the capillary wave to hit the bottom. And the upper boundary is set by the angle of the cavity. When the amplitude of the second to last wave is large enough, the same mechanism may apply for second to last wave entrainment.

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5

Oblique drop impact onto a deep liquid pool^{*} †

Oblique impact of drops on a solid or liquid surface is frequently observed in nature. Most studies on drop impact and splashing, however, focus on perpendicular impact. Here, we study oblique impact onto a deep liquid pool, where we quantify the splashing threshold, maximal cavity dimensions and cavity collapse by high-speed imaging above and below the water surface. Three different impact regimes are identified: smooth deposition onto the pool, splashing in the direction of impact only, and splashing in all directions. We provide scaling arguments that delineate these regimes by accounting for drop impact angle and Weber number. The angle of the axis of the cavity created below the water surface follows the impact angle of the drop independent of the Weber number, while cavity depth and its displacement with respect to the impact position depend on the Weber number. Weber number dependency of both the cavity depth and displacement is modeled using an energy argument.

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5.1 Introduction

In nature, oblique drop impact is ubiquitous. It is for example encountered in rain drop impact [1], where it triggers air entrainment [2] and aerosol generation [3], both drive the global gas/liquid exchange. In agriculture, e.g. in pesticides crop spraying [4], oblique drop impact and the subsequent splashing and droplet rebound [5] is important. In industrial applications oblique impact occurs in e.g. metal spray deposition [6] and direct fuel injection internal combustion engines [7]. In many of these applications, splashing is an unwanted side effect after impact; it decreases deposition efficiency and may lead to widespread contamination.

While drop impact and splashing is a topic widely studied (see e.g. [8–22]), surprisingly only few papers (e.g. [23–26]) deal with non-perpendicular or oblique impact. Drop splashing upon perpendicular impact has been studied on a solid substrate [12, 13], a thin liquid film [14–16] and a deep liquid pool [17–22]. In the latter case, a cavity is formed under the water surface [2]. Collapse of this cavity [27, 28] may lead to the pinch-off of small droplets [1, 29]. These droplets emerge from a high-speed microjet, and can reach velocities higher than the impact velocity [30]. The main parameter that governs splashing is the Weber number, $We = \frac{\rho D U^2}{\sigma}$ with drop diameter D , drop velocity U , liquid surface tension σ and liquid density ρ . Also important is the Reynolds number, $Re = \frac{UD}{\nu}$, with ν the kinematic viscosity. A standard splashing threshold has the form of $We^{1/2} Re^{1/4} = K$ [8, 13] where K is a constant depending on surrounding pressure [31], on the thickness of the liquid layer and on surface roughness when impacting on a rigid substrate.

To study oblique impact, experiments on dry tilted plates [23–26] and on a moving plate [32] were performed. Bird et al. [32] showed that by moving the substrate, the velocity of the ejecta sheet and therefore the splashing threshold changes. Only a few studies [33–37] focus on oblique impact on wetted surfaces and thin liquid films. Gao and Li [37] quantify a splashing threshold for impact on a moving thin liquid film, but due to experimental complications, such as liquid inertia, this approach is not feasible for a deep liquid pool.

In numerical simulations, oblique impact on a wetted substrate [38–40] shows a transition from crown splashing to single-sided splashing. In addition, crown formation and cavity formation [40] are studied. Due to the aforementioned experimental limitations these simulations lack validation. For oblique drop impact on a wetted or dry surface, there is a clear influence of the impact angle, but to the best of our knowledge, no experiments of oblique drop impact on a liquid pool have been reported in literature until now.

Here, we present an experimental study of oblique drop impact onto a quiescent deep liquid pool. We provide details of our experimental method in section 5.2 and discuss a typical result for the impact and splashing phenomena in section 5.3.1.

The angle of impact is varied systematically to quantify the splashing threshold and we present a model to explain our observations in section 5.4. We also quantify cavity formation and a scaling law for the cavity formation is presented in section 5.5.

5.2 Experimental Methods

To study oblique drop impact for a wide range of Weber numbers and impact angles onto a deep liquid pool, two steps are important: (i) creation of a single drop and (ii) rotation of the drop generator to obtain oblique impact. To create single drops a method previously described in [41] is used, which we adopt for oblique impact. For clarity, we briefly describe this method to isolate single drops from a stream of drops here, which is schematically drawn in Fig. 5.2.

Drops are generated by pumping (Shimadzu LC-20AD HPLC pump) demineralized water ($<0.1\%$ of ammonia added for conductivity, negligible effects on surface tension) through a micropipette (Microdrop AD K-501). The continuous jet breaks up into drops by applying a piezo acoustic pressure on the jet, which transforms the jet into a stream of monodisperse drops with equal velocity. Drops of size of $100 \pm 10 \mu\text{m}$ and velocity up to 25 m/s are generated. For these settings We ranges from 40 to 1056. This method allows for the creation of drops with a wide range of velocities and with a large enough drop spacing to prevent disturbances to the impact site due to earlier drop impact.

The drop train is directed through a ring-shaped charging electrode, where a periodic high-voltage pulse charges all passing drops except one every 10 milliseconds. Next, the drops pass another region with high electric field ($E \geq 100 \text{ kV/m}$) in between two deflection plates. The electric field separates the charged drops from the uncharged ones, and allows for the uncharged drops to continue straight and impact onto the pool. A glass container is filled to the top with water to minimize the influence of a disturbing meniscus at the container wall, which may limit the optical imaging quality. The charged drops are caught further on their path and then disposed. To create an oblique impact, the micropipette, charge electrode and deflection plates are tilted. With this method impact angles up to 80° from perpendicular can be obtained.

Figure 5.2 shows a sketch of drop velocity U and impact angle α . These parameters are varied systematically, while keeping the drop diameter D constant. Drop impact is recorded in a side view using high-speed imaging (Photron SA-X2, operated at frame rates up to 100 kfps) and backlight illumination (Olympus ILP-2). From the recordings, we extract D , U and α ($\tan\alpha = \left(\frac{U_{\parallel}}{U_{\perp}}\right)$, where $U_{\parallel}$ and $U_{\perp}$ are the tangential and perpendicular component of U , respectively) using customized image-processing analysis. The impact velocity is measured from the

Figure 5.1: a) Sketch of the experimental setup. A continuous stream of drops is created by piezoelectric-actuation of a microjet. A charging electrode charges the drops which are subsequently deflected by the deflection plates. Every 10 milliseconds a single drop is left uncharged and can be separated from the stream. By tilting the micropipette, charging electrode and deflection plates, the impact angle α of the drops onto the pool can be varied. b) Drop diameter D , velocity U and impact angle α are controlled by the experiment and cavity displacement d , height h are measured, which results in cavity angle α_c and the characteristic length of cavity L . Next to that, the angle of the jetted droplet out of the cavity α_j is measured. The shape of the splash and the collapse of the cavity are recorded using two separate high-speed recordings above or below the water surface, respectively.

penultimate frames just before impact to minimize the influence of air drag. We also characterize the direction of the splash and measure the angle of the jetted droplet emerging from the cavity α_j .

The dynamics of the cavity is characterized simultaneously with the impact event by collecting two separate recordings for each impact condition, one above and one below the water surface, in order to compensate for the different optical focal depth. From the cavity recordings, we measure the maximum cavity displacement d , maximum height h and define the cavity angle α_c , with $\tan\alpha_c = \left(\frac{d}{h}\right)$.

Data extracted from six combined recordings (above and below the water surface) is averaged for each impact condition. The average values allow us to relate parameters of the cavity with the impact conditions with a standard deviation of less than 5 percent. To quantify the splashing threshold above the water surface and the cavity formation and collapse below the water surface, we analyzed 1147 individual drop impacts.

5.3 Results and Interpretation

5.3.1 Typical features of oblique drop impact

Figure 5.2 shows the time series of a typical experiment with $We = 674$ and $\alpha = 28^\circ$. Qualitatively, the different stages of impact are comparable to perpendicular impact, with some differences for oblique impact, which we will now discuss.

Panel (a) shows a snapshot 0.02 ms after the drop impacts. The formation of a hemispherical cavity has started below the water surface and an asymmetric splash ejecting small droplets is visible above the water surface. At $t = 0.1$ ms, see panel (b), both crown and cavity are growing. In contrast to normal impact the droplets only detach on a single side. Just below the water surface, a wave crest is observed. At $t = 0.28$ ms (panel (c)) the wave crest is highlighted with an arrow while it travels downward along the cavity wall. Here it is clearly seen that the cavity is asymmetric. At this point in time, the cavity is still growing while surface tension causes the crown to retract and the rim to grow thicker. Panel (d) ($t = 0.66$ ms) shows the cavity at its deepest point, where the maximal cavity depth and displacement are reached. Now, the closure of the cavity starts and the capillary wave crest collides at the bottom of the cavity. During the collision of these waves, an air bubble can be entrapped [2]. The tip at the bottom of the cavity in (d) marks this air bubble just before pinch-off. The tip will retract quickly due to the high local curvature. The series of subsequent image frames suggest that the jetted droplet which is visible above water in panel (e) at $t = 0.77$ ms, originates from the rapid tip retraction. The droplets jetted out of the cavity reach velocities of the same order as the main drop impact velocity. Below the water (panel (e)) a

tiny bubble is clearly visible. At $t = 1.02$ ms (panel (f)) the cavity is completely collapsed, marked by the emergence of a thick Worthington jet [42].

5.4 Splashing Threshold

After drop impact three different phenomena can be observed above the water surface, as illustrated in Fig. 5.4. First, the drop can smoothly coalesce with the liquid pool. This behavior is identified as deposition. Secondly, a crown can be formed which destabilizes on a single side of the drop, resulting in the ejection of satellite droplets at the tips of the crown on this side of the drop. The direction of splashing corresponds to the direction of impact. This behavior is denoted as single-sided splashing. The final phenomenon observed is the ejection of small satellite droplets from the crown in all directions, which is indicated as omni-directional splashing. We quantify these phenomena in a phase diagram of α and We illustrated by three distinct regions, see Fig. 5.4. For a single value of α the impact behavior can change from deposition to single-sided splashing to omni-directional splashing by increasing We . Similarly, for a single value of We the impact behavior can change from omni-directional splashing to single-sided splashing to deposition for increasing α .

We now provide a scaling argument to explain these observations. To find the splashing threshold for oblique impact, we aim to describe the velocity of the crown in terms of U , D , α and We . We start by considering perpendicular (i.e. vertical) impact. A schematic view of the drop during splashing is shown in Fig. 5.4. In this case, the flow into the crown is distributed symmetrically. Once the entire drop has impacted, we can assume that the entire drop volume is proportional to the volume of the crown, which gives (per unit time)

$$D^2 U \sim e D V, \quad (5.1)$$

where e is the thickness of the crown at its origin. In line with a model for splashing of the fast ejecta on a thin liquid film [15], we assume that in order to obtain splashing upon impact on a liquid pool the velocity of the crown V has to be larger than the Taylor Culick velocity $V_{TC} \sim \sqrt{\frac{\sigma}{\rho e}}$. Thoroddsen [21] showed experimentally that $e \sim \sqrt{vt}$, consistent with dimensional analysis. Using $t \sim \frac{D}{U}$ in the expression for e and (5.1) to express V we find the splashing criterion

$$\frac{V}{V_{TC}} \sim We^{1/2} Re^{1/4} > K, \quad (5.2)$$

where K is the critical number for splashing [8, 9, 43]. The numerical value of K depends on specific experimental conditions, such as the flow profile inside the

Figure 5.2: Time series of a drop impacting onto the pool with a Weber number $We = 674$ and impact angle $\alpha = 28^\circ$, where $t=0$ marks the moment the drop makes first contact with the pool. Each image is composed of two independent recordings, one recording above the water to visualize the splash and the other one below the water surface to visualize the cavity, in order to compensate for the different depth of focus. Both recordings are however taken under identical experimental conditions. The images were then combined to show the entire impact process in a single time series. (a) The impacting drop causes a splash that fragments into droplets. Here, the splash is formed on a single side only. (b) A crown develops and remains more pronounced on one side. The cavity has a hemispherical form and is growing. (c) The splashing is finished and the cavity still grows. An arrow points to the wave crest, which travels downwards along the cavity surface, see [28]. The oblique impact leads to an asymmetry of the cavity. (d) The capillary wave focuses off-center at the bottom of the cavity. (e) Collapse of the wave leads to the entrainment of a bubble and retraction of the jet leads to the ejection of tiny droplets during closure of the cavity. The arrows indicate the position of the entrained bubble and the droplets. (f) A Worthington jet is formed during closure of the cavity.

Figure 5.3: a) Three types of impact behavior are observed above the water surface. From left to right: deposition (green), single-sided splashing (yellow) and omni-directional splashing (purple). b) A phase diagram of the impact behavior as a function of We and impact angle α . The color codes correspond to Fig. a): upward green triangles represent deposition, yellow squares represent single-sided splashing and downward purple triangles represent omni-directional splashing. The solid and the dashed line are derived from Eqn. (5.4) with $c = 0.33$, where the solid line represents the splashing threshold from deposition to single-sided splashing (Eqn. (5.4) with minus sign) and the dashed line indicates the splashing threshold from single-sided splashing to omni-directional splashing (Eqn. (5.4) with plus sign).

Figure 5.4: Schematic illustration of the mass distribution in the crown. Volume 1 indicates the part of the original drop that flows into crown volume 3. Volume 2 represents the part of the original drop that flows into crown volume 4. Volume 1 and 3, and 2 and 4 are identical due to mass conservation. a) For normal drop impact the volume of the original drop is distributed evenly over the crown, such that volume 1 and 2, and thus 3 and 4 are identical. b) For oblique impact, $U_{\parallel}$ will cause a shift (solid line) in drop position with respect to the normal impact (dashed line). This shift reduces volume 1 and increases volume 2 by an amount of $\pm \rho D U_{\perp} U_{\parallel} t$ (see text). As a result, the crown becomes asymmetrical.

crown [15]. Previous studies reported $K \approx 54$ for a rigid surface [9], $K \approx 160$ for a thin liquid film [14] and $K \approx 90$ for a deep liquid pool [20]. For our experiments we determine for perpendicular impact ($\alpha \simeq 0^\circ$) a critical Weber number for splashing of about $We_s \approx 400$ which leads to $K \approx 130$.

For oblique impact the mass is no longer symmetrically distributed over the crown: the mass flow into the crown increases on one side of the drop and decreases on the other side of the drop (Fig. 5.4). This unequal mass distribution originates from the contribution of the tangential component of the impact velocity $U_{\parallel} = U \cdot \sin \alpha$. On one side of the drop, $U_{\parallel}$ leads to an increased mass flow, while on the other side the mass flow is decreased. The added/reduced mass flow rate into the crown scales as $\pm \rho D U_{\perp} U_{\parallel} t$ respectively, where $U_{\perp} = U \cdot \cos \alpha$ sets the speed with which the drop is moving down into the pool.

To account for this additional mass flow, we can rewrite (5.1) as

$$D^2 U_{\perp} \pm D U_{\perp} U_{\parallel} t \sim e DV. \quad (5.3)$$

The (critical) velocity for crown splashing now depends both on $U_{\perp}$ and $U_{\parallel}$. From (5.3) it follows that $U_{\perp} \left[1 \pm \frac{U_{\parallel} t}{D} \right] \sim \frac{V_c}{D}$. We observe in experiment that splashing starts at the same time scale as the drop fully impacts the liquid pool $t \sim c \frac{D}{U_{\perp}}$, which leads to a splashing threshold:

$$\frac{V}{V_{TC}} \sim We^{1/2} Re^{1/4} \frac{(\cos \alpha_s)^{5/4}}{c^{1/4}} [1 \pm c \tan \alpha_s] > K, \quad (5.4)$$

where c is a fitting parameter. In (5.4), we use $K = 130$ (as determined for perpendicular impact) to find the splashing threshold as a function of We and α_s . These considerations suggest that there are two transitions in the phase space of Fig 5.4: (i) the solid line defines where V_{TC} is reached on a single side of the drop, which indicates the transition from deposition to single-sided splashing. (ii) The dashed line defines where V_{TC} is reached for splashing in all directions, which indicates the transition from single-sided splashing to omni-directional splashing. In Fig. 5.4 we plot these transitions and we find good agreement with the experimental data using $c \approx 0.33$.

5.5 Cavity Formation

From our recordings we extract the angle α_c (as defined in fig 5.2) and the dimensions of the cavity (h and d). In Fig. 5.5 the cavity angle α_c is plotted as a function of impact angle α . For small impact angles α_c equals the impact angle. This is expected since the momentum of the drop is transferred to the cavity in the direction of impact. However, for impact angles larger than $\alpha \gtrsim 30^\circ$, α_c gets increasingly

Figure 5.5: Cavity angle α_c as a function of the impact angle α . The solid line has slope of unity. For increasing α the distinctness of d and h decreases. Therefore, the error in measuring α_c increases with α and is estimated to be 10% of α .

smaller as compared to α . The measurement error of α_c is estimated to be 10 % of α ; however, this does not explain the observed decrease. A possible explanation for the deviation in proportionality of α_c with α for larger impact angles could come from additional wave drag [44] for drops moving close to parallel to the surface (i.e. large α). Due to the build-up of waves in the direction tangential to the surface, part of the energy can be dissipated into those waves for larger impact angles which leads to a smaller increase in cavity angle. We also analyzed the cavity angle as a function of We , but no significant dependency was found (data not shown).

We now present data on the maximum cavity depth and maximum displacement. The maximum cavity depth h and displacement d is plotted in Fig. 5.6 and Fig. 5.7 as a function of We based on the normal impact velocity $We_{\perp} = \frac{\rho D U_{\perp}^2}{\sigma}$ and the tangential velocity $We_{\parallel} = \frac{\rho D U_{\parallel}^2}{\sigma}$, respectively. We observe that h scales with $We_{\perp}^{1/2}$ while d scales with $We_{\parallel}^{1/2}$.

The observed scaling can be explained from energy conservation. We consider that the kinetic energy of the impacting drop is proportional to the additional surface energy to create the cavity, given by

$$\rho D^3 U^2 \sim \sigma L^2, \quad (5.5)$$

where L is the characteristic length of the cavity, as shown in Fig. 5.2. For oblique impact experiments, L depends on both h and d since the maximum depth of the cavity is displaced by d as a consequence of the tangential component of the

Figure 5.6: Double logarithmic plot of the maximum cavity depth h as a function of the Weber number, based on the normal impact velocity $We_{\perp}$. The dashed line corresponds to Eqn. (5.6) with slope 1/2 and a prefactor of 0.18.

Figure 5.7: Double logarithmic plot of the maximum horizontal cavity displacement d as a function of the Weber number based on the tangential impact velocity $We_{\parallel}$. The dashed line corresponds to Eqn. (5.7) with slope 1/2 and a factor of 0.09.

impact velocity, resulting in $L \sim \sqrt{h^2 + d^2} \sim h\sqrt{1 + \tan^2 \alpha}$. Here we have assumed $\tan \alpha = \frac{d}{h}$, which will introduce a small error for $\alpha \gtrsim 30^\circ$, as seen in Fig. 5.5.

From (5.5) we find $\frac{L}{D} \sim We^{1/2}$. Using the geometrical relation for L leads to $\frac{h}{D} \sim We^{1/2} \cos \alpha$, and hence

$$\frac{h}{D} \sim We_{\perp}^{1/2}. \quad (5.6)$$

Fig. 5.6 shows (5.6) together with the experimentally determined maximum cavity depth, where we used a prefactor of 0.18. For the maximum cavity displacement we find $\frac{d}{D} \sim \frac{h}{D} \tan \alpha \sim We^{1/2} \sin \alpha$, which gives

$$\frac{d}{D} \sim We_{\parallel}^{1/2}. \quad (5.7)$$

We plot in Fig. 5.7 the experimentally determined maximum cavity displacement with slope 1/2 and a prefactor of 0.09 which provided the best fit to the experimental data.

5.6 Discussion on Cavity Collapse

The collapse of a cavity is sometimes accompanied by the jetting of a droplet out of the cavity. In this subsection we aim to quantify the direction of the jetting behavior. Figure 5.2 depicts the angle α_j at which the droplet is jetted out of the cavity. Not every impact leads to the observation of a jetted droplet, which could mean that no droplets were pinched off from the jet, or that droplets remain trapped inside the cavity. Given the small droplet size and the limited depth of field of our imaging system, the ejected droplet could also have moved out of the optical focal plane.

Another observation is that the trajectory of the droplet is curved when it moves out of the cavity, as shown in Fig. 5.6. This may be an indication that the initial α_j , directly after pinch-off from the jet, is different from the α_j measured above the closing cavity, where it may be hindered by a disturbing airflow. To better quantify the jet direction, we change our focus to the cavity collapse and deduce the retraction angle α_r , as depicted in Fig. 5.6, where we assume that the onset of retraction of the cavity sets the initial direction of the jetted drop. From a series of frames α_r is measured where the first frame was taken when the cavity is at its maximum size. In the next frames we follow the direction of cavity collapse by tracking this point to give the retraction angle.

As shown in Fig. 5.6, α_j and α_r are indeed similar. Over the entire range, α_j is slightly smaller than α_r , which is probably due to the influence of a disturbing airflow, as discussed above. α_r follows α up to about 25° . For $\alpha > 25^\circ$, α_r flattens and reaches a maximum of 30° . The deviation from α occurs around the same α

Figure 5.8: a) The trajectory of two jetted droplets for $We = 371$ and $\alpha = 0.3^\circ$ shows a deflection when the droplets leave the cavity. This deflection is indicated by the arrow. b) Snapshot of the cavity at its maximal dimensions (top), and snapshot of the cavity approximately 14 microseconds later (bottom), where the collapse and tip retraction have started. The red arrows indicate the direction in which the cavity starts to collapse, used as a measure for the direction of the jetted droplet out of the cavity.

Figure 5.9: (a) Angle of the droplet jetted out of the cavity α_j (upward black triangles, each triangle corresponds to a single measurement) and retraction angle of the cavity α_r (downward red triangles, each triangle corresponds to the average taken over six measurements below and above the water surface) as a function of the impact angle α . (b) Schematic view of the cavity angle and the retraction angle. When a drop impacts obliquely, one expects a cavity consisting of the white area, due to symmetry. However, since surface tension prohibits the existence of the sharp corner, the hatched area is pulled outward. Therefore, an asymmetric cavity forms resulting in a retraction angle which is smaller compared to the cavity angle.

for both α_c and α_r . However, the build-up of drag waves in front of the impacting drop is not enough to account for the flattening observed for α_r . We speculate that this saturation is caused by a maximum angle that the cavity can make with the surface. For perpendicular impact, the cavity is symmetric and hemispherical, which is energetically most favorable. For oblique impact, the cavity is no longer hemispherical but one side of the cavity wall starts to move inwards (see fig 5.6). At the other side the cavity opening grows due to surface tension. The sharp edge rapidly retracts by surface tension, much more than at the opposite side of the cavity which is shown in Fig. 5.6 by the hatched area. This effect breaks the symmetry and therefore poses a limit on the maximum value of α_r .

5.7 Discussion and Conclusions

We present an experimental study of oblique drop impact onto a quiescent deep liquid pool. We performed quantitative experiments where oblique drops impact onto a deep liquid pool for a wide range of We and impact angles α and analyzed the splashing behavior, the cavity formation and the cavity collapse.

In analogy to previous studies e.g. [8, 9, 15, 43], we found that the crown velocity has to be larger than the Taylor-Culick velocity to obtain splashing. For oblique drop impact this crown velocity is influenced by the tangential velocity of the impacting drop, leading to an asymmetry in the crown and giving rise to an asymmetry in the splashing threshold [32]. In contrast to the model presented in [32] where oblique drop impact on a dry substrate was studied, we cannot describe our data by simply adding/subtracting the tangential velocity to the crown velocity. Instead, we assume the tangential velocity influences the amount of mass squeezed into the crown. We quantify this effect using scaling arguments and derived a model that is consistent with our measurements for a wide range of We and α . Thus, the model gives valuable insight into splashing occurrence and direction.

For small impact angle α the cavity angle directly equals the impact angle and does not depend on We , for large α a decrease in α_c with respect to α was observed. The magnitude of the cavity displacement depends on the tangential Weber number and the cavity depth depends on the perpendicular Weber number. The scarce numerical simulations on oblique impact [40] confirm this We -dependence of the cavity displacement. However, since [40] described drop impact on a thin liquid film, no information on the cavity depth is available.

In conclusion, our study provides the first quantitative overview of the events following oblique impact onto a deep quiescent liquid pool. The results allow to predict under what impact velocity and in what direction drops can splash after impact. In our experiment, data from a single measurement plane is obtained. It

would be interesting to obtain the full 3D profile above and below the water surface using holographic microscopy [45] or by extended numerical simulations.

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6

Bouncing droplets: A classroom experiment to visualize wave-particle duality on the macroscopic level * †

This chapter brings a recently discovered macroscopic phenomenon with wave-particle characteristics into the classroom. The system consists of a liquid droplet levitating over a vertically shaken liquid pool. The droplets allow visualization of a wave-particle system in a directly observable way. We show how to interpret this macroscopic phenomenon and how to set up and carry out this experiment. A class of students performed single slit diffraction experiments with droplets. By scoring individual droplet trajectories students find a diffraction pattern. This pilot application in the classroom shows that students can study and discuss the wave-particle nature of the bouncing droplet experiment. The experiment therefore provides a useful opportunity to show wave-particle behavior on the macroscopic level.

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6.1 Introduction

Over the past decades, quantum mechanics has found its way from laboratories and universities into high school classrooms. Even though it has not lost its gleam of mystery and being difficult, it has become part of the standard curriculum in high school physics education [1]. This poses the problem for teachers of explaining to their students phenomena that are not even fully understood by scientists. While the demonstration of appropriate experiments is a useful way to introduce and explain physics to students, few such experiments are available to help understand quantum mechanics. Many of the experiments conducted in the history of quantum mechanics are only feasible in a dedicated laboratory, so teachers commonly resort to the use of applets. Recently, Couder and coworkers demonstrated that millimeter-size oil droplets bouncing on a vibrating fluid bath can exhibit quantum-like behavior at a scale visible to the human eye. The experiment, working principle, and all of the physics behind it have been explained in detail [2–4]. We establish that this experiment can be used in a classroom setting to visualize a wave-particle system, and certain other aspects of quantum mechanics. We begin by introducing the experiment and explaining the connection between the experiment and wave-particle duality, indicating both similarities but also limitations. We then describe the experimental setup, followed by a description of a pilot held with high school students. We conclude by presenting the results of this pilot, highlighting that this experiment can be executed in a class-room setting, by students.

6.2 Walkers

When a drop of liquid is dropped on a bath of the same liquid, one naturally expects the droplet to merge with the bulk of the liquid. However, when the bath of liquid is shaken in the direction perpendicular to the liquid surface, something surprising can happen: the droplet survives, bouncing over the surface of the liquid bath for a prolonged time. The vertical motion of the bath creates and maintains a thin layer of air between the droplet and the surface of the bath, preventing their coalescence. Something even more extraordinary happens when the droplet size, liquid type and shaking parameters are chosen just right: the bouncing droplet then creates waves at the surface of the bath, interacts with these waves, and is propelled horizontally across the surface [5]. Such a droplet/wave combination is called a ‘walker’, a term that will be used throughout this chapter. An example of walkers is shown in Figure 6.1. For a walker to exist, a coupling must be established between the shaking frequency of the bath and the droplet’s bounce time such that the droplet always hits the sloping side of the wave. This causes the droplet to be propelled over the surface with speed of ≈ 1 cm/s [6].

Figure 6.1: An image taken during the single-slit experiments, showing a walker taken from supplementary movie 1. Under the liquid surface, the structure forming the slit is visible. More details about this setup can be found in figure 6.3 and surrounding text.

Walkers can only occur when the liquid bath is vibrated at an acceleration close to the Faraday transition: When at rest, the liquid bath will have a flat, hydrostatic surface. Any disturbance of the flat interface will be counteracted by gravity and surface tension. However, if we shake the liquid bath following

$$\gamma = \gamma_m \cos(2\pi f_0 t), \quad (6.1)$$

in the direction perpendicular to the fluid surface, the liquid is accelerated up and down. In Equation 6.1 f_0 is the oscillation frequency in Hz, t is the time in seconds, and γ_m is the acceleration of the bath. For certain accelerations of the liquid bath (note that the acceleration is directly related to the frequency and amplitude of the harmonic motion), the liquid surface destabilizes and enters a state of parametric resonance. The standing waves that develop on the liquid surface are called Faraday waves, see Figure 6.2 for a typical example of Faraday waves. An everyday example of Faraday waves can be observed in a glass of wine, which is made to 'sing' by rubbing a wet finger over its rim. The acceleration of the liquid bath above which Faraday waves appear is called the Faraday transition acceleration. At accelerations above this threshold, Faraday waves form on the entire surface. As it is the acceleration that determines if Faraday waves develop, we will describe the oscillatory motion of the liquid bath in terms of the acceleration, rather than by its frequency and amplitude.

The surface of the bath will be flat as long as it is oscillating at accelerations just *below* the transition acceleration. However, the impact of a bouncing droplet can be sufficient to trigger the formation of waves in the direct vicinity of the droplet. The developing waves are characterized by a wavelength, which can be

Figure 6.2: An image showing Faraday waves.

calculated from the dispersion relation [7]

$$(2\pi f_0)^2 = \left\{ gk + \left(\frac{\sigma}{\rho} \right) k^3 \right\} \tanh(kh_0), \quad (6.2)$$

where k is the wavenumber from which the wavelength $\lambda = \frac{2\pi}{k}$ can be calculated, g is gravitational acceleration, σ and ρ are the surface tension and density of the liquid, respectively, and h_0 is the depth of the liquid bath. The current experiment uses soybean oil, which has a surface tension of $\sigma = 27$ mN/m, and a density of $\rho = 0.92$ g/ml [8]. Keeping in mind that the frequency of Faraday waves f_F is half that of the driving oscillation f [6], $f = 50$ Hz converts to a wavelength of $\lambda = 4.45$ mm, which will be used later in the design of the experimental setup.

With an acceleration below the transition value and without agitation of the surface, the surface will remain flat. However, as the droplet is bouncing repeatedly on the interface, it will trigger the formation of Faraday waves at regular intervals (at the moment of impact), creating a pattern of waves. Eddi et al. [7] simulated the wave patterns on the surface, created by the subsequent bounces. Their results show that already after a few bounces the wavelengths close to the Faraday wavelength are dominant, while the others quickly dissipate. Moreover, they show that the wave pattern acts as a “memory” containing the information from previous bounces [9].

6.3 Relation with Quantum Mechanics

The macroscopic droplet and the wave that supports it show some interesting behavior, with resemblance to wave-particle duality in quantum mechanics. When barriers are placed in or nearby the path of the walker, the walker behaves as a combination of a wave and a particle. Although the system is macroscopic and hence not quantum-physical, the similarities can be exploited to explain that systems can show the kind of duality.

As an example, the walker system can be used to demonstrate wave diffraction in a single slit experiment (see figure 6.1): when a bouncing droplet is send through

a slit, it will change its direction upon passing through the slit. When the direction at which multiple walker leave the slit is measured and added, a diffraction pattern is measured that would be similar as one obtained when plane waves pass through the slit.

Whereas this observed behavior is clearly an interesting example of wave-particle duality, care should be taken in showing the limits of this particular analogy with quantum mechanics. First, as mentioned above, this is a macroscopic system where the velocity and position of the walkers is known at all time, in obvious contrast with quantum mechanics where accurate simultaneous measurement of position and momentum is excluded. Second, the role of the wave that supports the droplet is obviously different from a quantum mechanical wave. In quantum mechanics, the wave represents a probability amplitude, meaning that its squared amplitude denotes the probability density of the particle being at a certain position. In the case of the walker the waves form a kind of supporting structure for the bouncing droplet. This means that the wave shows behavior similar to the pilot wave interpretation of the de Broglie-Bohm theory [10, 11]. This interpretation considers the motion of quantum particles to be guided by a single associated pilot wave, described by the Schrödinger equation. This is in contrast with the Copenhagen interpretation, in which a measurement collapses the wave function to force the system to take one state explicitly. In the Bohm interpretation, the guiding wave is already present prior to any measurement. The randomness of the outcome of experiments in pilot-wave theory is therefore caused by the initial conditions and not by the dynamics.

The latter is true for the walker system: small variations in the initial conditions of the system cause the probabilistic behavior of the walkers. Because quantum physics education teaches the Copenhagen interpretation rather than the de Broglie-Bohm interpretation, care should be taken not to overstretch the walker analogy. A strong emphasis on the relation of the walker system with quantum mechanics, could lead to an implicit acceptance of the pilot wave interpretation. Although these two important differences exist between walkers and quantum mechanical experiments, walkers are suitable to display the basic principles of wave particle duality, and hence can function as an analogy for this phenomenon.

6.4 Experimental Design

An experimental setup was constructed following the description by Couder et al. [5]. A sketch and photograph of the setup are shown in figure 6.3. An aluminum basin is mounted on top of a linear actuator. The actuator shakes the basin vertically with adjustable acceleration and frequency. The design used in this current chapter

is suited for the single-slit diffraction experiment. Different designs are possible to perform other experiments [12]. Previous research used silicon oil, which can

Figure 6.3: Top and side view of the oil bath for bouncing droplets. Droplets are generated in the lower left-hand side. The arrow indicates a possible droplet trajectory. After the droplet passes through the opening, students record the section in which the droplet arrives. Each section covers 10 degrees. The side view shows that the liquid bath has a uniform depth, with a barrier and ridge along the edges covered by a thin layer of oil.

be ordered at a specified viscosity. One drawback is that silicon oil is expensive and notoriously difficult to remove from any surface. Soybean oil was therefore used as a cheap and widely available alternative, which can be obtained from the local supermarket and can be removed using water and a mild detergent. All fluid properties were mentioned in section 2.1. The design of the basin is crucial, because the depth of the oil bath, the type of oil, the size of the droplets, the shaking frequency and the slit-width are all interrelated. In the present chapter, the basin, milled from a single piece of aluminum, had internal dimensions of $80 \times 80 \times 15 \text{ mm}^3$. A small ridge with a width of 5 mm was made around the edges, with the same height as the barriers (10 mm). The two aluminum barriers were detachable

and placed diagonally, as shown in figure 6.3. These barriers were 4 mm wide and 10mm high. As it is advised to use a slit with a width of about 3 times the Faraday wavelength [5], the distance d between the barriers was $d = 18$ mm. The basin was filled with 70 ml of oil to obtain a liquid depth of 11 mm. The barriers and ridge were thus covered by 1 mm of oil, required to reflect the walkers away from the walls to avoid collision of the droplets and the wall. The rigidity of the basin was found to be important: flexing and bending of the basin during oscillation affects the local amplitude of oscillation, marring the experiment. The basin also needs to be properly leveled, as any tilt in the basin floor will create a non-uniform liquid depth. The basin was connected to an actuator (TiraVib 50301) that could be set to the desired oscillation frequency and acceleration. The values used were $50 \text{ Hz} \pm 2 \text{ Hz}$, with an acceleration of $4 - 5g$. The range of frequencies in this experiment, $20 - 100 \text{ Hz}$, allows for replacing the professional shaker and electronics by a shaker constructed from audio speakers and amplifiers. This has recently been done successfully by a group of high school students, with assistance from the authors. Small droplets were generated by swiftly dipping a thin tip, e.g. a needle or a toothpick, in and out of the liquid. Droplets of ≈ 1 mm were found to be stable and display the desired walker behavior for the parameters used. Supplementary movie 2 shows a high speed recording of this process.

6.4.1 Setting the right parameters

Bouncing droplets are easily formed for a wide range of droplet sizes and liquids, bath frequencies and accelerations. The desired walkers are only formed in a much narrower parameter space.

The acceleration was set just below the threshold for onset of Faraday instability as shown in figure 6.4. This allows for more droplet sizes behaving as walkers. The value of the acceleration had to be adjusted regularly during the experiment to maintain good results.

6.5 Application in the Classroom

After the setup was constructed and tested, a group of ten high-school students (ages ranging from 16 to 18 years old) was invited to perform the single slit experiment. An introductory lesson was given to introduce or remind the students of the main concepts involved. In this lesson wave-particle duality was introduced by showing diffraction grid patterns and discussing single and double slit experiments. Single-particle diffraction patterns were shown and explained. Excellent applets are available to support this introduction [14]. One week later, the experimental setup was prepared for the students. The experiment was executed in a laboratory space

Figure 6.4: Parameter space for acceleration and droplet size D , indicating various behavior, including bouncing (B), period doubling to bouncing (PDB) and period doubling to chaos (PDC), intermittent behaviour (Int) and the desired walker regime (W). The gray right border indicated with F indicates the onset of Faraday waves at higher acceleration. Results are shown here for silicon oil ($\mu = 50$ mPa·s) and a bath oscillation frequency of 50 Hz. Reproduced with permission from [13] .

at the university. After a brief introduction, the students were divided into five pairs; each pair worked for 10-15 minutes on the experiment. One student in each pair focused on generating bouncing droplets by dipping a needle into the surface. The other student checked the trajectories of the walkers and wrote down the section in which the walkers arrived after passing through the single slit, see figure 6.3. After 5 minutes, they switched tasks. The students were observed by one of the authors, who supervised the experiment. The supervisor also took notes on the students' behavior, and recorded remarks made by the students. Directly after the experiment, each pair of students was interviewed. The interviews were videotaped, and transcribed later on to score the students' understanding [15]. After the experiments, the students were assembled in a classroom. The observations of droplet trajectories by the individual groups were combined to form a probability histogram. This histogram, shown in figure 6.5 shows the number of droplets as a function of the angle of diffraction θ , and is compared to the expected distribution

$$N = \left| \text{sinc} \left(\frac{d\pi \sin(\theta)}{\lambda} \right) \right| \quad (6.3)$$

which is plotted in the same figure, taking a slit width $d = 22$ mm. This is somewhat wider than the 18 mm at which the slit was constructed, possibly because the barriers had rounded edges, leading to a larger effective width of the slit. Some skewness in the histogram can be observed, likely due to improper alignment of the liquid bath. It is noteworthy that this skewness and misalignment was noted by some of the students, and coupled to ‘one part of the liquid being wavy’.

Figure 6.5: Histogram showing 233 data points collected by the students in the experiment. The expected distribution (equation (6.3)) is plotted in red, using an effective slit width $d = 22$ mm.

6.5.1 Observations and Discussion

Giving each student a task worked well, although most students focused strongly on performance of the prescribed task, enthusiastically urging the droplets to pass through the slit. This was confirmed by answers given in the interview conducted directly after the experiment. Students referred mostly to performing the tasks: Generating droplets that had to go through the slit. However, two groups of students remarked that they were measuring the behavior of the droplets and checking for diffraction patterns of arriving droplets. The focus on the task of ‘making droplets’, caused many of the students to overlook other interesting behavior. Interaction between droplets, droplets hopping over the barrier and reflection from the walls

was often observed by the supervising author, but not by the students. The focus on a recipe is a well-known effect that can occur in practicals [16]. Most students may also lack an adequate framework of reference, which results in the unexpected going unnoticed. After the experiment, and upon asking, all students showed that they understood that diffraction patterns were looked for. All of them were also able to recognize that the data they collected actually resembled a diffraction pattern. More and better explanation of the analogy with other slit experiments and what is observed in the experiment is likely to be advisable as 50% of the students thought that either the wave or the droplet was responsible for the diffraction pattern. The other 50% of the students saw correctly that it was in fact the combination of both droplet and the wave. Future work should involve a larger number of students which will allow for pre- and post-test application of concept tests to show growth in understanding [17]

6.6 Conclusion

The exciting and relatively new ‘bouncing droplet’ experiment provides a way to give students a glimpse of the combination of simultaneous particle and wave behavior. The experiment is stable enough to be performed by students in a classroom setting. Students worked in pairs and collected data from the single slit experiment. Their combined data clearly showed a diffraction pattern. Discussions with students showed that it is important to guide learners as the simultaneous particle and wave behavior is not automatically recognized by all. The next step is to bring the experimental setup to more classrooms in a robust version. In doing this, we will provide teachers with hints for introducing the experiment, and for follow-up peer and classroom discussions. This will help ensure that student understanding of duality is scaffolded as intended.

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7

Conclusions and Outlook

Droplets are everywhere in nature and they have many applications in industry, e.g. in EUV-lithography, inkjet printing and medical spray formation. This thesis focusses on droplet coalescence during flight, drag on droplet trains and droplet impact on a deep pool and the subsequent cavity formation.

7.1 Conclusions

In chapter 2 we show that the superposition of two Rayleigh-Plateau-unstable modes is an efficient and robust method to generate a periodic stream of droplets, that allows for control of the droplet sizes and distances. The method is efficient because it uses the available surface energy of the continuous jet to induce the coalescence. The method is robust because the wave numbers of the employed Rayleigh-Plateau-unstable modes are close to the fastest growing mode. By tuning the phase difference between the two modes, the coalescence pattern is controlled. The phase difference with the shortest merge time is not the one for which the two perturbations are in phase at a minimum, due to the presence of satellite droplets. The final droplet size, spacing and merge time increase with the beat number n .

We continue in chapter 3 with an experimental study on drag and coalescence on a monodisperse droplet train. The results show quantitative agreement with H.C. Lee's theory [1] a volume averaged droplet distribution for a range of droplet actuation wavenumbers. However when we zoom in on the level of single droplets, a dispersion in drag depending on the spacing towards the preceding droplet is

observed. We interpret these results as a shielding effect where the droplet which has moved closer towards the preceding droplet, loses less kinetic energy to the surrounding air, as expected. Surprisingly, despite this shielding effect and intricate details of the flow, the drag on the ensemble-averaged system is very well captured by Lees theory, until coalescence breaks down the description.

Chapter 4 deals with our experimental, numerical and modelling results of perpendicular droplet impact on a deep pool. We have extended the work of Oguz and Prosperetti to the regime beyond the terminal velocity under ambient conditions for droplets smaller than a millimeter in size. Results show that in the velocity, diameter parameter space, a region exists where the movement of the cavity bottom and the incoming capillary waves is such that a bubble is entrained. The dynamics of the cavity are well described by a 1D Rayleigh-Plesset model including surface tension of the cavity. The cavity is closed by a capillary wave which has a velocity set only by the droplet size. Next to the observed regular bubble entrainment, there also exists a region in which the second to last wave entrains a bubble, this is also confirmed experimentally.

We present in chapter 5 an experimental study of oblique drop impact onto a quiescent deep liquid pool. We performed quantitative experiments where oblique drops impact onto a deep liquid pool for a wide range of We and impact angles α and analyzed the splashing behavior, the cavity formation and the cavity collapse. In analogy to previous studies e.g. [2–5], we found that the crown velocity has to be larger than the Taylor-Culick velocity to obtain splashing. For oblique drop impact this crown velocity is influenced by the tangential velocity of the impacting drop, leading to an asymmetry in the crown and giving rise to an asymmetry in the splashing threshold [6]. The magnitude of the cavity displacement depends on the tangential Weber number and the cavity depth depends on the perpendicular Weber number.

7.2 Outlook

7.2.1 Drag and Coalescence

Using the results from chapter 3, improvements can be made on the matching between the numerical simulations and experiments in Fig. 2.10. Implementation of spacing-dependent drag can be done by following the steps from section 2.5. Open questions remain, for example on the onset of coalescence. Lee's model including spacing-dependent drag does not take into account size differences present in coalescing droplet streams, while this influences the flow profile around the droplets. A way to include this in a model could be to give droplets an individual $\bar{\rho}$. Individual droplet behavior can be estimated with this approach, possibly leading to a new ensemble description. As this point, the model does

not include any details of the influence of the size distribution on the recirculation zone behind the droplet. A tool to study the flow field in air would be a full three dimensional simulation. Initial conditions for the simulation could be a train of droplets with a narrow velocity distribution as simulated in section 3.5, with the Lee boundary layer surrounding the droplet train. The results would then be a starting point for deriving a theoretical relation for the curves observed in Fig 3.14. For the range of Re as used in our experiments, the collapse of the spacing dependent drag curves indicates that the flow behavior is similar. For a larger range of Re this will probably not be the case as the recirculation zone will go to different phases towards instability as flow around a cylinder does [7]. An extension of the experiments in a direction interesting for industrial applications would be to disturb the droplet steam with lateral flows to characterize the system response.

7.2.2 Impact

For chapters 4 and 5, many approaches are possible to increase the understanding of the phenomena observed after droplet impact on a deep pool. For oblique impact, the splashing threshold contrasts the model presented in [6] where oblique drop impact on a dry substrate was studied. We cannot describe our data by simply adding/subtracting the tangential velocity to the crown velocity. Instead, we assume the tangential velocity influences the amount of mass squeezed into the crown. So the influence of tangential velocity on the splashing threshold appears to be different for impact on a pool and impact on a solid surface. An open questions is why exactly this is the case and what is the flow profile inside the liquid causing this difference. Also a difference in timing should be observable in the release time of the droplets, since the mechanism proposed in chapter 5, based on mass flow, should take more time than direct coupling of the droplet velocity into the lamella or crown. In our experiment, data from a single measurement plane is obtained. It would be interesting to obtain the full 3D profile above and below the water surface using holographic microscopy [8] or by extended numerical simulations.

For industrial applications, the influence of pressure on the splashing threshold is of interest and it is known to have an influence on the splashing threshold K_s . In literature, there is a discussion on the topic since the pioneering work from the Nagel group [9], for impact on a smooth surface. Not only the splashing threshold, but also the size of the droplets, their flight direction and velocity are of interest. More recent theoretical and numerical developments for impact on a thin film show that the pressure influences the timing at which a radial sheet emerges from the droplet, before splashing. It is this timing that is crucial for setting the splashing threshold [10–13].

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Summary

In chapter 2 we experimentally, numerically, and theoretically demonstrate a novel method of producing a stream of widely spaced high-velocity droplets by imposing a superposition of two Rayleigh-Plateau-unstable modes on a liquid jet. The wavelengths of the two modes are chosen close to the wavelength of the most unstable mode. The interference pattern of the two superimposed modes causes local asymmetries in the capillary tension. The velocity of the initial droplets depends on these local asymmetries. Due to their different velocities, the droplets coalesce to produce a stream of larger droplets spaced at a much larger distance than the initial droplets. We analytically derive the perturbations that robustly induce this process and investigate the influence of the non-linear interactions of the two Rayleigh-Plateau-unstable modes on the coalescence process. Experiments and numerical simulations demonstrate that the jet breakup and the subsequent droplet merging are fully governed by the selected modes.

Air drag on droplets in a droplet train can lead to axial and lateral spreading of the droplets eventually leading to coalescence and loss of monodispersity. A popular study by H.C. Lee (IBM J.Res. Dev. 21 (1),1977) has successfully modeled drag within a droplet train, however, the presented theory does not contain a physical description of the destabilization. Here in chapter 3 we present experimental data to show a mechanisms responsible for destabilization. We use ultrafast high-speed imaging and advanced image processing to capture and track individual monodisperse droplets created by piezo-acoustic actuation of a microjet. Our results show that the change in droplet spacing within the droplet train is a direct result of air drag. The measured drag coefficients critically depend on the spacing between droplets, where the closely spaced droplets have a significantly lower drag coefficient, as expected. Surprisingly, despite this shielding effect and intricate details of the flow, the drag on the ensemble-averaged system is very well captured by Lees theory, until coalescence breaks down the description. Our results apply directly to inkjet printing, medical spray formation and 3D printing.

Droplets impacting on a liquid pool and the subsequent bubble entrainment has been extensively studied for mm-sized droplets, such as rain droplets. Here in chapter 4 we study droplet impact and bubble entrainment in an entirely new parameter regime. We continue the pioneering work of Oguz & Prosperetti, for

microdroplets with a velocity beyond their terminal velocity. Cavity dynamics were found to be surface tension dominated, in analogy to the hydrostatically dominated cavity in the case of mm-sized droplets. We model the cavity with a 1D Rayleigh-Plesset type model and decouple the capillary wave travelling down. A capillary wave is formed at the crown tip, therefore we account for the formation time and height of the crown in predicting the cavity collapse. Analysis of numerical simulations in Gerris showed that the capillary wave has a wavelength corresponding to half the circumference of the impacting droplet. This sets the velocity of the capillary wave and combining it with the Rayleigh-Plesset type model, the collapse time and depth of the cavity can be estimated. Both high-speed imaging experiments and numerical simulations validate the simple empirical model.

Oblique impact of drops on a solid or liquid surface is frequently observed in nature. Most studies on drop impact and splashing, however, focus on perpendicular impact. Here, in chapter 5 we study oblique impact onto a deep liquid pool, where we quantify the splashing threshold, maximal cavity dimensions and cavity collapse by high-speed imaging above and below the water surface. Three different impact regimes are identified: smooth deposition onto the pool, splashing in the direction of impact only, and splashing in all directions. We provide scaling arguments that delineate these regimes by accounting for drop impact angle and Weber number. The angle of the axis of the cavity created below the water surface follows the impact angle of the drop independent of the Weber number, while cavity depth and its displacement with respect to the impact position depend on the Weber number. Weber number dependency of both the cavity depth and displacement is modeled using an energy argument.

Chapter 6 brings a recently discovered macroscopic phenomenon with wave-particle characteristics into the classroom. The system consists of a liquid droplet levitating over a vertically shaken liquid pool. The droplets allow visualization of a wave-particle system in a directly observable way. We show how to interpret this macroscopic phenomenon and how to set up and carry out this experiment. A class of students performed single slit diffraction experiments with droplets. By scoring individual droplet trajectories students find a diffraction pattern. This pilot application in the classroom shows that students can study and discuss the wave-particle nature of the bouncing droplet experiment. The experiment therefore provides a useful opportunity to show wave-particle behavior on the macroscopic level.

Samenvatting

Druppels vinden we overal in de natuur en ze kennen daarnaast veel industriële toepassingen, bijvoorbeeld in EUV-lithografie, inkjetprinting en medische spray formatie. In deze thesis kijken we naar druppelsamensmelting, luchtwrijving op druppelstromen en druppel inslag op een vloeistofbad.

In hoofdstuk twee laten we experimenteel, numeriek en theoretisch een nieuwe methode zien om een stroom druppels met zowel een grote onderlinge afstand als een hoge snelheid te produceren. Dit doen we door middel van superpositie van twee Rayleigh-Plateau instabiele golven op een langgerekte vloeistof straal. De golflengtes van de twee golven zijn dicht bij de meest instabiele golflengte gekozen. Het interferentiepatroon van twee opgetelde golven zorgt voor lokale asymmetrie in de capillaire spanning. De beginsnelheid van de druppels hangt af van deze lokale asymmetrie. Door de opgelegde snelheidsverschillen smelten de druppels samen en zo ontstaan grotere druppels en druppels met een grotere onderlinge afstand dan die aan het begin. We leiden analytisch af hoe de verstoringen dit proces op een robuuste manier in gang zetten en we onderzoeken de invloed van niet-lineaire interacties van de twee Rayleigh-Plateau instabiele golven op het samensmeltingsproces. Experimenten en numerieke simulaties laten zien dat de opbreking van de langgerekte waterstraal en de daaropvolgende samensmelting van de druppels volledig door de vooraf ingestelde golven wordt beheerst.

Luchtwrijving op druppels in een druppelstroom kan leiden tot axiale en laterale spreiding van de druppels en leidt uiteindelijk tot spontane samensmelting en een verlies van een monodisperse druppelgrootte. Een welgeciteerde publicatie van H.C. Lee (IBM J.Res. Dev. 21 (1),1977) heeft geleid tot het succesvol modelleren van wrijving op een druppeltrein, maar deze theorie bevat geen fysische beschrijving van de destabilisatie. In hoofdstuk drie presenteren we onze experimentele data om de onderliggende mechanismen die aanzetten tot destabilisatie uit de doeken te doen. We gebruiken hogesnelheidsopnames en geavanceerde beeldverwerking om individuele monodisperse druppels te volgen, die zijn gemaakt met piëzo-akoestische aandrijving van een microscopische langgerekte waterstraal. Onze resultaten laten zien dat een kleine verandering in de onderlinge afstand tussen de druppels wordt versterkt door onderliggende luchtwrijving. De gemeten luchtwrijvingscoëfficiënten hebben een kritische afhankelijkheid van de onderlinge

druppelafstand, waarbij een kleine onderlinge afstand een flink lagere luchtwrijvingsoëfficiënt tot gevolg heeft. Het was verrassend dat de druppels, ondanks dit effect waarbij druppels elkaar uit de wind houden, zich gemiddeld wel houden aan de theorie van Lee, totdat samensmelting van druppels leidt tot ineenstorting van de beschrijving. Onze resultaten zijn direct toe te passen bij inkjetprinting, verstuiwing van medicijnen voor inhalatie en driedimensionaal printen.

De inslag van een druppel op een vloeistofbad en het achtereenvolgens invangen van een bel zijn uitgebreid bestudeerd voor regendruppels. In hoofdstuk vier bestuderen we druppelinslag en het invangen van belletjes in een compleet nieuw deel van de parameterruimte. We werken verder aan het baanbrekende werk van Oguz & Prosperetti maar nu voor sub-millimeter druppeltjes met snelheden boven de terminale eindsnelheid van regendruppels. We zien dat oppervlaktespanning de dominante kracht is in de vorming van de holte, in tegenstelling tot de door zwaartekracht gedomineerde vorming van de holte in het geval van regendruppels. De numerieke simulaties laten zien dat de golf die de holte sluit een golflengte heeft die gelijk is aan de halve omtrek van de druppel die inslaat. Bij het modelleren van het gedrag van deze golf nemen we de vorming van de kroon mee. Gebruikmakend van een eendimensionaal Rayleigh-Plesset model voorspellen we de tijden lengteschalen van de holte. Zowel hogesnelheidsmicroscopie als numerieke simulaties in Gerris valideren ons model.

Schuine inslag van druppels op een vaste stof of vloeistof is een fenomeen wat regelmatig te zien is in de natuur. De meeste onderzoeken naar druppelinslag en -spatgedrag leggen echter de nadruk op loodrechte inslag. In hoofdstuk vijf bestuderen we schuine inslag op een diep waterbad waarna we kwantitatief het omslagpunt van niet naar wel spatten bepalen, de maximale dimensies van de holte meten en de ineenstorting van de holte bestuderen door middel van hogesnelheidsmicroscopie zowel boven als onder het wateroppervlak. Drie verschillende vormen van inslag gedrag zijn daarbij te onderscheiden: het eenvoudigweg opgaan van de druppel in waterbad, het spatten enkel in de richting van de inslag en spatten in alle richtingen. We leiden een schalingsargument af in termen van het Weber-getal en de hoek van inslag die deze drie mogelijkheden afbakent in de parameterruimte. De holtehoek die onder water is gevormd volgt de hoek van inval, ongeacht het Weber-getal, terwijl de holte-diepte en de holte-verplaatsing ten opzichte van het punt van inslag, afhankelijk zijn van het Weber-getal. De afhankelijkheid van het Weber-getal voor beide holte parameters is met behulp van een energiebalans uitgerekend.

Hoofdstuk zes laat een recent ontdekt macroscopisch fenomeen met zowel eigenschappen van een golf als die van een deeltje debuteren in het klaslokaal met als doel kwantummechanische begripsvorming. Het systeem bestaat uit een druppel (het deeltje) zwevend boven een verticaal op en neer bewegend vloeistof

bad en een golf die ontstaat op het oppervlak van het bad door het stuiteren van de druppel. Leerlingen hebben enkele slit diffractie experimenten met deze druppels uitgevoerd. Door elke druppel individueel te volgen vinden de leerlingen een diffractie patroon. Deze toepassing in het klaslokaal laat zien dat leerlingen golf-deeltjes eigenschappen van het experiment met druppeltjes kunnen bestuderen en dat het discussie uitlokt. Het experiment is daarom een nuttige mogelijkheid om golf-deeltjes gedrag op grotere schaal te laten zien.

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About the author

Cornelis Pascal Sleutel was born on February 12th, 1987 in Hoorn, The Netherlands. He graduated from high school (Gymnasium) at Oscar Romero in Hoorn in 2005. In the same year he started his studies in Applied Physics (Technische Natuurkunde) at the University of Twente in Enschede and moved to the campus. His bachelor assignment was performed in the Physics of Fluids group and Med-spray on "Characterizing the flow resistance in a Medspray nozzle". After that he started the master programme Fluid Physics. His internship was performed in JNCASR, Bangalore, India, under supervision of Prof. Meheboob Alam; resulting in a report titled "Granular gases and their stability". In March 2012 he received his masters degree in the Physics of Fluids group on "Transport phenomena in dental microchannels", supervised by Bram Verhaagen and Michel Versluis and resulting in a publication in "Microfluidics and Nanofluidics". In September 2012 he started his PhD research in the Physics of Fluids group, under supervision of Michel Versluis, Detlef Lohse and Frits Dijkman and in close collaboration with ASML. The results of this are presented in this thesis.

During his study time, Pascal worked as a exam preparation Physics Teacher in Leiden. These experiences led him to strive for a grade as a teacher, parallel to his PhD research. He obtained his licence to teach (1e graad natuurkunde) with an internship in "CSG Het Noordik" and a graduation on "Quantummechanica zichtbaar voor het oog: Een analogie om golf-deeltjes te zien en onbepaaldheid te begrijpen" resulting in chapter six and a publication in "European Journal of Physics".

Physics of Fluids

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Stellingen

behorend bij het proefschrift

Druppeltjes: Luchtwrijving, Samensmelting en Inslag

1. Het bijeenvegen van druppels door oppervlaktespanning optimaal in te zetten heeft als nuttige bijvangst een druppelstroom zonder satellieten, mits de fase goed gekozen is.

Hoofdstuk 2 van deze thesis

2. Een afstandsafhankelijke wrijvingscoëfficiënt is cruciaal als toevoeging op de theorie van Lee bij het modelleren van individuele druppels in druppelstromen die meer dan 200 maal hun eigen straal afleggen.

Hoofdstuk 3 van deze thesis

3. Het ontdekken van de diepere oorzaak voor snelle, terugschietende druppeltjes na druppelinslag vraagt om een ontkoppeling van de relevante fysische fenomenen.

Hoofdstuk 4 en 5 van deze thesis

4. Druppels die springen op Faraday-golven zijn in principe geschikt om middelbare scholieren kwantummechanica bij te brengen, maar voorlopig is het zowel voor de druppels als voor de leerlingen een sprong te ver.

Hoofdstuk 6 van deze thesis

5. Het is reuze jammer dat lesgeven aan VWO en Havo leerlingen bij docenten vooral een beroep doet op niet-wetenschappelijke vaardigheden als orde houden, disciplineren en het afleren van verkeerde gewoontes.

6. Het door de overheid gestimuleerd omtoveren van promovendi tot tech-ondernemers schiet zijn doel voorbij.

7. Promovendi die samenwerken met bedrijven zien de beknutting van hun academische vrijheid beloond met een kijkje in de keuken van diezelfde bedrijven.

8. Het toepassen van fysische wetten in de keuken komt de smaak ten goede, zolang men daardoor maar niet vergeet zout toe te voegen.

9. Een kwalijk effect van thuiswerken is een verzakelijking van sociale verhoudingen en takenpakketten op de werkvloer.

10. De bijzondere aandacht voor alledaagse zaken als koffie, lunches en diners bij natuurkunde conferenties en zomerscholen is cruciaal om het meest menselijke in wetenschappers naar boven te halen.

Propositions

accompanying the thesis

Droplets: Drag, Coalescence and Impact

1. Coalescence in a droplet stream by efficient use of surface tension eliminates all satellites from the stream, if the right phase is chosen.

Chapter 2 of this thesis

2. A spacing dependent drag coefficient is crucial as an addition to Lee's theory in modelling individual droplets in droplet streams that travel more than 200 times their own radius.

Chapter 3 of this thesis

3. Discovering the exact cause for fast backfiring droplets after drop impact requires a decoupling of relevant physical phenomena.

Chapters 4 and 5 of this thesis

4. Droplets bouncing on Faraday waves are suitable for teaching quantum mechanics to high school scholars but at the moment it is for both students and droplets a bridge too far.

Chapter 6 of this thesis

5. It is very sad that teaching to VWO and Havo scholars mainly requires teachers to use non-scientific skills like peacekeeping, disciplining and changing habits.

6. The government policy to turn PhD-students into tech entrepreneurs is wrong.

7. PhD-students that cooperate with companies see the limitation of their academic freedom rewarded with a view in the machine room of industry.

8. Applying the laws of physics in the kitchen improves taste as long as it does not make you forget to add salt.

9. Negative effects caused by working from home are a more businesslike relationship among colleagues and the formalisation of division of work.

10. The special attention paid to daily matters like coffee, lunches and dinner at physics conferences and summer schools reveals the most humane characteristics from scientists.