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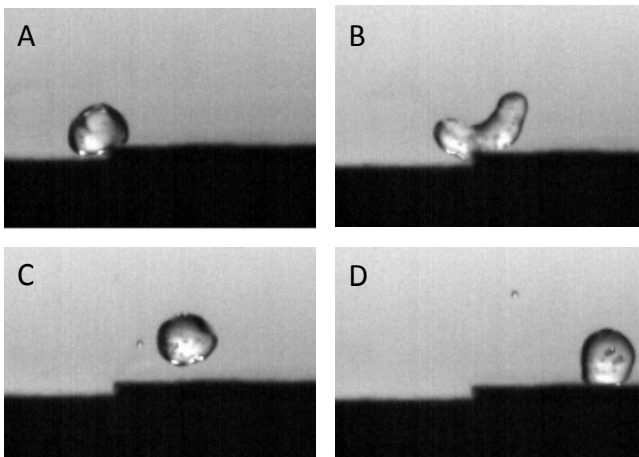
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**Jumping Leidenfrost droplets**

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2012





# JUMPING LEIDENFROST DROPLETS

MASTER THESIS

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# Abstract

A leidenfrost droplet forms when a liquid droplet is placed on a very hot surface. In the case that there is a thin layer of insulating material as a step on the substrate, under specific conditions Leidenfrost droplets interact with the edge of the step. The result of this interaction is sometimes lateral detachment or jumping from the surface. Specifically, the best jumping can be observed when liquid nitrogen droplets move on a brass substrate with a piece of plastic tape as a step. This thesis experimentally studies the specific physics of the jumping process.

We examined the effect of the step material, liquid property and the step height on the jumping process. We hypothesize that the Marangoni effect, due to thermal gradients contributes to this type of motion. This is evident by comparing the estimated Marangoni flow and measurement flow rate.

Key words: Leidenfrost the effect, Marangoni effect, Surface tension, Liquid solid interaction, Heat transfer



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# List of acronyms and notations

$\alpha$	Change in surface tension due to temperature
$\gamma$	Surface tension
$\gamma_0$	Specific value of surface tension for each liquid
$\gamma_{LG}$	liquid-gas interfacial tension
$\gamma_{SG}$	Solid-gas interfacial tension
$\gamma_{SL}$	Solid-liquid interfacial tension
$\kappa$	Thermal diffusivity
$\mu$	Dynamic viscosity
$\nu$	Kinematic viscosity
$\rho$	Density
$B_O$	Bound number
$CM$	Center of the mass
$g$	Gravity



$L$	Heat latent
$Re$	Reynolds number
$S$	spread coefficient
$T_c$	Critical temperature
$\alpha_t$	Coefficient of thermal expansion
$\theta_c$	Contact angle
$L_c$	Capillary length
$T$	Temperature



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# Chapter 1

## Introduction

The Leidenfrost phenomenon is observed when a liquid comes in contact with a surface significantly hotter than the boiling point of the liquid. In this case, rapid evaporation of the liquid at the contact point, provides a vapor layer between the liquid and surface. This vapor layer prevents the liquid to touch the surface and insulates it from the heat transfer. Particularly a very small volume of liquid, makes a spherical droplet over a Leidenfrost surface which is known as a Leidenfrost droplet. A Leidenfrost droplet levitates over its own vapor film by supporting of vapor pressure gradient in the vapor flowing underneath the droplet. The vapor flow beneath the droplet is a laminar flow [1]. Leidenfrost droplets perform a self-propel motion on an asymmetric saw-tooth shape (ratchetlike) surface. The self-propel motion is observed when droplet is in Leidenfrost regime. The vapor flow between droplet and surface exerts a viscous drag force which drives the liquid motion [2]. The same behavior is also observed for Leidenfrost solid that proves the deformation of the liquid is not necessary for its motion [3].

A liquid nitrogen droplet on a flat brass surface in room temperature experiences the Leidenfrost effect. When a moving Leidenfrost droplet hits a piece of tape with an initial velocity, the interaction with the edge of the tape is interesting and not well understood. As a result of this interaction,

the droplet will accelerate on the step, a phenomenon reported by H.Linke et al. in 2006 [2](See figure 1.1). The result of this interaction is a creeping motion over the step and sometimes also detachment from the step.

A set of six sequence pictures in figure 1.1 displays the behavior of a Leidenfrost droplet on a brass surface when it impacts a piece of the plastic tape. The step thickness is roughly  $0.2mm$ . Figure (a) shows a Leidenfrost droplet which moves towards the step, there is no contact with the surface due to Leidenfrost effect. Figure (b) shows the case when the droplet touches the top of the edge. It takes  $18ms$  between frames *a* and *b*, the droplet is moving with roughly  $44mm/s$  velocity. In figure (c); boiling is observed due to the contact between the droplet and the edge of the step. Figure (d) shows the droplet's behavior when it's on top of the step before the detachment. The interaction with the edge takes roughly  $10ms$ . Figure (e) and (f) exhibit the droplet jumping and come back to the surface and it takes roughly  $10ms$  between the detachment from the surface and when it is back on the surface again.

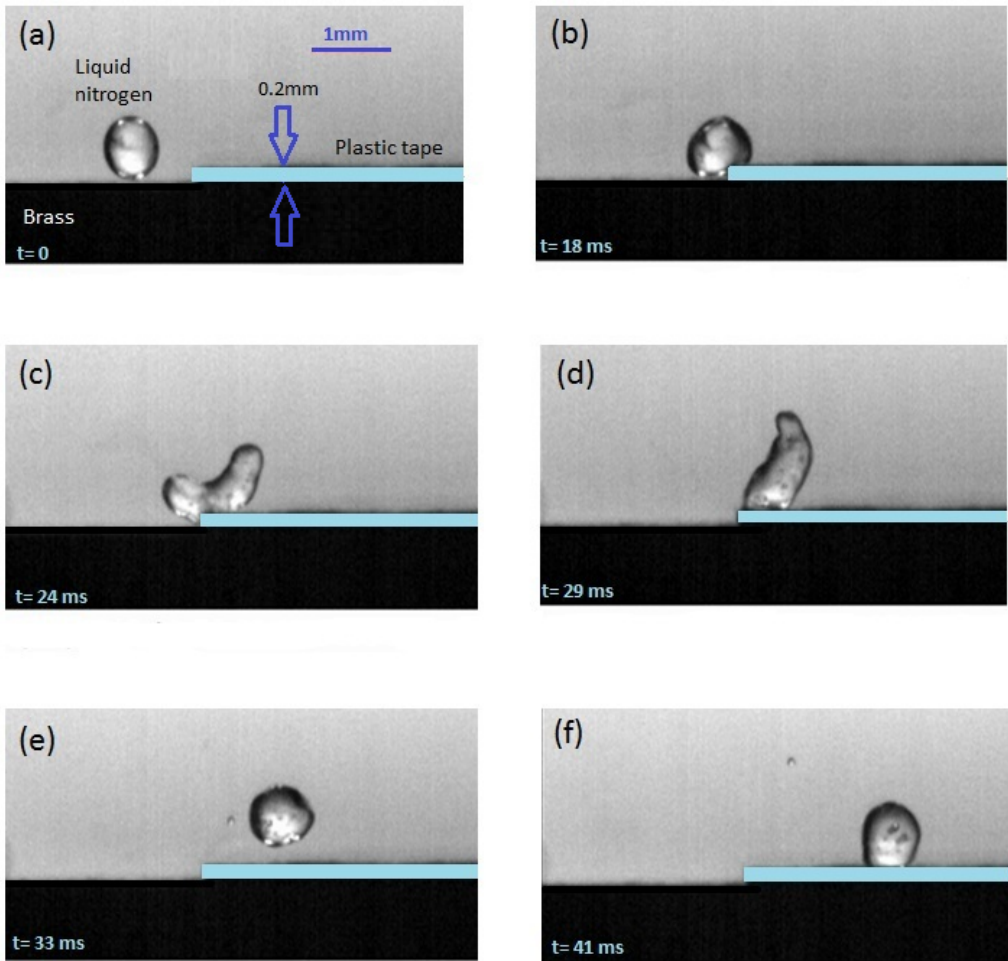
In order to explain our observation, we hypothesize that the Marangoni flow generates this motion, which affects the droplet's shape. Furthermore, we believe that when a droplet hits the edge of the step the vapor layer around the droplet is broken. It cools the edge down quickly, below the Leidenfrost temperature. As a consequence of cooling the edge of the step, droplet experiences a nucleate boiling regime. We propose that this causes the temperature gradient on the droplet and as a result a Marangoni flow on the droplet surface. This may be the reason for the droplet's deformation; along with it's out of plan jumping.

The objectives of this thesis are to understand the specific physical processes of the observed liquid nitrogen behavior and characterize the effective parameters of the jumping process. Further, this work will examine the effects of initial velocity, droplet size, liquid properties as well as the step material properties.

This study has been organized in the following way. Chapter 2 begins by explaining the basic physics of the phenomenon studied here; including heat transfer mechanism, surface tension, Leidenfrost effect, the basic concept of the impact between a liquid and a solid, together with the Marangoni



effect. Chapter 3 provides an overview of different jumping mechanisms of droplets reported in the literature and the physics of the bouncing droplets. In this chapter I reviewed some articles that discuss different mechanisms of jumping for different ranges in droplet size. Further, some articles about the outcome of the droplet-solid hitting process in bouncing droplets are discussed. Chapter 4 presents the experimental set-up both with liquid nitrogen and other liquids. In the experimental methods, details about data gathering for the different experiments are explained and finally, the method used for analyzing data is illustrated. In chapter 5, the experimental results are discussed for different material properties. Our goal is to examine how the velocities of the droplets change for various step materials. When the different energy variations of a typical droplet are given, we will compare the results from experiments with three different step heights of the same material. Eventually, the collected data for several liquids can be compared to find out how the properties of liquid affect the jumping process. Our hypothesis and proposed model is presented in chapter 6, we also present an estimation on Marangoni flow rate. Finally in chapter 7, conclusions and summary of the study are given. In addition, recommendations for future work on different liquids, specifically water are discussed.



**Figure 1.1:** Six sequence images showing the liquid nitrogen jumping process: (a) shows a liquid nitrogen droplet moves on a brass substrate in room temperature (Leidenfrost regime). (b) the droplet impacts the edge of the step. (c) the droplet deforms and starts boiling due to the impact edge of the step. (d) the droplet started to detaching from the step. (e) shows laterally jump of the droplet and (f) shows droplet reaches to the surface again.

# Chapter 2

## Basic Physics

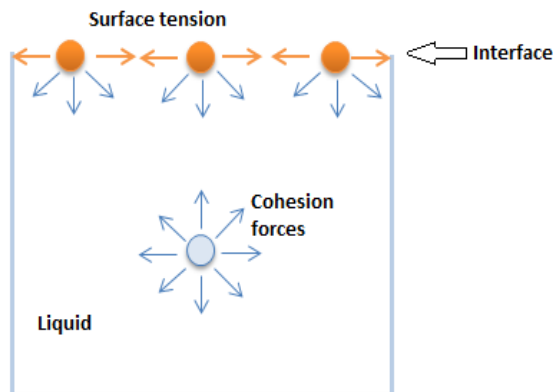
This chapter presents physics relevant to jumping Leidenfrost droplets phenomenon. It begins with explaining surface tension, three phase contact, the impact process, and finally a short discussion about the heat transfer mechanism and Marangoni effect is given, which is likely to play a role in the jumping Leidenfrost droplets phenomenon.

### 2.1 Surface Tension

Surface tension is a liquid property which causes the surface of a liquid to act like a stretched elastic sheet. Specifically, this property of water has significant effects on life. For instance, surface tension influences the upward movement of liquids through tiny tubes (capillary effect), which allows water to move from roots to tops of trees. It also allows insects to walk on water and some of them to breathe under water [4]. Some methods can be used for measuring the surface tension of a liquid depending on the nature and the condition of the liquid. Some of these methods are listed here. The surface tension measurement can be arranged into five groups: The first group represents a direct interfacial measurement using a microbalance, for instance, Wilhelmy plate and ring methods are in this group. Interfacial measurement using of capillary pressure is employed in second

group, as an example, maximum bubble pressure can be mentioned for this group technique. The third group are techniques in which interfacial tensions are determined from a balance between capillary and gravity forces; capillary rise method is an example of this group. Group four represents interfacial measurement by analysis of gravity distorted drops (pendant drop method). The fifth group commonly used for ultra-low surface tension by distorting the shapes of the droplet by centrifugal forces sniping drop technique [5].

There are strong forces between same molecules in a bulk, called cohesive forces, which keep the molecules close together. Molecules in bulk fluid experience these forces in all directions from the neighboring molecules so that the net forces on each molecule is zero.



**Figure 2.1:** A schematic of the unbalanced cohesive forces between the interior and exterior of the liquid molecules.

However, as figure 2.1 shows, exterior molecules only experience some of the attractive interactions because there are no liquid molecules above this layer. Then molecules at the surface are pulled by interior ones, It is clear that at the surface of a liquid, molecules are undergoing a net

force. Therefore, surface tension can be defined as a force that is parallel to the liquid's free surface and acts per unit length. It is denoted by  $\gamma$  and common units of measurement are mN/m or dyn/cm , [6]. Some parameters such as temperature and additive surfactant (organic compound that contains hydrophilic and hydrophobic groups, such as soap or other detergents) influence the surface tension of liquids. Generally, surface tension decreases with increasing temperature until a critical temperature ( $T_c$ ), is reached then it goes to zero. (At this temperature, the density of the liquid is identical to its vapor). Katayama and Guggenheim obtained an empirical relation between temperature and surface tension which is defined as follows:

$$\gamma = \gamma_0 \left(1 - \frac{T}{T_c}\right)^n \quad (2.1)$$

Where,  $\gamma$  is the surface tension,  $\gamma_0$  is a constant, specific for each liquid and  $n$  is an empirical value for which  $n = 1$  is a good approximation for some liquids. However, for non-hydrogen and non-polar liquids  $n$  shows a non-linear relation for the temperature and surface tension of liquids.  $n = \frac{11}{9}$  is derived for organic liquids like alcohol and acetone from the theory of close-pack non-polar and non-hydrogen bounding liquids [7].

Surface tension can also be defined as the work required to increase the surface area by a small amount,  $dA$ , for a given volume. Therefore another unit of measurement is J/m<sup>2</sup> [6].

When a liquid comes in contact with another material (liquid or solid) the surface tension depends on both materials. For homogeneous matter, when the temperature is constant, the surface energy is uniform at the interface. However, any thermal gradient on the interface or any gradient on the chemical composition of the surface will cause a surface tension gradient on the interface [8].

### 2.1.1 Pressure inside droplets

Surface tension tends to contract a liquid droplet in order to make the smallest surface area possible for a given volume. This causes an extra pressure inside the droplet. There is a characteristic length of the droplets

which defines the range of their size, where the surface tension dominates over gravity, this is called the capillary length. This characteristic length is obtained by comparing hydro-static pressure ( $2\rho gR$ ) with Laplace pressure ( $2\gamma/R$ ).

$$L_c = \sqrt{\frac{\gamma}{\rho g}} \quad (2.2)$$

A droplet which is smaller than the capillary length forms a spherical shape and in this range, the gravitational force can be ignored. Moreover, a small drop does not collapse due to the force of the surface tension, this is established by the pressure inside the droplet which is greater than the pressure outside the droplet. Then,  $(P_{inside} - P_{outside})$  defines the excess pressure of a droplet. Lets consider increasing the radius of a droplet from  $R$  to  $R + \Delta R$ ; the force acting on the droplet from the outside is obtained by:

$$F_{outside} = (P_{inside} - P_{outside})4\pi R^2 \quad (2.3)$$

Thus the work done by the excess pressure is:

$$dW = F_{outside}dR = (P_{inside} - P_{outside})4\pi R^2 dR \quad (2.4)$$

This should be balanced with the surface energy:

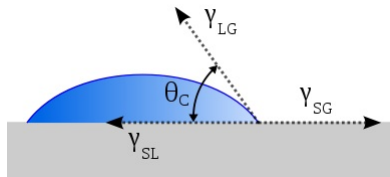
$$\gamma dA = (P_{inside} - P_{outside})4\pi R^2 dR \quad (2.5)$$

$$\gamma 8\pi R dR = \Delta P 4\pi R^2 dR \implies \Delta P = \frac{2\gamma}{R} \quad (2.6)$$

Equation 2.6 shows that surface tension causes higher pressure for smaller droplets. Any changes of the surface tension on a droplet with a certain size affects the pressure inside the droplet. It is known that surface tension decreases with temperature which means that a droplet's internal thermal gradients can vary with its surface tension and consequently result in a change of the droplet's internal pressure [6].

## 2.2 Three phase contact

When two different phases of materials are in contact with a layer in between them that separates them, the interface and surface tension in this area is characterized by interfacial tension. The mutual interaction between the two materials has a significant influence of the statics or dynamics behavior of the two liquids or the liquid on the solid.



**Figure 2.2:** A schematic of liquid-solid-gas interface which shows three interfacial tensions, figure taken from [9].

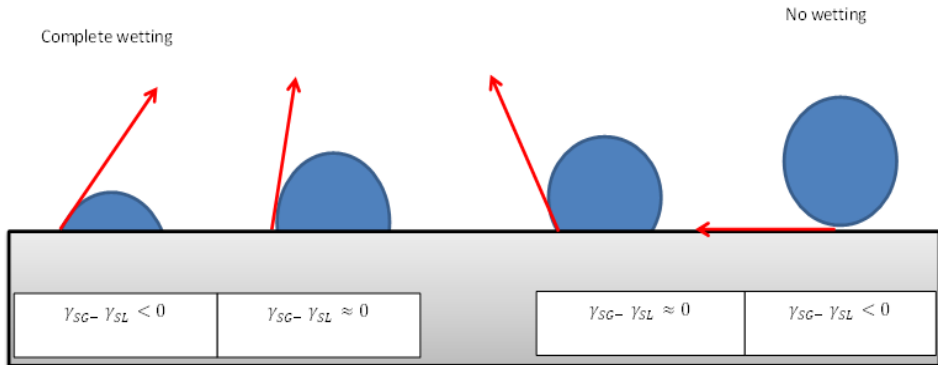
Considering a liquid droplet on a solid surface, as shown in figure 2.2, three different interfacial tensions become important for specifying the shape of the droplet. Young's equation defines the relation between these interfacial tensions as well as the contact angle ( $\theta_c$ ), Where  $\gamma_{SL}$  is solid-liquid,  $\gamma_{LG}$  liquid-air and  $\gamma_{SG}$  solid-air surface tension. Three interfacial tensions also determine the spread coefficient, which is denoted by  $S$ .

$$\gamma_{SG} = \gamma_{LG} \cos \theta + \gamma_{SL} \quad (2.7)$$

The wettability of the surface is determined by the spreading coefficient, see figure 2.3.

$$S = \gamma_{SG} - (\gamma_{SL} + \gamma_{LG}) \quad (2.8)$$

If  $S > 0$ , the liquids will spread and wet the surface. On the other hand if  $S < 0$  the liquid will make a spherical shape on the solid surface without wetting it, see figure 2.3. In figure 2.3, the main assumption is that the size of the droplet is much smaller than capillary length (see equation 2.2)  $R \ll L_c$  [10].



**Figure 2.3:** Three phase interface. Different contact angles of a liquid on the surface defines the wettability of the surface. A droplet smaller than its capillary length makes a spherical shape on non wetting surface,  $\theta_c = 180$ .

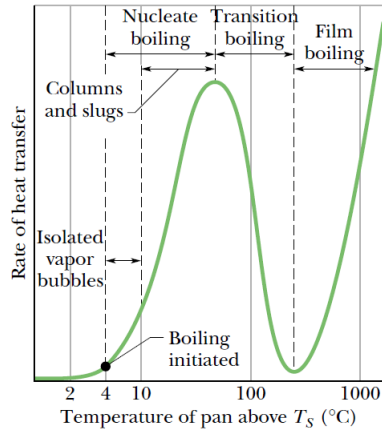
## 2.3 Leidenfrost effect

The Leidenfrost effect is easily observable without advanced equipment. This phenomenon occurs when a small liquid droplet is placed on a surface with a temperature much higher than the liquid's boiling point, and was reported for the first time by Hermann Berhave in 1732, 24 years later Johann Gottlob Leidenfrost published an explanation of the phenomenon [11].

When a liquid droplet comes in contact with a hot surface, if the temperature is lower than its boiling point, it will spread on the surface and there will be heat transfer to the droplet due to conduction. Consequently, the droplet will evaporate in just a few seconds. However, when the temperature is increased to above the boiling point, the rate of heat transfer decreases with temperature until it reaches a minimum value, at a certain temperature, which is called the Leidenfrost temperature. In this regime, due to the considerably high temperature, evaporation at the bottom of the droplet makes a vapor layer, which insulates the droplet from heat transfer. Walker demonstrated that the boiling process in water has three



stages, depending on the increase in temperature.



**Figure 2.4:** The figure shows the water boiling curve. As temperature increases the rate of heat transfer increases until it reaches the boiling temperature. When temperature increases to more than the boiling point, the heat transfer is decreasing function of temperature. This decreasing function continues until a certain temperature which the heat transfer reaches to a minimum value; this temperature is known as Leidenfrost temperature. Finally, when temperature increases to more than Leidenfrost temperature, heat transfer increases with temperature again. Figure taken from [11]

Figure 2.4 shows the rate of heat transfer for water as a function of temperature. A maximum value of the rate of heat is observed in the boiling point of water and minimum value of the rate of heat transfer defines Leidenfrost point. For temperature above the Leidenfrost point the rate of heat transfer shows a decreasing function of temperature due to the effect of heat radiation [11].

### 2.3.1 Leidenfrost droplets

The shape of a droplet on a Leidenfrost surface is described by its capillary length ( see equation 2.2). A droplet with a radius which is smaller than the capillary length would have an almost spherical shape but larger droplets make an ellipsoid shape, due to gravity. The vapor layer underneath the droplets reduces the forces of friction, which makes the droplets remarkably mobile. However, particularly, crenelated shape of the surface increases the friction and makes a remarkable deceleration on the motion [12].

For a droplet with radius  $R$ ,  $\lambda$  defines a region where the droplet and the surface are parallel. For a small droplet,  $\lambda$  is characterized by the following relation::

$$\lambda \sim \frac{R^2}{L_c} \quad (2.9)$$

However for a large droplet  $\lambda$  is almost identical to  $R$  and the thickness of the droplet is roughly  $2L_c$ .

A critical value for the radius of a droplet ( $R_c$ ) defines that bubbles appear in the center of the droplet, which burst when they reach the surface of the droplet if the droplet's radius is larger than  $R_c$ . The vapor layer at the bottom of the droplet rises due to Archimadas thrust and causes a deformation beneath the droplet. Therefore, the author expected that the maximum size  $R_c$  of a droplet is related to the capillary length,  $R_c = 3.84L_c$ , [13].

In order to demonstrate a Leidenfrost droplet, we consider a spherical droplet on a really hot solid surface. Several processes can be considered that take place on the droplet simultaneously. In the Leidenfrost regime, heat transfers to droplet by conduction between the hot surface and lower part of the droplet through the vapor film and also by radiation from hot surface to the entire surface of the droplet. On the other hand, the droplet loses it's mass due to evaporation of the lower part close to the surface and by effusion from the top of the droplet.

### 2.3.2 The effective parameters on Leidenfrost temperature

There are various parameters which effectively can change a Leidenfrost temperature, some of them are introduced here, such as a liquid's purity, surface roughness, surface impurity and pressure.

Purity; The purity of a liquid is one of the more important parameters when discussing Leidenfrost temperatures. Huang et al. demonstrated that dissolved salt in water increased the Leidenfrost temperature. When a solvent droplet is placed on a brass surface, at a temperature close to the Leidenfrost point, it starts evaporating at the bottom. This increases the concentration of the salt at the interface, which changes the liquid properties. Consequently, the vapor layer becomes thinner. As a result, higher temperature is needed in order to reach the Leidenfrost point [14]. Surface roughness also has a key role in the Leidenfrost temperature. On rougher surface, higher temperature is needed in order to keep the droplet floating on its own vapor. Surface impurity can also be effective because it can change the roughness of a surface [15]. However, if a droplet impacts the surface, the effect is quite different. A rougher surface yields a lower Leidenfrost temperature because the microscopic structure of the rough surface breaks the vapor layer beneath the droplet. It considerably decreases the pressure underneath the droplet and consequently, the Leidenfrost temperature is reduced [16]. It seems this is similar to what happens to a Leidenfrost droplet when it touches the edge of the step. Here, the velocity of the impact, which we call initial velocity, may have an important role in breaking down the vapor layer around the droplet. It is postulated that the droplets need some minimum velocity in order to affect the process, but not more than that, since I didn't find any correlation between initial velocity and the height of the jump. The Leidenfrost point of water is also dependent on pressure, however it does not depend considerably on thermal diffusivity [17]. According to these last two parameters, we can deduce that the way a droplet is placed on the surface could be one effective factor.

## 2.4 Impact droplet and heat transfer

When a droplet impacts the surface, even at the Leidenfrost point, it will be in direct contact with surface. When a droplet impacts an isotherm surface, the dynamics of the droplet, the Reynolds ( $Re$ ) and Weber ( $We$ ) numbers should be considered in order to determine the dynamics. The Weber number defines the ratio between inertial force and the surface tension force. The Reynolds number is the ratio of the inertia and viscous forces. A large ( $We$ ) number means that the droplet spreads to a maximum radius until have a large surface area. On the other hand, a large ( $Re$ ) number defines viscous effects which are limited to the thin layer close to the solid surface [18].

$$Re = \frac{\rho v_0 d_0}{\mu}, We = \frac{\rho v_0^2 d_0}{\gamma} \quad (2.10)$$

where  $\mu$  is the dynamic viscosity of the liquid,  $\rho$  is density of the liquid,  $d_0$  is a characteristic length, typically droplet diameter and  $v_0$  is velocity. In Leidenfrost regime we need to consider a droplet's initial temperature and also the liquid solid contact [19]. According to a droplet's temperature, we could divide the impact process into two types, droplets with initial temperatures higher than their boiling points and droplets with temperatures less than their boiling point. In the first step of determining the impact process, the Weber number is a useful dimensionless number. For  $We < 30$ , surface tension dominates over the inertia force, and is strong enough to stop the spreading process. Droplets in this case recoil and rebound without disintegration. For  $30 < We < 80$  spreading and recoiling are the same as when  $We < 30$  but in the rebounding process, droplets may disintegrate, which makes the shape of the droplet unstable [20]. For  $We > 80$ , impact inertia dominates and a droplet will break into several smaller droplets.

In the second case, it has been demonstrated that in the Leidenfrost regime for low Weber numbers the dynamics of a droplet is not dependent on the material. However, for high Weber numbers, the surface properties influence the droplets disintegration [16]. The impact angle also can influence the process. The results of a study by Shi Chao Yao established

that the impact angle can change the result of the process for high Weber numbers for normal impacts when the Weber number is high, the droplet breaks up. However, if a droplet hits a very hot surface at a small angle, a tangential relative velocity destabilizes the droplet and reduces the contact area of the impact. Further, there is a decreased effect on the Leidenfrost temperature when the impact angle is small [21].

### 2.4.1 Heat transfer

When a liquid droplet is in contact with a hot solid surface, the properties of both the liquid and the solid play a main role in controlling the process of the heat transfer to the liquid. The surface temperature is usually related to as the most important parameter in defining different regimes of heat transfer. Four different distinct regimes are defined for heat transfer regarding the temperature of the surface, see figure 2.4. The pattern of the solid-liquid interaction is different for each regime, and subsequently the heat transfer will be different. In the film boiling regime, heat flux reaches a minimum value due to significantly high surface temperature (Leidenfrost regime) In this regime, the heat transfer is controlled by the vapor layer that is forming at the lower surface of the droplet. As the temperature decreases, the heat flux increases until it reaches a critical value where wetting occurs. In the lower boundary in the nucleate boiling regime, heat transfer, as well as bubble formation, decrease with decreasing temperature. Finally, in the film evaporation regime, heat conduction occurs due to liquid film formation. This regime can exist at several degrees above the liquid saturation temperature and it depends on a liquid's properties and the surface roughness [22].

When a liquid droplet touches a hot surface, heat transfer from the surface to the liquid is one of the most important characteristic parameters. In this case heat may cause a thermal shock on the hot solid surface [23]. This will increase the temperature of the liquid and causes evaporation from underneath the droplet. If the heat transfer is large enough, the liquid evaporation at the base of the droplet forms a vapor cushion which can repel the droplet [22, 24] Otherwise the droplet is in direct contact with

the surface [25].

## 2.5 Marangoni effect

The Marangoni effect can be defined as a phenomenon that causes a fluid flow due to a local gradients in the surface tension of a liquid. When the surface tension varies across a droplet's surface, the resultant force will not be balanced, which causes a net force on the droplet. Since surface tension varies with temperature, any temperature gradients across a droplet's surface causes a surface tension gradient, and consequently Marangoni effect is observed. A surface tension gradient is generated either by thermal or concentration gradients which are called thermocapillary and solutal Marangoni, respectively [26].

When a spherical droplet is placed on a heated super hydrophobic surface, the fluid is set in motion due to Marangoni effect. In the conditions of small Marangoni number( $Ma$ ) and  $Re$  are small the convective motion and energy transport are negligible. The effect of gravity as well is small enough to neglect then the effect of buoyancy and buoyancy flow is negligible against Marangoni flow [27]. For a droplet on a hot surface three different mechanisms are possible that can drive convection in the droplet: buoyant convection, Marangoni convection or mass flow from a pinned point together with nonuniform evaporation. The thermal gradient across the spherical droplet causes both a surface tension gradient at interface (Marangoni convection) and a gradient in density which formed a heavy fluid on top of a light fluid (buoyant convection). The Rayleigh ( $Ra = \frac{\alpha_t g R^3 \Delta T}{\nu \kappa}$ ) and Marangoni number ( $Ma = \frac{\alpha R \Delta T}{\kappa \mu}$ ) are useful to determine which effect that dominates. Where  $\kappa$  is thermal diffusivity,  $\alpha$  is change in surface tension due to temperature,  $\nu$  is kinematic viscosity and  $\alpha_t$  is coefficient of thermal expansion. A critical Ra number determines the stability of the buoyancy convection which is on the order of  $10^3$  and it's different depending on geometry. According to the boundary conditions, Marangoni velocity is a result of balance between shear stress, and surface tension gradients in the  $X$  direction. Based on this, we can find a scale for

Marangoni velocity in the following way:

$$\frac{\mu \Delta U}{\Delta X} = \frac{\Delta \gamma}{\Delta X} = \frac{\Delta \gamma \Delta T}{\Delta T \Delta X} \quad (2.11)$$

where,  $\Delta U$  is the liquid's velocity and  $T$  is temperature inside the droplet. Then we obtain:

$$\Delta U_{Ma} = \frac{\alpha \Delta T}{\mu} \quad (2.12)$$

where  $\alpha$  defines the variation in surface tension according to temperature.

$$\alpha = \frac{\Delta \gamma}{\Delta T} \quad (2.13)$$

A balance between viscous dissipation and the rate that potential energy is gained, gives a characteristic velocity of buoyancy effect:

$$U_b = \frac{\alpha_t \Delta T \rho g R^2}{\mu} \quad (2.14)$$

According to definition of Ma and Ra number, Marangoni flow is expected for small droplets and for larger droplets the effect of the buoyancy is dominated against Maragoni effect [28]. R.Monti et al. studied Marangoni effect on pendant droplet. They showed a small droplet on a solid surface at the same temperature wet the surface. However, when there is a heated disk above the droplet or the substrate is cooled, it does not wet the surface anymore. Even if we press it against the surface, it will deform similar to an elastic material [29].

# Chapter 3

## Related phenomena

Small liquid droplets under specific conditions are able to jump from a non wetting surface. This will happen in different ranges of droplet size. The first section of this chapter discusses different mechanisms of jumping droplets reported in the literature. The second section focuses on the impact process and important effective parameters.

### 3.1 The jumping mechanism

#### 3.1.1 Spontaneous jump of a droplet

When a droplet comes in contact with a solid surface, the surface tension causes a shear stress in the solid at the contact line in perimeter of the droplet. When condensate droplets form on a solid which is close to its melting point, the solid substrate melts locally under the droplet due to latent heat. This causes a change in wetting property of the substrate. The released substrate strain due to melting will enable a lateral lift of the droplet [30]. This type of the jumping happen for droplets with a  $\mu m$  radius and take place in any direction and the distance between each jump is in order of the radius of the droplet.

At the three line interface the three forces due to surface tension are in



balance at the contact line. when the substrate is solid the resultant vertical force is not zero, and it is in balance with a reaction force of substrate. This force causes an elastic deformation of the substrate subsequently a storage of energy. The melting process suppresses the reaction force and this makes unstable droplets. The supplied elastic energy can move the unstable droplet [30]. However in this process, the amount of the surface energy of the droplet is remarkably larger than both the viscous energy and the elastic energy. Author proposed that the melting is what powers this jump by changing the surface energy and elastic energy is not enough to drive this motion. In this case, the sign and magnitude of  $\Delta\gamma$  is important.

$$\Delta\gamma = \gamma_{SC} - \gamma_{LSC} - \gamma_{SG} + \gamma_{LSG} \quad (3.1)$$

Here  $\gamma_S$  denotes surface tension of solid substrate after,  $\gamma_{LS}$  is surface tension of solid-liquid,  $\gamma_{SG}$  is surface tension of the solid-gas and  $\gamma_{LSG}$  is the interfacial tension of the solid liquid gas. In this formula index  $C$  means condensation.

If  $\Delta\gamma > 0$ , the condition is in favor of jumping. The droplet lies on the substrate and after locally melting of the substrate has taken place beneath the droplet, it will get a flow that covers the entire contact line.

If  $\Delta\gamma < 0$ , the condition is not sufficient for jumping and the droplet does not lie on the surface. When the substrate starts to melt, it will create a flow to an uncovered substrate and the droplet will roll on to the melting part of the substrate. This means lying on the surface is necessary for jumping process [31].

### 3.1.2 Self-propelled dropwise condensate on super hydrophobic surface

A self-propelled jumping motion on a super-hydrophobic surface happens due to condensation. The small condensate droplets move in any direction on a super-hydrophobic surface. They merge together and make a bigger droplet and when they reach to a critical size self-propel jumping is observed. Below the critical size viscose dissipation dominates during

the coalescence, and the energy release is not enough for the jumping process. The jumping process happens for a certain range of the droplet sizes. Because condensate droplets, jump in any direction finally they leave the surface spontaneously and the process occurs continually without exert any external forces. Such a jumping is due to the released surface energy resultant of coalescence of small droplets [32].

If we assume that all of the released surface energy changes to the kinetic energy ( $KE = E_s$ ) then the velocity of the merged droplets can be calculated by:

$$\frac{1}{2}mv^2 = \gamma area \quad (3.2)$$

$$R = (R_1^3 + R_2^3)^{\frac{1}{3}} \quad (3.3)$$

The area in equation 3.2 can be defined as difference between the area of the coalescence droplet and area of the two droplets.

$$v = \sqrt{\frac{6\gamma(R_1^2 + R_2^2 - (R_1^3 + R_2^3)^{2/3})}{\rho(R_1^3 + R_2^3)}} \quad (3.4)$$

if two droplets with the same radius merge together. Then  $R = R_1 = R_2$

$$v = \sqrt{\frac{\gamma}{\rho R}} \quad (3.5)$$

This equation shows that droplets with a smaller radius jump much higher than larger droplets, however jump starts from a critical radius. For very small droplets, the effect of the viscosity is comparable with released surface energy. The same phenomenon is possible to happen when two Leidenfrost droplets merge together. In this case, a liquid bridge forms between the surface and the droplet. The impact of the liquid bridge causes an upward force which can separate droplet from the surface [32].

The surface energy of the surface has a significant role on the size of the departure droplets. Dietz et al. showed experimentally that with surface energy reduction of the surface, smaller droplet tend to leave the surface.

This may cause a permanent dropwise condensation and consequently effective heat and mass transfer [33].

Self-propelling start in a certain range of radius of the droplet and the velocity of coalescence droplet increase with increasing the size of the droplet until a maximum radius. After that velocity shows a decrease behavior with increasing the size. The jumping velocity is increased with increasing size to a maximum value and after that it is a decrease function of the size. This type of the jumping is observed for a radius around several micrometer to a few millimeter, where the gravitational force and dissipation energy is negligible compared to surface energy [33].

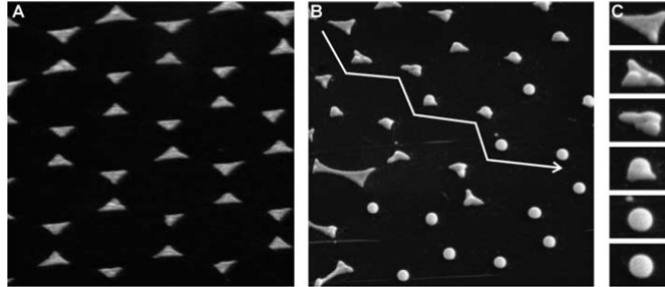
The jumping phenomenon in nano-scale has an application for deposition of nano-particles onto a second substrate. It is demonstrated that different metals can be deposited in a size ranging from a minimum of tens up to a maximum of several hundreds of nanometers [34].

Two different methods can be used to observe a nano droplet jumping.

In first method, colloidal lithography is used to deposit the triangular gold solid nano-structures on a non wetted surface. By heating the gold triangles with adequate laser energy near the gold melting point, they will change phase to liquid. In liquid phase, a small amount of liquid tends to has a spherical shape and due to have a minimum surface energy. Solid nano structure starts melting from cones of the triangles toward the center. This causes a movement in center of mass of the nano structure to up ward and a change in surface energy. In this method, surface deformation energy changes to the kinetic energy. In fact, when droplet forms a huge surface energy released due to change on shape of the nano structure.

Figure 3.1 shows different step of melting process. When the shape of the droplet changes from a flat triangle to a sphere, center of the mass(CM) moves upward and this can detach droplet from the substrate. The measurement value of the detachment velocity shows that there is a dissipation energy and also it will decrease with the decreasing released surface energy. In the dynamic of this system inertia dominates to the viscous dissipation. Thus detachment is possible when the kinetic energy(KE) of the droplet's center of mass dominates the adhesion energy [35].

In the second methods, the laser used to melt the nano prism is a



**Figure 3.1:** Scanning electron microscope picture of triangular gold nanostructures, shown before using laser beam(A)and after that(B). Figure C shows how the triangles melt, figure is from [35].

femtosecond laser pulses in a resonance with a strong surface plasmon oscillation. When the laser intensity is high enough, the rates of absorption is more than the rate of melting gold nano structures. This causes nano prism lift off from the surface while the shape of the nano prisms do not change. SEM and AFM show that nanoprisms bisectors and thickness change due to this process and they occupy a smaller area after jumping. The authors believe that the gold nanoprisms sublimation can explains this phenomenon. They observed when the laser intensity is  $5.1 \text{ mJ/cm}^2$  the nano prisms fly away. The fast gold sublimation causes an ultrahigh gold atomic pressure beneath the prisms, which is able lift the prisms off the substrate [14].

## 3.2 Bouncing droplet

When a low viscous droplet touches a Leidenfrost surface it will rebound. The rebounding process is divided into two regimes. These two regimes of the bouncing droplet are characterized by  $We$  number. Experimental results confirm that for high Weber numbers, the rebounding process shows low elasticity because of the high velocity. For low Weber numbers droplets bounce a lot and they always return to the same height (high elasticity). The bouncing mechanism is described as follows: When a liquid droplet

touches a solid surface, it loses its energy due to the spreading on the surface and division of the energy for droplet oscillation. Since larger droplets osculate more than smaller droplets, they show low elasticity because these droplets spend more energy on oscillation.

Droplets larger than capillary length can not restore enough energy to take off. Consequently, they are stuck to the surface and just oscillate without bouncing [36].

When two different phases are in contact, such liquid-liquid or liquid-solid, interfaces should be considered when describing the motion of a droplet. The shape of a sitting droplet on a solid can be described with the Bo number which is:

$$Bo = \frac{Mg}{4\pi\gamma R} \quad (3.6)$$

The effect of gravity present when the  $Bo$  is larger than one unit, consequently the droplet will make an ellipsoid shape. A bouncing droplet can be considered like a oscillating system. Viscosity is a damping process and surface tension a restoring force. If Ohnesorge number is much smaller than the viscosity, damping is negligible and the resonance is critical [37]. In impact phenomena with solid surface three types of behavior are observed: splashing, spreading, bouncing

Splashing occurs when the shock energy dominates to cohesion energy, droplet breaks up to several smaller droplets. If a drop touches a hydrophobic surface with low velocity, bouncing is observable and spreading is observed when surface is hydrophilic. When droplet touches the surface due to impact, a shock wave spreads near the surface, and it causes deformation increasing surface area. Subsequently the surface energy increases due to this process [38].

## Chapter 4

# Experimental Methods

This chapter describes the experimental methods used to study the motion and jumping of liquid droplets at the edge of the step (here the step is a thin layer which is placed over a substrate) at Leidenfrost regime. It starts by giving a description of the problem as well as explaining the setup of the experiment and the data gathering method. Lastly, the method used for analyzing data is explained.

The experiments can be classified in two main groups:

One set of experiments has been performed with liquid nitrogen where different materials with low thermal conductivity like plastic and paper tape, PDMS, parafilm and glass have been used as a step over a brass substrate. Moreover, brass and bronze have been used as a step on the brass substrate to compare the influence of high and low thermal conductivity of the materials. In addition, experiments with liquid nitrogen for different step heights of the same materials are examined. The step heights varies between roughly 0.2mm and 0.6mm.

The objectives of this set of experiments is to explain the behavior of liquid nitrogen when different materials are used as a step, as well as to find out what affect the physical properties of these materials have on both the interaction with the step and the jumping process. The effect of the height of the step on the jumping process is probed as well.

I used three different liquids for the second set of experiments: alcohol, acetone and liquid nitrogen on a step height of around 0.2mm. The same height of a step made from glass was examined. The purpose of this set of experiments was to study how the liquid properties influence the process. All the experiments have been performed in the typical laboratory conditions (room temperature and local atmospheric pressure).

#### 4.0.1 Recording system

The recording system consists of a high speed camera of the model Motion-BLITZ EoSens Cube mini, light, a tripod and a laptop. This high speed camera has a CMOS sensor with maximum 298,000 fps which we used maximum 3000 fps of this, maximum resolution of 1696 pixels  $\times$  1710 pixels and a minimum exposure time of 100  $\mu s$ . Different magnifications are possible using a spacer between the lens and camera. To record the droplet behavior on the step, the camera is adjusted by a tripod for getting a side view of the droplet, parallel to the surface so that the optical axis of the camera is perpendicular to the side of the surface.

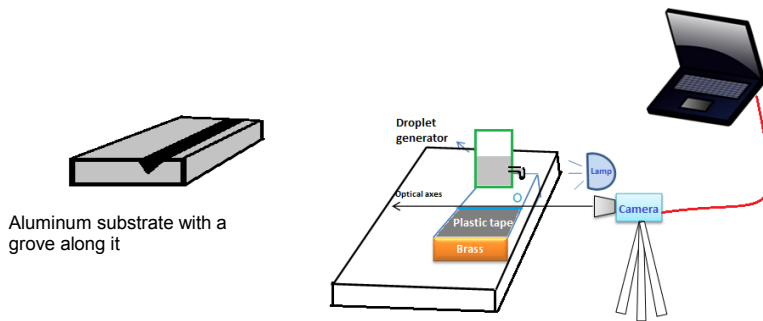
For a high speed camera with a fast frame rate since the exposure time is very short, the emitted light should be strong enough to obtain clear images. For a 3000 *fps* frame rate, 300  $\mu s$  exposure time was used and adequate light was provided by two desk light in order to bright behind the droplets. Finally, the recorded data will directly be imported to a laptop.

Recording the movie at high frame rate limits the time of the recording. This specification makes possible to take movie roughly 5 seconds. On the other hand, when saving the movie, droplets keep coming from the droplet generator and we miss to record droplets of the same size and velocity. Therefore, in order to get recordings with the same droplet size, the experiment needs to be done several times.

## 4.1 Liquid nitrogen

### 4.1.1 Experimental setup

A schematic diagram of the experimental setup for liquid nitrogen is demonstrated in figure 4.2. It comprises of a high speed camera, a tripod, a laptop



**Figure 4.1:** A schematic of the experimental setup which is used for liquid nitrogen droplets.

that is used for recording the motion of the droplets. In addition, two desk lights have used to improve the contrast of the movies. A brass substrate at room temperature is used as a Leidenfrost surface. In order to get small droplets and to give their motion a sufficient direction, an aluminum surface has been used which has a drilled groove (the groove depth is  $0.5\text{mm}$ ) along the surface. The aluminum surface is placed at the same level with brass and droplet moving inside the groove towards the brass surface, during which time it evaporates, decreasing in size. To create a step on the substrate, a thin layer of different materials was placed on the brass substrate.



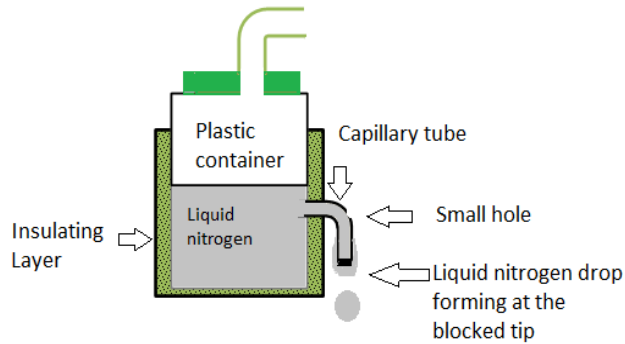


**Figure 4.2:** A picture of the experimental setup which is used for liquid nitrogen droplets.

### 4.1.2 Droplet generator

The extremely low boiling point ( $77K$ ) of liquid nitrogen makes it impractical to use simple devices such as a dropper or a small syringe to produce droplets. As soon as the liquid nitrogen comes in contact with the needle of the syringe, it evaporates very fast and freezes the syringe or dropper. However the most important problem is the explosive boiling when it comes from needle of a syringe. For this reason, in order to create small liquid nitrogen droplets a simple droplet generator [39] used which is shown in figure 4.3.

This simple liquid nitrogen droplet generator consists of a foam layer as an insulator around a plastic container, together with a small bent capillary tube (stainless steel tube with outer diameter of  $1.6mm$ ) connected to the container  $25mm$  from the bottom. There is a small drilled hole ( $0.5mm$  in diameter) in the capillary tube, which is bent at right angles. After pouring liquid nitrogen inside the plastic container it cools the container



**Figure 4.3:** A liquid nitrogen droplet forms in the hole at the end of the capillary tube and falls down when gravity overcomes the surface tension.

and nitrogen gas comes out from the small hole. Once the tubes walls cooled enough liquid come out through the small hole onto the outer surface of the tube moves toward the blocked tip. The accumulated liquid falls down due to gravity when it reaches a certain size. Different droplet sizes and velocities are produced by this droplet generator as the level of liquid nitrogen inside the container changes.

When the blocked tip is close to the brass or aluminum surface the liquid nitrogen that has accumulated between the surface and the tip forms a larger droplet. Therefore, in order to get small droplets the blocked tip should be placed far enough from the surface so that the droplet can fall down. Typically, the smallest produced droplet size is around  $2mm$  in diameter. However, challenging to make droplet is range of required size which is not controllable with this device. The droplets fall down close to the surface, so that the impact We number is low enough that the droplets do not break to smaller droplets.

### 4.1.3 Methods

It is important to keep the temperature constant, because different ranges of temperature will give different ranges of the energy gain. Since both cleanliness and roughness of the surface affect the Leidenfrost temperature [15, 40] consequently it will affect a droplet's behavior in this regime. Before beginning the experiment special care should be taken to protect the surface from dirt, oil and anything else that can change the roughness of the surface. The first step in cleaning is to polish the brass surface with chimerical brass polish, followed by rinsing the surface with de-ionized water.

For experiment with PDMS as a step we start as follow:

In the beginning, a clean brass surface (with 6 *mm* in thickness and 120mm in length, 42 mm length and width respectively ) is placed on an adjustable table, parallel to the edge of the table. A thin layer of PDMS with a thickness roughly with a 0.2 mm is fabricated by soft lithography (see appendix). A piece of this PDMS is placed on the brass surface as a step. Initially droplets are released onto an aluminum plate with a groove along it which has been placed on a level surface with brass substrate. We capture side-view images of the droplets using a high speed camera, at a sampling rate between 1000 and 3000 fps depending on the details which we want to get from the movie. After adjusting the setup, a ruler with millimeter resolution is put on the top of the surface in the same direction a moving droplet and a picture of the ruler is used as a scale. For a number of droplets, the motion when passing the step is filmed and the recorded data is imported to a laptop. The same method is used for the glass, parafilm, tape, brass and bronze. For each droplet, the velocity before and on the step, as well as the height of the jump is extracted. The data will be analyzed using the software ImageJ for each few millisecond. For this data, we aimed the analyzing data to find out how velocity of the droplets changes over the step for each material.

Different step height:

To understand how the jumping phenomenon depends step height, a piece of plastic tape was placed on the brass as a step. I started using one

layer of plastic tape (VWR) with a thickness of 0.2 mm, which was placed on a brass substrate, on the path of the liquid nitrogen droplets. In order to increase the step height two or three layers of the tape are placed on top of each other and each one is measured in the same way.

## 4.2 Different liquids

### 4.2.1 Set-up of the experiment

We used a hot plate with a temperature range of up to 500 degrees. Since the surface temperature is usually different from the temperature of the hot plate, a thermocouple of model Testo 925 was used for measuring the surface temperature. A syringe with a small needle size was used for creating small droplets. The size of the droplets for these liquids is relatively large in comparison to the nitrogen droplets. Making small droplets for these liquids is a challenge. The size range of the alcohol droplets is greater than the ones for liquid nitrogen and acetone. Data for all of the droplets was gathered for a temperature gradient of about 150 °C for both alcohol and acetone (different temperature between the Leidenfrost point which experiments done and boiling temperature).

### 4.2.2 Method

To prove what effect liquid properties have on the process, two different liquids were used for comparison with the results from the experiments with liquid nitrogen. For this we used alcohol and acetone, with boiling points of 79°C and 54 °C respectively. In order to reach the Leidenfrost point of these liquids, we need to heat up the surface to 200 °C for acetone and alcohol 230 °C. The experiments with acetone and ethanol were done in fume hood, the vapor or fume of those liquids. For this set of experiments glass was used as a step, because the parafilm, tape and also PDMS change their shape and some of them melt in high temperatures. It is possible to warm up the glass to high temperature, but since it is very thin any fast changing temperature can change its shape to a curvy shape hence we

should increase its temperature slowly. In that case that glass shape change in high temperature, the step height will vary with the droplets position and we cannot be sure that the droplets go over the step in the same place.

### 4.3 Analysis data

At this stage of the experiment, the kinetic energy (KE) before and after the step was compared for each droplet. The energy gained due to the interaction versus the volume of the droplets plotted.

For the first set of the experiment, the difference in velocity before and after the step, versus radius of the droplets is plotted in a graph (results are presented in next chapter). For this purpose, the velocity of the droplet is obtained by considering the position of the droplet at two different times using following equation:

$$\begin{aligned}x_1 &= x_1(t_1) \\x_2 &= x_2(t_2)\end{aligned}$$

$$v(t) = \frac{x_2 - x_1}{t_2 - t_1} = \frac{\Delta x}{\Delta t} \quad (4.1)$$

Two different types of errors can be defined: systematic and measurement errors.

I estimated the error of measurement, at most two pixels. A ratio of these two pixels to the total number of pixels defines the relative error of measurement. For these experiments the relative error varies between 7 and maximum 30 percent when measuring the size of the droplets.

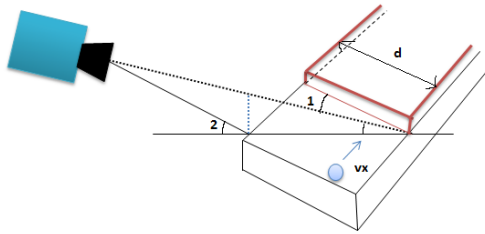
To estimate the error of the calculation for velocity we need to consider errors of both position and time. Using the following formula for velocity and its derivative we can define it.

$$v = \frac{x}{t} \quad (4.2)$$

$$\left(\frac{\delta v}{v}\right)^2 = \left(\frac{\delta x}{x}\right)^2 + (-1)^2 \left(\frac{\delta t}{t}\right)^2 \quad (4.3)$$

Due to the fast frame rate, the time between two frames is roughly 0.3 ms which makes the error smaller than 1 percent; for this reason we can ignore the second part of the equation and we obtain:

$$\frac{|dv|}{|v|} = \frac{|dx|}{|x|} \quad (4.4)$$



**Figure 4.4:** A schematic of systematic error.

The estimated value of the error in velocity is roughly 0.02.

Systematic error:

Both angles of 1 and 2 with the step and substrate (see figure 4.4) make errors when measuring the height of the jump and the velocity respectively. However for small angles the error due to this is negligible. Then by using the following equation we can find a value of the height of the jump.

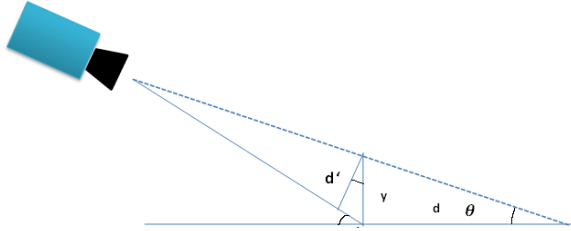
$$d' = d \sin \theta \quad (4.5)$$

where  $d$  is the width of the substrate measured with a ruler, and  $d'$  is the width of the substrate measured from video.

$\theta$  is the angle between the optical axis with surface substrate. By using a calculated value of  $\theta$  we can find the height of the jump.

$$y' = y \cos \theta \quad (4.6)$$

Here  $y'$  is the maximum height of the jump measured from the video and  $y$  is the a corrected value of the height of the jump.



**Figure 4.5:** A schematic of systematic error.

To estimate error of velocity measurement we need to consider angle number 1 (see figure 4.4 ).

$$v_x = v'_x \cos \theta \quad (4.7)$$

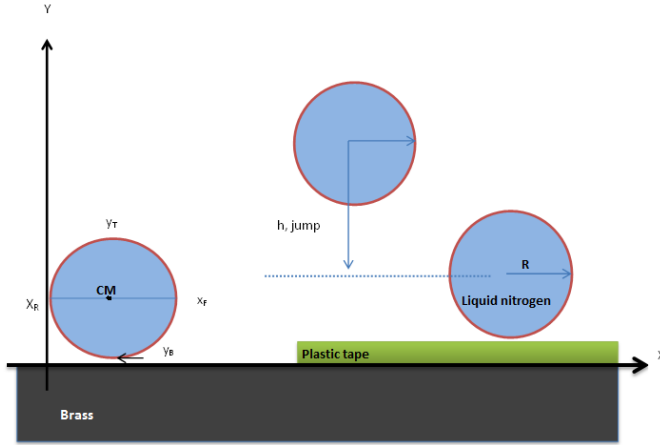
where  $\theta$  can be measured from video, and  $v'_x$  is the measurement value from video. The correction coefficient of velocity for systematic error is very small and negligible.

To obtain more detail about energy change in the droplet due to the interaction with the edge of the step, a 3000 fps movie of a typical droplet was analyzed as follows: The position coordinates for the rear ( $x_R$ ), front ( $x_F$ ), top ( $y_T$ ), and bottom ( $y_B$ ) of the droplet were determined separately and for each frame (see figure 4.6). The time of each frame was considered for each position and all the data were inserted into an Excel sheet for calculation energy of the droplets. Using the scale, we will convert pixels into mm, then, for each data series, position of the center of mass (CM) in  $x$  and  $y$  direction is calculated using the following equations:

$$x_{CM}(t) = \frac{x_F(t) - x_R(t)}{2} + x_R(t) \quad (4.8)$$

and

$$y_{CM}(t) = \frac{y_T(t) - y_B(t)}{2} + y_B(t) \quad (4.9)$$



**Figure 4.6:** A schematic of a droplet on a brass surface. Difference between CM in highest position of the droplet and CM of the droplet on the step when it made a spherical shape again defines the height of the jump.

Finally, by using:

$$v_x(t) = \frac{\Delta x_{CM}}{\Delta t} = \frac{x_{CM}(t_2) - x_{CM}(t_1)}{t_2 - t_1} \quad (4.10)$$

Using the equation 4.10 we can obtain the different kinetic energy of the droplet before and after the step as following:

$$\Delta K = \frac{1}{2}mv_{x_{final}}^2 - \frac{1}{2}mv_{x_{initial}}^2 \quad (4.11)$$

and by adding potential energy of the droplet proportional to the step height, the gained energy obtained:

$$Gainenergy = \frac{1}{2}mv_{x_{final}}^2 - \frac{1}{2}mv_{x_{initial}}^2 + PE \quad (4.12)$$

$$v_y(t) = \frac{\Delta y_{CM}}{\Delta t} = \frac{y_{CM}(t_2) - y_{CM}(t_1)}{t_2 - t_1} \quad (4.13)$$



the velocity components of CM in the  $x$  and  $y$  directions were calculated. Since frame by frame analyzing make scattering on velocity points, in order to reduce fluctuation in velocity points we considered a mean value for the velocity at each position. result for smoothing data points for every three points.

$$v(t) = \frac{x(t_3) - x(t_1)}{t_3 - t_1} \quad (4.14)$$

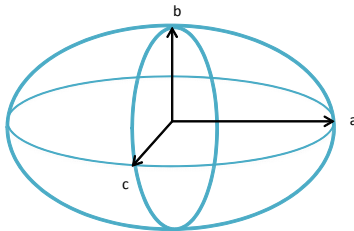
Finally, for each data point, the kinetic and potential energies were calculated (see appendix). We obtained the additional energy due to interaction by:

$$E_{Total} = K_x + PE + K_y \quad (4.15)$$

which  $K_x$ ,  $PE$  and  $K_y$  are obtained from the following equations:

$$K_x = \frac{1}{2}mv_x^2, K_y = \frac{1}{2}mv_y^2, PE = mgh \quad (4.16)$$

Here,  $K_x$  is the kinetic energy in  $x$  direction,  $PE$  is potential energy due to motion of the CM in  $Y$  direction,  $h$  is the maximum position of the CM and  $K_y$  is the kinetic energy in  $y$  direction.



**Figure 4.7:** A schematic of droplet shape during interaction

To estimate the droplet's surface area we assume an ellipsoid shape (see figure 4.7). Where,  $a$ ,  $b$  and  $c$  are the ellipsoids half axes. We can measure

$(a, b)$  by analysing movie. However in order to find the third dimension of the radius ( $c$ ), we considered the volume of the droplet to be constant during the process. The calculated value of the rate of evaporation is very slow, approximately  $0.2 \text{ mm}^3/\text{s}$ , and the jumping process takes only 30 ms at most. Therefore, we can consider a constant volume for a droplet during this process. For estimating the third radius we can use the following equations:

$$V_e = \frac{4}{3}\pi abc \quad (4.17)$$

$$V_s = \frac{4}{3}\pi R^3 \quad (4.18)$$

$V_e = V_s \implies c = \frac{R^3}{ab}$  where  $R$  is the radius of the droplet while the droplet shape is spherical. The assumption is a Leidenfrost droplet which is much smaller than capillary length with no bouncing motion makes a spherical shape.  $V_s$  is the volume of the spherical droplet and  $V_e$  defines volume of the ellipsoid. The following formula gives an estimation of the ellipsoid's area.

$$E = 4\pi \left( \frac{a^p b^p + a^p c^p + b^p c^p}{3} \right)^{\frac{1}{p}} \quad (4.19)$$

Here,  $p = 1.6075$  is a constant which makes a the least relative error around 1.06 percent for ellipsoid area calculation [41].

The surface energy can be calculated by;  $E_s = \gamma E$  and the vertical distance of the CM in each time defines potential energy of the droplet by  $U = mgh$ , where  $h$  is the vertical distance of CM from the  $x$  axis (see figure 4.6). The potential energy of the droplet is considered zero when the droplet is spherical on the brass substrate.

## Chapter 5

# Experimental observations

This chapter presents the experimental observations of three different sets of experiments. It starts by presenting how different materials and heights of the step affect the jumping process. Finally, the results of experiments with different liquids are described. The absolute error for measuring radius of the droplet is considered one pixel which was roughly 0.03 mm for the analyzed movies. For the relative error of the velocity, just the error of the distance was considered because the error of the time measurement is very small in the range of  $10^{-4}$ s. The ratio of the one pixel error to the number of counted pixels for measuring the distance multiply to velocity give the relative error of the velocity roughly 0.02 ( See equation 4.4). I just will show the errors of the measurement for some of the data because there are a lot of data dots on some of the graphs. The symbols which were used for all the graphs is defined as following:

- ◇ illustrates the droplets that detached from the surface (jumping).
- △ illustrates the droplets which do not show jumping our nucleate boiling. They just hit the step and bounce back.
- means the droplet can go over the step due to their initial momentum.
- × illustrates the droplet which boil due to contact with the step (Nucleate boiling).
- shows the droplet which splits into two droplets.

## 5.1 Different materials of the step

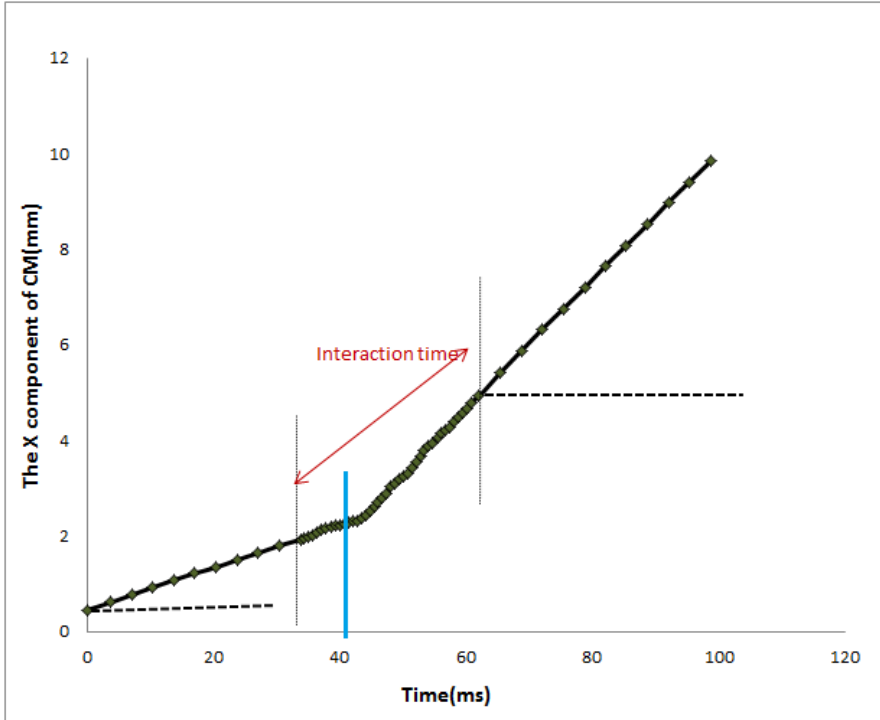
In this section the experimental results of PDMS, plastic tape, brass, glass, and parafilm are presented. The substrate is always brass except the figure 5.7 which the tape has been used as both substrate and step.

### 5.1.1 Plastic tape on brass substrate

Figure 5.1 presents the motion of  $CM_x$  of a typical droplet while the step is one layer plastic tape. The experimental data points specified as dotted curve and solid line present the best line through the data. At time  $t \geq 35$  ms and  $60 \text{ms} \leq t$  the movie has been analyzed for time interval of 3.5 ms. The interaction time is specified as time interval between the first contact of the droplet to the step until it is on top of the step again. In order to obtain more detail of the CM in the interaction time the movie has been analyzed with time interval 0.7 ms. A linear behavior of the  $CM_x$  in time domains  $t \leq 35$  ms and roughly  $60 \text{ms} \leq t$  is observed but with different slopes corresponding to different velocities. A gap between initial and final velocities which shows increasing behavior of the droplet during crossing the step proves that the droplet accelerates during the interaction with the step.

The vertical blue line indicates the time at which the CM of the droplet is placed over the edge. The  $CM_y$  of the droplet is presented in appendix B. Figure 5.2 shows the velocity evolution of the droplet which is derived from the figure 5.1. In order to reduce the noise, the graph has been smoothed in a window of three data point. It is obvious that the droplet experiences higher velocity after interaction with the edge. The blue arrow shows the time that the droplet touched the step. The droplets with various initial velocities of 10 mm/s to 80 mm/s and different radii have been studied for a step of plastic tape.

The result of velocity change versus radius is shown in figure 5.3. For instance the error of few of the data point has been indicated in the graph. As it is obvious, there is no significant correlation between the velocity change and droplet size.

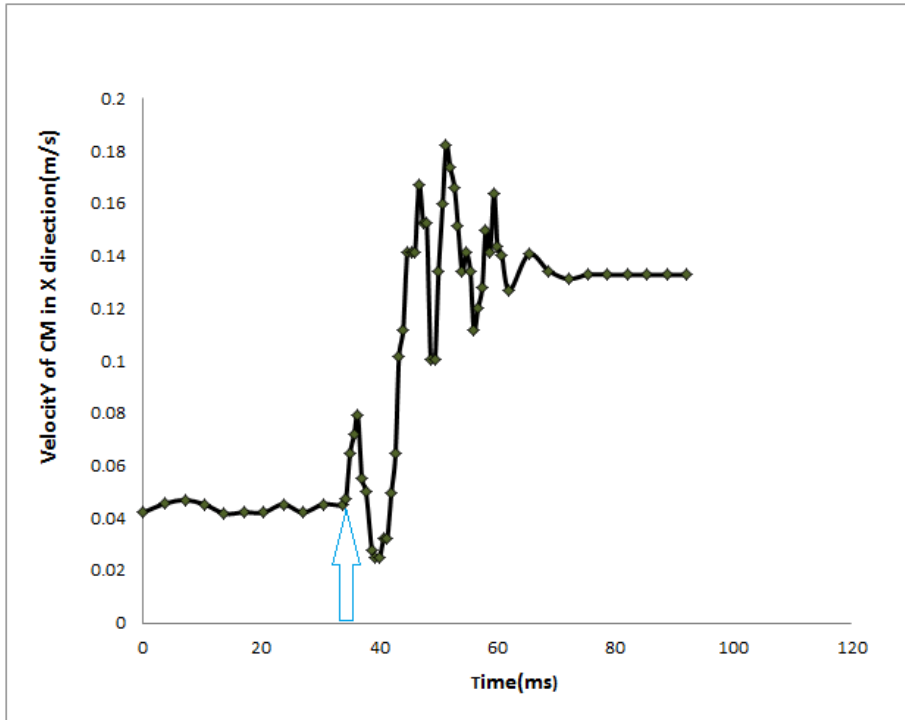


**Figure 5.1:** Motion of the  $CM_x$  of a typical liquid nitrogen droplet on a brass substrate with a layer plastic tape versus time. The green dots indicate data and the black line present best fitted line through the data. The blue line shows the time which the droplet touches the step and the dash lines show the different angles of the fitted line before and after the interaction with the step.

### 5.1.2 Brass step on brass substrate

Figures 5.4, 5.5, 5.6 show behavior of the liquid nitrogen droplets on the brass as step with a thickness of 0.2mm.

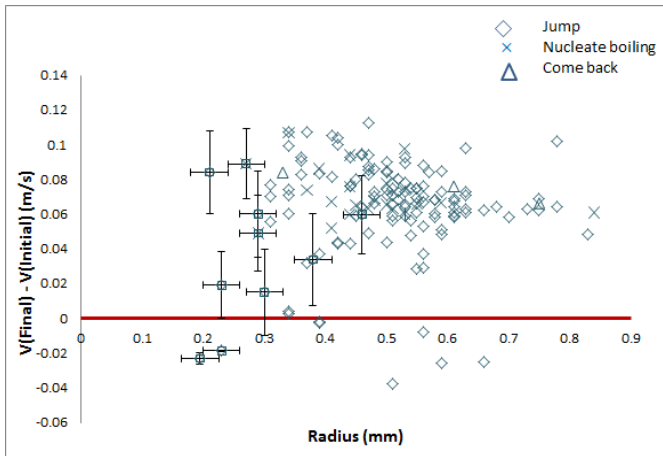
The motion of  $CM_x$  of the droplet exhibits different slopes before and



**Figure 5.2:** velocity of the CM of the liquid nitrogen droplet before and after the interaction with the edge of the step. This graph is derivative of the data points of the figure 5.1

after the edge of the step. The angles 1 and 2 show the slopes of the fitted line through the data points before and after the edge of the step, and  $a$  shows different between angles 1 and 2. The smaller slope proves a slower motion of the droplet on the step. The dash line in the graph shows the slope of the motion when droplet is on the step.

Figure 5.5 shows also clearly that the velocity of the droplet reduces



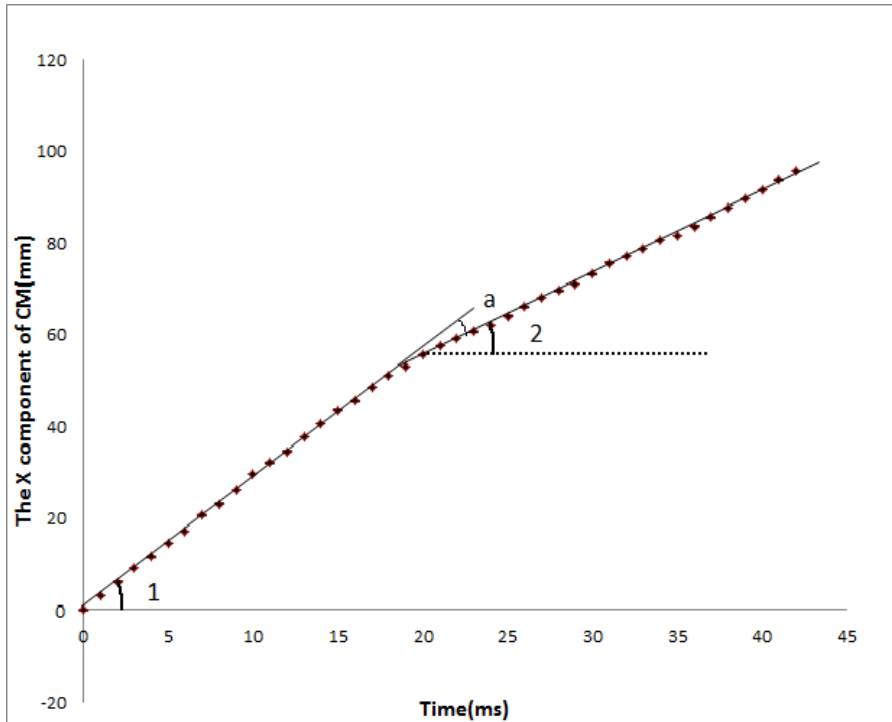
**Figure 5.3:** The velocity change of the different droplet sizes when step is plastic tape.

after interacting with the edge of the step. This proves that when the material of step is metal and the same as the substrate, droplets do not experience higher velocity over the step and they will lose part of their kinetic energy. The blue dash lines on figure 5.5 shows the average velocity before and after the edge of the step.

Figure 5.6 shows the velocity change of the droplet versus the droplet radius. The red circle indicates no nucleate boiling and no jump. In fact, the final kinetic energy of the droplets drops due to the potential energy of the step. The triangles symbol show, the droplets don't have enough energy, they only hit the wall and come back.

### 5.1.3 Paper tape on paper tape substrate

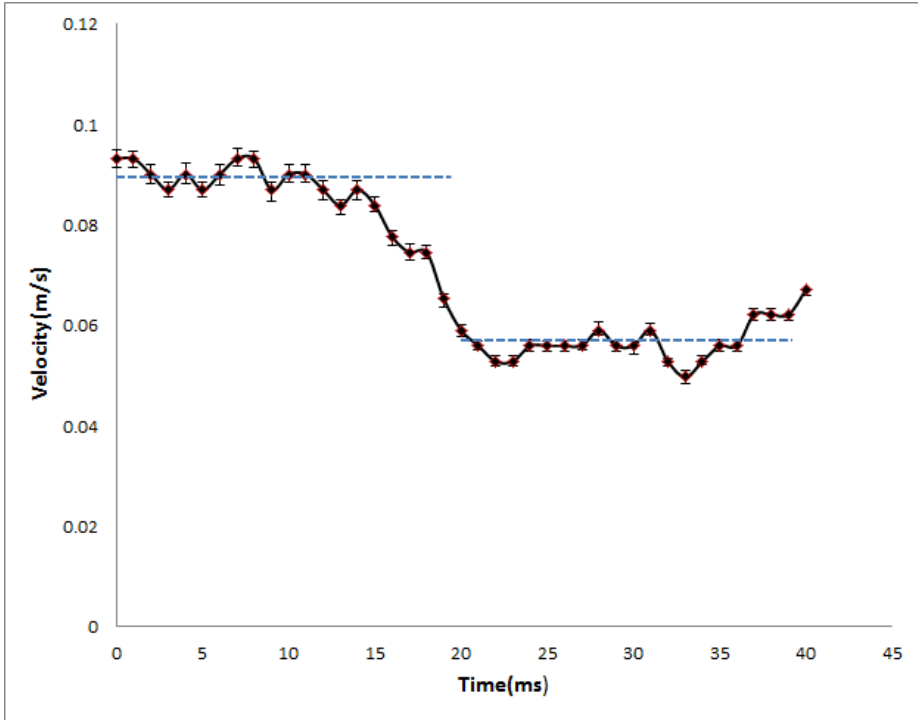
Another experiment has been done on a substrate and step made of the same insulating material. Figure 5.7 shows behavior of the liquid nitrogen droplets when the step and substrate made of paper tape. The brass sub-



**Figure 5.4:** The motion of  $CM_x$  of a typical droplet on a brass step when substrate is brass. The dash line shows the slope of the fitted line on the data. The angles 1 and 2, and show the sloped of the fitted line on the data points before and after the step and angle a shows the different between 1, and 2.

strate was covered by a layer of tape to provide an insulating substrate then one layer of the same paper tape was used to create a step on the insulating substrate. The cross symbols indict nucleate boiling of the droplet at edge of the step. It is obvious that droplet velocity has been increased and no jump has been observed. The same experiment was done with PDMS and





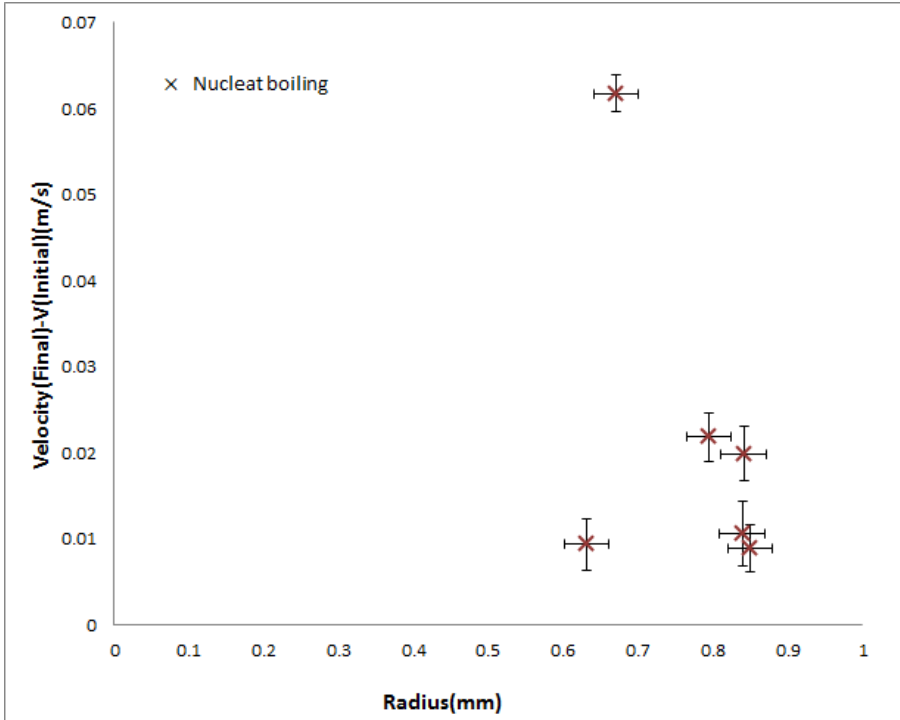
**Figure 5.5:** The velocity evolution of the droplet on a brass step. This graph is derivative of the graph in figure 5.4

glass and the nucleate boiling was observed for this case as well.

#### 5.1.4 PDMS on brass substrate

The figure 5.8 demonstrates the change in velocity of the different sizes of liquid nitrogen droplets while the step material is PDMS on the brass substrate. An important result of this experiment is that there is nucleate

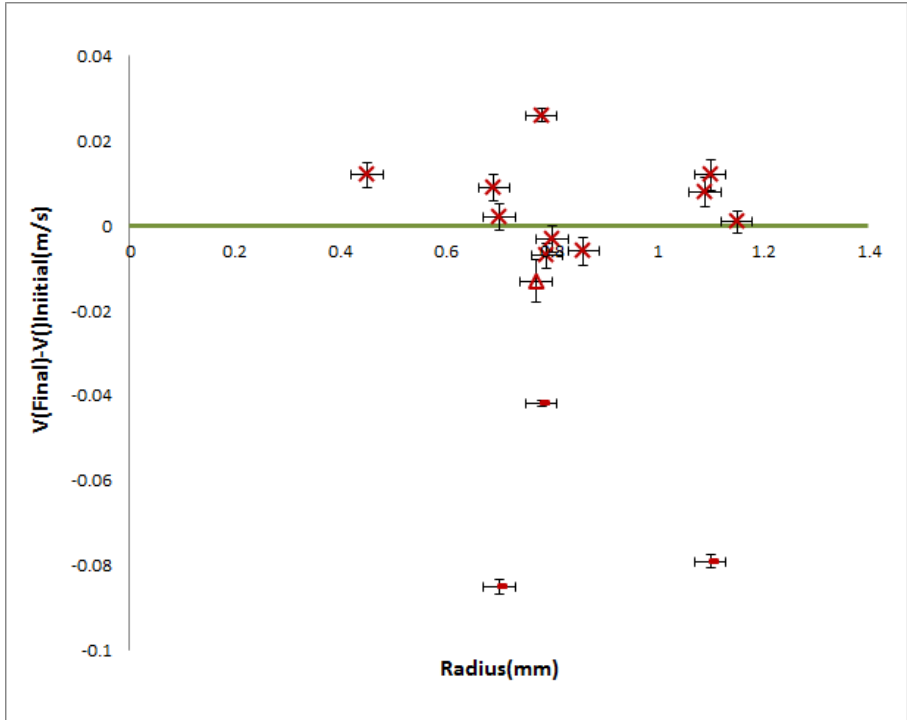




**Figure 5.7:** The result of the droplets interaction when substrate and step are made of paper tape. For these data the brass substrate was covered by layer paper tape and one another layer of the same paper tape was used to make a step on the tape substrate.

smaller droplets, figure 5.9.

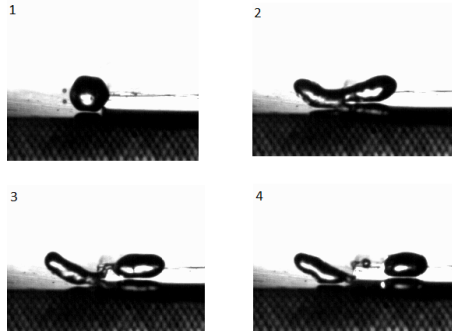
Figure 5.9 shows that the droplet split into two droplets when PDMS is used as a step.



**Figure 5.8:** The velocity variation of the different sizes of liquid nitrogen droplets before and after the step. The step material for these data series is a thin layer of PDMS with  $\sim 0.2\text{mm}$  in thickness.

**Table 5.1:** Materials properties [42]

Properties	Brass	Bronze	PDMS	Plastic	Glass
Thermal conductivity (W/mK)	109-121	15-60	0.15	0.03	0.8-1.4
Specific heat (J/kgK)	380	435	1460	1670	840-750



**Figure 5.9:** A typical droplet under a specific condition splits into two droplets due to high elongation on the PDMS.

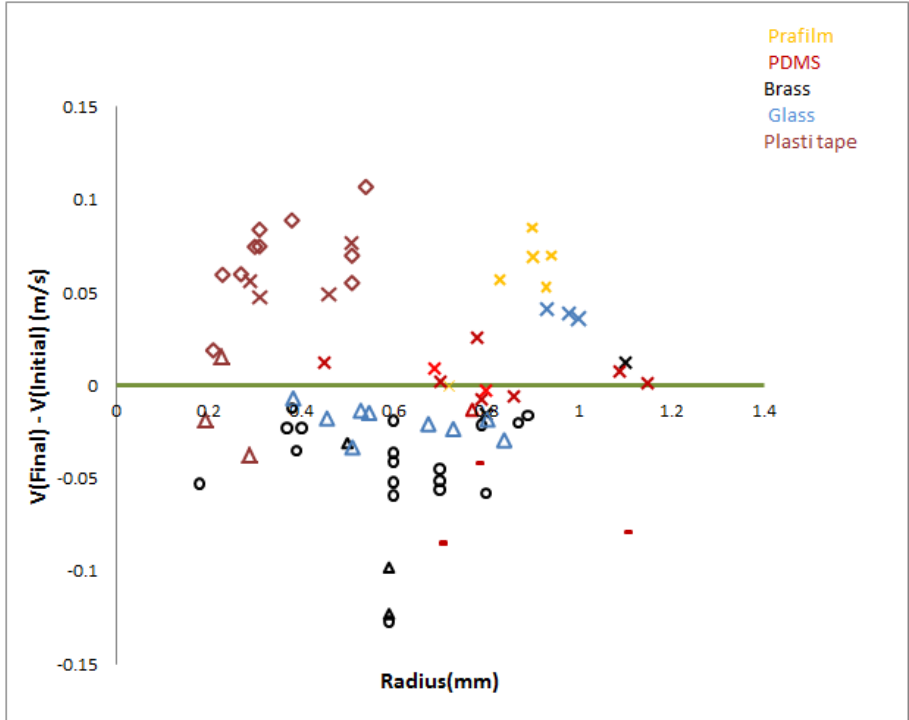
### 5.1.5 A comparison of different material on brass

Figure 5.10 shows a comparison between the results of the experiment for several step materials: glass, PDMS, parafilm, and brass on brass substrate. Droplets on the glass step rarely indicate an increase in velocity however, PDMS and parafilm cause a relatively high variation in velocity for the droplets as a result of the interaction with the edge of the step.

In addition, the experimental result shows that materials with low conductivity and high specific heat give a stronger interaction with the edge (see table 5.1).

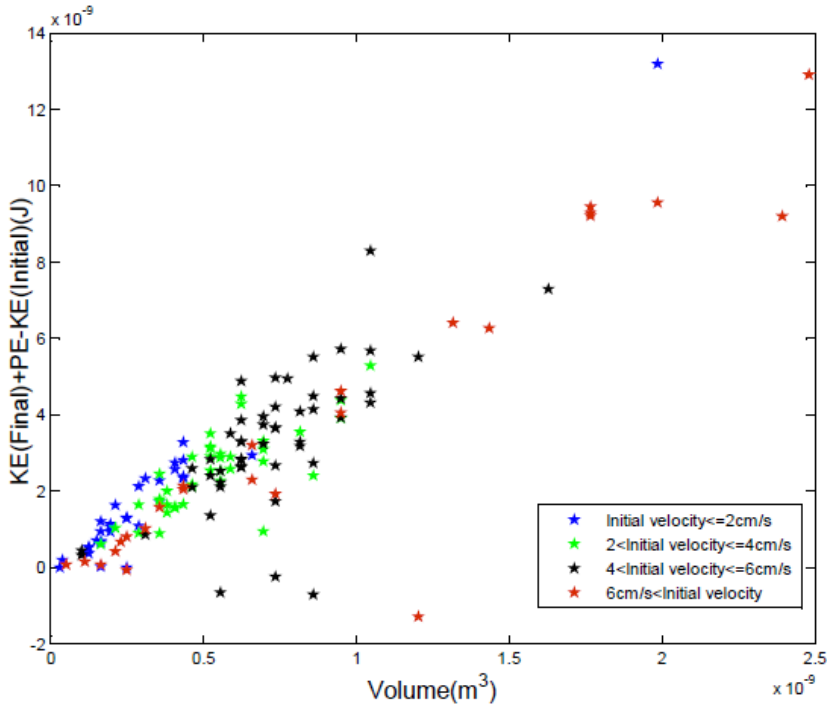
### 5.1.6 Gained energy

The experiment of plastic tape on brass substrate presented in figure 5.3, has been investigated with more experimental data. Looking at the figure 5.11, it is obvious that liquid nitrogen droplets usually show strong interaction and high jumping off the step. This behavior is observed for numerous droplets with different ranges in velocity and size. The height



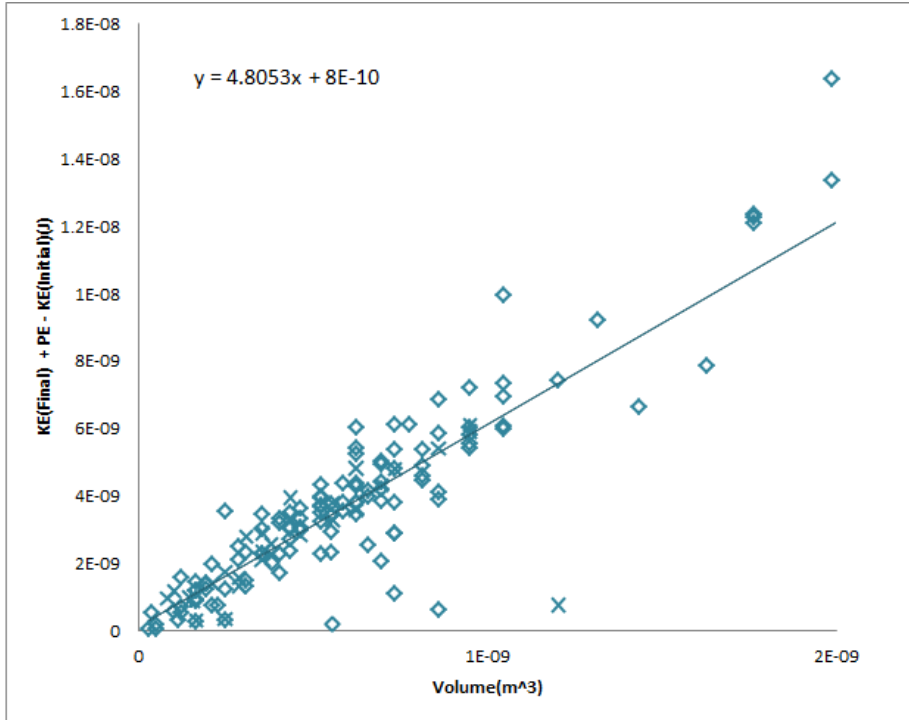
**Figure 5.10:** A comparison of the variation in velocity as a function of size for liquid nitrogen droplets on a brass substrate with different materials as a step.

of the jumps were plotted against velocity and size separately, but no correlate of these parameters was found (see appendix D). The energy gain has been calculated by equation 4.12. A linear relation is clearly observed between energy gain and the volume of the droplets. The droplets speeds has been categorize in different velocity domains. The velocity domain  $2 \text{ cm/s} < v \leq 4 \text{ cm/s}$  in blue color,  $4 \text{ cm/s} < v \leq 5 \text{ cm/s}$  in green color,  $5 \text{ cm/s} < v \leq 6 \text{ cm/s}$  in black color and  $v > 6 \text{ cm/s}$  in red color. The data



**Figure 5.11:** More than 140 droplets with different ranges in radius and velocity show a linear behavior for energy gain versus volume. The different colors show the different range of the droplet's velocity. The linear behavior is observed for all different velocities of the droplets.

in all the different radii and velocities of the droplets demonstrate a linear relation of change in kinetic energy with the volume.



**Figure 5.12:** The energy gain for liquid nitrogen droplets of different sizes and different initial velocity on a brass substrate when the step is plastic tape with a thickness of roughly  $\sim 0.2$  mm.

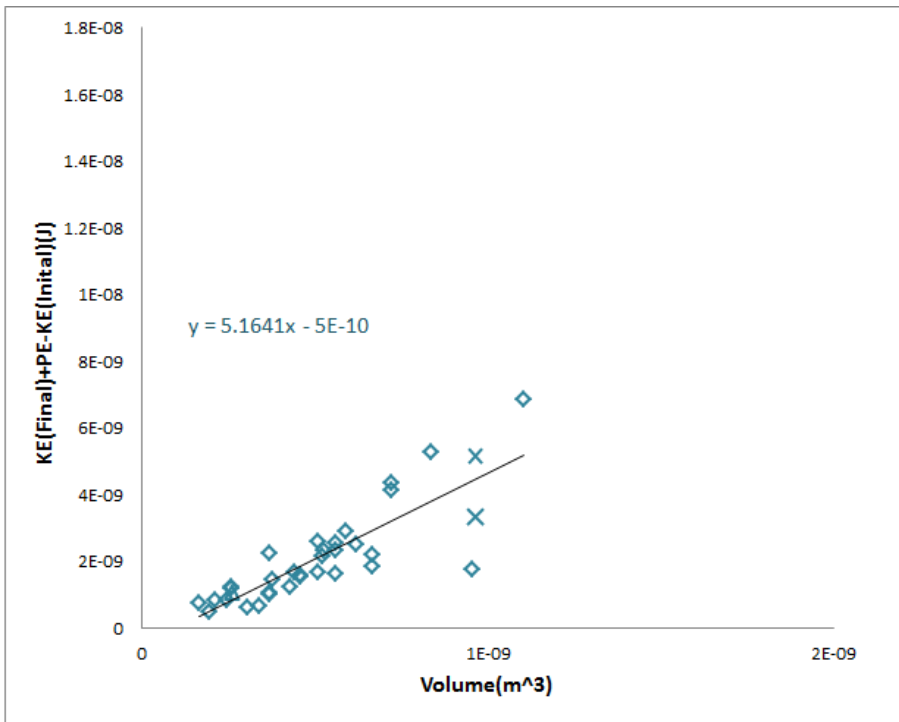
## 5.2 Different height of the step

In order to understand how a droplet behaves under different height of the step, the experiment with three different step heights is performed while the step material is plastic tape.

In these experiments, the initial droplet velocity is between 10 mm/s

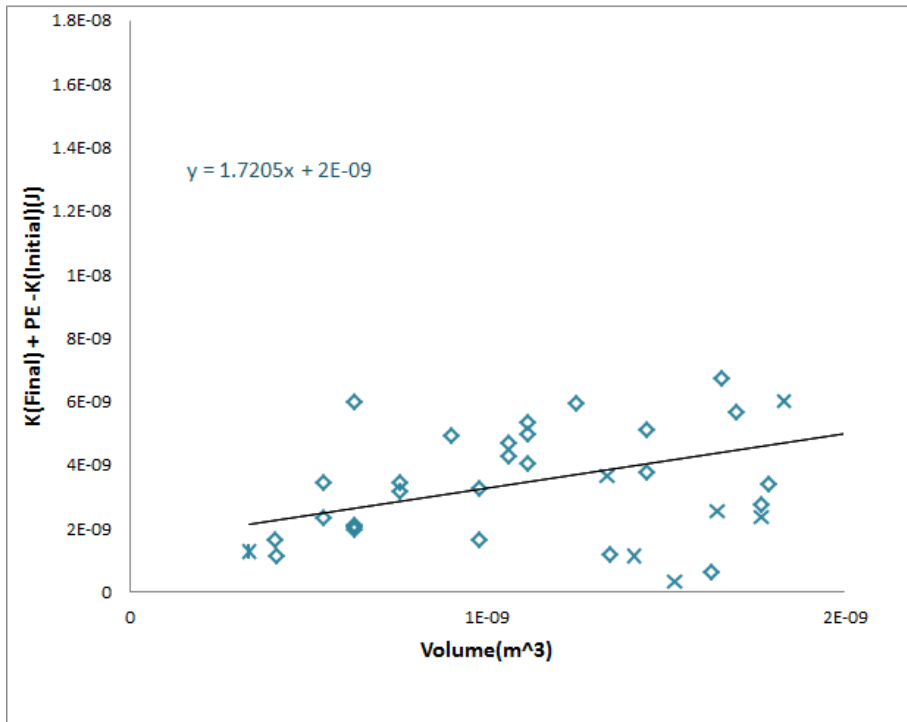


and 130 mm/s, the radius range of the droplet is between 0.2mm and 0.9mm. Using equation 4.12 the energy gain was calculated before and on the step while moving with a uniform velocity. As figure 5.12 shows, most of the droplets which have a volume smaller than 1mm<sup>3</sup> experience significant detachment from the surface as a result of the interaction with the edge of the step.



**Figure 5.13:** The gain in energy for liquid nitrogen droplets on a brass substrate with a step height of roughly  $\sim 0.44mm$ , made from plastic tape.

Another aspect of this graph is the behavior of the larger droplets.



**Figure 5.14:** The behavior of liquid nitrogen droplet on three layer plastic tape with a thickness of  $\sim 0.59$  mm.

They all gained more energy compared to the smaller droplets but only a few could detach from the surface. The majority of the larger droplets even the ones with high energy gain, only interacted with the edge of the step and experienced boiling during the contact with the surface. Figure 5.13 shows a similar behavior for changes in energy gain for two layer plastic tape with a total thickness around 0.44 mm.

The velocities of these droplets vary between 12 mm/s and 85 mm/s,

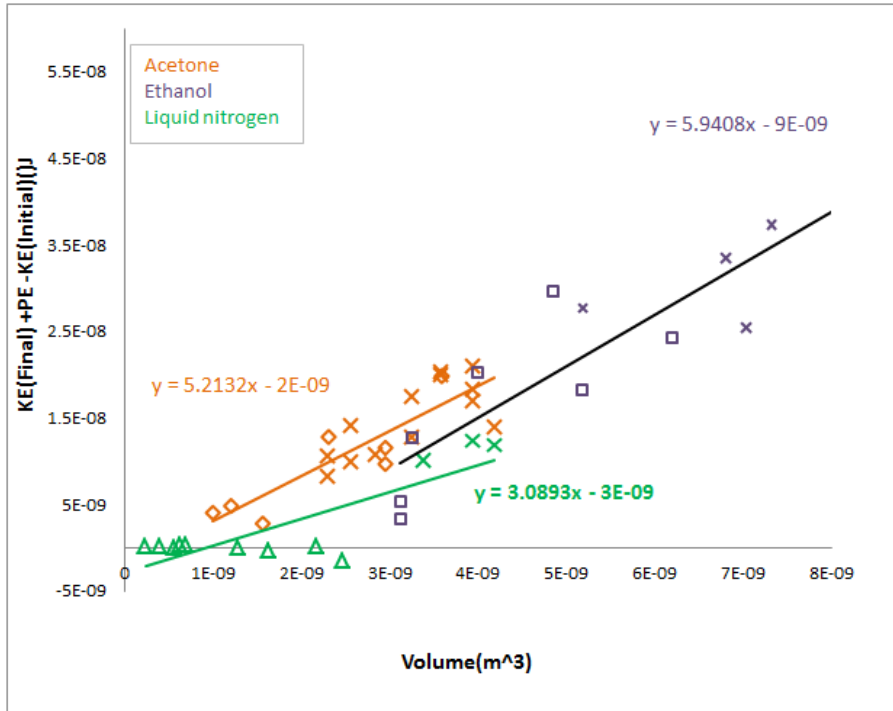
and the range of the energy gain is the same as for one layer plastic tape, roughly  $\simeq 6 \times 10^{-9} \text{J}$ . As a result, the slope of the linear fitted on the data points, for both one and two layer plastic tape are roughly the same. However, when plastic tape with a thickness of  $0.59 \text{mm}$  is used as a step, liquid nitrogen droplets of various sizes behave differently. In this case, the relation is not quite linear. Instead, scatter data is observed for droplets with volumes larger than  $1 \text{mm}^3$ . When the volume is smaller than  $1 \text{mm}^3$  the range of the gained energy is  $\sim 4 \times 10^{-9} \text{J}$ , which is less than for thinner layers of plastic tape, figure 5.14.

### 5.3 Different liquids

We examined three different liquids on the same step material of glass. The temperature required for Leidenfrost is slightly different for each liquid due to the different boiling temperature. Data were gathered when the surface temperature was roughly  $230^\circ$  for ethanol and around  $200^\circ \text{C}$  for acetone. However, all the data for liquid nitrogen were taken at room temperature ( $20^\circ \text{C}$ ). The sizes of the droplets varied for different liquids. The larger droplets for ethanol and acetone are still smaller than the capillary length. Since the surface tensions for acetone and ethanol are higher than for liquid nitrogen, it is quite difficult to get small droplets of these liquids by using syringe. The symbols for these data have the same definition as for the other graphs. The graph 5.15 shows the data gathered for a wide range of droplet sizes of acetone and ethanol on a glass as a step at a temperature of  $230^\circ \text{C}$ . It illustrates the difference in energy gain.

Both nucleate boiling and jumping are observed for both ethanol and acetone.

Turning to details, the energy gain by acetone and ethanol droplets for the same range in size is roughly the same. Table 5.2 shows that the liquid properties of these two are not very different, except the viscosity which is larger for ethanol. Liquid nitrogen droplets rarely interact with the edge of the glass. The green graph in figure 5.15 shows few interacting droplets and neither of them jump. As a fact, for liquid nitrogen experiments the



**Figure 5.15:** Different sizes of ethanol droplets (90 percent) on a glass step at a temperature of roughly 230 °C and acetone in roughly 200 °C experience a Leidenfrost regime.

substrate and ambient have the same temperature while it is different for ethanol and acetone droplets.

**Table 5.2:** Liquids properties [42]

Liquids	Liquid nitrogen	Ethanol	Acetone
Boiling point( $K$ )	77	351	329
density ( $kg/m^3$ )	808	789	790
Surface tension ( $mN/m$ )	8.4	22.4	23.3
Viscosity ( $kg/ms$ )	$1.58 \times 10^{-4}$	$1.07 \times 10^{-3}$	$3.07 \times 10^{-3}$
Specific heat( $J/kgK$ )	2042	2300	2150

## Chapter 6

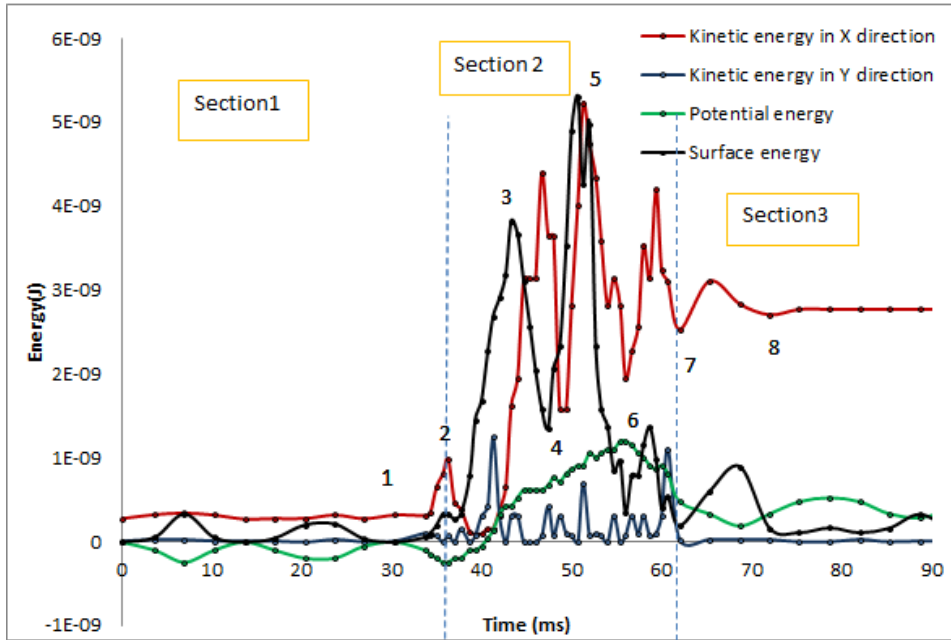
# Interpretation mechanism and model

It is known that for droplets on relatively hot substrate as our studies, differential surface tension between two points on the surface of the droplets can be introduced by the temperature gradient and create a tangential force. Such an effect causes a flow on the droplet from the hot point to the cold point which is known as Marangoni flow.

When the droplet touches a step with a low thermal conductivity, it will experience a nucleate boil and start boiling due to the heat transfer of the droplet. A movie of a typical droplet was analyzed frame by frame during crossing the step in order to obtain more information about the evolution of the energy of the droplet during the jumping process.

Figure 6.1 shows energy evolution of the droplet with low resolution of  $3ms$  in sections one and three and higher resolution in section two. The figure is divided by three sections. The surface energy and potential energy have been calculated with respect to the spherical shape of the droplet. The eight picture of figure 6.2 shows the droplet deformations during the motion. The pictures of figure 6.2 correspond to the sections of figure 6.1 as following:

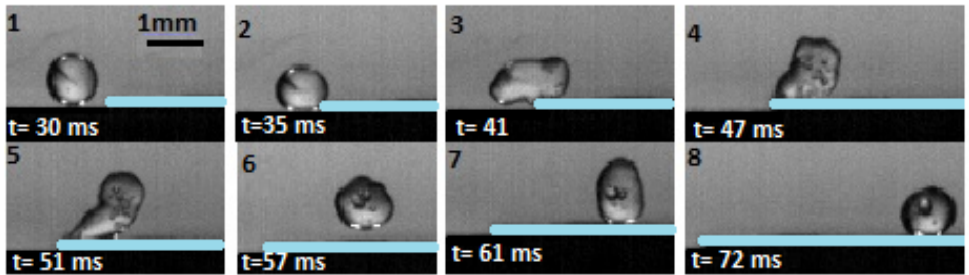
Pictures (1,2), pictures (3,4,5,6) and pictures (7,8) have been taken



**Figure 6.1:** A comparison of the droplet energies over time. All the energies are plotted in metric units.

when the droplet was in the sections one, two, and three of figure 6.1 respectively. In figure 6.1, the kinetic energy part due to motion in  $x$  direction, the kinetic energy part due to motion in  $y$  direction, the potential energy and the surface energy are demonstrated by red, blue, green and black colors. The fluctuations of the energy diagrams are due to the bouncing motion of the droplet which are more relevant to sections one and two.

The almost constant kinetic energy in sections one and three is due to



**Figure 6.2:** Sequence of eight pictures illustrating the jumping process of a liquid nitrogen droplet with a radius 0.45mm and an initial velocity of around 40 mm/s.

the constant velocity of the droplet. High fluctuation is observed when the droplet interacts with the edge of the step (section 2). The important point seen in this graph is a high gap between initial and final kinetic energy, which proves that the droplet gained energy during passing the step.

The black graph displays the variations of the surface energy of the droplet as a function of time. Weak fluctuation is also observed in section one and three due to the weak bouncing of the droplet. Increasing behavior of the surface energy is observed while droplet touches the edge  $t=35\text{ms}$  until it detaches of the step  $t=51\text{ms}$ . Then the surface tension decreases rapidly to a minimum value while the droplet reaches to highest height  $t=57\text{ms}$ . When the droplet is at a maximum distance from the surface it has lost contact with the surface and is restored to its minimum surface. When droplet is at maximum distance of the surface it means that the droplet lost its surface energy as a result of the contraction. A maximum value of the surface energy happens before the separation due to high elongation of the droplet.

Over all, due to the interaction, the droplet gains roughly  $5.5 \times 10^{-9}\text{ J}$



in surface energy which it later loses after detachment from the edge.

The potential energy of the droplet shows an increasing behavior while the droplet touches the step until reaches to maximum distance from the surface. The potential energy of the droplet reaches to a constant value at  $t > 70ms$  more than initial value because of the step height at  $t < 35ms$ . The kinetic energy in  $y$  direction shows a weak fluctuation during the interaction  $35 < t < 60ms$ .

Additionally, the kinetic energy due to motion in the  $x$  direction increases remarkably while the droplet is on the step. It demonstrates that an extra force acting on the droplet as a result of interaction. We believe that the effect of the interfacial tension due to thermal gradient causes a surface flow from a hot point to a cold point on the droplet. This generates a hydrodynamic pressure force, named the Marangoni force, which acts on the droplet surface and might affect the shape of the droplet [43].

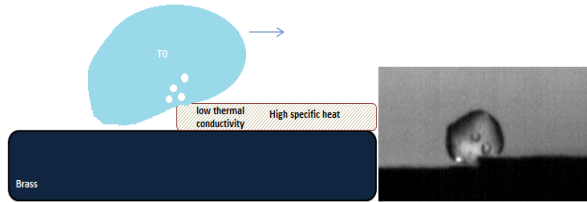
We hypothesize that the Marangoni flow generates the motion of the droplet and its source of energy is the thermal gradient inside the droplet.



**Figure 6.3:** A liquid nitrogen droplet in the film boiling regime moves toward an insulating step, which is lower than its radius.

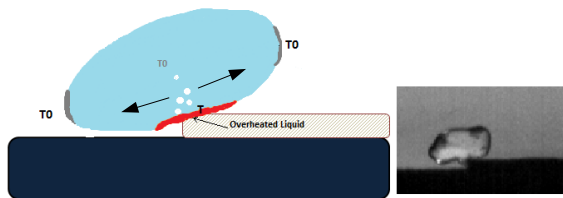
Figure 6.3 shows a droplet on a Leidenfrost surface, in which a piece of the plastic tape was placed on the brass surface. The height of the step is defined by thickness of the tape which is roughly 0.2 mm.

When a droplet touches the edge of the step with an initial momentum (if it is enough to break down the insulating layer around the droplet) it



**Figure 6.4:** A liquid touches the edge of the step, which causes nucleate boiling in the contact point.

starts to deform and boiling. This is evidenced by generation of several bubbles inside the droplet (see figure 6.4).

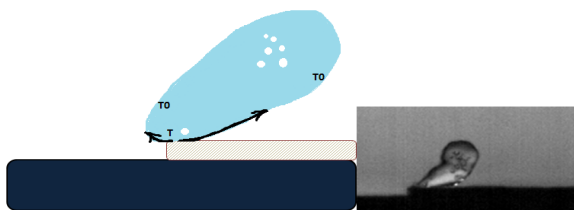


**Figure 6.5:** The Marangoni flow resultant of the thermal gradient drive the fluid in forward and backward direction.

In the contact point, temperature is much above the other part of the droplet  $T \gg T_0$ . This generates a Marangoni flow in all directions at the interface. While molecules of the liquid are in contact with a solid surface, adhesion forces (forces between different molecules) keep the droplet in contact with the edge. The droplet would be in contact with the surface until the surface tension overcomes the adhesion forces. Here, the main effect of

the Marangoni flow could be the droplet deformation (see figure 6.5).

While droplet moves over the step (figure 6.6) the net Marangoni flow will be in forward direction. We propose the vapor pressure underneath the droplet applies a force which can push the droplet in the upward direction. This causes a cooler point at the bottom of the droplet. In this case, at the cooler point, the surface tension is greater compared to other parts and cause a great pressure on that part of the droplet (see equation 2.6). Due



**Figure 6.6:** The symmetry breaking and whole the fluid flow in forward direction.

to this pressure gradient inside the droplet, the liquid pump to the lower pressure on the other side.

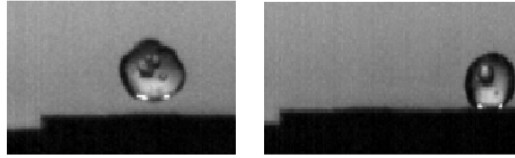
When a droplet surface tension overcomes the adhesive forces, it will be released from the edge and again contract to a spherical shape. This will add momentum to the center of mass, which may push the droplet forward and sometime upwards as well, see figure 6.7.

In order to support our hypothesis the Marangoni flow rate is estimated as follows. Daniel et al. presented a method which estimates a scale of Marangoni convection in water droplet on a super-hydrophobic surface [44].

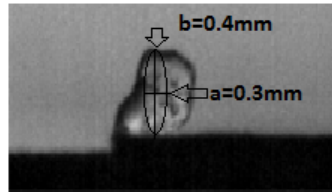
The scale formula for Marangoni flow is defined as follows:

$$U_{MNitrogen} \sim \frac{\alpha \Delta T}{\mu} \quad (6.1)$$

They measured the Marangoni velocity inside a water droplet with a



**Figure 6.7:** Two pictures show when droplet is in highest height and when it returns to the surface.



**Figure 6.8:** Shows when droplet is on the step, this case was considered for calculating the flow rate.

radius of 0.5 mm and 10 K temperature gradient between the bottom and top of the droplet on a super-hydrophobic surface. The measured velocity is around  $U_{measrment} = 1\text{mm/s}$ . The equation 2.14 and  $U_{measrment}$  are used as a scale for calculating the Marangoni velocity on a liquid nitrogen droplet.

$$U_{MH2O} \sim \frac{\alpha\Delta T}{\mu} \quad (6.2)$$

Using the data of table 6.1, the scale value of the Marangoni velocity is calculated for water; the result is 1.55mm/s. For liquid nitrogen a droplet with a maximum temperature gradient 200 K, the scale velocity is around

240.515 m/s. Now, by using the following equation we can estimate the Marangoni velocity on a liquid nitrogen droplet.

$$U_{measrmentH2O}/U_{measrmentN_2} = \frac{\frac{\alpha\Delta T}{\mu}}{\frac{\alpha\Delta T}{\mu}} \quad (6.3)$$

$$(1mm/s)/(U_{measrmentN_2}) = 1.55/240.5 \quad (6.4)$$

Then the Marangoni velocity in a liquid nitrogen droplet is calculated to be 155.2mm/s. In order to estimate the flow rate due to the Marangoni effect the following equation is used:

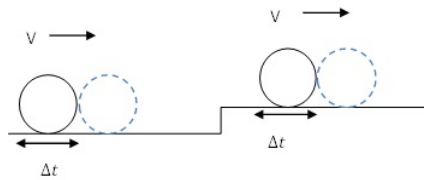
$$flowrate = U_{measrmentN_2} \times Area \quad (6.5)$$

and

$$Area = \pi ab \quad (6.6)$$

where  $a = 0.3$  mm and  $b = 0.4$  mm, half of ellipsoid axes. The area will be approximately  $3.8 \times 10^{-7}m^2$ , and the flow rate due to the Marangoni flow is estimated to be  $5.9 \times 10^{-8}m^3s^{-1}$ .

For obtaining the experimental flow rate according to figure 6.9, the flow rate before and on top of the step are estimated. The effect of the initial momentum on the flow rate has been subtracted.



**Figure 6.9:** A schematic of the droplet motion towards the step.

Flow rate before the step is:

$$flowrate\left(\frac{m^3}{s}\right) = \frac{Volume}{\Delta t} = 2.6 \times 10^{-8} \quad (6.7)$$

$\Delta t$  defines time interval for front and rear of the droplet pass one point. The flow rate on top of the step is:

$$flowrate\left(\frac{m^3}{s}\right) = 7.4 \times 10^{-8} \quad (6.8)$$

The flow rate due to the interaction with the edge of the step is obtained by:

$$flowrate\left(\frac{m^3}{s}\right) = 4.8 \times 10^{-8} \quad (6.9)$$

In comparison, the flow rate due to the Marangoni effect is in the same range as the measured value.

**Table 6.1:** Liquid nitrogen properties [45], [42], [46]

Thermal conductivity, $\kappa_w$ ( $W/mK$ )	$25.83 \times 10^{-3}$
Specific heat, $C_p$ ( $J/kgK$ )	2042
Thermal diffusivity, $\kappa$ ( $m^2s^{-1}$ )	$1.6 \times 10^{-8}$
Thermal expansion coefficient, $\alpha_t$ $K^{-1}$	$7.53 \times 10^{-3}$
Dynamic viscosity, $\mu$ ( $kg/ms$ )	$1.58 \times 10^{-4}$
Kinematic viscosity, $\nu = \mu/\rho$ ( $m^2/s$ )	$1.9 \times 10^{-7}$
Latent heat of vaporization, $L_v$ ( $J/kg$ )	$1.9838 \times 10^3$
Density, $\rho$ ( $kg/m^3$ )	808

## Chapter 7

# Conclusion and future work

The purpose of this study is to understand the specific physical mechanism of jumping Leidenfrost droplets, as well as the effective parameters of the process. This final chapter will begin with summarizing the most important results. Moreover, our model for the result of the Marangoni flow rate estimation is presented. Finally, recommendations for future work are proposed.

### 7.1 Summary and results

In summary, the experimental result of different materials as a step over a brass substrate showed that jumping occurs for liquid nitrogen droplets on different insulating step materials. Further, no nucleate boiling has been observed when the droplets moved on a metal. A metal substrate with a step from the same material remained steady in temperature and even the edge of the step was in Leidenfrost regime. The temperature stayed constant because of the high thermal conductivity of brass; even during an impact process a droplet could not cool down the edge.

We also observed how the droplets behaved when the substrate and the step were covered with an insulate layer (like PDMS or tape) in which case the droplets exhibited nucleate boiling when it touched the edge of the step.

The low thermal conductivity of these materials makes a possible thermal gradient as a result of the impact process. Different materials such as glass, PDMS, parafilm, paper tape and plastic tape with a thickness of less than roughly 0.3 mm were utilized in probing the effect of the material properties on the jumping phenomenon. The measured velocity ranges between those materials, which means that the energy gained by the droplets were different. Using PDMS as a step showed a strange behavior in the sense that a droplet would split into two smaller droplets after elongating and splashing, which was observed in some cases. A comparison of the results for different materials showed that great speed variation is observed for plastic tape. This means that more energy is gained and as a result relatively high jumping is observed for droplets interacting with this material. This means that higher jumping can be observed for materials that have lower thermal conductivity and higher specific heat.

Numerous liquid nitrogen droplets were analyzed using plastic tape as a step. The results can be summarized as follows:

We observed that the energy gain varied for liquid nitrogen droplets of the same size on a specific step material. Further the same energy gain could show difference in jumping. The results also showed that droplets of the same size and initial speed had different energy gains. These results indicate that the microscopic structure of the material might be important for the jumping. However, more experiments need to be done under well controlled conditions to be able to confirm this idea. All droplets regardless of the difference in size and speed, gave a linear relation for energy gain versus volume. We found no correlation neither between initial velocity and the height of the jump nor between the size and height of the jump. For a typical liquid nitrogen droplet, a 3000 fps movie has been analyzed for studying the details of the change in energy due to the interaction with plastic type on a brass surface. By comparing the changes in energy of a droplet it is shown that the maximum surface energy released due to the jumping process was in the same range of the total droplet energy. Analyzing the speed variation over time for a typical droplet, demonstrated a higher velocity on the step, which proves that a force is acting on the droplet and accelerates it. The investigated material, which different thickness of



the step showed following:

The droplets energy gain was not remarkably different for a step with twice the thickness. For a higher step liquid nitrogen exhibited an almost scattering behavior with a weak linear trace. This means that a critical value for the step height could probably be found when the behavior of the droplets is not linear anymore. But more data is needed for different step heights between 0.2 mm and roughly 0.6mm. Finally, the last results present a comparison between alcohol, acetone and liquid nitrogen.

We have demonstrated that the jumping phenomenon happens for each of these three liquids when they are in the Leidenfrost regime. We also observed that alcohol and acetone experienced a higher energy gain in comparison to liquid nitrogen. Liquid nitrogen rarely showed any nucleate boiling with the edge of the glass step, but the other liquids showed both nucleate boiling and jumping. The same thermal gradients were considered for all of the three liquids. There is a significant difference in surface tension for liquid nitrogen compared to both ethanol and acetone. One idea is that the difference in energy gains for these liquids could be a result of the difference in surface tension. If we compare the viscosity of these liquids, we notice that a significant difference in viscosity for ethanol and acetone didn't effect on the energy gain. We hypothesized liquid motion generated by Marangoni effect on the droplet due to thermal gradients which causes surface tension gradients. We also estimated Marangoni flow rate which was in the range of the measured liquid flow rate.

## 7.2 Future direction

This phenomenon is a complicated process with a number of effective variables. For future work it is essential to improve the control of the varying parameters in order to specify how each of them influences the process. We also need to have more data for different liquids with different properties to be able to emphasize our results. Another suggestion could be to investigate the microscopic structure of the edges since we obtained droplets with roughly the same size and initial speed but with different energy gains.

One idea is that this might be the result of the droplets motion before the impact or the structure of the edge of the step. The most interesting direction of future work would be to do the experiments with water droplets. We tried to get data for this; however the high temperature that is needed for reaching the Leidenfrost point limits what step materials can be used. Furthermore, I think that using a tracer inside the droplet, could be useful when measuring the Marangoni velocity on the surface of the droplet. Using an infrared camera would be helpful for finding how the temperature inside and on the surface of the droplet changes due to the interaction. In addition, designing a setup that keeps the environmental condition constant and controllable is suggested.

## Chapter 8

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## Appendix A

# PDMS Fabrication

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To use PDMS as a step on the brass we need to make a sufficient thickness of it. Here I explain the process of the making of 0.2 mm thickness of PDMS. PDMS is a liquid with high viscosity that is optically transparent, completely flexible and which in solid phase shows a hydrophobic property. To transform it into a flexible solid, a required amount of curing agents should be added to the PDMS base. This helps to form internal cross links, which function as a branching point. Step by step PDMS fabrication is explained as follow:

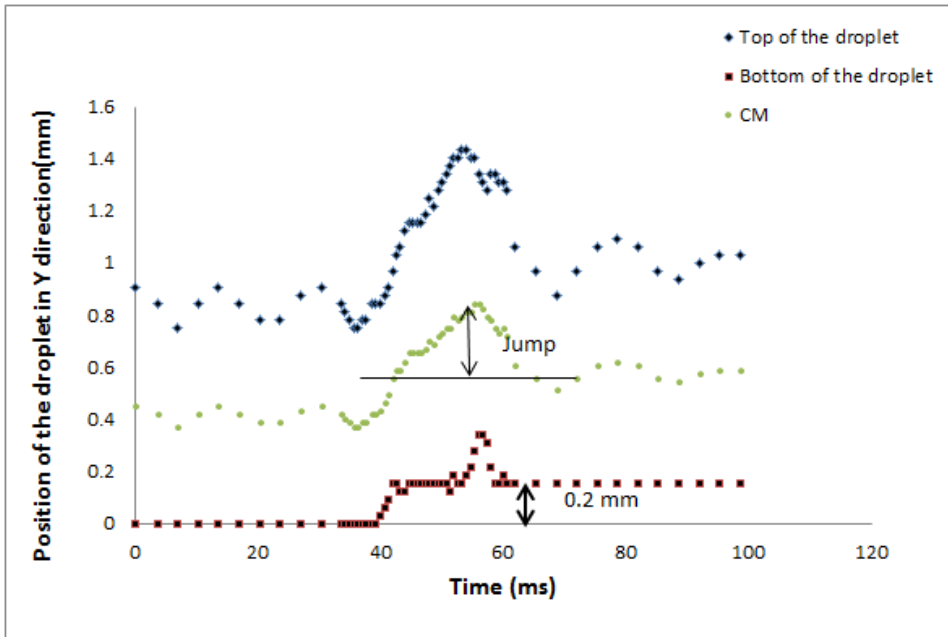
In the beginning, we should put a clean container on the electronic weighing machine which is adjusted to zero. Then we add 1:10 curing agent and PDMS base. For the next step, they should be mixed until we can be sure that the curing agent is uniformly distributed in the PDMS base. A lot of bubbles are produced due to the mixing, which should be completely removed from the mixture by degassing it in vacuum. When a uniform mixture without bubbles is obtained, we should spin it over  $700rpm/s^2$  for around 60 seconds to obtain a PDMS with a thickness of 0.2 mm. The last step is baking the PDMS in 90 °C for around one hour [47].



## Appendix B

# Appendix B

Figure shows the motion of the top, bottom, and CM of the droplet versus time. Low fluctuation is observed on the graph which is because of the bouncing of the droplet. The height of the detachment from the surface is observed on the graph of the CM.

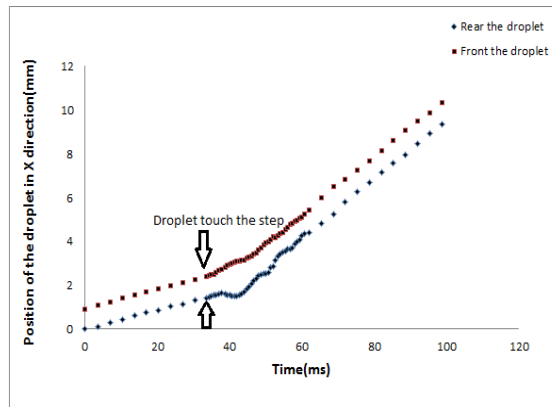


**Figure B.1:** Position of the top, bottom and CM of the droplet versus time. A typical droplet moving on a brass substrate with a step height roughly 0.2 mm made from plastic tape.

## Appendix C

# Appendix C

The motion of rear and front of the droplet is observed in figure C.1. After  $t=40$  ms elongation of the droplet is clear in graph while the droplet is in touch with the edge of the step.

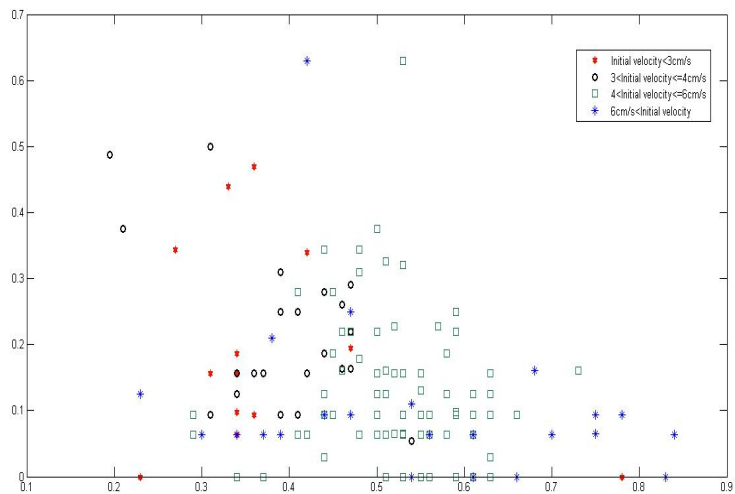


**Figure C.1:** position of the rear and front of the droplet as a function of time. A typical droplet moving on a brass substrate with a step height roughly 0.2 mm made from plastic tape.

## Appendix D

# Appendix D

Figure shows no correlation between size of the droplets and height of the jump.



**Figure D.1:** The maximum position of CM in  $y$  direction versus radius for a range of the velocities. Data gathered for a layer plastic tape on the brass substrate show no correlation between droplet size and height of the jump.



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