Advanced Irradiation Schemes for Target Shaping in Droplet-Based Laser-Produced Plasma Light Sources

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Abstract

The presented work is in the field of droplet-based laser produced plasma light sources. The preshaping of the liquid droplet target before it’s irradiation by the plasma generating main pulse has been of growing interest in the field of EUV and soft x-ray light sources. Research in this field has already proven that improvements in system performance are achievable in terms of conversion efficiency and debris mitigation when the target is carefully preshaped before the primary irradiation. The increase in conversion efficiency of the source leads to higher throughput for these light sources, whereas the improvement of debris mitigation leads to a reduction in the cost of ownership.

The neutral cluster debris dynamics of a droplet-based laser-produced plasma generated by a single nanosecond scale laser pulse is studied experimentally and analytically. Experiments were done imaging the debris with a high-speed shadowgraph system and using image processing to determine the droplet debris mean radial velocity $V$ dependence on laser pulse irradiance $E_c$. The data shows a power law dependence between the mean radial debris velocity and the incident irradiance giving $V \propto E_c^n$ with $n \approx 0.65$. A scaled analytical model was derived modeling the plasma ablation pressure on the droplet surface as the primary momentum exchange mechanism between the unablated droplet material and the laser pulse. The relationship between droplet debris trajectory and the droplet alignment with the laser was quantified analytically. The derived analytical model determines that the neutral cluster debris trajectory for an ablated droplet is a function of the laser profile $f_L$, the droplet diameter $d_0$ and the axial misalignment $\psi$ between the laser axis and the droplet center. The analytical calculations from these models were found to be in good agreement with the measurements. This analysis has practical significance for understanding ablated droplet debris, droplet
deformation by laser pulsing, and droplet breakup from very short timescale
shocks. Liquid Sn droplets were irradiated with shaped bursts of picosecond laser
pulses. The shape of the deforming droplets following the impact of the re-
coil pressure induced by these bursts were imaged using a high speed shadow-
graph system. It is observed that the modified Weber number $We_s$ describes
the unruptured sheet expansion over a wide variation in burst shapes. The
splash Weber $We_s$ scales as $\sim 0.5N_p^{1-2n}(N_p + 1)$ relative to the nominal
Weber number $We$ by distributing the laser energy evenly among $N_p$ pulses,
where the laser pulse peak ablation pressure $P_s$ is exponentially proportionate
by a factor of $n$ to the laser irradiance $I_0$ as $P_s \propto I_0^n$. The deforming droplet
sheet forms various cup shapes whose depth is dependent upon $N_p$. The
timing and energy arrangement of pulses within the burst has been shown
to influence the shape of the cup formed by the expanding droplet sheet.
Cavitation within the droplets are observed evidenced by hydrodynamically
focused microjets ejected behind droplet. The threshold conditions for ca vi-
tation are approximated from the ablation pressure and gain and loss terms
for the focused acoustic wave when it reaches the droplet center. The pre-
dicted cavity size and growth rate are compared against the dimensions of
the ejected microjets and found to be in good agreement. The rupture time
$\tilde{t}_b$ of the droplet expanding as a thin fluid film was measured for each case. A
Rayleigh-Taylor instability analysis is done in order to determine the dependen-
ces governing $\tilde{t}_b$. The evidence supports the hypothesis that the initial
perturbations of the developing Rayleigh-Taylor instabilities are on the or-
der of the ablation depth and that there is a lower cutoff wavelength of these
initial perturbations of $\sim 10 \mu m$.
Multiphase CFD simulations are constructed of a droplet impacted by the
ablation pressure wave of a pulsed laser. These simulations were performed
in Fluent 18.0 using the multiphase Volume of Fluid with the coupled level
set model, explicit formulation, the geo-reconstruct scheme, and adaptive
meshing. The pressure and velocity distribution from the laser ablation in-
duced shockwave was calculated analytically and set as the initial condition
for the droplet. The laser pulse irradiance is varied across several cases.
The cases were run until the droplet fragmentation ceased. Final fragment
size distributions and mass flux distributions are compared for the cases. It
was determined that the fragment size of the disintegrating droplet is deter-
mined by the splash Weber number $W{e_s}$, and the initial states of the surface
perturbations wavenumber $k$, and amplitude $\eta_0$, which are calculated ana-
lytically with the Rayleigh-Taylor instability model. This analytical model
is validated by the numerical simulations and the experimental results.
Zusammenfassung


Die neutrale Cluster-Trümmerdynamik eines tropfenbasierten Laserplasmas, das durch einen Laserpuls im Nanosekundenbereich generiert wird, wird experimentell und analytisch untersucht. Die Trümmerausbreitung wurde mit einem Hochgeschwindigkeits Schattenbildverfahren abgebildet und ausgewertet, um die mittlere radiale Geschwindigkeit der Tropfenfragmente $V$ in Abhängigkeit von der Laserpuls-Bestrahlungsstärke $E_c$ zu bestimmen. Aus den Resultaten ergibt sich der, auf dem Potentialgesetz basierende, Zusammenhang zwischen der mittleren radialen Trümmergeschwindigkeit und der einfallenden Bestrahlungsstärke, $V \propto E_c^n$ mit dem experimentell bestimmten Exponenten $n \approx 0,65$. Es wurde ein skaliertes analytisches Modell abgeleitet, das den Plasmaablationsdruck auf der Tropfenoberfläche als primären Impulsaustauschmechanismus zwischen dem ruhenden Tropfenmaterial und dem Laserpuls modellierte. Die Beziehung zwischen der Tropfenflugbahn und der Tropfenausrichtung gegenüber dem Laser wurde analytisch quantifiziert. Das abgeleitete analytische Modell bestimmt, dass die Trajektorie der neutralen Cluster-Trümmer für ein abgespaltenes Tropfenfragment eine Funktion des Laserprofils $f_L$, des Tropfendurchmessers $d_0$ und der axialen Versetzung $\psi$ zwischen der Laserachse und dem Tropfenmittelpunkt ist. Die analytischen

Flüssige Sn-Tröpfchen wurden durch getaktete Pikosekunden-Laserpulse bestrahlt. Die Verformung der Tropfen nach dem Aufprall des durch diese Stöße induzierten Rückstofldrucks wurde unter Verwendung eines Hochgeschwindigkeiten-Schattenbildsystems abgebildet. Es wird beobachtet, dass die modifizierte Weber-Nummer \( W_{es} \) die zusammenhängende Tropfenscheibenausbreitung über eine große Variation der Stoßformen beschreibt. Der Splash-Weber \( W_{es} \) skaliert als \( \sim 0,5N_p^{1-2n}(N_p+1) \) relativ zur nominellen Weber-Nummer \( W_e \), indem die Laserenergie gleichmäßig auf die Anzahl der Pulse \( N_p \) verteilt wird. In den Momenten, in denen der Laserpuls auftritt, ist der Spitzenablationsdruck \( P_s \) mit dem Faktor \( n \) exponentiell proportional zur Laserbestrahlungsstärke \( I_0 \), wie in \( P_s \propto I_0^n \). Die sich verformende Tropfenscheibe bildet verschiedene Becherformen aus, deren Tiefe von \( N_p \) abhängt. Es wurde gezeigt, dass die Becherform des Weiteren von der zeitlichen Abstimmung und der Energieverteilung der Impulse innerhalb des Impulspakets abhängt. Kavitationseffekte spielen sich innerhalb der Tröpfchen ab, welche sich indirekt durch fokussierte, hydrodynamische Mikrojets nachweisen lassen, die hinter den Tropfen ausgestoßen werden. Die Grenzwerte für Kavitationserscheinungen werden aus dem Ablationsdruck und den Verstärkungs- und Verlustbedingungen für eine fokussierte akustische Welle ermittelt, welche das Tropfenzentrum erreicht. Die vorhergesagte Hohlraumgröße und die Wachstumsrate stimmen gut mit den gemessenen Mikrojets überein. Die Tropfenausfallzeit \( \tilde{t}_b \) des sich zu einem dünnen Flüssigkeitsfilm ausbreitenden Tropfens wurde für alle Fälle gemessen. Eine Rayleigh-Taylor Instabilitätsanalyse wurde durchgeführt, um die Abhängigkeiten für \( \tilde{t}_b \) zu bestimmen. Die Resultate stützen die Hypothese, dass die anfänglichen Störungen der sich entwickelnden Rayleigh-Taylor-Instabilitäten in der Größenordnung der Ablationstiefe liegen und dass eine untere Grenzwellenlänge dieser anfänglichen Störungen von \( \sim 10 \mu m \) existiert.
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<tr>
<td>A-BI</td>
<td>Actinic Blank Inspection</td>
</tr>
<tr>
<td>AIMS</td>
<td>Aerial Image Measurement System</td>
</tr>
<tr>
<td>ALPS</td>
<td>Applied Laser Plasma Science</td>
</tr>
<tr>
<td>AOM</td>
<td>Acousto-Optic Modulator</td>
</tr>
<tr>
<td>A-PI</td>
<td>Actinic Pattern Inspection</td>
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<tr>
<td>CE</td>
<td>Conversion Efficiency</td>
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<td>CFD</td>
<td>Computational Fluid Dynamics</td>
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<tr>
<td>DPP</td>
<td>Discharge Produced Plasma</td>
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<tr>
<td>ETH</td>
<td>Eidgenössische Technische Hochschule</td>
</tr>
<tr>
<td>EUV</td>
<td>Extreme Ultraviolet</td>
</tr>
<tr>
<td>EUVL</td>
<td>Extreme Ultraviolet Lithography</td>
</tr>
<tr>
<td>FFT</td>
<td>Fast Fourier Transform</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full Width Half Maximum</td>
</tr>
<tr>
<td>HVM</td>
<td>High Volume Manufacturing</td>
</tr>
<tr>
<td>IB</td>
<td>Inverse Bremsstrahlung</td>
</tr>
<tr>
<td>IC</td>
<td>Integrated Circuit</td>
</tr>
<tr>
<td>LEC</td>
<td>Laboratory for Energy Conversion</td>
</tr>
<tr>
<td>LED</td>
<td>Light Emitting Diode</td>
</tr>
<tr>
<td>LPP</td>
<td>Laser Produced Plasma</td>
</tr>
<tr>
<td>MOPA</td>
<td>Master-Oscillator Power Amplifier</td>
</tr>
<tr>
<td>MD</td>
<td>Molecular Dynamics</td>
</tr>
<tr>
<td>SNR</td>
<td>Signal-to-Noise Ratio</td>
</tr>
<tr>
<td>SPH</td>
<td>Smoothed Particle Hydrodynamics</td>
</tr>
<tr>
<td>VOF</td>
<td>Volume of Fluid</td>
</tr>
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</table>
Nomenclature

Notation

\dot{} \quad (-) notation for first time derivative
\ddot{} \quad (-) notation for second time derivative
\sim \quad (-) dimensionless parameter

Latin symbols

\(a_0\) \quad (-) scaling prefactor for the center axial momentum transfer to the fluid
\(A\) \quad \text{(m}^2\) area
\(A_a\) \quad \text{(g/mol)} atomic mass
\(A_1\) \quad \text{ (?)}
\(C\) \quad \text{(\mu m)} circumference of the expanding sheet concentric with the sheet center axis
\(C_D\) \quad (-) drag coefficient
\(c_i\) \quad \text{(m/s)} average ion speed
\(c_s\) \quad \text{(m/s)} speed of sound in droplet fluid
\(d\) \quad \text{(\mu m)} fragment diameter
\(d_0\) \quad \text{(\mu m)} initial droplet diameter
\(d_{xx}\) \quad \text{(\mu m)} droplet fragment mean diameter; \(d_{ij} = \frac{\sum d^i}{\sum d^j}\)
\(d_{10}\) \quad \text{(\mu m)} droplet fragments mean diameter
\(d_{32}\) \quad \text{(\mu m)} droplet fragments Sauter mean diameter
\(d_{43}\) \quad \text{(\mu m)} droplet fragments mass mean diameter
\(d_c\) \quad \text{(\mu m)} cavity diameter
\(d_{h,\text{err}}\) \quad \text{(\mu m)} perforation diameter
\(D_r\) \quad \text{(\mu m)} deforming droplet cup diameter
\(E_c\) \quad \text{(GW/cm}^2\)} laser irradiance of a single laser pulse in a burst
\(E_B\) \quad \text{(\mu J)} total laser burst energy
### Contents

\( E_K \) (J) total kinetic energy of the impacted droplet  
\( E_{K,cm} \) (J) kinetic energy of the fluid center of mass of the impacted droplet  
\( E_{K,d} \) (J) kinetic energy of the deformation of the impacted droplet  
\( E_p \) (\( \mu \)J) single laser pulse energy  
\( f \) (-) wave perturbation shape function  
\( \overrightarrow{F} \) (N) force vector  
\( f_s \) (-) deformed droplet sheet shape factor  
\( f_N \) (-) acoustic f-number  
\( f_{\text{rare}} \) (-) ratio of the magnitude of tensile stress to peak pressure of the laser ablation shock wave traveling through the droplet  
\( f_{\text{rep}} \) (Hz) frequency of pulses within the picosecond burst  
\( F_{\text{vol}} \) (N/m\(^3\)) volume force  
\( \overrightarrow{g} \) (m/s\(^2\)) gravitational acceleration  
\( G_{\text{focus}} \) (-) gain factor of the shock wave pressure due to acoustic focusing  
\( h \) (\( \mu \)m) sheet thickness approximated from the conservation of mass  
\( i \) (\( \mu \)m) index for time step  
\( I_0 \) (GW/cm\(^2\)) laser irradiance of a single laser pulse in a burst; \( I_0 = 2 \ln(2)E_e \)  
\( k \) (rad m\(^{-1}\)) perturbation wavenumber  
\( K \) (-) concavity of the expanding droplet sheet \((Z - d_0)/D_r\)  
\( K_1 \) (-) single pulse concavity  
\( k_b \) (rad m\(^{-1}\)) wavenumber corresponding to the fastest growing wavemode  
\( k_c \) (rad m\(^{-1}\)) capillary wavenumber  
\( k_f \) (m\(^{-1}\)) acoustic wavenumber  
\( k_l \) (-) lithography process constant  
\( k_{\text{max}} \) (rad m\(^{-1}\)) maximum wavenumber for the laser accelerated droplet surface  
\( Kn \) (-) Knudsen number  
\( l^0_{ac} \) (\( \mu \)m) the nominal distance between the ablated surface and the critical density surface
\( L \) (m) the distance the debris travels from the plasma to the chamber wall

\( L_{\text{atten}} \) (-) attenuation factor due to acoustic losses of the shock wave

\( L_f \) (-) smallest visible length scale in a shadowgraph system

\( L_s \) (-) distance between the maximum absorption region of the plasma and the spot of peak intensity at the droplet surface for \( \alpha_d = 0^\circ \)

\( L_w \) (nm) minimum achievable line width on a wafer

\( m \) (kg) mass accelerated during the pressure pulse duration

\( m'' \) (kg s\(^{-1}\) sr\(^{-1}\)) debris mass flux per steradian

\( Ma \) (-) Mach number

\( \dot{m}_{qp} \) (kg/s) mass transfer from phase \( q \) to phase \( p \)

\( \dot{m}_{pq} \) (kg/s) mass transfer from phase \( p \) to phase \( q \)

\( n \) (-) exponent corresponding to exponential dependence of peak ablation pressure on pulse irradiance; \( P_s \propto I_0^n \)

\( \hat{n} \) (-) normalized surface normal; \( \hat{n} = \frac{\vec{n}}{|\vec{n}|} \)

\( n_{fg} \) (-) shadowgraph foreground intensity level

\( n_{bg,0} \) (-) nominal shadowgraph background intensity level

\( n_P \) (-) Gamma distribution parameter

\( NA \) (-) numerical aperture

\( N_p \) (-) number of pulses in a burst

\( N_s \) (-) CCD background noise level

\( Oh \) (-) Ohnesorge number

\( p \) (-) alternate phase to \( q \) in 2-phase VOF model

\( P \) (Pa) pressure

\( PDF \) probability density function

\( P_l \) (-) Legendre polynomial

\( P_{\text{max}} \) (Mbar) peak ablation pressure of a single laser pulse in a burst

\( P_s \) (Pa) peak ablation pressure of a single laser pulse in a burst

\( q \) (-) alternate phase to \( p \) in 2-phase VOF model

\( r \) (\( \mu \text{m} \)) distance from expanding sheet center to a point on the sheet surface, tracing the sheet surface
Contents

$R$ \hspace{20pt} (\mu m) \text{ distance from expanding sheet center to the sheet rim, tracing the sheet surface}

$r_0$ \hspace{20pt} (\mu m) \text{ initial droplet radius}

$r_b$ \hspace{20pt} (\mu m) \text{ radial location on deforming droplet sheet of hole perforation}

$r_c$ \hspace{20pt} (\mu m) \text{ cavity radius}

$R_c$ \hspace{20pt} (\mu m) \text{ expanding sheet radius of curvature}

$r_{cm}$ \hspace{20pt} (\mu m) \text{ coordinate of the expanding sheet center of mass relative to the } r

$r_d$ \hspace{20pt} (\mu m) \text{ spherical radial coordinate of initial droplet interior}

$Re$ \hspace{20pt} (-) \text{ Reynolds number}

$s$ \hspace{20pt} (\mu m) \text{ the length coordinate tangent to the droplet surface}

$St$ \hspace{20pt} (-) \text{ Strouhal number}

$t$ \hspace{20pt} (s) \text{ time}

$t_b$ \hspace{20pt} (s) \text{ time for first hole appearance relative to the last pulse in the burst}

$t_{b,err}$ \hspace{20pt} (s) \text{ time of the hole growth calculated from Taylor-Culick}

$t_f$ \hspace{20pt} (s) \text{ time of flight}

$t_t$ \hspace{20pt} (s) \text{ time until thin sheet criteria is reached for each wavenumber}

$t_{max}$ \hspace{20pt} (s) \text{ time when the deforming droplet sheet reaches its maximum rim radius}

$u$ \hspace{20pt} (m/s) \text{ velocity of the fluid relative to the fluid center of mass}

$\bar{u}$ \hspace{20pt} (m/s) \text{ mass mean velocity of droplet fluid after laser impact}

$u_0$ \hspace{20pt} (m/s) \text{ initial rim velocity of the expanding droplet sheet}

$u_c$ \hspace{20pt} (m/s) \text{ velocity of the expanding cavity wall}

$u_{cm}$ \hspace{20pt} (m/s) \text{ velocity of impacted droplet center of mass}

$U_f$ \hspace{20pt} (m$^3$/s) \text{ volume flux through the face, based on normal velocity}

$u_r$ \hspace{20pt} (m/s) \text{ rate of change in the sheet length } dR/dt

$u_R$ \hspace{20pt} (m/s) \text{ speed of expanding rim perpendicular to the laser center axis}

$u_q$ \hspace{20pt} (m/s) \text{ velocity of fluid of phase } q
Contents

\(u_x\) (m/s) velocity of the fluid along the direction of the sheet center axis

\(u_y\) (m/s) velocity of the fluid perpendicular to and away from the direction of the sheet center axis

\(V\) (m/s) fragment velocity

\(\nabla\) (m/s) total mass mean velocity of droplet fluid after laser impact

\(V_0\) (m/s) initial fragment velocity

\(V_c\) (m³) volume of cell

\(w\) (\(\mu\)m) \(e^{-2}\) beam waist radius

\(W\) (J) work

\(We\) (-) Weber number with \(\nabla\) as the velocity scale

\(We_s\) (-) splash Weber number with \(u_0\) as the velocity scale

\(We_{s,cr}\) (-) minimum splash Weber number necessary for sheet fragmentation

\(x\) (\(\mu\)m) lateral axis of the droplet position relative to the laser focus parallel to the laser direction

\(x_{cm}\) (\(\mu\)m) coordinate of the expanding sheet center of mass along the sheet center axis relative to the sheet center of curvature

\(y\) (\(\mu\)m) lateral axis of the droplet position relative to the laser focus perpendicular to the laser direction

\(z\) (\(\mu\)m) longitudinal axis of the droplet position relative to the laser focus perpendicular to the laser direction

\(Z\) (\(\mu\)m) deforming droplet cup depth

\(Z_a\) (-) atomic number

**Greek symbols**

\(\alpha\) (°) debris trajectory angle relative to the laser center axis

\(\alpha_d\) (°) mean droplet debris deflection angle

\(\alpha_f\) (-) acoustic geometric factor

\(\alpha_{lim}\) (°) maximum droplet debris trajectory angle

\(\alpha_m\) (neper/cm) acoustic attenuation coefficient

\(\alpha_{min}\) (°) minimum droplet debris deflection angle

\(\alpha_{max}\) (°) maximum droplet debris deflection angle

\(\beta\) (°) shadowgraph camera view angle relative to the laser center axis
Contents

\( \delta \)  (nm) ablation depth
\( \Delta \alpha \)  (°) debris spread angle
\( \Delta t \)  (\( \mu \)s) time delay between two pulses in a burst
\( \Delta t_f \)  (\( \mu \)s) shadowgraph flash duration
\( \Delta x \)  (\( \mu \)m) mesh spatial resolution
\( \dot{\varepsilon} \)  [s\(^{-1}\)] local strain rate
\( \eta \)  (\( \mu \)m) fluid surface perturbation amplitude
\( \Gamma \)  (-) gamma distribution
\( \kappa_q \)  (m\(^{-1}\)) surface curvature of phase \( q \)
\( \lambda \)  (nm) laser wavelength
\( \lambda_h \)  (\( \mu \)m) ruptured sheet perforation spacing
\( \lambda_p \)  (nm) lithography projection wavelength
\( \mu \)  (Pa s) dynamic viscosity
\( \nu_d \)  (kg/kg) mass fraction relative to the initial droplet volume
\( \omega \)  (s\(^{-1}\)) shock wave frequency
\( \omega_1 \)  (s\(^{-1}\)) wave mode growth rate for droplet surface acceleration phase
\( \omega_2 \)  (s\(^{-1}\)) wave mode growth rate for droplet surface during infinite medium conditions
\( \omega_3 \)  (s\(^{-1}\)) wave mode growth rate for droplet surface during thin film conditions
\( \phi_e \)  (\( \mu \)m) laser spot diameter \((1/e^2)\)
\( \phi_f \)  (\( \mu \)m) laser spot diameter (FWHM)
\( \psi \)  (\( \mu \)m) misalignment distance of droplet center with laser center axis
\( \rho \)  (kg/m\(^3\)) density of droplet fluid
\( \rho_\infty \)  (kg/m\(^3\)) density of the background gas
\( \rho_q \)  (kg/m\(^3\)) density of phase \( q \)
\( \sigma \)  (N/m) surface tension
\( \sigma_{cav} \)  (Pa) tensile stress seen by fluid at location of cavitation
\( \sigma_{G} \)  (rad) width of the pressure pulse on the droplet surface in radians
\( \sigma_{Sn} \)  (Pa) spall strength of fluid
\( \tau_B \)  (s) duration between the first and last peak in a laser burst
\( \tau_c \)  (s) capillary time of the droplet
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \tau_i )</td>
<td>(s) inertial timescale of the droplet deformation</td>
</tr>
<tr>
<td>( \tau_p )</td>
<td>(s) laser pulse duration (FWHM)</td>
</tr>
<tr>
<td>( \theta )</td>
<td>(rad) half-angle beam divergence</td>
</tr>
<tr>
<td>( \theta_0 )</td>
<td>(rad) angular coordinate of initial droplet fluid</td>
</tr>
<tr>
<td>( \Theta )</td>
<td>(rad) full angle of the expanding sheet rim relative to the sheet center of curvature</td>
</tr>
<tr>
<td>( \nu )</td>
<td>(-) volume fraction</td>
</tr>
<tr>
<td>( \nu_q )</td>
<td>(-) volume fraction of phase ( q )</td>
</tr>
<tr>
<td>( \nu_{q,f} )</td>
<td>(-) face value of the ( q^{th} ) volume fraction</td>
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Chapter 1

Introduction

The presented work is in the field of droplet-based laser produced plasma (LPP) light sources. The Laboratory for Energy Conversion (LEC) at the Swiss Federal Institute of Technology (also known as Eidgenössische Technische Hochschule (ETH) Zürich) has been engaged in the research and development of droplet-based LPP sources for over 10 years. The Applied Laser Plasma Science (ALPS) group is the research group within the LEC that focuses on this field and has been investigating the “preshaping” of the droplet target for the past five years. The preshaping of a liquid droplet target within a droplet-based LPP light source has been of growing interest in the field of EUV and soft x-ray light sources as a method for increasing the lifetime and efficiency of these sources. Optimizing this target preshaping process requires working knowledge of the laser produced plasma physics and fluid dynamics that governs it. Due to this need there has been a growing body of knowledge surrounding the effect of laser-induced shock waves interacting with droplets. This includes the work put forth here, which expands on experimental and theoretical work done by others in the field. The main conclusions of this work have shown that the recoil pressure of the expanding plasma acting on the fluid droplet surface determines the deformation pattern of the remaining unablated fluid. Therefore, by deliberate choice of the laser prepulse parameters irradiating the initial spherical target the droplet can be deformed into specific shapes and have fragmentation distributions that have the potential to substantially improve the light source lifetime and efficiency.
1.1 Droplet-based laser produced plasma sources

This work is done within the context of improving the operation and lifetime of droplet-based laser produced plasma (LPP) light sources. The sources enable the generation of high brightness extreme ultraviolet (EUV) light for use in the semiconductor industry. The development of extreme ultraviolet lithography (EUVL) has created the opportunity for the high volume manufacturing (HVM) of wafers with etched node sizes below 10 nm. EUV sources are a critical part of inspection tools used in the semiconductor industry. These inspection tools have a multitude of applications such as overlay measurements, critical dimension control, patterned and unpatterned wafer inspection, and mask inspection [1, 2]. There are challenges with the operation of these sources that stem from the droplet/laser interaction that provide fertile ground for new scientific research. These challenges will be discussed in the next sections.

1.1.1 Laser-produced plasmas

The development of peak laser powers upwards of $10^9$ W/cm$^2$ since the 1960’s has led to the opening of the field of laser-produced plasmas (LPP) [3]. Laser produced plasma is created when a target material is irradiated by intense laser radiation. This work is focused on laser produced plasmas in the range from $10^9 - 10^{13}$ W/cm$^2$, with pulse durations from $26$ ns to $\sim 40$ ps (FWHM). Within this range the plasma is generated through several processes that occur through the duration of the laser irradiation. First there is absorption of the laser radiation within the absorption depth of the target material, which is typically a fraction of the laser wavelength. The absorption layer will continue to heat up and melt and/or evaporate. In parallel and during the first several picoseconds of the pulse, when the laser pulse intensity reaches the threshold for multiphoton ionization, fast electrons are ejected from the material and ionization occurs transforming the vapor into a charged gas known as a “plasma,” also known as the fourth state of matter [4]. As the material vaporizes it is ejected away from the target surface creating a high pressure gradient that feeds vaporized material to the plasma. The laser intensity continues to increase and the plasma continues expanding. Eventually this
leads to the transition of the plasma as the main absorber of the laser energy shielding the target surface from the majority of the proceeding laser radiation. There is an electron density gradient that is high at the target surface and decreases towards the plasma. There is a critical density layer that corresponds to the laser wavelength where the majority of the radiation is reflected, shielding the target surface. The majority of the laser absorption process then occurs in the plasma due to inverse Bremsstrahlung (IB) absorption [5]. As energy is continually absorbed by the plasma, the plasma is remitting this energy as radiation, which continues to heat the target and expand the plasma. The plasma can reach peak temperatures upwards of 30 eV when being used to generate EUV [6]. The reemitted radiation from this hot dense plasma is one of the major byproducts of LPP’s that drives the interest of this field. During this process the plasma can generate very high recoil pressures potentially reaching peaks in the $10^{12}$ Pa range on the target surface. This recoil pressure transfers momentum to the target surface. The absorption and remission of laser energy by the plasma will continue in a quasi-stable state until the laser irradiation ceases. After the irradiation ceases the plasma will continue to expand and cool.

LPP’s have a wide range of applications from nuclear physics (for inertial confinement fusion), material science, soft X-ray microscopy, and semiconductor manufacturing [7, 8, 9, 10]. Plasma-based light sources have been particularly advantageous for generating radiation that cannot be generated with conventional means. Light wavelengths in the extreme ultraviolet (EUV) and soft x-ray region are highly absorbed by all matter and therefore need to be generated in a vacuum. This property also precludes the use of refraction optics eliminating many conventional means of generating radiation such as lamps or lasers. The most reliable method for generating this radiation is either through synchrotrons or discharge plasmas. Synchrotrons have a high cost of ownership compared to LPP light sources and discharge produced plasmas cannot reach the source brightness or stability compared with LPP sources [6].
1.1. Lithography in semiconductor manufacturing

In semiconductor manufacturing the lithography process is used to create integrated circuits (IC), i.e. “microchips.” This is done through the repetitive copying of submicrometer spatial patterns into interconnected layers of semiconductor material, which form an electronic structure \[11\]. The copying of these submicrometer patterns is done by projection of said pattern onto a wafer surface. A function diagram of an optical projection lithography system is shown in Fig. 1.1. The system consists of a light source, such as an ArF (193 nm) laser and a condenser (represented as a single lens) with a numerical aperture of \( NA_{\text{cond}} = \sin(\theta_{\text{cond}}) \), which illuminates a mask pattern attached to a transmissive reticle. The objective focusing optics (represented as a single lens) collects the light scattered from the mask within \( NA_{\text{obj}} = \sin(\theta_{\text{obj}}) \) and focuses the mask image onto the wafer with demagnification of typically 4:1 \[11\]. Using this lithography process in conjunction with various chemical processes IC’s are manufactured in vast quantities.

The methods for building the semiconductor layers on IC’s can be grouped into four general categories of deposition, removal, modification of electrical properties, and patterning. For deposition material is transferred to the
1.1. Droplet-based laser produced plasma sources

wafer, which can be done through physical vapor deposition, chemical vapor deposition, electrochemical deposition, and others. The removal process is any process that removes material from the wafer, such as through etching. Modification of electrical properties is the process of doping transistor sources and drains. Patterning or lithography is the altering of the deposited material, which is typically called a resist. There are positive and negative resists. When a positive resist is used, the exposed material is intended to be dissolved away leaving the unexposed resist. When a negative resist is used, the exposed material will remain and the unexposed will be dissolved away [12]. A sketch of the dry etching process is shown in Figs. 1.2(a) - 1.2(g) as an example of a typical process for creating a structure on a Si wafer. In Fig. 1.2(a) the wafer with the deposited oxide layer is cleaned and prepared usually by elevating its temperature in order to remove moisture. In Fig. 1.2(b) the photoresist is applied using spin coating and then prebaked to remove excess resist solvent. In Fig. 1.2(c) a photomask is aligned with the wafer, which is transmissive to the light of wavelength $\lambda_p$ in the desired etching pattern. In Fig. 1.2(d) the photoresist is exposed to shorter wavelength light through the mask, selectively exposing the resist. The light changes the chemical structure of the resist, which for positive resists allows the removal of the exposed areas with a solvent leaving the remaining area as depicted in Fig. 1.2(e). Figure 1.2(f) shows the result of the etching of the oxide layer that is not protected by the resist. The etching process involves the application of a chemical agent that can either be liquid (“wet” etching) or with a plasma (“dry” etching). The final part of the process shown in Fig. 1.2(g) is the resist removal, which can be done using a “resist stripper”, which chemically alters the resist making is possible to remove from the substrate or else an oxidizing plasma can be used [13].

The features that can be printed onto the photoresist during the exposure process are limited by the diffraction limit of the exposure light. This is quantified as the minimum achievable line width $L_w$ for the process determined by [11]

$$L_w = k_l \frac{\lambda_p}{NA},$$

(1.1)

where $k_l$ is the lithography process constant, $\lambda_p$ is the projection light wavelength, and $NA$ is the numerical aperture. From this definition it can be seen that $\lambda_p$ is a critical factor in determining the amount of electrical com-
1.1. Droplet-based laser produced plasma sources

Figure 1.2: A sketched representation of the wet/dry etching process with a positive photoresist, which is used in semiconductor lithography. The representation of the stages of the process are shown as (a) layer preparation, (b) application of the photoresist, (c) alignment of the photomask, (d) exposure of the mask pattern to the photoresist with the lithography projection wavelength $\lambda_p$, (e) the removal of the exposed resist, (f) the etching of the oxide layer in the area not protected by the resist, and (g) finally the removal of the resist [13]. The linewidth $L_w$ represents the minimum feature size the process is capable of etching.
1.1. Droplet-based laser produced plasma sources

Figure 1.3: Microprocessor Transistor counts for 1971-2011 and Moore’s Law [15].

Components that can be concentrated onto an IC. Moore’s Law has stated that the number of transistors per volume that can fit in an IC should double every 2 years [14]. This law has been a major goal setter for the semiconductor industry since the 1970’s (see Fig. 1.3). A large part of the advancement of Moore’s Law has been due to reduction in $\lambda_p$ in semiconductor manufacturing. Lithography light sources for the high volume manufacturing (HVM) have typically been lasers, with the latest being 193 nm Argon Fluoride excimer lasers. Alternatives for decreasing $\lambda_p$ have included 157 nm Fluorine (F$_2$) lasers, but the mirror manufacturing for this wavelength is costly for marginal benefit [11]. Synchrotrons are capable of providing very short wavelength light, but they are very costly and occupy a large foot print.

One of the latest movements in the industry for the reduction of the stan-
standard $\lambda_p$ for HVM is extreme ultraviolet (EUV) light at 13.5 nm within a 2% bandwidth. This wavelength has been chosen based on the creation of Molybdenum/Silicon multilayer (ML) mirrors, which are capable of efficiently reflecting 13.5 nm with a theoretical maximum efficiency of 70% utilizing Bragg reflection [16].
1.1. Droplet-based laser produced plasma sources

Figure 1.4: Function diagrams of two types of EUV sources. (a) A discharge produced plasma (DPP) source with a grazing incidence collector and (b) an LPP source with a normal incidence collector [16]. Both systems require a debris mitigation system between the plasma and the collector.
1.1.3 EUV lithography

EUV light has the advantage of having a $\lambda_p$ that is an 15x shorter than the current wavelength used in semiconductor HVM. According to Eq. (1.1) this would allow for the printing of $\sim 15x$ smaller feature sizes on a silicon wafer. There are several challenges that are traded in exchange for this short wavelength. One major challenge is the high absorptivity of EUV radiation by all matter. Therefore, the entire process path from the light source to the object wafer needs to be in a vacuum. The light is collected and focused by ML mirrors, which direct the light to the mask and wafer, following the same principle shown in Fig. 1.1 except with only reflective optics. There are many auxiliary processes that must be adapted in parallel with the EUV lithography setup. This includes the manufacturing of the reflective ML optics at exceptional tolerances for surface roughness and the inspection of the mask for defects at the actinic wavelength.

There are several types of mask inspection methods. These include actinic blank inspection (A-BI) for checking the quality of multilayer stack, actinic pattern inspection (A-PI), and aerial image measurement system (AIMS) to determine defect printability [17].

As is shown in Fig. 1.1 the first component in an optical lithography system is the light source, which generates radiation at $\lambda_p$. The work researched in the LEC ALPS group is focused primarily on sources. Specifically, light sources for EUV and soft x-ray. Two functional diagrams of EUV source layouts are shown in Figs. 1.4(a) and 1.4(b). The two major methods that have been explored to generate EUV have been discharge produced plasmas (DPP) and LPP. Figures 1.4(a) and 1.4(b) also show the two major approaches to collector design, which are the grazing incidence and normal incidence collectors.

A current example of an EUV source that is on the market today is the TWINSAN NEXE:3400B manufactured by ASML and shown in Fig. 1.5 [18]. This source operates using a Sn droplet based LPP with a normal incidence ML collector. It uses a CO$_2$ laser as the pump laser. Once the EUV is generated at the plasma it is focused by reflective condenser optics to the reticle. Sn is used as a fuel, because it has an efficient line emission at 13.5 nm [16]. The industry is heavily focused on droplet-based LPP sources, because there are several advantages to using droplet fuel targets that will be
elaborated on in the following section.

1.1.4 Droplet based laser produced plasmas

Droplet-based LPPs use micrometer-sized liquid droplets as the fuel target. The main advantage of droplet targets is that they consist of a regenerative and mass limited target. A regenerative target translates to a very high source stability and a mass limited target translates to a minimization of the excess fuel debris proceeding the plasma generation [19, 20]. The radiation generation process with a mass limited target follows the same sequence described in Sec. 1.1.1. For the generation of EUV in a droplet based LPP source the droplet ablation process starts with a pulsed nanosecond laser heating the liquid Sn droplet surface. For metallic targets generally the laser light is absorbed by the electrons of the conduction band. As the electrons are heated, electron-phonon coupling transfers energy to the metal lattice. The laser energy is absorbed between the target surface and the skin depth of the material [21]. Since the thermal diffusivity is typically low in comparison to the nanosecond timescales of the laser pulse, only a $< 1 \mu m$ layer of the target material is expected to be heated [22]. The temperature of the heated
material increases until a phase transition occurs leading to ablation of the surface layer. Most of the absorbed laser energy energizes free electrons, which then ionize neutral vapor atoms leading to a state of plasma. The main plasma heating mechanism in the nanosecond pulse regime is Inverse Bremsstrahlung (IB) absorption [19, 23].

There have been many studies done exploring which fuels are capable of generating line emission in the 13.5 nm ±2% bandwidth range. These fuels include Sn, Xe, and Li [24, 25, 26, 27]. Sn is found to have the highest conversion efficiency [28, 29, 30]. The temperature range of the plasma needed in order to emit this wavelength efficiently is between 30 - 40 eV, which can be generated with 1.06 μm laser irradiances on the order of 10^{11} W/cm² or 10.6 μm (CO₂ laser) laser irradiances on the order of 10^9 - 10^{10} W/cm² [31, 22, 32, 33, 34, 35, 36, 37].

1.1.4.1 Plasma ablation pressure

The rapid vaporization and heating of the plasma leads to a sharp pressure gradient. The pressure gradient drives the expansion of the plasma plume into the vacuum. This plume expansion interacts with the remaining liquid target material, breaking it into fragments which are propelled normal to the droplet surface [23, 38, 39]. The peak pressure reached by the plasma is known as the “ablation pressure”. This property has been studied extensively for nanosecond timescales in relation to LPP generated shock waves in condensed matter [40, 41, 42]. Harrach et al. [40] derived a relation for ablation pressure $P_{max}$ based on 2D hydrocode calculations that is fit for an irradiance range of $I_0 = 10^{13} - 10^{15}$ W/cm²

$$P_{max} = 8 \left( \frac{I_0}{10^{14}} \right)^{0.7} \left[ 1 + \frac{l_{ac}^{(0)}}{w} \frac{\sin(\theta)}{\tau_p} \left( \frac{I_0}{10^{14}} \right)^{0.3} \right]^{-1.4},$$

where $P_{max}$ is in units of Mbar, $w$ is the $1/e^{-2}$ beam waist radius in μm, $\tau_p$ is laser pulse duration in ns, $I_0$ is the laser nominal peak intensity in W/cm² and is $I_0 = E_p/(\tau_p \pi w^2)$, $E_p$ is the laser pulse energy, $l_{ac}^{(0)}$ is the nominal distance between the ablated surface and the critical density surface in μm, and $\theta$ is the half-angle beam divergence. For picosecond laser pulses acting
on Sn droplets the peak ablation pressure $P_s$ for an individual laser pulse has been derived as \[43, 44\]

\[ P_s = 0.4 \left( \frac{I_0}{\lambda} \right)^{2/3} \left( \frac{A_a}{2 Z_a} \right)^{1/3}, \]  

(1.3)

where the pulse irradiance is $I_0$ in TW/cm$^2$, $\lambda$ is the laser wavelength in $\mu$m, $A_a$ is the atomic mass, and $Z_a$ is the atomic number.

### 1.1.4.2 Plasma debris

Mass limited targets not only limit the fuel consumption, but also limit the amount of generated plasma debris. When the droplet target is irradiated by the pump laser the plasma is produced and the fuel is dispersed as debris. Plasma debris must be mitigated to prevent the contamination of the optics or any other critical surfaces, such as sensors, of the LPP light source. In general, debris is grouped into three classes for droplet-based LPP; high-energy ions, low energy neutral particles, and neutral clusters composed of liquid or solid fuel fragments \[19, 45\]. The formation and dynamics of the plasma debris has been a major focus in the ALPS group. This work focuses exclusively on the formation and distribution of neutral clusters. Neutral clusters are formed when the expanding plasma plume interacts with the remaining unablated liquid fuel with a recoil effect, accelerating the droplet surface \[23, 38, 39\]. The accelerated droplet expands away from the plasma zone forming splashes of liquid fuel.

### 1.1.4.3 Debris mitigation methods

In the ALPS laboratory in the LEC-ETH Zürich debris behavior has been studied extensively \[46, 45, 47\]. Debris mitigation systems at the LEC and elsewhere have been designed to reduce the contamination of optics and sensors from plasma debris. Ions have been mitigated through the use of a background gas at a certain vacuum level \[48, 49\] or by magnetic fields \[50, 51\]. Unlike charged ion debris, which hold the possibility of electro-
magnetic methods of debris mitigation, neutral debris does not hold a charge that can be used to repel it. Fast neutrals that result from ions recombining with high density background gas are still a major challenge as they are not easily deflected away from sensitive optics [52]. At the LEC fast neutrals have been mitigated with supersonic jet curtains [53].

Neutral cluster debris mitigation has typically been done with either gas curtains that deflect the fragments or gas etching [54, 23]. Both approaches are less effective against larger neutral clusters. Tin deposition onto optics from neutral clusters has been mitigated by etching the debris from the surface with hydrogen gas creating tin hydride. These systems are less effective against larger particles [54, 55, 56, 57]. It can be expected that a larger particle splash will require a higher etch rate to keep the optics clean due to the smaller ratio of etching surface to mass. These etching systems are also very complex and one would prefer to avoid them. For gas curtains the larger fragments are deflected less than the smaller fragments and therefore less effective against larger splash particles [58, 59, 54]. The required velocity \( u_{\text{def}} \) and density \( \rho_\infty / \rho_l \) for deflecting gas curtains scales with fragment diameter \( d \) on the order of \( u_{\text{def}} \propto \sqrt{d} \) and \( \rho_\infty / \rho_l \propto d \) [60]. Larger splashes that deposit on surfaces also require longer periods of etching for the same etch rate [59, 54]. To improve the current debris mitigation techniques there is a need to understand which relations govern the neutral cluster breakup and how to reduce the size and number of these fragments.

### 1.2 Droplet deformation

A body of research has been produced in recent years investigating the dynamics of the droplet deformation from a single laser pulse. Work that has been done already in an effort to map out the dynamics of this phenomena have included droplets hitting an impactor and expanding in free space. Villermaux and Bossa [61] determined experimentally the dynamic equations for a droplet hitting an impactor with an initial velocity \( u_0 \) and an initial droplet diameter \( d_0 \), and splashing in free space. The droplet crushes on the impactor and spreads out radially and perpendicular to the impact surface as a fluid film with a radius \( R(t) \). The asymmetric balance between the inertia
1.2. Droplet deformation

Figure 1.6: The splashing droplet dimensions are approximated as an expanding thin disc of radius $R$ and thickness $h$ with equivalent volume to the initial droplet.

of the expanding fluid and capillary restoring forces leads to a trajectory of the rim that can be described by [61]

$$\frac{R - r_0}{r_0} = \sqrt{\frac{W_e \ t}{2 \ \tau_c}} \left(1 - \frac{\sqrt{3}}{2} \ \frac{t}{\tau_c}\right)^2,$$

where $W_e$ is the Weber number defined as $W_e = \rho d_0 u_0^2 / \sigma$, $\rho$ is density, $\sigma$ is surface tension, and $\tau_c$ is the droplet capillary number defined as $\tau_c = \sqrt{\rho d_0^3 / (8 \sigma)}$. The maximum radius $R_{max}$ is found with $dR(t_{max})/dt = 0$ to be $t_{max} = 2/(3\sqrt{3}) \tau_c \approx 0.385 \tau_c$. This model uses an approximation of the splashing droplet dimensions (see Fig. 1.6) as an expanding thin disc of radius $R$ and thickness $h$ with equivalent volume to the initial droplet where

$$h \approx \frac{d_0^3}{6R^2}.$$

There have been computational and analytical investigations of the deformation of the droplet due to laser-pulse impact by Gelderblom et al. [62].
1.2. Droplet deformation

Figure 1.7: The kinetic energy partition formulated by Gelderblom et al. [62] as a function of the pressure impulse size $\sigma_G$.

In this work Gelderblom et al. [62] formulates the distinction between the kinetic energy of the deforming fluid relative to the fluid center of gravity $E_{K,d}$ and the kinetic energy of the fluid center of gravity $E_{K,cm}$, denoted as the kinetic energy partition. Together these two values are the total impact kinetic energy $E_K = E_{K,d} + E_{K,cm}$. The kinetic energy partition was found to be a function of the pressure profile size on the droplet surface, which is approximated in this work from a polynomial fit of the analytical solution as

$$\frac{E_{K,d}}{E_K} \approx -1.2\sigma_G^4 + 4.6\sigma_G^3 - 5.5\sigma_G^2 + 1.3\sigma_G + 0.91,$$  \hspace{1cm} (1.6)

where $\sigma_G$ is width of the pressure pulse on the droplet surface in radians (see fig. [1.7]). The initial velocity field for a droplet impacted in the incompressible regime was also formulated and is described in detail in Sec. 5.1.2.

Experiments have been done with inked water droplets shot with a laser pulse by Klein et al. [63]. This work showed experimentally that the droplet impacted by a short impulse will follow similar dynamics to (1.4). It quantified the momentum coupling mechanism between the laser impulse and the
droplet as the work transfer to the fluid from the vaporization recoil pressure. In the case of surface vaporization, the mass mean velocity $V$ was proportionate to the laser pulse energy $E_p$. Further work was published for liquid Sn droplets impacted by a laser impulse by the author of this work [60]. In this publication a model of the momentum coupling mechanism to the droplet from a laser ablation plasma is experimentally validated. This model is described in detail in Chap. 3.

The droplet deformation in the compressible regime was investigated analytically by Reijers et al. [64]. They determined that the droplet shape evolution will alter considerably, keeping momentum transfer constant, as the pulse duration is reduced into the compressible flow regime.

The latest work extends into the dynamics for picosecond and femtosecond laser pulses. Basko et al. [65] conducted experiments and numerical simulations of the droplet interaction with a single laser pulse of varying pulse durations between 50 fs and 5.3 ps. This work details the cavitation and spallation that occurs within the droplet by the focused shockwave generated by these ultrashort pulses. Krivokorytov et al. [44] investigates similar dynamics for a 0.8 ps pulse irradiating a 56 μm droplet. They use the velocity doubling rule in order to estimate the tensile stress within the droplet at the moment of rupture [66].

\[ v_r = 2u = \frac{2P_r}{\rho c_s}, \]  

where $v_r$ is the speed of the body surface after the release of the shock wave at the free surface, $u$ is the mass speed of the material behind the shock front, and $P_r$ is the pressure at the front of the shockwave [67]. To the knowledge of the author there has been no investigations of the droplet deformation due to bursts of laser pulses, beyond a recent investigation with the droplet deformation from a picosecond pulse pair, which demonstrated the deformation of the droplet into an “acorn-like” shape [68].
1.3 Droplet fragmentation

Knowing the parameters that dictate the fragment size and distribution of the neutral clusters is of key importance in optimizing debris mitigation within droplet-based LPP sources. One of the foundational works that is an important step in this understanding is by Villermaux and Bossa [61] where the fragment distribution of droplets hitting an impactor and expanding in free space is measured and correlated to $We$. For this work the impacted droplet expands as a radial expanding sheet with a rim accumulating mass. The rim radius reaches a maximum $R_{\text{max}}$ where the capillary forces retracting the sheet overcome outwards radial momentum. Before $R_{\text{max}}$ is reached the rim is being stretched, which dampens the growth of corrugations on the rim. After $R_{\text{max}}$ is reached the corrugations grow into ligaments retracting the sheet inwards towards the center. The ligaments extend out radially from the center of the impact and fragment into droplets with a mean diameter $d_{10}$ of $d_{10} \sim d_0 We^{-1/2}$. The fragment size distribution follows a Gamma distribution, which is covered in more detail in Sec. 5.2.4. The work by Villermaux and Bossa [61] provides key principles to aid in the approach to droplets fragmented by a laser induced shockwave, but there are some key differences that will be explored in this work. Namely the droplet deformation will be dictated by the kinetic energy partition [62] and a significant part of the droplet fragmentation will occur due to rupture of the droplet as an expanding sheet. The sheet rupture dynamics are investigated in depth in Chs. 4 and 5.

Bremond and Villermaux [69] conducted an experimental investigation of bursting thin films, which was used to validate a Rayleigh-Taylor instability model to predict the rupture time and perforation spacing of the sheet. The time of the first hole appearance was found to scale with $We^{-1/2}$. A similar analysis for the droplet sheet rupture time is done in Sec. 4.4. Other investigations into first hole perforation time have been done by Klein [70], Kurilovich et al. [71], where the rupture time is shown to have an exponential dependence on the impact Weber number.

Other nonlinear effects can play a major role in the droplet target fragmentation dynamics. Especially with fragmentation from picosecond and femtosecond pulses. These effects include cavitation and spallation of the droplet fluid.
due to the propagation of higher Mach number (\(Ma\)) laser induced shockwaves. Grigoryev et al. [72] recently showed how a shockwave induced by a picosecond laser pulse irradiating a droplet can generate a tensile stress that will cavitate the droplet fluid if it exceeds the spall strength. A cavity may form in the interior of the droplet and could also cause spallation of the droplet rear wall if the rarefaction wave is intense enough. Grigoryev et al. [72] also demonstrated that molecular dynamics (MD) simulations can play an important role for determining the phase transitions during the shockwave propagation. The MD simulations aid in determining the spall strength of the shocked material depending upon the local strain rate and the fluid parameters, which in turn determines the location and expansion of the induced cavity. MD simulations are very useful for calculating these threshold rupture states of the fluid, but they are limited both spatially and temporally [73]. This prevents them from being used for simulating the complete droplet fragmentation process without advances in computing power. For modeling the complete droplet fragmentation, the smoothed particle hydrodynamics (SPH) method has been investigated and shows results that are comparable with experimental work. Grigoryev et al. [74] used the SPH method to simulate the cavitation of a 2 \(\mu\)m diameter droplet from a 10 fs laser pulse. Koukouvinis et al. [75] used a similar SPH method to simulate the cavitation of a small 50 \(\mu\)m diameter droplet from an ultrashort laser pulse. The deformation dynamics of a shocked droplet have shown good comparison to similar experimental results, but to the author’s knowledge there has been no work published comparing the predicted fragment size distribution from these simulations to experiment results.

### 1.4 Prepulsing

One of the major methods being developed in the LPP source field for increasing the debris mitigation and CE is the implementation of prepulsing as a method of changing the target shape and density profile before the main laser pulse interacts with target. Nishihara et al. [76] proposed the prepulse scheme in 2008 for increasing the CE of a CO\(_2\) laser plasma. Fujimoto et al. [77] later showed an increase in CE from <2% to >4.5% with the use of a 10 ps prepulse and a CO\(_2\) main laser. Schafgans et al. [55] has also shown an
increase in CE by a factor of 4 with the use of a prepulse and a CO$_2$ main laser in a master-oscillator power amplifier (MOPA) configuration. Prepulsing is also used as a method of reducing the size neutral clusters and for deflecting the neutral clusters away from sensitive optics, but the published experimental research on these phenomena is sparse [78].

1.5 Motivation

Droplet-based LPP x-ray light sources are a key technology in the future of semiconductor manufacturing. At the heart of these systems is the interaction between a fuel droplet and a high energy laser pulse. The efficiency of this process and the spectrum of debris emanating from it are critical factors in terms of determining the lifetime and cost of ownership of these sources. A working knowledge of this process is essential for pushing the performance of these sources.

The deformation and fragmentation of a droplet impacted by a laser pulse is a critical piece of this knowledge for two reasons. The first reason is the prediction of the neutral cluster distribution based on the system operating parameters. Debris mitigation systems are more efficient at removing or deflecting neutral clusters that are smaller. Knowledge of how the input laser parameters effect the trajectory of the neutral clusters is also critical for insuring that the flux of neutral clusters to sensitive optics is minimized. The second reason for mapping out this knowledge is to determine what laser parameters are required in order to create a specific intended target shape for target preshaping. Preshaped targets have the potential to reduce the mean neutral cluster size, debris flux, and they have the potential to increase the coupling between the main laser pulse and the target. The ultimate goal of the target preshaping is the improvement of key source parameters, which are a decrease in neutral cluster debris, an increase in conversion efficiency, and an increase in source stability.

The motivation of this work is to map out the deformation and fragmentation dynamics of a droplet impacted by one or more ablating laser pulses. Identifying the causal relationship between the laser pulse parameters and the droplet shape after impact terminating in a distribution of neutral clusters provides a novel contribution to the scientific community.
1.6 Research objectives

The project scope consists of exploring various irradiation schemes that can influence droplet target dynamics in an LPP light source. Specifically, this work seeks to determine which schemes are relevant for application in metrology applications and EUV science. A method needs to be developed for characterizing the droplet breakup dynamics. It must be determined which physical factors influence droplet debris distribution and what methods may be available for preshaping the target in order to optimize the source performance. This is to be done in service of the greater goal with the field of droplet-based LPP light source development, which is to increase the coupling of the droplet debris to the source debris mitigation systems and to increase the source stability and conversion efficiency.

The major approaches that are investigated involve the irradiation of liquid metal droplets with one or more laser pulses of varying energy and duration from the nanosecond to the picosecond scale. Shaped bursts of picosecond pulses are of special interest to this work as they have higher order effects due to the energy distribution within the burst. It is expected that these higher order effects will prove to be a useful tool in accessing target shapes not achievable with single pulses.

The experimental work utilizes both the ALPS I and the ALPS II prototype sources with all of their available control and diagnostic systems. A major tool that is used and improved upon in this work is the high speed shadowgraph imaging system, which is capable of capturing the shape and velocity of the deforming droplet target.

The priority goal of this work is to derive a generalized model of the droplet deformation and breakup given known parameters of the irradiating laser pulse(s). Theoretical and computational modelling are to be performed to explain the knowledge gained by the experimental work. The various models developed are one dimensional, 2-dimensional axisymmetric, and 3-dimensional in time, depending upon the resolution required for the modeled phenomena. A methodology is to be developed whereby the optimum target irradiation conditions can be found for achieving a specific target shape depending on the target parameters such as droplet size, the fuel material, and laser type.
1.7 Thesis outline

This work is organized such that it follows the sequence spanning the initial laser irradiation and subsequent momentum transfer with the droplet, to the deformation of the impacted droplet, and finally to the fragmentation of the droplet.

Chapter 1 provides the context for this work in its scientific field and its relevance for industry. It also gives an informative overview of the previous scientific works that this work builds from.

Chapter 2 presents the main experimental apparatus used to acquire the presented experimental results.

Chapter 3 describes an analytical model for predicting neutral cluster debris momentum and trajectory for single laser pulses with a duration of nanoseconds. Experimental results are shown that validate this model.

Chapter 4 describes an analytical model for predicting the deformation and rupture time of a droplet irradiated by a burst of picosecond pulses. Experimental results are shown that validate this model. This model provides a basis for Chapter 5.

Chapter 5 presents simulations of a droplet fragmented by a laser induced shockwave. The analytical model derived in Chapter 4 is extended to predict the fragment size distribution of the impacted droplet based on the simulation initial conditions. The analytical model of the droplet fragment distribution is validated by the simulation results.

Chapter 6 concludes the presented work and provides several suggested directions for future work in the field.
Chapter 2

Experimental Apparatus

The ALPS laboratory comprises two droplet-based LPP light sources [20, 17]. The LPP source prototypes are named ALPS I and ALPS II. The experimental results presented in Chap. 3 were acquired in both ALPS I and ALPS II using laser pulse durations on the order of nanoseconds. The experimental results presented in Chap. 4 were performed exclusively within ALPS II, but with laser pulse durations on the order of picoseconds. Most of the fluid dynamic experimental results in this work were acquired using a high speed shadowgraph system, which capture the shape and velocity of the deforming droplets upon impact from the laser produced plasma.

2.1 ALPS I experimental setup

The ALPS I test facility consists of a spherical vacuum chamber, a vacuum pump, a droplet dispenser mounted to a 3D positioning system, and a 10 Hz laser system. The photograph of the ALPS I test facility is shown in Fig. 2.1 and a schematic representation of the interior is shown in Fig. 2.2.

This test facility is operated with an Nd:YAG laser, with a repetition rate 10 Hz and a typical pulse duration of 10 ns and a pulse energy up to 300 mJ per pulse. The laser pulse energy for this system is adjustable between 100 $\mu$J to 300 mJ through the use of adjustable polarizing beam splitters. The laser beam is focused within the vacuum chamber through a plano convex lens to spot sizes of 35-45 $\mu$m, which provide an irradiance range from $8 \times 10^8$
2.1. ALPS I experimental setup

Figure 2.1: The ALPS I test facility consists of a spherical vacuum chamber with a droplet dispenser located within it. The 10 Hz Nd:YAG laser sits on an optical table located on the left side. The monitoring, data acquisition systems, and user interface are located on the right side.

Figure 2.2: A schematic representation of the ALPS I experimental setup.
to $2.4 \times 10^{12} \text{W/cm}^2$. The vacuum levels typically maintained in this system are $\sim 0.1 \text{ mbar}$ with Ar background gas.

2.2 ALPS II Setup

The main prototype droplet-based LPP light source in the LEC is the ALPS II prototype source, which is shown in Fig. 2.3. This source is equipped with a large capacity droplet dispenser and a high power (kW), high repetition rate Nd:YAG laser operating at the fundamental wavelength of 1064 nm. The Nd:YAG laser has an average power of 1.6 kW and operates at frequencies up to 20 kHz, with pulse energies of up to 200 mJ, and pulse durations down to 24 ns. The laser beam is focused into the vacuum chamber through a plano convex lens to spot sizes of 50-70 $\mu\text{m}$. The image of the laser spot is shown in Fig. 2.4 with the Gaussian profile plotted in Fig. 2.5. The peak irradiance for each laser pulse is up to about $2 \times 10^{11} \text{W/cm}^2$ [79]. The vacuum levels typically maintained in this system are $<0.1 \text{ mbar}$ with Ar background gas.

2.2.1 Droplet dispenser

The droplet dispenser generates a mono-dispersed and coherent stream of droplets as shown in Fig. 2.6. The droplet dispenser is mounted to a 3-D motion stage and coupled to a closed loop control system able to keep the position of the droplet train at the main laser focus position. The control system compensates for lateral instabilities with a spatial resolution of $\pm 0.5 \mu\text{m}$ [80]. A piezoelectric actuator determines the droplet frequency [81].

2.2.2 Burst picosecond laser

A custom built laser system is utilized in this work that is capable of generating bursts of ultrashort laser pulses. The laser is a master oscillator pulsed amplifier (MOPA) Nd-doped Vanadium (Nd:VAN) system with a wavelength of 1064 nm. A schematic representation of the system is shown in Fig. 2.7.
Figure 2.3: The ALPS II test facility consists of a cuboid vacuum chamber with a large capacity droplet dispenser mounted within it. The 1.6 kW Nd:YAG laser sits on an optical table located within an enclosure behind the vacuum chamber shown in the foreground. The monitoring, data acquisition systems, and user interface are located on the left side.
2.2. ALPS II Setup

Figure 2.4: Normalized image of the laser spot of the ALPS II 1.6 kW Nd:YAG laser.

Figure 2.5: Normalized intensity profile of the laser spot of the ALPS II 1.6 kW Nd:YAG laser. The profile follows a typical Gaussian shape.
The master oscillator is a pulsed laser diode that generates pulses with a duration of $\tau_p = 43$ ps (FWHM). This laser diode has a trigger jitter of $< 10$ ps and is capable of repetition rates from single pulses up to 100 MHz. Coupled to the solid-state amplifiers, the system is capable of reaching single pulse energies up to $E_p \leq 2$ mJ. An acousto-optic modulator (AOM) is placed before the amplifier stages in order to clip bursts of pulses emitted from the laser diode. The clipped burst is then amplified, reaching burst energies up to $E_B \leq 2.7$ mJ at the laser output window. A programmable arbitrary waveform generator with an output frequency of 1 GHz creates the analog signal that controls the AOM clipping, which is synchronized with the laser diode trigger and the shadowgraph system. The AOM response time from closed to full open is $\sim 90$ ns. For the tests in this work the laser spot was focused to a spot diameter of $\phi_f = 22 \, \mu$m (FWHM).
2.3 Shadowgraph imaging system

The droplet fragmentation process in this work was imaged using a high speed shadowgraph system. A schematic representation of the typical shadowgraph layout for imaging droplets is shown in Fig. 2.8. The shadowgraph system consists of a high speed camera (SONY ICX625ALA/AQA) capable of up to 20 fps. The high speed flash is a high powered LED pulsed with a pulse duration of $\Delta t_f = 0.5 - 2\mu s$ FWHM. The system trigger signals for the main laser, camera, and flash are synchronized such that during the source operation the delays between these signals can be changed with 0.25 $\mu s$ resolution.

The fragment speeds in this work can exceed well over 100 m/s [60]. When small fragment sizes reach high speeds, they can approach the limit of the shadowgraph system to record, depending upon the hardware and setup limitations of the shadowgraph. This sensitivity range can be quantified analytically. The imaging resolution of the shadowgraph system will be dependent upon the length scale of the imaged features $L_f$, the velocity $V_\alpha$ of the imaged features, and the duration of the shadowgraph flash $\Delta t_f$ according to the relation

$$L_f = \Delta t_f V_\alpha \frac{N_s\text{SNR}}{n_{fg} - n_{bg,0} - N_s\text{SNR}},$$  

(2.1)
2.3. Shadowgraph imaging system

Figure 2.8: Typical shadowgraph experimental layout utilized in this work. The camera and flash are timed such that droplet shape is captured for a predetermined time after the laser pulse irradiates the droplet.

where $N_s$ is the image noise level (see Fig. 2.9), SNR is the minimum acceptable signal-to-noise level, $n_{fg}$ is the signal level in the foreground, and $n_{bg,0}$ is the nominal signal level in the background. Equation (2.1) infers that there is minimum length scale $L_f$ moving at a velocity perpendicular to the image plane $V_\alpha$, which is capable of being imaged above the minimum SNR for a given test. The shadowgraph hardware configuration will determine the values of $\Delta t_f$, $N_s$, $n_{fg}$ and $n_{bg,0}$, which ultimately determines the particle range that will be detected. Droplet fragments traveling at $V_\alpha$ with a cross section smaller than $L_s(V_\alpha)$ will be invisible to the shadowgraph system. This is illustrated in Fig. 2.10 where the range of detectable length scales are highlighted. The light purple region represents the length scales that can be imaged at $\Delta t_f = 2\mu s$. By reducing this value to $\Delta t_f = 0.5\mu s$, while leaving the other parameters constant, the range of visible length scales are substantially increased. Other methods for increasing this range include the reduction background image noise $N_s$, as well as increasing the difference between $n_{fg}$ and $n_{bg,0}$ by providing more energy into the flash.
Figure 2.9: A plot of the intensity across a line of a shadowgraph image.

Figure 2.10: The range of visible fragment sizes the shadowgraph is capable of detecting according to Eq. (2.1). This range will also be limited by the image pixel size. By decreasing $\Delta t_f$ this range can be increased substantially.
Chapter 3

Droplet deformation from a nanosecond laser pulse

One of the major approaches to investigate the debris generation process and spatial expansion over time has been to image the produced debris with different time delays from the main laser pulse. From the time resolved images one can determine the speed, the distribution and the scale of the droplet fragments. The most typical technique to image debris is the shadowgraph technique with a fast exposure. Other methods include LIF and PIV systems [54, 82]. Most of these experiments have been done with planar targets or solid droplet targets [82, 83, 84, 85]. Limited studies have been done to examine the breakup dynamics of liquid droplets during continuous source operation mode, looking mainly at target cross-section expansion and distribution [77, 86]. The other major approach to understand the droplet dynamics has been through computational models. Two phase solvers have been written to approximate the ablated droplet target deformation [87, 88, 89, 90].

From experiments carried out at the LEC, it is clear that breakup and debris distribution is dependent on the droplet to laser alignment [20, 23, 82]. When operating a droplet-based LPP source, the debris distribution is a function of the alignment range in the system. Alignment between the droplet and the laser focus must occur for three separate degrees of freedom as shown in Fig. 3.1. The droplet lateral positions are described by the $x$ and $y$ axes respectively. At the LEC, lateral misalignments are compensated with the use of a droplet tracking system coupled to a 3-D jet droplet train positioning
Figure 3.1: Droplet position layout describing the three alignment axes. The three axes origins intersect at the laser focus. The $x$ and $y$ axes are the lateral axes. The $x$-axis is coaxial to the laser pulse axis. The $y$-axis is horizontal and perpendicular to the laser axis. The $z$-axis (longitudinal axis) is parallel to the droplet jet direction.

stage [80]. The longitudinal droplet position is described by the $z$-axis in Fig. 3.1. Longitudinal misalignments can occur through variations in the spacing between the droplets, known as drop-to-drop jitter, causing a desynchronization between the droplet and the laser frequency [20]. These longitudinal misalignments are compensated for by synchronizing the laser frequency to the droplet through the so-called laser triggering [20]. In this chapter the debris dynamics caused by longitudinal misalignment along the $z$-axis are investigated. This is accomplished by deactivating the laser triggering system for individual droplets.

The debris velocity and distribution are investigated with an analytical model. Additionally, some of the principles for debris formation and momentum exchange in a droplet-based LPP source are experimentally validated. Trucano and Grady [42] modelled the deformation of a slab target from an ablating laser pulse. A scaling relation was derived on the flat surface by
the ablation pressure of the laser plasma. In this chapter a similar scaling analysis is done with an analytical model for the case of a spherical droplet surface. The model is presented in Sec. 3.3 and is validated by the experimental results.

Measurements were taken of the debris velocity and trajectory over a range of irradiances. The results of the analytical model are compared to experimental results to determine the validity of the models assumptions. The relevance of these results for the shaping of liquid droplet targets and for determining the spatial debris distribution are discussed. The ultimate intent of the work in this chapter is to predict within a useful approximation the mass distribution of neutral cluster debris for given system parameters. The relations derived are built upon in the subsequent chapters towards a debris distribution model.

3.1 Experimental setup

The results presented in this chapter are acquired using the droplet-based LPP sources ALPS I and ALPS II as described in Sec. 2.1 and 2.2. The ALPS I facility is operated with a Nd:YAG laser, running at 10 Hz with a pulse length of 10 ns FWHM and a pulse energy up to 300 mJ per pulse. The laser pulse energy for this system is adjustable between 100 μJ to 300 mJ through the use of adjustable polarizing beam splitters, providing an irradiance range from $8 \times 10^8$ to $2.4 \times 10^{12}$ W/cm$^2$.

For all of the ALPS II measurements in this chapter, the laser is operated at 6 kHz with droplet sizes of 50-60 μm. The vacuum pressure was maintained at 0.1 mbar filled with a background of Ar. The laser beam is focused into the vacuum chamber through a plano convex lens to spot sizes of 50-70 μm. The droplet source is operated at a frequency of 23 kHz. The droplet dispenser is mounted to a 3-D motion stage and coupled to a closed loop control system able to keep the position of the droplet train at the main laser focus position. The control system compensates for lateral instabilities with a spatial resolution of ±5 μm [80]. A piezoelectric actuator determines the droplet frequency [81].
3.1. Experimental setup

Figure 3.2: Experimental layout with shadow graph imaging system for ALPS I and II; (a) Top view, the view angle $\beta = 90^\circ$ in ALPS I and in ALPS II $\beta = 108.4^\circ$ for the recorded tests, (b) side view.

3.1.1 Imaging system

For the experiments in ALPS I and II the debris was imaged using a high speed shadowgraph system [see Figs. 3.2(a)-3.2(b)] as described in Sec. 2.3. In ALPS II the high speed flash duration is $\Delta t_f = 1 \mu s$. The system trigger signals for the main laser, camera, and flash are synchronized such that during the source operation the delays between these signals can be changed with $1 \mu s$ resolution. The flash/camera axis was mounted at $\beta = 108.4^\circ$ relative to the laser axis.

For the experiments acquired in ALPS I the system is also equipped with a 3-D motion stage [see Figs. 3.2(a)-3.2(b)]. For the tests conducted in this facility the shadowgraph system, identical to the one described for ALPS II, is mounted at $\beta = 90^\circ$ to the laser axis. The flash LED in this facility was pulsed with a $2 \mu s$ pulse width.

3.1.2 Image processing

In Fig. 3.3(a) a typical shadowgraph image displaying the interaction of the main laser pulse (coming from the left as indicated by the red arrow) with
the droplet target is shown. The image shows the general debris position for the set flash delay. The camera exposure window is chosen such that it captures the overexposed region where the plasma is located. An imaging processing code was written in MATLAB, which utilized mainly MATLAB’s Image Processing Toolbox. This code processed the shadowgraph images and extracted the main debris dynamics parameters such as the angular distribution and the velocity [see Fig. 3.3(b)]. For each test case hundreds of images are processed using the in-house code. The images are first filtered such that images were removed where the laser missed the droplet. Droplet misses were likely since the triggering system needed to remain off in order to observe the effects of misalignments. This allowed for a collection of data representative of the maximum possible range of misalignments.

The plasma saturation region was utilized in this code for finding the approximate coordinates of the debris point of origin. There is no information pertaining to the intensity distribution of the plasma within the over exposed area, but the boundary of the saturation region does offer useful information. The centroid of the plasma saturation region is measured in the image [see Fig. 3.4(a)]. When comparing the horizontal origin coordinate measured from the plasma location to the droplet jet axis visible in the image, the two values correlate within a standard deviation of ±11 μm. With horizontal debris distances from the origin on the order of 1000 μm, this standard deviation is considered small (±1-2%). The vertical coordinate of the point of origin measured from the plasma spot is assumed to have a standard of deviation of the same order. The vertical origin coordinate is offset to compensate for the time delay between the plasma spot and the shadowgraph flash, which accounts for the initial downwards momentum of the jet present in the debris field.

A numerical Wiener filter is used to remove the high spatial frequency pixilated noise added by the CCD chip [see Fig. 3.4(b)]. The background variation in the intensity of the flash field is removed by subtracting a highly smoothed version of the image created using a binary image morphological operation [91]. After the noise removal, the debris field will remain and can be digitally isolated with level filters. A connectivity function isolates the debris regions based on their size and location [see Fig. 3.4(c)]. The debris position is transformed from the x-y coordinate system to the r-α polar coordinate system with the pivoting axis located at the debris point of origin.
3.2 Experimental results

Two different types of studies are performed in this chapter. The first study is carried out to image the droplet debris over a range of irradiances from 5-130 GW/cm², which is a range of relevance for EUV generation [22]. These results are acquired in the ALPS I facility. The goal of this study is to observe how the neutral cluster breakup is influenced by the laser irradiance,
3.2. Experimental results

Figure 3.4: (a) The boundary of the image region saturated by the plasma has a centroid (blue cross) with a displacement to the droplet initial position that can be approximated. (b) The small scale image noise originating from the CCD is subtracted from the image. (c) The debris region is isolated using level filters and measured in reference to the initial droplet position (blue cross).
3.2. Experimental results

in terms of fragmentation, velocity and trajectory. The recorded images for each irradiance were processed with an image processing code that quantifies the debris radial velocity and trajectory for each ablated target.

The second study is conducted within the ALPS II facility with the goal to image the droplet debris for a large sampling of images (approx. 1000 per case). For these experiments the main laser is operated at 6 kHz repetition rate. The pulse duration for this laser is 24 ns FWHM. The delay between the main pulse and the flash pulse was varied between 5-15 \( \mu s \). The optimal delay for image processing is where the debris field has cleared the saturation spot of the plasma and still retains a density capable of being discriminated by the image processing software. This optimum delay was 10 \( \mu s \) for the images recorded in ALPS II. The goal of these studies was to determine some of the governing relations dominating the neutral cluster debris distribution.

3.2.1 Mean velocity vs laser irradiance

In the ALPS I facility the distribution and velocity of the debris was analyzed for varying irradiance by imaging the debris at irradiances \( E_e \) between 5-130 GW/cm\(^2\) and processing the images with the in-house image processing code. The ALPS I facility was utilized for these tests, because of its greater tunable range of laser pulse energy. For each irradiance case approximately 100 images were taken. Example images at each irradiance value are shown in Figs. 3.5(a) - 3.5(g). The peak irradiance is reached on the droplet surface at \( \alpha_d = 0^\circ \) where the maximum mean debris velocity \( \bar{V} \) is measured. The velocity \( \bar{V} \) used is the mean value of \( V_{avg}(\alpha) \) taken between \( \alpha_{max} \) and \( \alpha_{min} \) [see Fig. 3.3(b)]. This value \( \bar{V} \) readily lends itself to us as a velocity scale term for the Weber number \( We \), which can then be written as

\[
We = \frac{\rho d_0 \bar{V}^2}{\sigma},
\]

where \( \rho \) is the liquid density, \( \sigma \) is the surface tension, and \( d_0 \) is the initial droplet diameter. \( We \) compares the droplet impact kinetic energy to the surface energy. The droplet breakup dynamics from the ablative laser pulse are similar in behavior to a droplet impacting a surface. Using \( \bar{V} \) as the velocity term for this number has shown validity already for characterizing
3.2. Experimental results

Table 3.1: Corresponding properties for each image are listed in the table where $E_e$ is the pulse irradiance, $t$ is the flash delay, $We$ is the Weber number, and $\tilde{t}$ is time normalized to the capillary time scale.

<table>
<thead>
<tr>
<th>Fig 3.5</th>
<th>(a)</th>
<th>(b)</th>
<th>(c)</th>
<th>(d)</th>
<th>(e)</th>
<th>(f)</th>
<th>(g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_e$ (GW/cm²)</td>
<td>5</td>
<td>11</td>
<td>12</td>
<td>25</td>
<td>51</td>
<td>85</td>
<td>124</td>
</tr>
<tr>
<td>$We$</td>
<td>81</td>
<td>265</td>
<td>300</td>
<td>767</td>
<td>1873</td>
<td>3225</td>
<td>5439</td>
</tr>
<tr>
<td>$\tilde{t}$</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.3</td>
</tr>
</tbody>
</table>

these types of dynamics, which includes the use of a non-dimensional time scale based on the capillary time scale $\tau_c = \sqrt{\rho r_0^3/\sigma}$, where $r_0$ is the initial droplet radius [63, 61].

In order to determine the relation between the irradiance and the debris velocity, the mean debris velocity within a range of $-5^\circ < \alpha_d < 5^\circ$ for each case was measured and plotted versus the different laser irradiances. The measured irradiance $E_e$ vs $V$ are plotted in Fig. 3.6. An exponential dependence is observed in the data as $V \propto E_e^n$, with $n \approx 0.65$. Many sources cite a power law dependence on ablation pressure $P_{\text{max}} \propto E_e^n$, with $n \approx 0.67-0.82$, which indicates that the debris velocity may have a dependence on ablation pressure [92, 40, 93, 94, 95]. As mentioned above, in this chapter a scaling analysis of an ablative laser pulse acting on a spherical droplet surface is done, which is validated with the experimental results.

3.2.2 Debris velocity and trajectory measurements

In the ALPS II facility the debris distribution is imaged over the maximum possible range of laser/droplet misalignment by leaving the laser triggering system off. This is done in order to observe the relationship between debris deflection and target misalignment. Examples of the typical images processed are shown for varying $\alpha_d$ in Fig. 3.7. Figure 3.8 shows the data extracted from 92 images of varying laser-droplet alignments. Figure 3.8(a) plots the measured neutral cluster debris velocity $\bar{V}$ versus the average debris deflection angle $\alpha_d$. As expected, there is a decrease in the debris velocity, and therefore the debris kinetic energy, as the absolute deflection angle increases. The deflection angle is expected to be proportional to the droplet-laser misalignment. In the next section the dependence between $\bar{V}$, $\alpha_d$, and droplet-laser
3.2. Experimental results

Figure 3.5: Debris image sequences recorded in the ALPS I test facility. The laser is entering from the left with $\beta = 90^\circ$. The corresponding properties for each image are listed in the Tab. 3.1. *The image shown in (d) is the image with the lowest deflection angle (-10°) available for the set irradiance. The mean radial velocity is expected to be within the standard of deviation of the 0° deflection case (see Fig. 3.8).
3.2. Experimental results

Figure 3.6: Debris $\bar{V}$ measurements vs irradiance $E_e$. The flash exposure time introduces an uncertainty in the velocity measurements between ±7-10%. The irradiance values are measured with an uncertainty of ±15%.

Figure 3.7: Debris image sequences recorded in the ALPS II test facility. The laser is entering from the left with $\beta = 108.4^\circ$; (a) $\alpha_d = 25.5^\circ$ (b) $\alpha_d = 15.0^\circ$ (c) $\alpha_d = -1.0^\circ$ (d) $\alpha_d = -14.0^\circ$ (e) $\alpha_d = -23.5^\circ$. 

45
misalignment $\psi$ will be derived and validated against these experimental results. Figure 3.8(b) shows the measured debris cone angle denoted as $\Delta \alpha = \alpha_{\text{max}} - \alpha_{\text{min}}$ over the corresponding deflection angle. This figure shows that the neutral cluster region is distributed within a conical area between $\sim 70$-$110^\circ$ that is referenced to $\alpha_d$. The angular distribution of these neutral clusters is in good agreement with recent hydrodynamic modeling of a Sn droplet deformed by pulsed laser ablation [87]. An analytical model is developed to validate the second set of obtained data. The model is presented in Sec. 3.3.

### 3.3 Theoretical models

The experimental results described in the previous section are modeled analytically in order to identify the parameters determining the neutral cluster velocity distribution versus the laser irradiance and the angular distribution. This is done by approximating the pressure impulse distribution over the droplet surface. By quantifying this interaction, it is possible to use conservation of energy in order to bridge the laser pulse parameters with the debris dynamics.

#### 3.3.1 Mean velocity vs laser irradiance model

It is first assumed that the laser beam is centered on the droplet target and has an axisymmetric intensity profile (see Fig. 3.9). It is assumed that the ablation pressure profile induced by this pulsed laser on the unablated droplet surface is also radially symmetric. The work $W$ transferred to the unablated droplet surface can be approximated by the expression [41]

$$ W = \frac{\tau_p}{4\rho c_s} \int \overline{P}^2 \, dA, \quad (3.2) $$

where $\tau_p$ is the laser pulse duration (FWHM), $c_s$ is the speed of sound in the fluid, $\rho$ is the fluid density, and $\overline{P}$ is the radial pressure profile, which for this
3.3. Theoretical models

Figure 3.8: (a) Neutral cluster debris mean velocity measurements and analytical prediction plotted versus the mean trajectory $\alpha_d$. (b) $\Delta\alpha = \alpha_{\text{max}} - \alpha_{\text{min}}$ plotted versus the measured deflection angle $\alpha_d$. Measurements were done with the laser triggering system deactivated in order to maximize the recorded misalignment ranges.
work is approximated by

\[ P = P_{\text{max}} \left( 1 - \left( \frac{r}{w} \right)^2 \right) \sqrt{1 - \left( \frac{r}{r_0} \right)^2}, \]  

(3.3)

where \( w \) is the \( 1/e^2 \) beam waist radius [related to the focused beam spot size \( \phi_f \) by \( w = \phi_f / \sqrt{2\ln(2)} \)], \( r_0 \) is the droplet radius, and \( P_{\text{max}} \) is the peak value of the ablation pressure. The \( \sqrt{1 - (r/r_0)^2} \) term accounts for the variation in the laser incidence angle over the droplet surface \( \lim_{r_0 \to \infty} P = P_{\text{max}}[1 - (r/w)^2] \) \[49\]. The surface element \( dA \) that is shown in Fig. 3.9 is given as

\[ dA = 2\pi \left[ 1 - \left( \frac{r}{r_0} \right)^2 \right]^{-1/2} r \, dr. \]  

(3.4)

The work expression \( W \) can be written as

\[ W = \frac{\pi \tau_p P_{\text{max}}^2}{2 \rho c_s} \int_0^{r_{\text{max}}} \left[ 1 - \left( \frac{r}{w} \right)^2 \right]^2 \sqrt{1 - \left( \frac{r}{r_0} \right)^2} r \, dr. \]  

(3.5)

Due to the limits imposed by the droplet surface, two different \( r_{\text{max}} \) limits will need to be integrated depending upon the value of \( r_0/w \). For \( r_0/w \leq 1 \), \( r_{\text{max}} = r_0 \) and for \( r_0/w > 1 \), \( r_{\text{max}} = w \). Solving for these integrals, the work expression can be written as:

\[ W = \frac{\tau_p P_{\text{max}}^2}{\rho c_s} \frac{4\pi r_0^2}{105} f \left( \frac{r_0}{w} \right); \]  

(3.6)

\[ f \left( \frac{r_0}{w} \right) = \begin{cases} \left( \frac{r_0}{w} \right)^4 - \frac{7}{2} \left( \frac{r_0}{w} \right)^2 + \frac{35}{8}, & \frac{r_0}{w} \leq 1 \\ \left\{ 1 - \left[ 1 - \left( \frac{r_0}{w} \right)^{-2} \right]^{7/2} \right\} \left( \frac{r_0}{w} \right)^4 - \frac{7}{2} \left( \frac{r_0}{w} \right)^2 + \frac{35}{8}, & \frac{r_0}{w} > 1 \end{cases}. \]  

(3.7)

Using the conservation of energy, the ablation pressure work \( W \) is equated with the approximate kinetic energy of the debris \( \rho(\pi/12)d_0^3V^2 \):

\[ \frac{\tau_p P_{\text{max}}^2}{\rho c_s} \frac{4\pi r_0^2}{105} f \left( \frac{r_0}{w} \right) \approx \rho \frac{\pi}{12} d_0^3V^2. \]  

(3.8)
3.3. Theoretical models

Equation (3.9) simplifies to a dimensionless form as

$$\frac{c_s r_0 \rho^2 \nabla^2}{\tau_p P_{\text{max}}^2} \approx \frac{2}{35} f \left( \frac{r_0}{w} \right); \quad (3.9)$$

$$\nabla \approx \frac{P_{\text{max}}}{\rho} \sqrt{\frac{\tau_p}{r_0 c_s} \frac{2}{35} f \left( \frac{r_0}{w} \right)}. \quad (3.10)$$

A generalized model predicting the peak ablation pressure for 1.06 μm laser light has already been derived [40]

$$P_{\text{max}} = 8 \left( \frac{I_0}{10^{14}} \right)^{0.7} \left[ 1 + \frac{l_{\text{ac}}^{(0)} \sin(\theta) \tau_p^{0.9}}{w} \left( \frac{I_0}{10^{14}} \right)^{0.3} \right]^{-1.4}, \quad (3.11)$$

where $P_{\text{max}}$ is in units of Mbar, $w$ is in μm, $\tau_p$ is in ns, $I_0$ is the laser nominal peak intensity in W/cm$^2$ and is related to $E_e$ as $I_0 = 2 \ln(2) E_e$, $l_{\text{ac}}^{(0)}$ is the nominal distance between the ablated surface and the critical density surface in μm, and $\theta$ is the half-angle beam divergence. This simple 2-D corrected model has been validated for irradiance ranges from $10^{13} - 10^{15}$ W/cm$^2$ and the uncorrected 1-D model version of this relation has shown consistency with experiments in the $10^{10} - 10^{12}$ W/cm$^2$ range [41].

3.3.2 Debris mean trajectory vs axial misalignment analysis

When a nanosecond laser is focused on a droplet target, the neutral cluster debris generally expands and distributes along the direction of the main beam, opposite to the plasma forward expansion [54, 89, 90]. From observations made at LEC and others, the analytical model was derived with the assumption that the average direction of the back expanding debris depends strongly on the laser alignment to the target [82].

It is assumed that the ablation pressure acting on the droplet surface occurs on a much faster timescale than the droplet deformation. Therefore the initial pressure force acting on the unablated material acts approximately normal to the droplet surface. This results in an ejection of the neutral debris in a direction dependent on the incident irradiance profile and the droplet sur-
3.3. Theoretical models

Figure 3.9: Sketch of the approximate ablation pressure profile over the droplet surface used in the analytical model.

\[ \bar{P} = P_{\text{max}} \left[ 1 - \left( \frac{r}{w} \right)^2 \right] \sqrt{1 - \left( \frac{r}{r_0} \right)^2} \int \left[ 1 - \left( \frac{r}{r_0} \right)^2 \right]^{-\frac{1}{2}} dr \]
face normal \cite{38,39}. Misalignment of the droplet with the Gaussian intensity profile of the laser pulse leads to a change of the incident irradiance profile on the droplet and to a consequent deflection in the debris trajectory from the normal direction.

A simplified model of a laser pulse is considered with a radially Gaussian intensity profile interacting with a spherical droplet \cite[see Fig. 3.10(a)]{3.1}. For this model all of the length scales are nondimensionalized with respect to \( r_0 \) and signified with the notation "\( \tilde{~} \)". The intensity profile and the droplet surface are analyzed on the \( \tilde{x} - \tilde{r} \) plane, which intersects the center axis of the laser pulse and the center of the spherical droplet. The laser profile is considered axisymmetric around the \( \tilde{x} \)-axis and the droplet profile is considered axisymmetric around the line \( \tilde{r} = \tilde{\psi} \). The objective of this model is to locate the point on the surface of the droplet of highest irradiance. Therefore, one can exclude the points on the droplet surface off the \( \tilde{x} - \tilde{r} \) plane, which will have greater corresponding angles of incidence than those on the \( \tilde{x} - \tilde{r} \) plane. In this work the asymmetric irradiance profile on the droplet surface is neglected and it is assumed that the location of the maximum irradiance on the droplet surface estimates the ejected mean debris trajectory. The impact of neglecting asymmetry will be investigated in the next sections by comparison with the experimental results.

The laser spot size is described by its \( 1/e^2 \) beam waist radius \( \tilde{w} \). The laser intensity profile is described by the radial Gaussian function

\[
f_L(\tilde{r}) = e^{-2\tilde{r}^2/\tilde{w}^2},
\]

where \( f_L(\tilde{r}) \) is the normalized laser intensity as a function of \( \tilde{r} \) and \( \tilde{w} \). The droplet surface profile in the \( \tilde{x} - \tilde{r} \) plane is simply an offset semicircle function described by the equation

\[
\tilde{x} = \sqrt{1 - (\tilde{\psi} - \tilde{r})^2},
\]

where \( \tilde{\psi} \) is the nondimensional droplet misalignment. As the laser projects onto the surface of the droplet, the angle of incidence changes as a function of the spherical surface profile. This leads to a local irradiance maximum on the droplet surface that is a function of \( d_0 \), \( f_L \) and \( \psi \). The droplet surface normal angle \( \alpha \) \cite[see Fig. 3.10(a)]{3.10} is the angular coordinate of the droplet.
3.3. Theoretical models

Figure 3.10: (a) Dimensionless profiles of laser pulse intensity and droplet surface axially misaligned by distance $\tilde{\psi}$. (b) Debris deflection calculations at varying misalignments.

surface and can be written as a function of $\tilde{r}$ as

$$\alpha = \tan^{-1}(\frac{d\tilde{x}}{d\tilde{r}}) = \sin^{-1}(\tilde{\psi} - \tilde{r});\ -\frac{\pi}{2} < \alpha_d < \frac{\pi}{2} \quad (3.14)$$

In order to determine the location of the maximum irradiance on the droplet surface we calculate first the normalized intensity onto the projected surface of the droplet, which we now write as a function of $\alpha$

$$\frac{I(\alpha)}{I_0} = \cos(\alpha)f_L(\alpha) = \cos(\alpha)\exp\left\{-2\left[\tilde{\psi} - \sin(\alpha)\right]^2/\tilde{w}^2\right\} \quad (3.15)$$

where $I$ is the laser intensity on the droplet surface as a function of $\alpha$ and $I_0$ is the nominal peak laser intensity. The location of the maximum surface irradiance on the droplet ($\alpha_d$) can be found by taking the derivative of Eq. (3.15) and setting it equal to 0, which reduces to a cubic equation as

$$\sin(\alpha_d)^3 - \tilde{\psi}\sin(\alpha_d)^2 - (\tilde{w}^2 + 1)\sin(\alpha_d) + \tilde{\psi} = 0;\ -\frac{\pi}{2} < \alpha_d < \frac{\pi}{2}. \quad (3.16)$$

Solving numerically for $\alpha_d$ the theoretical estimate of the deflection angle of the droplet with respect to the laser center axis is determined [see Fig. 3.10(b)]. In Equation (3.16) the debris deflection angle $\alpha_d$ is dependent only
on the droplet misalignment $\tilde{\psi}$, the laser beam radius $\tilde{w}$ and the droplet radius $r_0$. In Figs. 3.11(a)-3.11(b) the relationship between these three parameters, $\alpha_d$ and $I/I_0(\alpha_d)$ and $\tilde{\psi}$ is plotted. The results intuitively show that as $\tilde{w}$ increases, the debris will have a smaller deflection $\alpha_d$ for the same misalignment $\tilde{\psi}$ than for smaller $\tilde{w}$. Therefore, for an LPP source, a larger laser spot will in general reduce the spread in the debris distribution.

In order to compare the validity of relation (3.16) to the experimental results, the decrease in the ablation pressure interaction with the droplet surface due to the alignment offset must be approximated. A scaling term is derived to account for the radially-cubed dependence of the ablation pressure acting on the droplet surface. The distance between the maximum absorption region of the plasma and the spot of peak intensity at the droplet surface for $\alpha_d = 0^\circ$ is defined as $\tilde{L}_s$. As the droplet is offset, the distance between the maximum absorption region and the peak intensity position ($\tilde{r}, \tilde{x}, \alpha_d$) is assumed to increase. This behavior is approximated with the following relation

$$P_{offset} \approx \tilde{L}_s^3 \left( \left\{ \tilde{L}_s + [1 - \cos(\alpha_d)]^2 \right\}^2 + \left\{ \tilde{\psi} - [1 - \sin(\alpha_d)]^2 \right\} \right)^{-3/2} \quad (3.17)$$

In order to account for the droplet-laser misalignment, the $P_{offset}$ factor and the peak surface intensity $I(\alpha_d)$ will be inserted into (3.11) as

$$P_{max} \approx P_{offset} \cdot \frac{I(\alpha_d)}{10^{14}}^{0.7} \left\{ 1 + \frac{l_{ac} \sin(\theta)}{w} \cdot 10^{0.9} \left[ \frac{I(\alpha_d)}{10^{14}} \right]^{0.3} \right\}^{-1.4} \quad (3.18)$$

According to Eq. (3.15) the shape of the projected laser spot onto the droplet will be distorted and attenuated by the increase in $\tilde{\psi}$ when $\tilde{w} \gtrsim 1$, due to the change in the incidence angle profile relative to the laser spot. This leads to a reduction in the droplet surface area that is above the ablation threshold fluence, which leads to a proportionate decrease in the spot size seen by the droplet. This effect is illustrated in Fig. 3.12 where Eq. (3.15) versus $\alpha$ is plotted for different values of $\tilde{\psi}$. The reduction in the projected $\tilde{w}$ will have a narrowing of the pressure profile on the droplet surface. The work of Gelderblom et al. [62] predicts that the narrower pressure profile will lead to an increase in the kinetic energy partition $E_{K,d}/E_K$ according to Eq. (1.6) described in Sec. 1.2. The relationship between $E_{K,d}/E_K$ and the velocity
Figure 3.11: (a) The calculated mean droplet deflection $\alpha_d$ as a function of the droplet-laser misalignment $\tilde{\psi}$ for various values of $\tilde{\omega}$. (b) The maximum normalized intensity $I/I_0(\alpha_d)$ calculated for given $\tilde{\psi}$ for various values of $\phi_f$. 
3.3. Theoretical models

Figure 3.12: The laser intensity profile $I/I_0(\alpha)$ on droplet surface will narrow as the laser/droplet misalignment $\tilde{\psi}$ is increased.

The laser intensity profile is described by \cite{62}

$$\frac{E_{K,d}}{E_K} \approx \frac{u_R^2}{u_R^2 + 2u_{cm}^2} = \left[1 + 2\left(\frac{u_{cm}}{u_R}\right)^2\right]^{-1}, \quad (3.19)$$

where $u_R$ is the initial rim velocity of the expanding droplet sheet perpendicular to the sheet center axis and $u_{cm}$ is the velocity of the droplet center of mass. From this relation the spread angle of the debris can approximated as

$$\Delta \alpha \approx 2\tan^{-1}\left(\frac{u_R}{u_{cm}}\right), \quad (3.20)$$

For the results in Fig. 3.8(b) Eq. (3.15) is used to calculate the change in $\sigma_G$ (denoted as $\sigma$ in \cite{62} and further explained in Sec. 5.1.2) as a function of $\alpha_d$ and is shown in Fig. 3.13.
3.4 Discussion

In order to validate the analytical models, the experimental test cases were calculated for the given test conditions. First the measurements versus the analytical model calculations for the mean debris velocity dependence on irradiance are compared. The same comparison is done for the debris velocity-trajectory dependence.

Using the Eqs. (3.10) and (3.11), the predicted average debris velocity for each test case is approximated, the results of which are shown in Fig. 3.14. One can observe that the predictions are in good agreement with the experimental results, with the analytical model predicting $n \approx 0.65$ for $V \propto E_c^n$. This validates our hypothesis that the dominant momentum exchange mechanism accelerating on the droplet neutral cluster debris is the ablation pressure. It is interesting to note that this power law dependence with $n \approx 0.65$ is present when an ablating plasma is the momentum exchange mechanism, but when the momentum exchange is dependent on recoil from evaporation, then $n \approx 1$ [63].
Using the Eqs. (3.10), (3.16) and (3.18), the velocity $\bar{V}$ versus the mean trajectory $\alpha_d$ is predicted for the given test conditions and plotted versus the measured results in Fig 3.15. For these cases the nominal maximum absorption distance $L_s$ was fitted from the data as $L_s \sim 109 \, \mu m$. This value is consistent with previously measured distances of $115 \, \mu m$ at $200 \, \text{GW/cm}^2$ and $135 \, \mu m$ at $600 \, \text{GW/cm}^2$, [96, 97] as well as from calculations performed with the LEC in-house analytical plasma model [23, 98]. The analytical model is in good agreement with the experimental results. From these results one can make the conclusion that although the mean neutral cluster velocity is only a first order quantity of the debris dynamics, it validates the hypothesis that the neutral cluster debris is ejected normal to the droplet surface. There is also strong evidence to support that the ablation pressure interaction with the droplet is the primary momentum exchange mechanism propelling the neutral cluster debris. This is also the primary mechanism deforming the droplet. It is assumed at this point that as the irradiance drops, there is a range where the recoil from evaporation dominates the kinetic energy exchange and the dynamic approaches $n \approx 1$ [63].
Figure 3.15: Neutral cluster debris mean velocity measurements and the analytical prediction of Eq. (3.10) plotted versus the mean trajectory $\alpha_d$. Measurements were done with the laser triggering system deactivated in order to maximize the recorded misalignment ranges.
3.4. Discussion

Figure 3.16: $\Delta \alpha$ is predicted as a function of $\alpha_d$ for the experimental cases according to Eq. (3.20).

The expected magnitude of $\Delta \alpha$ is calculated and using Eq. (3.20) and compared with the experimental data in Fig. 3.16 and found to be in good agreement. The surface intensity attenuation due to the offset incidence angles over the droplet is not expected to have a measurable impact on the irradiance seen by the plasma for deflections on the order of $\psi < 1$ for typical EUV source operating conditions. When $\sim 2\%$ of the laser pulse energy reaches the target it ablates a layer of material from the surface, creating an ionic plume that quickly propagates to the center of the beam absorbing the major portion of the laser pulse energy. This can be verified when one considers the following scaling relation [23]

$$\tau_p > \frac{\sqrt{2}\psi}{c_i}$$

(3.21)

where $c_i$ is the average ion speed, which for these test conditions is chosen to be approximately $20$ km/s [45, 99]. The right hand term is the estimated amount of time for the initial plasma to propagate to the center of the laser pulse. For an offset of half the droplet size (i.e. $\sim 30$ $\mu$m for our experimental
3.4. Discussion

conditions) Eq. (3.21) gives 24 ns > 2.1 ns, which means that the plasma plume can be expected to reach the laser spot in time to interact with ~98% of the laser pulse energy. Therefore, a misalignment of this order is not expected to have a substantial effect on the plasma laser coupling and source location, but will have a substantial effect on the neutral cluster trajectory and velocity [39].

From what has been observed in the studies at the LEC, similar to many other fluid dynamic studies, [61, 100, 101, 102] there are consistent droplet breakup dynamics that can be grouped into different Weber number regimes. The higher Weber number cases seen in our experiments are similar to droplet breakup induced by shockwaves [100, 101, 102]. The primary difference seen between droplet breakup from a planar shockwave versus an ablative laser pulse is the radial momentum component, caused by the radial expansion of the plasma plume.

The scope of this work is concerned mainly with the droplet breakup dynamics within the first 10-20 μs after the main laser pulse. It is interesting here to investigate briefly the neutral cluster fragment evolution from the irradiation site to the chamber wall. Mainly two dominant factors could alter the debris trajectory before it reaches the wall, which are drag force from the background gas and gravity. The influences of localized debris mitigation systems such as barrier plates or gas curtains are excluded. Under the given test conditions described in Sec. 3.2.2, which are within the typical test conditions in ALPS II, the Weber number for α_d = 0° is \( W_e \approx 10^000 \). Within less than 10 μs, the droplet flattens into a thin film and then breaks up into small droplet fragments. These fragments will then travel away from the point of origin with their own individual momentum (initial velocity of \( V_0 \sim 100 \) m/s) and trajectory. The fragments in the size range of 0.1 μm < d < 50 μm are considered. The distance the debris travels from the plasma to the chamber wall is given as \( L \approx 70 \) cm. First looking at the influence of drag force a standard force balance on a spherical droplet fragment with a diameter \( d \) is done writing

\[
\frac{\pi}{8} \rho_\infty V^2 C_D d^2 = -\frac{\pi}{6} \rho d^3 \frac{dV}{dt},
\]

where \( V \) is the fragment velocity, \( \rho_\infty \) is the density of the background gas, \( C_D \) is the fragment drag coefficient, and \( \rho \) is the fragment fluid density. For the chosen fragment size range, the Knudsen number is \( 40 < K_n < 19^000 \),
which is on the order of the molecular flow regime. Using relations derived by Sengers et al. [103] for the drag coefficient of a sphere in the nearly free molecular flow regime the drag coefficient is approximated as $C_D \approx 15$. Time of flight for the fragment in a frictionless environment can be defined as $t_{f,0} \approx L/V_0$. Solving the force balance gives

$$\frac{t_f}{t_{f,0}} = \left[ \exp \left( \frac{3 \rho_\infty C_D L}{4 \rho d} \right) - 1 \right] \frac{4 \rho d}{3 \rho_\infty C_D L},$$

(3.23)

where $t_f$ is the final fragment velocity in a drag environment. Equation (3.23) is plotted as a function of the dimensionless fragment diameter $\bar{d} = d/r_0$ in Fig. 3.17 for two different ambient pressures. $C_D$ is held constant for these approximations. Figure 3.17 illustrates how smaller fragments are decelerated much more rapidly compared to larger fragments. It is also clear that the ambient pressure will have a significant influence on how much the neutral clusters are decelerated before they reach the chamber wall. Due to the high particle velocities gravity is expected to play a minor role in the particle deflection.

3.5 Summary

Experiments were done imaging the debris produced during the generation of a droplet-based LPP. The laser/droplet interaction was imaged with a high-speed shadowgraph system with a synchronized delay. Image processing code was written to measure the mean droplet debris velocity $V(\alpha_d = 0^\circ)$ and trajectory. Several cases were recorded imaging the droplet deformation for varying laser pulse irradiance $E_e$ and laser spot size. Further experiments were performed where the droplet misalignment was deliberately introduced into the system. Images were collected of the misaligned droplet debris and the images were processed to correlate the debris velocity to its average trajectory.

A scaled analytical model was derived modeling the plasma ablation pressure on the droplet surface as the primary momentum exchange mechanism between the neutral cluster debris and the laser pulse. A non-dimensional relation was derived that describes the debris-plasma momentum exchange
Figure 3.17: Equation (3.23) is plotted as a function of the dimensionless fragment diameter $\tilde{d} = d/r_0$ for two ambient pressures. The drag coefficient is held constant at $C_D \approx 15$. 

$\frac{t_f}{t_{f0}}$ vs $\tilde{d}$
as a function of the ratio of the droplet size to the beam waist radius. This dimensionless number is the ratio of debris kinetic energy to shockwave energy of the propagating plasma. Relations were derived to describe analytically the droplet deflection as a function of droplet size, laser spot size, and laser-droplet misalignment.
Chapter 4

Droplet deformation dynamics from multiple picosecond laser pulses

A body of research has been produced in recent years investigating the dynamics of the droplet deformation from a single laser pulse. Work that has been done already in an effort to map out the dynamics of this phenomena have included droplets hitting an impactor and expanding in free space [61]. Experiments have been done with inked water droplets shot with a laser pulse [63] and with liquid Sn [60, 70, 104]. This work has focused strongly on quantifying the momentum coupling mechanism between a laser pulse and a fluid droplet. There has been computational and analytical investigations of the deformation of the droplet due to laser-pulse impact [62]. The newest work is extending into the dynamics for picosecond and femtosecond laser pulses [65, 44, 67].

To the authors knowledge there has been no investigations of the droplet deformation due to bursts of laser pulses, beyond a recent investigation with a picosecond pulse pair [68]. In this chapter liquid Sn droplets are irradiated by a broad array of picosecond laser bursts. The deformation dynamics are observed using a high speed shadowgraph imaging system, which images the deforming droplet shape at different times. This work provides an opportunity for investigating the governing principles of droplet deformation by
observing what relationships maintain themselves over a wide range of variability in terms of the burst parameters. This includes the kinetic energy transfer, the shape of the expanding fluid sheet, and the effects of cavitation. Droplet irradiation by picosecond bursts are important to study, because they have the potential to provide higher order mechanisms to influence the deformation process. Thereby allowing for targets shapes not achievable with single pulse deformation.

4.1 Experimental Setup

The experiments in this chapter were conducted within a vacuum chamber at 0.02 mbar with Ar background gas (see Fig. 4.1). A droplet dispenser generates a mono-dispersed and coherent stream of droplets of liquid Sn. The droplet frequency is determined by a piezoelectric actuator and was operated between 20-33 kHz for the various cases. The droplet sizes typically vary between 30-100 μm. The droplet dispenser is mounted to a 3-D motion stage and coupled to a closed loop control system able to keep the position of the droplet train at the main laser focus position. The control system compensates for lateral instabilities with a spatial resolution of ±0.5 μm [80]. The laser is a MOPA Nd:YAG system with a wavelength of 1064 nm. The master oscillator is a pulsed laser diode that generates pulses with a duration of $\tau_p = 43$ ps (FWHM) that are amplified through several stages. It is capable of single pulse to 100 MHz repetition rates with single pulse energies up to $E_p \leq 2$ mJ. Using an acousto-optic modulator (AOM) the oscillator output is modulated in order to shape bursts with energies up to $E_B \leq 2.7$ mJ. The laser spot was focused to a spot diameter of $\phi_f = 22$ μm (FWHM).

The deforming droplet was imaged using a high speed shadowgraph system. The shadowgraph system consists of a high speed camera (SONY ICX625ALA/AQA) capable of up to 20 fps. The image resolution was 0.93 μm/pixel. The flash is a high powered light emitting diode (LED) pulsed at 500 ns duration. The system trigger signals for the laser, camera, and flash are synchronized such that during the source operation the delays between these signals can be changed with 0.25 μs resolution. The flash/camera axis was mounted at 90° relative to the laser axis.
4.1. Experimental Setup

Figure 4.1: The functional layout of the experiment. The deforming droplets were imaged at 90° relative the laser axis. The 500 ns LED flash was delayed at controlled time intervals relative to the laser pulse.
4.1. Experimental Setup

4.1.1 Experimental parameters

In total 40 unique burst interactions were imaged. For each case an average of five time steps were imaged and for each time step \( \sim 100 \) images were acquired. Each image is a unique droplet impact. For each time step only images that had a deflection angle within \( \pm 5^{\circ} \) relative to the laser center axis were taken as representative of each data point, since the droplet deflection corresponds to a misalignment range of the laser center axis with the droplet center of \( \pm 4\% \) of the initial droplet diameter [60, 105]. The dimensions \( Z \) and \( D_r \) (see Sec. 4.2.1) were measured for these images and were found to vary with a maximum standard of deviation of \( 8\% \) relative to the mean. Therefore, the images for each time step are considered highly repeatable with regards to the investigated parameters for this work; mainly sheet expansion rate, sheet shape and concavity, first hole appearance time, and jetting. The fluid dynamic features that varied broadly image-to-image were the position of the receding rim ligaments and the location on the sheet of the hole appearances, i.e. features dependent on the phase of initial fluid perturbations. The variation in these features were not expected to influence the investigated parameters in this work.

For the investigated cases, the total burst energy \( E_B \) and the pulse energy \( E_p \) are nondimensionalized relative to the initial surface energy as

\[
\tilde{E}_B = \frac{E_B}{\sigma d_0^2}; \quad \tilde{E}_p = \frac{E_p}{\sigma d_0^2}; \quad (4.1)
\]

where \( \sigma = 0.55 \text{ N/m} \) is surface tension, \( d_0 \) is the original droplet diameter, and \( E_B \) was varied from \( E_B = 220 - 1'860 \ \mu\text{J} \), or \( \tilde{E}_B = 6.0 \times 10^4 - 5.6 \times 10^5 \). Throughout this work all of the length scales are non-dimensionalized (indicated by the “~” notation) with respect to the initial droplet radius \( r_0 \) and the time scales with the droplet capillary time \( \tau_c = \sqrt{\rho r_0^3/\sigma} \), where \( \rho \) is the fluid density.

Examples of the burst shapes used for this work are shown in Fig. 4.2(a)-4.2(f). Certain burst shapes are named according to their characteristics, such as double pulse for two pulse bursts, flat top (FT) for bursts with pulses that were within \( \pm 20\% \) of the mean \( E_p \), rising saw tooth (RST) for bursts with steadily increasing pulse energies, falling saw tooth (FST) for bursts...
with steadily decreasing pulse energies, Gaussian for bursts with a Gaussian burst envelope, and double burst for bursts with two distinct envelopes within a burst. For the double pulses the ratio between the pulse energies was varied from 50/50 to 10/90. The time interval between pulses within the burst could be chosen arbitrarily. For bursts with pulses more than 2, the delay between pulses was maintained at either 50 ns or 12.5 ns (with the exception of the time duration between the burst envelopes in the double bursts).

### 4.2 Droplet sheet expansion

The breakup dynamics of liquid droplets impacted by a laser can vary depending upon the laser impulse parameters. For the experimental cases investigated here the droplet deformation is dominated by either surface acceleration or cavitation from shock waves traveling through the droplet. The dynamics involving cavitation observed in these experiments are investigated in Sec. 4.3. For the cases where surface acceleration dominates the dynamics, after the laser impacts the droplet it flattens into a sheet of varying concavity. Concavity in this work is defined as the ratio of the sheet depth $Z$ to sheet diameter $D_r$ described in Sec. 4.2.1. Surface acceleration refers to the droplet surface being accelerated to an initial velocity by the recoil pressure of the expanding laser plasma, at the location of the peak ablation pressure. As the sheet flattens the rim of the expanding sheet will decelerate from the initial velocity until it starts to recede, ejecting fragments, or until holes form in the sheet that will grow into a web of ligaments that then fragment into neutral clusters [61, 62, 70]. It can be assumed that $u_0$ is equal to the velocity of the droplet surface after acceleration due to mass conservation and conservation of energy [106, 61]. The sheet expansion is determined by measuring the expansion of the rim contour (see Fig. 4.3) over a series of time steps. It is found that the rim expansion $R(t)$ follows the dynamics for droplet impact described by Villermaux and Bossa [61]

$$
\tilde{R}(\tilde{t}) = 1 + \sqrt{\frac{1}{2}We_s \tilde{t}} \left(1 - \frac{\sqrt{3}}{2} \tilde{t}\right)^2,
$$

(4.2)
4.2. Droplet sheet expansion

Figure 4.2: Examples of burst profiles implemented in this work for (a) double pulse, (b) flat top (FT), (c) rising saw tooth (RST), (d) falling saw tooth (FST), (e) Gaussian, (f) double burst. Each peak represents one laser pulse of the shown dimensionless pulse energy $\tilde{E}_p$ at the time from the beginning of the burst $\tilde{t}$.
where $W_{e_s} = \rho d_0 u_0^2 / \sigma$ is a modified Weber number, where the velocity scale is $u_0 = \dot{R}(t \approx 0)$. The dynamics described by Eq. (4.2) operate within the deformation partition $E_{K,d}$ of the kinetic energy partition. This is differentiated from the portion of kinetic that corresponds to the movement of the droplet center of mass $E_{K,cm}$, which has no influence on the droplet deformation [62]. For each case $W_{e_s}$ and $u_0$ are derived from the measurement of $R(t)$ using (4.2) (see Fig. 4.4). In Fig. 4.4 the first hole time $\tilde{t}_b$ is marked for the plotted cases. For each case $\tilde{t}_b$ was measured by finding the earliest time within the acquired images where a hole was visible within the area half the distance to the rim from the sheet center. For all cases where $W_{e_s} < 5'000$ there was no observable perforation. This limit corresponds to a maximum $\tilde{t}_b$ of $\sim 0.4$, which is near the maximum value of $R(t)$ reached at $\tilde{t} \sim 0.385$ [61]. The values of $\tilde{t}_b$ for all of the measured cases are scattered within a region represented by the highlighted area in Fig. 4.4.

A few factors must be considered when comparing a droplet impacted by a shock wave versus a droplet impacting a surface. The period of fluid acceleration for a droplet impacting a surface is $\sim d_0 / u_0$. For a droplet accelerated by a laser burst there is a period of intermittent acceleration that occurs within the burst duration $\tau_B$ (defined as the time between the first and last pulse in the burst). There is no correlation in this case between $\tau_B$ and $d_0 / u_0$. The velocity component of $W_{e_s}$ corresponds to the surface velocity $u_0$ directly after the acceleration phase and the shock waves have propagated through the
Figure 4.4: Example cases of varying $We_s$. The markers represent the measured $R(t)$ and the lines represent the corresponding fit of (4.2) to measure $We_s$. The perforation times $\tilde{t}_b$ for the plotted cases are also shown. The highlighted area marks the range where $\tilde{t}_b$ is observed across all measured cases.
4.2. Droplet sheet expansion

droplet. \( u_0 \) must be distinguished from the mass averaged velocity \( \bar{u} \), which corresponds to the total deformation kinetic energy \( E_{K,d} = (\pi/12)\rho d_0^3 \bar{u}^2 \). \( u \) is the fluid velocity relative the fluid center of mass and therefore \( \bar{u} \) is approximated by modeling the expanding sheet as an expanding shell of constant thickness of the dimensions \( Z \), \( D_r \), and \( u_0 \). The full formulation is included in Sec. \( \Delta \) and the polynomial fit is given as

\[
\frac{u_0}{\bar{u}} \approx -\left( \frac{Z}{D_r} \right)^3 + 2.1 \left( \frac{Z}{D_r} \right)^2 - 0.45 \frac{Z}{D_r} + 1.5. \tag{4.3}
\]

One can see that the proportion of \( u_0/\bar{u} \) for a thin expanding disc is 3/2. For the above approximation the sheet thickness \( h \) is calculated with the conservation of mass and the rim radius from Eq. \( \Delta \) and gives \[61\]

\[
\tilde{h}(\tilde{t}) \sim \frac{4}{3} \tilde{R}^{-2}(\tilde{t}). \tag{4.4}
\]

For a droplet impacting a surface with a velocity \( u_0 \) the inertial timescale would typically be \( \tau_i \sim r_0/u_0 \). For droplets deformed by an impulse where the impact timescale is different than the droplet crush time (i.e. \( \tau_B \neq r_0/u_0 \)), the inertial timescale will be chosen as the time when the approximate sheet thickness \( h \) is equivalent to the initial droplet radius \( r_0 \), i.e. \( h/r_0 \sim 1 \). Substituting \( \Delta \) into \( \Delta \) and using this criteria defines the inertial timescale as

\[
\tilde{\tau}_i \sim 0.22 \text{We}_{s}^{-1/2}, \tag{4.5}
\]

where the factor 0.22 is an approximation of \( \sqrt{2}(2/\sqrt{3} - 1) \). Here it can be determined that since \( \text{We}_s \gg 1 \) in all cases, then \( \tau_i \ll \tau_c \) and the dynamics are primarily effected by inertial and surface tension forces and viscous forces are negligible. In the cases investigated here the shock wave propagates on the sonic timescale \( \tau_s = r_0/c_s \), where \( c_s \) is the fluid speed of sound. \( \tau_s \ll \tau_i \) in all cases here. Therefore, it can be assumed that the dynamics during the droplet deformation are not influenced by pressure or density variations in the fluid.

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4.2. Droplet sheet expansion

Figure 4.5: An example sequence illustrating the deep cup shape formed by a burst ($N_p = 53, \bar{E}_B = 7.5 \times 10^4$, Gaussian burst, 80 MHz). The timing from left to right is $t = 0.057, 0.095, 0.17, 0.28, 0.40, 0.59, \text{ and } 0.78$. The third frame displays the definition of the deformation dimensions $D_r(t)$ and $Z(t)$, where $D_r$ is diameter of the sheet rim perpendicular to the laser center axis and $Z$ is the distance between the rim and the sheet back along the laser center axis. This case can be contrasted with the one shown in Fig. 4.3, which has $N_p = 30$.

4.2.1 Sheet concavity

It is observed experimentally that the concavity of the expanding droplet sheet $K$, defined as $K = (Z - d_0)/D_r$, varies with the burst parameters (see Fig. 4.5). The definition of $K$ is chosen as the ratio of the initial expansion speeds of $Z$ and $D_r$ before deceleration is non-negligible, where $Z(t = 0) \sim d_0$ and $D_r(t = 0) \sim 0$. If $\ddot{Z}$ and $\ddot{D}_r$ are plotted in time for some example cases [see Fig. 4.6(a)-4.6(b)], it is seen that both parameters have an initial velocity and as the sheet expands, the film becomes thinner and deceleration from surface tension plays a dominant role. For the cases observed, $\ddot{Z}$ reaches its peak faster relative to $\ddot{D}_r$ as the radius of curvature of the cup back surface $R_c$ (defined in Sec. A) decreases relative to the rim radius of curvature defined as $D_r/2$.

In order to determine the scaling parameters of $K$, the momentum transfer during the burst is approximated first. The final velocity of the portion of the droplet surface accelerated by the laser plasma is assumed to be $\sim u_0$, which is derived for each case by the fit of (4.2). The thickness of the droplet center at the end of the burst is approximated as $h(t \sim \tau_B/2)$, where $h$ is calculated from (4.4). If $h(\tau_B/2) > r_0$, then $h$ is approximated as the droplet diameter minus the distance traveled by the interface during the
4.2. Droplet sheet expansion

Figure 4.6: Sheet cup dimensions $\tilde{Z}$ and $\tilde{D}_r$ plotted in time for two burst cases shown in (a) Fig. 4.3 with $N_p = 30$ and $\tilde{E}_B = 8.0 \times 10^4$ and (b) Fig. 4.5 with $N_p = 53$ and $\tilde{E}_B = 7.5 \times 10^4$. It can be noted that although both of them have Gaussian envelope shapes and similar burst energies, (b) has a maximum $\tilde{Z}$ of nearly double that shown in (a).
burst as \( h \approx d_0 - \tau_B u_0/2 \). The average momentum of the fluid, which is moving at the velocity \( \dot{Z} \), along the droplet/laser center axis is assumed to be dependent on the total duration of the acceleration \( \sim N_p \tau_p \) and the final surface velocity \( u_0 \) as

\[
\dot{Z} = a_0 \frac{u_0}{u_0} \frac{c_s N_p \tau_p}{c_s N_p \tau_p + h(t_s)},
\]

(4.6)

where \( c_s N_p \tau_p \) represents the fluid depth reached by the transferred momentum during the acceleration phase, and \( a_0 \) is a scaling prefactor. Equation (4.6) allows a proportionality check of \( Z \) relative to \( D_r \). During the initial expansion the local fluid velocities dominate \( Z \) and \( D_r \) relative to the deceleration from surface tension. Therefore the concavity is approximated by \( K \sim \dot{Z}/D_r \) where \( \dot{D}_r \sim D_r/t \) for \( t \ll 0.385 \). This gives an expression for the proportionality of \( Z \) and \( D_r \) as

\[
K \sim a_0 u_0 \frac{c_s N_p \tau_p}{c_s N_p \tau_p + h(t_s)} \frac{t}{D_r} + K_1,
\]

(4.7)

where an offset term \( K_1 \) needs to be added to account for the shape contribution of pressure wave diameter relative to the droplet size [62]. This factor is measured over the range \( \tilde{E}_p = 7.6 \times 10^4 - 3.1 \times 10^5 \) from single pulse cases, which gave \( K_1 = 0.29 \pm 0.07 \). Figure 4.7 plots (4.7) vs the experimentally measured \( K \) as a function of \( N_p \). \( K \) reaches a limit in the experiments where the back of the sheet stretches until it perforates and the burst essentially "drills" through the droplet converting it into an expanding tube concentric with the laser axis (see Fig. 4.8). The dynamic described by (4.7) implies that there is a degree of manipulation that is possible in the cup development depending upon the burst envelope shape. This manipulation is more clearly seen in Fig. 4.9(a)-4.9(b) by observing a two cases where the burst was divided into two distinct burst envelopes separated by a delay [see Fig. 4.2(f)]. As described in this figure, the burst shape influence can be seen in the shape of the expanding sheet where regions are created with different stretch rates. For double burst profiles this leads to an "umbrella" like shape, since the more stretched part of the film will be thinner leading to different rates of contraction in the later half of the deformation.
4.2. Droplet sheet expansion

Figure 4.7: The droplet sheet concavity $K$ dependency with the number of pulses $N_p$. The inset compares directly the measured ratio to that calculated from (4.7), using a prefactor of $a_0 = 0.67$. The dashed red linear regression line has a coefficient of determination $R^2 = 0.91$. The error bars represent the max/min range of the data points.
4.2. *Droplet sheet expansion*

Figure 4.8: An example sequence illustrating the droplet being "drilled" through and continuing as an expanding tube ($N_p = 82$, $\tilde{E}_B = 1.8 \times 10^5$, Gaussian burst, 80 MHz). The timing from left to right and top to bottom is $\tilde{t} = 0.04, 0.06, 0.10, 0.14, 0.18$, and 0.30. Similar breakup dynamics were observed for three other burst cases which had $N_p = 82 - 245$ and $\tilde{E}_B = 1.8 \times 10^5 - 4.6 \times 10^5$. 
Figure 4.9: Example breakup sequences of double burst profiles where distinct stretch regions in the expanding sheet are seen. For (a) the timing from left to right is $\tilde{t} = 0.09, 0.17, 0.28, 0.34, 0.45, \text{ and } 0.62$ ($N_p = 52, \tilde{E}_B = 6.0 \times 10^4$, double burst, 80 MHz). For (b) the timing from left to right is $\tilde{t} = 0.09, 0.15, 0.26, 0.36, 0.43, \text{ and } 0.50$ ($N_p = 44, \tilde{E}_B = 9.2 \times 10^4$, double burst, 80 MHz). The small jet ejected behind the sheet forms at roughly the time of the maximum sheet extension $\tilde{t} \sim 0.385$. It is the result of the contraction of the sheet center, reversing the geometric damping of the sheet instability amplitude growth rate \cite{61, 67}. This is a separate phenomena from the fast microjets produced by cavitation and discussed in Sec. 4.3.
4.2. Droplet sheet expansion

4.2.2 Kinetic energy transfer

It is necessary to determine the governing parameters of the kinetic energy transfer to the droplet from the ablation pressure impulses. According to the definition of the Weber number as the ratio of kinetic to surface energy, the splash Weber number is related to the droplet deformation kinetic energy as

\[ W_{es} = \left( \frac{u_0}{\bar{u}} \right)^2 12 \frac{E_{K,d}}{\sigma \pi d_0^2}. \]  

(4.8)

According to Gelderblom et al. [62] the droplet impact kinetic energy \( E_K \) can be partitioned into two separate components, which are a deformation component \( E_{K,d} \) and a center of mass displacement component \( E_{K,cm} \) (note that \( E_{K,d} + E_{K,cm} = E_K \)), the ratio of which is referred to as the kinetic energy partition \( E_{K,d}/E_{K,cm} \) and is a function of \( \phi_f/d_0 \). In the experimental cases done in this work \( \phi_f/d_0 \) is such that \( E_{K,d}/E_K \sim 0.91 - 0.97 \) [62], therefore for this work \( W_{es} \) is treated as representative of the total kinetic energy of the experimental cases in order to investigate scaling parameters. It should be noted that it is not possible to get an accurate measurement of \( E_{K,cm} \) for most of the cases due to that lack of transparency of the deforming fluid sheet. In order to model the kinetic energy transfer the approach is taken to estimate the sum of the work done on the droplet by each individual pulse in the burst as

\[ E_{K,d} \approx \sum_{i=1}^{N_p} \Delta KE_i, \]  

(4.9)

where \( \Delta KE_i \) is the kinetic energy transferred to the droplet surface by the corresponding pulse in the laser burst. This kinetic energy is approximated by

\[ \Delta KE_i = \frac{1}{2} m (\Delta u_i + u_{s,i})^2 - \frac{1}{2} m u_{s,i}^2 \]  

(4.10)

where the mass \( m \) accelerated by the subsequent pressure pulse during the irradiation is approximated by

\[ m = \rho \frac{\pi}{4} \phi^2 c_s \tau_p \]  

(4.11)
4.2. Droplet sheet expansion

and the increase in the velocity $\Delta u_i$ of $m$ after the pressure impulse is \[41\]

\[\Delta u_i = \frac{P_{s,i}}{\rho c_s}. \quad (4.12)\]

The peak ablation pressure $P_{s,i}$ for an individual laser pulse acting on the droplet surface is given as \[43\]

\[P_{s,i} = 0.4 \left( \frac{I_{0,i}}{\lambda} \right)^{2/3} \left( \frac{A_a}{2 Z_a} \right)^{1/3}, \quad (4.13)\]

where the pulse irradiance is $I_{0,i} = E_{p,i}/(\tau_p \pi \phi_e^2)$ in TW/cm$^2$, $\lambda$ is the laser wavelength in $\mu$m, $A_a$ is the atomic mass, $Z_a$ is the atomic number, and $\phi_e$ is the $1/e^2$ laser spot diameter. The surface velocity at the beginning of the pulse irradiation $u_{s,i}$ is approximated by

\[u_{s,i} \approx \sum_{j=1}^{i-1} \frac{1}{\tau_e} + \frac{1}{\rho c_s} P_{s,j} \quad (4.14)\]

where $t_i$ and $t_j$ are the arrival time of the corresponding laser pulse within the burst. $\tau_e = d_0/c_s$ is the sonic timescale of the droplet and represents the time scale of the diffusion of momentum from the droplet surface into the interior. Equation (4.8) then derives to

\[W_e = \left( \frac{u_0}{\overline{u}} \right)^2 \frac{3 \phi_j^2}{2 \frac{d_0^2}{\sigma \rho c_s}} \frac{\tau_p}{2} \times \sum_{i=1}^{N_p} \left[ \left( \frac{P_{s,i}}{\frac{1}{\tau_e} + \frac{1}{\rho c_s}} \right)^2 - \left( \sum_{j=1}^{i-1} \frac{1}{\tau_e} + \frac{1}{\rho c_s} P_{s,j} \right)^2 \right] \quad (4.15)\]

where $u_0/\overline{u}$ is given from (4.3). The experimental value of $W_e$ measured by fitting the measured $R(t)$ to (4.2) is compared against (4.15) in Fig. 4.10.

Possible sources of error are suspected to be the deviation in the shape of the expanding sheet from the shape approximation taken in Sec. A. The effects of cavitation within the droplet have also not been considered for this model. This model provides insight into the scaling of the kinetic energy transfer.
4.3 Cavitation and jetting

that comes with dividing the laser energy into multiple pulses. A way to conceptualize this is to take the example where there is a burst of $N_p$ pulses of equal energy $E_{p,i} = E_B/N_p$. We also simplify by only considering bursts where the total duration of the burst is $\ll \tau_o$. The scaling is calculated as the ratio of $We_s$ for $N_p$ pulses versus $We_s$ for a single pulse of the equivalent burst energy

$$\frac{We_s}{We_{s,N_p=1}} \propto \frac{\sum_{i=1}^{N_p} \left[ \left( \frac{E_B}{N_p} \right)^n \sum_{j=1}^{i} \left( \frac{E_B}{N_p} \right)^n \right]}{E_B^{2n}} = N_p^{-2n} \frac{N_p(N_p + 1)}{2}. \tag{4.16}$$

This relation shows that one can expect an increase $\sim 0.5 N_p^{2/3}$ in the kinetic energy transfer as a result of the fact that the ablation pressure has an exponential dependence on pulse energy as $n = 2/3$. This scaling is not infinite since as the pulse irradiance decreases below the ionization threshold, the ablation pressure is no longer dependent on the plasma absorption of laser energy and is instead dependent of recoil pressure from vaporization. Therefore, the surface velocity would become proportionate to the pulse energy, i.e. $n \sim 1$ and the proportionality of (4.16) is no longer valid [63].

The major proportionality factors influencing $We_s$ can be compared across all measured cases by rearranging (4.16) and denoting the gain from $N_p$ as

$$f(N_p) = \frac{1}{2} N_p^{1-2n}(N_p + 1). \tag{4.17}$$

The measured cases are plotted in comparison to (4.17) and the other varying parameters from (4.15) in Fig. 4.11 in order to show the proportionality of these factors with $We_s$.

4.3 Cavitation and jetting

Many of the cases investigated have features that are instigated by cavitation bubbles forming in the droplet interior. One of these features is the presence of high speed microjets that are ejected behind the droplet relative to the
Figure 4.10: $We_s$ is predicted by equation (4.15) and compared to the measured $We_s$ using (4.2). The coefficient of determination of the logarithms of (4.15) and (4.2) is $R^2 = 0.91$. 

\[
\begin{align*}
\text{Equation (4.2)} & \quad \text{(dashed line)} \\
\text{Equation (4.15)} & \quad \text{(solid line)}
\end{align*}
\]
4.3. Cavitation and jetting

Figure 4.11: $We_s$ is compared against the derived scaling factors shown in the horizontal axis for $n = 2/3$, where the variation in the individual pulse energies is ignored in favor of $f(N_p)$. For cases with $N_p > 82$ the droplet was drilled through and it was not possible to measure a comparable $We_s$. The coefficient of determination for the logarithms of the red dashed regression line is $R^2 = 0.90$
4.3. Cavitation and jetting

Figure 4.12: Examples of high speed jets ejected out of the back of the droplet along the laser center axis. Image (a) is a FT burst with $N_p = 8$, $\tilde{E}_B = 1.2 \times 10^5$, 20 MHz and $\tilde{t} = 0.13$. Image (b) is double pulse burst with $N_p = 2$, $\tilde{E}_B = 1.2 \times 10^5$, 250 ns between pulses and $\tilde{t} = 0.23$. Image (a) is the case with the lowest peak pulse energy of $\max(E_{p,i}) \sim 60 \mu J$ where this jetting is still observed. Therefore, 60 $\mu J$ is identified as the lower threshold pulse energy required for cavity formation within the droplet with the given experimental parameters.

laser impact [see Fig. 4.12(a)-4.12(b)]. The process of this microjetting has already been studied extensively [107, 108, 109, 110] and the sequence of the jetting dynamic is explained in Fig. 4.13. These microjets do not appear in breakup sequences where the pulse energies are below $E_{p,i} \sim 60 \mu J$ [the case shown in Fig. 4.12(a) has the lowest maximum pulse energy that still exhibits microjetting]. It is well known that cavitation will be caused by the rarefaction of the focused shock wave propagating through the droplet [65, 44, 72]. When the tensile stress of the rarefaction wave exceeds the spall strength of the liquid Sn, defined as $\sigma_{Sn}$, and the velocity field is divergent the material separates and cavitation occurs [65].

In the single pulse cases this microjetting was not be observed, because
4.3. Cavitation and jetting

Figure 4.13: A sketch describing the microjetting process within the droplet for a two pulse process. (a) The shock wave from the first pulse is focused towards the droplet center due to the curvature of the droplet surface. (b) An expanding cavitation bubble is formed near the droplet center at the point where the velocity field diverges. (c) The shock wave from the second pulse is also focused. (d) A portion of the shock wave interacts with the cavitation bubble hydrodynamically focusing a jet perpendicular to the shock wave plane, with the remaining shock wave traveling around the cavitation bubble to transmit momentum to the rear of the droplet as shown in (e). (f) The high speed hydrodynamically focused jet penetrates through the back of the droplet through the crown center.
there is no secondary shock wave to generate the microjet.

4.3.1 Cavitation criteria

For all burst cases with $\max(E_{p,i}) > 60 \mu J$ one or more small high velocity jets are ejected behind the droplet as shown in Fig. 4.12. This phenomena is caused by laser induced shock waves interacting with cavities within the droplet [109]. A precise investigation of the threshold conditions for this jetting would require finite element models like those done by Basko et al. [65], Grigoryev et al. [72], which is beyond the scope of this work. A scaled approach for approximating the cavitation threshold is taken here instead in an effort to understand the order of magnitude of the dominant factors influencing cavitation within the droplet and how they counterbalance each other. Namely the gain from the acoustic focusing of the shock wave, the acoustic attenuation of the propagating shock wave, and the limit between the rarefaction component of the shock wave and the tensile strength of the liquid.

The cavitation threshold is defined here as the threshold conditions required in order to form a cavitation bubble within the droplet. This criteria is checked by approximating the rarefaction wave stress intensity seen at the center of the droplet. The shock wave peak intensity evolution is modeled with three superimposed gain and loss terms

$$
\sigma_{cav} = P_s f_{rare} G_{focus} L_{atten} < \sigma_{Sn}, 
$$

(4.18)

where $\sigma_{cav}$ is the tensile stress seen by the fluid near the droplet center by the shock rarefaction wave, $P_s$ is the peak pressure interacting with the droplet surface calculated by Eq. (4.13), $f_{rare}$ is the ratio of the rarefaction wave peak to the shock wave peak pressure near the droplet center, $G_{focus}$ is the acoustic gain of the shock wave into the droplet caused by geometric focusing, $L_{atten}$ is the loss factor from acoustic attenuation, and $\sigma_{Sn}$ is the local spall strength of the fluid. In order to calculate precisely the ratio of the peak negative pressure and the peak positive pressure of the shock wave, it would require high-resolution FE hydrodynamic simulations that solve the equation of state for tin during the shock wave evolution. Earlier experimental and
numerical studies show a range between $-5\%$ to $-20\%$ and therefore it is approximated here as $f_{\text{rare}} \sim -10\%$ \cite{111, 112, 113, 114, 72}. For a 60 $\mu$J pulse $P_s \sim 10$ GPa. The acoustic focusing gain $G_{\text{focus}}$ is calculated using the method described by Baac et al. \cite{115}, Krivokorytov et al. \cite{67}

\begin{equation}
G_{\text{focus}} = k_f r_0 \alpha_f,
\end{equation}

where $k_f$ is the acoustic wavenumber $k_f = 2\pi/(c_s \tau_p)$, and $\alpha_f$ is a geometric factor $\alpha_f = 1 - \sqrt{1 - 1/4 f_N^2}$ depending upon the acoustic f-number $f_N \approx r_0/\phi_f$. For the given experiment $f_N \sim 1.8$, which gives $G_{\text{focus}} \approx 92$. The attenuation loss factor is approximated from Stokes law of sound attenuation \cite{116}

\begin{equation}
L_{\text{atten}} = e^{-\alpha_m r_0},
\end{equation}

where $\alpha_m$ is the frequency dependent acoustic attenuation coefficient, which is calculated as $\alpha_m = 2\mu \omega^2/(3\rho c_s^3)$. $\mu$ is the dynamic viscosity and $\omega$ is the frequency of the shock wave defined as $\omega = 2\pi/\tau_p$. For the given experiment $L_{\text{atten}} \approx 0.0037$. From Eq. (4.18) $\sigma_{\text{cav}} \sim -350$ MPa. It is known that the spall strength of a fluid is dependent on the local strain rate. This dependence can be approximated for surface tension dominated spallation as $\sigma_{S_n} = c_s \rho^{2/3}(6\dot{\varepsilon})^{1/3}$, where $\dot{\varepsilon}$ is the local strain rate \cite{117}. This dependence was more recently investigated by Grigoryev et al. \cite{72} specifically for tin shocked by a laser pulse. From the results of Eq. (4.18) the strain rate at the cavitation threshold conditions is within a range of $\dot{\varepsilon} \sim 3 \times 10^6 - 2 \times 10^7$ s$^{-1}$.

4.3.2 Jetting

Jetting as shown in Fig. 4.12(a)-4.12(b) is caused by one or more subsequent planar shock waves interacting with a cavity within a fluid and has been studied already extensively by Obreschkow et al. \cite{107}, Antkowiak et al. \cite{108}, Thoroddsen et al. \cite{109}, Ohl et al. \cite{110} (see figure 4.13). This dynamic can be investigated in more detail by looking at two cases where only two
pulses were used with a time gap of 225—250 ns, giving the cavity more time to grow [see Fig. 4.14(a)-4.14(b)]. In each case a "crown" is visible encircling the ejected jet. In the experimental work by Thoroddsen et al. [109, p. 12] the diameter $d_c$ of this crown was found to correspond to $\sim 72\%$ of the cavity diameter that the shock wave interacted with. The same scaling will be used for this work. From the cavity radius $r_c = d_c/1.44$ and the timing between the laser pulses $\Delta t$, the cavity growth rate $u_c$ can be estimated as $u_c \sim r_c/\Delta t$. With the shocked material velocity relation \[ \sigma_{cav} = -\frac{1}{2}\rho u_c c_s, \] the tensile stress at the point of rupture can be calculated from the crown diameter. Comparing $\sigma_{cav}$ calculated for the cases in Fig. 4.14(a) and 4.14(b) from Eq. (4.21) with that calculated using Eq. (4.18) we see a correlation within 16\% between the two methods in Tab. 4.1.

### 4.4 Sheet rupture time

The observation of the experiments shows that after the droplet is impacted by the laser burst the sheet expands into a thin liquid sheet. Depending upon the burst profile and energy the sheet will either expand to a maximum rim radius and then contract due to rim destabilization or the sheet will rupture during the rim evolution. When the sheet ruptures a number of holes will perforate the sheet. The holes then expand converting the sheet into a web of ligaments that break into fragments (Fig. 4.15). The time from the last pulse in the burst until the first hole appearance is defined as the breakup time.

Table 4.1: Comparison between the calculations for the tensile stress at the droplet center using the acoustic attenuation model (4.18) vs calculating the tensile strength from the cavity growth rate measured indirectly from the hydrodynamically focused crown and jet (4.21).  

<table>
<thead>
<tr>
<th></th>
<th>$r_c$((\mu\text{m}))</th>
<th>$u_c$(m/s)</th>
<th>Eq. (4.18)(MPa)</th>
<th>Eq. (4.21)(MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fig. 4.14(a)</td>
<td>38</td>
<td>150</td>
<td>-1'500</td>
<td>-1'300</td>
</tr>
<tr>
<td>Fig. 4.14(b)</td>
<td>79</td>
<td>350</td>
<td>-2'800</td>
<td>-3'000</td>
</tr>
</tbody>
</table>
Figure 4.14: Two cases with double pulses where it is possible to measure the crown and microjet phenomena. Image (a) has $\tilde{E}_B = 2.5 \times 10^5$ at $\tilde{t} = 0.076$, with the first pulse energy $\tilde{E}_{p,1} = 1.2 \times 10^5$ and (b) has $\tilde{E}_B = 5.7 \times 10^5$ at $\tilde{t} = 0.079$, with the first pulse energy $\tilde{E}_{p,1} = 2.9 \times 10^5$. The crown diameter was measured at the points where the crown breaks from the back droplet surface.
4.4. Sheet rupture time

time $t_b$. For each case $t_b$ is measured by identifying the earliest time when a hole appears on the sheet as shown in Figs. 4.16(a)-4.16(b). As seen in these figures the holes can appear grouped together in clusters while other parts of the film remain unbroken. The measurement error in $t_b$ includes shadowgraph exposure time and the estimate of the hole growth rate using Taylor-Culick as $t_{b,\text{err}} = d_{h,\text{err}}\sqrt{\rho h/(8\sigma)}$, where $d_{h,\text{err}}$ is the uncertainty in the diameter of the observed first hole(s). In order to determine the rupture criteria for the expanding sheet a similar approach to that done by Bremond and Villermaux [69] for a bursting fluid thin film is derived. This approach was further adapted for laser impacted droplets by Klein [70], which will be emulated in this work, except for some key differences in assumptions and formulation. The key differences lay in the formulation of the acceleration term $a$, the handling of the threshold time $t_t$ for the thin sheet criteria, and the initial perturbation amplitude and wavenumber range for a laser ablation accelerated target.

The sheet expansion is divided into three separate time phases. The first phase is for the time from $t = -\tau_B$ to the end of the burst $t = 0$, which is the duration of intermittent fluid acceleration. The second phase is the period before the droplet has flattened into the thin sheet $t = 0 \rightarrow t_t(k)$, where $t_t$ is a function of the specific wavenumber $k$. The third phase is defined as the remaining time to the sheet rupture $t = t_t(k) \rightarrow t_b$, where $t_b$ is denoted as the time for the first hole appearance in the droplet.

The droplet surface is accelerated by a laser ablation plasma, which instigates the growth of Rayleigh-Taylor hydrodynamic instabilities. Since material is being removed from the droplet surface through ablation, it is assumed that the initial amplitude $\eta_0$ of the perturbations on the droplet surface after the laser pulse will be on the order of the ablation depth. The ablation depth is expected to be in the range of $\eta_0 \sim 60 - 80$ nm [72]. Another phenomena has been observed for laser ablation accelerated targets where there is a lower cutoff wavelength of $\lambda \sim 10 \mu m$ for surfaces accelerated with 1064 nm light due to the damping effects caused by the ablation [118, 119]. This means there is a maximum wavenumber $\bar{k}_{\text{max}} = 2\pi/\lambda$ excited during the acceleration.

The initial modulations of the surface waves are represented as Fourier modes [69]

\[
\tilde{\eta} = \tilde{\eta}_0 f(\tilde{t}) e^{ik\tilde{s}}, \tag{4.22}
\]
4.4. Sheet rupture time

Figure 4.15: An example sequence illustrating the rim expansion and sudden rupture of the expanding sheet into a web of ligaments \(N_p = 8, \ E_B = 2.4 \times 10^5\), RST, 20 MHz. The timing from left to right is \(\tilde{t} = 0.071, 0.13, 0.20, 0.27, 0.41, \) and 0.59. The third frame displays \(R(t)\), which is defined as the path length along the sheet from the sheet center to the rim.

Figure 4.16: Example cases showing first hole perforation, where (a) \(N_p = 27\) and \(E_B = 2.3 \times 10^5\), and (b) \(N_p = 8\) and \(E_B = 3.3 \times 10^5\).
4.4. Sheet rupture time

where \( \eta_0 \) is the initial surface wave amplitude, \( \tilde{s} \) is the length coordinate tangent to the droplet surface, and \( f(\tilde{t}) \) is the evolution of the amplitude scale for the specific wavenumber \( \tilde{k} \) as a function of time. Throughout this analysis \( \tilde{k} \) refers to the wavenumber of the initial wave mode on the droplet surface before surface stretching occurs. Effects of stretching on \( \tilde{k} \) will be compensated for in the model separately as explained further below.

The acceleration of the droplet surface during the burst interaction is approximated by

\[
a \approx \frac{u_0}{(N_p \tau_p)} = \frac{W e_s^{1/2} r_0}{(\tau_c N_p \tau_p)},
\]

where \( N_p \tau_p \) is the duration of the acceleration period for the burst. The velocity scale \( u_0 \) was chosen for the Weber number since the surface will be accelerated to \( \dot{R}(t = 0) \sim u_0 \) [106 61].

The acceleration phase of the droplet is assumed to be impulsive with \( \tilde{\omega}_{1,im} \ll (N_p \tau_p)^{-1} \), where \( \tilde{\omega}_{1,im} \) is the imaginary part of the wave mode growth rate \( \tilde{\omega}_1 \). The capillary wavenumber for the accelerated surface is defined as

\[
\tilde{k}_c = (N_p \tau_p)^{-1/2} W e_s^{1/4},
\]

which will determine the growth rates of the wave modes in the first phase. For the cases investigated, during the impulsive acceleration \( \tilde{k}_c \gg \tilde{k}_{max} \gg 1 \) and the dispersion relation for the surface waves is given as [69]

\[
\tilde{\omega}_1^2 = (\tilde{k}^3 - \tilde{k}_c^2 \tilde{k}). \tag{4.23}
\]

The initial conditions of the shape function \( f(\tilde{t}) \) in equation (4.22) are given such that [69]

\[
f_1(\tilde{t} = -\tilde{\tau}_B) = 1; \quad \dot{f}_1(\tilde{t} = -\tilde{\tau}_B) = 0. \tag{4.24}
\]

All unstable modes for \( 1 \leq \tilde{k} \leq \tilde{k}_{max} \) grow with the same initial amplitude and zero initial growth rate. The shape function of \( f_1(\tilde{t}) \) is governed by [69]

\[
\ddot{f}_1 = -\tilde{\omega}_1^2 f_1(\tilde{t}). \tag{4.25}
\]

Using (4.23) and (4.24) the differential equation (4.25) is solved for as

\[
f_1(\tilde{t}) = \cos[\tilde{k}_c^{3/2}(\tilde{t} + \tilde{\tau}_B)]
+ \frac{W e_s^{1/2}}{\tilde{k}^2 N_p \tau_p} \{1 - \cos[\tilde{k}_c^{3/2}(\tilde{t} + \tilde{\tau}_B)]\}; \tag{4.26}
\]
\[ f_1(\tilde{t}) = -\tilde{k}^{3/2}\sin[\tilde{k}^{3/2}(\tilde{t} + \tilde{\tau}_B)] \]
\[ + \frac{We_s^{1/2}}{\tilde{k}^{1/2}N_p \tilde{\tau}_p} \sin[\tilde{k}^{3/2}(\tilde{t} + \tilde{\tau}_B)]. \]

(4.27)

It is assumed that the pauses between pulses within the burst are short enough that there is not a substantial increase in the surface wave amplitudes, and therefore the duration of phase 1, originally defined as \( \tilde{\tau}_B \), is treated as \( \sim N_p \tilde{\tau}_p \). Assuming impulsive acceleration, the duration of phase 1 is approximated \( N_p \tilde{\tau}_p \to 0 \) and the small angle approximation is used for the amplitude and growth rate at the end of the acceleration phase as \( f_1(\tilde{t} = 0) \approx 1 \) and \( \dot{f}_1(\tilde{t} = 0) \approx We_s^{1/2} \tilde{k} \).

At the end of phase 1 each surface wave mode will evolve characteristic of an infinite medium where \( \tilde{k}_c = 0 \), until the flattening fluid film reaches the thin layer criteria \( \tilde{k}\tilde{h} \ll 1 \) \([69]\). Since \( \tilde{h} \) is a function of time, each wave mode corresponding to \( \tilde{k} \) will reach the flat sheet criteria at a different time denoted as \( \tilde{t}_l \). The effect of the sheet surface expansion must be accounted for by self-similar stretching, since \( \tilde{R} > 1 \) for the wave modes already during phase 2. The stretching of the wavenumber is accounted for by dividing \( \tilde{k} \) by \( \tilde{R} \), giving a proportionate decrease in the wavenumber for an increase in the sheet length \([120]\). Therefore, \( \tilde{t}_l \) is the time that satisfies \( \tilde{k}\tilde{R}^{-1}\tilde{h} = 1 \) \( (\tilde{R}^{-1} \) accounts for self-similar stretching), which by combining with (4.2) and (4.4) is

\[ \tilde{t}_l \approx \left[ \left( \frac{4}{3} \tilde{k} \right)^{1/3} - 1 \right] \sqrt{2} We_s^{-1/2}. \]

(4.28)

Accounting for the infinite medium and self-similar stretching the dispersion relation for the second phase is defined as

\[ \tilde{\omega}_2^2 = \left( \frac{\tilde{k}}{\tilde{R}} \right)^3. \]

(4.29)

The initial conditions for phase 2 are the final conditions at the end of phase 1 where \( f_2(\tilde{t} = 0) = f_1(\tilde{t} = 0) \) and \( \dot{f}_2(\tilde{t} = 0) = \dot{f}_1(\tilde{t} = 0) \) \([69]\). Solving for \( \ddot{f}_2 = -\tilde{\omega}_2^2 f_2(\tilde{t}) \) produces

\[
\dot{f}_2(\tilde{t}) = \cos \left[ \left( \frac{\tilde{k}}{\tilde{R}} \right)^{3/2} \tilde{t} \right] \\
+ We_s^{1/2} \tilde{k} \left( \frac{\tilde{k}}{\tilde{R}} \right)^{-3/2} \sin \left[ \left( \frac{\tilde{k}}{\tilde{R}} \right)^{3/2} \tilde{t} \right];
\]

(4.30)
4.4. Sheet rupture time

\[ \dot{f}_2(\tilde{t}) = -\left(\frac{\tilde{k}}{R}\right)^{3/2} \sin \left[ \left(\frac{\tilde{k}}{R}\right)^{3/2} \tilde{t} \right] \]

\[ + We_{s}^{1/2} \tilde{k} \cos \left[ \left(\frac{\tilde{k}}{R}\right)^{3/2} \tilde{t} \right]. \]

(4.31)

Using (4.30), (4.31), and (4.28) \( f_2 \) and \( \dot{f}_2 \) at the end of the second phase are written as functions of \( \tilde{k} \) as

\[ f_2(\tilde{t}) = \cos \left( \tilde{k} \sqrt{\frac{3}{4}} \tilde{t} \right) \]

\[ + \sqrt{\frac{4}{3}} We_s \sin \left( \tilde{k} \sqrt{\frac{3}{4}} \tilde{t} \right); \]

\[ \dot{f}_2(\tilde{t}) = -\tilde{k} \sqrt{\frac{3}{4}} \sin \left( \tilde{k} \sqrt{\frac{3}{4}} \tilde{t} \right) \]

\[ + We_{s}^{1/2} \tilde{k} \cos \left( \tilde{k} \sqrt{\frac{3}{4}} \tilde{t} \right). \]

(4.33)

The third phase is characterized by the evolution of the wave mode on a thinning sheet until the amplitude of the mode is of the order of the sheet thickness. The mode that fulfills this condition for the shortest time is the breakup wave mode characterized by the breakup wavenumber \( \tilde{k}_b \). This value is found computationally by substituting (4.28) into (4.33) and solving for

\[ \dot{f}_2(\tilde{k}_b) \approx \max \left( We_{s}^{1/2} \tilde{k} \right) \]

\[ \times \cos \left\{ \sqrt{\frac{3}{2}} We_{s}^{-1/2} \tilde{k} \left[ \left(\frac{4}{3}\right)^{1/3} - 1 \right] \right\}. \]

(4.34)

The thin sheet dispersion relation in the third phase, which includes self-similar stretching, is given as [69]

\[ \tilde{\omega}_3^2 = \frac{\tilde{h}}{2} \left( \frac{\tilde{k}}{R} \right)^4. \]

(4.35)
The amplitude function in phase 3 can now be derived with the same general solution as phase 2 for $\ddot{f}_3 = -\tilde{\omega}_3^2 f_3$ as \[ f_3(\tilde{t}) = f_2(\tilde{t}) \cos \left[ \sqrt{\frac{\tilde{h}}{2}} \left( \frac{\tilde{k}}{\tilde{R}} \right) \right. \left. \left( \tilde{t} - \tilde{t}_t \right) \right] + \dot{f}_2(\tilde{t}) \sqrt{\frac{2}{\tilde{h}}} \left( \frac{\tilde{k}}{\tilde{R}} \right)^{-2} \sin \left[ \sqrt{\frac{\tilde{h}}{2}} \left( \frac{\tilde{k}}{\tilde{R}} \right) \right. \left. \left( \tilde{t} - \tilde{t}_t \right) \right]. \] (4.36)

Substituting (4.2), (4.4), (4.28), (4.32) and (4.33) and using the small angle approximation this expression conveniently simplifies to

\[ f_3(\tilde{t}) \approx W e_s^{1/2} \tilde{k} \tilde{t}. \] (4.37)

The complete mathematical explanation for the step between Eq. (4.36) and (4.37) is given in Sec. 3. The time to the appearance of the first perforation $\tilde{t}_b$ is the time at which the amplitude of the fastest growing wavenumber $\tilde{k}_b$ is on the order of the sheet thickness.

There will be a contraction of the perturbation amplitude due to mass conservation resulting in $\tilde{\eta}_0/\tilde{k}^2 \sim \tilde{\eta}_{0,3}/(\tilde{k}/\tilde{R})^2$, where $\tilde{\eta}_{0,3}$ is the initial perturbation amplitude attenuated by self-similar stretching \[120\]. Using (4.2) and (4.4) an expression is found for the stretched perturbation amplitude in relation to the sheet thickness \[70\]

\[ \frac{\tilde{\eta}_{0,3}}{\tilde{h}} = \frac{3}{4} \tilde{\eta}_0. \] (4.38)

Substituting (4.38) into (4.22) gives the relation to solve for $\tilde{t}_b$

\[ \frac{\tilde{\eta}_{0,3}}{\tilde{h}} f_3(\tilde{k}_b, \tilde{t}_b) = \frac{3}{4} \tilde{\eta}_0 f_3(\tilde{k}_b, \tilde{t}_b) = 1. \] (4.39)

Solving for $\tilde{t}_b$ reveals

\[ \tilde{t}_b \approx \frac{4}{3} \tilde{\eta}_0^{-1} \tilde{k}_b^{-1} W e_s^{-1/2}. \] (4.40)

In Fig. 4.17 the experimental $\tilde{t}_b$ was compared against (4.40) as a function of $W e_s$ where $\tilde{t}_b$ was measured for all test cases where hole perforation occurs and the droplet has not been drilled through. The initial wave amplitude was chosen to fit the data as $\eta_0 = 70$ nm, which is within the expected range.
4.4. Sheet rupture time

Figure 4.17: The breakup time $\tilde{t}_b$ plotted as a function of $W e_s$ and compared with Eq. (4.40). Rupture of the sheet does not appear for $\tilde{t}_b > 0.385$, which corresponds to the time of the maximum rim radius. The highlighted area marks the variation in Eq. (4.40) for $\eta_0 = 60 - 80$ nm. The coefficient of determination for the logarithms of experimental values compared to Eq. (4.40) is $R^2 = 0.90$. 
4.5 Summary

Experiments were performed imaging sequences of shadowgraphs of droplets irradiated by bursts of picosecond laser pulses. The bursts parameters were varied widely among 40 cases in terms of the number of pulses $N_p = 1 - 245$, the burst energy $\tilde{E}_B = 6.0 \times 10^4 - 5.6 \times 10^5$, the pulse energy distribution within the burst, and the timing between pulses. The sheet rim radius $R(t)$ is measured for each case and compared to a model of a splashing droplet deforming in free space, where a modified Weber number is defined as $We_s = \rho d_0 u_0^2 \sigma^{-1}$. A relation is derived in order to estimate the proportionality between the mass mean velocity relative to the fluid center of mass $\bar{u}$ and velocity term for $We_s$, which is $u_0 = \dot{R}(t \approx 0)$ for a given concavity $K$. The inertial timescale $\tau_i$ is defined as a function of $We_s$. An analytical expression is derived of the conservation of energy between the laser burst and the droplet and is compared to the experimental results. The rupture time of the expanding droplet sheet is measured as a function of $We_s$. A Rayleigh-Taylor instability analysis is done of the laser accelerated droplet surface in order to predict the time of the first hole appearance. High speed jets are observed ejecting behind the droplet relative to the laser irradiation zone. An analytical model is derived in order to predict the conditions for the microjet ejection.
Chapter 5

Droplet fragmentation dynamics from a laser induced shockwave

One of the major approaches to understand the debris generation process and spatial expansion over time has been to image the produced debris with different time delays from the main laser pulse with the use of a shadowgraph. From the time resolved images one can determine the speed, distribution, and scale of the droplet fragments. This has been done in previous work and in order to determine the momentum coupling mechanism between the laser plasma and the unablated droplet fluid (see Chap. 3).

In this work the droplet breakup process from a pulsed laser is simulated using volume of fluid (VOF) CFD. The strategy is to produce high-resolution representations of the breakup processes that occur across a range of parameters. The simulations can then be used in order to determine certain scaling relations for the neutral cluster size and spatial distribution. In this work the laser to droplet momentum coupling relations determined in previous work are used as an initial condition for several VOF CFD simulations of the impacted droplet. The separation of timescales is employed for this work where $\tau_p \ll \tau_i \ll \tau_c$, where $\tau_p$ is the laser pulse duration and the subsequent timescale for the plasma expansion, the inertial timescale of the droplet deformation is $\tau_i \sim 0.22 W e_s^{-1/2}$, and the droplet capillary timescale
is $\tau_c = \sqrt{\rho \cdot r_0^3 / \sigma}$. The velocity field for the ablated droplet is calculated as an initial condition where the droplet is assumed to remain spherical during the recoil pressure wave.

The goal of the simulations is to use the results in order to determine the primary parameters and dynamics which govern the fragment size and spatial distribution of the splashes from the impacted droplet. Determining what factors could reduce the size of the fragments would result in progress towards solving some of the challenges described. For the figures in this chapter, the orientation of the laser irradiation is indicated by $\rightarrow$.

### 5.1 Model description and cases

Simulations of a droplet impacted by a laser plasma induced shockwave were performed for several cases of varying laser pulse intensities. The laser profile was modelled as a Gaussian profile irradiating the droplet surface (see Fig. 5.1). The modelled process is that of a laser pulse ablating a thin layer of material ($\delta/d_0 \sim 0.006$) off of the surface of the droplet [22], converting the material into a high temperature plasma, which then applies a recoil pressure to the remaining droplet fluid [60]. The velocity field imposed within the droplet by the ablation pressure shockwave was calculated analytically utilizing the method employed by Gelderblom et al. [62] and used as the initial condition for the VOF simulations. The simulations were run until fragmentation of the droplet had ceased.

The simulations were performed in Fluent 18.0. The Volume of Fluid (VOF) model enables the simulation of fluid flows with two or more phases and interactions between these fluids and their surfaces by solving a single set of momentum equations and tracking the phases through the domain [121]. The formulation of the VOF model included the coupled level set method for tracking the surface interface with the sharp interfacing method. The droplet surface interface was constructed using the piecewise linear interpolation scheme called Geo-reconstruct. The laminar viscous model is used. The finite volume approach was used with conservation equations listed in Tab. 5.1.

The parameters for the eight cases are listed in Tab. 5.2. The splash Weber number is defined as $We_s = \rho d_0 u_0^2 / \sigma$, where $d_0$ is the initial droplet diame-
5.1. Model description and cases

Figure 5.1: Droplet irradiated by the laser pulse. The resulting shockwave is calculated analytically and the velocity field is used as the VOF simulation initial condition. The vector indicates the direction of radiation.

Table 5.1: Numerical models solved for two phase the VOF model [121]. The nomenclature for the parameters are defined in the Nomenclature section.

<table>
<thead>
<tr>
<th>Equation Type</th>
<th>Mathematical Expression</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume Fraction equation</td>
<td>$\frac{1}{\rho_q} \left[ \frac{\partial}{\partial t} (v_q \rho_q) + \nabla \cdot (v_q \rho_q \vec{u}<em>q) \right] = m</em>{pq} - m_{qp}$ (a)</td>
</tr>
<tr>
<td>Explicit Scheme</td>
<td>$v_{q,i}^{i+1} \rho_{q,i}^{i+1} - v_{i}^{i} \rho_{q}^{i} \over t_{i+1} - t_{i} \cdot V_c + \sum_{f} \left( \rho_q U_f^i v_{q,f}^i \right) = \left( m_{pq} - m_{qp} \right) \cdot V_c$ (b)</td>
</tr>
<tr>
<td>Material Properties</td>
<td>$\rho = v_q \rho_q + (1 - v_q) \rho_p$ (c)</td>
</tr>
<tr>
<td>Momentum Equation</td>
<td>$\frac{\partial}{\partial t} (\rho \hat{u}) + \nabla \cdot (\rho \hat{u} \hat{u}) = - \nabla p + \nabla \left[ \mu \left( \nabla \hat{u} + \nabla \hat{u}^T \right) \right] + \rho \vec{g} + \vec{F}$ (d)</td>
</tr>
<tr>
<td>Surface Tension (implicit body force)</td>
<td>$F_{vol} = \frac{\sigma \kappa_q}{\frac{1}{2} (\rho_q + \rho_p)} \nabla \cdot v$ (e)</td>
</tr>
</tbody>
</table>
5.1. Model description and cases

ter, $\rho$ is the droplet fluid density, $\sigma$ is surface tension and $u_0 = \dot{R}(t = 0)$ is the deforming droplet rim initial extension rate. The Reynolds number is defined as $\rho d_0 \overline{V} / \mu$, where $\mu$ is the droplet viscosity and $\overline{V}$ is the impacted fluid mass mean velocity. The Ohnesorge number is defined as $Oh = Re^{-1} \sqrt{We}$. The dimensionless parameters for the laser pulse energy $E_p$ and irradiance $E_e$ are nondimensionalized with respect to the initial droplet surface energy as $\tilde{E}_p = E_p / (\sigma d_0^2)$ and $\tilde{E}_e = E_e \sqrt{\rho d_0^3 / \sigma^3}$. These parameters are relevant for a 1064 nm laser pulse interacting with liquid Sn. The maximum rim extension rate $u_0$ is taken from the simulation results.

The ambient pressure for the simulations were set at 1 bar. For lower pressures Fluent 18.0 struggled to converge, because the criteria for continuum flow was not being met. This pressure level is not expected to effect the fragmentation process of the expanding droplet sheet. The criteria for ambient gas influencing the sheet fragmentation is derived by Villermaux and Bossa \cite{61} as

$$We_s < We^*_s = \left( \frac{20 \pi \rho}{0.12 \rho_\infty} \right)^{2/3}$$ (5.1)

where $We^*_s$ is the minimum splash Weber number the droplet needs to reach for the ambient gas to influence the fragmentation process. For all of the cases measured or simulated in this dissertation $We_s \ll We^*_s$.

5.1.1 VOF geometry

All cases were run in a rectangular cuboid domain with hexahedral meshing (see Fig. 5.2). The size of the domain was chosen based upon the extent of the droplet breakup expansion. The velocity component of the droplet center of mass is removed from the simulation initial condition, such that the domain is a moving reference frame. Adaptive meshing was required in order to complete the simulation at a resolution of $\Delta x / d_0 \sim 0.012$. The mesh refinement was maintained at $\Delta x / d_0 \sim 0.012$ for regions where the fluid volume fraction was $\vartheta_l > 10^{-6}$. There were up to 6 levels of refinement between the coarse and fine cell sizes (see Fig. 5.3). The meshing was refined or coarsened after each time step, such that the refined mesh would follow
Table 5.2: Simulation case parameters including Weber number $We$, Reynolds number $Re$, Ohnesorge number $Oh$, density and dynamic viscosity ratio of the liquid phase “$l$” and the gaseous phase “$g$”, Knudsen number of the gas phase $Kn$, laser parameter $\sigma_G$ defined by equation (5.10), the dimensionless parameters for laser pulse energy $\tilde{E}_p$ and irradiance $\tilde{E}_e$, and the ratio of the maximum radial velocity $u_0$ to the mean velocity $\overline{V}$ calculated from the simulations.

<table>
<thead>
<tr>
<th>Case</th>
<th>$We_s$</th>
<th>$Re$</th>
<th>$\sigma_G$</th>
<th>$\tilde{E}_p$</th>
<th>$\tilde{E}_e$</th>
<th>$u_0/\overline{V}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>165</td>
<td>2’601</td>
<td>0.42</td>
<td>$6.9 \times 10^4$</td>
<td>$2.0 \times 10^9$</td>
<td>2.7</td>
</tr>
<tr>
<td>II</td>
<td>270</td>
<td>3’099</td>
<td>0.42</td>
<td>$9.1 \times 10^4$</td>
<td>$2.7 \times 10^9$</td>
<td>2.9</td>
</tr>
<tr>
<td>III</td>
<td>680</td>
<td>4’731</td>
<td>0.42</td>
<td>$1.8 \times 10^5$</td>
<td>$5.3 \times 10^9$</td>
<td>3.0</td>
</tr>
<tr>
<td>IV</td>
<td>1’800</td>
<td>8’521</td>
<td>0.42</td>
<td>$4.7 \times 10^5$</td>
<td>$1.4 \times 10^{10}$</td>
<td>2.7</td>
</tr>
<tr>
<td>V</td>
<td>4’100</td>
<td>13’612</td>
<td>0.42</td>
<td>$1.1 \times 10^6$</td>
<td>$3.1 \times 10^{10}$</td>
<td>2.5</td>
</tr>
<tr>
<td>VI</td>
<td>14’000</td>
<td>21’359</td>
<td>0.42</td>
<td>$2.3 \times 10^6$</td>
<td>$6.8 \times 10^{10}$</td>
<td>3.0</td>
</tr>
<tr>
<td>VII</td>
<td>24’000</td>
<td>29’493</td>
<td>0.42</td>
<td>$4.2 \times 10^6$</td>
<td>$1.2 \times 10^{11}$</td>
<td>2.8</td>
</tr>
<tr>
<td>VIII</td>
<td>14’000</td>
<td>58’707</td>
<td>0.72</td>
<td>$1.7 \times 10^7$</td>
<td>$4.5 \times 10^{10}$</td>
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</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$Oh$</th>
<th>$\rho_l/\rho_g$</th>
<th>$\mu_l/\mu_g$</th>
<th>$Kn$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.002</td>
<td>4’400</td>
<td>48</td>
<td>0.10</td>
</tr>
</tbody>
</table>

The sides of the rectangular cuboid were set to pressure outlet conditions with constant pressure. In order to convert the laser-droplet interaction into the initial conditions for the simulation, the spatial and temporal pressure profile acting on the unablated droplet surface needs to be determined for a given $E_e$ and $E_p$. A method has been validated for calculating the peak pressure $P_{\text{max}}$ generated on a liquid Sn droplet surface from a pulsed Nd:YAG laser for similar $We$ in Chap. 3 [60]. The same relation is used to approximate $P_{\text{max}}$ for the given $E_p$ and $E_e$ for cases I – VIII [40].

\[
P_{\text{max}} \approx 8 \left( I_0 / 10^{14} \right)^{0.7} \left[ 1 + \frac{l_{ac}^{(0)} \sin(\theta)}{w} \tau_p^{0.9} \left( I_0 / 10^{14} \right)^{0.3} \right]^{-1.4}, \tag{5.2}
\]
where $P_{\text{max}}$ is in Mbar, $I_0$ is in W cm$^{-2}$, $\tau_p$ is in ns, $l_{ac}^{(0)}$ is in $\mu$m, and $w$ is in $\mu$m. The surface pressure from the Gaussian laser profile projected onto the droplet spherical surface is approximated in Sec. 3.3 by the function

$$P(r, t) = P_{\text{max}} \cdot 2^{-4(t/\tau_p)^2} \left[ 1 - \left( \frac{r}{w} \right)^2 \right] \sqrt{1 - \left( \frac{r}{r_0} \right)^2}.$$

(5.3)

This relation is converted to spherical coordinates where the $P$ is a function of $\theta_0$ (see Fig. 5.4) using

$$r = r_0 \sin(\theta_0)$$

(5.4)

resulting in

$$P(\theta_0, t) = P_{\text{max}} \cdot 2^{-4(t/\tau_p)^2} \cos(\theta_0) \left[ 1 - \left( \frac{T_0}{w} \right)^2 \sin^2(\theta_0) \right].$$

(5.5)

In order to determine the velocity and pressure profile within the droplet volume after the shock has ceased we use the model derived by Gelderblom
5.1. Model description and cases

Figure 5.3: The adaptive mesh is refined/coarsened after each time step to follow the droplet fluid phase. Cross-section (a) shows an example center contour of the density distribution of the simulation with the mesh overlaid after the simulation has run to the first rim detachment. Cross-section (b) shows a magnified view of the rim cross section.

Figure 5.4: Layout of spherical coordinates for initial condition formulation.
et al. [62], Antkowiak et al. [122], which provides the following

\[ P(r_d, \theta_0) = \sum_{l=0}^{\infty} A_l r_d^l P_l[\cos(\theta_0)] \] (5.6)

\[ A_l = \frac{2l + 1}{2} \int_0^\pi f(\theta_0) P_l[\cos(\theta_0)] \sin(\theta_0) d\theta_0 \] (5.7)

\[ f(\theta_0) = P_{\text{max}} e^{-\theta_0^2/(2\sigma_G^2)} \] (5.8)

\[ \vec{u} = -\frac{\tau_p}{\rho} \nabla P(r_d, \theta_0) \] (5.9)

Where the laser profile parameter \( \sigma_G \) is determined from (5.5) by numerically solving

\[ \int_0^{\pi/2} \sin(\theta_0) \cos(\theta_0) \left[ 1 - \left( \frac{r_0}{w} \right)^2 \sin^2(\theta_0) \right] d\theta_0 = \int_0^\pi \sin(\theta_0) e^{-\theta_0^2/(2\sigma_G^2)} d\theta_0 \] (5.10)

The resulting volumetric velocity field is mapped to the droplet simulation for \( t = 0 \) (Fig. 5.5).

## 5.2 Results and discussion

In Figs. 5.6 - 5.9 the breakup sequence over time is shown for cases II, III, VI, and VIII as representative examples of the fragmentation process. We define the dimensionless time scale for these cases as \( \tilde{t} = t/\tau_c \), with \( \tau_c \) as the droplet capillary timescale \( \tau_c = \sqrt{\rho r_0^3/\sigma} \). The simulations were run until the fragments had ceased dividing. The first noticeable feature of these sequences is how \( R \) reaches a critical radius beyond which holes form at the rim which grow to release ligaments from the rim. The separated ligaments then continue to stretch until they break into smaller fragments. The perforations then propagate towards the center of the sheet. This breakup pattern is reminiscent of expanding sheet dynamics observed in splash plate atomizers with high Weber numbers [123]. For the lower \( We_s \) cases the finer fragments are ejected from the rim first and that there are progressively larger fragments that form from ligaments originating from the center. This will be more
5.2. Results and discussion

Figure 5.5: The velocity field of the simulation initial condition calculated for $\sigma_G = 0.42$. The darkness of the field is proportionate to the magnitude of the velocity.

clearly described in Sec. 5.2.5. At the highest $We_s$ cases $VII$ and $VIII$ there is a transition to burst dynamics where the film contracts into a web of ligaments that eventually fragment. These dynamics are reminiscent of a bursting thin liquid film, where as the film stretches it is perforated by a number of holes. These holes then grow until they connect with each other forming a web of ligaments that break into droplets [69, 124].

As shown in Fig. 5.10 the rim radius $R$ for each simulation is specified as the length of the line tracing the sheet midpoint from the sheet center to the rim. The splash Weber number $We_s$ is determined for each case with the same method used in Sec. 4.2 where $We_s$ is fitted to $\tilde{R}(\tilde{t})$ before sheet rupture occurs (shown in Fig. 5.11).

One observation from the simulations is that the local sheet thickness $h_r(r)$ follows a self-similar function that can be written as

$$\frac{h_r(r)}{h_{0,r}} = \frac{1}{2} \left[ 1 + \cos(f_s \frac{r}{R}) \right],$$

(5.11)
Figure 5.6: Simulation breakup sequence for case $II$ with $We_s = 270$ and $\sigma_G = 0.42$. The original droplet dimensions and position are included for scale.
Figure 5.7: Simulation breakup sequence for case $III$ with $We_s = 680$ and $\sigma_G = 0.42$. The original droplet dimensions and position are included for scale.
Figure 5.8: Simulation breakup sequence for case VI with $We_s = 14'000$ and $\sigma_G = 0.42$. The original droplet dimensions and position are included for scale.
Figure 5.9: Simulation breakup sequence for case VIII with \( We_s = 14'000 \) and \( \sigma_G = 0.72 \). The original droplet dimensions and position are included for scale.
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Figure 5.10: The deformed droplet cross section. $R$ is measured as the length of the fluid film center from the center to the rim. The mean film thickness $h(r)$ at point of rupture is included in the calculation of $\tilde{t}_b$.

Figure 5.11: $We_s$ is determined for each case by fitting $\tilde{R}(\tilde{t})$ to (4.2).
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where \( f_s \) is a function of the \( \sigma_G \), and \( h_{0,r} \) is \( h_r(r = 0) \). Figure 5.12 shows the plot of several case states compared with Eq. (5.11). As implied by Eq. (5.11), this self-similarity is not dependent upon \( \tilde{t} \) or \( We_s \), but will vary with \( \sigma_G \). The center thickness \( h_r(r = 0) \) is solved for as a function of \( \tilde{R} \) and \( f_s \) by integrating the volume of the expanding sheet from 0 to \( \tilde{R} \) and using conservation of mass such that

\[
\int_0^{\tilde{R}} 2\pi r h_r(r)dr \approx \frac{4}{3}\pi r_0^3, \tag{5.12}
\]

which solves in the dimensionless form as

\[
\tilde{h}_{0,r} = \tilde{h}\frac{f_s^2}{\pi}, \tag{5.13}
\]

where \( \tilde{h} \) is calculated from the expanding thin disc approximation Eq. (1.5).

5.2.1 Kinetic energy partition

Experimental studies were done under similar conditions to cases \( III - VIII \) in Chap. 3 [60]. The test parameters for these cases are described in Tab. 5.3. Initial comparison of the kinetic energy partition \( E_{K,d}/E_K = u_0^2/(u_0^2 + 2u_{cm}^2) \) as described in Gelderblom et al. [62] is shown in Fig. 5.13. \( u_{cm} \) is the velocity of the impacted droplet center of mass. Figure 5.13 shows that \( E_{K,d}/E_K \) for the simulation cases are comparable to the Gelderblom model and are close to the similar experimental cases in Hudgins et al. [60]. The kinetic energy partitions for the experimental results and the simulations are well predicted by the Gelderblom model. The fragmentation pattern observed by the experiments in Sec. 3.2 are different to the simulation cases. This is due to the difference in the initial perturbations of the simulation droplet surface compared to the experimental cases, which will be elaborated in detail in the following sections.
5.2. Results and discussion

Figure 5.12: In the CFD cases the sheet thickness $h_r(r)$ is observed to obey the self-similarity Eq. (5.11) for the part of the sheet that has not been perforated.

Table 5.3: Experimental case parameters from Chap. 3 and Hudgins et al. [60].

<table>
<thead>
<tr>
<th>Case</th>
<th>$We$</th>
<th>$Re$</th>
<th>$\sigma_G$</th>
<th>$E_p$</th>
<th>$E_c$</th>
<th>$u_0/V$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$iii$</td>
<td>81</td>
<td>5'030</td>
<td>0.42</td>
<td>$2.7 \times 10^5$</td>
<td>$7.9 \times 10^9$</td>
<td>2.3 ± 0.4</td>
</tr>
<tr>
<td>$iv$</td>
<td>300</td>
<td>9'670</td>
<td>0.42</td>
<td>$6.6 \times 10^5$</td>
<td>$1.9 \times 10^{10}$</td>
<td>1.9 ± 0.3</td>
</tr>
<tr>
<td>$v$</td>
<td>767</td>
<td>15'500</td>
<td>0.42</td>
<td>$1.3 \times 10^6$</td>
<td>$3.9 \times 10^{10}$</td>
<td>1.7 ± 0.2</td>
</tr>
<tr>
<td>$vi$</td>
<td>1'870</td>
<td>24'200</td>
<td>0.42</td>
<td>$2.7 \times 10^6$</td>
<td>$7.9 \times 10^{10}$</td>
<td>1.4 ± 0.2</td>
</tr>
<tr>
<td>$vii$</td>
<td>3'230</td>
<td>31'700</td>
<td>0.42</td>
<td>$4.5 \times 10^6$</td>
<td>$1.3 \times 10^{11}$</td>
<td>1.4 ± 0.1</td>
</tr>
<tr>
<td>$viii$</td>
<td>10'600</td>
<td>52'400</td>
<td>0.72</td>
<td>$2.1 \times 10^7$</td>
<td>$5.5 \times 10^{10}$</td>
<td>1.0 ± 0.02</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$Oh$</th>
<th>$\rho_l/\rho_g$</th>
<th>$\mu_l/\mu_g$</th>
<th>$Kn$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.002</td>
<td>$\rightarrow \infty$</td>
<td>48</td>
<td>500</td>
</tr>
</tbody>
</table>
5.2. Results and discussion

Figure 5.13: Kinetic energy partition comparison between the VOF simulation results for cases $I - VIII$, the model by Gelderblom et al. \cite{62} and experimental results in Hudgins et al. \cite{60}.
5.2. Results and discussion

Figure 5.14: A cross-sectional center slice of the initial droplet surface for wave mode analysis (black line). The mean radius $r_0$ is traced in red.

5.2.2 Rupture time

The rupture time of the droplet can be predicted following the same analysis as that derived in Sec. 4.4. The surface shape of the initial conditions are spherical with perturbations due to the grid resolution and the interpolation method. The initial wave modes can be analyzed by sampling the surface of a slice of the initial droplet surface (see Fig. 5.14). The radial deviation of the surface from $r_0$ is denoted as $H$ and nondimensionalized with respect to $r_0$ as $\tilde{H}$. The nondimensional length coordinate tangent to the droplet surface is $\tilde{s}$. In Figure 5.15 $\tilde{H}$ is plotted with respect to $\tilde{s}$ from the isoline data. $\tilde{H}(\tilde{s})$ is processed with a fast Fourier transform (FFT) outputting $\tilde{\eta}_0(k)$. In Figure 5.16 $\tilde{k}\tilde{\eta}_0$ is plotted versus $\tilde{k}$ in order to show the peak value that corresponds to the fastest growing wave mode $\tilde{k}_b$ according to (4.40) in Sec. 4.4 shown here again as

$$\tilde{t}_b \approx \frac{4}{3}(\tilde{\eta}_0\tilde{k}_b)^{-1}W e_{s^{-1/2}}.$$  \hfill (5.14)
5.2. Results and discussion

Figure 5.15: Dimensionless surface deviation from $r_0$ versus the tangential length coordinate $\tilde{s}$ taken from the isoline data in Fig. 5.14

Figure 5.16: The maximum product of the wave number $\tilde{k}$ and the perturbation amplitude $\tilde{\eta}_0$ determines the minimum breakup time $\tilde{t}_b$. 
As discussed in Sec. 5.2 the initial perforations appear near the rim of the deforming droplet where the sheet is thinnest. Since there is a non-negligible difference between the mass mean sheet thickness $\tilde{h}$ and the sheet thickness at the point of rupture $\tilde{h}_r(\tilde{r}_b)$, the mean sheet thickness at the point of rupture needs to be accounted for in order to calculate $\tilde{t}_b$. The radial location of the perforation appearance is denoted $\tilde{r}_b$. $\tilde{h}_r(\tilde{r}_b)$ is taken from the CFD results (see Fig. 5.10) and incorporated into (4.40) as

$$\tilde{t}_b \approx \frac{\tilde{h}_r(\tilde{r})}{\tilde{h}} \frac{4}{3} \tilde{\eta}_0^{-1} \tilde{k}_b^{-1} W e_s^{-1/2}. \quad (5.15)$$

The rupture time for the sheet center ($r_b = 0$) is also calculated from (5.15) and compared with the CFD results. At this point it is not possible to measure the equivalent simulation rim radius $\tilde{R}$ in order to calculate $\tilde{h}$, since many rim ligaments have already detached. Therefore, Eq. (1.4) and (1.5) are used to calculate $\tilde{h}(\tilde{t}_b, \text{sim})$ to find the mass mean sheet thickness.

In Figures 5.17 and 5.18 the simulated results are compared against the calculation of (5.15) across two orders of magnitude of $W e_s$ and found to be well predicted both for the rim rupture time and the center rupture time.

### 5.2.3 Fragment mass distributions

The simulation results were post-processed sorting each fragment by its volumetric diameter $d$ and the position and velocity of its center of gravity, which is represented schematically in Fig. 5.19. The fluid mesh was resolved to $\Delta \tilde{x} \approx 0.024$. It is noted that at this resolution the final break of the fluid will be numerical, because in reality the dynamics governing the fluid pinch off occurs on molecular length and time scales. The error produced in the VOF method used here will occur on the scale of the grid size. Higher levels of multi-scale modelling would be computationally cost prohibitive for this work. Since the major focus of this work is for the scaling of the larger fragments from the initial conditions, we ignore the calculated fragments on the smaller scale range of the fragment distribution accounting for $< 0.4\%$ of the total droplet mass. This is the fragment range of $\Delta \tilde{x}$. It was observed in the simulations that the size of the ligaments detached...
Figure 5.17: The comparison between $\tilde{t}_{b,\text{sim}}$ occurring within the simulation is compared against $\tilde{t}_{b}$ [Eq. (5.15)] calculated from the initial conditions across two orders of magnitude of $We_s$. The dependency of $\tilde{t}_{b}$ on the local sheet thickness at the rupture location is shown clearly in the difference in times between the rim $\tilde{t}_{b}$ and the $\tilde{t}_{b}$ at the center. The points sitting outside of the lines correspond to case VIII, where $\sigma_G = 0.72$ whereas the other cases have $\sigma_G = 0.42$. The larger $\sigma_G$ leads to a reduced difference between $\tilde{h}_r(\tilde{r} = 0)$ and $\tilde{h}_r(\tilde{r} \approx \tilde{R})$. 
Figure 5.18: A direct comparison between $\tilde{t}_{b,\text{sim}}$ occurring within the simulation is compared against $\tilde{t}_b$ [Eq. (5.15)] calculated from the initial conditions across two orders of magnitude.
5.2. Results and discussion

Figure 5.19: The simulations were post processed such that each fragment was cataloged by its volumetric diameter $d$, its location in space, and its velocity vector $\vec{V}$. The angle $\alpha$ is defined as the angle of the debris fragment velocity vector relative to the laser center axis.

from the rim is dependent on the sheet thickness of the point of rupture. Since the sheet thickness varies radially, the ligament sizes also vary depending upon which radial location of the sheet they detached from. This is illustrated by the mass fraction $\nu_d$ distribution of the fragments shown in Figs. 5.20(a) - 5.20(h), where the fragments are divided by trajectory. The fragment trajectory is specified by the $\alpha$, which is the angle of the debris fragment velocity vector relative to the laser center axis. At first glance it can be seen that as $We_s$ increases more of the fluid mass is distributed in the smaller fragment sizes. The figures show that the fragments ejected near the rim are smaller on average than the fragments ejected near the sheet center region. This is expected since the sheet is thicker in the center than the rim and the ligament diameter is proportionate to the sheet thickness when it is detached [125]. By reducing the variability of the ligament size dependence on the separation location, the parameters influencing the fragmentation dynamics can be more clearly identified in the following section.
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Figure 5.20: Mass fraction of droplet fragments $\nu_d$ vs $d$ size for each simulation case.
5.2.4 Fragment size distributions

The fragment size distributions were analyzed by plotting the probability density function (PDF) of the fragment sizes derived from the simulation results after the breakup had ceased. The PDF’s for cases III - VIII are shown in Figs. 5.21(a) - 5.21(f). For the PDF’s shown in this work, data points were plotted with a confidence interval of 95% [126]. It has been found that the droplet fragment distributions are well represented by gamma distributions defined as [127, 61]

\[
\Gamma(n_P, x = d/d_{10}) = \frac{n_P^n}{\Gamma(n_P)} x^{n-1}e^{-nx}; \quad \Gamma(n_P) = \int_0^\infty x^{n-1}e^{-nx}dx;
\]

\[
PDF(d) = \frac{1}{d_{10}} \Gamma(n_P, x = d/d_{10}),
\]

where \(d_{10}\) is the mean splash fragment diameter and the parameter \(n_P\) determines the width of the PDF. From Dombrowski and Johns [125] the ligament diameters for the disintegrating sheet can be calculated as

\[
\tilde{d}_L = \sqrt{4\tilde{h}_r(\tilde{r}_b)\frac{\tilde{R}}{\tilde{k}_{max}}},
\]

where \(\tilde{d}_L\) is the mean ligament diameter and \(\tilde{k}_{max}\) is divided by \(\tilde{R}\) in order to account for self-similar stretching. From the ligament diameter the mean droplet diameter \(d_{10}\) can be calculated using conservation of mass as [125]

\[
\tilde{d}_{10} = \left(2\pi d_L^2\frac{\tilde{R}}{\tilde{k}_{max}}\right)^{1/3} = [8\pi \tilde{h}_r(\tilde{r}_b)]^{1/3} \left(\frac{\tilde{R}}{\tilde{k}_{max}}\right)^{2/3}.
\]

Equation (5.19) predicts \(d_{10}\) within an 18% standard of deviation across cases III-VIII. Cases I and II did not have enough fragments for a proper
5.2. Results and discussion

Statistical comparison. \( n_P \) is calculated as \[ 128 \] \[ 127 \]

\[
n_P = n_P(\infty) e^{\gamma \sqrt{\gamma^2 + 1}},
\]

\[
n_P(\infty) \approx \left\{ \frac{d_L}{2 \max[\eta(t = 0)]} \right\}^2, \text{ and}
\]

\[
\gamma = \frac{u_0}{R(t_b)} \sqrt{\frac{\rho d_L^3}{8\sigma}},
\]

where \( \gamma \) is the dimensionless ligament stretch rate.

It is observed that there is not a large variation of the mean fragment size \( d_{10} \) ejected from the rim of the droplets over the range of \( W_e \). Utilizing the Rayleigh Taylor instability model derived in Sec. 4.4, the perforation time for all of the wave modes are calculated from Eq. (4.40) and shown in Fig. 5.22. It can be seen here that a spectrum of modes will puncture the sheet up to \( \sim \tilde{k}_{\max} \), which in the cases run is limited by the grid resolution.

The breakup wavenumber \( \tilde{k}_b \) will determine the first hole appearance time of the sheet, but the population of the perforations will be determined by a superposition of all of the wavemodes where \( \tilde{t}_b(\tilde{k}) < \tilde{t}_{\max} \). From Fig. 5.22 it can be seen that the largest wave mode \( \sim \tilde{k}_{\max} \) in the initial conditions will determine the dominant perforation spacing since \( \tilde{t}_b(\tilde{k}_{\max}) \) is on the same order of the lower wave modes.

A mesh sensitivity study was done where the mesh size for Case IV was varied by \( \Delta \tilde{x} = 0.024, 0.048, \) and \( 0.096 \) and the fragment size distribution was predicted from the simulation initial surface conditions using Eqs. (5.17) and (5.19). The predicted results from these equations are compared against the simulation results in Figs. 5.23 and 5.24. The simulation results are well predicted by the equations. These results demonstrate that the initial surface perturbations on the droplet simulation surface, which are an artifact of the VOF grid resolution, are one of the primary determinants of the simulation fragment size distribution.

5.2.5 Fragment spatial distributions

The dependence of the fragment distribution on the laser parameters is an element of interest in this work. The fragment spatial distributions are plot-
Figure 5.21: Droplet fragment size PDF’s plotted for (a) case III, (b) case IV, (c) case V, (d) case VI, (e) case VII, (f) case VIII. The dotted lines are calculated from Eq. (5.17).
Figure 5.22: The time $\tilde{t}_b(\tilde{k})$ for each initial wave mode of the droplet surface initial condition to rupture the sheet for case VII. The red dotted line demarcates $\tilde{t}_{max} = 0.385$. 
Figure 5.23: $\tilde{d}_{10}$ is plotted for Case IV simulation results with a mesh resolution of $\Delta \tilde{x} = 0.024$, 0.048, and 0.096. The prediction of Eq. (5.19) from the simulation initial conditions is compared to the simulation results and found to be in close agreement.
Figure 5.24: PDF($\tilde{d}$) is plotted for Case IV simulation results with a mesh resolution of $\Delta \tilde{x} = 0.024, 0.048$, and 0.096. The prediction of Eq. (5.17) from the simulation initial conditions is compared to the corresponding simulation results and found to be in close agreement.
5.2. Results and discussion

related with respect to the angle $\alpha$, which is the angle of the fragment velocity relative to the laser center axis vector (see Fig. 5.19). In Figures 5.25(a)-5.25(f) the mass flux of the splashed droplet is plotted for cases III-VIII. For cases III - VII there is a general weighting of the fragments towards the center axis, with the majority of the mass ejected at angles below $80^\circ$. For case VIII the majority of the mass is ejected within a range of $\alpha < 45^\circ$. This is clearly the influence of the laser pressure profile parameter $\sigma_G$ (see Tab. 5.2). When the splash range is calculated as $\alpha_{lim} = \tan^{-1}(u_0/u_{cm})$ for cases III - VIII the range is $\alpha_{lim} = 74 \pm 5^\circ$. For case VIII $\alpha_{lim} = 45^\circ$. This dependency of the fragment trajectory on the pressure profile has been covered in relation to the kinetic energy partition in Sec. 5.2.1. For case III the mass distribution is the most concentrated than the higher cases towards the center. It can be more clearly understood from the results in Figs. 5.26(a)-5.26(f) where the droplet sizes are plotted as a function of $\alpha$. As mentioned in Sec. 5.2.4, the perforating wave modes will be dependent on the whether the wave modes can grow fast enough to puncture the sheet, before sheet retraction reverses the local sheet thinning. This dynamic is highlighted in Fig. 5.26(a) for case III at the range of $\alpha < 60^\circ$ where the sheet thickness becomes high enough to diminish the influence of the higher wave modes, leading to larger fragments that can be seen in the local mass mean fragment diameter $\bar{d}_{43}$ and the Sauter mean diameter $\bar{d}_{32}$. The fragments formed from the ligaments ejected from the rim are typically the smallest with the narrowest size distribution, as seen from the converging of the three diameter terms towards the $\alpha_{lim}$.

5.2.6 Splash threshold

The splash criteria is defined as the conditions where the droplet deformation leads to the ejection of fragments. Using the model detailed in Sec. 5.2.2, it is known that if the rupture time exceeds the time for the expanding sheet to reach the maximum rim extension, then the sheet thickness overcomes the perturbation growth and sheet puncture will not occur. This limit can be represented as

$$\tilde{t}_{max} \approx \frac{4}{3}\tilde{\eta}_0^{-1}\tilde{k}_b^{-1}We_{s,cr}^{-1/2},$$  (5.21)
Figure 5.25: Average mass fraction of fragments per steradian \( m'' \) versus the angle relative to the laser center axis for (a) case III, (b) case IV, (c) case V, (d) case VI, (e) case VII, and (f) case VIII. The fragment distribution is substantially narrower for case VIII, which has a 71% wider \( \sigma_G \) than the other cases.
5.2. Results and discussion

Figure 5.26: Droplet size distributions versus the angle relative to the laser center axis for a) case I, b) case II, c) case III, d) case IV, e) case V, f) case VI.
where $W_{e_{s,cr}}$ is the splash Weber number at which the impacted droplet can be expected to eject fragments. Equation (5.21) is rewritten as

$$W_{e_{s,cr}} \approx 12\tilde{\eta}_{0}^{-2}\tilde{k}_{b}^{-2}. \quad (5.22)$$

Solving for the critical splash Weber number $W_{e_{s,cr}}$ for cases $I - VII$ gives

$$W_{e_{s,cr}} \approx 154. \quad (5.23)$$

Cases were run close to $W_{e_{s,cr}}$ in order to observe this threshold. The results are shown in Fig. 5.27. It can be seen that for $W_{e_{s}} < W_{s,cr}$ the rim expands to a maximum amplitude and then retracts without fragmentation. For $W_{e_{s}} \sim W_{s,cr}$ perforations form near the rim at the maximal amplitude, but the droplet does not eject fragments and eventually the fluid retracts back together. For $W_{e_{s}} > W_{s,cr}$ the first ligament detachment from the rim can be seen.

### 5.3 Summary

The fragmentation of droplets were modelled using multi-phase three dimensional VOF simulations, where high resolution is achieved through adaptive meshing. The adaptive meshing provides the advantage of substantially reducing computational costs for computing breakup dynamics within domain volumes many orders of magnitude greater than the droplet fluid volume. Several cases were run for increasing laser pulse intensity. The deforming droplet shape evolution over time was analyzed as function of the laser input parameters. The kinetic energy partition of the simulations were compared against similar experimental cases and theoretical models. The initial surface perturbations of the simulations were extracted from the CFD model using an FFT. From the initial surface conditions, the splash Weber number $W_{e_{s}}$, and the sheet thickness $h_{f}(r_{b})$, the first hole appearance time $\tilde{t}_{b}$ was calculated from the derived analytical model and compared against the $\tilde{t}_{b}$ observed in the simulations. The diameters and trajectories of the droplet fragments at the end of the fragmentation process were extracted for each case. The mass fraction of the droplet fluid was sorted by fragment size and
Figure 5.27: The splash transition shown with three cases for \( We_s < We_{s,cr} \), \( We_s \sim We_{s,cr} \), and \( We_s > We_{s,cr} \) (\( \sigma_G = 0.42 \) and \( Oh = 0.0019 \)).
compared across all cases. The droplet fragment diameter PDF’s were plotted for each case and compared against the expected Gamma distribution. The mean fragment diameter is compared against a theoretical model for the disintegration of a fluid sheet. A relation is derived to predict the droplet splash threshold as a function of the initial conditions.
Chapter 6

Conclusions and recommendations for the future

The research objectives of this work are to determine the dynamics that govern the deformation and fragmentation of a fluid droplet from one or more series of laser pulses. The dynamics of a droplet fragmented from an ns scale pulse and for a series of picosecond bursts have been described by an analytical model that stems from the initial ablation pressure wave interaction with the droplet. The analytical models and formulas in this work have been validated for the corresponding laser and droplet conditions. They have not been validated for any other conditions not described here. Nevertheless, a multivariate analysis has been completed correlating several key droplet and laser impulse parameters over a range of measured dynamics. The findings of this work identify potential avenues of future investigation in the field of droplet target shaping.

6.1 Studies

Several experimental and numerical investigations were conducted throughout this work that facilitated the development of a simplified analytical
model capable of describing the complex fluid dynamic processes governing laser/droplet interaction. The first set of studies investigated the droplet breakup dynamics from a single nanosecond scale laser pulse. The second set of studies investigated the more complex dynamics of varying bursts of picosecond pulses interacting with tin droplets. The last set of investigations were done through numerical modeling of a droplet fragmented by a nanosecond pulse.

### 6.1.1 Single pulse dynamics

Experiments were done imaging the interaction between single laser pulses in the nanosecond range interacting with a droplet. By measuring the mean droplet debris velocity of the ejected fragments $\bar{V}$ in comparison to the irradiance of the laser pulse $E_e$, the data shows a power law dependence of $\bar{V} \propto E_e^n$ with $n \approx 0.65$. A scaled analytical model is derived modeling the conservation of energy between the unablated droplet and the plasma recoil pressure. This model predicts the neutral cluster kinetic energy and is validated from the experimental results.

From the experimental observations of neutral debris clusters generation and expansion, it was demonstrated that there is a relationship between droplet debris trajectory and the droplet alignment with the laser. The derived analytical model determines that the neutral cluster debris trajectory for an ablated droplet is a function of the laser profile $f_L$, the droplet diameter $d_0$ and the axial misalignment $\psi$ between the laser axis and the droplet center. From these three parameters the mean debris trajectory $\alpha_d$ can be estimated.

The ablation pressure acting on the droplet surface occurs on a much faster timescale than the droplet deformation. Therefore the initial pressure force acting on the unablated material acts approximately normal to the droplet surface. This results in an ejection of the neutral debris in a direction dependent on the incident irradiance profile and the droplet surface normal. Although the neutral cluster debris trajectory is shown to have a measurable dependence on laser-droplet alignment, the coupling dependence between the laser and the plasma supports that no measurable impact on the plasma radiation emission for the debris dynamics should be expected.
6.1.2 Picosecond burst dynamics

Experiments were performed imaging sequences of shadowgraphs of droplets irradiated by bursts of picoseconds laser pulses that varied in the number of pulses, the energy distribution of the pulses within the burst, and the total burst energy. From the experimental observation a multitude of different breakup dynamics were observed, which can be demarcated into three general categories. These categories are defined in this work as sheet expansion, cavitation, and jetting.

For sheet expansion the droplet spreads after impact as a thin fluid sheet where the fluid rim radius follows the dynamics of a droplet impact in free space, with no noticeable deviation due to sheet concavity. The modified Weber number $We_s$ is the main parameter describing the rim evolution until the sheet fragmentation, where the velocity scale is $u_0 = \dot{R}(t = 0)$. This has also lead to a distinction of the definition of the inertial timescale for a laser impacted droplet as $\tilde{\tau}_i \sim 0.22 We_s^{-1/2}$ from the condition $h(\tau_i)/r_0 \sim 1$.

It is shown how the kinetic energy transferred to the droplet can be approximated by the sum of the work deposited to the droplet surface by each individual pulse when the peak ablation pressure is known. It is further shown that when there is an exponential dependence of the ablation pressure on the pulse irradiance of $n < 1$, then the kinetic energy transferred to the target will scale by $\sim 0.5 N_p^{1-2n}(N_p + 1)$, when the burst energy is distributed evenly among $N_p$ pulses. Increasing $N_p$ has also been shown to increase the depth of the cup of the deformed sheet, by increasing the duration of the acceleration seen by the droplet surface.

The next category of droplet breakup dynamics is the effect of cavitation within the droplet due to the focused rarefaction wave within the droplet. The shock wave intensity increases with single pulse energy. The threshold condition for cavitation is explained by modeling the rarefaction wave intensity within the droplet when it reaches the region near the droplet center. When the rarefaction wave intensity exceeds the tensile strength of the fluid where the velocity field is divergent, then cavitation occurs. The cavity growth rate is proportionate to the tensile stress of the rarefaction wave. The faster the cavity growth rate, the more the cavitation dynamics will be superimposed with the sheet expansion dynamics. This is further applied to the cases where the cavitation bubble growth can be predicted and thereby leads
6.1. Studies
to the explanation of the size and appearance of high velocity jets ejected behind the droplet. With multiple high intensity shock waves small high speed jets are ejected behind the droplet. This phenomena is attributed to hydrodynamic focusing of subsequent shock waves interacting with the previously formed cavity within the droplet.

The timing of the first hole appearance $\tilde{t}_b$ in the droplet sheet is measured for the range of cases. The derived analytical model connects $\tilde{t}_b$ with the evolution of Rayleigh-Taylor instabilities initiated by the acceleration of the droplet surface and the removal of droplet material. The experimental data supports the claim of damping of the shorter wavelength hydrodynamic instabilities during the laser ablation induced droplet surface acceleration. One of the first apparent uses of this model is in the field of droplet target shaping in order to predict the maximum target surface size for the given laser parameters before disintegration. The model is also a key component in the prediction of the fragment sizes of the fragmenting droplet. The experimental results imply that the ablation depth is the linking factor to the initial perturbation amplitude of the droplet surface. The rupture time model offers an explanation for why the droplet has a tendency to rupture in the center of the expanding sheet instead of the sheet rim where the sheet is thinner. The laser profile is imprinted on the droplet surface and propagates with the sheet expansion. Therefore, the initial perturbations near the sheet center are expected to be large enough to compensate for the thicker sheet center in terms of where rupture occurs first. This is also indicated in the work by Klein [70, see p. 143].

Current EUV sources in the field today use a combination of an Nd:YAG pre-pulse and a main pulse from a CO$_2$ laser. The results here warrant a brief comment on the application of laser bursts for target shaping in these systems. By switching from single high energy pico/femto-second pulses to a burst of pulses, there is a potential to reduce the required pre-pulse laser power for a given target expansion speed due to the kinetic energy scaling. This includes a reduction in the fast ion/neutrals debris generated by the pre-pulse [68]. It should also be noted that the dimensions reached by the targets shown in Figs. 4.5 and 4.9(a) are close to the cavity dimensions described in Ueno et al. [37] where the CE of the laser/target interaction was increased from 2.2% to 4%. These EUV sources may be able to take advantage of the inertial confinement properties of these deep cup target shapes and generally
the finer control over the target shape that shaped laser bursts provide.

### 6.1.3 Droplet fragmentation

Several CFD simulations were run of droplets impacted by a single laser pulse using a multi-phase three dimensional VOF approach, where high resolution is achieved through adaptive meshing. These simulations provide a useful basis for deriving and validating analytical fluid dynamic models of the droplet fragmentation process. The major advantage is the resolution at which the fluid dynamics can be captured in terms of timing, fragment size and fragment trajectory.

A first observation is that the thickness profile of the expanding droplet sheet, absent perforations, is found to hold a self-similarity that is independent of $t$ or $We_s$. It is clear that the location of the initial surface perturbations play a key role in the location of the first sheet hole perforations. In the CFD simulations the initial surface perturbations are evenly distributed around the droplet surface. As the droplet deforms and these perturbations evolve, it is expected that the thinnest part of the sheet will rupture first as observed numerically. Due to the sheet thickness $h_r(r)$ being thinnest near the rim, the perforations appear near the rim first and populate into the sheet as it expands and thins.

Experimentally it is most often observed that the center of the sheet perforates first. This supports the conclusions made in Sec. 4.5 that the initial perturbation amplitude is directly dependent on the ablation depth, which is also intuitively correct. Therefore, one would expect that the ablation zone perturbations, located at the sheet center, would grow as the sheet expands and thins. These perturbations would be ($\mathcal{O}$) $\sim$100 nm compared to the typical initial fluid perturbations on the order of the Van der Waals radius for the fluid ($\mathcal{O}$) $\sim$0.2 nm. From this principle it is reasonable to expect the sheet center to rupture first.

The fragment size distribution is well predicted from analytical models of a disintegrating fluid sheet when the breakup wavemodes are known. The derived Rayleigh-Taylor model allows the identification of the relevant breakup wavemode. This model has further provided the relation for the predicting the rupture threshold specified as $We_{s,cr}$. 

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6.2 Recommendations for future studies

ANSYS was used for the investigation of an incompressible flow problem. It should be noted that the ANSYS commercial code is not considered suitable for modeling the compressible deformation of the droplet in the picosecond pulse impact regime. The commercial code is closed source and will thereby be difficult to adapt to include the multi-scale complex processes occurring from one or more ultrashort laser pulse induced shock waves. For future simulation work of this nature an open source simulation code such as OpenFOAM would be preferable.

6.2 Recommendations for future studies

There are several worthwhile avenues of investigation that can be explored using this work as a spring board. The deep cup shapes and umbrella-like targets that are created with the picosecond laser bursts show potential for effecting the plasma evolution and the debris distribution relative to a spherical or disc-shaped target. Specifically, one would expect inertial confinement effects to influence the plasma absorption when the main laser irradiates the inner surface of the cup, which may extend the duration and CE of the plasma. A thin film target would be expected to have a smaller fragment distribution due to the wavenumber cutoff found in laser ablation accelerated targets. One could approach the analysis by approximating the expanded sheet, which is disintegrating after the impact of the main pulse, as a web of ligaments whose spacing is determined by $\lambda_h$ as shown in Fig. 6.1. Using conservation of mass the ligament size could be approximated as

$$\tilde{d}_L \approx \sqrt{\frac{4}{\pi \sqrt{3} \tilde{R}(\tilde{t}_b)^2}}$$

The wavemode $k_{max}$ that is excited during the main laser pulse would not be not be subject to self-similar stretching as is the case of a spherical target, which means a reduction of $\lambda_h$ by a factor of $1/\tilde{R}(\tilde{t}_b)$. This would imply a reduction in the ligament diameter $\tilde{d}_L$ according to Eq. 6.1 of $\sim [\tilde{R}(\tilde{t}_b)]^{-3/2}$. Measurements of the difference in debris distribution would need to be done in order to confirm if such a scaling is indeed the case. These measurements could be done with a shadowgraph system or with a system measuring the
Figure 6.1: The geometric approximation of the web of ligaments formed by the perforated sheet, where $\lambda_h$ is the approximate hole spacing and $d_L$ is the ligament diameter.
6.2. Recommendations for future studies

fragment size utilizing Mie scattering [54]. The analytical models presented in this work should be useful tools for choosing a measurement system of the appropriate spatial and temporal resolution to capture the debris distribution.

More work is required to create a CFD simulation of the droplet with initial surface conditions, in terms of the initial perturbation amplitudes, that approximate well the experimental ablated surface. Work in this direction would facilitate a more thorough theoretical bridging between the laser/droplet interaction and the droplet fragmentation.

A more fundamental investigation that is of practical significance to the optimization of droplet-based LPP sources is the deflection of the target before, during, or after the main pulse, such that the heaviest distribution of neutral clusters is deflected away from the collector optics, without trading in other aspects of the system performance.

Finally, there is a lot of potential for further study that is open here in the field of fluid dynamics. The picosecond burst shaping has shown that there is a degree of programmability into the shape of the deforming droplet depending upon the burst profile, evidenced by the umbrella-like shapes. This phenomenon warrants further investigation into what extremes this dynamic can be pushed in terms of the complexity of shapes that can be created. This includes the investigation of different laser profile shapes beyond a Gaussian intensity profile. For example, polygon shapes, such as triangles and hexagons etc. would be expected to deform the target favoring these shapes. Further studies could investigate the influence of multiple shaped profiles for changing the droplet form, such as a triangle spot impulse followed by a 60°-rotated triangular spot after a short delay.
Appendix A

Droplet sheet rim velocity vs mass averaged fluid velocity

In order to calculate the expected $W_{es}$ from the laser parameters it is necessary to approximate the mean fluid velocity $\bar{u}$ from the measurable parameters of the expanding fluid sheet $Z$, $D_r$, and $u_0$. The mean velocity is calculated using a volume integral as

$$\bar{u} = \frac{\int_0^R s(r) h u(r) \, dr}{\int_0^R s(r) h \, dr}, \quad (A.1)$$

where $r$ is the path length along the sheet surface from expanding sheet center towards the rim, $s(r)$ is the sheet circumference concentric with the laser center axis, and $u(r)$ is the fluid velocity relative to the sheet center of mass. The dimensions of the expanding sheet are approximated as a spherical cap as shown in Fig. A.1. When $Z/D_r = 0.5$ this is a hemisphere shape and for $Z/D_r > 0.5$ the circumference remains constant for $r > \pi/(4D_r)$. The angle $\Theta$ is the full angle of the spherical cap rim relative to the cap radius of
curvature $R_c$ and is related to $Z$ and $D_r$ as

$$\frac{Z}{D_r} = \frac{1 - \cos\left(\frac{\Theta}{2}\right)}{2 \sin\left(\frac{\Theta}{2}\right)}; \Theta < \pi. \quad (A.2)$$

$\Theta$ can be approximated as a polynomial fit of (A.2) given as

$$\Theta \approx \begin{cases} -4.6\left(\frac{Z}{D_r}\right)^2 + 8.6 \frac{Z}{D_r} - 0.02, & \frac{Z}{D_r} \leq 0.5 \\ \pi, & \frac{Z}{D_r} > 0.5 \end{cases}. \quad (A.3)$$

The dimensions $R_c$, $R$, and $s(r)$ are calculated in succession as

$$R_c = \begin{cases} \frac{D_r}{2 \sin\left(\frac{\Theta}{2}\right)}, & \frac{Z}{D_r} \leq 0.5 \\ \frac{D_r}{2}, & \frac{Z}{D_r} > 0.5 \end{cases}, \quad (A.4)$$

$$R = \begin{cases} R_c \frac{\Theta}{2}, & \frac{Z}{D_r} \leq 0.5 \\ \frac{D_r}{2} \left(\frac{\pi}{2} - 1\right) + Z, & \frac{Z}{D_r} > 0.5 \end{cases}, \quad (A.5)$$

and

$$s(r) = \begin{cases} 2\pi R_c \sin\left(\frac{r}{R_c}\right), & \frac{r}{R_c} \leq \frac{\pi}{2} \\ \pi D_r, & \frac{r}{R_c} > \frac{\pi}{2} \end{cases}. \quad (A.6)$$

The location of the sheet center of mass will lie on the laser center axis. The distance of the sheet center of mass relative to the sheet center of curvature is approximated as

$$x_{cm} = \begin{cases} \frac{R_c}{2} \frac{\sin^2\left(\frac{\Theta}{2}\right)}{1 - 2 \cos\left(\frac{\Theta}{2}\right)}, & \frac{Z}{D_r} \leq 0.5 \\ \frac{1}{2}(D_r - Z), & \frac{Z}{D_r} > 0.5 \end{cases}. \quad (A.7)$$
Figure A.1: The schematic of the dimensions approximation of the fluid expanding sheet as a function of $Z$ and $D_r$. The shaded region is representative of an expanding fluid sheet of the shown $Z$ and $D_r$. The circumference $s(r)$ is constant at $\pi D_r$ for $\Theta > \pi$. 
Figure A.2: Calculation of $u_0/\bar{u}$ by numerical integration in comparison with the polynomial curve fit shown in Eq. (A.14). In comparison, for a flat expanding fluid disc $u_0/\bar{u} = 3/2$. 
The center of mass location with respect to \( r \) is calculated as

\[
r_{cm} = \begin{cases} 
R_e \cos^{-1}\left(\frac{x_{cm}}{R_e}\right), & x_{cm} \geq 0 \\
\frac{\pi}{2} R_e + x_{cm}, & x_{cm} < 0 
\end{cases}.
\]  
(A.8)

The velocity of the fluid with respect to the droplet center of mass can now be calculated as

\[
u_{cm} = \begin{cases} 
u_0 \frac{r_{cm}}{R} \sin\left(\frac{r_{cm}}{R_e}\right), & x_{cm} \geq 0 \\
u_0 \frac{r_{cm}}{R}, & x_{cm} < 0 
\end{cases}.
\]  
(A.9)

The velocity \( u_r(r) \) is the velocity component relative to the local direction of \( r \) and is related to the rim velocity \( u_0 \) as

\[u_r(r) = u_0 \frac{r}{R}.
\]  
(A.10)

From this relation the velocity components of the fluid relative to the sheet center of mass are calculated as

\[
u_x = \begin{cases} 
u_0 \frac{r}{R} \sin\left(\frac{r}{R_e}\right) - u_{cm}, & \frac{r}{R_e} \leq \frac{\pi}{2} \\
u_0 \frac{r}{R} - u_{cm}, & \frac{r}{R_e} > \frac{\pi}{2} 
\end{cases},
\]  
(A.11)

\[
u_y = \begin{cases} 
u_0 \frac{R}{R} \sin\left(\frac{r}{R_e}\right), & \frac{r}{R_e} \leq \frac{\pi}{2} \\
u_0 \frac{R}{R}, & \frac{r}{R_e} > \frac{\pi}{2} 
\end{cases}.
\]  
(A.12)

Whereby the fluid velocity magnitude can be calculated

\[u(r) = \sqrt{u_x^2 + u_y^2}.
\]  
(A.13)

These relations allow for the computational solving of Eq. (A.1) as a function of \( Z/D_r \), where a polynomial fit is generated for simplicity as

\[
\frac{u_0}{u} \approx - \left(\frac{Z}{D_r}\right)^3 + 2.1 \left(\frac{Z}{D_r}\right)^2 - 0.45 \frac{Z}{D_r} + 1.5.
\]  
(A.14)
The polynomial fit of $u_0/\bar{u}$ is shown in Fig. A.2 over the range relevant for this work.
Appendix B

Rupture time derivation simplification

In Section 4.4 the simplification step between Eq. (4.36) and (4.37) can be done, because the test conditions permitted the approximation. The approximation was checked in this work by comparing the results of Eq. (4.37) to the complete solution of Eq. (4.36) (solved computationally) and found to have negligible differences. There may be test conditions, where the complete solution is necessary, therefore the full derivation is provided here specifying where the small angle approximation is used and which terms are neglected thereafter.

The final amplitude function $f_3(\tilde{t})$ for phase 3 in Sec. 4.4 is derived as

$$f_3(\tilde{t}) = f_2(\tilde{t}_t) \cos \left[ \sqrt{\frac{\tilde{h}}{2}} \left( \frac{\tilde{k}}{\tilde{R}} \right)^2 (\tilde{t} - \tilde{t}_t) \right]$$

$$+ \dot{f}_2(\tilde{t}_t) \sqrt{\frac{2}{\tilde{h}}} \left( \frac{\tilde{k}}{\tilde{R}} \right)^{-2} \sin \left[ \sqrt{\frac{\tilde{h}}{2}} \left( \frac{\tilde{k}}{\tilde{R}} \right)^2 (\tilde{t} - \tilde{t}_t) \right].$$

(B.1)

For simplicity of the handling of the formulas certain terms are substituted with the following coefficients:

$$a_1 = \sqrt{\frac{3}{4} \tilde{t}_t \tilde{k}},$$

(B.2)
\[
a_2 = \sqrt{\frac{4}{3}} W e_s, \quad (B.3)
\]
\[
a_3 = -\sqrt{\frac{3}{4}} \tilde{k}, \quad (B.4)
\]
\[
a_4 = \sqrt{W e_s \tilde{k}}, \quad (B.5)
\]
\[
a_5 = \sqrt{\frac{3}{2}} R^3 \tilde{k}^{-2}, \quad (B.6)
\]
\[
a_6 = (\tilde{t} - \tilde{t}_t) \sqrt{\frac{2}{3}} R^{-3} \tilde{k}^2. \quad (B.7)
\]

These coefficients are substituted into the terms in (B.1) as

\[
f_2(\tilde{t}_t) = \cos(a_1) + a_2 \sin(a_1), \quad (B.8)
\]
\[
f_2(\dot{\tilde{t}}_t) = a_3 \sin(a_1) + a_4 \cos(a_1), \quad (B.9)
\]
\[
f_3(\tilde{t}) = f_2(\tilde{t}_t) \sin(a_6) + f_2(\dot{\tilde{t}}_t) a_5 \sin(a_6). \quad (B.10)
\]

This allows Eq (B.11) to be rewritten as

\[
f_3(\tilde{t}) = [\cos(a_1) + a_2 \sin(a_1)] \sin(a_6) + a_5 [a_3 \sin(a_1) + a_4 \cos(a_1)] \sin(a_6), \quad (B.11)
\]

which reduces to

\[
f_3(\tilde{t}) = \frac{1 - a_3 a_5}{2} \cos(a_1 + a_6) + \frac{1 + a_3 a_5}{2} \cos(a_1 - a_6) \ldots \quad (B.12)
\]
\[
+ \frac{a_2 + a_4 a_5}{2} \sin(a_1 + a_6) + \frac{a_2 - a_4 a_5}{2} \sin(a_1 - a_6).
\]

For the experimental test parameters the trigonometric terms are

\[
a_1 \pm a_6 = \mp \sqrt{\frac{2}{3}} \tilde{k}^2 \tilde{t} + (\tilde{k} \sqrt{\frac{3}{4}} \pm \sqrt{\frac{2}{3}} \tilde{k}^2) \tilde{t}_t \ll \frac{\pi}{2}. \quad (B.13)
\]
Therefore the small angle approximation is used as

\[
f_3(\tilde{t}) \approx \frac{1}{2} - \frac{1}{2} a_3 a_5 \left(1 - \frac{1}{2} (a_1 + a_6)^2\right) + \frac{1}{2} a_3 a_5 \left(1 - \frac{1}{2} (a_1 - a_6)^2\right) \ldots
\]

\[
+ \frac{a_2 + a_4 a_5}{2} (a_1 + a_6) + \frac{a_2 - a_4 a_5}{2} (a_1 - a_6),
\]

(B.14)

which algebraically reduces to

\[
f_3(\tilde{t}) \approx 1 - \frac{1}{2} a_1^2 - \frac{1}{2} a_6^2 + a_1 a_6 a_3 a_5 + a_1 a_2 + a_6 a_4 a_5.
\]

(B.15)

When substituting the coefficients back into Eq. \(\text{(B.15)}\)

\[
f_3(\tilde{t}) \approx 1 + \left(\frac{3}{8} - \frac{1}{3} \tilde{R}^{-6} \tilde{k}^2 \tilde{t}_i^2\right) \tilde{k}^2 \tilde{t}_i^2 - \frac{1}{3} \tilde{R}^{-6} \tilde{k}^4 \tilde{t}_i^2 \ldots
\]

\[
+ \left(\frac{2}{3} \tilde{R}^{-6} \tilde{k}^2 - \frac{3}{4}\right) \tilde{k}^2 \tilde{t}_i \tilde{t} + We_s^{1/2} \tilde{k} \tilde{t}.
\]

(B.16)

Looking at typical values for \(We_s, \tilde{R}, \tilde{t}, \) and \(\tilde{k}\) present in the experiments there is the following comparative scaling

\[
\frac{1}{3} \tilde{R}^{-6} \tilde{k}^4 \tilde{t}_i^2 \sim 1,
\]

(B.17)

\[
\frac{3}{8} \tilde{k}^2 \tilde{t}_i^2 \sim 10,
\]

(B.18)

\[
\frac{3}{4} \tilde{k}^2 \tilde{t}_i \tilde{t} \sim 10,
\]

(B.19)

\[
We_s^{1/2} \tilde{k} \tilde{t} > 100.
\]

(B.20)

Therefore

\[
f_3(\tilde{t}) \approx We_s^{1/2} \tilde{k} \tilde{t}.
\]

(B.21)
Bibliography


[7] O. A. Hurricane, D. A. Callahan, D. T. Casey, P. M. Celliers,


[38] L. Lásk a, J. Krása, M. Pfeifer, K. Rohlenta, S. Gammino, L. Torrisi, L. Andò, and G. Ciavola. Angular distribution of ions emitted from


[87] I. Yu. Vichev, V. G. Novikov, M. M. Basko, V. V. Ivanov, and V. V. Medvedev. Modeling of target deformations due to pre-pulse with


[105] Sten A. Reijers, Dmitry Kurilovich, Francesco Torretti, Hanneke Gelderblom, and Oscar O. Versolato. Laser-to-droplet alignment sensitivity relevant for laser-produced plasma sources of extreme


[115] Hyoung Won Baac, Jong G. Ok, Adam Maxwell, Kyu-Tae Lee,


