Droplet impact on superheated surfaces

Khavari, Mohammad

2017

Khavari, M. (2017). Droplet impact on superheated surfaces. Doctoral thesis, Nanyang Technological University, Singapore.

<http://hdl.handle.net/10356/72216>

<https://doi.org/10.32657/10356/72216>

NANYANG
TECHNOLOGICAL
UNIVERSITY

DROPLET IMPACT ON SUPERHEATED SURFACES

MOHAMMAD KHAVARI

**SCHOOL OF MECHANICAL AND AEROSPACE
ENGINEERING**

A thesis submitted to the Nanyang Technological University
in partial fulfilment of the requirement for the degree of
Doctor of Philosophy

2017

Abstract

The impact of droplets on heated substrates is a crucial process in a diverse range of technological and industrial applications such as thermal spray, electronics cooling and material processing. Of particular importance is the situation in which the substrate is heated above the liquid boiling point. In such case, hydrodynamic behaviors of the impacting droplets, such as spreading and possible splashing, are accompanied by boiling processes, such as liquid-vapor phase transition, bubble generation, and droplet ejection. These intangible physical processes make it challenging to understand and control quantities of practical interests such as the heat transfer rate from the surface to the liquid. In a typical boiling process, the liquid absorbs heat from the surface, vaporizes, and the generated vapor is carried away by natural convection. The heat flux remains increasing with temperature as long as the liquid maintains contact with the surface; it drops abruptly when excessive vapor completely covers the surface. The former case is commonly referred to as the contact boiling regime, while the latter one is known as the Leidenfrost regime. The transition to the Leidenfrost regime therefore is directly linked to the upper limit of boiling heat transfer. Current theories of the Leidenfrost transition assume a priori existence of the vapor layer, thus focusing on its hydrodynamics without making reference to the contact boiling regime. The main aim of this research is to obtain a physical understanding of the Leidenfrost transition. To this end, we systematically study the boiling phenomena of droplet impinging on superheated substrates made of materials having low and high thermal conductivities, namely glass and sapphire. The difference in thermal conductivity of impacted substrates results in distinct boiling behaviors of the liquids: we observe new regimes, i.e., fingering boiling and oscillating boiling regimes for substrate of low and high thermal conductivities, respectively. In particular, detailed observation and analysis of the oscillating boiling regime allows us to elucidate a new mechanism of the Leidenfrost transition based on competition between two effects: separation of liquid from the heated surface due to localized boiling, and rewetting. We show that the predicted values of the Leidenfrost temperature are consistent with the experimentally measured ones for various liquids having widely different properties, suggesting that complex entanglement between the involving hydrodynamic and thermodynamic processes of the Leidenfrost phenomenon can be understood under the newly proposed mechanism. Our findings offer a new theoretical framework to treat the Leidenfrost transition, a crucial step towards complete control of the Leidenfrost phenomenon.

Acknowledgments

First and foremost, I am extremely grateful to my supervisor, Dr. Tuan Tran, for not only accepting me as his first PhD student at NTU, but also giving me his full advisory and friendly support during my entire PhD. He has had such high standards for research that made me work much harder than I could have ever imagined. I learned so much from him, not only in working professionally in the lab, dealing with technical equipment, performing accurate experiments, supervising FYP students, etc., but also in expressing our scientific results in the simplest possible manner. I am incredibly honored to be his student.

I also want to thank my co-supervisor, Dr. Mohammad S.M. Saifullah (IMRE, A*STAR) for his continuous support during my PhD.

Finally I want to sincerely appreciate the Agency for Science, Technology and Research (A*STAR) for granting me the SINGA (Singapore International Graduate Award) scholarship to pursue my PhD at NTU. Studying in Singapore has been the best experience of my life.

Table of Contents

Abstract.....	ii
Acknowledgments.....	iii
List of Figures.....	vi
List of Tables	xiv
Nomenclature	xv
Chapter 1: Introduction and Motivations.....	1
1.1 Some Historical Notes	2
1.1.1 Droplet Impact	2
1.1.2 Leidenfrost Phenomenon	2
1.2 Applications.....	2
1.3 Motivations and Objectives	3
1.4 Outline of Thesis.....	5
Chapter 2: Literature Review	6
2.1 Droplet Impact on Unheated Solid Surfaces.....	7
2.1.1 Non-Dimensional Groups in Droplet Impact.....	7
2.1.2 Spread Factor	8
2.1.3 Other Experimental Studies	18
2.1.4 Air Layer beneath the Drop	20
2.2 Droplet Impact on Heated Solid Surfaces.....	21
2.2.1 Leidenfrost Temperature.....	21
2.2.2 Features of Leidenfrost Drops	23
2.2.3 Recent Studies.....	31
Chapter 3: Impact on Glass Surface	34
3.1 Experimental Method.....	35
3.2 Experimental Setup.....	37
3.3 Condition of Total Internal Reflection	41
3.4 Experiments on Glass Substrate.....	42
3.5 Image Post-Processing.....	43
3.5.1 Side-View Analysis	43
3.5.2 Bottom-View Analysis	44
3.6 Impact Behavior on Glass Substrate.....	50
3.7 Results of Impact on Glass Substrate	63
3.7.1 Phase Diagram	63

3.7.2 Contact Area	66
3.7.3 Length of Contact Line	67
3.7.4 Rate of Spreading	70
3.7.5 Maximum Values	72
3.7.6 Number of Fingers	75
Chapter 4: Impact on Sapphire Surface	77
4.1 Experimental Method and Setup	78
4.2 Oscillating Behavior: Acetone	80
4.3 Oscillating Behavior: Ethanol	84
4.4 Oscillating Behavior: IPA	89
4.5 Oscillating Behavior: Water	93
4.6 Oscillating Behavior: FC-72	97
4.7 Frequency of Oscillations	98
4.8 Vapor Thickness	100
4.9 Determination of Leidenfrost Temperature	101
Chapter 5: A Theory for Transition to Leidenfrost	103
5.1 Oscillating Boiling: A New Boiling Behavior	104
5.2 Heterogeneous Nucleation	106
5.3 A Proposed Theory for Transition to Leidenfrost	111
5.3.1 Phase Diagrams	112
5.3.2 Dynamic Leidenfrost Temperature	115
5.4 Discussion	118
Chapter 6: Conclusions and Future Work	119
6.1 Conclusions	120
6.2 Future Work	121
Appendices	122
Appendix A. Image Transformation	123
Appendix B. Thermophysical Properties of Liquids	124
References	128

List of Figures

Fig. 2.1 a) The classical pool boiling curve; b) The lifetime (evaporation time) of a sessile drop (taken from Ref. [187]).	22
Fig. 3.1 a) The incident and refracted beams governed by Snell's law; b) Critical angle corresponding to a 90° refracted angle and c) incident angles larger than the critical angle corresponding to total internal reflection.	35
Fig. 3.2 Schematic of experimental setup.	37
Fig. 3.3 Schematic of the laser and beam expander.	38
Fig. 3.4 a) 3D drawing of holder and prism; b) The holder and the prism inside it.	39
Fig. 3.5 A schematic of the experimental setup.	40
Fig. 3.6 The actual experimental setup.	41
Fig. 3.7 a) Snapshots of side-view of a droplet impacting the surface for measuring drop size and impact velocity; b) Detected circles as the droplet in order to find the center and radius of the droplet. The last image shows how the droplet diameter is found. Here the velocity is 1.89 m/s, temperature $T = 60^\circ\text{C}$, drop size 2.1 mm and $We = 258$.	43
Fig. 3.8 Temporal variation of droplet center from which impact velocity as the slope can be found.	44
Fig. 3.9 A sample of an image of wetted area from bottom-view which shows the stretched image because of passing through Dove prism.	44
Fig. 3.10 The cross sign used for calibration of bottom-view. The sizes of cross wings are clearly not equal which shows the image is stretched.	45
Fig. 3.11 a) A recorded bottom-view image before projecting into normal plane; b) The image projected back to the normal plane and after background subtraction. Here the velocity is 0.93 m/s, temperature $T = 20^\circ\text{C}$, drop size 2.1 mm and $We = 64$.	45
Fig. 3.12 a) Original gray scale image; b) Binary image. The velocity is 0.93 m/s, temperature $T = 20^\circ\text{C}$, drop size 2.1 mm and $We = 64$.	46
Fig. 3.13 a) Original image before cropping and b) The image with detected center and selected segment for analysis. The velocity is 1.34 m/s, temperature $T = 40^\circ\text{C}$, drop size 2.1 mm and $We = 132$.	47
Fig. 3.14 Snapshot at two different times, a) $t = 1.3\text{ ms}$ and b) $t = 3.1\text{ ms}$, for distinguishing between true dry regions and interference patterns. The arrows show the interference fringes and dry areas (outside the wetted region) are also specified. The velocity is 2.85 m/s, temperature $T = 20^\circ\text{C}$, drop size 2.03 mm and $We = 584$.	48
Fig. 3.15 Snapshot for distinguishing between true dry regions and interference patterns at a higher temperature. The arrows show the interference fringes and dry areas (due to bubble nucleation). The velocity is 1.87 m/s, temperature $T = 180^\circ\text{C}$, drop size 2.04 mm and $We = 253$.	49
Fig. 3.16 Snapshot for the entire process of image processing. a) Original gray scale image; b) Image checked for interference patterns and modified to be treated as	

wetted are; c) Binary format of the original image; d) Binary format of the modified image; e) Cropped binary image ready to calculate the contact area and f) The detected contact length from the binary image used for calculating the perimeter of the wetted region. The velocity is 1.34 m/s, temperature $T = 40\text{ }^{\circ}\text{C}$, drop size 2.1 mm and $We = 132$50

Fig. 3.17 Snapshot for a case of striking fingering patterns observed via TIR. a) Gray scale image; b) Binary image and c) Detected perimeter of the binary image. These photos show qualitatively how the wetted area and length are measured. The velocity is 2.58 m/s, temperature $T = 200\text{ }^{\circ}\text{C}$, drop size 2.1 mm and $We = 484.50$

Fig. 3.18 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 65$; b) $We = 131$; c) $We = 259$; d) $We = 371$; e) $We = 488$ and f) $We = 584$. Here the temperature $T = 20\text{ }^{\circ}\text{C}$ is the lowest temperature tested and the drop size for all cases is $D0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Images in the same column were taken at the same time after impact.52

Fig. 3.19 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 65$; b) $We = 131$; c) $We = 259$; d) $We = 385$; e) $We = 478$ and f) $We = 598$. Here the temperature is $T = 80\text{ }^{\circ}\text{C}$ and the drop size for all cases is $D0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Images in the same column were taken at the same time after impact.53

Fig. 3.20 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 62$; b) $We = 129$; c) $We = 253$; d) $We = 376$; e) $We = 485$ and f) $We = 593$. Here the temperature is $T = 180\text{ }^{\circ}\text{C}$ and the drop size for all cases is $D0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Images in the same column were taken at the same time after impact.54

Fig. 3.21 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 63$; b) $We = 128$; c) $We = 252$; d) $We = 364$; e) $We = 484$ and f) $We = 582$. Here the temperature is $T = 200\text{ }^{\circ}\text{C}$ and the drop size for all cases is $D0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Images in the same column were taken at the same time after impact.55

Fig. 3.22 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 63$; b) $We = 132$; c) $We = 254$; d) $We = 369$; e) $We = 478$ and f) $We = 595$. Here the temperature is $T = 240\text{ }^{\circ}\text{C}$ and the drop size for all cases is $D0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Images in the same column were taken at the same time after impact.56

Fig. 3.23 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 280\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 63$ ($H = 5\text{ cm}$) and the drop size for all cases is $D0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Note the Leidenfrost regime (no wetted area) for case (f) at $T = 280\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.57

- Fig. 3.24 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 280\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 130$ ($H = 10\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 280\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.58
- Fig. 3.25 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 254$ ($H = 20\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.59
- Fig. 3.26 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 371$ ($H = 30\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.60
- Fig. 3.27 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 484$ ($H = 40\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.61
- Fig. 3.28 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 584$ ($H = 50\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.62
- Fig. 3.29 Phase diagram showing four major boiling regimes: spreading (blue squares), bubbly boiling (red circles), fingering boiling (purple asterisks) and Leidenfrost (green stars). Several points with transitional behaviors (right triangles) were observed between the Leidenfrost and the fingering boiling regimes: the wetting patterns only appear for a very short time ($\approx 1\text{ ms}$) and do not have the “fingering” shapes. The dashed line indicates the transition between the fingering boiling and the Leidenfrost regimes.64
- Fig. 3.30 Qualitative results of spreading of droplet upon impact on glass prism surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 484$ ($H = 40\text{ cm}$) and the drop size for all

cases is $D0 = 2.05 \pm 0.05$ mm. All the inset scale bars represent the length scale of 2 mm. Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300$ °C....	64
Fig. 3.31 Temporal variation of contact area for different We and temperatures: a) $T = 20$ °C; b) $T = 180$ °C; c) $T = 200$ °C; d) $T = 260$ °C.....	66
Fig. 3.32 Temporal variation of contact area for different We and temperatures: a) We = 64; b) We = 130; c) We = 255; d) We = 372; e) We = 481 and f) We = 591.	68
Fig. 3.33 Temporal variation of contact length for different We and temperatures: a) $T = 20$ °C; b) $T = 180$ °C; c) $T = 200$ °C and d) $T = 260$ °C.....	69
Fig. 3.34 Temporal variation of contact length for different We and temperatures: a) We = 64; b) We = 130; c) We = 255; d) We = 372; e) We = 481 and f) We = 591.	70
Fig. 3.35 Calculation of the rate of spreading (slope of linear section of contact area) obtained for $T = 40$ °C and We = 65. a) The original contact area; b) Fitted line to the data in $0 < t < 1$ ms; c) Fitted line to the data in $0 < t < 1.5$ ms and d) Fitted line to the data in $0 < t < 2$ ms. The equation of the fitted line is also shown as inset in each plot. The final slope for this case is the average of the slope of these three fitted lines.....	71
Fig. 3.36 Variation of rate of spreading with temperature for different Weber numbers.	72
Fig. 3.37 Variation of equivalent spreading factor with temperature for different We. Inset: log-log plot of ξe vs. We for spreading and bubbly boiling regimes. The solid line indicates the best fitted line for the empirical data whose slope is 0.16.....	73
Fig. 3.38 Variation of normalized contact length with temperature for different We. Inset: log-log plot of L vs. We for spreading and bubbly boiling regimes. The solid line indicates the best fitted line for the empirical data whose slope is 2.7.....	74
Fig. 3.39 Variation of the number fingers with We for different temperatures. Inset: log-log plot of N vs. We. The solid line indicates the best fitted line for the empirical data whose slope is 0.5.....	76
Fig. 4.1 a) Simplified experimental setup for impact on a heated sapphire prism; b) Snapshot of droplet from side-view recordings before reaching the heated substrate; c) Snapshot of droplet from side-view recordings after contacting with the heated substrate and d) A typical snapshot from bottom-view recordings via TIR technique which distinguishes between dry (bright) and wetted (black) areas. All scale bars indicate the length scale of 1 mm.	78
Fig. 4.2 The oscillating behavior observed during impact of an acetone droplet on a heated sapphire prism. All inset numbers show the real time in μ s. Surface temperature is $T = 140$ °C and impact velocity is $U0 = 1.85$ m/s. Scale bar indicates the length scale of 1 mm.....	80
Fig. 4.3 Series of snapshots showing the oscillating behavior of acetone. All inset numbers show the real time in μ s. Surface temperature is $T = 150$ °C and impact velocity is $U0 = 1.85$ m/s. Scale bar indicates the length scale of 1 mm.	81
Fig. 4.4 Series of snapshots showing the oscillating behavior of acetone. All inset numbers show the real time in μ s. Surface temperature is $T = 160$ °C and impact velocity is $U0 = 1.85$ m/s. Scale bar indicates the length scale of 1 mm.....	81

- Fig. 4.5 Comparison of the rewetting process of acetone for different surface temperatures and same impact velocity ($U_0 = 1.85$ m/s). Each column shows successive snapshots for the same temperature. Scale bars indicate the length scale of 1 mm. The rewetting process is observed as the three-phase contact line moves towards the center, but tends to occur more irregularly as T is increased.83
- Fig. 4.6 Comparison of oscillating behavior of acetone for same surface temperature ($T = 140$ °C) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bars indicate the length scale of 1 mm.....84
- Fig. 4.7 Series of snapshots showing the oscillating behavior of ethanol. All inset numbers show the real time in μ s. Surface temperature is $T = 150$ °C and impact velocity is $U_0 = 2.66$ m/s. Scale bar indicates the length scale of 1 mm.85
- Fig. 4.8 Series of snapshots showing the oscillating behavior of ethanol. All inset numbers show the real time in μ s. Surface temperature is $T = 160$ °C and impact velocity is $U_0 = 2.66$ m/s. Scale bar indicates the length scale of 1 mm.85
- Fig. 4.9 Series of snapshots showing the oscillating behavior of ethanol. All inset numbers show the real time in μ s. Surface temperature is $T = 170$ °C and impact velocity is $U_0 = 2.66$ m/s. Scale bar indicates the length scale of 1 mm.86
- Fig. 4.10 Comparison of the rewetting process of ethanol for different surface temperatures and same impact velocity ($U_0 = 2.66$ m/s). Each column shows successive snapshots for the same temperature. Scale bar indicates the length scale of 1 mm.87
- Fig. 4.11 Comparison of oscillating behavior of ethanol for same surface temperature ($T = 150$ °C) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bar indicates the length scale of 1 mm.....88
- Fig. 4.12 Series of snapshots showing the oscillating behavior of IPA. All inset numbers show the real time in μ s. Surface temperature is $T = 150$ °C and impact velocity is $U_0 = 2.58$ m/s. Scale bar indicates the length scale of 1 mm.89
- Fig. 4.13 Series of snapshots showing the oscillating behavior of IPA. All inset numbers show the real time in μ s. Surface temperature is $T = 160$ °C and impact velocity is $U_0 = 2.58$ m/s. Scale bar indicates the length scale of 1 mm.89
- Fig. 4.14 Series of snapshots showing the oscillating behavior of IPA. All inset numbers show the real time in μ s. Surface temperature is $T = 170$ °C and impact velocity is $U_0 = 2.58$ m/s. Scale bar indicates the length scale of 1 mm.90
- Fig. 4.15 Comparison of the rewetting process of IPA for different surface temperatures and same impact velocity ($U_0 = 2.58$ m/s). Each column shows successive snapshots for the same temperature. Scale bar indicates the length scale of 1 mm.91
- Fig. 4.16 Comparison of oscillating behavior of IPA for same surface temperature ($T = 160$ °C) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bar indicates the length scale of 1 mm.....92

Fig. 4.17 Series of snapshots showing the oscillating behavior of water. All inset numbers show the real time in μs . Surface temperature is $T = 260\text{ }^{\circ}\text{C}$ and impact velocity is $U_0 = 2.78\text{ m/s}$. Scale bar indicates the length scale of 1 mm.	93
Fig. 4.18 Series of snapshots showing the oscillating behavior of water. All inset numbers show the real time in μs . Surface temperature is $T = 280\text{ }^{\circ}\text{C}$ and impact velocity is $U_0 = 2.78\text{ m/s}$. Scale bar indicates the length scale of 1 mm.	94
Fig. 4.19 Series of snapshots showing the oscillating behavior of water. All inset numbers show the real time in μs . Surface temperature is $T = 300\text{ }^{\circ}\text{C}$ and impact velocity is $U_0 = 2.78\text{ m/s}$. Scale bar indicates the length scale of 1 mm.	94
Fig. 4.20 Comparison of the rewetting process of water for different surface temperatures and same impact velocity ($U_0 = 2.78\text{ m/s}$). Each column shows successive snapshots for the same temperature. Scale bar indicates the length scale of 1 mm.	95
Fig. 4.21 Comparison of oscillating behavior of water for same surface temperature ($T = 260\text{ }^{\circ}\text{C}$) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bar indicates the length scale of 1 mm.	96
Fig. 4.22 Series of snapshots showing the oscillating behavior of FC-72. All inset numbers show the real time in μs . Surface temperature is $T = 100\text{ }^{\circ}\text{C}$ and impact velocity is $U_0 = 2.59\text{ m/s}$. Scale bar indicates the length scale of 1 mm.	97
Fig. 4.23 Fluctuations of image intensity at the center point vs. time for a typical case. Liquid is water, impact velocity is $U_0 = 1.88\text{ m/s}$ and surface temperature is $= 260\text{ }^{\circ}\text{C}$	98
Fig. 4.24 Frequency of oscillations vs. impact velocity for different surface temperatures: a) acetone; b) ethanol; c) IPA and d) water. It can be seen that the frequency is almost independent of impact velocity.	99
Fig. 4.25 Dependence of averaged frequency of oscillations on surface temperature for all liquids.	100
Fig. 4.26 Total number of the wetted points vs. time at two surface temperatures. a) At $T = 170\text{ }^{\circ}\text{C}$, since $Nt \neq 0$, there is still contact between liquid and solid and b) At higher temperature ($T = 190\text{ }^{\circ}\text{C}$), there is no wetted point on the substrate during the entire impact time, therefore the system is in Leidenfrost regime. Insets show typical snapshots of the impact. Liquid is acetone and impact velocity is $U_0 = 2.59\text{ m/s}$. Scale bars indicate the length scale of 1 mm.	102
Fig. 5.1 Wetted area in the oscillating boiling regime. The liquid is acetone, impact velocity is $U_0 = 2.74\text{ m/s}$ and surface temperature is $T = 150\text{ }^{\circ}\text{C}$. Snapshots showing a) The boiling process and b) The rewetting process; c) Wetted area A_{wvs} . t in the early stage of a droplet impacting on a heated sapphire surface and d) Wetted area A_w vs. t during the entire impact process; the shaded area corresponds to the wetted area shown in (c). Inset snapshots show side-view (upper panel) and bottom-view (lower panel) of the impact at different times. All scale bars indicate the length scale of 1mm.	105
Fig. 5.2 Comparison of the variation rate of generation of embryos J (from Eq. 5.5) and with surface temperature T for acetone, ethanol and IPA. The corresponding plot for water is shown as inset due to its higher range of surface temperature. ...	107

- Fig. 5.3 Heterogeneous boiling and rewetting processes. a) Vapor embryos in a heterogeneous boiling process; b) Critical size rc of a vapor embryo (blue solid line) and evaporation time τh (red dash line) versus surface temperature T . Inset: schematic of a vapor embryo on a flat surface surrounded by liquid and c) Snapshots showing the rewetting process for acetone droplets impacting with velocity $U0 = 2.3$ m/s at low temperature (left panel) and high temperature (right panel); arrows indicate the motion of the three phase contact line. Scale bars show the length scale of 0.3 mm. 108
- Fig. 5.4 a) Rewetting velocity V_{re} versus surface temperature T for acetone droplets for different impact velocities; solid line shows the prediction according to Eq. 5.15 and b) V_{re} vs. T for different impact velocities for ethanol, IPA, and water. 110
- Fig. 5.5 Comparison of the variation of vapor embryo's critical size rc (from Eq. 5.4) with surface temperature T for acetone, ethanol and IPA. The corresponding plot for water is shown as inset due to its higher range of surface temperature. 110
- Fig. 5.6 Variation of evaporation time τh (from Eq. 5.14) with surface temperature T for a) acetone; b) ethanol; c) IPA and d) water. 111
- Fig. 5.7 Phase diagram of acetone based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes three predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16, the one measured from TIR recordings (red dashed line) and the one detected from side-view images (blue dashed-dotted line). 112
- Fig. 5.8 Phase diagram of ethanol based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes three predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16, the one measured from TIR recordings (red dashed line) and the one detected from side-view images (blue dashed-dotted line). 113
- Fig. 5.9 Phase diagram of IPA based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes three predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16, the one measured from TIR recordings (red dashed line) and the one detected from side-view images (blue dashed-dotted line). 114
- Fig. 5.10 Phase diagram of water based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes two predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16 and the one measured from TIR recordings (red dashed line). Note that for water, we were not able to detect the Leidenfrost transition from side-view recordings. 114
- Fig. 5.11 Predicted Leidenfrost temperature (Eq. 5.16) vs. impact velocity for the four tested liquids in this study: acetone (dashed line), ethanol (solid line), IPA (dotted line) and water (dash-dot line). 115

Fig. 5.12 Comparison between the dynamic Leidenfrost temperatures obtained from theory and experiments for various liquids: acetone (square), ethanol (circle), IPA (diamond), water (pentagram star), HFE7100 (right triangle), and heptane (hexagram star). The empty markers on the diagonal solid lines indicate theoretical values of TL for different liquids. All upward solid triangles mark the measured Leidenfrost temperature from TIR, while the downward solid ones mark those measured from side-view recordings. All other solid markers show the dynamic Leidenfrost temperature experimentally determined in other studies [24, 76, 260, 272, 273, 293, 339]. The shaded area indicates $\pm 15\%$ deviation from the theoretical values.....117

List of Tables

Table 3.1 Refractive index of the required materials [312].	37
Table 3.2 Values of penetration depth for red light ($\lambda_0 = 640$ nm) and different incident angles.	37
Table 3.3 Physical properties of ethanol at room temperature [317].	42
Table 4.1 Comparison of physical properties of N-BK7 glass [326] and sapphire [327] at $T = 20$ °C.	79
Table 4.2 Physical properties of the liquids at $T = 20$ °C and $P_0 = 101$ kPa.	79
Table 4.3 Experimental conditions of the entire tests for impact on sapphire prism.	79
Table 4.4 Incident angle and penetration depth for all the liquids studied in this chapter.	100
Table 5.1 Predicted Leidenfrost temperature from our theory (Eq. 5.16) for different liquids. The shaded rows show the ones for which the experimental values of dynamic Leidenfrost temperature were available in the literature.	116

Nomenclature

a	Capillary length
A	Contact area
Bo	Bond number
Ca	Capillary number
C_p	Specific heat at constant pressure
D_0	Droplet diameter before impact
D_{max}	Maximum diameter of droplet
d_p	Penetration depth
f	Frequency
F	Correction factor
Fr	Froude number
g	Gravity
h	Thickness
h_{fg}	Latent heat
h_v	Vapor thickness
I	Image intensity
J	Generation rate of vapor embryos per unit time per unit area
k	Thermal conductivity
k_B	Boltzmann constant
L	Contact length
m	Mass of one molecule
$\bar{M}$	Molecular mass
n	Index of refraction
N_A	Avogadro constant
N_b	Density of bubble nucleation
Oh	Ohnesorge number
P	Pressure
P_l	Liquid pressure
P_{sat}	Saturation pressure
P_v	Vapor pressure
R	Droplet radius
R	Specific gas constant

$\bar{R}$	Universal gas constant
r_c	Critical radius of vapor embryo
Re	Reynolds number
S	Spreading rate
St	Stokes number
t	Time
T	Surface temperature
T_b	Boiling temperature
T_L	Leidenfrost temperature
T_L^M	Measured Leidenfrost temperature
T_L^P	Predicted Leidenfrost temperature
U_0	Impact velocity
V	Volume
V_{re}	Rewetting velocity
We	Weber number

Greek Letters

α	Incident angle
α_c	Critical angle
κ	Curvature
μ	Dynamic viscosity
ν	Kinematic viscosity
ρ	Density
σ	Surface tension
τ_d	Droplet lifetime
τ_h	Duration to populate a unit area by vapor embryo
θ	Contact angle

Chapter 1:

Introduction and Motivations

1.1 Some Historical Notes

1.1.1 Droplet Impact

The impact of liquid droplets on solid surfaces is ubiquitous in everyday life: water drops dripping from a kitchen faucet onto a sink, raindrops falling on windshields, paint drops from spray gun. Although the impact process appears simple, it is an inherently complex physical problem that dates back to more than 100 years ago [1]. Accompanied by its numerous industrial applications, this problem has been attracting extensive research efforts from scientists and engineers of diverse disciplines.

Worthington was possibly one of the first who carried out a systematic study of the droplet impact phenomenon [1-5]. With simple setups using strobe light, he studied the impact for different liquid droplets, e.g., water, mercury, alcohol, and different impact velocities on smoked glass surface. He observed various behaviors of the droplets upon impact such as fingers formation at the spreading front, and splashing [1].

With the advancements in photographic technologies, in particular the rapid development of high-speed cameras and image processing techniques, physical insights into the impact phenomenon has been becoming more accessible to researchers. The field of droplet impact currently is an active one, with numerous papers published in impactful journals such as *Nature* [6-16] and *Physical Review Letters* (PRL) [17-43]. Many review papers in the field [44-54] demonstrate that there are still various fundamental questions left unanswered.

1.1.2 Leidenfrost Phenomenon

Water droplet floats on a glowing hot iron spoon. This peculiar phenomenon still remains elusive despite that its first systematic study dates back to the middle of the eighteenth-century [55]. For a droplet in this so-called *Leidenfrost regime*, the temperature of the heated surface must be higher than a critical value, the Leidenfrost temperature, to generate a thin vapor layer underneath the droplet to sustain its weight. The vapor layer both separates and insulates the droplet from the heated surface [56, 57], thus keeping it from being instantly vaporized. At lower surface temperatures, insufficient vapor generation causes the liquid to wet the surface, and the droplet is said to be in the *contact boiling regime*.

1.2 Applications

As mentioned above, one of the main reasons for extensive studies of the droplet impact phenomenon is its crucial role in many industrial and technological applications. These include:

Liquid atomization [58], inkjet printers [59, 60], sprinklers for fire suppression [47, 61, 62], thermal spray [35, 63-69], spray painting [47], spray cooling [70], blood drop spreading in forensic investigations [71], electrowetting [45, 72], accumulation of ice on wires and airplanes [47, 61], spray cooling of electronic devices [73, 74] and semiconductors chips, gas turbine blades [75], incinerators [47, 76-78], cooling in metallurgical processes [47, 76, 79], solder bumps on printed circuit boards [47, 61, 80, 81], quenching processes [47, 82], food applications [83, 84], internal combustion engines [85-88], PolyJet additive manufacturing (3D printing) [89], soil erosion [49, 90], and pesticide deposition [47, 49, 91, 92].

Since its discovery, the Leidenfrost phenomenon and related processes have been an integral part of numerous applications with diverse requirements. On the one hand, for those involving heat transfer from a heated surface to the surrounding liquid, it sets the upper limits for both the surface's operating temperature and the heat transfer rate. Such limitations come from the poor thermal conductivity of vapor: as soon as the Leidenfrost phenomenon sets in, a vapor layer is formed and considerably inhibits the heat transfer rate from the surface causing its temperature to quickly reach the damaging point. Thus, in such applications, high Leidenfrost temperature is preferred. On the other hand, in applications aiming at eliminating solid-liquid contact such as drag reduction [31] and fluidic control [13, 93], low Leidenfrost temperature is required. Nonetheless, optimization of these widely diverse applications necessitates an accurate prediction and efficient ways to control the occurrence of the Leidenfrost phenomenon.

1.3 Motivations and Objectives

The impact of droplets on solid surfaces remains an active research area with various long standing questions. One of the challenges in obtaining a physical understanding of the phenomenon originates from a large number of involving parameters including droplet size, impact velocity, liquid properties, e.g., density, viscosity, and surface tension, as well as surface properties, e.g., roughness and wettability. Variation in surface temperature further complicates the problem due to possible liquid-vapor phase transition that occurs upon impact. Nonetheless, the impact phenomenon involving heated surfaces has been extensively investigated due to its vital role in diverse industrial and technological applications, in particular to those utilizing impacting droplets for heat removal. Various correlations have been proposed to both predict and enhance the heat removal rate. Although these correlations have been applied with success in several isolated cases, a physical understanding of the entangled hydro- and thermo-dynamical processes during impact and the resulting heat transfer is still missing.

In classical boiling studies, the heat transfer between a heated surface and the surrounding liquid is a function of both surface temperature and contact area. The heat transfer rate reaches a maximum at a critical temperature, which marks the end of the

nucleate boiling regime, and drops to a minimum values at the so-called Leidenfrost temperature. At temperature higher than the Leidenfrost temperature, a vapor film is formed and completely separates the liquid from the surface. Thus, the Leidenfrost temperature is the boundary between the contact boiling regime, where there is direct contact between the liquid and the solid surface, and the film boiling (Leidenfrost) regime, where the vapor film completely eliminates the liquid-surface contact. The heat removed from the heated surface is therefore considerably diminished due to the low thermal conductivity of vapor.

The contact area between the liquid and the heated surface therefore is a key factor that affects the heat transfer. It strongly depends on formation and growth of vapor bubbles at the solid-liquid interface. It is however particularly challenging to obtain experimental measurements and detailed analysis of the contact area because it is optically inaccessible and fluctuates with high frequency as a result of the extremely fast dynamics of the vapor bubbles. In order to understand the physical mechanism of the heat transfer process, as well as the transition from the contact boiling to the Leidenfrost regimes, it is imperative to obtain the exact values of the contact area between the impinging droplet and the surface.

In this research project, we investigate the impact of liquid droplets on superheated surfaces with the aim to obtain detailed measurements of the contact area using the Total Internal Reflection (TIR) technique. By combining high-speed imaging with the TIR technique, we systematically obtain temporally and spatially resolved measurements of the contact area for varying surface temperature, substrates, and liquid properties. These measurements considerably enhance our insight into the impact and boiling processes compared to those obtained by the conventional side-view high-speed imaging as they contain information of the moving contact lines caused by vapor bubbles generated spontaneously during impact.

The transition to the Leidenfrost regime is another key factor that drastically influences the heat transfer from the heated surfaces. This transition often sets in abruptly as a result of a sudden formation of a vapor layer separating the liquid and the heated surfaces. Due to vapor's poor thermal conductivity, the heat transfer in the Leidenfrost regime is substantially diminished causing the surface temperature to increase rapidly to a damaging point (burn-out). Thus a study of the Leidenfrost transition is inseparable to that of the heat transfer in the contact boiling regime. We aim to obtain a physical understanding of the Leidenfrost transition by experimentally and analytically investigating the boiling phenomenon of the liquid at temperatures close to the Leidenfrost regime.

1.4 Outline of Thesis

In chapter 2, we cover the literature review for droplet impact on unheated and superheated surfaces, as well as studies dedicated to the Leidenfrost phenomenon. We focus on the most recent and important studies for these topics.

In chapter 3, we discuss details of the first part of this research; impact of ethanol droplets on heated prisms made of *glass*, which is a material with low thermal conductivity. The chapter includes a full description of the experimental setup; the post-processing procedure for analyzing images obtained from high-speed recording, and qualitative results of the impact behaviors.

In chapter 4, we present experimental measurements for the impact of droplets of different liquids (acetone, ethanol, IPA and water) on heated prisms made of *sapphire*, which is a material with high thermal conductivity and therefore is more relevant to practical situations. The chapter includes a short description of the experimental setup; qualitative description of a new boiling regime, called oscillating boiling regime, for different impact velocities and surface temperatures and different liquids. We provide detailed analysis of the experimental results including quantities that characterize the oscillating boiling regime such as the frequency of oscillation of the contact area and vapor thickness for different liquids.

In chapter 5, we propose a new theory for the Leidenfrost transition of droplets impacting superheated surfaces at high velocity based on our observations provided in the previous chapter and experimental validations of the theory.

Finally, in chapter 6, we present conclusions drawn from this research and several remarks for future work.

Chapter 2:

Literature Review

In this chapter, a review of the most important studies on droplet impact is presented. Because of the huge number of the published works and the vastness of this field, it is a bit challenging to include all the related works. Therefore, the whole problem of droplet impact is divided into several sections in order to give a comprehensive review of the published papers. First, the problem of droplet impact on unheated solid surfaces is considered in which the focus is mainly on the correlations, physical concepts and pioneer works on this field. Next, the case of droplet impact on heated surfaces which might lead to Leidenfrost regime will be explained and most important works will be mentioned. At the end, we will also review the most recent works.

2.1 Droplet Impact on Unheated Solid Surfaces

Droplet impact has been studied extensively in the literature throughout history and even recently, there have been many papers still discussing the fundamental physics behind the problem [71]. Rein (1993) [49] and Yarin (2006) [47] have reviewed the important works on droplet impact on both (unheated) solid and liquid surfaces and studied different behaviors of spreading, splashing, deposition, receding, etc. Yarin's work had mostly focused on droplet impact on wetted surface and the transition to splashing as well as splashing threshold.

2.1.1 Non-Dimensional Groups in Droplet Impact

The most useful non-dimensional groups encountered in the study of droplet impact on solid surfaces used throughout this report are:

$$We = \frac{\rho U_0^2 D_0}{\sigma}, \quad \text{Weber Number} \quad 2.1$$

$$Re = \frac{\rho U_0 D_0}{\mu}, \quad \text{Reynolds Number} \quad 2.2$$

$$Ca = \frac{\mu U_0}{\sigma} = \frac{We}{Re}, \quad \text{Capillary Number} \quad 2.3$$

$$Oh = \frac{\mu}{\sqrt{\rho D_0 \sigma}} = \frac{\sqrt{We}}{Re}, \quad \text{Ohnesorge Number} \quad 2.4$$

$$Bo = \frac{\rho g D_0^2}{\sigma} = \frac{D_0^2}{a^2}, \quad \text{Bond Number} \quad 2.5$$

$$Fr = \frac{U_0^2}{g D_0} = \frac{We}{Bo}, \quad \text{Froude Number} \quad 2.6$$

$$St = \frac{\rho U_0 D_0}{\mu_g}, \quad \text{Stokes Number} \quad 2.7$$

$$a = \sqrt{\frac{\sigma}{\rho g}}. \quad \text{Capillary Length} \quad 2.8$$

The parameters used in the above equations are: D_0 is droplet diameter just before impact, U_0 is impact velocity and g is the gravitational acceleration. The liquid properties include density (ρ), viscosity (μ) and surface tension (σ). μ_g is viscosity of the surrounding gas. The capillary characteristic length a is obtained by comparing the Laplace pressure and hydrostatic pressure in a liquid. Thus, each liquid would have its distinct a value. If droplet radius, $R_0 < a$, surface tension dominates the gravity (capillary regime), but if $R_0 > a$, (for example in the case of droplet deposited on a glass surface) gravity tends to flatten the droplet and a puddle shape will form [94].

Most importantly, We number is the ratio of liquid inertia to its surface tension; Re is the ratio of liquid inertia to the viscous force, Ca is the ratio of viscous force to surface tension and St is the ratio of liquid inertia to surrounding gas (air) viscous force which will appear in study of air layer generated beneath the drop upon impact on the surface [23].

2.1.2 Spread Factor

One of the most important objectives in studies regarding droplet impact is to find the maximum diameter of an impacting droplet on a solid surface (D_{max}). This quantity is also known as maximal extension or maximal deformation [95]. An equivalent parameter is defined as the ratio of maximum deformation to the initial drop diameter, $\xi_{max} = D_{max}/D_0$, which is also called spread factor or degree of flattening [82]. Various empirical and theoretical models for ξ_{max} have been presented in the literature in the past 40 years. The fact that even recently [71, 96], new correlations for the very same parameter (ξ_{max}) are suggested, shows the complexity of this phenomenon. Bennett and Poulikakos [82] reviewed some of the models proposed for spread factor which will be described here shortly along with some other models.

The main discussion in derivation of the models of spreading is how the kinetic energy of impacting drop is treated. Most of the models are based on whether the kinetic energy is transferred to surface energy or viscous energy. Jones [97] gave one of the first models in which he ignored the effect of surface tension in terminating the spreading and gave the following relation for spread factor, based solely on viscous dissipation as the main source of spreading termination [82]:

$$\xi_{max} = \sqrt{\frac{4}{3}} \text{Re}^{1/8}. \quad 2.9$$

But the experimental results for ξ_{max} were much larger, around 5 times, than the above prediction [82, 97, 98].

Conversely, Collings *et al.* [99] neglected the viscous dissipation effect in energy loss and by considering surface energy of the spreading droplet, while ignoring droplet's initial surface energy (which is not true and is indeed 25% of the total surface energy [82]) and assuming the equilibrium contact angle to be $\theta_e = \pi$, obtained the following relation for spread factor:

$$\xi_{max} = \sqrt{\frac{1}{6}} We^{1/2}. \quad 2.10$$

The experimental data does not verify above relation either, but it gives the simple scaling of spread factor for the case of energy transfer by surface tension: $\xi_{max} \sim We^{1/2}$. Although for so many years, it has been believed that this scaling does not fully capture the observed behavior for the maximum extension of the droplet [95], recently Laan *et al.* [71] showed that this scaling is applicable for the capillary regime.

Chandra and Avedisian [76] in a highly referred paper, gave a model for ξ_{max} based on both surface and viscous force (by order-of-magnitude) analysis as responsible for energy loss. They offered the following correlation [76, 82]:

$$\frac{4.5\xi_{max}^4}{Re} + \frac{3[(1 - \cos \theta_e)\xi_{max}^2 - 4]}{We} = 1, \quad 2.11$$

which overpredicted the empirical data by 20-40% [76, 82]. Bennett and Poulikakos attributed this to the suggested term for the viscous loss.

Madejski [100] considered both viscous and surface energy effects as well as solidification of splat theoretically and gave this correlation in the case of no solidification for $Re > 100$ and $We > 100$:

$$\frac{3\xi_{max}^2}{We} + \frac{1}{Re} \left(\frac{\xi_{max}}{1.2941} \right)^5 = 1. \quad 2.12$$

For the special cases, if the surface tension is ignored:

$$\xi_{max} = 1.2941(Re + 0.9517)^{1/5}, \quad 2.13$$

which for $Re > 100$ will be simplified to:

$$\xi_{max} = 1.2941Re^{1/5}, \quad 2.14$$

On the other hand, if the viscous dissipation is ignored:

$$\xi_{max} = \sqrt{\frac{1}{3}} We^{1/2}, \quad 2.15$$

which is similar to that of Collings *et al.*, Eq. 2.10 except that here $\theta_e = \pi/2$. Madejski's model was seen to have good agreement with experiments of McPherson for very high We numbers [98].

After reviewing the above models, Bennett and Poulikakos [82] attempted to select the best of the them and finally came with a modified model by combining the effect of viscous energy dissipation and modifying the treatment of surface energy by including initial surface tension of the droplet (for $Re > 100$):

$$\frac{\left(\frac{\xi_{max}}{1.2941} \right)^5}{Re} + \frac{3[(1 - \cos \theta_e)\xi_{max}^2 - 4]}{We} = 1. \quad 2.16$$

Like Chandra and Avedisian [76], Bennett and Poulikakos [82] emphasized the effect of choosing the correct contact angle in above relations. For $\theta_e = \pi/2$, they simplified Eq. 2.16 and by calculating two correction factors for each of the limiting cases (ignoring either surface energy or viscous energy), tried to find the transition between the two regimes. They found that if the spreading behavior is assumed to be dominated by only surface energy or viscous energy (i.e., ignoring the other), the error of this treatment will be less than 20%. They also claimed that the effect of surface tension in viscous domain is still important, while the effect of viscous dissipation is negligible in surface tension domain. They stated that if the following condition is satisfied, the surface tension is responsible for spreading to stop:

$$We < 2.80Re^{0.457}. \quad 2.17$$

Delplanque and Rangel [101] modified Madejski's model by incorporating a velocity distribution for spreading of droplet (by Markworth and Saunders [102]) to improve the accuracy of the model and proposed the following relation for the viscous regime (while their relation for capillary regime was the same as Madejski (Eq. 2.15)):

$$\xi_{max} = 1.1626Re^{1/5}, \quad Re > 140 \quad 2.18$$

and thus for the general case:

$$\frac{3\xi_{max}^2}{We} + \frac{1}{Re} \left(\frac{\xi_{max}}{1.1626} \right)^5 = 1. \quad \begin{matrix} Re > 140, \\ We > 670 \end{matrix} \quad 2.19$$

Zhang [103] developed the models of Madejski [100] and Delplanque and Rangel [101] by including the wetting effects through a parameter m_1 and arrived at the following relation:

$$\frac{3(1 - m_1)(\xi_{max}^2 - 1)}{We} + \frac{1}{Re} \left(\frac{\xi_{max}}{1.18} \right)^5 = 1, \quad 2.20$$

where $m_1 = \cos \theta_{eq}$ is the wetting coefficient and θ_{eq} is the equilibrium contact angle. Therefore, for the capillary and viscous regimes, it would respectively simplify to:

$$\xi_{max} = 1.18Re^{1/5}, \quad 2.21$$

$$\xi_{max} = \sqrt{\frac{We}{3(1 - m_1)}} + 1. \quad 2.22$$

Some other studies also gave similar results for the viscous regime which mostly differ in the numerical coefficient and most of them compare their results with that of Madejski [100]. Chronologically, they are as follows:

Hamatani *et al.* [104]:

$$\xi_{max} = 0.83Re^{1/5}, \quad 2.23$$

Trapaga and Szekely [105]:

$$\xi_{max} = 1.0\text{Re}^{1/5}, \quad 2.24$$

Fukanuma [106]:

$$\xi_{max} = 1.234\text{Re}^{1/5}, \quad 2.25$$

Watanabe *et al.* [107]:

$$\xi_{max} = 0.82\text{Re}^{1/5}, \quad 2.26$$

Yoshida *et al.* [108]:

$$\xi_{max} = 0.83\text{Re}^{0.21}, \quad 2.27$$

Bertagnolli *et al.* [109]:

$$\xi_{max} = 0.925\text{Re}^{1/5}, \quad 2.28$$

Liu *et al.* [110]

$$\xi_{max} = 1.04\text{Re}^{1/5}. \quad 2.29$$

Scheller and Bousfield [111] carried out many experiments of droplet impact with liquids of different viscosities, focusing primarily on viscous liquids (up to 300 mPa.s), as well as different surfaces. They found that the increase of D_{max} with U_0 is more profound for low viscous liquids than high viscous ones. Also they found the following correlation for the spread factor based on their experimental results which is stated in terms of Ohnesorge number, since it only shows the liquid properties:

$$\xi_{max} = 0.61(\text{Re}^2\text{Oh})^{0.166} = 0.61\left(\frac{\text{We}}{\text{Oh}}\right)^{0.166} \quad 2.30$$

Furthermore, they also offered an analytical model based on squeezing flow model (modeling drop spreading as a cylinder, with a volume double that of the droplet, which is squeezed) which finally after regression is named as base model and obtained as:

$$\xi_{max} = 1.0(\text{Re}^2\text{Oh})^{0.123}, \quad 2.31$$

This had a much lower slope (with respect to Re^2Oh) than that of the experiments and also overpredicted the results at lower Re^2Oh (high viscosities) and underpredicted them at higher Re^2Oh (low viscosities). They then modified this model to have better agreement with the experiments by changing the contact line velocity. After regression, the following relation was obtained and named as free spreading model:

$$\xi_{max} = 0.91(\text{Re}^2\text{Oh})^{0.133} \quad 2.32$$

This had a higher slope and was closer to their empirical correlation $(0.61(\text{We}/\text{Oh})^{0.166})$. The amendment improved the underprediction at higher Re^2Oh , but the overprediction at lower ones still existed.

Pasandideh-Fard *et al.* [112] used the same approach as Chandra and Avedisian [76] and tried to improve the model by replacing the length scale (which was assumed

to be the splat thickness in Chandra and Avedisian's model [76]) with the boundary layer thickness (δ). Another difference is that they used the advancing contact angle and not the equilibrium one in their model and arrived at this relation:

$$\xi_{\max} = \sqrt{\frac{We + 12}{3(1 - \cos \theta_a) + 4\left(\frac{We}{\sqrt{Re}}\right)}}, \quad 2.33$$

where θ_a is the advancing contact angle. If $We \gg \sqrt{Re}$ and if $We \gg 12$, the above equation will be simplified to the much simpler scaling relation which ignores the capillary effects:

$$\xi_{\max} = 0.5 Re^{1/4}. \quad 2.34$$

They also mentioned that time τ for reaching the maximum spreading diameter is independent of the impact velocity, but other studies showed its dependency on the impact velocity and drop size [113-116]. Pasandideh-Fard *et al.* [117] later developed this relation for a droplet which is also solidifying during impact.

Healy *et al.* [118] compared the available experimental data with different models for spread factor in order to suggest the best of them. They evaluated the models of Madejski [100], Chandra and Avedisian [76], Kurabayashi-Yang [119], Bechtel *et al.* [120], Naber [121], Ford and Furmidge [122] and Shi and Chen [123]. They finally concluded that the model of Kurabayashi-Yang is the most accurate model among them, although it cannot predict the transient spreading. This model is a version of original Kurabayashi [124] model modified by Yang [119]:

$$\frac{We}{2} = \frac{3}{2} \left[1 + \frac{3We}{Re} \left(\xi_{\max}^2 \ln \xi_{\max} - \frac{\xi_{\max}^2 - 1}{2} \right) \left(\frac{\mu_{\text{drop}}}{\mu_{\text{wall}}} \right)^{0.14} \right] - 6, \quad 2.35$$

where μ_{drop} and μ_{wall} are the liquid viscosities evaluated at initial droplet temperature and wall temperature respectively. As mentioned by Ukiwe and Kwok [113], Healy *et al.* [125] later modified Kurabayashi-Yang model by including the contact angle effects through the following computationally obtained correction factor:

$$\xi_{KY, \text{corr}} = \xi_{KY} \left(\frac{45}{\theta} \right)^{0.241}. \quad 2.36$$

Roisman *et al.* [114] analytically solved the mass and momentum equations for the rim to obtain the evolution of droplet and finally gave the theoretical relations for both maximum spreading diameter as well as the time of reaching this extension. Their model included inertia, surface tension, wettability (dynamic contact angle) and viscous dissipation. They compared their analytical model with their own experiments as well as others and observed almost good agreement and concluded that their model gives good predictions for $Re \gg We \gg 1$ [114]. The final analytical expression for the spread factor is:

$$\xi_{max} = \frac{\bar{D}_1}{4} + \frac{\sqrt{6}A_1\bar{D}_1^2}{8} \sqrt{\frac{1 - \cos \theta_d}{We}} + \frac{\sqrt{6}}{48A_1} \sqrt{\frac{We}{1 - \cos \theta_d}}, \quad 2.37$$

where

$$\bar{D}_1 = \frac{D_1}{D_0} = \sqrt{\frac{2}{3\bar{h}_1}} \quad 2.38$$

A_1 is a dimensionless parameter defined as:

$$A_1 = \frac{1}{4\sqrt{6}} \left[\frac{1}{12} + \frac{1}{We} - \frac{1}{Re} \left(\frac{1}{20\bar{h}_1^3} + \frac{3}{5\bar{h}_1} \right) - \frac{\bar{D}_1^2(1 - \cos \theta_d)}{4We} - \frac{\bar{D}_1\bar{h}_1}{We} \right]^{-1/2}, \quad 2.39$$

and $\bar{h}_1$ is computed from:

$$3We + 5(1 - \cos \theta_d)Re \bar{h}_1 = 10Re We \bar{h}_1^3 \quad 2.40$$

The contact angle θ_d in above equations is the dynamic contact angle computed from Hoffman's law [113, 114, 126]:

$$\theta_d = f_{Hoff} \left(Ca + f_{Hoff}^{-1}(\theta_0) \right), \quad 2.41$$

where f_{Hoff}^{-1} is the inverse of the function f_{Hoff} defined as:

$$f_{Hoff}(x) = \arccos \left(1 - 2 \tanh \left[5.16 \left(\frac{x}{1 + 1.31x^{0.99}} \right)^{0.706} \right] \right), \quad 2.42$$

and θ_0 is the static contact angle. For calculating the Capillary number of the spreading film, Roisman *et al.* [114] used the impact velocity as the velocity scale. But Ukiwe and Kwok questioned this choice, since the propagation speed is much larger than the impact velocity [113]. Also based on their theoretical solution, the dimensionless time at which the droplet reaches its maximum is:

$$\bar{t}_m = 1 + \sqrt{\frac{We}{24(1 - \cos \theta_d)}} - \frac{A_1\bar{D}_1}{2}, \quad 2.43$$

where $\bar{t} = tD_0/U_0$. It is obvious that this time depends on impact velocity and drop diameter. Roisman *et al.* later used this model to consider the impact of multiple drops on solid surfaces as well [127].

Roisman *et al.* [128], by an analytical self-similar solution and fitting experimental results [112, 114, 129], proposed the following semi-empirical equation:

$$\xi_{max} \approx 0.87 Re^{1/5} - 0.40 Re^{2/5} We^{-1/2}. \quad 2.44$$

Ukiwe and Kwok [113] reviewed the most of the above models, compared their own carefully done experimental results with them, suggested the best model and finally presented a new model based on the previous ones. They conducted experiments for two fluids, water and formamide which have almost similar surface

tensions (78 and 59 mN/m) and three different surfaces (PMMA, P(*n*BMA/MMA) and P*n*BMA) for 5 different velocities and they measured the droplets' diameter by weighting them. According to their results, Madejski's model (Eq. 2.12) and also Bennett and Poulikakos [82] suggested model (Eq. 2.16) were not accurate and gave more than 50% error. They also evaluated Roismon's model [114] and found it to be almost in agreement with their results. Based on the study of Healy *et al.* [118], they also compared their results with the modified Kurabayashi-Yang model [119] which showed a good agreement. Pasandideh-Fard's model (Eq. 2.33) was the next one evaluated by them which gave the best agreement [113].

Ukiwe and Kwok then modified Pasandideh-Fard's model by considering the surface energy of the droplet in the maximum spreading state and obtained the following relation for spread factor [113]:

$$(We + 12)\xi_{\max} = 8 + \xi_{\max}^3 \left[3(1 - \cos \theta_Y) + 4 \left(\frac{We}{\sqrt{Re}} \right) \right], \quad 2.45$$

where θ_Y is Young equilibrium contact angle which they believe is equivalent to the advancing contact angle. Thus by presenting a good discussion about the conditions and importance of choosing the correct contact angle, they suggested the replacement of the dynamic contact angle at the maximum spreading by the experimental advancing contact angle (θ_a). This correlation gave the best agreement with their experimental data with mean error of 5%. It should be mentioned that based on Bennett and Poulikakos' [82] approach, Ukiwe and Kwok tried to improve further the above model by applying Madejski's treatment of viscous dissipation into their model, but the resulting model was not satisfactory and lead to mean error of more than 59%. Furthermore, they found that the time in which the droplet reaches its maximum extension is not constant and depends on the impact velocity and drop size; this is also mentioned in other studies [114-116] and is in contrast with what Pasandideh-Fard claimed [112]. Although their reviewing and evaluating of different models were interesting and comprehensive, their experimental results only covered two liquids of similar surface tension and they were not very viscous. Also their frame rate was only 2500 frames per second.

Vadillo *et al.* [130] studied in detail the effect of dynamic contact angle for a wide range of surface wettability and also for three liquids of different viscosities, but similar surface tensions. They modified Ukiwe and Kwok's model [113] (Eq. 2.45), by replacing the advancing contact angle in their model with the plateau (dynamic advancing) contact angle (θ_p), defined as "the mean value of the apparent dynamic contact angle during the spreading phase" [130]. The reason was that they observed the dependency of θ_p on viscosity (θ_p increases with increasing viscosity) and therefore it would include the competition of inertia and hydrodynamics. Thus they obtained the following relation which showed better agreement with experimental results, especially at low Re numbers:

$$(We + 12)\xi_{\max} = 8 + \xi_{\max}^3 \left[3(1 - \cos \theta_p) + 4 \left(\frac{We}{\sqrt{Re}} \right) \right] \quad 2.46$$

They concluded that for surfaces with low wettability ($\theta_{eq} > 70^\circ$), advancing contact angle (θ_a) and plateau contact angle (θ_p) are so close and thus it is uncertain that which one should be chosen.

Asai *et al.* [131] presented the following semi-empirical correlation for impact of ink droplets on paper:

$$\xi_{\max} = 1 + 0.48We^{0.5} \exp(-1.48We^{0.22} Re^{-0.21}). \quad 2.47$$

Mao *et al.* [132] also proposed the following correlation for the spread factor based on their experimental data:

$$\left(\frac{1}{4}(1 - \cos \theta) + 0.2 \frac{We^{0.83}}{Re^{0.33}}\right) \xi_{\max}^3 - \left(\frac{We}{12} + 1\right) \xi_{\max} + \frac{2}{3} = 0. \quad 2.48$$

An and Lee [133] studied the impact of shear-thinning droplets on solid surfaces and compared their results to that of the Newtonian liquids. They observed that the maximum spreading diameter of shear-thinning liquid was between that of the Newtonian liquid with same zero-shear viscosity and Newtonian one with infinite zero-shear viscosity. They also corrected the Scheller and Bousfield model [111] to agree with their experimental data.

Clanet *et al.* [95] obtained the following simple and interesting relation for droplet impact on superhydrophobic surfaces in capillary regime which they claim to be still correct for wettable surfaces as well:

$$\xi_{\max} = 0.9 We^{1/4}. \quad 2.49$$

This relation is valid only if $We > 1$ (or the crashing time $t_{cr} \sim D_0/U_0$ must be smaller than the contact time of the droplet $t_{cont} \sim \sqrt{\rho D_0^3/\sigma}$ [134]). They obtained the same scaling relation analytically as well by replacing gravitational acceleration in the definition of capillary length (Eq. 2.8) by an artificial acceleration obtained from scaling $a' \sim U_0^2/D_0$ and using this capillary length as the scale of the puddle thickness in volume conservation. Note that balancing of kinetic energy and surface energy leads to the power-law scaling $We^{1/2}$ which is not directly verified by the experiments, but recently, Laan *et al.* [71] believed that it is the true dependence in the capillary regime.

For the viscous regime, they obtained the following relation and confirmed not only with their own results for viscous silicone oils, but also with those of Marmanis and Thoroddsen [135]:

$$\xi_{\max} \sim Re^{1/5}, \quad 2.50$$

This has weaker dependency on Re than that of Pasandideh-Fard *et al.* [112]. The two different scaling relations for spread factor in viscous regime ($Re^{1/4}$ and $Re^{1/5}$) can be obtained from balancing the kinetic energy of impacting liquid with the viscous dissipation term and they only differ in scaling the viscous loss term. For the transition between the two regimes, Clanet *et al.* [95] defined the following parameter as *impact number*:

$$P = \frac{We}{Re^{4/5}} \Rightarrow P \sim \rho^{1/5} D_0^{1/5} U_0^{6/5} \mu^{4/5} \sigma^{-1}. \quad 2.51$$

If $P < 1$, the viscosity can be ignored (inviscid droplet) and Eq. 2.49 applies, but if $P > 1$, the viscous force will dominate and surface tension can be ignored and Eq. 2.50 will describe droplet spreading. So, the transition between the regimes will occur at $P = 1$.

Kim and Chun [136] gave an analytical model for recoiling of droplet impact via a variation principle by assuming two shapes, cylinder and truncated-sphere for modeling the droplet. The former was development from the study by Kendall and Rosehnow [137] by including wetting and viscous effects and the latter was a modification of the work by Bechtel *et al.* [120] which included the viscous (friction) effects. They also compared their model with their experiments for water, ink and silicone oil and observed good agreement. They concluded that for high Oh numbers ($Oh = 0.006$), the truncated-sphere model is good, while for low Oh numbers ($Oh < 0.002$), the cylindrical model is accurate.

Fedorchenko and Wang [115] modeled the formation of blob (thick borders) formed at the free edges induced by drop impact by considering the liquid to be inviscid and arrived at the following relations for spread factor and the time for reaching the maximum extension:

$$\xi_{max} = 0.92 \sqrt{\frac{2}{9} \frac{We}{(1 - \cos \theta_a)}}, \quad 2.52$$

$$t_m = 0.96 \sqrt{\frac{We}{6(1 - \cos \theta_r)}}, \quad 2.53$$

where θ_a and θ_r are the advancing and receding contact angles, respectively. Fedorchenko *et al.* [116] later continued the study by considering the capillary and viscous effects and concluded that at the late stage of the drop spreading, both capillary and viscous forces must be taken into consideration. They believed that if $We/Re^{1/2} \ll 1$, the capillary force is dominant and spreading is controlled by We number, while for $We/Re^{1/2} \gg 1$, Re is the governing the spreading. For the viscous regime, they offered the following correlation for spread factor:

$$\xi_{max} = 1.455(1 + 0.262Re)^{1/5} \quad 2.54$$

and if $Re \gg 1$:

$$\xi_{max} = 1.113Re^{1/5} \quad 2.55$$

which is similar to that of Madejski (Eq. 2.14). Many similar scaling laws ($\xi_{max} \sim Re^{1/5}$) but with different numerical coefficients were proposed in the literature which were also mentioned by Fedorchenko *et al.* [116].

Eggers *et al.* [138] developed an analytical model, based on mass and momentum equations, for time dependent droplet spreading for both high Re and We numbers and

compared the results with their full numerical simulations of Navier-Stokes equations. Their model ignores both the wetting effects (i.e., the surface repels the liquid completely) and the effects of surrounding air (air bubble entrapment beneath the drop [23, 139-141]). The numerical scheme used for validation of their model was that of Popinet and Zaleski [142]. Through their model, they were able to describe the droplet evolution in four different steps (initial inviscid spreading, growth of boundary layer and film formation, film spreading and finally formation of the rim and retraction) [138]. By a scaling analysis, they also proposed the following law for minimal film thickness:

$$h_f \sim \frac{R}{\text{Re}^{2/5}}. \quad 2.56$$

The results of their model for the minimal film thickness confirm the above scale law and not the previously proposed one as ($h \sim \text{Re}^{-1/2}$) [35]; because they opine that the Re number in Schroll *et al.* [35] study was small and also ignored the initial hyperbolic flow [138].

Most importantly, they discussed the two different scaling laws in the literature for spread factor in capillary regime ($\text{We}^{1/2}$ and $\text{We}^{1/4}$), and based on their results, they believed that the suggested power-law by Clanet *et al.* [95], Eq. 2.49, is not valid for small values of impact number ($P = \text{We}/\text{Re}^{4/5}$). They also mentioned that data from Clanet *et al.* are only for an intermediate regime (between the main two capillary and viscous regimes) and cannot be used for reliable distinction between the two debatable laws for the capillary regime. Hence, they suggested that in the small viscosity limit and high velocities, the true scaling law for this regime would be:

$$\xi_{\max} \sim \text{We}^{1/2}, \quad 2.57$$

which is still debatable.

Lagubeau *et al.* [143] used a Fourier transform profilometry technique in their experiments on droplet impact in order to study in detail the transition between the capillary and viscous regimes. According to their observations, the minimal film thickness scales as $\text{Re}^{-2/5}$ (as in Eggers *et al.* model [138]) and the transition between the two regimes scales as $\text{Re}^{1/5}$. Also they found the proper impact parameter as:

$$P = \frac{\text{We}}{\text{Re}^{2/5}}, \quad 2.58$$

which was already obtained numerically by Eggers *et al.* [138].

Bayer and Megaridis [144] on their experiments for water droplet impact on surfaces with different wettability, suggested the following relation for spread factor which shows no sign of contact angle effect on the maximum spreading diameter:

$$\xi_{\max} = 1.73\text{We}^{0.14}. \quad 2.59$$

Sen *et al.* [145] found their results for impact of biofuel droplets on smooth stainless steel surfaces to agree with model of Ukiwe and Kwok [113], Eq. 2.45, and proposed the following scaling relation for the spread factor with a new exponent:

$$\xi_{max} = 0.72(\text{ReWe}^{1/2})^{0.14}, \quad 2.60$$

which shows much weaker dependency on both Re and We numbers.

Laan *et al.* [71] used a similar approach of Eggers *et al.* for deriving a scaling law for their experiments on water, water-glycerol and blood for forensic applications and eventually proposed the following relation for the spread factor:

$$\xi_{max} = \frac{P^{1/2}}{A + P^{1/2}} \text{Re}^{1/5}, \quad 2.61$$

where $P = \text{We}/\text{Re}^{4/5}$ is the impact number. They believed that the spreading behavior cannot be described by only one parameter and all the three forces (inertia, surface tension and viscous forces) are important in the analysis. Like Eggers *et al.* [138], they also concluded that in the viscous regime, the true scaling would be $\text{We}^{1/2}$ in the capillary regime and $\text{Re}^{1/5}$ in the viscous regime, while water was the liquid in their experiments which follow $\text{We}^{1/4}$ in the capillary regime. They also observed that shear-thinning properties of the blood do not affect the maximum diameter of the drop.

Lee *et al.* [96] applied numerous experiments for different liquids (water, ethanol and three glycerol mixtures) and different surfaces (smooth (glass, steel and parafilm) and rough (with various grit sizes)). They considered the effect of dynamic wetting behavior at low impact velocity by finding the maximum extension of droplet at zero impact velocity ($\xi_{U_0 \rightarrow 0}$) from their experimental data.

By incorporating its effect in the energy balance and applying the same approach as Laan *et al.* [71], they could collapse all of their data into one single scale as:

$$(\xi_{max}^2 - \xi_{U_0 \rightarrow 0}^2)^{\frac{1}{2}} = \frac{\text{We}^{1/2}}{A + \text{We}^{1/2}} \text{Re}^{1/5}, \quad 2.62$$

where A is a fitting parameter.

2.1.3 Other Experimental Studies

Besides the studies mentioned so far, there are many experimental works on droplet impact on solid surfaces. Some of them include Harlow and Shannon [146], Cheng [129], Tanner [147], Karl *et al.* [148], Stow and Hadfield [149], Fukai *et al.* [150], Mao *et al.* [132], Mundo *et al.* [151], Schiaffino and Sonin [152], Rioboo *et al.* [153, 154], Sikalo *et al.* [155-157], An and Lee [133], Luu and Forterre [21], Pittoni *et al.* [158], Antonini *et al.* [19], Lee *et al.* [159], Bertola and Haw [160], Liu *et al.* [6] as well as numerous papers on the complicated subject of splashing [12, 18, 22, 27, 29, 32, 33, 38, 41, 161-173]. Some of the important ones are considered here in more details.

Rioboo *et al.* [153] have systematically studied the impact of droplets on dry solid surfaces with focusing on droplet behavior after impact and the possibility of splashing. Their study covers many liquids of different viscosities and surface tensions as well as surfaces with different roughnesses. Based on this comprehensive study, they gave an elegant classification of possible behaviors for droplet after impinging on

the surface. Depending on the various parameters, six different scenarios might appear upon drop impact: deposition, prompt splash, corona splash, receding breakup, partial rebound and full rebound. Also they discussed the effects of different parameters, varied in their experiments including impact velocity, drop size, roughness amplitude and wavelength, viscosity and finally surface tension, on droplet behavior. Unfortunately, they gave no data on maximum spreading, etc., and did not discuss about scaling of the parameters.

In another study, however, Rioboo *et al.* [154] presented the local spread factor ($\xi(t) = D(t)/D_0$) variation in time for different liquids and surfaces and discussed the effects of similar parameters on it. They classified the temporal evolution of spread factor in four different regions: kinematic ($R \sim \sqrt{t}$), spreading, relaxation and wetting/equilibrium phases. For the kinematic phase (very early stages of impact), they found all their data (on glass surface) to fall into one curve fitted by $\xi \sim 2.8\sqrt{t^*}$ where $t^* = tU_0/D_0$. They observed the increase in local spread factor in spreading phase by either increasing drop size and impact velocity or decreasing viscosity, while it was not so sensitive to surface wettability, except for the late final stage which is both in agreement and disagreement by other studies [154]. Fukai *et al.* [150] reported the effect of wetting effects on spreading, while Mao *et al.* [132] observed no significant effect of static contact angle on maximum spreading. Sikalo *et al.* [155] reported that the contact angle effect on maximum spreading is less significant at higher Re and We numbers. Rioboo *et al.* [154] also observed the temporal spread factor to increase more with increasing Re (at constant We) than increasing We (at constant Re).

Bartolo *et al.* [40] showed experimentally how the non-Newtonian properties of droplets, gained through polymer additives, would prevent rebounding and bouncing of droplet on hydrophobic surfaces (which is usually observed for the Newtonian droplets). Smith and Bertola [36] later showed that the reason for rebound prevention is not the extensional viscosity (as assumed before) but is the stretching of the polymer molecules.

Bird *et al.* [39] studied the initial stages of spreading upon droplet impact and found the inertia dominating the wetting, droplet size and surface hydrophobicity being important in this early regime and the radius of spreading droplet following a power-law whose exponent depends on the equilibrium contact angle.

Luu and Forterre [21] observed much larger maximum extension (twice) for impact of complex fluids on rough surfaces at high impact velocities than those for the Newtonian droplets which leads to significant drag reduction.

Recently, Lee *et al.* [159] studied experimentally the impact of both millimeter and micrometer droplets containing graphene nanoparticles (GNP) on solid surfaces and found their results for the maximum spreading factor to be well agreed with Pasandideh-Fard's model [112], when the surface wetting effects are taken into account. Their results for maximum spread factor for millimeter sized GNP droplets also follow the model of Clanet *et al.* [95] ($We^{1/4}$).

2.1.4 Air Layer beneath the Drop

An important aspect of droplet impact on solid surfaces that has recently become so attractive to the researchers [23, 26, 30, 139-141, 174-181] is the existence of air layer and formation of bubble beneath the droplet. Upon droplet impact, the air layer between drop and surface is highly squeezed which leads to pressure buildup and this increased pressure then causes the formation of a dimple in the drop and consequently entrapment of an air bubble [23].

Van der Veen *et al.* [140] measured the temporal variation of the complete air layer profile beneath the droplet upon impact on solid surfaces using color interferometry method and found the thickness of the dimple formed there. They observed the reduction of dimple height as drop spreads and measured the velocity of dimple center (which was 2 orders smaller than the impact velocity (around 1 – 2 mm/s) as well as the air film horizontal velocity (which is around 20 – 60 mm/s) and showed that by getting close to the rim (higher radius), this velocity is increased [140].

Bouwhuis *et al.* [23] measured the height and volume of the bubble entrapped beneath the drop experimentally (by interference technique) and numerically (using Boundary Integral method for solving potential flow for the liquid coupled by lubrication approximation for air flow) in order to find the critical impact velocity corresponding to maximum dimple height (H_d) and bubble volume (V_b). They also presented scaling relations as $H_d/R \sim St^{1/2}$ and $V_b/R^3 \sim St$ for capillary (low U_0) regime and $H_d/R \sim St^{-2/3}$ and $V_b/R^3 \sim St^{-4/3}$ for inertial (high U_0) regime where $St = \rho_l R U / \mu_g$ is the Stokes number. The crossover between the two regimes corresponding to the maximum bubble height, occurred for $U_0 = 0.25$ m/s for their experimental conditions, since for larger velocities, the inertial force tries to flatten the droplet, while at lower ones, the capillary force tries to take the dimple out and keep the droplet spherical; thus between these two cases, there must be a maximum.

Recently, Van der Veen *et al.* [180], using the same technique, studied the influence of hydrophobic micropatterns on the dimple height and air film formation. When the distance between the pillars was reduced, the dimple height was seen to increase, but it was almost not sensitive to pillars' height.

Lee *et al.* [30] used ultrafast x-ray phase-contrast imaging to track the morphological changes of liquid-air interface and observed that the evolution of air film into bubble happens in a sequential way consisting of three different stages: 1) inertial retraction of the air film; 2) contraction of top air surface into bubble and 3) pinch-off a daughter droplet in the bubble [30]. These may be accompanied by two interesting features: i) generation of daughter droplet because of energy transfer and ii) detachment of bubble from the surface because of solid surface wettability. They reported that the air film radius decreased exponentially with time and that the daughter droplet generation depended only on Oh number, but did not depend on surface wettability (contact angle). However, final bubble detachment depended on contact angle and did not happen for $\theta > \theta_{cr} \sim 40 \pm 5^\circ$ (i.e., it remained adhered to the surface).

By focusing on dynamics of the air layer trapped under the drop, Liu *et al.* [174] studied the air compression as well as the contraction of air film into bubbles and investigated the effects of velocity and viscosity on these phenomena. They observed that before contact, the air is significantly compressed with central pressure being much higher than atmospheric pressure (the degree of compressibility depends on V_0), but after impact, the air pressure is reduced in tens of μs , expands vertically at the edge and generates a thick rim which contracts into bubbles governed by interaction of viscous stress, surface tension and fluid inertia.

By using high speed dual wavelength interferometry, de Ruiter *et al.* [26] found the complete air profile and studied the evolution and collapse of the air layer for different μ, σ, ρ . They observed distinct air profiles (with two or one minimum) at different V . Their results confirmed the visco-inertial theory [38, 164, 165] regarding the formation and evolution of the dimple [26], but they believed that the air layer is not responsible for splashing, contrary to the theory of Mandre *et al.* [38] and consistent with results of Driscoll and Nagel [32].

2.2 Droplet Impact on Heated Solid Surfaces

In previous section, many of the studies done on impact of droplet on solid surfaces were reviewed and some were discussed in more details to show the direction on which state-of-the-art droplet impact research is moving. However, another aspect of this vast phenomenon is when the solid surface is already heated which opens an increasingly wide area of study as it might lead to Leidenfrost state.

The study of droplet impact on heated or superheated (heated to a temperature higher than boiling temperature of the impacting liquid) goes back to more than 250 years ago, even older than that of unheated surfaces, when Dr. Johann Gottlob Leidenfrost [55, 182], a German physician, observed the non-adhesion of water droplets on the heated surface and that they do not evaporate quickly [44, 183]. Since then, this problem has been widely studied both because of its complicated physical aspects and its applications in industry such as spray quenching [184].

2.2.1 Leidenfrost Temperature

If the temperature of the surface over which the droplet is impacting becomes higher than a certain temperature, the droplet will no longer have any contact with the solid surface, a thin vapor layer (film) will form underneath the droplet and the droplet will then levitate on this vapor cushion. This is called Leidenfrost phenomenon and the certain temperature is known as the Leidenfrost temperature (T_L). One of the simplest and most important parameters in this problem is having some knowledge of this T_L , its dependence on different parameters including droplet size, impact velocity (in case of impacting droplet), surface material, surface roughness, purity of the liquid, method of deposition etc. Many studies in the literature address these dependencies.

The different regimes in boiling phenomenon and Leidenfrost state can be described according to the classic boiling curve, shown in Fig. 2.1(a) [185]. Pool

boiling curve is classically a plot of wall heat flux (q'') vs. wall (solid surface) temperature (T_w) or wall superheat ($\Delta T_{sup} = T_w - T_{sat}$) where T_{sat} is the saturation temperature [185, 186]. In the case of droplet deposition on the solid surface, i.e., with almost zero impact velocity, the classical method of measuring the Leidenfrost temperature, which in this case is known as static Leidenfrost temperature (to be distinct from dynamic one which is associated with a droplet impacting the surface with a given velocity), is measuring the lifetime of the droplet until it has fully evaporated. The typical behavior of sessile drop evaporation time (lifetime) is also shown in Fig. 2.1(b). Here we discuss the different regimes that a droplet, deposited on a heated surface, may encounter based on heat flux and evaporation time.

Fig. 2.1 a) The classical pool boiling curve; b) The lifetime (evaporation time) of a sessile drop (taken from Ref. [187]).

Usually there are four regimes in a boiling curve and its corresponding evaporation time curve including single phase regime (conduction/natural convection), nucleate boiling regime, transition regime and film boiling regime (Leidenfrost regime). In single phase regime, associated with low heat flux and long evaporation time, increasing the surface temperature (wall superheat) will increase the heat transfer from the solid to the liquid (almost slowly [186]), and thus the droplet will evaporate more quickly. Heat is also dissipated by evaporation through liquid-gas interface [185]. In this regime, the droplet temperature is usually lower than that of the solid surface, thus the main mechanism of heat transfer would be conduction, while natural convection may also occur due to the surface tension gradients [187]. As the surface temperature is increased, system will arrive in another distinct regime, nucleate boiling, in which bubble nucleation occurs at solid-liquid interface, vapor bubbles are generated, heat flux increases significantly, more cavities (source of bubble nucleation) become active and hence the droplet lifetime decreases even more. This process continues until the surface reaches a certain temperature at which the heat flux is at its maximum (Critical

Heat Flux, CHF), and on the other hand, the droplet lifetime would be at its minimum. Experimentally, for example for the case of water droplet on duralumin (an Al alloy) plate, the lifetime of droplet at this temperature (around 100 °C), was seen to be 200 ms and so the droplet would evaporate quickly [57].

When the surface temperature is increased further, the system will arrive at a transition regime in which gradually, a thin unstable non-continuous vapor film starts to form under the droplet. This would, therefore, decrease the amount of heat transfer (heat flux), act as an insulating vapor layer and increase the lifetime of droplet. At the end of this regime, the vapor layer becomes stable and thick enough to avoid any contact between the droplet and hot solid surface. This point, the moment at which the contact between solid and liquid is lost, is corresponding to a minimum heat flux and maximum lifetime of the droplet on the surface, and thus it is called as Leidenfrost point and its corresponding temperature is called the Leidenfrost temperature. After this point, further increasing the surface temperature will add up to heat flux because of conduction through vapor film and the liquid will levitate on the vapor film [44, 185, 186]. This regime is also called the film boiling regime in which the boiling is occurred without any contact between the solid and liquid. Recently, Quere [44] gave a review of the Leidenfrost phenomenon including time and heat transfer discussions and scales, the Leidenfrost drop's shape, friction in the Leidenfrost state, self-propulsion features and a short discussion of impact effects.

2.2.2 Features of Leidenfrost Drops

A liquid drop resting, usually deposited on the hot surface so gently with almost zero velocity, is called a Leidenfrost drop which is generally a non-wetting situation. Shape of the drop in Leidenfrost regime, depends mainly on its radius and is determined by the competition between gravitational energy ($\rho g R^4$) and surface energy (σR^2) [44].

If $R < a$ where a is the capillary characteristic length (Eq. 2.8), the Leidenfrost drop will be of spherical or semi-spherical shape, interface curvature is $\kappa = 2/R$ and the height is $H = 2R$ [44, 57]. In this case, the bottom surface of the drop would also be slightly flattened due to gravity. The length scale of the contact region for this flattened area was found by Mahadevan and Pomeau [188] to be $l_c \sim R^2/a$.

If $R > a$, the Leidenfrost drop will have a shape of pancake (gravity puddle [95]) which is modeled simply by a cylindrical disk of radius R and thickness H whose shape equation is [44]:

$$\kappa = \kappa_0 + \frac{H - z}{a^2}, \quad 2.63$$

where κ is the interface curvature and z is measured from substrate surface. So, the curvature at top liquid surface would be zero and at the bottom would be H/a^2 . The scale of the puddle thickness will also be $H \sim 2a$.

The shape and stability of the liquid-vapor interface also depend on drop size. For small drops, it would be almost flat, while for large ones, there would be a vapor pocket like a wetting drop which is hanging from the ceiling [44]. For large drops, the interface might become unstable which is proposed [44, 57] to be of Rayleigh-Taylor instability [189].

The main mechanism of heat transfer in vapor film is the conduction heat transfer, and the convection and radiation will be important only at very high temperatures [44, 56]. In order to find some typical values and proper scales of the vapor thickness (h_v) below the drop, lifetime of the droplet (τ_d) and the rate of evaporation of the droplet $\dot{M}$, the puddle is assumed to be a disk as described above, and a Poiseuille flow and lubrication approximation are considered for the vapor flow. Then, it can be shown that for large droplets ($R > a$) [44]:

$$h_v \sim R^{1/2}, \quad 2.64$$

$$\dot{M} \sim R^{3/2}, \quad 2.65$$

$$\tau_d \sim R^{1/2} \text{ or } \tau_d \sim \Delta T_{sup}^{-3/4}. \quad 2.66$$

But for the small droplets, the vapor thickness and droplet lifetime can be scaled as [57]:

$$h_v \sim R^{4/3}, \quad 2.67$$

$$\tau_d \sim R^2 \text{ or } \tau_d \sim \Delta T_{sup}^{-1}. \quad 2.68$$

Biance *et al.* [57] showed that drop radius will decrease in a parabolic trend, while vapor thickness decreases linearly and that the droplet has to have a maximum radius (around 1cm) before any bubbles are formed in its center that will lead to interface instability [57].

There are also many other, mostly experimental, studies on interactions of droplet with hot surfaces and Leidenfrost dynamics [57, 79, 190-207] including studies on bubble nucleation [208-227] and especially many recent papers [9, 10, 13, 24, 28, 31, 34, 42, 43, 93, 228-260] from which some are discussed here in more details.

Gottfried *et al.* [56] studied the Leidenfrost phenomenon both analytically and empirically for impact of several liquids on steel plate mostly in the film boiling regime to find the total evaporation time. They, in their model, considered different mechanisms of heat transfer in this problem including the conduction through the vapor film, radiation from the solid surface to droplet upper and lower surfaces as well as evaporation from the droplet. They assumed the droplet to be isothermal and at saturation temperature. They finally gave an empirical relation for the total evaporation time of droplet which was within 20% of the experimental data.

Wachters *et al.* [261] studied the evaporation of sessile drop deposited on a hot surface experimentally, and by theoretically assuming the droplet to have a flat bottom, found a good agreement. They later [262] developed this model for the case

of droplet impact, but the empirical results and those of the theory were not found to be in agreement.

Bernardin and Mudawar [185] reviewed the different models for calculating the Leidenfrost temperature as well as carrying out many experiments to find the sensitivity of this temperature to different parameters. They finally concluded that the old model of Baumeister and Simon [206] is a good model for T_L . This semi-empirical correlation was a modified version of an earlier model by Spiegler *et al.* [263] and is given as:

$$T_L = T_f + \frac{0.844T_c \left\{ 1 - \exp \left(-0.016 \left[\frac{(\rho_s)^{1.33}}{At} \right]^{0.5} \right) \right\} - T_f}{\exp(3.066 \times 10^6 \beta') \operatorname{erfc}(1758\sqrt{\beta'})}, \quad 2.69$$

where T_f is the liquid temperature, T_c is the critical temperature, ρ_s is the density of solid surface (wall), At is the atomic weight of the surface material and $\beta' = (k_s \rho_s C_{p,s})^{-1}$ in which k_s and $C_{p,s}$ are thermal conductivity and specific heat at constant pressure of the solid surface respectively.

In literature, the temperature measured and considered to be the Leidenfrost temperature is commonly the temperature of the solid surface, since this temperature is easier to measure. The true T_L , however is the temperature of solid-liquid interface which is slightly smaller than that of the substrate [185]. But, this interface temperature can also be calculated by the following relation [264]:

$$T_{int} = \frac{\sqrt{(k\rho C_p)_s} T_{s_0} + \sqrt{(k\rho C_p)_f} T_{f_0}}{\sqrt{(k\rho C_p)_s} + \sqrt{(k\rho C_p)_f}}, \quad 2.70$$

where subscript 's' stands for solid and 'f' stands for liquid and T_{s_0} and T_{f_0} are solid and liquid initial temperatures, respectively.

Based on their experiments for different liquids and surfaces of various characteristics, Bernardin and Mudawar [185] showed that T_L does not depend on drop size, liquid sub-cooling and liquid degassing; surface roughness would increase T_L and different metallic surfaces (aluminum, silver and nickel) will almost have the same T_L , but the Leidenfrost temperature of copper was higher than the others.

A few years later, Bernardin and Mudawar [203] proposed a model for the Leidenfrost temperature for a sessile drop that was developed based on characterization of the surface cavity size and criteria for bubble nucleation and growth, and interaction (merging). To generalize the model to become applicable for spraying problems as well, they later [204] developed this model to include the effects of impact as well (the only difference between the impact model and that of sessile drop is the estimation of liquid properties at the liquid-solid interface) and gave the

following relation for the Leidenfrost temperature for polished surfaces as a function of impact velocity [204]:

$$T_L = 162.0 + 24.3U_0^{0.64}. \quad 2.71$$

This relation does not give an upper limit for T_L , while there is an upper limit for it which is called thermodynamic superheat temperature (maximum temperature that a liquid can have before spontaneously turning into vapor) [186, 204]. For $U_0 = 0$, this relation gives the value of $T_L = 162^\circ\text{C}$ for water which is close to the experimentally measured one for sessile water drop on a polished aluminum plate [185] as well as those obtained by their previous model for sessile drops [203].

Wang *et al.* [194] studied the Leidenfrost dynamics upon impact of two types of ethanol droplets, medicinal (95%) and industrial (99.5%), and distinguished between the Leidenfrost temperature as the wall temperature at which droplet lifetime is maximum and “dry impact” temperature (T_{dry}) as the minimum temperature over which there would be no contact between liquid and solid surface. They observed different regimes during impact and reported that the Leidenfrost temperature does not depend on drop size (which was also reported before [56, 185, 265, 266]) and it is also independent of purity of the liquid. They found the Leidenfrost temperature to be $T_L = 158^\circ\text{C}$ for medicinal ethanol which was only 1°C larger than that of industrial droplet. This is also very close to the value (160°C) obtained in this study for impact on glass substrate for low We number (see chapter 3). They concluded that critical $T_{dry} = T_L$ for $We \approx 0$ and as We is increased, T_{dry} will become smaller than T_L .

Tran *et al.* [24] studied the effect of impact dynamics (Weber number) on the dynamic Leidenfrost temperature (T_L) and developed classification by comprehensive study of impacts of water and FC-72 on superheated surfaces by observing three different regimes during boiling process. In contact-boiling regime (low surface temperatures, T), there is a partial contact between liquid and solid surface, and thus, high heat transfer rate and growth of vapor bubbles were observed. The vapor pressure suddenly increases and leads to disruption of bottom surface of liquid and a vigorous and violent droplet ejection happens because of venting of vapor bubbles. In gentle film-boiling regime (high T), there is no contact of liquid and solid because of sufficient thickness of vapor layer and thus no droplet ejection occurs. Finally in spray-boiling regime (very high T), there is still no contact between solid and liquid, but some spraying like ejection of tiny droplets happen from liquid top surface, but not as violent and vigorous as the first regime [24]. According to their report for mapping regimes, for the transition between contact boiling and film boiling regimes, by increasing the Weber number, T_L also increases (because of competition of liquid dynamic pressure and vapor pressure). The model of Bernardin and Mudawar [204] and the experiments of Yao and Cai [79] also showed the increase of Leidenfrost temperature with Weber number, while Celata *et al.* [260] observed an opposite behavior for low Weber numbers. However, for transition between the film boiling and spraying film boiling regimes, opposite trend is observed; T_L decreases when Weber number is increased (because of ejections of tiny droplets from liquid's top

surface). They observed that all of their data for the spread factor for film boiling regime would fall into one single plot with the scaling of $\xi_m \sim We^{0.39}$.

The very short time interval between droplet's first touch of the hot surface and the time when it leaves the surface is called the resident time. During film boiling regime, over the Leidenfrost temperature, the droplet may bound on the vapor film. It was commonly accepted that the resident time scale would be the same as the period of free oscillating droplet:

$$t_{os} \sim \sqrt{\frac{\rho D_0^3}{\sigma}} \quad 2.72$$

The proportionality coefficient was found by Lord Rayleigh [267] to be $\pi/4$. This value would give smaller resident time than that observed in the experiments of Chandra and Avedisian [76]. Chen *et al.* [199] studied the impact of compound droplets (diesel and water) on inclined hot surfaces well beyond Leidenfrost temperature in order to find the resident time and found the coefficient to be 1.12 for both pure diesel fuel and the compound liquid (if the correct definition of Weber number for the compound liquid is used). In another paper [198], they did similar tests for pure diesel on inclined superheated surfaces and gave the following relation for maximum spreading factor:

$$\xi_{max}^3 - 2(0.052We_n + 1)\xi_{max} + \frac{4}{3} = 0, \quad 2.73$$

where $We_n = \rho U_n^2 D_0 / \sigma$ and U_n is the normal component of impact velocity.

Xie and Zhou [258] gave a theoretical model for boiling of droplet impacted on heated surfaces near the Leidenfrost point and presented the following semi-empirical correlation for total evaporation lifetime:

$$\tau_l = \frac{3.76 \left[\left(\frac{D_0}{2} \right)^{2.25} - \frac{9}{16} C \frac{R_m A_2^{0.25}}{A_1^{0.25}} D_0^2 \right]^{5/9}}{A_1^{0.75} A_2^{0.25}} + \frac{C D_0^2}{A_1}, \quad 2.74$$

where $A_1 = k\Delta T / L\rho_l$, $A_2 = \rho_v g / \mu_v$, $C = 0.0024$ and R_m is the dimensional maximum spreading radius ($R_m = \xi_{max} \cdot D_0$) calculated from Eq. 2.37, proposed by Roisman *et al.* [114]. Myers and Charpin [259] also gave a model for Leidenfrost axisymmetric droplets which included the evaporation from droplet upper surface and also a heat transfer coefficient between the hot solid surface and vapor film.

Linke *et al.* [93] showed that in the Leidenfrost regime, a droplet placed on some heated asymmetric ratchet like surfaces can interestingly exhibit self-propelled motion induced possibly because of viscous forces of the vapor flow. This later motivated others [13, 34, 43, 268-270] such as Cousins *et al.* [271] who constructed "ratchet traps" which was a kind of new Leidenfrost spoon as suggested by Quere [44].

In their study of droplet impact on hot surfaces, Biance *et al.* [230] studied the mechanism of tiny droplets disintegration and proposed that this drop fragmentation in the Leidenfrost regime is due to the formation of a hole (which can be provoked by a defect) and its growth and thus it might be different with classic splashing mechanism.

Celestini and Kirstetter [231] studied the effect of electric field on the Leidenfrost phenomenon and observed that the higher the voltage is, the smaller the thickness of the vapor layer would be. Thus, the vapor layer can collapse after a threshold value for voltage.

Kwon *et al.* [244] studied the possibility of increasing the Leidenfrost point (LFP) by using textured surfaces. They used deionized water gently deposited ($We \sim 1$) on silicon surfaces with microposts of given width and height and different spacing $b = 3.3 - 100 \mu m$ as well as smooth surfaces. For smooth surfaces, T_L was between $270^\circ C$ and $300^\circ C$. For textured surfaces, they observed that surfaces with larger spacing (bigger b) have higher T_L . Thus, the textures can increase the Leidenfrost temperature by enhancing the capillary pressure, but they would also restrict this enhancement. Furthermore, for micro-nano hierarchical textured surface, the Leidenfrost temperature can be increased even more, since they will enhance the capillary pressure without promoting resistance to the vapor flow [244].

Vakarelski *et al.* [10] in their experiments on both heating and cooling of surfaces with different hydrophobicity, observed an interesting phenomenon in which for the superhydrophobic surfaces, the vapor layer associated with Leidenfrost regime would stay on surface and will not collapse by temperature change, as it is expected to fall. In their experiments for inverse Leidenfrost problem (inserting a hot object in a cold liquid) -for a hot steel ball inserted in a water bath- and also for a heated cylinder, they observed that for the case of superhydrophobic surfaces, there was no collapse of vapor layer and so the film boiling regime would be preserved down to the bath temperature. However, for similar hydrophobic and hydrophilic surfaces, the vapor film collapsed at certain temperatures and the surface cooled down more quickly at nucleate boiling regime. This was also seen in heating experiments. Thus, they suggested that for superhydrophobic surfaces, the classical boiling curve ($q'' - \Delta T_{sup}$), which contains a maximum heat flux (end of nucleate boiling regime) and a minimum heat flux (end of transition regime and beginning of Leidenfrost regime), i.e., the transition between film-boiling and nucleate-boiling, shall be altered to a continuously increasing curve. This is because, for superhydrophobic surfaces, the heat flux is smoothly increasing with superheat temperature due to the vapor layer retained on them.

In an another similar study for inverse Leidenfrost phenomenon involving inserting preheated solid balls into cold liquid (Fc-72), Vakarelski *et al.* [31] showed that because of the existence of vapor film in Leidenfrost regime, a huge amount of reduction in drag coefficient can be achieved, especially for high Re numbers ($Re > 1 \times 10^5$). For low Re numbers ($Re \sim 2 \times 10^4$), the Leidenfrost vapor film has no effect on drag coefficient and for moderate ones, drag coefficient reduces with increasing Re.

They attributed this drag reduction to the postponement of flow separation and altering of pressure distribution [31].

Adera *et al.* [9] observed non-wetting droplets (high contact angle $\theta \sim 160^\circ$) sustained on textured hydrophilic (superhydrophilic) surfaces at low superheat temperatures ($\Delta T = T_{wall} - T_{sat}$) of 5 – 50 °C, which is much lower than Leidenfrost range. They believed that this is because of the high pressure region below the droplet due to micropillars. By using surfaces with pillars of different height (h_p), spacing (b_p) and diameter (d_p) as well as a one-dimensional force model, they found that the superheat temperature scales as $\Delta T \propto b_p$, $\Delta T \propto h_p$, $\Delta T \propto 1/d_p$ and $\Delta T \propto 1/R_{base}^2$ where R_{base} is the droplet base radius (analogous to radius of contact region [44, 57, 188]). Finally they discussed that the non-wetting droplets observed in their experiments are distinct from classic Leidenfrost problem in which there is no solid-liquid contact due to the vapor layer formation [9].

Weickgenannt *et al.* [233] showed that by using polymer nanofiber mats, the Leidenfrost phenomenon can be suppressed as nanofiber mats can increase heat transfer rate from the heated solid surface to the contacting liquid. They had already shown that these surfaces could also hinder the receding motion and bouncing of droplet impacting on them [234].

Tran *et al.* [243] studied the effect of microstructures on the dynamic Leidenfrost temperature by doing series of experiments of water droplet impacting on silicon microstructured surfaces (with different pillar heights and spacing) for a large range of We number between 1 – 1000. They observed that dynamic Leidenfrost temperature decreases as the height of the pillars is increased, because larger pillar height corresponds to larger area and thus higher evaporation rate which lowers the temperature of film boiling regime. They also confirmed that Leidenfrost temperature increases as the We number is increased for all structured surfaces [243] as well as for smooth surfaces [24]. The resident time of the droplet was also measured and seemed to be followed by Rayleigh period for free oscillating drop, Eq. 2.72, and so to be independent of impact velocity. All of their data for the maximum spread factor (ξ_{max}) were seen to fall on one universal curve and this agreed well with their analytically obtained scaling law given by:

$$\xi_{max} \sim We^{0.3}, \quad 2.75$$

whose exponent of the power-law is slightly lower than the one obtained before in their experiments for smooth surface (0.39) [24].

Nair *et al.* [242] later continued this work for Fc-72 droplet impact on surfaces covered with carbon-nanofiber (CNF). Curiously, they observed an opposite behavior, in which the dynamic Leidenfrost temperature is larger for CNF surfaces than that of smooth ones (whose T_L was already higher than microstructured surfaces [243]) and also showed that T_L is increased as the nanofiber length is enhanced. This is because the CNF is cooled down by the vapor flow before the drop reaches it. They confirmed this by calculating typical values for time scales of fibers cool-down (τ_c), their

exposure to vapor flow (τ_e) and also that of heat flow within the CNF (τ_h) and found them to be $\tau_h \gg \tau_e > \tau_c$. Regarding the spread factor (ξ_{max}), the data for CNF surfaces also followed the same trend as microstructured ones, i.e., Eq. 2.75.

Using the interference technique, Burton *et al.* [42] measured accurately the curvature, radius and shape of vapor cushion below droplet in the Leidenfrost regime and curiously found that vapor shape does not depend on surface temperature but depends on droplet diameter.

Agapov *et al.* [240] observed that for droplet impact on (asymmetric) tilted nanopillar arrays (TNPAs), no preferred direction of Leidenfrost droplets is observed in steady-state conditions and concluded that for transition regime, the asymmetric wettability of the nanostructured surface is responsible for the directional rebound [240].

Celestini *et al.* [238] studied the Leidenfrost drops in pressures lower than atmospheric pressure and found that the lifetime of the droplet increases magnificently, when the pressure is reduced and also verified their previously proposed [235] correlation for evaporation rate:

$$\dot{M} \propto R^{12/5} P^{12/5} \Delta T^{4/5}, \quad 2.76$$

for droplet radius within the range of $R_i = (R^3 a^4)^{1/7}$ and a where a is the capillary length.

Celestini *et al.* [28] reported that the thickness of the vapor film is increased (opposite to what has been reported before) when the droplet radius is reduced and for drop sizes smaller than a critical value, the droplet would take off from the solid surface because of failure of the lubrication approximation [28].

Nagai and Nishio [272] studied the impact of different liquid droplets (water, acetone, ethanol, R113 and cyclohexane) on heated sapphire and steel plates using total internal reflection technique to measure and compare the Leidenfrost temperature on them. They found no significant difference in Leidenfrost temperature on these two substrates if they have similar wettability. Their experiments were performed at a fixed impact velocity ($We = 17$ for water).

Recently, Orejon *et al.* [253] studied the effect of ambient pressure on the Leidenfrost temperature and observed that T_L will decrease, as the surrounding pressure is reduced for the range of pressures both above and below atmospheric pressure and also by using an analogy with simplified Clausius-Clapeyron equation, proposed the following simple correlation for T_L in terms of pressure:

$$\frac{1}{T_L} = A' - B' \log_{10} P, \quad 2.77$$

where A' and B' are empirical constants which depend on surface properties and were reported in their paper for different surfaces [253].

2.2.3 Recent Studies

Shirota *et al.* [273] studied the impact of ethanol and FC-84 on heated sapphire substrate, placed over a glass prism using the total internal reflection (TIR) technique. They categorized the dynamics of impact on heated surfaces into three regimes: contact boiling, transition boiling and Leidenfrost boiling, depending on the type of the contact between liquid and solid. They observed a weak dependence of Leidenfrost temperature on impact velocity (particularly for ethanol). They reported that the length scale for impact of droplets on heated substrate is associated to the radius of a circular region (neck) scaled as $R_n/R \sim St^{-1/3}$ where St is the Stokes number. The corresponding time scale was reported by them to be the time of the dimple formation $t_d \sim H_d/U_0$. Therefore, $t_d U_0/R \sim St^{-2/3}$ where H_d is the dimple height [23] and R is the droplet radius before impact. By considering the light reflection from the droplet, they also provided a relation for finding the incident angle of TIR, using the optical geometry.

Using the same technique, van Limbeek *et al.* [274] studied the impact of ethanol droplets on a heated glass substrate by focusing on vapor cooling of the substrate. They reported similar phases of impact dynamics as Shirota *et al.* [273] and measured the vapor thickness. By using the 1-D conduction model, they provided a relation for Leidenfrost temperature for a solid surface with small conductivity. They believed that if the time scale of the impact ($\tau_i \sim D_0/U_0$) is smaller than the time scale of substrate (glass) cooling (τ_h), i.e., $\tau_i \ll \tau_h$, then the temperature of the heated solid surface remains constant; otherwise, the substrate would cool down while the droplet is spreading on it [274]. Bertola [275] carried out numerous experiments of impact of water droplets on heated aluminum surface with low impact velocities (maximum We number of 160) and presented a phase diagram based on the outcome of the impact. He categorized the impact dynamics into three independent regimes (secondary atomization, rebound, and splashing) and two combined regimes.

Grishaev *et al.* [276] studied the effect of adding particles on impact of water droplets on hydrophilic and hydrophobic surfaces and observed that the particles make splashing occur earlier. Clavijo *et al.* [277] also investigated the impact on superhydrophobic surfaces considering the effects of slip (both isotropic and anisotropic) and observed that the slip can significantly affect the impact behavior at high We numbers, but its effects can be neglected at low We numbers. Wu *et al.* [278] numerically simulated the impact of droplets on solid surface using Lattice Boltzmann method.

Castanet *et al.* [279] studied the impact of different liquids on a solid surface heated beyond the Leidenfrost point and gave an analytical model based on an inviscid asymptotic solution for the fluid flow inside the liquid lamella. For maximum spreading factor, their data for impact of very viscous liquids (water-glycerol mixtures) were well fitted by following correlation:

$$\xi_{max} \sim 1 + 0.077 Re^{1/5} We^{1/2}. \quad 2.78$$

While for water and ethanol, this relation was obtained:

$$\xi_{max} \sim 1 + 0.23We^{1/2}. \quad 2.79$$

Hamdan *et al.* [280] investigated the impact of water droplets on an inclined surface heated beyond the Leidenfrost point and obtained a relation for the size of the secondary droplets.

Chabicovsky *et al.* [281] studied the effects of the existence of an oxide layer with different thicknesses (on top of the heated steel plate) on the Leidenfrost temperature both experimentally and numerically. They reported that when the thickness of the oxide layer is increased or when its thermal conductivity is reduced, the effective Leidenfrost temperature is increased. Raux *et al.* [282] theoretically and experimentally studied the instability of a Leidenfrost droplet trapped among two hot plates.

Staat *et al.* [283] performed numerous experiments of ethanol droplet impacting on heated sapphire surface and presented a phase diagram including deposition, contact-splash, bounce and film-splash regimes. They also modeled the contact-splash transition using the theory given by Riboux and Gordillo [18, 284].

Ng *et al.* showed that the Leidenfrost point could be postponed by either vibrating the heated surface with low frequency (~ 100 Hz) [285] or applying acoustic waves with high frequency in the droplet [286]. Gauthier *et al.* [287] investigated the levitation of gently deposited droplet (aerodynamic Leidenfrost effect) on a moving plate which was analogous to Leidenfrost phenomenon.

Raufaste *et al.* [288] experimentally studied Leidenfrost droplets reactivity through contacting of different liquids on heated copper plate and observed the copper oxide layer being cleaned by the reactive Leidenfrost flow which can be useful in many applications.

Liang *et al.* [289] reported their experimental data for the impact of ethanol, butanol and water droplets on heated stainless steel surface mostly in the contact boiling regime and at low impact velocities ($We < 80$). Zhang *et al.* [290] studied the impact of water droplets on hot micro-patterned surfaces and reported different modes of impact and found the instability of the droplet due to the rate of vapor generation and the exhaust condition to be responsible for different impact modes.

Moon *et al.* [291] studied the impact of water droplets on hot textured copper plate at low impact velocities ($We < 85$) focusing on wetting behavior of droplet and presented modified relations for the absorbed total (thermal) energy by the droplet for different wetting states. Wildeman and Sun [292] applied an external electric field to Leidenfrost droplets and observed higher jumping of the droplet over the hot surface and attributed it to the droplet's gravity and electrical force and presented an analysis of the droplet's kinetic and potential energies.

Jung *et al.* [293] studied the impact of single water liquid droplet on a heated sapphire plate at a fixed and small impact velocity ($U_0 = 0.44$ m/s) and high surface

temperatures and simultaneously used an IR-camera to measure the temperature change during impact. Using their empirically measured time-dependent temperature profile and transient heat conduction equation, they numerically calculated the heat flux as well. They presented the profile of vapor thickness as well as heat transfer during impact and effective heat transfer area and finally reported the Leidenfrost temperature to be 335 °C.

Vakarelski *et al.* [294] showed that for viscous liquids, the Leidenfrost vapor film can lead to drag reduction for a wide range of liquid viscosities and Re numbers.

Maquet *et al.* [295] studied the effect of gravity on Leidenfrost regime and observed that the Leidenfrost temperature slightly increases with gravity. Shi *et al.* [296] observed the “cold Leidenfrost” phenomenon by putting an ice device over the surface of the water (at room temperature) and could develop a self-propelled hovercraft.

Takata *et al.* [297] studied the impact of series of microdroplets on a heated inclined surface with different surface roughnesses. They observed that increasing the surface roughness will decrease the quenching time, but will enhance both the contact time and spreading diameter of the droplet.

Chen and Bertola [298] studied the Leidenfrost regime for the impact of viscoelastic droplets on hot surfaces at low impact velocities ($We < 120$). From their observations, they reported that the principle contribution in rebound of the droplets comes from the surface forces. In another work [299], they studied symmetry breakup of Leidenfrost drops.

Cordeiro and Desai [300] studied the Leidenfrost phenomenon in nanoscale by molecular dynamic simulations and observed that the Leidenfrost regime is reached at lower surface temperature than that of macro or micro scales.

Villegas *et al.* [301] studied numerically the impact on heated surfaces beyond the Leidenfrost point at low We numbers, observed good agreement with experimental results regarding the evolution of the shape of the droplet and found the temporal change of heat transfer coefficient during impact as well.

Qiu *et al.* [302] studied the impact of a train of water droplets on heated surfaces in terms of steady-state spreading diameter and angle of stable splashing and observed that both of them decrease by increasing the surface temperature.

And recently, Liang and Mudawar [54] reviewed the most important works on droplet impact on heated surfaces focusing on film boiling, critical heat flux and transition boiling.

Reviewing various studies of impact on heated surfaces in this chapter shows the need for a deeper understanding of transition to Leidenfrost regime. This thesis aims to perform a systematic study of droplet impact on heated surfaces by focusing on high surface temperatures in order to obtain a more detailed comprehension of transition to Leidenfrost regime.

Chapter 3:

Impact on Glass Surface

We are interested in examining the true contact area during impact and studying its dependence on different factors such as the temperature and droplet kinetic energy. This can be achieved by the principle of total internal reflection (TIR) technique [17, 25, 303-310]. This chapter introduces this technique, the experimental setup, and the results for impact of droplets on substrates made of glass, a material having low thermal conductivity.

3.1 Experimental Method

Total internal reflection is a well-known method in optical physics [311], but it has also been used in other fields such as biology [303, 308] and fluid dynamics [304]. Kolinski *et al.* [25] used this technique to study the droplet impact on unheated solid surfaces and focused mostly on the role of air film beneath the droplet on the impact dynamics. Recently, they used the same technique to study the lift-off mechanism [17] and also bouncing of droplet on superhydrophilic surfaces [306].

Here, we use the TIR technique for the study of droplet impact on superheated surfaces. This method enables us to measure the exact contact (wetted) area between an impinging droplet and the hot surface during impact. The contact area is of crucial importance in both understanding the impact dynamics and boiling process.

When an incident laser beam is directed to an interface between two transparent media with different indices of refraction (for instance, from glass to air), the beam will be either reflected back to the incident medium or refracted into the other medium depending on the incident angle and the indices of the two media. Fig. 3.1 shows a simple schematic to illustrate the different scenarios that could happen.

Fig. 3.1 a) The incident and refracted beams governed by Snell's law; b) Critical angle corresponding to a 90° refracted angle and c) incident angles larger than the critical angle corresponding to total internal reflection.

The incident beam in medium n_1 is hitting the interface at angle α_i , measured from the vertical direction, and is then refracted in the second medium n_2 with angle α_r (Fig. 3.1(a)). The diversion of refracted angle is simply calculated *via* the Snell's law:

$$\frac{\sin \alpha_r}{\sin \alpha_i} = \frac{n_1}{n_2}. \quad 3.1$$

Thus, the value of refracted angle in a given medium depends on incident angle and is inversely proportional to that medium's index of refraction. As the incident angle is increased, the refracted angle also increases, until for a given incident angle, the refracted angle would be the right angle (90°). The incident angle for which the refracted angle is $\pi/2$ is called the critical angle obtained from (Fig. 3.1(b)):

$$\alpha_c = \sin^{-1} \left(\frac{n_2}{n_1} \right). \quad 3.2$$

When this incident angle α_i is equal or larger than this critical angle, the beam will be totally internally reflected back into the incident medium. That is the basis of TIR used in this research. It is considered that $n_a (= n_2)$ is the index of refraction of the less dense medium (air) and $n_g (= n_1)$ is that of the denser medium (glass). Although the beam is totally reflected back into the incident medium, at the interface however, there is a part of the incident energy that penetrates through the medium of lower index of refraction (air). This so-called evanescent wave [311] propagates in the less dense medium parallel to the interface and its intensity decays exponentially with the distance from the interface as [303, 304]:

$$I = I_0 \exp \left(-\frac{z}{d_p} \right), \quad 3.3$$

where I_0 is the intensity of the electric field at the interface ($z = 0$), z is the normal distance from the interface, and d_p is the penetration depth which depends on the incident angle, indices of refraction of the two mediums and the wavelength of incident light in vacuum, λ_0 :

$$d_p = \frac{\lambda_0}{4\pi} \frac{1}{\sqrt{n_1^2 \sin^2 \alpha_i - n_2^2}}. \quad 3.4$$

The penetration depth decreases with increasing α_i and its typical value is usually of the order of the light wavelength in vacuum (λ_0), except for $\alpha_i = \alpha_c$ for which $d_p \rightarrow \infty$ [303].

The index of refraction of a medium is a function of wavelength [312] and (depends weakly on) temperature [313-315]. The values of refractive index for different materials used in this chapter, i.e., air, ethanol and glass (N-BK7), at room temperature and wavelength $\lambda_0 = 640\text{nm}$ (red laser light) are given in Table 3.1. From this table and using Eq. 3.2, the critical angle of air-glass interface and that of glass-liquid (ethanol) interface can be found to be $\alpha_{ga} = 41.3^\circ$ and $\alpha_{gl} = 63.9^\circ$ respectively. Also, the values of penetration depth for different incident angles and the wavelength $\lambda_0 = 640\text{nm}$ can be calculated from Eq. 3.4 and are listed in Table 3.2. The

implementation of TIR method in study of droplet impact will be elaborated in the next section following the experimental setup description.

Table 3.1 Refractive index of the required materials [312].

Material	Air	N-BK7 Glass	Ethanol
Index of Refraction (n)	1	1.5148	1.3601

Table 3.2 Values of penetration depth for red light ($\lambda_0 = 640$ nm) and different incident angles.

Incident Angle (α_i)	42°	45°	50°	55°	60°
Penetration Depth d_p (nm)	307	132.7	86.5	69	59.9

3.2 Experimental Setup

A schematic of the experimental setup is shown in Fig. 3.2. It consists of various components to create and satisfy the required conditions for TIR.

Fig. 3.2 Schematic of experimental setup.

First we describe the path of the illumination light for the TIR recording: Laser beam (L) is first expanded by a beam expander, reflects at the first mirror (M1), enters the prism's side panel (left), illuminates the upper surface of prism, then is totally internally reflected (from the dry areas) and is directed to the high-speed camera BC (SAX2, Photron Inc.). The camera is connected to an appropriate long distance microscope with proper magnification. The camera then captures the wet and dry areas on the surface. There is also another camera, SC (APX, Photron Inc.), which records the images from side to calculate droplet diameter D_0 and impact velocity U_0 from which Weber number can be calculated. In the following, the detailed description of each part of the TIR experimental setup is presented.

i) Laser: A red light laser (iBeam Inc.) with wavelength $\lambda = 640$ nm was used for capturing the data presented in this report. The diameter of the light leaving the laser source was 1.3 mm.

ii) Beam Expander: In order to expand the laser light to cover larger area of the prism and therefore being able to observe more of the wetted area, a system of beam expander was used. This system was composed of a first diverging ($f = 25$ mm) and then converging lenses ($f = 250$ mm) where f is the focal length. The system can expand the beam diameter 10 times. A schematic of how the beam expander works is shown in Fig. 3.3.

Fig. 3.3 Schematic of the laser and beam expander.

iii) Plane Mirrors: Since the laser light is horizontal, as is the beam expander, and since the light should be hitting the side and finally top surfaces of the prism with a special angle, the incoming light should be ordinated somehow to satisfy the condition of TIR at prism's upper surface. To achieve this, two plane square mirrors (50×50 cm, Thorlabs Inc.), M1 and M2 are used in either side of the prism. Mirror M1 is orientated in such a way that when the collimated laser light hits it at a given angle, the reflected light from the mirror will have the required angle which will finally lead to necessary TIR angle at the prism's top surface. A second plane mirror M2 is orientated in such a way that when the reflected laser light hits it at a given angle, the reflected light from the mirror will be illuminated to the high-speed camera (BC) horizontally.

iv) Prism: The prism is the main part of TIR experiment. A Dove prism made from N-BK7 glass (Thorlabs Inc.) was used throughout the experiments reported in this chapter. The prism angle at the corners is 45° and its width (top surface) is 15 mm. A schematic of the prism and holder and a photo of the prism inside the machined holder are respectively shown in Fig. 3.4(a) and (b). The surface roughness of N-BK7 glass has been reported to be between $0.03 - 0.06 \mu\text{m}$ [316].

v) Holder: Based on the prism dimensions, a holder was designed and made in order to hold the prism as well as heating it up. The schematic and CNC-manufactured holder is shown in Fig. 3.4. The holder has two holes at its sides for embedding two cartridge heaters which would heat the prism from side symmetrically. There is also a hole at the side of the holder where the temperature probe is inserted. The thermometer

is connected to the temperature controller for controlling the temperature of the prism and holder.

Fig. 3.4 a) 3D drawing of holder and prism; b) The holder and the prism inside it.

vi) High-Speed Cameras: Two high-speed cameras, SAX2 and APX, both from Photron Company were used for recording the impact from bottom and side-views, respectively. Each camera was equipped with appropriate lenses. A long distance microscope (Optem Inc.), including camera mount (C-mount), camera tube, core optical module, lower function module and lower lens was mounted on to the SAX2 camera which totally produced the required magnification for bottom-view imaging. The pixel size for the side-view camera was $27.56 \mu\text{m}$ and that of the projected bottom-view was $11.6 \mu\text{m}$.

vii) Syringe Pump: The syringe pump (Cole-Parmer 74900 Series) was used to pump the working liquid (ethanol) through the plastic tube to the flat-tip needle (with inside diameter of 0.41 mm and outside diameter of 0.64 mm) with a very small flow rate (0.025 ml/min) in order to generate the droplets. The flow rate is set sufficiently small to allow the droplet to fall solely due to its weight. The surface tension keeps the liquid from falling, while more and more liquid is supplied by the pump. When the weight of the drop is large enough to overcome the surface tension, the droplet will be detached from the needle tip and impacts the surface of the prism.

viii) Temperature Controller: The heaters and thermocouple were connected to a temperature controller (3.5W, E5CWL, Omron Corp.) in order to control the temperature of the holder and consequently that of the prism. The main purpose of the system is setting the temperature of the prism surface to a desired value by pumping heat *via* the two cartridge heaters embedded in the holder. Note that the heaters are symmetrically inserted in the holder. For using this controller in order to handle the prism temperature, the desired temperature is set at the controller and the system controls the necessary current through the 2-PID algorithm. If the temperature exceeds the set point, the controller will reduce the current to decrease the heat transfer from the heaters to the holder. Since the temperature used for connecting to the controller is the temperature of the holder through the probe inserted in the side wall of the holder (see Fig. 3.4) and because of heat loss to the surroundings from all sides, the temperature of the prism surface is not necessarily the same as the holder. That is why a K-type surface temperature probe was used to measure the true temperature of the

prism and the probe was calibrated for a range of holder's temperature to obtain the required plot for calibration.

ix) Surface Temperature Probe: In order to measure the temperature of the upper surface of the prism, a K-type surface probe (TME Ltd.) was used. This probe was mounted on the surface of the prism to measure the true temperature of the prism that is different, especially at high temperatures, with that of the holder measured with a thermocouple.

A schematic of the whole experimental setup including almost every component described above is shown in Fig. 3.5. The pump drives the liquid to the needle held perpendicular to the surface; finally the weight of the drop overcomes its surface tension and falls vertically and spherically (especially at low impact velocities) on the surface; the side-view camera (SC) captures the images for which the necessary white light is provided by the light source shown at the left; the laser light - after being reflected back from the prism through TIR mechanism as described in detail earlier - reaches horizontally the bottom-view camera (BC); the two cameras are connected to a PC for recording the images. A photo of the setup constructed in the lab is shown in Fig. 3.6.

Fig. 3.5 A schematic of the experimental setup.

Fig. 3.6 The actual experimental setup.

3.3 Condition of Total Internal Reflection

The fundamentals of total internal reflection technique and the experimental setup were described in the previous sections. Here the detailed process of how TIR method works and how it is implemented to attain the true wetted area by the impacting liquid is described.

The expanded laser beam hits the side surface of the prism so that the refracted beam in the glass medium made an incident angle α_i to the prism's upper surface. In order for the TIR condition to be satisfied, this incident angle should be larger than the critical angle of glass-air interface. If this happens, then when the droplet impinges on the surfaces, the area where the surface is dry, i.e., not wetted by the liquid, will be shown in the camera as grey (bright) region. But this is not the only condition that must be satisfied. This angle must also be smaller than the critical angle of glass-liquid (herein glass-ethanol) interface, so that the light will penetrate into the liquid and be refracted there; thus the area where the liquid wets the surface will be observed in the camera as dark (black) areas. In summary, the necessary condition for distinguishing between the dry (bright) and wetted (dark) areas is that the light must be totally internally reflected in the glass-air interface, but not in the glass-liquid interface. Therefore, the incident angle should be such that $\alpha_{ga} < \alpha < \alpha_{gl}$, where $\alpha_{ga} = 41.3^\circ$ is the critical angle of glass-air interface and $\alpha_{gl} = 63.9^\circ$ is the critical angle of glass-liquid interface. With this arrangement (see Fig. 3.2), a beam incident on a dry area of the upper surface is totally reflected back to the glass medium and is directed to a high-speed camera, whereas a beam incident on a wetted area will be refracted into the liquid medium; thus, the dry area will appear bright and the wetted one dark in the recorded images. The sensitivity of the TIR method is extremely high, since it is capable of distinguishing a dry area from a wet one as long as the distance between the

glass and the liquid is larger than the evanescent wave's penetration depth d_p . Herein, since the whole system (mirrors, prism etc.) is arranged in such a way that the beam reaches the prism top surface with angle $\alpha \approx 50^\circ$, according to Table 3.2, the penetration depth is $d_p = 86$ nm.

Together with the bottom-view camera for the TIR recordings, another high-speed camera (APX, Photron Inc.) was used to capture the droplets from the side. From the side-view recordings, the velocity V and droplet diameter D_0 before impact can be obtained to enable computation of the Weber number. The working liquid used in the experiments reported in this chapter is ethanol. We used the typical physical properties of ethanol listed in Table 3.3 for our results in this chapter.

Table 3.3 Physical properties of ethanol at room temperature [317].

Physical Property	Value
Density ρ (kg/m ³)	789
Surface Tension σ (mN/m)	22.39
Kinematic Viscosity ν (m ² /s)	1.4×10^{-6}
Dynamic Viscosity μ (Pa.s)	1.1×10^{-3}
Boiling Point T_b (°C)	78.4

3.4 Experiments on Glass Substrate

When the droplet dispensed by the syringe pump becomes large enough that its weight dominates the surface tension force, the droplet is detached from the needle. The setup was adjusted so that the droplet would fall on the part of the solid surface that was illuminated by the evanescent wave, as described in the previous sections. The temperature probe was calibrated and the setup was tested to capture the wetted area using the high-speed recordings. Experiments were then carried out for different temperatures and impact velocities. The experiments were done for six different impact velocities (which were achieved by changing the needle height) and fifteen different temperatures. The heights tested were $H = 5, 10, 20, 30, 40$ and 50 cm and the temperatures used were $20 - 300$ °C with 20 °C intervals. Each test was carried out two times to ensure the repeatability of the experiment. The surface temperature was changed by using the temperature controller described in previous section. All the experiments reported in this chapter were done using ethanol as the working liquid and were carried out at lab conditions with constant humidity, atmospheric pressure and constant temperature ($T \approx 20$ °C).

In order to minimize the effect of surface roughness on the wetting characteristics and impact dynamics, the surface of prism was cleaned once before starting the whole experiments by standard method of washing with ethanol, IPA, rinse with DI water and then drying with new and clean lens paper. After each test, especially at high T at which cleaning is difficult, the surface was cleaned gently by new and wet lens paper.

3.5 Image Post-Processing

The entire image processing in this research has been done in MATLAB. Many different codes have been written for processing the different images to get the desired quantities. Here, the process of analyzing the data will be described.

3.5.1 Side-View Analysis

As mentioned before, one of the high-speed cameras recorded the images from side-view in order to find the two quantities: droplet radius just before impact (D_0) and impact velocity (U_0). They were used to calculate the Weber number. The pixel-size calibration of side-view images was carried out using an object with known dimensions such as a needle.

A series of snapshots for side-view analysis for one experiment is shown in Fig. 3.7. These images are obtained for $T = 60\text{ }^\circ\text{C}$ and $H = 20\text{ cm}$ which corresponds to $U_0 = 1.89\text{ m/s}$ and $We = 258$. This figure shows how the droplet during impact is captured as a circle, its center is detected and the slope of the plot of its center position versus time would simply give the impact velocity. The droplet size just before impact (D_0) is also obtained similarly by capturing the droplet as a circle for the frame just before impact (last image of Fig. 3.7(b)). Note that the time origin $t = 0$ is taken at the frame in which the droplet first makes contact with surface. For detecting the circles, a MATLAB function called `imfindcircle` is used whose algorithm is based on Circular Hough Transform (CHT) [318-320].

Fig. 3.7 a) Snapshots of side-view of a droplet impacting the surface for measuring drop size and impact velocity; b) Detected circles as the droplet in order to find the center and radius of the droplet. The last image shows how the droplet diameter is found. Here the velocity is 1.89 m/s , temperature $T = 60\text{ }^\circ\text{C}$, drop size 2.1 mm and $We = 258$.

The vertical position of the droplet's center can be plotted as a function of time and after fitting the best line to the data using the Least Square method, the slope (the impact velocity) can be obtained. Fig. 3.8 shows a sample of such a plot for the above case. Note that the time scale has been considered as positive values for the sake of simplicity. The error of fitting for plot shown in Fig. 3.8 was $\pm 0.5\%$.

Fig. 3.8 Temporal variation of droplet center from which impact velocity as the slope can be found.

All the other cases of side-view tests were similarly analyzed to find the impact velocities and We numbers. The droplet size was almost constant for all cases: $D_0 = 2 \pm 0.01$ mm, impact velocity U_0 range was from 0.93 m/s to 2.86 m/s and Weber number varied from 64 to 590. For the cases of very small impact velocities for which the droplet upon impact is not perfectly spherical, the droplet diameter was found by averaging the horizontal and vertical diameters.

3.5.2 Bottom-View Analysis

The analysis of bottom-view images was one of the most important parts of the processing. This analysis consists of different steps described below. Due to optical property of the Dove prism, an image passing through it is flipped and inverted, but is not transposed laterally. Thus, when images are recorded by the bottom-view camera, they are already elongated; in other words a circle there would be seen as an ellipse (see Fig. 3.9). Thus, the images should first be projected back in the normal plane.

Fig. 3.9 A sample of an image of wetted area from bottom-view which shows the stretched image because of passing through Dove prism.

i) Pixel-Size Calibration: Since the images from the bottom-view are elongated, thus unlike the side-view, the pixel-size (PS) in horizontal (x) and vertical (y) directions are no longer equal. In order to get the images back to the normal plane, an accurate calibration must be done. For that, a small board with a tiny cross sign was used for

calibration as shown in Fig. 3.10. The true size of the cross itself was already measured using the side-view camera.

Fig. 3.10 The cross sign used for calibration of bottom-view. The sizes of cross wings are clearly not equal which shows the image is stretched.

ii) Projection: When the PS in both directions of bottom-view images are known from the previous step, the ratio of PS in x and y directions can be found and using that, the image can be projected back into the normal plane.

iii) Background Subtraction: In order to find the correct values of contact area and removing clutter (noise) from the images, the background information was also subtracted from the images. Since all the images are digital (numeric), this can easily be done by subtracting the information of first image (the one before the contact which contains no wetted area and only contains the surface, i.e., the background) from all the images. This process helps the analysis significantly. Fig. 3.11 (a) and (b) show a sample image before projecting to normal plane and after both projection and background subtraction, respectively. Next, the center of the contact area is found (using previously mentioned methods or fitting circle manually to the wetted area) which will be used later for analysis. The MATLAB code used for this process is given in Appendix A.

Fig. 3.11 a) A recorded bottom-view image before projecting into normal plane; b) The image projected back to the normal plane and after background subtraction. Here the velocity is 0.93 m/s, temperature $T = 20\text{ }^{\circ}\text{C}$, drop size 2.1 mm and $We = 64$.

iv) Binary: In image processing, gray level is defined as the intensity of a monochrome image at any location. This gray level has to be within a certain range called the gray scale. This scale is then usually shifted to be in a format such as $[0, L' - 1]$ where $L' = 2^{k'}$ is the number of gray levels and k' is a positive integer. Then the lowest value in gray scale (0) represents the black and the maximum value ($L' - 1$) is considered as white and all the values among them would be different gray shades between black and white [321]. An image having $2^{k'}$ gray levels is referred to as a k-bit image; thus an 8 bit image would have 256 gray levels where 0 is black and 255 is white.

The next step for analyzing the images is converting a digital image from one class such as gray scale (either 8 bit or 16 bit images) into another class, binary, in which all the values representing the image will be 0 or 1 where 0 refers to black and 1 refers to white regions. This can be done *via* different functions in MATLAB such as `im2bw` [322]. When the images are converted into binary, there might also be some clutters around the true wetted area. Before calculating the wetted area, these clutters should be removed, and in other words, the image should be cleaned. Fig. 3.12 (a) and (b) show a sample image in its original gray scale and its corresponding binary image, respectively.

Fig. 3.12 a) Original gray scale image; b) Binary image. The velocity is 0.93 m/s, temperature $T = 20^\circ\text{C}$, drop size 2.1 mm and $We = 64$.

v) Cropping: Since laser light through the TIR can only cover a part of the prism surface, therefore, only a portion of the wetted area is fully visible in the recorded images. Hence, in order to get the wetted area (as well as contact length) as accurately as possible, first the center of wetted area is found. Then, a portion (the maximum possible visible region at late times) was cropped from the original image and eventually the entire wetted area (length) was calculated based on the angle of the portion selected. Fig. 3.13 shows a case of image before cropping (part (a)) and the image with specified area for further analysis (part (b)). The contact area or length will then be calculated based on the angle of this analyzed region.

Fig. 3.13 a) Original image before cropping and b) The image with detected center and selected segment for analysis. The velocity is 1.34 m/s, temperature $T = 40^\circ\text{C}$, drop size 2.1 mm and $We = 132$.

vi) Interference Patterns: Interference is the superposition or overlapping of two coherent (having the same phase difference or frequency) waves leading to formation of so-called interference patterns which include alternatively dark and bright fringes [311, 323]. Generally if the phase difference between the two waves is $0, 2\pi, 4\pi, \dots$ or the waves are in phase, the interference is called to be constructive (corresponds to bright regions) and if it is $\pi, 3\pi, \dots$ or the waves are out of phase, it is called destructive (corresponds to dark regions) [311]. This was originally seen in Young's double-slit experiment in which the dark and bright fringes were formed on screen due to interference of two light waves from the two slits. However, interference may also occur for the case of thin films (dielectric films) in which the light hits a thin film that is deposited over another medium. Depending on the refractive indices of the mediums and thickness of the film, interference might happen. In this case, the phase change can be due to both optical path as well as the reflection. The incoming light will undergo a phase change (π or $\lambda/2$ where λ is the light wavelength) if it is travelling from a less dense (lower index of refraction) medium to a denser one. For the opposite case, there would be no phase change. Also, the wavelength in any medium is $\lambda = \lambda_0/n$ where λ_0 is the wavelength in vacuum and n is the index of refraction of the medium [323].

In TIR recordings, interference patterns can be observed. In regions where the droplet has wet the surface, the laser light hitting the prism's upper surface (glass-liquid interface) is reflected inside the prism, while part of the beam is also refracted into the liquid phase, hitting the upper surface of the liquid (liquid-air interface). This light is reflected back inside the liquid and then refracted into the prism. These two light beams create interference patterns as described above which look like dark and bright fringes on the TIR recorded images. These fringes can be constructive or destructive depending on the film thickness, laser wavelength and values of indices of refraction. Therefore, these interference patterns are indeed representative of the

wetted areas; while they might appear similar to dry areas due to the higher brightness. One of the challenges in analyzing bottom-view images to obtain the wetted area is to distinguish between the true dry area and interference patterns that look like dry area. Detection of the wetted area, therefore, is done manually by observing the dark and bright fringes. This task is particularly difficult at higher temperatures ($T = 160 - 200\text{ }^{\circ}\text{C}$) at which boiling process has started with bubbles being nucleated at different regions. The interference fringes are more profound at higher Weber numbers.

Fig. 3.14 shows two snapshots of low temperature and high Weber number at two different times for distinguishing dry area and interference patterns. The arrows show the interference patterns with dark and bright fringes. These regions are recognized and categorized as wetted areas and different from the true dry areas as specified in the snapshots.

Fig. 3.14 Snapshot at two different times, a) $t = 1.3\text{ ms}$ and b) $t = 3.1\text{ ms}$, for distinguishing between true dry regions and interference patterns. The arrows show the interference fringes and dry areas (outside the wetted region) are also specified. The velocity is 2.85 m/s , temperature $T = 20\text{ }^{\circ}\text{C}$, drop size 2.03 mm and $We = 584$.

Fig. 3.15 shows a similar snapshot taken at a higher temperature $T = 180\text{ }^{\circ}\text{C}$. The arrows distinguish the difference between the interference patterns and true dry areas. As mentioned above, the process of recognizing these two different types of regions was done manually for all the cases in order to have higher accuracy.

Fig. 3.15 Snapshot for distinguishing between true dry regions and interference patterns at a higher temperature. The arrows show the interference fringes and dry areas (due to bubble nucleation). The velocity is 1.87 m/s, temperature $T = 180\text{ }^{\circ}\text{C}$, drop size 2.04 mm and $We = 253$.

v) Extraction of the wetted area and length of the contact line: One of the goals of the research project is to find the true wetted area as well as the length of this contact region, known as contact length. From an image processing approach this simply means calculation of the black regions, either area or perimeter. When the images were converted to binary, wetted area is calculated by simply counting the number of pixels containing the value 0. For the case of contact length (perimeter), the function `bwperim` was used to extract the borders (edges) of the contact region. Fig. 3.16 summarizes the process of analyzing an image from the original one to the clutter-removed binary one and the one with detected perimeter. The original image (Fig. 3.16(a)) was first read, then checked for interference patterns and if these fringes existed in it, they were detected and filled as wetted area to be distinguished with dry area (Fig. 3.16(b)). The image was then converted into binary format (Fig. 3.16(d)). Note that Fig. 3.16(c) is not necessarily required, if there was no interference pattern present. Then the image was cropped according to the method described earlier and finally the perimeter of the contact region, i.e., the contact length is detected. Fig. 3.16(e) was used for calculating the wetted area. Fig. 3.16(f) was utilized for calculation of the wetted length (perimeter of the wetted region) and then the total area and length is simply computed based on the angle of the cropped region.

A striking wetted pattern was recorded by using the TIR technique. This type of wetted patterns, to the best of our knowledge, is observed for the first time in the case of droplet impact and is an important contribution of this research project. These *fingering* patterns are observed for temperatures higher than $200\text{ }^{\circ}\text{C}$ for most of the Weber numbers. The higher the We , the more noticeable these fingering channels are. A sample of such patterns is shown in Fig. 3.17. This shows another example of detected wetted area and length. As it can be seen from the figure, the algorithms are well capable of detecting the area and length of the wetted zones containing regular and irregular patterns.

Fig. 3.16 Snapshot for the entire process of image processing. a) Original gray scale image; b) Image checked for interference patterns and modified to be treated as wetted are; c) Binary format of the original image; d) Binary format of the modified image; e) Cropped binary image ready to calculate the contact area and f) The detected contact length from the binary image used for calculating the perimeter of the wetted region. The velocity is 1.34 m/s, temperature $T = 40\text{ }^{\circ}\text{C}$, drop size 2.1 mm and $We = 132$.

Fig. 3.17 Snapshot for a case of striking fingering patterns observed via TIR. a) Gray scale image; b) Binary image and c) Detected perimeter of the binary image. These photos show qualitatively how the wetted area and length are measured. The velocity is 2.58 m/s, temperature $T = 200\text{ }^{\circ}\text{C}$, drop size 2.1 mm and $We = 484$.

3.6 Impact Behavior on Glass Substrate

In the previous sections, details of the experimental setup and the steps of image processing were described. In this section, some snapshots of different experiments of both side and bottom-views are presented to give a better idea of the quantitative results that will be presented in the following section. The qualitative results here are exhibited for different temperatures and impact velocities (We).

Fig. 3.18 shows different snapshots for the lowest (room) temperature, $T = 20\text{ }^{\circ}\text{C}$ tested and all six different impact velocities (Weber numbers) at different times. As time goes on, the spreading radius increases for higher We . For the highest Weber number, some ejection of tiny droplets from the liquid rim is visible. The interference patterns are also clearly visible in most of the cases, especially for high impact velocities. Since the surface is not heated; there is no effect of the heated wall on the spreading regime yet.

Fig. 3.19 shows similar snapshots for the same We numbers and an intermediate temperature, $T = 80\text{ }^{\circ}\text{C}$. Here since the temperature is higher than the results in Fig. 3.18, the spreading diameter at similar time and We would be larger due to the surface tension reduction. However the temperature is not high enough to cause boiling and start the nucleation of bubble.

Fig. 3.20 shows similar snapshots for the boiling regime for different Weber numbers and $T = 180\text{ }^{\circ}\text{C}$ at which boiling process and bubble nucleation are obvious. Fig. 3.21 shows snapshots for the striking fingering patterns for different Weber numbers at $T = 200\text{ }^{\circ}\text{C}$. This is the first temperature at which fingers are observed. More discussions about the fingers and their formation and dependence on temperature and Weber number will be presented in the next section. Fig. 3.22 shows snapshots for different Weber numbers at $T = 240\text{ }^{\circ}\text{C}$. The fingers are still observed particularly at higher Weber numbers. The Leidenfrost regime, zero wetted area, is observed at $T = 280\text{ }^{\circ}\text{C}$ for the first two heights and for the remaining three larger heights (impact velocities), the dynamic Leidenfrost temperature was seen at $300\text{ }^{\circ}\text{C}$.

The next snapshots compare the effect of temperature for one fixed Weber number. Fig. 3.23 presents the snapshots for $We = 63$ (average of Weber number between the considered cases) for six different temperatures including the Leidenfrost temperature. As it can be seen from Fig. 3.23(f), at $T = 280\text{ }^{\circ}\text{C}$, the Leidenfrost regime is reached and there is no contact between the liquid and solid surface and therefore there is no area being wetted by the liquid, i.e., the bottom-view images indicate no dark region. Figures 3.24-3.28 show similar snapshots for $We = 130, 254, 371, 489$ and 584 respectively for different temperatures including the Leidenfrost temperature. Note that in all these cases, the Leidenfrost temperature is $T = 300\text{ }^{\circ}\text{C}$.

Fig. 3.18 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 65$; b) $We = 131$; c) $We = 259$; d) $We = 371$; e) $We = 488$ and f) $We = 584$. Here the temperature $T = 20^\circ\text{C}$ is the lowest temperature tested and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Images in the same column were taken at the same time after impact.

Fig. 3.19 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 65$; b) $We = 131$; c) $We = 259$; d) $We = 385$; e) $We = 478$ and f) $We = 598$. Here the temperature is $T = 80^\circ\text{C}$ and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Images in the same column were taken at the same time after impact.

Fig. 3.20 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 62$; b) $We = 129$; c) $We = 253$; d) $We = 376$; e) $We = 485$ and f) $We = 593$. Here the temperature is $T = 180^\circ\text{C}$ and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Images in the same column were taken at the same time after impact.

Fig. 3.21 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 63$; b) $We = 128$; c) $We = 252$; d) $We = 364$; e) $We = 484$ and f) $We = 582$. Here the temperature is $T = 200^\circ\text{C}$ and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Images in the same column were taken at the same time after impact.

Fig. 3.22 Qualitative results of spreading of droplet upon impact on glass surface for a fixed temperature and different We numbers: a) $We = 63$; b) $We = 132$; c) $We = 254$; d) $We = 369$; e) $We = 478$ and f) $We = 595$. Here the temperature is $T = 240\text{ }^{\circ}\text{C}$ and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Images in the same column were taken at the same time after impact.

Fig. 3.23 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20^\circ\text{C}$; b) $T = 100^\circ\text{C}$; c) $T = 180^\circ\text{C}$; d) $T = 200^\circ\text{C}$; e) $T = 240^\circ\text{C}$ and f) $T = 280^\circ\text{C}$. Here the average Weber number is $We = 63$ ($H = 5 \text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05 \text{ mm}$. All the inset scale bars represent the length scale of 2 mm. Note the Leidenfrost regime (no wetted area) for case (f) at $T = 280^\circ\text{C}$. Images in the same column were taken at the same time after impact.

Fig. 3.24 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 280\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 130$ ($H = 10\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 280\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.

Fig. 3.25 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20^\circ\text{C}$; b) $T = 100^\circ\text{C}$; c) $T = 180^\circ\text{C}$; d) $T = 200^\circ\text{C}$; e) $T = 240^\circ\text{C}$ and f) $T = 300^\circ\text{C}$. Here the average Weber number is $We = 254$ ($H = 20 \text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05 \text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300^\circ\text{C}$. Images in the same column were taken at the same time after impact.

Fig. 3.26 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 371$ ($H = 30\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.

Fig. 3.27 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 484$ ($H = 40\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.

Fig. 3.28 Qualitative results of spreading of droplet upon impact on glass surface for a fixed Weber number and different temperatures: a) $T = 20\text{ }^{\circ}\text{C}$; b) $T = 100\text{ }^{\circ}\text{C}$; c) $T = 180\text{ }^{\circ}\text{C}$; d) $T = 200\text{ }^{\circ}\text{C}$; e) $T = 240\text{ }^{\circ}\text{C}$ and f) $T = 300\text{ }^{\circ}\text{C}$. Here the average Weber number is $We = 584$ ($H = 50\text{ cm}$) and the drop size for all cases is $D_0 = 2.05 \pm 0.05\text{ mm}$. All the inset scale bars represent the length scale of 2 mm . Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300\text{ }^{\circ}\text{C}$. Images in the same column were taken at the same time after impact.

3.7 Results of Impact on Glass Substrate

In this section, the results of analysis of the experiments of TIR for droplet impact on low-thermal-conductive substrate (glass) including contact area, contact length, spreading rate, spread factor and number of fingers are presented¹.

3.7.1 Phase Diagram

In order to describe the different types of impact behaviors and investigate the conditions for each behavior, we constructed a phase diagram. Fig. 3.29 is the phase diagram that represents the different regimes observed during the impact of ethanol on glass prism. Based on the observations from TIR, four regimes corresponding to four distinct behaviors were constructed on a $T - We$ phase space. In order to illustrate the different regimes better, several series of snapshots for $We = 484$ are shown in Fig. 3.30. Note that for having a more comprehensive phase diagram, extra tests were done for small range of Weber numbers ($8 < We < 50$) to obtain a more accurate description of different regimes. These tests for small Weber numbers are only considered and discussed in phase diagram here.

i) Spreading Regime: The first expected regime in droplet impact is clearly the spreading regime in which the droplet starts wetting the surface while spreading on it. In this regime, the temperature is relatively low and an impacting droplet spreads on the surface without any noticeable boiling effect, i.e., no vapor bubbles are observed during the spreading time (see Fig. 3.30(a)). This regime corresponds to the region of blue squares in Fig. 3.29. The temperature range of this regime is $T < 140\text{ }^{\circ}\text{C}$ and is almost insensitive to Weber number, i.e., the liquid inertia has no effect on the upper limit of this regime.

ii) Bubbly Boiling Regime: As the temperature is increased further, bubbles are nucleated on the surface during the spreading stage of impacting droplet. The impacts having this behavior are said to be in the *bubbly boiling* (contact boiling) regime. In this regime, the TIR recordings show tiny dry areas caused by the nucleating vapor bubbles on the glass-liquid interface (Fig. 3.30(b) at $t = 0.9\text{ ms}$). Subsequently, these dry areas may either disappear due to detachment of bubbles from the surface or merge with the neighboring ones to create larger dry patches. The density of the bubble nucleation is observed to grow with temperature which is consistent with previous studies of nucleate pool boiling [208]. The relation between the nucleation density and temperature has also been used in theoretical modeling of the transition to the Leidenfrost regime. Nucleate boiling at the solid-liquid interface is the mechanism for diminishing the wetted area and the eventual formation of the vapor layer [56, 185, 204, 205]. The bubbly boiling regime in our experiments was observed for $140\text{ }^{\circ}\text{C} < T < 180\text{ }^{\circ}\text{C}$ for almost all Weber numbers, except for the very small Weber ($We = 8$) in which we directly observed Leidenfrost regime at $T = 160\text{ }^{\circ}\text{C}$. This value is close

¹ Some of the results of this section have been published in: Khavari, Mohammad, Chao Sun, Detlef Lohse, and Tuan Tran. "Fingering patterns during droplet impact on heated surfaces." *Soft matter* 11, no. 17 (2015): 3298-3303.

to the experimentally measured value (158°C) which was already obtained for ethanol [194].

Fig. 3.29 Phase diagram showing four major boiling regimes: spreading (blue squares), bubbly boiling (red circles), fingering boiling (purple asterisks) and Leidenfrost (green stars). Several points with transitional behaviors (right triangles) were observed between the Leidenfrost and the fingering boiling regimes: the wetting patterns only appear for a very short time (≈ 1 ms) and do not have the “fingering” shapes. The dashed line indicates the transition between the fingering boiling and the Leidenfrost regimes.

Fig. 3.30 Qualitative results of spreading of droplet upon impact on glass prism surface for a fixed Weber number and different temperatures: a) $T = 20$ °C; b) $T = 100$ °C; c) $T = 180$ °C; d) $T = 200$ °C; e) $T = 240$ °C and f) $T = 300$ °C. Here the average Weber number is $We = 484$ ($H = 40$ cm) and the drop size for all cases is $D_0 = 2.05 \pm 0.05$ mm. All the inset scale bars represent the length scale of 2 mm. Note the Leidenfrost regime (no wetted area) for case (f) at $T = 300$ °C.

iii) Fingering Boiling Regime: As the temperature is increased, fingering-like patterns were observed upon impact of the droplet. This is called the *fingering boiling* regime and corresponds to situations in which the liquid creates many channels to escape from the center of the wetted area. It is interesting to note that it is due to the capabilities of the TIR technique in showing the wetted areas that these patterns were observed. The fingering boiling regime was observed in the experiments for $200 < T < 240$ °C; the most obvious fingering-like patterns were observed at $T = 200$ °C. As seen in Fig. 3.29, these patterns were weekly dependent on We . For very small We , fingers were not observed and the impact directly transitions from the bubbly boiling regime to Leidenfrost regime without going through any fingering boiling regime.

In the fingering boiling regime, it was observed that after the liquid makes contact with the surface, the wetted pattern quickly evolves to consist mainly of two parts: the “fingering” section which keeps advancing outwards, and the “bubbly” boiling part at the center (Fig. 3.30(c) at $t = 0.5$ & 0.9 ms). In the phase diagram in Fig. 3.29, the fingering boiling regime is shown as asterisks. The dynamics of the fingering patterns change with temperature and Weber number. The delay time of the fingering patterns, measured from the frame in which wetting is first observed ($t = 0$), to the one in which the fingering pattern first appears, decreases with increasing temperature and Weber. The number of fingers also depends significantly on both T and We .

If the temperature is increased further, the fingers will also disappear due to high rate of heat transfer from the heated surface. For the cases of high Weber number, ($We > 64$), we observed a transitional behavior, neither perfectly bubbly boiling nor perfectly fingering, in which there were small dispersed regions wet by the liquids. This is considered to be a transition to the Leidenfrost regime and shows a very fast dynamic in which the system is approaching the Leidenfrost regime where there would be no contact between the liquid and solid surface.

iv) Leidenfrost Regime: Finally, for very high surface temperatures, the system will reach the Leidenfrost regime in which there is absolutely no contact between the liquid and solid surface, and therefore, no wetted area will be seen from the bottom-view (see Fig. 3.30(d)). For the impact on glass substrate, the transition to the Leidenfrost regime is observed to depend weakly on Weber number (Fig. 3.29). For very high We , it seems to be independent of droplet inertia. In our case, for the highest Weber number ($We = 584$), the dynamic Leidenfrost temperature was observed to be $T = 300$ °C, while for the lowest one ($We = 8.5$), it was seen to be $T = 160$ °C. This clearly shows the effect of liquid inertia on Leidenfrost temperature. It was recently shown that increasing Weber number will increase the dynamic Leidenfrost temperature [24].

3.7.2 Contact Area

One of the first objectives of this study and the main advantage of using TIR technique in study of droplet impact was an accurate measurement of the true wetted area. The detailed method of how contact area was observed through TIR and how this quantity was calculated from the recorded images were described in previous sections. Here the main results regarding this parameter for different temperatures and Weber numbers are presented.

Contact Area at Different We and Same T : Fig. 3.31 shows the temporal variation of contact area for different temperatures and each for different Weber numbers. Here four different plots are shown for four distinct regimes ($T = 20^\circ\text{C}$, $T = 180^\circ\text{C}$, $T = 200^\circ\text{C}$, $T = 260^\circ\text{C}$,) to illustrate the variation of wetted area at each regime. These plots include the variation of the scaled contact area, normalized with drop initial diameter (D_0^2) with time. Furthermore, the corresponding log-log plot of the scaled contact area vs. non-dimensional time, time normalized by crashing time D_0/U_0 , is shown as inset in each plot. The error bars in the following figures have been obtained by finding the standard deviation from different number of experiments for each case (each experiment has usually been done two times).

Fig. 3.31 Temporal variation of contact area for different We and temperatures: a) $T = 20^\circ\text{C}$; b) $T = 180^\circ\text{C}$; c) $T = 200^\circ\text{C}$; d) $T = 260^\circ\text{C}$.

As it can be seen and as expected, the contact area is increased by increasing We for each T especially at low temperatures. In fingering boiling regime ($T = 200\text{ }^{\circ}\text{C}$), although the contact area is initially higher for higher We , but at later times and because of the fingers formation and boiling process, the contact area is decreased. This can be seen for $T = 260\text{ }^{\circ}\text{C}$ in which the contact area is much smaller than lower temperatures. Note the different values of vertical axis in Fig. 3.31(c and d) and Fig. 3.31(a and b). Note that for each case, the log-log plot of the normalized parameters almost fall into one single curve. The TIR recordings clearly show how We and surface temperature affect the contact area and its sharp reduction at very high temperature, close to that of Leidenfrost.

Contact Area at Different T and Same We : Fig. 3.32 shows the temporal variation of normalized contact area for different Weber numbers and each for different temperatures. The plots are presented ascendingly for all the six We numbers. In order to avoid crowded plots, only several temperatures are shown including room temperature and one temperature in each regime; the bubbly boiling, fingering boiling and Leidenfrost regimes. By increasing the temperature, the wetted area first slightly increases because of the reduction of surface tension (compare the curve for $T = 20\text{ }^{\circ}\text{C}$ and $T = 120\text{ }^{\circ}\text{C}$ in all plots of Fig. 3.32). When the temperature is high enough, (bubbly boiling regime) boiling is started which causes considerable reduction of the contact area. In the fingering regime, due to formation of fingers and bubble nucleation (mostly at the center), the contact area decreases significantly. For very high temperatures ($T > 260\text{ }^{\circ}\text{C}$), the wetted area becomes very small at all We numbers. Eventually, the system reaches the Leidenfrost regime (corresponding to zero contact area) at $T \geq 280\text{ }^{\circ}\text{C}$ for $We \leq 130$ and at $T = 300\text{ }^{\circ}\text{C}$ for $We > 130$.

3.7.3 Length of Contact Line

The second quantity measured in the experiments was the total length of the three-phase contact line denoted here as the contact length (L). This parameter is especially useful at higher temperatures, bubbly and fingering boiling regimes, to help us understand how bubbles are nucleated and contribute to the boiling process. Similar to contact area, the variation of contact length is also presented for different temperatures and Weber numbers.

Fig. 3.32 Temporal variation of contact area for different We and temperatures: a) $We = 64$; b) $We = 130$; c) $We = 255$; d) $We = 372$; e) $We = 481$ and f) $We = 591$.

Length of Contact Line at Different We and Same T : Fig. 3.33 shows the temporal variation of contact length for different temperatures and each for different Weber numbers. Here four different plots are shown for four distinct regimes ($T = 20^\circ\text{C}$, $T = 180^\circ\text{C}$, $T = 200^\circ\text{C}$, $T = 260^\circ\text{C}$,) to illustrate the variation of contact length with time at each regime. The contact length is normalized by drop initial diameter (D_0). Furthermore, the corresponding log-log plot of the scaled contact length vs. non-dimensional time is shown as inset of each plot. Note that, in some cases and for the purposes of clarity, the plots of only three We are shown, so comparison would be possible. In the spreading regime (Fig. 3.33(a)), the contact perimeter increases monotonically with time, since the droplet is simply spreading on the surface. Here also all the plots of different Weber numbers fall into one single curve in log-log plot which shows the universal behavior of contact length. When the temperature is high enough for the boiling to occur (Fig. 3.33(b)), the length of the wetted region first reaches a maximum at some intermediate time and then starts decreasing, almost for

all Weber numbers. The fluctuations of the contact length as later time are due to the capillary waves. In the fingering boiling regime (Fig. 3.33(c)), because of formation of the fingers, the contact length increases significantly; reaches a maximum at an intermediate time and then is gently reduced for all We . At very high temperatures and close to the Leidenfrost regime (Fig. 3.33(d)), the contact length is decreased as expected.

Fig. 3.33 Temporal variation of contact length for different We and temperatures: a) $T = 20^\circ\text{C}$; b) $T = 180^\circ\text{C}$; c) $T = 200^\circ\text{C}$ and d) $T = 260^\circ\text{C}$.

Length of Contact Line at Different T and Same We : Fig. 3.34 shows the temporal variation of normalized contact length for different Weber numbers and each for different temperatures. The plots are presented ascendingly for all the six We numbers and in order to avoid crowded plots, only several temperatures are shown which include room temperature, temperatures associated with bubbly boiling and fingering boiling regimes and the Leidenfrost temperature. At low temperatures, the length of the wetted region increases monotonically with time for all We . When the temperature is increased up to boiling regime, the contact length is increased sharply and reaches a maximum at the fingering regime; the fingers contribute significantly to length of the contact line. If the temperature is increased further, the contact length is reduced and finally would drop to zero at Leidenfrost state. The temporal evolution of the contact length is almost similar for all Weber numbers.

Fig. 3.34 Temporal variation of contact length for different We and temperatures: a) $We = 64$; b) $We = 130$; c) $We = 255$; d) $We = 372$; e) $We = 481$ and f) $We = 591$.

3.7.4 Rate of Spreading

Besides contact area and length, it would be interesting to know how fast the liquid is wetting the solid surface and how this rate is influenced by temperature and We . This rate is calculated as the rate of contact area at early stages of impact. For that, since in most cases (different temperatures and We), the initial variation of wetted area (see Fig. 3.31 and Fig. 3.32) is almost linear, the slope of this linear section is calculated for each case for three different times: $t = 1, 1.5$ and 2 ms and their average is considered as the spreading rate of the liquid for that case. Fig. 3.35 summarizes how the mean slope is calculated for each experiment. This figure has been obtained for $T = 40^\circ\text{C}$ and $We = 65$ and includes the equation of the fitted line for each case.

Fig. 3.35 Calculation of the rate of spreading (slope of linear section of contact area) obtained for $T = 40^\circ\text{C}$ and $We = 65$. a) The original contact area; b) Fitted line to the data in $0 < t < 1$ ms; c) Fitted line to the data in $0 < t < 1.5$ ms and d) Fitted line to the data in $0 < t < 2$ ms. The equation of the fitted line is also shown as inset in each plot. The final slope for this case is the average of the slope of these three fitted lines.

Now we compare the spreading rate for different surface temperatures and We numbers. The slope is defined as $S = A/t$ where A is the contact area and t is the time. Fig. 3.36 shows the variation of slope with surface temperature for different We numbers in different regimes. We observe that the rate of spreading in the first two regimes, spreading and bubbly boiling, does not change with temperature (even at high temperatures such as $T = 180^\circ\text{C}$) and it is increased by We . This rate is then decreased sharply as the system arrives at fingering regime which shows the sudden reduction of spreading, since the boiling process and formation of fingers slow it down. Hence, the liquid spreads equally fast for a wide range of surface temperatures (the first two regimes).

Fig. 3.36 Variation of rate of spreading with temperature for different Weber numbers.

3.7.5 Maximum Values

We now calculate the maximum values of contact area and contact length and study their dependence on surface temperature and We number. As discussed in chapter 2, the maximum extension of the droplet spreading on the surface is traditionally calculated from side-view imaging. Here, however, the TIR technique enables us to calculate the accurate contact area. We define the equivalent diameter, D^e obtained from the maximum contact area as:

$$D_m^e = \sqrt{\frac{4A_{max}}{\pi}}, \quad 3.5$$

And thus the equivalent spreading factor is:

$$\xi^e = \frac{D_m^e}{D_0}, \quad 3.6$$

This parameter is more accurate than the one calculated from side-view. Fig. 3.37 shows the variation of ξ^e with temperature for all six different We.

Fig. 3.37 Variation of equivalent spreading factor with temperature for different We . Inset: log-log plot of ξ^e vs. We for spreading and bubbly boiling regimes. The solid line indicates the best fitted line for the empirical data whose slope is 0.16.

As shown in Fig. 3.37, in the spreading regime, ξ^e slightly increases with T for a fixed We due to a reduction in the liquid's surface tension. As the impacts enter the bubbly boiling regime, ξ^e starts to decrease gradually because higher T causes more bubble nucleations and therefore a reduction in the wetted area. However, at any fixed We , the deviation of ξ^e caused by variation in T in these two regimes is less than 20% and can be considered insignificant. Thus, generally, the equivalent spread factor is almost independent of temperature for the first two regimes: spreading and bubbly boiling. In the fingering boiling regime, the spread factor decreases sharply with temperature due to violent and fingering-type boiling. Therefore, the dependence of ξ^e on We can be approximated by a single power law $\xi^e \sim We^\alpha$ in the first two regimes. The inset plot of Fig. 3.37 shows the log-log plot of the spread factor versus We in the spreading and bubbly boiling regimes. Here, the independency of ξ^e with temperature is shown clearly in these regimes; the dependence of ξ^e on We is best fitted by the following scaling relation:

$$\xi^e \sim We^{0.16}, \quad 3.7$$

The exponent coefficient of the above power-law is much smaller than that obtained from energy equation (0.5) [95], that observed experimentally for superhydrophobic surfaces (0.25) [95, 168] and that for the impacts of droplets on surfaces in Leidenfrost (film-boiling) regime where the vapor provides an extra driving mechanism for the spreading liquid (0.3) [24, 243]. That is because this exponent is not representative of the *apparent* spreading diameter under the liquid, but the one resulted from the *absolute* wetted area.

The effect of boiling can be further assessed by examining the total length L of the three phase contact line of the wetted pattern (as discussed in section 3.6). The variation of L as a function of time was already shown from which we determine the maximum value. Fig. 3.38 shows a plot of the normalized contact length $\bar{L} = L/D_0$ versus T for different We .

Fig. 3.38 Variation of normalized contact length with temperature for different We . Inset: log-log plot of $\bar{L}$ vs. We for spreading and bubbly boiling regimes. The solid line indicates the best fitted line for the empirical data whose slope is 2.7.

Fig. 3.38 clearly shows that the contact line length does not change significantly with temperature in the spreading regime due to the absence of vapor bubbles, while the bubble nucleation in the bubbly boiling regime causes an abrupt increase of $\bar{L}$. To elaborate the process responsible for the increase in $\bar{L}$ in the bubbly boiling regime, a scaling relationship between $\bar{L}$ and T is obtained as follows. The normalized superheat temperature is defined as:

$$\bar{T} = \frac{T - T_b}{T_b}, \quad 3.8$$

where T_b is the boiling temperature of the liquid. If we denote N_b as the density of the bubble nucleation, the total number of bubbles scales as $N_b D_m^{e^2}$. Thus the total length of the contact line will scale as:

$$L \sim N_b D_m^{e^2} l_b, \quad 3.9$$

where l_b is the typical size of the vapor bubbles. Thus:

$$\bar{L} = \frac{L}{D_0} \sim \frac{N_b D_m^{e^2} l_b}{D_0}, \quad 3.10$$

Now, N_b can be related to the normalized superheat temperature using a generally accepted scaling relation resulted from nucleate boiling theory:

$$N_b \sim \bar{T}^p, \quad 3.11$$

Where p is an empirically determined coefficient [208]. The dependence of l_b on T is considered weak [208] and can be neglected. Similarly, since D_m^e was shown insensitive to changes in T (in both the spreading and the bubbly boiling regimes; see Fig. 3.37), it is considered here as a function of We only which was obtained before in Eq. 3.7 ($D_m^e \sim D_0 We^{0.16}$). Thus:

$$\bar{L} \sim We^{0.32} \bar{T}^p, \quad 3.12$$

Hence, the log-log plot of $\bar{L} We^{-0.32}$ vs. $\bar{T}$ for different We is plotted as an inset in Fig. 3.38 as well. A best fit to the data would give $p = 2.7 \pm 0.9$. Therefore:

$$\bar{L} \sim We^{0.32} \bar{T}^{2.7}, \quad 3.13$$

This value for p is within and at the lower end of the reported range $2 < p < 6$ [208, 224, 324]. It should be noted here that the large error of the estimation of p in the experiments is due to the limited range of $\bar{T}$ tested here. The wide range of the reported values of p may originate from many attributing factors including the material of the surface and how it is prepared, but it is generally accepted that p is smaller for smoother surfaces [208, 325]. Together with the experimental observation, this strongly suggests that the dominant thermal process in the bubbly boiling regime is the *nucleate boiling*. As discussed before, the transition between the bubbly boiling and the fingering boiling regimes marks a distinctive change in the dependence of ξ^e on T and We (Fig. 3.37): ξ^e becomes insensitive to changes in We but decreases with increasing T and eventually diminishes at the Leidenfrost temperature. It is also remarkable that exactly at this transition, $\bar{L}$ starts decreasing with increasing temperature (Fig. 3.38). This implies that the dominant thermal process is no longer the nucleate boiling and the heat transfer rate from the surface to the liquid starts decreasing and eventually minimizes at the Leidenfrost transition. Thus, the continuous reduction of ξ^e and $\bar{L}$ in the fingering boiling regime, which spans a significant temperature range, quantitatively shows that the Leidenfrost transition is continuous and should be considered with mass transport of vapor taken into account.

3.7.6 Number of Fingers

Although the fingering patterns can only be observed within a limited range of temperature $200^\circ\text{C} < T < 260^\circ\text{C}$, the geometry of these patterns change substantially with temperature. At lower temperature $T = 200^\circ\text{C}$, the fingers appear more slender in larger quantity, whereas at higher temperature ($T = 240^\circ\text{C}$), they are more fragmented and have larger width and in smaller quantity. Varying the Weber number from 130 to 591 does not seem to significantly change the geometry of these patterns. To quantitatively characterize this behavioral change of the patterns, the number of “fingers” for each impact experiment is counted (by user) as a function of time and then the time-averaged number N is obtained. In Fig. 3.39(b), plot of N vs. We for

different temperature is shown. All the snapshots were taken at the same time (0.9 ms) after impact. It can be seen that N increases with We but decreases with temperature. The inset plot shows the corresponding log-log plot along with the best fitted line to the data whose slope is 0.5. Although it is not clear to us what causes this dependence of N on We and temperature, we conjecture that the formation of these patterns is related to the instability of the vapor flow as it is squeezed out from the gap between the droplet and the solid surface. Therefore this phenomenon calls for more thorough experimental and theoretical investigations at the onset of impact.

Fig. 3.39 Variation of the number fingers with We for different temperatures. Inset: log-log plot of N vs. We . The solid line indicates the best fitted line for the empirical data whose slope is 0.5.

Chapter 4:

Impact on Sapphire Surface

In this chapter, we present the experimental results of a systematic study for droplet impact of different liquids on a heated substrate with high thermal conductivity. To this end, we use a sapphire (Aluminum oxide, Al_2O_3) prism.

4.1 Experimental Method and Setup

The experimental method used here is the Total Internal Reflection (TIR) technique as described in the previous chapter (impact on glass substrate), except that here we use a sapphire prism (equilateral prism of $15 \times 15 \times 15 \times 10$ mm, Meller Optics Inc.) as the heated substrate. A simplified schematic of the experimental setup is shown in Fig. 4.1. Two high-speed cameras are used for recording the bottom-view (SA-X2, Photron) and side-view (SA5, Photron) images during impact. For these series of experiments, we use high frame rates: 300000 and 540000 fps for bottom-view and 10000 fps for side-view recordings.

Fig. 4.1 a) Simplified experimental setup for impact on a heated sapphire prism; b) Snapshot of droplet from side-view recordings before reaching the heated substrate; c) Snapshot of droplet from side-view recordings after contacting with the heated substrate and d) A typical snapshot from bottom-view recordings via TIR technique which distinguishes between dry (bright) and wetted (black) areas. All scale bars indicate the length scale of 1 mm.

Table 4.1 compares the physical properties of glass (NB-K7) and sapphire. Besides the difference in their optical properties ($n_{\text{sap}} > n_{\text{glass}}$), note the *significant* difference between their thermal properties ($k_{\text{sap}} \gg k_{\text{glass}}$). Sapphire's thermal conductivity is higher than that of stainless steel ($k_{\text{steel}} \approx 16 \text{ W/(m.K)}$). This leads to a completely different behavior during droplet impact, in particular at temperatures close to Leidenfrost temperature. For these series of tests, we use four different liquids: acetone, ethanol, IPA and water¹. Experiments are done for a wide range of surface temperature, mostly close to Leidenfrost regime.

¹ Some preliminary tests were done for FC-72 as well.

Table 4.2 presents the physical properties of the tested liquids. Table 4.3 summarizes the experimental conditions for the entire experiments reported in this chapter for these liquids including the range of surface temperature, impact velocity, drop size and critical angle of each liquid with sapphire.

Table 4.1 Comparison of physical properties of N-BK7 glass [326] and sapphire [327] at $T = 20\text{ }^{\circ}\text{C}$.

Material	N-BK7 Glass	Sapphire
Index of Refraction (n)	1.5148	1.7656
Conductivity, k (W/(m. K))	1.114	31.4 ¹
Density, ρ (kg/m ³)	2510	3980
Specific Heat, C_p (kJ/(kg. K))	0.858	0.757
Melting Point ($^{\circ}\text{C}$)	557	2050

Table 4.2 Physical properties of the liquids at $T = 20\text{ }^{\circ}\text{C}$ and $P_0 = 101\text{ kPa}$.

Property \ Liquid	Acetone [329]	Ethanol [329]	IPA [317]	Water [329]
Boiling point, T_b ($^{\circ}\text{C}$)	56.3	78.3	82.3	100.0
Density, ρ (kg/m ³)	790	800	781	999
Surface tension, σ (mN/m)	23.7	22.8	21.7	72.9
Latent heat, h_{fg} (kJ/kg)	552	1030	755	2454
Specific heat, C_p (kJ/(kg. K))	2.16	2.40	2.60	4.18
Viscosity, μ (mPa. s)	0.323	1.198	2.038	1.002
Molar mass (kg/kmol)	58.1	46.0	60.1	18.0
Index of refraction	1.36	1.36	1.37	1.33

Table 4.3 Experimental conditions of the entire tests for impact on sapphire prism.

Property \ Liquid	Acetone	Ethanol	IPA	Water
Critical angle with sapphire	50.4 $^{\circ}$	50.3 $^{\circ}$	51.3 $^{\circ}$	48.9 $^{\circ}$
Droplet size, D_0 (mm)	2.0	2.0	2.0	2.3
Impact velocity, U_0 (m/s)	0.62 – 2.74	0.53 – 2.66	0.52 – 2.67	0.54 – 2.78
Surface temperature, T ($^{\circ}\text{C}$)	20 – 370	20 – 210	20 – 230	20 – 520

¹ Sapphire has different properties at different orientations; the above value is the mean of $k_{c\perp} = 30.3$ and $k_{c\parallel} = 32.5$ W/(m. K) for the optical axis perpendicular and parallel, respectively, to the c-axis of the crystal. Some other references report smaller values for sapphire thermal conductivity: $k_{c\perp} = 23$ and $k_{c\parallel} = 25.1$ W/(m. K) [328].

4.2 Oscillating Behavior: Acetone

Upon impact of liquid droplets on a heated *sapphire* prism, we observe a new behavior which is completely different from impact on glass substrate. For high surface temperatures after the contact (bubbly) boiling regime [24, 273, 274, 330] and before the Leidenfrost regime (no contact between solid and liquid), we observe a striking *oscillating behavior* (see Fig. 4.2) in the wetted area. This becomes more evident by using high frame rates (300000 and 540000) for TIR recordings. We will argue how this new behavior can contribute to our understanding of Leidenfrost transition in chapter 5. In this section, several snapshots of droplet impact on sapphire prism for different liquids, T and U_0 are presented to illustrate this newly oscillating behavior. To focus on the oscillating behavior successfully detected from TIR, in these snapshots, only the bottom-view images are shown.

Fig. 4.2 shows a series of snapshots from TIR recordings for an acetone droplet impacting with moderate impact velocity, $U_0 = 1.85$ m/s, on the heated sapphire with a nearly high surface temperature, $T = 140$ °C. Each snapshot includes the real time (in μ s) during impact (the frame that shows the first contact of liquid and solid is always taken as $t = 0$). The first row shows clearly the recurring process of drying (spontaneous boiling) and rewetting (wetted areas grow toward center of impact) which illustrates a fluctuating behavior. The second and third rows (with the small time interval of 3.3μ s) show the rewetting process in more details. At later time, some fingering patterns are observed ($t = 1250 \mu$ s). These fingering patterns are not exactly as those observed on glass substrate, as they are not formed instantaneously upon impact; they are rather formed after the suppression of oscillating behavior. To observe the oscillating behavior in the following figures, one should follow the liquid-vapor interface (the boundary between dark and bright regions in snapshots).

Fig. 4.2 The oscillating behavior observed during impact of an acetone droplet on a heated sapphire prism. All inset numbers show the real time in μ s. Surface temperature is $T = 140$ °C and impact velocity is $U_0 = 1.85$ m/s. Scale bar indicates the length scale of 1 mm.

To further investigate this oscillating behavior and the possibility of its universality, we perform similar experiments for other liquids (ethanol, IPA, water and FC-72) and observe this behavior for them as well. We also study this phenomenon for different surface temperatures and impact velocities of each liquid (see Table 4.3).

Fig. 4.3 and Fig. 4.4 show similar oscillating behavior of acetone droplet at higher surface temperatures and the same impact velocity. The oscillating behavior is still observed, but due to higher surface temperature, the behavior becomes more irregular than lower T and the liquid film is evaporated at higher rate.

Fig. 4.3 Series of snapshots showing the oscillating behavior of acetone. All inset numbers show the real time in μs . Surface temperature is $T = 150^\circ\text{C}$ and impact velocity is $U_0 = 1.85\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.4 Series of snapshots showing the oscillating behavior of acetone. All inset numbers show the real time in μs . Surface temperature is $T = 160^\circ\text{C}$ and impact velocity is $U_0 = 1.85\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.5 compares the rewetting process at different surface temperatures and same impact velocity ($U_0 = 1.85$ m/s). While the rewetting process occurs regularly towards the center at lower temperature ($T = 140$ °C), it becomes more irregular as the surface temperature is increased.

Fig. 4.6 shows the oscillating behavior of acetone droplets for same surface temperature ($T = 140$ °C) and 6 different impact velocities. Here, successive snapshots (3.3 μ s interval) of a typical rewetting-evaporation process are shown. Each column gives successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Comparing the snapshots of different columns gives no significant difference between them, especially at high impact velocities. Thus, the initial kinetic energy of the droplet does not seem to influence the oscillating behavior. This will be discussed quantitatively in next chapter. In the upcoming sections, similar groups of snapshots for other liquids will be presented to show the oscillating behavior for them as well.

Fig. 4.5 Comparison of the rewetting process of acetone for different surface temperatures and same impact velocity ($U_0 = 1.85 \text{ m/s}$). Each column shows successive snapshots for the same temperature. Scale bars indicate the length scale of 1 mm. The rewetting process is observed as the three-phase contact line moves towards the center, but tends to occur more irregularly as T is increased.

Fig. 4.6 Comparison of oscillating behavior of acetone for same surface temperature ($T = 140\text{ }^{\circ}\text{C}$) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bars indicate the length scale of 1 mm.

4.3 Oscillating Behavior: Ethanol

In this section, the oscillating behavior of impact of ethanol droplets on heated sapphire prism are presented. Fig. 4.7 shows series of snapshots of ethanol droplet with $U_0 = 2.66\text{ m/s}$ and $T = 150\text{ }^{\circ}\text{C}$ indicating the oscillating behavior. Rows 3-5 include successive snapshots ($3.3\mu\text{s}$ interval) during impact. Fig. 4.8 and Fig. 4.9 show similar snapshots for the same impact velocity and $T = 160\text{ }^{\circ}\text{C}$ and $T = 170\text{ }^{\circ}\text{C}$ respectively.

Fig. 4.7 Series of snapshots showing the oscillating behavior of ethanol. All inset numbers show the real time in μs . Surface temperature is $T = 150^\circ\text{C}$ and impact velocity is $U_0 = 2.66\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.8 Series of snapshots showing the oscillating behavior of ethanol. All inset numbers show the real time in μs . Surface temperature is $T = 160^\circ\text{C}$ and impact velocity is $U_0 = 2.66\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.9 Series of snapshots showing the oscillating behavior of ethanol. All inset numbers show the real time in μs . Surface temperature is $T = 170^\circ\text{C}$ and impact velocity is $U_0 = 2.66 \text{ m/s}$. Scale bar indicates the length scale of 1 mm.

In Fig. 4.10, we compare the rewetting process at three different surface temperatures and the same impact velocity ($U_0 = 2.66 \text{ m/s}$). Fig. 4.11 shows the oscillating behavior of ethanol droplets for same surface temperature ($T = 150^\circ\text{C}$) and 5 different impact velocities. Each column gives successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. The general behavior of rewetting process seems not to depend on impact velocity.

Fig. 4.10 Comparison of the rewetting process of ethanol for different surface temperatures and same impact velocity ($U_0 = 2.66 \text{ m/s}$). Each column shows successive snapshots for the same temperature. Scale bar indicates the length scale of 1 mm.

Fig. 4.11 Comparison of oscillating behavior of ethanol for same surface temperature ($T = 150^{\circ}\text{C}$) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bar indicates the length scale of 1 mm.

4.4 Oscillating Behavior: IPA

In this section, snapshots illustrating the oscillating behavior for IPA are presented. Fig. 4.12 shows a typical case for IPA at $T = 150\text{ }^{\circ}\text{C}$ and $U_0 = 2.58\text{ m/s}$. Fig. 4.13 and Fig. 4.14 show similar snapshots for same impact velocity and $T = 160\text{ }^{\circ}\text{C}$ and $T = 170\text{ }^{\circ}\text{C}$ respectively.

Fig. 4.12 Series of snapshots showing the oscillating behavior of IPA. All inset numbers show the real time in μs . Surface temperature is $T = 150\text{ }^{\circ}\text{C}$ and impact velocity is $U_0 = 2.58\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.13 Series of snapshots showing the oscillating behavior of IPA. All inset numbers show the real time in μs . Surface temperature is $T = 160\text{ }^{\circ}\text{C}$ and impact velocity is $U_0 = 2.58\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.14 Series of snapshots showing the oscillating behavior of IPA. All inset numbers show the real time in μs . Surface temperature is $T = 170^\circ\text{C}$ and impact velocity is $U_0 = 2.58 \text{ m/s}$. Scale bar indicates the length scale of 1 mm.

In Fig. 4.15, we compare the rewetting process at different surface temperatures and same impact velocity ($U_0 = 2.58 \text{ m/s}$). The rewetting process is observed for the three temperatures similar to acetone and ethanol. Fig. 4.16 shows the oscillating behavior of IPA droplets for same surface temperature ($T = 160^\circ\text{C}$) and 6 different impact velocities. Each column gives successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Again, no qualitative difference in rewetting process, due to change in impact velocity is observed.

Fig. 4.15 Comparison of the rewetting process of IPA for different surface temperatures and same impact velocity ($U_0 = 2.58 \text{ m/s}$). Each column shows successive snapshots for the same temperature. Scale bar indicates the length scale of 1 mm.

Fig. 4.16 Comparison of oscillating behavior of IPA for same surface temperature ($T = 160^\circ\text{C}$) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bar indicates the length scale of 1 mm.

4.5 Oscillating Behavior: Water

Since boiling point and latent heat of water are much higher than the other three liquids used here (see Table 4.2), the oscillating behavior and consequently its Leidenfrost temperature would be observed at much higher surface temperatures. For instance, the oscillating behavior of water is first seen at $T = 260^\circ\text{C}$. In this section, similar snapshots of this behavior for water are presented. We used DI water for all the experiments in this research.

Fig. 4.17 shows series of snapshots of water at $T = 260^\circ\text{C}$ and $U_0 = 2.78\text{ m/s}$; the previously described oscillating behavior is observed for water as well. The rewetting process can be tracked by successive snapshots in rows 3-5. Fig. 4.18 and Fig. 4.19 show similar series for the same impact velocity and $T = 280^\circ\text{C}$ and $T = 300^\circ\text{C}$ respectively.

Fig. 4.20 compares the rewetting process in details for three surface temperatures and same impact velocity. Fig. 4.21 compares the rewetting process of water for the same surface temperature and 6 different impact velocities. The behavior seems not to vary with impact velocity.

Fig. 4.17 Series of snapshots showing the oscillating behavior of water. All inset numbers show the real time in μs . Surface temperature is $T = 260^\circ\text{C}$ and impact velocity is $U_0 = 2.78\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.18 Series of snapshots showing the oscillating behavior of water. All inset numbers show the real time in μs . Surface temperature is $T = 280^\circ\text{C}$ and impact velocity is $U_0 = 2.78 \text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.19 Series of snapshots showing the oscillating behavior of water. All inset numbers show the real time in μs . Surface temperature is $T = 300^\circ\text{C}$ and impact velocity is $U_0 = 2.78 \text{ m/s}$. Scale bar indicates the length scale of 1 mm.

Fig. 4.20 Comparison of the rewetting process of water for different surface temperatures and same impact velocity ($U_0 = 2.78 \text{ m/s}$). Each column shows successive snapshots for the same temperature. Scale bar indicates the length scale of 1 mm.

Fig. 4.21 Comparison of oscillating behavior of water for same surface temperature ($T = 260^\circ\text{C}$) and different impact velocities. Each column shows successive snapshots for the same impact velocity. Snapshots at each row belong to the same time after impact. Scale bar indicates the length scale of 1 mm.

4.6 Oscillating Behavior: FC-72

To further investigate the universality of the oscillating behavior, we even carried out experiments for FC-72, a liquid with much smaller physical properties than the other tested liquids: boiling point, $T_b = 56\text{ }^\circ\text{C}$; surface tension, $\sigma = 10\text{ mN/m}$ and latent heat $h_{fg} = 88\text{ kJ/kg}$. The result as a series of snapshots is shown in Fig. 4.22. It is quite fascinating that despite its low boiling point and small surface tension, the oscillating behavior was observed for FC-72 as well. We observed this behavior for FC-72 for all impact velocities ($0.62 - 2.73\text{ m/s}$) but for limited surface temperatures. It should be mentioned that since the empirical data of the temperature-dependent properties of FC-72 were not available in the literature, our study of droplet impact of this liquid is limited to this qualitative observation.

Fig. 4.22 Series of snapshots showing the oscillating behavior of FC-72. All inset numbers show the real time in μs . Surface temperature is $T = 100\text{ }^\circ\text{C}$ and impact velocity is $U_0 = 2.59\text{ m/s}$. Scale bar indicates the length scale of 1 mm.

The experimental results presented in sections 4-3 to 4-7 showed that the newly observed oscillating behavior could be a universal phenomenon. Therefore, we consider this behavior to be a new boiling regime and call it *oscillating boiling regime*. The oscillating boiling regime connects the bubbly boiling regime and the Leidenfrost regime. Detecting, quantifying and investigating this new regime can help us understand the transition to Leidenfrost regime.

4.7 Frequency of Oscillations

To quantify the striking oscillating behavior that was observed for all the tested liquids (acetone, ethanol, IPA and water), we measured the frequency of these oscillations and investigated its dependence on liquid properties, surface temperature and impact velocity. Hence, the temporal behavior of the mean image intensity (grayscale value) over a square zone (10×10 pixels which is less than 5% of the wetting area) at the center point of the impact for each experiment was considered for frequency measurement.

A typical plot of such an oscillating behavior is shown in Fig. 4.23. This plot was obtained for impact of a water droplet with $U_0 = 1.88$ m/s and at surface temperature $T = 260$ °C (see third column of Fig. 4.21 for the corresponding snapshots for this case). The figure shows that this new boiling regime has indeed an *oscillating* nature with a finite frequency. The frequency of this oscillation was simply measured to be 12711 Hz. The measurement of the frequency for the oscillating regimes of each T, U_0 and liquid was done similarly.

Fig. 4.23 Fluctuations of image intensity at the center point vs. time for a typical case. Liquid is water, impact velocity is $U_0 = 1.88$ m/s and surface temperature is $= 260$ °C.

Fig. 4.24 shows the variation of frequency of oscillations with impact velocity for different surface temperatures for the four liquids (acetone, ethanol, IPA and water). It can be seen that the frequency does not depend on impact velocity, but increases with surface temperature for all liquids.

Fig. 4.24 Frequency of oscillations vs. impact velocity for different surface temperatures: a) acetone; b) ethanol; c) IPA and d) water. It can be seen that the frequency is almost independent of impact velocity.

Since the frequency is almost independent of impact velocity for all liquids, an averaged frequency (for different V_0) was then calculated. We plotted this average frequency vs. surface temperature for all the liquids in Fig. 4.25. The plot shows, interestingly, that the frequency of the oscillations increases almost linearly with surface temperature. The range of oscillation's frequency for acetone is 15 – 33 kHz, ethanol, 15 – 24 kHz, IPA, 14 – 31 kHz and water, 13 – 23 kHz. From Fig. 4.25, for the same surface temperature:

$$f_{\text{Ace}} > f_{\text{IPA}} > f_{\text{Eth}}. \quad 4.1$$

Also the frequency of oscillations for water (while for higher surface temperatures) is smaller than the other three liquids.

Fig. 4.25 Dependence of averaged frequency of oscillations on surface temperature for all liquids.

4.8 Vapor Thickness

Total Internal Reflection (TIR) technique enables us to measure the vapor thickness during droplet impact on the heated surface. Using the equations for the intensity of the evanescent wave (decaying with distance from the surface), Eq. 3.3, and the penetration depth, Eq. 3.4, we can find the equation for measuring the vapor thickness [17, 25, 273]:

$$h_v = -d_p \ln \left(1 - \frac{I_r}{I_0} \right) \quad 4.2$$

where I_0 is the intensity before the droplet contacts the surface, I_r is the reflected light intensity and d_p is the penetration depth. The high-speed camera (SA-X2, Photron) connected to a long-distance microscope was used to collect the reflected light from the impacted surface. By considering reflection of the light from the droplet, Shirota *et al.* [273], presented a relation (from optics geometry) for calculating the true incident angle (with prism's top surface), α_i , which consequently leads to a more accurate estimation of the penetration depth. Table 4.4 gives the incident angle (based on their relation) and penetration depth for the four liquids used in our experiments on heated sapphire. For calculations of vapor thickness, we used the images from TIR recordings without background subtraction.

Table 4.4 Incident angle and penetration depth for all the liquids studied in this chapter.

Liquid	Acetone	Ethanol	IPA	Water
Property				
Incident angle	48.3°	48.5°	48.3°	46.6°
Penetration depth (nm)	58.4	58.0	59.3	63.1

4.9 Determination of Leidenfrost Temperature

By using Eq. 4.2, we measured the vapor thickness (and its evolution in time) at every point of impact region for surface temperatures close to Leidenfrost regime. This was done for all liquids and impact velocities. These estimations were used for determining the temperature at which the system is in Leidenfrost regime.

The relation between h_v and d_p affects the intensity I_r of the light reflected to the sapphire medium as follows: for $h_v \gg d_p$, $I_r \approx I_0$, the maximum value, whereas for $h_v \sim h$, I_r is attenuated due to optical coupling and loss to the liquid medium.

For a wetted impacted surface, i.e., $h_v = 0$, the condition for α_i ensures that the incident light refracts into the liquid medium, corresponding to a minimum value of I_r . We calculated h_v at each point on the impacted surface based on the recorded light intensity normalized by the maximum values (Eq. 4.2). Based on these estimations, we determine if a surface point is wetted or dry by comparing h_v with the surface roughness $h_r \approx 10 \text{ nm}$ ¹: $h_v < h_r$ or $h_v > h_r$ for wetted and dry points, respectively. We calculate the wetted area A_w based on the total number of wetted points, thereby constructing the dependence of A_w on time. An impact experiment is categorized in the *Leidenfrost regime* if $A_w = 0$ during the entire impact duration.

To illustrate this in a simple manner, we consider N_t to be the total number of the points in the impact region with vapor thickness $h_v < h_r = 10 \text{ nm}$, i.e., a wetted spot (a point is a pixel; its wetted area is simply $A_w = \text{PS}^2$ where PS is the pixel size: $\text{PS} \sim 20 \mu\text{m}$).

Fig. 4.26 shows the variation of total number of the wetted points (equivalent to wetted area) with time for a typical case of impact of acetone droplet at two different surface temperatures and same impact velocity: At $T = 170^\circ\text{C}$ (Fig. 4.26(a)), there are numerous number of points ($N_t \neq 0$) in the impact region (and during the entire impact time) where the liquid wets the surface. Therefore, there is still contact between liquid and solid and the system has not reached the Leidenfrost regime yet. At higher temperature ($T = 190^\circ\text{C}$) however, no wetted point ($N_t \neq 0$) on the solid during the entire impact time is detected (Fig. 4.26(b)). Hence, the system is considered to be in Leidenfrost regime. The plots also include snapshots and their corresponding time which show typical behavior of the impact. Therefore, we are able to determine the Leidenfrost temperature for all of our experiments. They will be used for constructing the final phase diagrams of impact behavior.

In next chapter, we will present a proposed theory for transition to Leidenfrost regime, based on the experiments described in this chapter.

¹ Sapphire roughness is 10 nm [273].

Fig. 4.26 Total number of the wetted points vs. time at two surface temperatures. a) At $T = 170^\circ\text{C}$, since $N_t \neq 0$, there is still contact between liquid and solid and b) At higher temperature ($T = 190^\circ\text{C}$), there is no wetted point on the substrate during the entire impact time, therefore the system is in Leidenfrost regime. Insets show typical snapshots of the impact. Liquid is acetone and impact velocity is $U_0 = 2.59$ m/s. Scale bars indicate the length scale of 1 mm.

Chapter 5:

A Theory for Transition to Leidenfrost

In this chapter, based on the experimental observations reported in previous chapter, we describe the oscillating boiling behavior in more details and propose a theory for transition to Leidenfrost regime. We finally compare the predicted values of dynamic Leidenfrost temperature of different liquids with those of the experiments from the present and past studies.

5.1 Oscillating Boiling: A New Boiling Behavior

Research efforts aiming at understanding the Leidenfrost phenomenon have mostly focused on the case of *static* Leidenfrost droplets, i.e., droplets approaching a heated surface with negligible or small initial velocity and subsequently floating on the surface. The floating mechanism in this case has been studied in great details: the vapor flow between a floating droplet and a heated surface generates a viscous stress sufficient to sustain the droplet's weight [56, 57, 185, 331]. Similar hydrodynamical arguments are also capable of explaining the bouncing behavior without contact of droplets falling on an unheated smooth surface with small velocity. Here the closing gap between a droplet and the unheated surface generates a gas flow, inducing a viscous stress that keeps the droplet from touching the surface [7]. Although these theories have been successfully used to explain the floating mechanism in the static case, they have been predicated on hydrodynamical arguments of the gas/vapor flow in the gap between the liquid and the solid, thus precluding reference to the contact boiling behavior and its role in the Leidenfrost transition. As a result, these theories cannot be used to reveal the connection between the Leidenfrost regime and the contact boiling regime. Here we show that the transition to the Leidenfrost regime occurs as a result of a competition between two competing processes at the liquid-solid interface: separation of liquid from the heated surface due to localized boiling and rewetting. The boiling process becomes dominant with increasing surface temperature and completely overwhelms the other process at temperature close to the Leidenfrost temperature. We provide direct observations of these processes and description of the underlying physics to elucidate their contributions to the Leidenfrost transition. To this end, we develop theoretical arguments to predict the Leidenfrost transition and provide experimental validation of this prediction.

As mentioned in chapter 4, for these series of experiments we use four different liquids: acetone, ethanol, isopropyl alcohol (IPA) and water. For each liquid, we vary the impact velocity U_0 of droplets and keep the droplet size fixed (see Table 4.3 for velocity range and droplet size for each liquid). Thus, the diversity of tested liquids, as well as the wide ranges of impact velocity and surface temperature, emphasize the generality of our experimental results.

The bottom-view recordings, coupled with the total internal reflection (TIR) technique (see Chapters 3 and 4 for more details), provided direct observations of the surface's wetted areas at high temporal- and spatial-resolutions, $3\mu\text{s}$ and $20\mu\text{m}$,

respectively. In each recorded grayscale image, the wetted areas appear dark and are well contrasted with the dry ones (Fig. 5.1(a) and (b)).

We measure the total wetted area A_w as a function of time t from the TIR recording of each impact experiment. For all impact velocities, liquids, and temperatures close to the Leidenfrost transition, we discover a new boiling behavior (see chapter 4) in which A_w oscillates with high frequency in the range 13 – 32 kHz; its repeating pattern typically consists of two distinct stages: an abrupt drop and a subsequent increase in A_w (Fig. 5.1(c)). In the first stage, exemplified in Fig. 5.1(a), tiny dry spots first appear spontaneously and rather uniformly in the wetted area, then expand and merge to create larger dry areas. This suggests that the decrease in A_w is primarily caused by *heterogeneous boiling*, a process of forming the vapor phase from the liquid phase on a solid surface at lower temperature compared to boiling in liquid (*homogeneous boiling*) [186, 332]. In the second stage, however, the wetted areas merge and invade the dry ones; the invading direction is perpendicular to the three-phase contact line signifying a rewetting process (Fig. 5.1(b)). On this basis, we postulate that the heterogeneous boiling and rewetting processes are fabric of the new boiling behavior, now termed as *oscillating boiling*; the rapid fluctuation of A_w is resulted from alternate domination of one process over the other. Spontaneous boiling pushes the system to Leidenfrost, while rewetting opposes it.

Fig. 5.1 Wetted area in the oscillating boiling regime. The liquid is acetone, impact velocity is $U_0 = 2.74 \text{ m/s}$ and surface temperature is $T = 150 \text{ }^\circ\text{C}$. Snapshots showing a) The boiling process and b) The rewetting process; c) Wetted area A_w vs. t in the early stage of a droplet impacting on a heated sapphire surface and d) Wetted area A_w vs. t during the entire impact process; the shaded area corresponds to the wetted area shown in (c). Inset snapshots show side-view (upper panel) and bottom-view (lower panel) of the impact at different times. All scale bars indicate the length scale of 1mm.

5.2 Heterogeneous Nucleation

In this section, we analyze the heterogeneous boiling process. On an ideally flat surface immersed in a liquid at temperature T_l and pressure P_l , vapor bubbles grow from embryos, or tiny vapor pockets in the form of spherical caps attaching to the surface (Fig. 5.3(a)).

For an embryo to grow into a vapor bubble, the pressure difference across the vapor-liquid interface must overcome the Laplace pressure. Equivalently, its radius must be larger than a critical value r_c [186]:

$$r_c = \frac{2\sigma_{lv}F}{P_v - P_l} \quad 5.1$$

where σ_{lv} is the surface tension of liquid-vapor interface and F is a correction factor to account for the partial spherical shape of the embryo with contact angle θ (Fig. 5.3(a)) [186]:

$$F = 0.5 + 0.75 \cos \theta - 0.25 \cos^3 \theta, \quad 5.2$$

Here, the vapor pressure P_v inside the embryo depends on T_l and P_l as follows:

$$P_v = P_{sat}(T_l) \exp \left\{ \frac{v_l [P_l - P_{sat}(T_l)]}{RT_l} \right\} \quad 5.3$$

where v_l is the specific volume of the liquid ($v_l = 1/\rho_l$ where ρ_l is the liquid density), $P_{sat}(T_l)$ is the saturation pressure at T_l , and the liquid pressure P_l is approximated using the atmospheric pressure ($P_l = P_0 + 0.5\rho U_0^2$). Thus, the critical radius (by assuming the bubble embryo to have a spherical shape and to be in thermodynamic equilibrium with surrounding liquid) can be rewritten as:

$$r_c = \frac{2\sigma_{lv}F}{P_{sat}(T_l) \exp \left\{ \frac{v_l [P_l - P_{sat}(T_l)]}{RT_l} \right\} - P_l} \quad 5.4$$

Owing to the very small size of embryos, we assume that they are well within the thermal boundary layer, or the temperature of liquid surrounding embryos is $T_l \approx T$. If we denote J the generation rate per unit area of embryos having radius r_c , J can be readily calculated using the thermodynamic conditions of the liquid [186]:

$$J = \frac{\rho_{N,l}^{2/3} (1 + \cos \theta)}{2F} \left(\frac{3F\sigma_{lv}}{\pi m} \right)^{1/2} \exp \left\{ \frac{-16\pi F\sigma_{lv}^3}{3k_B T_l [\eta P_{sat}(T_l) - P_l]^2} \right\} \quad 5.5$$

where

$$\eta = \exp \left\{ \frac{v_l [P_l - P_{sat}(T_l)]}{RT_l} \right\}, \quad 5.6$$

Here:

$$\rho_{N,l} = \frac{N_A}{v_l \bar{M}} \quad \text{Number density of liquid molecules per unit volume} \quad 5.7$$

$$m = \frac{\bar{M}}{N_A} \quad \text{Mass of one molecule (kg/molcule)} \quad 5.8$$

$$\bar{M} \quad \text{Molecular mass (kg/kmol)} \quad 5.9$$

$$N_A = 6.02 \times 10^{26} \quad \text{Avogadro constant (1/kmol)} \quad 5.10$$

$$k_B = 1.38 \times 10^{-23} \quad \text{Boltzmann constant (J/K)} \quad 5.11$$

$$R = \frac{\bar{R}}{\bar{M}} \quad \text{Specific gas constant (J/(kg. K))} \quad 5.12$$

$$\bar{R} = 8314.4 \quad \text{Universal gas constant (J/(K. kmol))} \quad 5.13$$

Fig. 5.2 shows the plot of J (from Eq. 5.5) for our four tested liquids. It should be noted that v_l , σ_{lv} and P_{sat} all depend strongly on liquid temperature. The temperature-dependent values of these parameters can be obtained from the reported data for each liquid. For θ of each liquid, we use the equilibrium contact angle measured for the impact of droplet of the corresponding liquid on an unheated substrate.

Fig. 5.2 Comparison of the variation rate of generation of embryos J (from Eq. 5.5) and with surface temperature T for acetone, ethanol and IPA. The corresponding plot for water is shown as inset due to its higher range of surface temperature.

For vapor embryos generated by rate J , the duration to populate a unit area by bubbles of radius r_c is:

$$\tau_h = \frac{1}{J r_c^2} \quad 5.14$$

In Fig. 5.3(b), we show the plots of r_c and $\log_{10}(\tau_h)$ versus T for acetone at the vicinity of Leidenfrost transition (see Fig. 5.5 and Fig. 5.6 for similar plots of r_c and τ_h for ethanol, IPA and water).

Fig. 5.3 Heterogeneous boiling and rewetting processes. a) Vapor embryos in a heterogeneous boiling process; b) Critical size r_c of a vapor embryo (blue solid line) and evaporation time τ_h (red dash line) versus surface temperature T . Inset: schematic of a vapor embryo on a flat surface surrounded by liquid and c) Snapshots showing the rewetting process for acetone droplets impacting with velocity $U_0 = 2.3$ m/s at low temperature (left panel) and high temperature (right panel); arrows indicate the motion of the three phase contact line. Scale bars show the length scale of 0.3 mm.

The reduction in r_c indicates a decrease in thickness of the vapor layer. Remarkably, we also observe a sharp drop in τ_h , implying that the required duration to generate a vapor layer becomes extremely small as T approaches the Leidenfrost transition. For instance, at $T = 180^\circ\text{C}$, $\tau_h = 1.6$ ns. This highlights the dominant role of the boiling process: a vapor layer is created immediately to separate the liquid from the surface if the liquid is sufficiently heated. However, we note that the boiling process takes away heat from the surface, causing its temperature to decrease. As a result, the heterogeneous boiling process is forced to stop if either the surface temperature is not sufficiently high, or the solid has low thermal conductivity and heat capacity.

As soon as the heterogeneous boiling process stops, the liquid in neighboring wetted areas merges and rewets the dry areas (Fig. 5.3(c)). From the contact line's motion, indicated in Fig. 5.3(c), we measured rewetting velocity V_{re} for different liquids and impact velocities U_0 . Generally, V_{re} is insensitive to U_0 and increases with surface temperature T (see Fig. 5.4(a) and (b)).

In order to understand this behavior, we note that a dry area resulted from heterogeneous boiling is covered by a vapor layer of thickness $h \approx 2r_c$. Since the tested liquids have small viscosities, the rewetting process is then driven by capillary pressure $\sigma/2r_c$ and resisted only by inertia $\rho_l V_{\text{re}}^2$. Here, both the surface tension σ and density ρ_l are functions of surface temperature T . Balancing the capillary pressure and inertia gives an estimate for the rewetting velocity:

$$V_{\text{re}} \sim \left(\frac{\sigma}{2\rho_l r_c} \right)^{\frac{1}{2}} = \left(\frac{P_v - P_l}{4\rho_l F} \right)^{\frac{1}{2}} \quad 5.15$$

In Fig. 5.4(a) and (b), we show several plots of V_{re} versus T for all tested liquids with impacting velocity U_0 varying from 0.62 – 2.74 m/s. We observe an excellent agreement with experimental results for acetone, ethanol, and IPA, but an overestimate for water.

We attribute possible surface temperature decrease during impact to such discrepancy [293]: owing to water's high latent heat and heat capacity compared to those of other liquids, the surface temperature in actuality may lessen considerably during impact. This causes an overestimation of the liquid temperature T_l , as well as the vapor pressure $P_v(T_l)$, leading to an overestimation of V_{re} (Eq. 5.15). Nonetheless, these results strongly suggest that the measured rewetting velocity is not only consistent with our theoretical arguments for the rewetting process, but also confirm that the vapor layer thickness is well described by the critical size of embryos.

We conclude that the liquid-vapor dynamics in the oscillating boiling regime consist of two main processes: heterogeneous boiling followed by capillary-inertial rewetting.

Fig. 5.4 a) Rewetting velocity V_{re} versus surface temperature T for acetone droplets for different impact velocities; solid line shows the prediction according to Eq. 5.15 and b) V_{re} vs. T for different impact velocities for ethanol, IPA, and water.

Fig. 5.5 compares the variation of r_c with T for our four tested liquids (acetone, ethanol and IPA in the main plot and water as inset due its higher range of surface temperature). It is seen that for the same surface temperature, acetone has smaller r_c than ethanol and IPA. For water, on the other hand, r_c has weaker dependency on T than the other three liquids. Fig. 5.6 shows the change of evaporation time (τ_h) with surface temperature for four liquids.

Fig. 5.5 Comparison of the variation of vapor embryo's critical size r_c (from Eq. 5.4) with surface temperature T for acetone, ethanol and IPA. The corresponding plot for water is shown as inset due to its higher range of surface temperature.

Fig. 5.6 Variation of evaporation time τ_h (from Eq. 5.14) with surface temperature T for a) acetone; b) ethanol; c) IPA and d) water.

5.3 A Proposed Theory for Transition to Leidenfrost

Based on the arguments given in previous sections and fundamentals of heterogeneous nucleation, here we propose our theory for transition to the Leidenfrost regime.

We derive an expression to determine the *dynamic* Leidenfrost transition, which marks the lower bound of the Leidenfrost regime for impacting droplets; the surface temperature at this transition is termed as *dynamic* Leidenfrost temperature T_L .

We argue that at $T = T_L$, the upward speed of vapor generation must be comparable to the downward speed U_0 of liquid. Since the heterogeneous boiling process generates a vapor layer of thickness $h = 2r_c$ during the time τ_h , the upward speed of vapor generation is $V_v \sim 2r_c/\tau_h$. Thus the dynamic Leidenfrost temperature T_L satisfies the following condition:

$$U_0 = \frac{2r_c(T_L)}{\tau_h(T_L)} \quad 5.16$$

This condition implicitly describes the dependence of T_L on U_0 and the liquid properties and we use it to determine the Leidenfrost transition for our tested liquids.

5.3.1 Phase Diagrams

Now that we have been able to predict the Leidenfrost temperature, we are capable of constructing our final phase diagram for droplet impact on heated surfaces. We calculate T_L for four tested liquids using Eq. 5.16 and in Fig. 5.7, we show a plot of T_L vs. U_0 (solid line) together with a phase diagram for acetone consisting of three different characteristic behaviors: Leidenfrost, oscillating boiling, and bubbly boiling [24, 330]. We identify and categories these behaviors using TIR recordings of numerous impact experiments.

We note that although both the oscillating boiling and bubbly boiling behaviors allow solid-liquid contact, only the former exhibits high-frequency switching between the heterogeneous boiling and rewetting processes, while the boiling dynamics of the latter are much slower owing to low surface temperature.

Fig. 5.7 Phase diagram of acetone based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes three predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16, the one measured from TIR recordings (red dashed line) and the one detected from side-view images (blue dashed-dotted line).

In the phase diagram, the boundary separating the Leidenfrost and oscillating boiling regimes indicates T_L determined experimentally from TIR recordings (dashed line). It is evident that the experimental values of T_L increases monotonically with U_0 and reaches a plateau value (we observed similar behavior for impact of droplets on glass substrate in chapter 3). For instance, for acetone (Fig. 5.7), the plateau value of

Leidenfrost temperature is $T_L = 190^\circ\text{C}$ at $U_0 = 1.33\text{ m/s}$, consistent with earlier studies [24, 273].

We also show the Leidenfrost temperature determined by *side-view* recordings (dashed-dotted line), a method incapable of directly detecting wetted areas during impact, but instead relying on ejection of small droplets as indication of wetted areas [24]. The discrepancy between values of T_L determined by side-view recordings and ones by TIR technique, although not significant, is therefore expected. Nevertheless, the plateaued values of T_L measured by both techniques are in remarkable agreement with the predicted value $T_L = 180^\circ\text{C}$, highlighting that the proposed prediction for T_L is valid for impacts at high velocity.

We emphasize that our prediction for T_L is not applicable for the case of low impact velocity, e.g., $U_0 < 1.3\text{ m/s}$, where liquid-solid separation is resulted from the viscous stress induced by air/vapor flows. This hydrodynamical mechanism has been shown responsible for phenomena such as dimple formation [24, 140], bubble entrapment [23], or bouncing of droplets at small impact velocity even for unheated surfaces [7]. Thus the deviation of T_L calculated using Eq. 5.16 from the experimental ones at low impact velocity is indicative of the region where hydrodynamical effects become the dominant mechanism for liquid-solid separation. Similar phase diagrams for ethanol, IPA and water are shown in Fig. 5.8-Fig. 5.10.

Fig. 5.8 Phase diagram of ethanol based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes three predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16, the one measured from TIR recordings (red dashed line) and the one detected from side-view images (blue dashed-dotted line).

Fig. 5.9 Phase diagram of IPA based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes three predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16, the one measured from TIR recordings (red dashed line) and the one detected from side-view images (blue dashed-dotted line).

Fig. 5.10 Phase diagram of water based on observations from TIR. The regimes include bubbly boiling (blue diamond) and the newly observed oscillating boiling (red square) before transition to Leidenfrost (black triangle). The phase diagram includes two predictions for Leidenfrost transition: the one from theoretical prediction (black solid line) from Eq. 5.16 and the one measured from TIR recordings (red dashed line). Note that for water, we were not able to detect the Leidenfrost transition from side-view recordings.

5.3.2 Dynamic Leidenfrost Temperature

Fig. 5.11 shows the change in Leidenfrost temperature, predicted from our proposed theory (Eq. 5.16), with impact velocity for the four tested liquids in this study: acetone (dashed line), ethanol (solid line), IPA (dotted line) and water (dash-dot line). For all of these liquids, the theoretically predicted Leidenfrost temperature is seen to be independent of impact velocity, in particular at high impact velocities. Also, comparing T_L for different liquids gives: $T_{L_{Ace}} < T_{L_{IPA}} < T_{L_{Eth}} \ll T_{L_{Wat}}$. It is noteworthy to mention that plateaued Leidenfrost temperature for ethanol on glass substrate ($T_L \sim 300^\circ\text{C}$) is much larger than that on sapphire substrate ($T_L \sim 210^\circ\text{C}$) which can be attributed to the significant difference in their thermal conductivities. It should be mentioned that since the surface tension σ , specific volume v_l , and saturation pressure P_{sat} of each liquid depend strongly on the liquid temperature T_l , we have interpolated their temperature-dependent values from the reported data for each liquid: acetone, ethanol and water [329] and IPA [333-335] (see Appendix B for the temperature-dependent properties of the liquids). We did not apply our theory for FC-72, since the empirical data of its temperature-dependent properties were not available in the literature.

Fig. 5.11 Predicted Leidenfrost temperature (Eq. 5.16) vs. impact velocity for the four tested liquids in this study: acetone (dashed line), ethanol (solid line), IPA (dotted line) and water (dash-dot line).

To further evaluate our theoretical prediction for dynamic Leidenfrost temperature, we applied our theory for a few other liquids for which the dependence of physical properties on temperature were available in the literature and studies of their Leidenfrost temperature have been carried out. These liquids (and the references used for their temperature dependent properties) include heptane, benzene and methanol [329], HFE7100 [336, 337] and HFE7300 [337, 338] (see Appendix B).

We now present the final results of this research. Table 5.1 gives the predicted values of dynamics Leidenfrost temperature from our theory (Eq. 5.16) for all 9 different liquids along with the experimental values measured either in this research (from both side-view and TIR) or other studies. However, the reported data for *dynamics* Leidenfrost temperature were only available for six of them (as shaded in Table 5.1). For the rest, only the static Leidenfrost temperature was reported.

Table 5.1 Predicted Leidenfrost temperature from our theory (Eq. 5.16) for different liquids. The shaded rows show the ones for which the experimental values of dynamic Leidenfrost temperature were available in the literature.

Liquid	T_L^P (°C) Predicted(Eq. 5.16)	T_L^M (°C) Measured	Remarks	Substrate
Acetone	180.2	170	Present study (Side-View)	Sapphire
		190	Present study (TIR)	Sapphire
Ethanol	209.9	180	Present study (Side-View)	Sapphire
		200	Present study (TIR)	Sapphire
		212.5 [273]	$U_0 = 0.4 - 4.3 \text{ m/s}^1$	Sapphire
		180 [272]	$U_0 = 0.6 \text{ m/s}$	Sapphire
IPA	203.9	190	Present study (Side-View)	Sapphire
		200	Present study (TIR)	Sapphire
Water ²	316.8	420	Present study (TIR)	Sapphire
		410 [24]	$U_0 = 0.13 - 4.2 \text{ m/s}^1$	Silicon
		250 [260]	$U_0 = 0.86 \text{ m/s}$	Stainless Steel
		200 [272]	$U_0 = 0.6 \text{ m/s}$	Sapphire
		335 [293]	$U_0 = 0.44 \text{ m/s}$	Sapphire
Heptane	215.2	210 [339]	$U_0 = 2 \text{ m/s}$	Stainless Steel
		210 [266]	$U_0 = 0.4 \text{ m/s}$	Stainless Steel
		200 [76]	$U_0 = 0.93 \text{ m/s}$	Stainless Steel
HFE7100	166	140 [339]	$U_0 = 2 \text{ m/s}$	Stainless Steel
Methanol	205.4	169.9 [340]	Static Leidenfrost	Ceramic
Benzene	247.1	175 [185]	Static T_L (polished surface)	Aluminum
		220 [185]	Static T_L (particle blasted)	Aluminum
		218 [185]	Static T_L (rough sanded)	Aluminum
HFE7300	205.2	167.9 [341]	Static Leidenfrost	Stainless Steel

¹ We averaged the reported T_L over the range of impact velocity.

² Only the dynamic Leidenfrost temperatures of water are reported here. There are numerous reported values for static one in the literature as well.

In Fig. 5.12, we show a plot comparing the experimentally measured *dynamic* Leidenfrost temperature (T_L^M) to the predicted one (T_L^P) for six different liquids: acetone, ethanol, IPA, water, heptane, and HFE7100 (see Table 5.1 for numeric values). These liquids have broadly different thermal and physical properties.

Fig. 5.12 Comparison between the dynamic Leidenfrost temperatures obtained from theory and experiments for various liquids: acetone (square), ethanol (circle), IPA (diamond), water (pentagram star), HFE7100 (right triangle), and heptane (hexagram star). The empty markers on the diagonal solid lines indicate theoretical values of T_L for different liquids. All upward solid triangles mark the measured Leidenfrost temperature from TIR, while the downward solid ones mark those measured from side-view recordings. All other solid markers show the dynamic Leidenfrost temperature experimentally determined in other studies [24, 76, 260, 272, 273, 293, 339]. The shaded area indicates $\pm 15\%$ deviation from the theoretical values.

For each of our four tested liquids, we use the plateaued value of T_L for the comparison shown in Fig. 5.12. The plot also consists of numerous experimental datasets from previous studies in which measurements of the dynamic Leidenfrost temperature were reported; most of them utilized the side-view technique to determine T_L . Except for the case of water, we observe that all the experimental values of T_L are consistent, within 15% deviation, with the predicted ones; typically the TIR measured values are closer to theory than those obtained by the side-view technique.

We note that the discrepancy between the theory and experimental data in the case of water may be originated from the so-called *vapor cooling* effect, whereby the high heat capacity of water causes the surface to cool down quickly, making it difficult to maintain the Leidenfrost state during the entire impact time [273]. This effect is indeed

supported by our overestimate of the rewetting velocity for water (Fig. 5.4 (b)), as well as the vastly disparate reported values for T_L , which range from 200 °C to 410 °C. An analytical prediction of T_L for water, therefore, must be derived on the ground of non-negligible fluctuation in surface temperature and therefore merits further investigation.

It is noteworthy that Eq. 5.16 does not have any adjustable parameters and provides remarkable results in predicting the Leidenfrost temperature. We note that while the surface temperature is assumed constant in our simplified theory, it fluctuates considerably in reality as a result of two competing heat transfer processes: one from the solid surface to the liquid, and the other from the solid bulk to the surface. In addition, variations in surface roughness and impact velocity may contribute to discrepancy between T_L^M and T_L^P .

5.4 Discussion

From our experimental measurements and analysis of T_L , we infer that the mechanisms for Leidenfrost transition are fundamentally different at two extremes of velocity. The mechanism at low impact velocity is mainly associated with hydrodynamical effects and has long been studied. By contrast, the mechanism for Leidenfrost transition at high impact velocity is dominated by the heterogeneous boiling process. The domination of boiling over hydrodynamical effects is the main cause of the plateau formation in T_L i.e., T_L becomes independent of U_0 at high impact velocity. The heterogeneous boiling process also causes the oscillating boiling behavior, i.e., high-frequency fluctuation of the wetted area due to alternative domination of either heterogeneous boiling or rewetting over the other.

The transition to Leidenfrost regime, therefore, reduces to sustaining capability of the heterogeneous boiling process. For practical applications, the two major mechanisms must be considered to account for the Leidenfrost transition. We conclude by highlighting that our findings offer a theoretical framework to treat the Leidenfrost transition, a crucial step towards achieving complete control of occurrence of the Leidenfrost phenomenon.

Chapter 6:

Conclusions and Future Work

6.1 Conclusions

We have investigated the transition to the Leidenfrost regime by systematically studying the boiling behaviors of droplets impacting on heated surfaces of materials having distinct thermal conductivities, i.e., glass and sapphire. We have observed in details the boiling activities at the solid-liquid interface using the Total Internal Reflection (TIR) technique, thereby obtained physical insights into characteristic boiling behaviors of the liquid during impact. Most importantly, we have established a mechanism of the Leidenfrost transition in the limiting case of high impact velocity and high thermal conductivity of impacted surfaces.

At the limit of low thermal conductivity, i.e., in the case of glass surfaces, the surface temperature at the impacted point is decreased substantially during impact. For a surface with initial temperature much lower than the Leidenfrost temperature, the liquid spreading is mainly driven by its initial inertia. As the surface temperature approaches the Leidenfrost temperature, the boiling behavior transitions into the fingering boiling regime in which the wetting patterns take the form of radial fingers. The shape change of the wetted patterns, which is caused either by liquid spreading or evaporation, mostly occurs at the three-phase contact line. Within the wetted area, bubble nucleation occurs in isolation as a result of the temperature drop. This is opposite to the boiling behavior on surface having high thermal conductivity: spontaneous boiling is repeatedly sustained after wetting because the surface temperature drops insignificantly. We have also quantitatively analyzed the wetted area, the total length of contact line, and their dependences on surface temperature and impact velocity. The results show that the Leidenfrost transition, if quantified by the wetted area, occurs continuously instead of abruptly as commonly thought.

At the limit of high thermal conductivity, i.e., in the case of sapphire surface, the surface temperature decreases insignificantly during impact and can be considered constant when considering the impacts of most liquids except water. We have carried out a comprehensive study of impact of various liquids, including ethanol, IPA, acetone, FC-72, and water. We have observed a new boiling behavior called *oscillating boiling* that preceded the Leidenfrost regime. Detailed TIR observations reveal that the fabric of the boiling behavior in this regime is a repetitive two-stage process: spontaneous nucleation and rewetting. With increasing surface temperature, the former one becomes increasingly dominant in causing solid-liquid separation, while the latter one opposes it with declining intensity.

We have estimated the velocity of the three-phase contact line during the rewetting process based on the assumption that rewetting is driven by capillary pressure and resisted by liquid inertia. The estimated velocity is consistent with the measured ones for all the liquids except for water. The discrepancy between the estimated and the measured wetting velocities in the case of water is resulted from its high latent heat and heat capacity, which effectively cause a much larger drop in surface temperature compared to that for other liquids.

We have derived a theoretical prediction of the *dynamic* Leidenfrost temperature. We compared our predictions with the measured Leidenfrost temperature both from side-view and TIR recordings for various liquids and found very good agreement between our theoretical predictions and measurements. From our theoretical and experimental study of dynamic Leidenfrost temperature, we have deduced that the mechanisms for Leidenfrost transition at the low and high velocity limits are fundamentally different: the former is mainly associated with hydrodynamical effects, while the latter is dominated by the heterogeneous boiling process. The domination of boiling over hydrodynamical effects is the main cause of the plateau formation in T_L at high impact velocity. Furthermore, the heterogeneous boiling process is responsible for the oscillating boiling behavior. The transition to Leidenfrost regime therefore reduces to the capability of the heated surface to sustain the heterogeneous boiling process.

6.2 Future Work

A more precise treatment of the Leidenfrost transition requires that both the hydrodynamical effects of the vapor flow and the heterogeneous boiling process be taken into account. We anticipate that our theoretical predictions and experimental insights will be useful for more sophisticated models of Leidenfrost transition. They may also open novel approaches to achieve better control of the heat transfer rate in heat management applications. In particular, a model incorporating conjugate heat transfer between the surface and the liquid is required in the case that variation in surface temperature is significant (as in the case of impacting water droplets). This requirement necessitates an experimental investigation into the evolution of the surface temperature and local heat transfer, as well as their relations to the wetted area during impact.

Appendices

Appendix A. Image Transformation

This MATLAB code projects the original images recorded from TIR into the normal plane and performs the background subtraction.

```

clc
clear
close all
% the address for folder containing original images:
default_folder = '....';
% path for creating Transformed folder:
default_folder2 = default_folder (1:end-1);
Img = 1;
[FileName,PathName] = uigetfile([default_folder '*.tif;*.tiff'],'Select
images'); % choose the first image
files = dir(PathName);
% to account for possible extra files existing in the folder:
N_files = length(files) - 4;
% ratio of pixels size in two directions used to project the image:
ratio = 1/ratio_value;
% Make new folder for transformed images:
Trans_folder_name = ['Trans_',FileName(1:end-10)];
mkdir(default_folder2,Trans_folder_name);
for I = 1:N_files
FileIn1 = [FileName(1:end-10) num2str(Img,'%06d') '.tif'];
FileOut = ['Trans_' FileName(1:end-10) num2str(I,'%06d') '.tif'];
image1 = imread([PathName FileIn1]);
image1double = double(image1);
disp(N_files-I)
FileIn2 = [FileName(1:end-10) num2str(I,'%06d') '.tif'];
image2 = imread([PathName FileIn2]);
image2double = double(image2);
% subtract the background:
image2_bg_sub_double = image2double - image1double +
mean(mean(image1double));
image2_bg_sub = uint8(image2_bg_sub_double);
[NY,NX] = size(image2_bg_sub);
% resize the image:
image2_bg_sub_final = imresize(image2_bg_sub,[NY/ratio,NX]);
% write the image:
imwrite(image2_bg_sub_final,[default_folder ,Trans_folder_name, '\
FileOut]);
end

```


Appendix B. Thermophysical Properties of Liquids

In this appendix, we provide the temperature-dependent properties of different liquids. We used these properties in our proposed theory for predicting the Leidenfrost temperature in chapter 5.

Table B.1 Thermophysical properties of acetone (taken from Ref. [329])

T Temp. °C	p_v saturation pressure (10 ³ Pa)	h_{lv} latent heat (kJ/kg)	ρ_l liquid density (kg/m ³)	ρ_v vapor density (kg/m ³)	μ_l liquid viscosity (10 ⁻³ N-s/m ²)	μ_v vapor viscosity (10 ⁻⁷ N-s/m ²)	k_l liquid thermal conductivity (W/m-K)	k_v vapor thermal conductivity ^a (W/m-K)	σ liquid surface tension (10 ⁻³ N/m)	$c_{p,l}$ liquid specific heat ^b (kJ/kg-K)	$c_{p,v}$ vapor specific heat ^c (kJ/kg-K)
-40	0.01	660.0	860.0	0.03	0.800	68.0	0.200		31.0	2.04	1.109
-20	0.03	615.6	845.0	0.10	0.500	73.0	0.189	0.0082	27.6	2.07	1.160
0	0.10	564.0	812.0	0.26	0.395	78.0	0.183	0.0096	26.2	2.11	1.215
20	0.27	552.0	790.0	0.64	0.323	82.0	0.181	0.0110	23.7	2.16	1.271
40	0.60	536.0	768.0	1.05	0.269	86.0	0.175	0.0126	21.2	2.22	1.328
60	1.15	517.0	744.0	2.37	0.226	90.0	0.168	0.0143	18.6	2.29	1.386
80	2.15	495.0	719.0	4.30	0.192	95.0	0.160	0.0161	16.2	2.39	1.444
100	4.43	472.0	689.6	6.94	0.170	98.0	0.148	0.0178	13.4	2.49	1.502
120	6.70	426.1	660.3	11.02	0.148	99.0	0.135	0.0195	10.7	2.61	1.560
140	10.49	394.4	631.8	18.61	0.132	103.0	0.126	0.0215	8.1	2.77	1.616

Table B.2 Thermophysical properties of ethanol (taken from Ref. [329])

T Temp. °C	p_v saturation pressure (10 ³ Pa)	h_{lv} latent heat (kJ/kg)	ρ_l liquid density (10 ³ kg/m ³)	ρ_v vapor density (kg/m ³)	μ_l liquid viscosity (10 ⁻³ N-s/m ²)	μ_v vapor viscosity (10 ⁻⁵ N-s/m ²)	k_l liquid thermal conductivity (W/m-K)	k_v vapor thermal conductivity ^a (W/m-K)	σ liquid surface tension (10 ⁻³ N/m)	$c_{p,l}$ liquid specific heat ^b (kJ/kg-K)	$c_{p,v}$ vapor specific heat ^c (kJ/kg-K)
0	0.012	1048.4	0.901	0.036	1.7990	0.774	0.183	0.0117	24.4	2.27	1.34
20	0.058	1030.0	0.800	0.085	1.1980	0.835	0.179	0.0139	22.8	2.40	1.40
40	0.180	1011.9	0.789	0.316	0.8190	0.900	0.175	0.0160	21.0	2.57	1.48
60	0.472	988.9	0.770	0.748	0.5880	0.959	0.171	0.0179	19.2	2.78	1.54
80	1.086	960.0	0.757	1.430	0.4320	1.030	0.169	0.0199	17.3	3.03	1.61
100	2.260	927.0	0.730	3.410	0.3180	1.092	0.167	0.0219	15.5	3.30	1.68
120	4.290	885.5	0.710	6.010	0.2430	1.157	0.165	0.0238	13.4	3.61	1.75
140	7.530	834.0	0.680	10.670	0.1900	1.219	0.163	0.0256	11.2	3.96	
160	12.756	772.9	0.650	17.450	0.1500	1.293	0.161	0.0272	9.0		
180	19.600	698.8	0.610	27.650	0.1200	1.369	0.159	0.0288	6.7		
200	29.400	598.3	0.564	44.480	0.0950	1.464	0.157	0.0395	4.3		
220	42.800	468.5	0.510	74.350	0.0725	1.618	0.155	0.0321	2.2		
240	60.200	280.5	0.415	135.500	0.0488	1.948	0.153		0.1		

Table B.3 Thermophysical properties of heptane (taken from Ref. [329])

T Temp. °C	p_v saturation pressure (10 ³ Pa)	h_{lv} latent heat (kJ/kg)	ρ_l liquid density (10 ³ kg/m ³)	ρ_v vapor density (10 ³ kg/m ³)	μ_l liquid viscosity (10 ⁻³ N-s/m ²)	μ_v vapor viscosity (10 ⁻⁷ N-s/m ²)	k_l liquid thermal conductivity (W/m-K)	k_v vapor thermal conductivity ^a (W/m-K)	σ liquid surface tension ^d (10 ⁻³ N/m)	$c_{p,l}$ liquid specific heat (kJ/kg-K)	$c_{p,v}$ vapor specific heat ^b (kJ/kg-K)
-20	0.003866	383.1	0.7172		0.6890		0.140	0.0084		2.10	0.83
0	0.0152	375.6	0.7005	0.000070	0.5260		0.134	0.0099		2.16	0.87
20	0.0472	366.0	0.6836	0.000200	0.4140		0.129	0.0115	20.86	2.23	0.92
40	0.1230	354.7	0.6665	0.000500	0.3380		0.123	0.0132	18.47	2.30	0.97
60	0.2800	342.6	0.6491	0.001100	0.2810		0.118	0.0151	16.39	2.39	1.02
80	0.5700	330.1	0.6311	0.002000	0.2390		0.113	0.0170	14.35	2.47	1.05
100	1.0606	316.7	0.6124	0.003597	0.1980	73.6		0.0189	12.47	2.57	1.09
120	1.8330	302.9	0.5926	0.006075	0.1672	78.2		0.0207	10.63	2.67	1.16
140	2.9790	287.4	0.5711	0.009785	0.1427	83.4		0.0228	8.87	2.78	
160	4.5990	269.5	0.5481	0.015110	0.1217	89.7		0.0251	7.19	2.89	

Table B.4 Thermophysical properties of water (taken from Ref. [329])

Property ^a	Temp.	α_0	α_1	α_2	α_3	α_4	α_5	Error (%)
p_v (10^5 Pa)	20–200 °C	-5.0945	7.2280	-2.8625-4 ^b	9.2341-7	-2.0295-9	2.1645-12	0.01
h_{lv} (kJ/kg)	20–200 °C	7.8201	-5.8906-4	-9.1355-6	8.4738-8	-3.9635-10	5.9150-13	0.05
ρ_l (kg/m ³)	20–200 °C	6.9094	-2.0146-5	-5.9868-6	2.5921-8	-9.3244-11	1.2103-13	0.02
ρ_v (kg/m ³)	20–200 °C	-5.3225	6.8366-2	-2.7243-4	8.4522-7	-1.6558-9	1.5514-12	0.01
μ_l (10^{-7} Ns/m ²)	20–200 °C	9.7620	-3.1154-2	2.0029-4	-9.5815-7	2.7772-9	-3.5075-12	0.02
μ_v (10^{-7} Ns/m ²)	20–200 °C	4.3995	3.8789-3	2.1181-5	-3.4406-7	1.6730-9	-2.8030-12	0.04
k_l (W/m-K)	20–200 °C	-5.6528-1	3.1743-3	-1.4392-5	-1.3224-8	2.5534-10	-6.4454-13	0.04
k_v (W/m-K)	20–200 °C	-4.0406	3.2288-3	5.3383-6	-6.7139-8	4.0967-10	-6.9579-13	0.07
σ (10^{-3} N/m)	20–200 °C	4.3438	-3.0664-3	2.0743-5	-2.5499-7	1.0377-9	-1.7156-12	0.03
$c_{p,l}$ (kJ/kg-K)	20–200 °C	1.4338	-2.2638-4	4.2819-6	-2.7411-8	1.4699-10	-2.2589-13	0.01
$c_{p,v}$ (kJ/kg-K)	20–200 °C	6.2084-1	3.1420-4	1.6110-6	4.0156-8	3.4841-11	-2.0709-13	0.01

^a Polynomial function: $\ln(\text{Property}) = \alpha_0 + \alpha_1 T + \alpha_2 T^2 + \alpha_3 T^3 + \alpha_4 T^4 + \alpha_5 T^5$ ^b The notation -2.8625-4 signifies -2.8625×10^{-4}

Table B.5 Thermophysical properties of methanol (taken from Ref. [329])

Property ^a	Temp.	α_0	α_1	α_2	α_3	α_4	α_5	Error (%)
p_v (10^5 Pa)	0–240 °C	-3.2260	5.2299-2 ^b	1.0307-4	-2.9204-6	1.4369-8	-2.2852-11	3.52
h_{lv} (kJ/kg)	0–240 °C	7.1001	-1.6650-3	4.2482-5	-8.2591-7	5.2087-9	-1.1710-11	0.39
ρ_l (10^3 kg/m ³)	60–220 °C	-2.0054-1	-5.5461-3	1.8492-4	-2.6081-6	1.4646-8	-2.8794-11	1.96
ρ_v (10^3 kg/m ³)	0–240 °C	-67.300	2.1944	-3.0858-2	2.1359-4	-7.2014-7	9.4732-10	6.77
μ_l (10^{-3} Ns/m ²)	0–240 °C	-2.0529-1	-1.8685-2	1.2225-4	-1.0701-6	4.5665-9	-7.2697-12	0.35
μ_v (10^{-7} Ns/m ²)	0–240 °C	4.4794	3.3930-3	4.0163-6	-7.3376-8	2.4641-10	-2.1042-13	0.28
k_l (W/m-K)	0–140 °C	-1.5847	-4.0650-4	1.1186-5	-2.2761-7	1.6087-9	-3.9220-12	0.03
k_v (W/m-K)	40–220 °C	-6.7646	8.9038-3	-3.5238-5	1.4232-7	-4.0030-10	5.0729-13	0.03
σ (10^{-3} N/m)	0–220 °C	3.2019	-6.2365-3	1.1123-4	-1.7652-6	1.0660-8	-2.4530-11	0.65
$c_{p,l}$ (kJ/kg-K)	0–40 °C	8.8377-1	6.2710-4	9.6297-6	0.0	0.0	0.0	0.01
$c_{p,v}$ (kJ/kg-K)	0–140 °C	3.1404-1	5.0100-3	-1.7365-4	3.4376-6	-2.5001-8	5.9705-11	0.40

^a Polynomial function: $\ln(\text{Property}) = \alpha_0 + \alpha_1 T + \alpha_2 T^2 + \alpha_3 T^3 + \alpha_4 T^4 + \alpha_5 T^5$ ^b The notation 5.2299-2 signifies 5.2299×10^{-2}

For IPA, we used the data presented in Table B.6 from Ref. [333] for density (ρ) and saturation pressure (P_{sat}) and from Ref. [334, 335] for surface tension (σ).

Table B.6 Thermophysical properties of IPA (taken from Ref. [333-335])

T (K)	σ (mN/m)	T (K)	ρ (kg/m ³)	P_{sat} (kPa)
287.15	22.3	275	798.8	1.186
293.15	21.74	300	779.55	6.481
298.15	21.22	325	757.02	25.931
303.15	20.72	350	731.84	81.349
308.15	20.23	375	702.89	210.416
308.55	20.1	400	669.36	466.18
311.35	19.8	425	630	911.89
313.15	19.71	450	581.99	1615.77
314.95	19.5	475	519.76	2654.77
318.15	19.21	500	425.22	4139.39
319.85	19			
323.15	18.69			
328.55	18.2			
336.75	17.4			
340.15	17.2			
344.15	16.9			
347.95	16.5			
350.65	16.2			
353.45	15.9			

For HFE 7100, we used the following relations for density (ρ) and surface tension (σ) [337] and saturation pressure (P_{sat}) [336]:

$$\rho = 2510.64 - 5.68904T + 1.32655 \times 10^{-2}T^2 - 1.80161 \times 10^{-5}T^3 \quad \text{B.1}$$

$$\sigma = 49.351 \times 10^{-3} \left(1 - \frac{T}{T_r}\right)^{1.26} \left[1 - 0.0527 \left(1 - \frac{T}{T_r}\right)^{0.5}\right] \quad \text{B.2}$$

$$P_{sat} = \exp\left(-\frac{3641.9}{T} + 22.415\right) \quad \text{B.3}$$

where $T_r = T/T_c$, T_c is the critical temperature and T is in K, ρ in kg/m³, σ in N/m and P_{sat} in Pa.

Similarly, for HFE 7300, we used the following relations for density (ρ) and surface tension (σ) [337] and saturation pressure (P_{sat}) [338]:

$$\rho = 2471.72 - 4.04355T + 0.74486 \times 10^{-2}T^2 - 102405 \times 10^{-5}T^3 \quad \text{B.4}$$

$$\sigma = 46.54 \times 10^{-3} \left(1 - \frac{T}{T_r}\right)^{1.26} \left[1 - 0.1134 \left(1 - \frac{T}{T_r}\right)^{0.5}\right] \quad \text{B.5}$$

$$P_{sat} = \exp\left(-\frac{6504.623709}{T + 350} + 21.151057\right) \quad \text{B.6}$$

where in Eq. B.6, T is in °C and P_{sat} is in mmHG.

For benzene, we used the following relations for density (ρ)[342]:

$$\rho = \frac{A}{B^{1+\left(1-\frac{T}{C}\right)^D}} \quad \text{B.7}$$

where $A = 80.3682$, $B = 0.266457$, $C = 561.65$, $D = 0.28797$ and T is in K. For saturation pressure (P_{sat}), we used the following relation [343]:

$$P_{sat} = 10^{A-\frac{B}{C+T}} \quad \text{B.8}$$

where $A = 7.2009$, $B = 1415.8$, $C = 248.028$, T is in °C and P_{sat} is in mmHG. Finally for surface tension (σ), we used the following relation [344]:

$$\sigma = A(1 - T_r)^B \quad \text{B.9}$$

where $A = 72.989$, $B = 1.2578$, $T_r = T/T_c$, T_c is the critical temperature, T is in K and σ is in mN/m.

References

1. Worthington, A., *On the forms assumed by drops of liquids falling vertically on a horizontal plate*. Proceedings of the royal society of London, 1876. **25**(171-178): p. 261-272.
2. Worthington, A.M. and R. Cole, *Impact with a liquid surface, studied by the aid of instantaneous photography*. Philosophical Transactions of the Royal Society of London. Series A, Containing Papers of a Mathematical or Physical Character, 1897: p. 137-148.
3. Worthington, A. and R. Cole, *Impact with a liquid surface studied by the aid of instantaneous photography. Paper II*. Philosophical Transactions of the Royal Society of London. Series A, Containing Papers of a Mathematical or Physical Character, 1900: p. 175-199.
4. Worthington, A.M., *A study of splashes*. 1908: Longmans, Green, and Co.
5. Worthington, A., *A Second Paper on the Forms Assumed by Drops of Liquids Falling Vertically on a Horizontal Plate*. Proceedings of the Royal Society of London, 1876. **25**(171-178): p. 498-503.
6. Liu, Y., L. Moevius, X. Xu, T. Qian, J.M. Yeomans, and Z. Wang, *Pancake bouncing on superhydrophobic surfaces*. Nature Physics, 2014.
7. de Ruiter, J., R. Lagraauw, D. van den Ende, and F. Mugele, *Wettability-independent bouncing on flat surfaces mediated by thin air films*. Nature Physics, 2014.
8. Bird, J.C., R. Dhiman, H.-M. Kwon, and K.K. Varanasi, *Reducing the contact time of a bouncing drop*. Nature, 2013. **503**(7476): p. 385-388.
9. Adera, S., R. Raj, R. Enright, and E.N. Wang, *Non-wetting droplets on hot superhydrophilic surfaces*. Nature communications, 2013. **4**.
10. Vakarelski, I.U., N.A. Patankar, J.O. Marston, D.Y. Chan, and S.T. Thoroddsen, *Stabilization of Leidenfrost vapour layer by textured superhydrophobic surfaces*. Nature, 2012. **489**(7415): p. 274-277.
11. Jung, S., M.K. Tiwari, N.V. Doan, and D. Poulikakos, *Mechanism of supercooled droplet freezing on surfaces*. Nature communications, 2012. **3**: p. 615.
12. Duez, C., C. Ybert, C. Clanet, and L. Bocquet, *Making a splash with water repellency*. Nature physics, 2007. **3**(3): p. 180-183.
13. Lagubeau, G., M. Le Merrer, C. Clanet, and D. Quéré, *Leidenfrost on a ratchet*. Nature Physics, 2011. **7**(5): p. 395-398.
14. Li, J., Y. Hou, Y. Liu, C. Hao, M. Li, M.K. Chaudhury, S. Yao, and Z. Wang, *Directional transport of high-temperature Janus droplets mediated by structural topography*. Nature Physics, 2016.
15. Schutzius, T.M., S. Jung, T. Maitra, G. Graeber, M. Köhne, and D. Poulikakos, *Spontaneous droplet trampolining on rigid superhydrophobic surfaces*. Nature, 2015. **527**(7576): p. 82-85.
16. Hao, C., J. Li, Y. Liu, X. Zhou, Y. Liu, R. Liu, L. Che, W. Zhou, D. Sun, and L. Li, *Superhydrophobic-like tunable droplet bouncing on slippery liquid interfaces*. Nature Communications, 2015. **6**.
17. Kolinski, J.M., L. Mahadevan, and S.M. Rubinstein, *Lift-Off Instability During the Impact of a Drop on a Solid Surface*. Physical review letters, 2014. **112**(13): p. 134501.
18. Riboux, G. and J.M. Gordillo, *Experiments of Drops Impacting a Smooth Solid Surface: A Model of the Critical Impact Speed for Drop Splashing*. Physical review letters, 2014. **113**(2): p. 024507.
19. Antonini, C., I. Bernagozzi, S. Jung, D. Poulikakos, and M. Marengo, *Water drops dancing on ice: how sublimation leads to drop rebound*. Physical review letters, 2013. **111**(1): p. 014501.
20. Eddi, A., K. Winkels, and J. Snoeijer, *Influence of droplet geometry on the coalescence of low viscosity drops*. Physical review letters, 2013. **111**(14): p. 144502.
21. Luu, L.-H. and Y. Forterre, *Giant Drag Reduction in Complex Fluid Drops on Rough Hydrophobic Surfaces*. Physical review letters, 2013. **110**(18): p. 184501.

22. Peters, I.R., Q. Xu, and H.M. Jaeger, *Splashing onset in dense suspension droplets*. Physical review letters, 2013. **111**(2): p. 028301.
23. Bouwhuis, W., R.C. van der Veen, T. Tran, D.L. Keij, K.G. Winkels, I.R. Peters, D. van der Meer, C. Sun, J.H. Snoeijer, and D. Lohse, *Maximal air bubble entrainment at liquid-drop impact*. Physical review letters, 2012. **109**(26): p. 264501.
24. Tran, T., H.J. Staat, A. Prosperetti, C. Sun, and D. Lohse, *Drop impact on superheated surfaces*. Physical review letters, 2012. **108**(3): p. 036101.
25. Kolinski, J.M., S.M. Rubinstein, S. Mandre, M.P. Brenner, D.A. Weitz, and L. Mahadevan, *Skating on a film of air: drops impacting on a surface*. Physical review letters, 2012. **108**(7): p. 074503.
26. de Ruiter, J., J.M. Oh, D. van den Ende, and F. Mugele, *Dynamics of collapse of air films in drop impact*. Physical review letters, 2012. **108**(7): p. 074505.
27. Latka, A., A. Strandburg-Peshkin, M.M. Driscoll, C.S. Stevens, and S.R. Nagel, *Creation of prompt and thin-sheet splashing by varying surface roughness or increasing air pressure*. Physical review letters, 2012. **109**(5): p. 054501.
28. Celestini, F., T. Frisch, and Y. Pomeau, *Take off of small Leidenfrost droplets*. Physical review letters, 2012. **109**(3): p. 034501.
29. Thoraval, M.-J., K. Takehara, T.G. Etoh, S. Popinet, P. Ray, C. Josserand, S. Zaleski, and S.T. Thoroddsen, *von Kármán vortex street within an impacting drop*. Physical review letters, 2012. **108**(26): p. 264506.
30. San Lee, J., B.M. Weon, J.H. Je, and K. Fezzaa, *How does an air film evolve into a bubble during drop impact?* Physical review letters, 2012. **109**(20): p. 204501.
31. Vakarelski, I.U., J.O. Marston, D.Y. Chan, and S.T. Thoroddsen, *Drag reduction by Leidenfrost vapor layers*. Physical review letters, 2011. **106**(21): p. 214501.
32. Driscoll, M.M. and S.R. Nagel, *Ultrafast interference imaging of air in splashing dynamics*. Physical review letters, 2011. **107**(15): p. 154502.
33. Thoroddsen, S., M.-J. Thoraval, K. Takehara, and T. Etoh, *Droplet splashing by a slingshot mechanism*. Physical review letters, 2011. **106**(3): p. 034501.
34. Dupeux, G., M. Le Merrer, C. Clanet, and D. Quéré, *Trapping Leidenfrost drops with crenulations*. Physical review letters, 2011. **107**(11): p. 114503.
35. Schroll, R.D., C. Josserand, S. Zaleski, and W.W. Zhang, *Impact of a viscous liquid drop*. Physical review letters, 2010. **104**(3): p. 034504.
36. Smith, M. and V. Bertola, *Effect of polymer additives on the wetting of impacting droplets*. Physical review letters, 2010. **104**(15): p. 154502.
37. Lagubeau, G., M.A. Fontelos, C. Josserand, A. Maurel, V. Pagneux, and P. Petitjeans, *Flower patterns in drop impact on thin liquid films*. Physical review letters, 2010. **105**(18): p. 184503.
38. Mandre, S., M. Mani, and M.P. Brenner, *Precursors to splashing of liquid droplets on a solid surface*. Physical review letters, 2009. **102**(13): p. 134502.
39. Bird, J.C., S. Mandre, and H.A. Stone, *Short-time dynamics of partial wetting*. Physical review letters, 2008. **100**(23): p. 234501.
40. Bartolo, D., A. Boudaoud, G. Narcy, and D. Bonn, *Dynamics of non-Newtonian droplets*. Physical review letters, 2007. **99**(17): p. 174502.
41. Xu, L., W.W. Zhang, and S.R. Nagel, *Drop splashing on a dry smooth surface*. Physical review letters, 2005. **94**(18): p. 184505.
42. Burton, J., A. Sharpe, R. van der Veen, A. Franco, and S. Nagel, *Geometry of the vapor layer under a Leidenfrost drop*. Physical review letters, 2012. **109**(7): p. 074301.
43. Würger, A., *Leidenfrost gas ratchets driven by thermal creep*. Physical review letters, 2011. **107**(16): p. 164502.
44. Quéré, D., *Leidenfrost dynamics*. Annual Review of Fluid Mechanics, 2013. **45**: p. 197-215.

45. Snoeijer, J.H. and B. Andreotti, *Moving contact lines: scales, regimes, and dynamical transitions*. Annual review of fluid mechanics, 2013. **45**: p. 269-292.
46. Quéré, D., *Wetting and roughness*. Annu. Rev. Mater. Res., 2008. **38**: p. 71-99.
47. Yarin, A., *Drop impact dynamics: splashing, spreading, receding, bouncing....*. Annu. Rev. Fluid Mech., 2006. **38**: p. 159-192.
48. Lin, S. and R. Reitz, *Drop and spray formation from a liquid jet*. Annual Review of Fluid Mechanics, 1998. **30**(1): p. 85-105.
49. Rein, M., *Phenomena of liquid drop impact on solid and liquid surfaces*. Fluid Dynamics Research, 1993. **12**(2): p. 61.
50. Prosperetti, A. and H.N. Oguz, *The impact of drops on liquid surfaces and the underwater noise of rain*. Annual Review of Fluid Mechanics, 1993. **25**(1): p. 577-602.
51. Korobkin, A. and V. Pukhnachov, *Initial stage of water impact*. Annual review of fluid mechanics, 1988. **20**(1): p. 159-185.
52. Lesser, M. and J. Field, *The impact of compressible liquids*. Annual review of fluid mechanics, 1983. **15**(1): p. 97-122.
53. Josserand, C. and S. Thoroddsen, *Drop impact on a solid surface*. Annual Review of Fluid Mechanics, 2016. **48**: p. 365-391.
54. Liang, G. and I. Mudawar, *Review of drop impact on heated walls*. International Journal of Heat and Mass Transfer, 2017. **106**: p. 103-126.
55. Leidenfrost, J.G., *De aquae communis nonnullis qualitatibus tractatus*. 1756: [Typis Joan. Sebast. Straube, Acad. typogr.] Impensis Hermanni Ovensi, Univers. bibliopolæ.
56. Gottfried, B., C. Lee, and K. Bell, *The Leidenfrost phenomenon: film boiling of liquid droplets on a flat plate*. International Journal of heat and mass transfer, 1966. **9**(11): p. 1167-1188.
57. Biance, A.-L., C. Clanet, and D. Quéré, *Leidenfrost drops*. Physics of Fluids (1994-present), 2003. **15**(6): p. 1632-1637.
58. Lefebvre, A.H., *Atomization and sprays*, 1989. Hemisphere, New York.
59. de Gans, B.J., P.C. Duineveld, and U.S. Schubert, *Inkjet printing of polymers: state of the art and future developments*. Advanced materials, 2004. **16**(3): p. 203-213.
60. Sirringhaus, H. and T. Shimoda, *Inkjet printing of functional materials*. MRS bulletin, 2003. **28**(11): p. 802-806.
61. Aziz, S.D. and S. Chandra, *Impact, recoil and splashing of molten metal droplets*. International journal of heat and mass transfer, 2000. **43**(16): p. 2841-2857.
62. Grant, G., J. Brenton, and D. Drysdale, *Fire suppression by water sprays*. Progress in energy and combustion science, 2000. **26**(2): p. 79-130.
63. Jiang, X., Y. Wan, H. Herman, and S. Sampath, *Role of condensates and adsorbates on substrate surface on fragmentation of impinging molten droplets during thermal spray*. Thin Solid Films, 2001. **385**(1): p. 132-141.
64. Fauchais, P., A. Vardelle, M. Vardelle, and M. Fukumoto, *Knowledge concerning splat formation: an invited review*. Journal of Thermal Spray Technology, 2004. **13**(3): p. 337-360.
65. Pasandideh-Fard, M., V. Pershin, S. Chandra, and J. Mostaghimi, *Splat shapes in a thermal spray coating process: simulations and experiments*. Journal of Thermal Spray Technology, 2002. **11**(2): p. 206-217.
66. Chandra, S. and P. Fauchais, *Formation of solid splats during thermal spray deposition*. Journal of thermal spray technology, 2009. **18**(2): p. 148-180.
67. Dhiman, R., A.G. McDonald, and S. Chandra, *Predicting splat morphology in a thermal spray process*. Surface and Coatings Technology, 2007. **201**(18): p. 7789-7801.
68. Xue, M., S. Chandra, and J. Mostaghimi, *Investigation of splat curling up in thermal spray coatings*. Journal of thermal spray technology, 2006. **15**(4): p. 531-536.

69. Li, C.-J. and J.-L. Li, *Evaporated-gas-induced splashing model for splat formation during plasma spraying*. Surface and Coatings Technology, 2004. **184**(1): p. 13-23.
70. Kim, J., *Spray cooling heat transfer: the state of the art*. International Journal of Heat and Fluid Flow, 2007. **28**(4): p. 753-767.
71. Laan, N., K.G. de Bruin, D. Bartolo, C. Josserand, and D. Bonn, *Maximum Diameter of Impacting Liquid Droplets*. Physical Review Applied, 2014. **2**(4): p. 044018.
72. Mugele, F. and J.-C. Baret, *Electrowetting: from basics to applications*. Journal of Physics: Condensed Matter, 2005. **17**(28): p. R705.
73. Sinha-Ray, S. and A.L. Yarin, *Drop impact cooling enhancement on nano-textured surfaces. Part I: Theory and results of the ground (1g) experiments*. International Journal of Heat and Mass Transfer, 2014. **70**: p. 1095-1106.
74. Sinha-Ray, S., S. Sinha-Ray, A.L. Yarin, C.M. Weickgenannt, J. Emmert, and C. Tropea, *Drop impact cooling enhancement on nano-textured surfaces. Part II: Results of the parabolic flight experiments [zero gravity (0g) and supergravity (1.8 g)]*. International Journal of Heat and Mass Transfer, 2014. **70**: p. 1107-1114.
75. Dalili, N., A. Edrisy, and R. Carriveau, *A review of surface engineering issues critical to wind turbine performance*. Renewable and Sustainable Energy Reviews, 2009. **13**(2): p. 428-438.
76. Chandra, S. and C. Avedisian, *On the collision of a droplet with a solid surface*. Proceedings: Mathematical and Physical Sciences, 1991: p. 13-41.
77. Panchagnula, M.V. and P.E. Sojka, *Spatial droplet velocity and size profiles in effervescent atomizer-produced sprays*. Fuel, 1999. **78**(6): p. 729-741.
78. Holder, A.L., E.P. Vejerano, X. Zhou, and L.C. Marr, *Nanomaterial disposal by incineration*. Environmental Science: Processes & Impacts, 2013. **15**(9): p. 1652-1664.
79. Yao, S.-C. and K.Y. Cai, *The dynamics and Leidenfrost temperature of drops impacting on a hot surface at small angles*. Experimental Thermal and Fluid Science, 1988. **1**(4): p. 363-371.
80. Liu, Q. and M. Orme, *High precision solder droplet printing technology and the state-of-the-art*. Journal of materials processing technology, 2001. **115**(3): p. 271-283.
81. Yu, D., J.B. Kwak, S. Park, and J. Lee, *Dynamic responses of PCB under product-level free drop impact*. Microelectronics Reliability, 2010. **50**(7): p. 1028-1038.
82. Bennett, T. and D. Poulikakos, *Splat-quench solidification: estimating the maximum spreading of a droplet impacting a solid surface*. Journal of materials science, 1993. **28**(4): p. 963-970.
83. Werner, S.R., J.R. Jones, A.H. Paterson, R.H. Archer, and D.L. Pearce, *Droplet impact and spreading: droplet formulation effects*. Chemical engineering science, 2007. **62**(9): p. 2336-2345.
84. Andrade, R., O. Skurtys, and F. Osorio, *Drop impact behavior on food using spray coating: Fundamentals and applications*. Food Research International, 2013. **54**(1): p. 397-405.
85. Golovitchev, V.I. and J. Yang, *Construction of combustion models for rapeseed methyl ester bio-diesel fuel for internal combustion engine applications*. Biotechnology advances, 2009. **27**(5): p. 641-655.
86. Arcoumanis, C. and J. Whitelaw, *Fluid mechanics of internal combustion engines—a review*. Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science, 1987. **201**(1): p. 57-74.
87. Stanton, D.W. and C.J. Rutland, *Multi-dimensional modeling of thin liquid films and spray-wall interactions resulting from impinging sprays*. International Journal of Heat and Mass Transfer, 1998. **41**(20): p. 3037-3054.
88. Moreira, A., A. Moita, and M. Panao, *Advances and challenges in explaining fuel spray impingement: How much of single droplet impact research is useful?* Progress in energy and combustion science, 2010. **36**(5): p. 554-580.

89. Gaynor, A.T., N.A. Meisel, C.B. Williams, and J.K. Guest, *Multiple-Material Topology Optimization of Compliant Mechanisms Created Via PolyJet Three-Dimensional Printing*. Journal of Manufacturing Science and Engineering, 2014. **136**(6): p. 061015.
90. Soto, D., A.B. De Larivière, X. Boutillon, C. Clanet, and D. Quéré, *The force of impacting rain*. Soft matter, 2014.
91. Wirth, W., S. Storp, and W. Jacobsen, *Mechanisms controlling leaf retention of agricultural spray solutions*. Pesticide science, 1991. **33**(4): p. 411-420.
92. Wallace, K. and K. Yoshida, *Determination of dynamic spread factor of water droplets impacting on water-sensitive paper surfaces*. Journal of Colloid and Interface Science, 1978. **63**(1): p. 164-165.
93. Linke, H., B. Alemán, L. Melling, M. Taormina, M. Francis, C. Dow-Hygelund, V. Narayanan, R. Taylor, and A. Stout, *Self-propelled Leidenfrost droplets*. Physical review letters, 2006. **96**(15): p. 154502.
94. De Gennes, P.-G., F. Brochard-Wyart, and D. Quéré, *Capillarity and wetting phenomena: drops, bubbles, pearls, waves*. 2004: Springer.
95. Clanet, C., C. Béguin, D. Richard, and D. Quéré, *Maximal deformation of an impacting drop*. Journal of Fluid Mechanics, 2004. **517**: p. 199-208.
96. Lee, J., N. Laan, K. de Bruin, G. Skantzaris, N. Shahidzadeh, D. Derome, J. Carmeliet, and D. Bonn, *Universal rescaling of drop impact on smooth and rough surfaces*. Journal of Fluid Mechanics, 2015. **786**: p. R4.
97. Jones, H., *Cooling, freezing and substrate impact of droplets formed by rotary atomization*. Journal of Physics D: Applied Physics, 1971. **4**(11): p. 1657.
98. McPherson, R., *On the formation of thermally sprayed alumina coatings*. Journal of Materials Science, 1980. **15**(12): p. 3141-3149.
99. Collings, E., A. Markworth, J. McCoy, and J. Saunders, *Splat-quench solidification of freely falling liquid-metal drops by impact on a planar substrate*. Journal of Materials Science, 1990. **25**(8): p. 3677-3682.
100. Madejski, J., *Solidification of droplets on a cold surface*. International Journal of Heat and Mass Transfer, 1976. **19**(9): p. 1009-1013.
101. Delplanque, J.-P. and R. Rangel, *An improved model for droplet solidification on a flat surface*. Journal of materials science, 1997. **32**(6): p. 1519-1530.
102. Markworth, A.J. and J.H. Saunders, *An improved velocity field for the Madejski splat-quench solidification model*. International journal of heat and mass transfer, 1992. **35**(7): p. 1836-1837.
103. Zhang, H., *Theoretical analysis of spreading and solidification of molten droplet during thermal spray deposition*. International Journal of Heat and Mass Transfer, 1999. **42**(14): p. 2499-2508.
104. Hamatani, H., T. Okada, and T. Yoshida, *Radio-frequency and hybrid plasma spraying of ceramics*. ICPS-9, Italy, 1989: p. 1527-1532.
105. Trapaga, G. and J. Szekely, *Mathematical modeling of the isothermal impingement of liquid droplets in spraying processes*. Metallurgical Transactions B, 1991. **22**(6): p. 901-914.
106. Fukanuma, H., *An analysis of the porosity producing mechanism*. Thermal Spray: International Advances in Coatings Technology, 1992: p. 767-772.
107. Watanabe, T., I. Kuribayashi, T. Honda, and A. Kanzawa, *Deformation and Solidification of a Droplet on a Cold Substrate*. Chemical engineering science, 1992. **47**(12): p. 3059-3065.
108. Yoshida, T., T. Okada, H. Hamatani, and H. Kumaoka, *Integrated fabrication process for solid oxide fuel cells using novel plasma spraying*. Plasma Sources Science and Technology, 1992. **1**(3): p. 195.
109. Bertagnolli, M., M. Marchese, and G. Jacucci, *Modeling of particles impacting on a rigid substrate under plasma spraying conditions*. Journal of thermal spray technology, 1995. **4**(1): p. 41-49.

110. Liu, H., E. Lavernia, and R. Rangel, *Numerical simulation of impingement of molten Ti, Ni, and W droplets on a flat substrate*. Journal of Thermal Spray Technology, 1993. **2**(4): p. 369-378.
111. Scheller, B.L. and D.W. Bousfield, *Newtonian drop impact with a solid surface*. AIChE Journal, 1995. **41**(6): p. 1357-1367.
112. Pasandideh-Fard, M., Y. Qiao, S. Chandra, and J. Mostaghimi, *Capillary effects during droplet impact on a solid surface*. Physics of Fluids (1994-present), 1996. **8**(3): p. 650-659.
113. Ukiwe, C. and D.Y. Kwok, *On the maximum spreading diameter of impacting droplets on well-prepared solid surfaces*. Langmuir, 2005. **21**(2): p. 666-673.
114. Roisman, I.V., R. Rioboo, and C. Tropea, *Normal impact of a liquid drop on a dry surface: model for spreading and receding*. Proceedings of the Royal Society of London. Series A: Mathematical, Physical and Engineering Sciences, 2002. **458**(2022): p. 1411-1430.
115. Fedorchenko, A.I. and A.-B. Wang, *The formation and dynamics of a blob on free and wall sheets induced by a drop impact on surfaces*. Physics of Fluids (1994-present), 2004. **16**(11): p. 3911-3920.
116. Fedorchenko, A.I., A.-B. Wang, and Y.-H. Wang, *Effect of capillary and viscous forces on spreading of a liquid drop impinging on a solid surface*. Physics of Fluids (1994-present), 2005. **17**(9): p. 093104.
117. Pasandideh-Fard, M., R. Bhola, S. Chandra, and J. Mostaghimi, *Deposition of tin droplets on a steel plate: simulations and experiments*. International Journal of Heat and Mass Transfer, 1998. **41**(19): p. 2929-2945.
118. Healy, W., J. Hartley, and S. Abdel-Khalik, *Comparison between theoretical models and experimental data for the spreading of liquid droplets impacting a solid surface*. International journal of heat and mass transfer, 1996. **39**(14): p. 3079-3082.
119. Yang, W.-J., *Theory on vaporization and combustion of liquid drops of pure substances and binary mixtures on heated surfaces*. Tokyo, University, Institute of Space and Aeronautical Science, Report no. 535, 1975. **40**: p. 423-455.
120. Bechtel, S., D. Boggy, and F. Talke, *Impact of a liquid drop against a flat surface*. IBM Journal of Research and Development, 1981. **25**(6): p. 963-971.
121. Naber, J.D., *Droplet impingement on a heated surface*. 1992: University of Wisconsin--Madison.
122. Ford, R. and C. Furmidge, *Impact and spreading of spray drops on foliar surfaces*. Soc Chem Ind Monogr, 1967. **25**: p. 417-32.
123. Shi, M. and J.C. Chen. *Behavior of a liquid droplet impinging on a solid surface*. in ASME Winter Annual Meeting. Preprint. 1983.
124. Kurabayashi, T., T. Karasawa, and K. Iida, *Impact of liquid droplets on a solid surface*. JSME, 1967(175): p. 153-156.
125. Healy, W., J. Hartley, and S. Abdel-Khalik, *Surface wetting effects on the spreading of liquid droplets impacting a solid surface at low Weber numbers*. International journal of heat and mass transfer, 2001. **44**(1): p. 235-240.
126. Kistler, S., *Hydrodynamics of wetting*. Wettability, 1993. **6**: p. 311-430.
127. Roisman, I., B. Prunet-Foch, C. Tropea, and M. Vignes-Adler, *Multiple drop impact onto a dry solid substrate*. Journal of colloid and interface science, 2002. **256**(2): p. 396-410.
128. Roisman, I.V., *Inertia dominated drop collisions. II. An analytical solution of the Navier-Stokes equations for a spreading viscous film*. Physics of Fluids (1994-present), 2009. **21**(5): p. 052104.
129. Cheng, L., *Dynamic spreading of drops impacting onto a solid surface*. Industrial & Engineering Chemistry Process Design and Development, 1977. **16**(2): p. 192-197.

130. Vadillo, D., A. Soucemarianadin, C. Delattre, and D. Roux, *Dynamic contact angle effects onto the maximum drop impact spreading on solid surfaces*. Physics of Fluids (1994-present), 2009. **21**(12): p. 122002.
131. Asai, A., M. Shioya, S. Hirasawa, and T. Okazaki, *Impact of an ink drop on paper*. The Journal of imaging science and technology, 1993. **37**(2): p. 205-207.
132. Mao, T., D. Kuhn, and H. Tran, *Spread and rebound of liquid droplets upon impact on flat surfaces*. AIChE Journal, 1997. **43**(9): p. 2169-2179.
133. An, S.M. and S.Y. Lee, *Maximum spreading of a shear-thinning liquid drop impacting on dry solid surfaces*. Experimental Thermal and Fluid Science, 2012. **38**: p. 140-148.
134. Richard, D., C. Clanet, and D. Quéré, *Surface phenomena: Contact time of a bouncing drop*. Nature, 2002. **417**(6891): p. 811-811.
135. Marmanis, H. and S. Thoroddsen, *Scaling of the fingering pattern of an impacting drop*. Physics of Fluids (1994-present), 1996. **8**(6): p. 1344-1346.
136. Kim, H.-Y. and J.-H. Chun, *The recoiling of liquid droplets upon collision with solid surfaces*. Physics of Fluids (1994-present), 2001. **13**(3): p. 643-659.
137. Kendall, G.E. and W.M. Rohsenow, *Heat transfer to impacting drops and post critical heat flux dispersed flow*. 1978, Cambridge, Mass.: Heat Transfer Laboratory, Dept. of Mechanical Engineering, Massachusetts Institute of Technology,[1978].
138. Eggers, J., M.A. Fontelos, C. Josserand, and S. Zaleski, *Drop dynamics after impact on a solid wall: theory and simulations*. Physics of Fluids (1994-present), 2010. **22**(6): p. 062101.
139. Thoroddsen, S., T. Etoh, K. Takehara, N. Ootsuka, and Y. Hatsuki, *The air bubble entrapped under a drop impacting on a solid surface*. Journal of fluid mechanics, 2005. **545**: p. 203-212.
140. van der Veen, R.C., T. Tran, D. Lohse, and C. Sun, *Direct measurements of air layer profiles under impacting droplets using high-speed color interferometry*. Physical Review E, 2012. **85**(2): p. 026315.
141. Korobkin, A., A. Ellis, and F. Smith, *Trapping of air in impact between a body and shallow water*. Journal of Fluid Mechanics, 2008. **611**: p. 365-394.
142. Popinet, S. and S. Zaleski, *A front-tracking algorithm for accurate representation of surface tension*. International Journal for Numerical Methods in Fluids, 1999. **30**(6): p. 775-793.
143. Lagubeau, G., M.A. Fontelos, C. Josserand, A. Maurel, V. Pagneux, and P. Petitjeans, *Spreading dynamics of drop impacts*. Journal of Fluid Mechanics, 2012. **713**: p. 50-60.
144. Bayer, I.S. and C.M. Megaridis, *Contact angle dynamics in droplets impacting on flat surfaces with different wetting characteristics*. Journal of Fluid Mechanics, 2006. **558**: p. 415-449.
145. Sen, S., V. Vaikuntanathan, and D. Sivakumar, *Experimental investigation of biofuel drop impact on stainless steel surface*. Experimental Thermal and Fluid Science, 2014. **54**: p. 38-46.
146. Harlow, F.H. and J.P. Shannon, *The splash of a liquid drop*. Journal of Applied Physics, 1967. **38**(10): p. 3855-3866.
147. Tanner, L., *The spreading of silicone oil drops on horizontal surfaces*. Journal of Physics D: Applied Physics, 1979. **12**(9): p. 1473.
148. Karl, A., K. Anders, and A. Frohn, *Experimental investigation of droplet deformation during wall collisions by image analysis*. ASME, NEW YORK, NY,(USA). 1993. **172**: p. 135-141.
149. Stow, C. and M. Hadfield, *An experimental investigation of fluid flow resulting from the impact of a water drop with an unyielding dry surface*. Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences, 1981. **373**(1755): p. 419-441.
150. Fukai, J., Y. Shiiba, T. Yamamoto, O. Miyatake, D. Poulikakos, C. Megaridis, and Z. Zhao, *Wetting effects on the spreading of a liquid droplet colliding with a flat surface: experiment and modeling*. Physics of Fluids (1994-present), 1995. **7**(2): p. 236-247.

151. Mundo, C., M. Sommerfeld, and C. Tropea, *Droplet-wall collisions: experimental studies of the deformation and breakup process*. International journal of multiphase flow, 1995. **21**(2): p. 151-173.
152. Schiaffino, S. and A.A. Sonin, *Molten droplet deposition and solidification at low Weber numbers*. Physics of Fluids (1994-present), 1997. **9**(11): p. 3172-3187.
153. Rioboo, R., C. Tropea, and M. Marengo, *Outcomes from a drop impact on solid surfaces*. Atomization and Sprays, 2001. **11**(2).
154. Rioboo, R., M. Marengo, and C. Tropea, *Time evolution of liquid drop impact onto solid, dry surfaces*. Experiments in Fluids, 2002. **33**(1): p. 112-124.
155. Šikalo, Š., M. Marengo, C. Tropea, and E. Ganić, *Analysis of impact of droplets on horizontal surfaces*. Experimental thermal and fluid science, 2002. **25**(7): p. 503-510.
156. Šikalo, Š., H.-D. Wilhelm, I. Roisman, S. Jakirlić, and C. Tropea, *Dynamic contact angle of spreading droplets: experiments and simulations*. Physics of Fluids (1994-present), 2005. **17**(6): p. 062103.
157. Šikalo, Š., C. Tropea, and E. Ganić, *Impact of droplets onto inclined surfaces*. Journal of colloid and interface science, 2005. **286**(2): p. 661-669.
158. Pittoni, P.G., H.-K. Tsao, and S.-Y. Lin, *Water drop impingement on graphite substrates with random dilute defects*. Experimental Thermal and Fluid Science, 2014. **53**: p. 142-146.
159. Lee, S.J., H.K. Huh, and D.H. Kwon, *Energy dissipation of graphene colloidal suspension droplets impacting on solid substrates*. RSC Advances, 2014. **4**(14): p. 7216-7224.
160. Bertola, V. and M.D. Haw, *Impact of concentrated colloidal suspension drops on solid surfaces*. Powder Technology, 2014.
161. Rioboo, R., C. Bauthier, J. Conti, M. Voue, and J. De Coninck, *Experimental investigation of splash and crown formation during single drop impact on wetted surfaces*. Experiments in fluids, 2003. **35**(6): p. 648-652.
162. Xu, L., *Liquid drop splashing on smooth, rough, and textured surfaces*. Physical Review E, 2007. **75**(5): p. 056316.
163. Xu, L., L. Barcos, and S.R. Nagel, *Splashing of liquids: interplay of surface roughness with surrounding gas*. Physical Review E, 2007. **76**(6): p. 066311.
164. Mani, M., S. Mandre, and M.P. Brenner, *Events before droplet splashing on a solid surface*. Journal of Fluid Mechanics, 2010. **647**: p. 163-185.
165. Mandre, S. and M.P. Brenner, *The mechanism of a splash on a dry solid surface*. Journal of Fluid Mechanics, 2012. **690**: p. 148-172.
166. Driscoll, M.M., C.S. Stevens, and S.R. Nagel, *Thin film formation during splashing of viscous liquids*. Physical Review E, 2010. **82**(3): p. 036302.
167. Tsai, P., R. CA van der Veen, M. van de Raa, and D. Lohse, *How micropatterns and air pressure affect splashing on surfaces*. Langmuir, 2010. **26**(20): p. 16090-16095.
168. Tsai, P., M.H. Hendrix, R.R. Dijkstra, L. Shui, and D. Lohse, *Microscopic structure influencing macroscopic splash at high Weber number*. Soft Matter, 2011. **7**(24): p. 11325-11333.
169. Thoroddsen, S., K. Takehara, and T. Etoh, *Micro-splashing by drop impacts*. Journal of Fluid Mechanics, 2012. **706**: p. 560-570.
170. Bischofberger, I., K.W. Mauser, and S.R. Nagel, *Seeing the invisible—Air vortices around a splashing drop*. Physics of Fluids (1994-present), 2013. **25**(9): p. 091110.
171. Kim, H., U. Park, C. Lee, H. Kim, M.H. Kim, and J. Kim, *Drop splashing on a rough surface: How surface morphology affects splashing threshold*. Applied Physics Letters, 2014. **104**(16): p. 161608.

172. Li, H., S. Mei, L. Wang, Y. Gao, and J. Liu, *Splashing phenomena of room temperature liquid metal droplet striking on the pool of the same liquid under ambient air environment*. International Journal of Heat and Fluid Flow, 2014. **47**: p. 1-8.
173. Liu, Y., P. Tan, and L. Xu, *Kelvin–Helmholtz instability in an ultrathin air film causes drop splashing on smooth surfaces*. Proceedings of the National Academy of Sciences, 2015. **112**(11): p. 3280-3284.
174. Liu, Y., P. Tan, and L. Xu, *Compressible air entrapment in high-speed drop impacts on solid surfaces*. Journal of Fluid Mechanics, 2013. **716**: p. R9.
175. Tran, T., H. de Maleprade, C. Sun, and D. Lohse, *Air entrainment during impact of droplets on liquid surfaces*. Journal of Fluid Mechanics, 2013. **726**: p. R3.
176. Thoroddsen, S., T. Etoh, and K. Takehara, *Air entrapment under an impacting drop*. Journal of Fluid Mechanics, 2003. **478**: p. 125-134.
177. Hicks, P.D. and R. Purvis, *Air cushioning and bubble entrapment in three-dimensional droplet impacts*. Journal of Fluid Mechanics, 2010. **649**: p. 135-163.
178. Wang, A.-B., C.-C. Kuan, and P.-H. Tsai, *Do we understand the bubble formation by a single drop impacting upon liquid surface?* Physics of Fluids (1994-present), 2013. **25**(10): p. 101702.
179. Vakarelski, I.U., D.Y. Chan, J.O. Marston, and S.T. Thoroddsen, *Dynamic Air Layer on Textured Superhydrophobic Surfaces*. Langmuir, 2013. **29**(35): p. 11074-11081.
180. van der Veen, R.C., M.H. Hendrix, T. Tran, C. Sun, P.A. Tsai, and D. Lohse, *How microstructures affect air film dynamics prior to drop impact*. Soft matter, 2014. **10**(21): p. 3703-3707.
181. Pittoni, P.G., R.-J. Wang, T.-S. Yu, and S.-Y. Lin, *Occurrence and formation mechanisms of bubbles entrapped into water drops impinging on graphite*. Journal of the Taiwan Institute of Chemical Engineers, 2014.
182. Leidenfrost, J.G., *On the fixation of water in diverse fire*. International Journal of Heat and Mass Transfer, 1966. **9**(11): p. 1153-1166.
183. Curzon, F., *The Leidenfrost phenomenon*. American Journal of Physics, 1978. **46**(8): p. 825-828.
184. Bernardin, J.D. and I. Mudawar, *Validation of the quench factor technique in predicting hardness in heat treatable aluminum alloys*. International journal of heat and mass transfer, 1995. **38**(5): p. 863-873.
185. Bernardin, J. and I. Mudawar, *The Leidenfrost point: experimental study and assessment of existing models*. Journal of Heat Transfer, 1999. **121**(4): p. 894-903.
186. Carey, V.P., *Liquid Vapor Phase Change Phenomena: An Introduction to the Thermophysics of Vaporization and Condensation Processes in Heat Transfer Equipment, Second Edition*. 2007: Taylor & Francis.
187. Rein, M., *Interactions between drops and hot surfaces*. 2002: Springer.
188. Mahadevan, L. and Y. Pomeau, *Rolling droplets*. Physics of Fluids (1994-present), 1999. **11**(9): p. 2449-2453.
189. Sharp, D.H., *An overview of Rayleigh-Taylor instability*. Physica D: Nonlinear Phenomena, 1984. **12**(1): p. 3-18.
190. Zhang, N. and W.-J. Yang, *Evaporation and explosion of liquid drops on a heated surface*. Experiments in Fluids, 1983. **1**(2): p. 101-111.
191. Anders, K., N. Roth, and A. Frohn, *The velocity change of ethanol droplets during collision with a wall analysed by image processing*. Experiments in fluids, 1993. **15**(2): p. 91-96.
192. Chaves, H., A.M. Kubitzek, and F. Obermeier, *Dynamic processes occurring during the spreading of thin liquid films produced by drop impact on hot walls*. International journal of heat and fluid flow, 1999. **20**(5): p. 470-476.

193. Karl, A. and A. Frohn, *Experimental investigation of interaction processes between droplets and hot walls*. Physics of Fluids (1994-present), 2000. **12**(4): p. 785-796.
194. Wang, A.-B., C.-H. Lin, and C.-C. Chen, *The critical temperature of dry impact for tiny droplet impinging on a heated surface*. Physics of Fluids (1994-present), 2000. **12**(6): p. 1622-1625.
195. Shi, M., T. Bai, and J. Yu, *Dynamic behavior and heat transfer of a liquid droplet impinging on a solid surface*. Experimental thermal and fluid science, 1993. **6**(2): p. 202-207.
196. Labeish, V., *Thermohydrodynamic study of a drop impact against a heated surface*. Experimental thermal and fluid science, 1994. **8**(3): p. 181-194.
197. Chandra, S. and S. Aziz, *Leidenfrost evaporation of liquid nitrogen droplets*. Journal of heat transfer, 1994. **116**(4): p. 999-1006.
198. Chen, R.-H., S.-L. Chiu, and T.-H. Lin, *On the collision behaviors of a diesel drop impinging on a hot surface*. Experimental Thermal and Fluid Science, 2007. **32**(2): p. 587-595.
199. Chen, R.-H., S.-L. Chiu, and T.-H. Lin, *Resident time of a compound drop impinging on a hot surface*. Applied thermal engineering, 2007. **27**(11): p. 2079-2085.
200. Chen, R.-H. and Y.-L. Huang, *Collision of a liquid drop on the edge region of a plate heated above the Leidenfrost temperature*. Experiments in fluids, 2009. **47**(2): p. 223-237.
201. Bernardin, J.D., C.J. Stebbins, and I. Mudawar, *Effects of surface roughness on water droplet impact history and heat transfer regimes*. International Journal of Heat and Mass Transfer, 1996. **40**(1): p. 73-88.
202. Bernardin, J.D., C.J. Stebbins, and I. Mudawar, *Mapping of impact and heat transfer regimes of water drops impinging on a polished surface*. International Journal of Heat and Mass Transfer, 1997. **40**(2): p. 247-267.
203. Bernardin, J.D. and I. Mudawar, *A cavity activation and bubble growth model of the Leidenfrost point*. Journal of heat transfer, 2002. **124**(5): p. 864-874.
204. Bernardin, J.D. and I. Mudawar, *A Leidenfrost point model for impinging droplets and sprays*. Journal of heat transfer, 2004. **126**(2): p. 272-278.
205. Bernardin, J.D. and I. Mudawar, *Transition boiling heat transfer of droplet streams and sprays*. Journal of Heat Transfer, 2007. **129**(11): p. 1605-1610.
206. Baumeister, K. and F. Simon, *Leidenfrost temperature—its correlation for liquid metals, cryogenes, hydrocarbons, and water*. Journal of Heat Transfer, 1973. **95**(2): p. 166-173.
207. Fujimoto, H., S. Watanabe, T. Okamoto, T. Hama, and H. Takuda, *Photographic study of hydrodynamics of drops of aqueous polymer solution impinging on hot solid*. Experimental Thermal and Fluid Science, 2015. **60**: p. 66-74.
208. Dhir, V., *Boiling heat transfer*. Annual review of fluid mechanics, 1998. **30**(1): p. 365-401.
209. Kocamustafaogullari, G. and M. Ishii, *Interfacial area and nucleation site density in boiling systems*. International Journal of Heat and Mass Transfer, 1983. **26**(9): p. 1377-1387.
210. Yang, S. and R. Kim, *A mathematical model of the pool boiling nucleation site density in terms of the surface characteristics*. International journal of heat and mass transfer, 1988. **31**(6): p. 1127-1135.
211. Barthau, G., *Active nucleation site density and pool boiling heat transfer—an experimental study*. International journal of heat and mass transfer, 1992. **35**(2): p. 271-278.
212. Hibiki, T. and M. Ishii, *Active nucleation site density in boiling systems*. International Journal of Heat and Mass Transfer, 2003. **46**(14): p. 2587-2601.
213. Benjamin, R. and A. Balakrishnan, *Nucleation site density in pool boiling of saturated pure liquids: effect of surface microroughness and surface and liquid physical properties*. Experimental thermal and fluid science, 1997. **15**(1): p. 32-42.
214. Basu, N., G.R. Warrier, and V.K. Dhir, *Onset of nucleate boiling and active nucleation site density during subcooled flow boiling*. Journal of heat transfer, 2002. **124**(4): p. 717-728.

215. Theofanous, T., J. Tu, A. Dinh, and T.-N. Dinh, *The boiling crisis phenomenon: Part I: nucleation and nucleate boiling heat transfer*. Experimental thermal and fluid science, 2002. **26**(6): p. 775-792.
216. Scharifker, B. and J. Mostany, *Three-dimensional nucleation with diffusion controlled growth: Part I. Number density of active sites and nucleation rates per site*. Journal of electroanalytical chemistry and interfacial electrochemistry, 1984. **177**(1): p. 13-23.
217. Mostany, J., J. Mozota, and B. Scharifker, *Three-dimensional nucleation with diffusion controlled growth: Part II. The nucleation of lead on vitreous carbon*. Journal of electroanalytical chemistry and interfacial electrochemistry, 1984. **177**(1): p. 25-37.
218. Zhang, J. and R.M. Manglik, *Additive adsorption and interfacial characteristics of nucleate pool boiling in aqueous surfactant solutions*. Journal of heat transfer, 2005. **127**(7): p. 684-691.
219. Dhir, V.K., *Mechanistic prediction of nucleate boiling heat transfer—achievable or a hopeless task?* Journal of Heat Transfer, 2006. **128**(1): p. 1-12.
220. Qi, Y. and J.F. Klausner, *Comparison of nucleation site density for pool boiling and gas nucleation*. Journal of heat transfer, 2006. **128**(1): p. 13-20.
221. Tarrad, A.H., *A Correlation for the Prediction of Nucleate Pool Boiling Performance of Pure Liquids from Enhanced Tubes*. JJMIE, 2011. **5**(2).
222. Sun, T. and W. Li, *Three-dimensional numerical simulation of nucleate boiling bubble by lattice Boltzmann method*. Computers & Fluids, 2013. **88**: p. 400-409.
223. Roh, H.-S., *Heat transfer mechanisms in pool boiling*. International Journal of Heat and Mass Transfer, 2014. **68**: p. 332-342.
224. Cheung, S., S. Vahaji, G. Yeoh, and J. Tu, *Modeling subcooled flow boiling in vertical channels at low pressures—Part 1: Assessment of empirical correlations*. International Journal of Heat and Mass Transfer, 2014. **75**: p. 736-753.
225. Yeoh, G., S. Vahaji, S. Cheung, and J. Tu, *Modeling subcooled flow boiling in vertical channels at low pressures—Part 2: Evaluation of mechanistic approach*. International Journal of Heat and Mass Transfer, 2014. **75**: p. 754-768.
226. Misyura, S.Y., *Nucleate boiling in bidistillate droplets*. International Journal of Heat and Mass Transfer, 2014. **71**: p. 197-205.
227. Attinger, D., C. Frankiewicz, A.R. Betz, T.M. Schutzius, R. Ganguly, A. Das, C.-J. Kim, and C.M. Megaridis, *Surface engineering for phase change heat transfer: A review*. MRS Energy & Sustainability, 2014. **1**: p. E4.
228. Castanet, G., T. Lienart, and F. Lemoine, *Dynamics and temperature of droplets impacting onto a heated wall*. International Journal of Heat and Mass Transfer, 2009. **52**(3): p. 670-679.
229. Orzechowski, T. and S. Wciślik, *Instantaneous heat transfer for large drops levitating over a hot surface*. International Journal of Heat and Mass Transfer, 2014. **73**: p. 110-117.
230. Biance, A.-L., C. Pirat, and C. Ybert, *Drop fragmentation due to hole formation during Leidenfrost impact*. Physics of Fluids (1994-present), 2011. **23**(2): p. 022104.
231. Celestini, F. and G. Kirstetter, *Effect of an electric field on a Leidenfrost droplet*. Soft Matter, 2012. **8**(22): p. 5992-5995.
232. Hashmi, A., Y. Xu, B. Coder, P.A. Osborne, J. Spafford, G.E. Michael, G. Yu, and J. Xu, *Leidenfrost levitation: beyond droplets*. Scientific reports, 2012. **2**.
233. Weickgenannt, C.M., Y. Zhang, S. Sinha-Ray, I.V. Roisman, T. Gambaryan-Roisman, C. Tropea, and A.L. Yarin, *Inverse-Leidenfrost phenomenon on nanofiber mats on hot surfaces*. Physical Review E, 2011. **84**(3): p. 036310.
234. Weickgenannt, C.M., Y. Zhang, A.N. Lembach, I.V. Roisman, T. Gambaryan-Roisman, A.L. Yarin, and C. Tropea, *Nonisothermal drop impact and evaporation on polymer nanofiber mats*. Physical Review E, 2011. **83**(3): p. 036305.
235. Pomeau, Y., M. Le Berre, F. Celestini, and T. Frisch, *The Leidenfrost effect: From quasi-spherical droplets to puddles*. Comptes Rendus Mecanique, 2012. **340**(11): p. 867-881.

236. Kruse, C., T. Anderson, C. Wilson, C. Zuhlke, D. Alexander, G. Gogos, and S. Ndao, *Extraordinary shifts of the Leidenfrost temperature from multiscale micro/nanostructured surfaces*. *Langmuir*, 2013. **29**(31): p. 9798-9806.
237. Piroird, K., B.D. Texier, C. Clanet, and D. Quéré, *Reshaping and capturing Leidenfrost drops with a magnet*. *Physics of Fluids (1994-present)*, 2013. **25**(3): p. 032108.
238. Celestini, F., T. Frisch, and Y. Pomeau, *Room temperature water Leidenfrost droplets*. *Soft Matter*, 2013. **9**(40): p. 9535-9538.
239. Orzechowski, T. and S. Wcislik, *Analysis of D2-law in case of Leidenfrost drop evaporation*. *Experimental Thermal and Fluid Science*, 2014. **59**(0): p. 230-237.
240. Agapov, R.L., J.B. Boreyko, D.P. Briggs, B.R. Srijanto, S.T. Retterer, C.P. Collier, and N.V. Lavrik, *Asymmetric Wettability of Nanostructures Directs Leidenfrost Droplets*. *ACS Nano*, 2013. **8**(1): p. 860-867.
241. Lastakowski, H., F. Boyer, A.-L. Biance, C. Pirat, and C. Ybert, *Bridging local to global dynamics of drop impact onto solid substrates*. *Journal of Fluid Mechanics*, 2014. **747**: p. 103-118.
242. Nair, H., H.J. Staat, T. Tran, A. van Houselt, A. Prosperetti, D. Lohse, and C. Sun, *The Leidenfrost temperature increase for impacting droplets on carbon-nanofiber surfaces*. *Soft matter*, 2014. **10**(13): p. 2102-2109.
243. Tran, T., H.J. Staat, A. Susarrey-Arce, T.C. Foertsch, A. van Houselt, H.J. Gardeniers, A. Prosperetti, D. Lohse, and C. Sun, *Droplet impact on superheated micro-structured surfaces*. *Soft Matter*, 2013. **9**(12): p. 3272-3282.
244. Kwon, H.-m., J.C. Bird, and K.K. Varanasi, *Increasing Leidenfrost point using micro-nano hierarchical surface structures*. *Applied Physics Letters*, 2013. **103**(20): p. 201601.
245. Celestini, F., T. Frisch, A. Cohen, C. Raufaste, L. Duchemin, and Y. Pomeau, *Two dimensional Leidenfrost droplets in a Hele-Shaw cell*. *Physics of Fluids (1994-present)*, 2014. **26**(3): p. 032103.
246. Maquet, L., P. Colinet, and S. Dorbolo, *Organization of microbeads in Leidenfrost drops*. *Soft matter*, 2014. **10**(23): p. 4061-4066.
247. Liu, F., G. Ghigliotti, J.J. Feng, and C.-H. Chen, *Self-propelled jumping upon drop coalescence on Leidenfrost surfaces*. *Journal of Fluid Mechanics*, 2014. **752**: p. 22-38.
248. Moon, J.H., D.Y. Kim, and S.H. Lee, *Spreading and receding characteristics of a non-Newtonian droplet impinging on a heated surface*. *Experimental Thermal and Fluid Science*, 2014. **57**: p. 94-101.
249. Shin, D.H., C.K. Choi, Y.T. Kang, and S.H. Lee, *Local aggregation characteristics of a nanofluid droplet during evaporation*. *International Journal of Heat and Mass Transfer*, 2014. **72**: p. 336-344.
250. Vakarelski, I.U., D.Y. Chan, and S.T. Thoroddsen, *Leidenfrost vapour layer moderation of the drag crisis and trajectories of superhydrophobic and hydrophilic spheres falling in water*. *Soft matter*, 2014.
251. Pan, Z., J.A. Weibel, and S.V. Garimella, *Influence of Surface Wettability on Transport Mechanisms Governing Water Droplet Evaporation*. *Langmuir*, 2014. **30**(32): p. 9726-9730.
252. Shahriari, A., J. Wurz, and V. Bahadur, *Heat transfer enhancement accompanying Leidenfrost state suppression at ultrahigh temperatures*. *Langmuir*, 2014. **30**(40): p. 12074-12081.
253. Orejon, D., K. Sefiane, and Y. Takata, *Effect of ambient pressure on Leidenfrost temperature*. *Physical Review E*, 2014. **90**(5): p. 053012.
254. Paul, G., P.K. Das, and I. Manna, *Droplet Oscillation and Pattern Formation during Leidenfrost Phenomenon*. *Experimental Thermal and Fluid Science*, 2014.
255. Negeed, E.-S.R., M. Albeirutty, and Y. Takata, *Dynamic behavior of micrometric single water droplets impacting onto heated surfaces with TiO₂ hydrophilic coating*. *International Journal of Thermal Sciences*, 2014. **79**(0): p. 1-17.

256. Yong Park Ji, G.A., King William P, Cahill, David G. , *Droplet Impingement and Vapor Layer Formation on Hot Hydrophobic Surfaces*. Journal of Heat Transfer, 2014. **136**(9).
257. Dash, S. and S.V. Garimella, *Droplet evaporation on heated hydrophobic and superhydrophobic surfaces*. Physical Review E, 2014. **89**(4): p. 042402.
258. Xie, H. and Z. Zhou, *A model for droplet evaporation near Leidenfrost point*. International Journal of Heat and Mass Transfer, 2007. **50**(25): p. 5328-5333.
259. Myers, T. and J. Charpin, *A mathematical model of the Leidenfrost effect on an axisymmetric droplet*. Physics of Fluids (1994-present), 2009. **21**(6): p. 063101.
260. Celata, G.P., M. Cumo, A. Mariani, and G. Zummo, *Visualization of the impact of water drops on a hot surface: effect of drop velocity and surface inclination*. Heat and mass transfer, 2006. **42**(10): p. 885-890.
261. Wachters, L., H. Bonne, and H. Van Nouhuis, *The heat transfer from a hot horizontal plate to sessile water drops in the spheroidal state*. Chemical Engineering Science, 1966. **21**(10): p. 923-936.
262. Wachters, L. and N. Westerling, *The heat transfer from a hot wall to impinging water drops in the spheroidal state*. Chemical Engineering Science, 1966. **21**(11): p. 1047-1056.
263. Spiegler, P., J. Hopenfeld, M. Silberberg, C. Bumpus Jr, and A. Norman, *Onset of stable film boiling and the foam limit*. International Journal of Heat and Mass Transfer, 1963. **6**(11): p. 987-989.
264. Eckert, E.R.G. and R.M. Drake Jr, *Analysis of heat and mass transfer*. 1987.
265. Patel, B.M. and K. Bell, *The Leidenfrost phenomenon for extended liquid masses*. 1965, Oklahoma State University.
266. Xiong, T. and M. Yuen, *Evaporation of a liquid droplet on a hot plate*. International journal of heat and mass transfer, 1991. **34**(7): p. 1881-1894.
267. Rayleigh, L., *On the capillary phenomena of jets*. Proceedings of the Royal Society of London, 1879. **29**(196-199): p. 71-97.
268. Dupeux, G., M. Le Merrer, G. Lagubeau, C. Clanet, S. Hardt, and D. Quéré, *Viscous mechanism for Leidenfrost propulsion on a ratchet*. EPL (Europhysics Letters), 2011. **96**(5): p. 58001.
269. Feng, R., W. Zhao, X. Wu, and Q. Xue, *Ratchet composite thin film for low-temperature self-propelled Leidenfrost droplet*. Journal of colloid and interface science, 2012. **367**(1): p. 450-454.
270. Ok, J.T., E. Lopez-Oña, D.E. Nikitopoulos, H. Wong, and S. Park, *Propulsion of droplets on micro-and sub-micron ratchet surfaces in the Leidenfrost temperature regime*. Microfluidics and nanofluidics, 2011. **10**(5): p. 1045-1054.
271. Cousins, T.R., R.E. Goldstein, J.W. Jaworski, and A.I. Pesci, *A ratchet trap for Leidenfrost drops*. Journal of Fluid Mechanics, 2012. **696**: p. 215-227.
272. Nagai, N. and S. Nishio, *Leidenfrost temperature on an extremely smooth surface*. Experimental thermal and fluid science, 1996. **12**(3): p. 373-379.
273. Shirota, M., M.A. van Limbeek, C. Sun, A. Prosperetti, and D. Lohse, *Dynamic Leidenfrost effect: relevant time and length scales*. Physical review letters, 2016. **116**(6): p. 064501.
274. van Limbeek, M.A., M. Shirota, P. Sleutel, C. Sun, A. Prosperetti, and D. Lohse, *Vapour cooling of poorly conducting hot substrates increases the dynamic Leidenfrost temperature*. International journal of heat and mass transfer, 2016. **97**: p. 101-109.
275. Bertola, V., *An impact regime map for water drops impacting on heated surfaces*. International Journal of Heat and Mass Transfer, 2015. **85**: p. 430-437.
276. Grishaev, V., C.S. Iorio, F. Dubois, and A. Amirfazli, *Complex Drop Impact Morphology*. Langmuir, 2015. **31**(36): p. 9833-9844.

277. Clavijo, C.E., J. Crockett, and D. Maynes, *Effects of isotropic and anisotropic slip on droplet impingement on a superhydrophobic surface*. Physics of Fluids (1994-present), 2015. **27**(12): p. 122104.
278. Wu, J., J. Huang, and W. Yan, *Lattice Boltzmann investigation of droplets impact behaviors onto a solid substrate*. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 2015. **484**: p. 318-328.
279. Castanet, G., O. Caballina, and F. Lemoine, *Drop spreading at the impact in the Leidenfrost boiling*. Physics of Fluids (1994-present), 2015. **27**(6): p. 063302.
280. Hamdan, K.S., D.-E. Kim, and S.-K. Moon, *Droplets behavior impacting on a hot surface above the Leidenfrost temperature*. Annals of Nuclear Energy, 2015. **80**: p. 338-347.
281. Chabičovský, M., M. Hnizdil, A. Tseng, and M. Raudenský, *Effects of oxide layer on Leidenfrost temperature during spray cooling of steel at high temperatures*. International Journal of Heat and Mass Transfer, 2015. **88**: p. 236-246.
282. Raux, P.S., G. Dupeux, C. Clanet, and D. Quéré, *Successive instabilities of confined Leidenfrost puddles*. EPL (Europhysics Letters), 2015. **112**(2): p. 26002.
283. Staat, H.J., T. Tran, B. Geerdink, G. Riboux, C. Sun, J.M. Gordillo, and D. Lohse, *Phase diagram for droplet impact on superheated surfaces*. Journal of fluid mechanics, 2015. **779**: p. R3.
284. Riboux, G. and J.M. Gordillo, *The diameters and velocities of the droplets ejected after splashing*. Journal of Fluid Mechanics, 2015. **772**: p. 630-648.
285. Ng, B.T., Y.M. Hung, and M.K. Tan, *Suppression of the Leidenfrost effect via low frequency vibrations*. Soft matter, 2015. **11**(4): p. 775-784.
286. Ng, B.T., Y.M. Hung, and M.K. Tan, *Acoustically-controlled Leidenfrost droplets*. Journal of colloid and interface science, 2016. **465**: p. 26-32.
287. Gauthier, A., J.C. Bird, C. Clanet, and D. Quéré, *Aerodynamic Leidenfrost effect*. Physical Review Fluids, 2016. **1**(8): p. 084002.
288. Raufaste, C., Y. Bouret, and F. Celestini, *Reactive Leidenfrost droplets*. EPL (Europhysics Letters), 2016. **114**(4): p. 46005.
289. Liang, G., X. Mu, Y. Guo, S. Shen, S. Quan, and J. Zhang, *Contact vaporization of an impacting drop on heated surfaces*. Experimental Thermal and Fluid Science, 2016. **74**: p. 73-80.
290. Zhang, W., T. Yu, J. Fan, W. Sun, and Z. Cao, *Droplet impact behavior on heated micro-patterned surfaces*. Journal of Applied Physics, 2016. **119**(11): p. 114901.
291. Moon, J.H., M. Cho, and S.H. Lee, *Dynamic wetting and heat transfer characteristics of a liquid droplet impinging on heated textured surfaces*. International Journal of Heat and Mass Transfer, 2016. **97**: p. 308-317.
292. Wildeman, S. and C. Sun, *Electric field makes Leidenfrost droplets take a leap*. Soft Matter, 2016. **12**(48): p. 9622-9632.
293. Jung, J., S. Jeong, and H. Kim, *Investigation of single-droplet/wall collision heat transfer characteristics using infrared thermometry*. International Journal of Heat and Mass Transfer, 2016. **92**: p. 774-783.
294. Vakarelski, I.U., J.D. Berry, D.Y. Chan, and S.T. Thoroddsen, *Leidenfrost vapor layers reduce drag without the crisis in high viscosity liquids*. Physical Review Letters, 2016. **117**(11): p. 114503.
295. Maquet, L., M. Brandenbourger, B. Sobac, A.-L. Biance, P. Colinet, and S. Dorbolo, *Leidenfrost drops: Effect of gravity*. EPL (Europhysics Letters), 2015. **110**(2): p. 24001.
296. Shi, M., X. Ji, S. Feng, Q. Yang, T.J. Lu, and F. Xu, *Self-Propelled Hovercraft Based on Cold Leidenfrost Phenomenon*. Scientific Reports, 2016. **6**.
297. Takata, Y., M. Kohno, S. Hidaka, T. Wakui, A.I. Majid, H.Y. Kuntoro, and A. Widyaparaga, *The effects of the surface roughness on the dynamic behavior of the successive micrometric*

- droplets impacting onto inclined hot surfaces*. International Journal of Heat and Mass Transfer, 2016. **101**: p. 1217-1226.
298. Chen, S. and V. Bertola, *The impact of viscoplastic drops on a heated surface in the Leidenfrost regime*. Soft Matter, 2016. **12**(36): p. 7624-7631.
299. Chen, S. and V. Bertola, *Jumps, somersaults, and symmetry breaking in Leidenfrost drops*. Physical Review E, 2016. **94**(2): p. 021102.
300. Cordeiro, J. and S. Desai, *The Leidenfrost Effect at the Nanoscale*. Journal of Micro and Nano-Manufacturing, 2016. **4**(4): p. 041001.
301. Villegas, L.R., S. Tanguy, G. Castanet, O. Caballina, and F. Lemoine, *Direct numerical simulation of the impact of a droplet onto a hot surface above the Leidenfrost temperature*. International Journal of Heat and Mass Transfer, 2017. **104**: p. 1090-1109.
302. Qiu, L., S. Dubey, F.H. Choo, and F. Duan, *The transitions of time-independent spreading diameter and splashing angle when a droplet train impinging onto a hot surface*. RSC Advances, 2016. **6**(17): p. 13644-13652.
303. Axelrod, D., T.P. Burghardt, and N.L. Thompson, *Total internal reflection fluorescence*. Annual review of biophysics and bioengineering, 1984. **13**(1): p. 247-268.
304. Huang, P., J.S. Guasto, and K.S. Breuer, *Direct measurement of slip velocities using three-dimensional total internal reflection velocimetry*. Journal of Fluid Mechanics, 2006. **566**: p. 447-464.
305. Sadr, R., H. Li, and M. Yoda, *Impact of hindered Brownian diffusion on the accuracy of particle-image velocimetry using evanescent-wave illumination*. Experiments in fluids, 2005. **38**(1): p. 90-98.
306. Kolinski, J., L. Mahadevan, and S. Rubinstein, *Drops can bounce from perfectly hydrophilic surfaces*. EPL (Europhysics Letters), 2014. **108**(2): p. 24001.
307. Zettner, C. and M. Yoda, *Particle velocity field measurements in a near-wall flow using evanescent wave illumination*. Experiments in Fluids, 2003. **34**(1): p. 115-121.
308. Axelrod, D., *Total internal reflection fluorescence microscopy in cell biology*. Traffic, 2001. **2**(11): p. 764-774.
309. Kihm, K., A. Banerjee, C. Choi, and T. Takagi, *Near-wall hindered Brownian diffusion of nanoparticles examined by three-dimensional ratiometric total internal reflection fluorescence microscopy (3-D R-TIRFM)*. Experiments in Fluids, 2004. **37**(6): p. 811-824.
310. Rubinstein, S.M., G. Cohen, and J. Fineberg, *Detachment fronts and the onset of dynamic friction*. Nature, 2004. **430**(7003): p. 1005-1009.
311. Hecht, E., *Optics*. 2002: Addison-Wesley.
312. Kedenburg, S., M. Vieweg, T. Gissibl, and H. Giessen, *Linear refractive index and absorption measurements of nonlinear optical liquids in the visible and near-infrared spectral region*. Optical Materials Express, 2012. **2**(11): p. 1588-1611.
313. Fontao, M. and M. Iglesias, *Temperature dependence of changes of refractive index in methyl acetate, ethanol and 2-propanol mixtures*. Physics and Chemistry of Liquids, 2000. **38**(6): p. 623-633.
314. Kamikawachi, R., I. Abe, A. Paterno, H. Kalinowski, M. Muller, J. Pinto, and J. Fabris, *Determination of thermo-optic coefficient in liquids with fiber Bragg grating refractometer*. Optics Communications, 2008. **281**(4): p. 621-625.
315. Wohlfarth, C., *Refractive index of ethanol*, in *Refractive Indices of Pure Liquids and Binary Liquid Mixtures (Supplement to III/38)*. 2008, Springer. p. 108-115.
316. Tsegaw, A.A., F.-J. Shiou, and S.-P. Lin, *Ultra-precision polishing of N-Bk7 using an innovative self-propelled abrasive fluid multi-jet polishing tool*. Machining Science and Technology, 2015. **19**(2): p. 262-285.
317. Lide, D.R., *CRC handbook of chemistry and physics*. Vol. 85. 2004: CRC press.

318. Atherton, T.J. and D.J. Kerbyson, *Size invariant circle detection*. Image and Vision computing, 1999. **17**(11): p. 795-803.
319. Davies, E.R., *Machine vision: theory, algorithms, practicalities*. 2004: Elsevier.
320. Yuen, H., J. Princen, J. Illingworth, and J. Kittler, *Comparative study of Hough transform methods for circle finding*. Image and Vision Computing, 1990. **8**(1): p. 71-77.
321. Gonzalez, R.C. and R.E. Woods, *Digital image processing*. 2002, Prentice hall Upper Saddle River, NJ:.
322. Gonzalez, R.C., R.E. Woods, and S.L. Eddins, *Digital image processing using MATLAB*. Upper Saddle River, N. J: Pearson Prentice Hall, 2004.
323. Pedrotti, L.S., *Basic physical optics*. Fundamentals of Photonics, Chandra Roychoudhuri, Ed. Bellingham: SPIE Press Book, 2008: p. 137.
324. Wang, C. and V. Dhir, *Effect of surface wettability on active nucleation site density during pool boiling of water on a vertical surface*. Journal of Heat Transfer, 1993. **115**(3): p. 659-669.
325. Cornwell, K. and E. Brown. *BOILING SURFACE TOPOGRAPHY!* in *6th Int. Heat Transfer Conference*. Toronto, Canada. 1978.
326. Crystran UK Ltd. *Optical Glass (N-BK7 and others)*. 2012; Available from: <http://www.crystran.co.uk/optical-materials/optical-glass-n-bk7-and-others>.
327. Dobrovinskaya, E.R., L.A. Lytvynov, and V. Pishchik, *Sapphire: material, manufacturing, applications*. 2009: Springer Science & Business Media.
328. Yoshizawa, T., *Handbook of optical metrology: Principles and Applications*. 2009: CRC Press.
329. Faghri, A. and Y. Zhang, *Transport phenomena in multiphase systems*. 2006: Academic Press.
330. Khavari, M., C. Sun, D. Lohse, and T. Tran, *Fingering patterns during droplet impact on heated surfaces*. Soft matter, 2015. **11**(17): p. 3298-3303.
331. Rein, M., *Drop-surface interactions*. 2002: Springer.
332. Cole, R., *Boiling nucleation*. Advances in Heat Transfer, 1974. **10**: p. 85-166.
333. Ambrose, D., J. Counsell, I. Lawrenson, and G. Lewis, *Thermodynamic properties of organic oxygen compounds XLVII. Pressure, volume, temperature relations and thermodynamic properties of propan-2-ol*. The Journal of Chemical Thermodynamics, 1978. **10**(11): p. 1033-1043.
334. Hoke Jr, B.C. and J.C. Chen, *Binary aqueous-organic surface tension temperature dependence*. Journal of Chemical and Engineering Data, 1991. **36**(3): p. 322-326.
335. Bank, D.D. *Surface Tension of 2-Propanol*. 2016; Available from: http://www.ddbst.com/en/EED/PCP/SFT_C95.php.
336. *3M Novec Engineered Fluid HFE-7100 Product Information*. 2016; Available from: http://www.3m.com/3M/en_US/novec/products/engineered-fluids/.
337. Rausch, M.H., L. Kretschmer, S. Will, A. Leipertz, and A.P. Fröba, *Density, Surface Tension, and Kinematic Viscosity of Hydrofluoroethers HFE-7000, HFE-7100, HFE-7200, HFE-7300, and HFE-7500*. Journal of Chemical & Engineering Data, 2015. **60**(12): p. 3759-3765.
338. *3M Novec Engineered Fluid HFE-7300 Product Information*. Available from: http://www.3m.com/3M/en_US/novec/products/engineered-fluids/.
339. Manzello, S.L. and J.C. Yang, *An experimental study of high Weber number impact of methoxy-nonafluorobutane C₄F₉OCH₃ (HFE-7100) and n-heptane droplets on a heated solid surface*. International journal of heat and mass transfer, 2002. **45**(19): p. 3961-3971.
340. Avedisian, C. and J. Koplik, *Leidenfrost boiling of methanol droplets on hot porous/ceramic surfaces*. International journal of heat and mass transfer, 1987. **30**(2): p. 379-393.
341. Hassebrook, A., C. Kruse, C. Wilson, T. Anderson, C. Zuhlke, D. Alexander, G. Gogos, and S. Ndao, *Effects of Droplet Diameter and Fluid Properties on the Leidenfrost Temperature of*

- Polished and Micro/Nanostructured Surfaces*. Journal of Heat Transfer, 2016. **138**(5): p. 051501.
342. Bank, D.D. *Benzene Density*. 2016; Available from: <http://ddbonline.ddbst.de/>.
343. Bank, D.D. *Benzene Vapor Pressure*. 2016; Available from: <http://ddbonline.ddbst.com>.
344. Bank, D.D. *Benzene Surface Tension*. 2016; Available from: <http://ddbonline.ddbst.com/>.