Explosive vaporization in microenclosures
and boiling phenomena
on submicron thin film strip heaters

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“Denken ist interessanter als Wissen, aber nicht als Anschauen”
Johann Wolfgang von Goethe
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Abstract

Explosive vaporization of a highly superheated liquid can be induced by electrically heating a microheater which is in contact with the liquid. This phenomenon is widely exploited as an actuator mechanism in Micro Electro Mechanical Systems (MEMS). For most applications, explosive vaporization must occur in a closed system. However, the majority of recent research focuses on explosive vaporization at atmospheric conditions.

The first part of this thesis presents the effect of confinement on explosive vaporization. The experiments were performed in a device composed of a 50 µm × 50 µm platinum heater on a glass substrate bonded to a silicone chip with an embedded fluidic cavity and channels. Experiments were performed first without the fluidic cover (unconfined). The rates of temperature rise obtained were as high as $70 \cdot 10^6 K \cdot s^{-1}$. The inflection point in the temperature evolution curve was used to identify the time and temperature associated with the formation of vapor on the heater. The maximum nucleation temperature measured was 273 °C. For the experiment in the composed device (confined) measurements revealed that the temperature evolution and the nucleation temperatures were not significantly altered by confinement.

Visualization of explosive vaporization on the microheaters was achieved by means of a pulsed image acquisition system using flash lamp illumination. Image times could be matched to the temperature evolution curves by synchronizing the two sets of data. The inflection point of the temperature curve coincides well with the instant when vapor covers the entire microheater surface. Visualization of explosive vaporization inside the cavity (confined) was also achieved and the confinement did not affect the bubble growth but it accelerated the bubble collapse process.

In the second part of the thesis, a method based on Ion Beam Milling, was applied
to fabricate a thin film platinum heater of $\sim 0.5 \, \mu m \times 3 \mu m$. The micro/nanofabricated structure can be simultaneously used as a heater and a temperature sensor. The temperature coefficient of resistance of the heater is $1.6 \times 10^{-3} \, K^{-1}$. Electrical pulses were used to increase the heater temperature in contact with deionized water. The temperature evolution of the heater subjected to pulsed heating showed a rapid transient behavior followed by a temperature plateau. If the temperature at the plateau is high enough, after few $\mu s$ a sudden increase of temperature indicates the formation of vapor on the heater. An unexpected oscillation in the temperature was observed following vapor formation. This is caused by subsequent bubble growth and collapse cycles taking place at a high frequency (MHz). The oscillation stops after 40 to 50 $\mu s$ when the temperature of the surrounding liquid increased. Once vapor forms on the heater the growth of the bubble is controlled by the energy balance at the liquid bubble interface. Energy is supplied to the bubble interface by conduction through the vapor phase. Since the liquid away from the hot surface is colder than in the vapor (saturation) conduction heat transfer in the liquid cools down the interface. The bubble cannot keep the Laplace pressure at certain bubble sizes since the heat removal is larger than the heat input. This makes the bubble to collapse and to re-growth again after touching the hot heater surface. Furthermore, the effect of the initial temperature of the liquid was studied showing that the oscillation frequency decreased and finally disappeared when the initial temperature of the liquid was increased. Visualization of vapor on the heater was achieved by synchronizing the heating pulse with a flash lamp and confirmed the formation of vapor on the heater. This vapor volume oscillation may be exploited in new applications of the thermal actuation.
Zusammenfassung


Chapter 1

Introduction

1.1 Introduction and Motivation

Micro Electro Mechanical Systems (MEMS) are present in many commonly used devices including air bags in automobiles, in portable blood analyzers, in automated drug dispensing systems and in ink jet printers. In the last example an integrated heater, in contact with the ink, is electrically excited to generate an intense localized temperature increase. The ink directly in contact with the heater, reaches temperature high enough to partially vaporize. Due to the fact that the vapor has a specific volume several orders of magnitude larger than the liquid, this sudden phase change is accompanied with a considerable increase of the volume of the system. As a result a droplet of ink if forced out of the cartridge towards the target. This kind of actuation is commonly known as thermal bubble actuation. This constitutes a very concentrated mechanical power source that could be exploited in several ways such as in MEMS actuators for micro valves and micro pumps.

The thermodynamics behind thermal bubble actuation has been extensively studied in the last decade but is not yet fully understood. The problem is a combination of intercoupled phenomena:

1. Thermal heat transfer from the heater to the liquid, mainly conditioned by materials and surface state,
2. nucleation mechanism mainly affected by temperature involved and heating rate,

3. bubble dynamics, affected by the geometry, the pressure the ambient temperature and the properties of the liquid.

The next section introduces different examples of microsystem using thermal actuation. The examples are divided into four groups, ink jet printing (for ink but also for other applications such as biological reagents), applications of the thermal bubble to microfluidics (microvalves and micropumps), application of the thermal bubble to external flows (micro fuel injectors and microthrusters) and optical switches.

### 1.2 Applications of thermal actuation in MEMS

**Ink jet printing**

The history of Ink jet printing began in the 1970’s. Curiously it was discovered by accident when a Canon researcher brought a soldering iron into contact with an ink-filled syringe. A bubble formed and forced ink to spurt from the nozzle. Canon patented what they called the Bubble Jet technology in 1979. At approximately the same time, researchers at Hewlett Packard (HP) also discovered the technology, which they called Thermal Ink Jet. This technology is based on a sequence of processes that can be summarized in 3 steps:

1. An electrical current is applied to a thin film heater in contact with the ink or the substance to deposit. The liquid directly in contact with the heater reaches a high temperature in a very short time and it vaporizes at a temperature close to the superheat limit.

2. The vapor bubble formed grows and displaces the liquid away from the heater towards a micro nozzle where a droplet of ink is ejected towards the desired substrate.

3. The chamber is automatically refilled and the system is ready to be actuated again.
As the previous description suggests the process does not only suit the ink printing process. In general the technique is known as drop on demand (DOD). It is one of the most promising technologies for the development of organic electronic devices where organic layers are deposited by an ink jet printer (Natori et al., 2005). It is also a promising substitute to the conventional patterning techniques because it offers many advantages for microelement fabrication and adhesive joining such as: precise volumetric control of dispensed material, low cost, high speed, and low environmental impact (Hayes et al., 1998).

Applications of the thermal bubble to microfluidics

The thermal bubble also has applications in internal control of microfluidic devices. We find it as an actuator mechanism for micro pumps and microvalves where thermal actuation is preferred to piezo actuators because of easier integration and larger stroke, (Nguyen and Wereley, 2002).

Microvalves

In a typical thermopneumatic microvalve, the working medium is enclosed in a volume in contact with a heater. A flexible wall separates this closed volume from the flow controlled by the valve. The closed position is obtained by actuating the heater which induces an increase in volume of the enclosed system deflecting the membrane. The working medium will depend on the application and on the required membrane displacements: It can be a solid, as paraffin, a liquid, with or without phase change or a gas.

Micropumps

A typical reciprocating micropump is sketched in Fig. 1.1. It is composed of a chamber with a variable volume and inlet/outlet conduits. Typically some rectifier valves need to be integrated in the design to avoid reverse flow. A thin-film resistive element heats the driver working fluid in a secondary chamber above the pump. The heated fluid expands, exerting pressure on the pump diaphragm forcing the liquid out of the chamber. When the heating of the secondary chamber stops, the pump chamber refills with the liquid.
Introduction

Figure 1.1: Reciprocating displacement micropump with thermopneumatic driver, in which a thin-film resistive element heats the driver working fluid in a secondary chamber above the pump. The heated fluid expands, exerting pressure on the pump diaphragm (Laser and Santiago, 2004).

It is also possible to design valve-less micropumps as demonstrated by Stemme and Stemme (1993). The concept of the valve-less micropump is illustrated in Fig. 1.2 where, instead of rectifier valves, the inlet and outlet make up a convergent-divergent nozzle pair with a preferred flow direction. The nozzle-diffuser pair can be easily integrated into microfabricated systems and thermal bubbles were demonstrated to be an effective actuation mechanism for these types of valveless pumps (Tsai and Lin, 2002; Kim et al., 2005).

Figure 1.2: Valveless nozzle-diffuser Reciprocating displacement micropump with thermopneumatic actuation. Instead of rectifier valves the inlet and outlet make up a convergent-divergent nozzle pair with a preferred direction. The thermal bubble can be also in this case used as actuation mechanism (Tsai and Lin, 2002).
Another interesting example of a micropump actuated by the thermal bubble has been proposed by Yin and Prosperetti (2005). In his prototype, an array of heaters are arranged on a quartz substrate along a microchannel. The heaters are actuated sequentially, starting with the closest to the inlet to produce peristaltic movement of the liquid.

Control of biological particles

Thermal bubbles have also been used to move and control particles for chemical and biochemical micro reactors. The mechanism presented by Maxwell et al. (2003) is sketched in Fig. 1.3. A well of about 15 \( \mu m \) side square (the capture well) is machined on the bottom of a microchannel. This well is connected to a larger chamber (100-500 \( \mu m \)) by a small duct. On the bottom of the larger chamber is a thin film microheater. When a back pressure is applied, a bioparticle is drawn into a capture well (Fig 1.3 A). The capture well is sized to accommodate only one particle. Then when a back flow is applied over the top of the device, all the uncaptured particles are swept away (Fig 1.3 B). To release the particle, a voltage is applied to the microheater and a bubble forms (Fig. 1.3 C) and the volume expansion in the bigger chamber pushes out a jet of fluid that ejects the bioparticle from the capture well where it is entrained in the flow and carried away (Fig. 1.3 D).

Applications to external flows: micro fuel injectors and microthrusters

Micro fuel injectors

In combustion engines the fuel needs to be injected in small droplets to ensure fast mixing with the oxidizer. Typical injector systems use high pressure injection through a nozzle to produce droplets of sizes ranging from 10 to 100 \( \mu m \) diameter. The use of thermally actuated microinjectors has been considered to optimize low pressure atomization in Lee et al. (1999) and Tseng et al. (1998). These micro injectors would be conceptually very similar to the ink ejectors used in ink jet printing. They would allow for reduction of the stresses due to high pressure cycling on the nozzle. Additionally, with the micro injector,
Figure 1.3: Control of bioparticle using the thermal bubble (Maxwell et al., 2003).

the droplet size is more uniform and it depends on the nozzle diameter. The operating frequency could reach 10 kHz.

**Microthrusters**

Vaporizing Liquid Microthrusters (VLM) for fine positioning of micro satellites have been also proposed. The prototype presented in Maurya et al. (2005) produced a 120 \( \mu \text{N} \) with 2W heater power at a water flow rate of 0.7 \( \mu \text{l s}^{-1} \). The design consists of a propellant inlet channel, the vaporizing chamber with an integrated heater, and a nozzle outlet. The liquid propellant enters the chamber and it is vaporized. The vapor escapes the chamber through a nozzle producing thrust.

**Optical switch**

Optical switching has been considered to be one of the most promising applications of MEMS. A simple microelectromechanical optical switch could be a combination of a
microactuator with a planar optical fiber. The more widespread technology in optical MEMS switches is based on micromirrors integrated with aligned optical fibers. In these switches the micromirror made of Silicon or a metal film is driven into or off the cross point of the fiber axis by an electrostatic actuator. Since the fiber is coupled with a gap space in these switches, the lack of a waveguide structure in distance of several tens of \( \mu m \) could cause a high transmission loss. Many researchers focus on new designs and optimized solutions for these kinds of all optical switches (Iyer et al., 2004).

Thermal actuation has been proposed for optical switching processes. A liquid matching the index of refraction of the wave guide can fill a trench positioned in the cross point. The light is directed to this microscopic trench filled with liquid. The light travels straight passes through the liquid in the trenches unimpeded; If a shift is required, a bubble forms in the trench and the light is reflected off the interface between the glass wall and the bubble in a new direction (Fouquet, 2000). Other thermally actuated MEMS switches use an air bubble entrapped in a liquid located in a trench that can be pushed into the light path. The air bubble can be displaced in and out of the cross point by thermocapillary (Makihara et al., 1999).

These examples show the enormous potential of the thermal bubble as actuator in MEMS devices. The bubble formation and growth in microsystems has been successfully applied but still it is not properly understood making optimization of the design difficult. As the thermal ink jet printer shows, a successful implementation of the technology depends on the smart combination of the available techniques and on a deep understanding of the thermal and phase-change phenomena involved. For some applications the effect of confinement should be further studied.

1.3 Precedent investigations on explosive boiling

An investigation of explosive boiling on a wire immersed in a pool of fluid was first conducted by Skripov and co-workers (Pavlov and Skripov, 1965; Skripov et al., 1965; Pavlov and Skripov, 1970; Skripov and Pavlov, 1970). A 20 \( \mu m \) diameter platinum wire was heated at a rate of \( 10^6 K/s \) using pulses of duration ranging from 25\( \mu s \) to 1000\( \mu s \). The temperature of the heating wire was monitored by measuring the variation in the resistance
of the wire and simultaneous images of the initial stages of the explosive boiling were acquired. The nucleation temperature was measured analyzing the resistance evolution curve. They found very good agreement with the kinetic theory of homogenous nucleation for various organic liquids and for water.

Avedisian et al. (1999) presented a method for measuring the average surface temperature of a small square thin metallic film deposited on a silicon substrate and immersed in subcooled water during a voltage pulse of short duration. The studied thin film was made of a mixture of tantalum and aluminum. It was a 65 $\mu m$ side and 0.2 $\mu m$ thick square. A bridge circuit with a dynamic amplifier was used to measure the evolution of the electrical resistance. A separate calibration of the thin film resistor was used to convert the electrical resistance to average surface temperature. This method allows the monitoring of the heater average temperature during the heating process. An inflection point in the evolution temperature of the heater indicates the nucleation temperature. The largest rate of temperature rise (heating rate) and the highest nucleation temperature were $0.25 \times 10^9$ K/s and 283 °C respectively.

Iida et al. (1994) used 250 $\mu m \times 100 \mu m$ and 0.2 $\mu m$ thick heaters made of platinum onto a quartz substrate. They studied boiling nucleation phenomena of several fluids by a pulsed heating method. They measured the average surface temperature and photographed the nucleation phenomena using a 10 ns pulse stroboscopic light. They used the acquired images to detect boiling incipience observing that the temperature at boiling incipience increases with the rate of temperature rise and it approaches the temperature predicted by the homogenous nucleation theory (except for water that was about 10 °C below the predicted value). For high rate of temperature rise a large number of tiny bubbles appeared on the heater. The density of nucleated bubbles was $2 \times 10^{10}$/m$^2$ in water.

Together with boiling incipience the growth of the bubble during and after applying the heating pulse was also investigated. Asai (1991) proposed a model for nucleation and bubble dynamics showing also experimental results matching his models (Asai, 1991; Asai et al., 1988). He explained the phenomena defining different regimes: First nucleation in the superheated liquid, then instantaneous formation of a vapor film, followed by rapid bubble growth due to the pressure impulse and bubble collapse.
Okuyama et al. (2004) studied the behavior of boiling bubbles generated on a 22\(\mu m\times 130\ \mu m\) polysilicon film heater used for thermal ink jet. The set-up was the one used in Iida et al. (1994). They studied the dependence of the size and lifetime of the coalesced bubble on the pulse power and the pulse width. The pressure impulse arising during the rapid evaporation was estimated from the size of the coalesced bubble. They concluded that the pressure impulse increases with the superheat in the liquid as the heat flux is increased. The same group (Okuyama et al., 2006) also studied the bubble dynamics of ethyl alcohol succeeding spontaneous nucleation. Also in this study a large number of tiny bubbles were generated in the initial phase of the vaporization which collapsed to form a vapor film. For the faster processes it becomes difficult to observe the tiny bubble and a vapor film seems to appear instantaneously on the heater.

Hong et al. (2004) presented an experimental study of the bubble nucleation and growth using pulse heating and laser stroboscopic system (30 ns temporal resolution). The experimental system allowed the acquisition of images of the phenomena from the top and from the side. In this work the microheater was made of polysilicon with dimensions of 25 \(\mu m\times 80\ \mu m\). The nucleation temperature was estimated using the one dimensional heat conduction equation for a multi-layered heater structure bounded by liquid on one side. The model gives the surface temperature and the nucleation temperature was estimated by the surface temperature at which the first vapor bubble appeared. The nucleation temperature obtained in this case was below the limit of superheat of water (260 °C) and showed a weak dependency on the heating rate. They obtained the time evolution of the bubble wall velocity and acceleration throughout the bubble growth and collapse cycle. The pressure inside the bubble and the extractable work where estimated using the model of Asai (1991).

Glod et al. (2002) investigated the pulsed heating of a thin (10\(\mu m\) in diameter) platinum wire. They obtained simultaneously temperature, pressure and visualization measurements of explosive vaporization during pulsed heating experiments. The maximum measured nucleation temperature was 302 °C for a rate of temperature rise of \(86\times10^6\) K/s. The visualization was achieved using a stroboscopic microscopy technique and a fast pressure transducer was used to capture the acoustic emission from the expanding vapor volume. The pressure inside the growing vapor layer was estimated to be 10 bar. and the maximum extractable work was 0.52 W. They also proved that the extractable mechanical energy
is much higher when the vaporization takes place at temperature close to the superheat limit.

In Lin (1998) the authors analyzed the bubble formation in polysilicon line shape resistors (50µm×2µm×0.53 µm). In this work an electrothermal model is presented and found to represent well the behavior of the studied heaters. It was possible to control the size of the generated bubbles.

Deng et al. (2004, 2005, 2006) studied the heater size effect on micro boiling. They used a series of micron/submicron thin film platinum heaters of different sizes ranging from 0.5 µm to 70 µm. In these experiments the pulse duration was kept constant at 1.66 ms (quite long compared with the perviously mentioned experiments with pulse duration ∼ 5 µs). They observed different patterns in the bubble shape: spherical for heater sizes less than 10 µm, and oblate for heater sizes larger than 10 µm. The higher nucleation temperature was 245 °C corresponding to the smaller heater. The reason for the oblate shape was attributed to the condensation effect of the vapor outside the superheated zone in water. It is not very clear why the nucleation temperatures measured by this group are so low but probably the heating rate are much lower than in the previously cited ones.

The effect of ambient pressure has been assessed by Pavlov and Skripov (1970) and later by Derewnicki (1985). The later presented experimental results of a heated platinum wire immersed in a pressurized chamber full of water. Iida et al. (1997) studied the effect of ambient pressure on pool boiling of ethyl alcohol heated by a planar platinum heater. Their experiments were performed inside a pressurized chamber with a transparent window facing the objective of the microscope.

All the systems mentioned in this section refer to either an open system or to a pressurized macroscopic chamber. A different approach was taken by Carey (2000). In his investigation, he focuses on a liquid enclosed in a cavity with a flexible wall as in a typical microvalve. He uses the Redlich-Kwong equation of state and the principles of thermodynamics to calculate the spinoidal and saturation points of the fluorinert liquids: FC 72 and FC 75. His analysis predicts that a confinement of the liquid being heated increases the boiling temperature and it may inhibit spontaneous boiling.
1.4 Outline of the project and goals

This thesis work is divided in two main parts. First we studied explosive vaporization in a liquid enclosed in a small cavity made of a transparent material with 50$\mu$m×50$\mu$m heaters. In the second part we used a much smaller heater (3 $\mu$m×0.5 $\mu$m), to study how a reduction in the heater size could affect the explosive vaporization phenomena including the nucleation temperature.

The heaters used in the first part of the work were square 50 $\mu$m×50 $\mu$m constructed of sputtered platinum on a glass substrate. The achieved rate of temperature rise was on the order of $10^7Ks^{-1}$. The average temperature on the heater was measured and the boiling phenomena inside the microcavity was visualized using a pulsed flash lamp with a temporal resolution of 100 ns.

In the second part, a method based on Ion Beam Milling was applied to fabricate a thin film platinum heater of 0.5 $\mu$m × 3$\mu$m size. The temperature evolution of the heater subjected to pulsed heating was measured and images of the heater were acquired using flash lamp illumination that confirmed the formation of vapor on the heater.

The thesis is structured in the following way: First, the thermodynamics aspects of explosive boiling will be introduced. Next, the set-up for the investigation of explosive boiling in microenclosures will be presented followed by a chapter explaining the fabrication of the device. The chapter covering the results is divided into temperature measurements and visualization. Next, the experiments performed with the submicron heater will be presented. Finally, the conclusions are drawn followed by an outlook.
Introduction
Chapter 2

Thermodynamic aspects of explosive vaporization

2.1 Thermodynamic limit of superheat

In classical thermodynamics, phase transitions are assumed to occur at the equilibrium saturation conditions. However, real phase transformations usually occur under nonequilibrium conditions. For example in real vaporization processes, at least some liquid in the system is superheated (heated above the saturation temperature). Figure 2.1 shows the saturation curve in a pressure versus specific volume diagram. We will refer to the states represented by the area under the saturation curve as the saturation dome. This region represents the conditions \((T,P)\) in which the liquid and the vapor phases coexist.

The analysis of the stability of a closed system containing a pure substance which is not in thermodynamic equilibrium can be used to determine the conditions under which a portion of such a system is likely to undergo a change of phase. This condition is called the criterion for mechanical stability (Carey, 1992):

\[
\left( \frac{\partial P}{\partial V} \right)_T < 0
\]

This condition is necessary and sufficient for the stability of a phase. Eq. 2.1 combined with the equation of state can be used to define the limit of intrinsic stability, also called
the spinodal limit. For a liquid heated at constant pressure above its equilibrium boiling temperature the spinodal limit is a maximum upper limit on the superheat that results from thermodynamic considerations. It is sometimes called the thermodynamic limit of superheat. The spinodal curve is the locus of spinodal limit points in the vapor dome (see Fig. 2.2).
A liquid at atmospheric pressure being heated (constant pressure) can in principle enter into the metastable region without experiencing phase change. Theoretically the liquid can be superheated till the spinodal temperature corresponding to its pressure. The spinodal represents the limit at which the phase change is certain to occur. It is not easy to establish the spinodal limit since the extrapolation of the existing equations of state in the metastable region is dubious. Avedisian [Avedisian, 1985] obtained a spinodal temperature for water at atmospheric pressure of 323°C using the Peng-Robinson equation of state.

The pressure corresponding to the stable vapor phase (vapor pressure) increases with the temperature. The higher the temperature at which vaporization occurs the larger the pressure difference between the metastable state and the final stable vapor state. This means that the higher the temperature at which the phase change takes place the more violent will be the phenomenon [Carey, 1992; Glod, 2001].

Thermodynamic stability criterions can be used to establish the absolute maximum temperature beyond which the phase change is certain to occur. However, the actual achievable temperature depends on the probability that a certain perturbation will initiate a phase transition, this depending on the molecular kinetics of the process. The next section will elucidate the kinetic factors that influence the phase change process.

2.2 Nucleation mechanism in explosive vaporization

Before introducing the kinetic factors that influence the phase change process we will differentiate between homogenous nucleation and heterogenous nucleation. Heterogenous nucleation is defined as the generation of vapor at an interface between the liquid and another phase (solid, impurity, dissolved gas, etc). In homogenous nucleation the generation of vapor occurs completely within a superheated liquid. Internal fluctuations take place always in a liquid system (even in thermodynamically stable system). These fluctuations, or localized transient deviations from the normal state, include fluctuations of the local molecular density in the liquid. While for subcooled liquids such fluctuations are likely to be within the limits of the liquid phase existence, for saturated or superheated liquids,
the density fluctuations may exceed this limits resulting in localized regions where the molecular density is that of saturated vapor.

In general the energy barrier that needs to be surpassed to initiate the phase change is higher for homogenous nucleation than for heterogenous nucleation. Because of that, homogenous nucleation is associated with higher superheats than heterogenous nucleation and the second is more commonly observed in real phase change phenomena.

It has been observed that liquids where homogenous nucleation takes place vaporize explosively while vaporization originated by heterogenous nucleation is less intense. However it is important to note that the intensity of the phase change phenomena depends on the superheat and not on the nucleation mechanism. Heterogenous nucleation can also produce vapor in a violent manner when occurring at high superheat. Homogenously nucleated liquids usually vaporize explosively because homogenous nucleation occurs at very high superheat.

2.3 The kinetic limit of superheat

The density fluctuations causing homogenous nucleation (heterophase fluctuations), give rise to small embryo of vapor within the liquid. As the liquid superheat increases, the energy of the molecules in the liquid also increase and thus regions of low density have higher probability of being generated (because of mutual repulsion of the molecules at higher energy). A small fraction of these unstable and hence short-lived embryos of vapor may grow to a critical size $r_c$, beyond which further growth becomes thermodynamically favorable.

The critical radius can be obtained knowing that the chemical potential of the liquid and the vapor at equilibrium are equal, $\mu_l = \mu_{ve}$, and using the Young-Laplace equation, $P_{ve} = P_l + \frac{2\sigma}{r_c}$. The Gibbs-Duhem equation ($d\mu = -sdT + v dP$) can be integrated to obtain the pressure inside the embryo $P_{ve}$ at equilibrium with the liquid at temperature $T_l$. Substituting the $P_{ve}$ into the Young-Laplace equation the critical radius $r_c$ can be
2.3 The kinetic limit of superheat

obtained:

\[ r_c = \frac{2\sigma}{P_{sat}(T_l)\exp\{\eta[P_l - P_{sat}(T_l)]/RT_l\} - P_l} \]  

(2.2)

Some concepts of statistical thermodynamics are required to establish the frequency at which radius of the critical size form in a superheated liquid. The parameter used to express this probability is the nucleation rate \( J \), or rate of nucleus formation per unit surface.

\[ J = N_l \left( \frac{3\sigma}{\pi m} \right) \exp \left\{ \frac{-16\pi\sigma^3}{3k_B T_l[\eta P_{sat}(T_l) - P_l]^2} \right\} \]  

(2.3)

where, \( N_l \) is the number of liquid molecules, \( m \) the mass of one molecule of the liquid, \( K_B \) the Boltzmann constant and

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

(2.4)

A numerical value for \( J \) allows to determine the nucleation temperature or the kinetic limit of superheat using Eq. 2.3. It is not easy to obtain a value of \( J \) but due to the exponential term in Eq. 2.3 there exists only a narrow range of temperatures below which homogenous nucleation does not occur and above which it occurs almost immediately.

The kinetic limit of superheat has been investigated by many authors, (Skripov, 1992; Eberhart, 1976; Blander and Katz, 1975; Blander, 1979). Recent theories question the validity of using macroscopic values of the surface tension \( \sigma \) when describing the microscopic nucleus (Kwak, 2004).
Chapter 3

Experimental set-up

The system to generate microbubbles is the core of the experimental set-up used for this thesis. It consists of a DC pulse generator and the microfabricated thin-film heater. The chip connector is permanently fixed onto a holder. This holder provides support for the BNC-female connection between the chip and the instrument.

The measurement of the heater temperature during bubble formation was one of the main goals of this work. We measured the average temperature of the heater using the heater itself as a temperature sensor. To measure the temperature the change in resistance induced by the change in temperature was monitored. This was made by acquiring the voltage drop and the current supplied to the device. A Matlab program calculated the average temperature from the two acquired signals and the temperature coefficient of resistance of the heater.

To measure the temperature using the heater as a sensor element we need to know its temperature coefficient of resistance, ($\alpha$). The thermal coefficient of resistance was measured during the calibration experiments. The calibration consisted in measuring the resistance of the heater at different temperatures. The value obtained during calibration differs from the known $\alpha$ value for bulk of platinum which justifies the need for calibration. Originally the calibration was done in a standard oven with an opening for the electrical connections. Since some of the components used for electrical connection with the chip do not withstand high temperature, the calibration was done only up to 110 °C. Therefore a dedicated oven was designed and built allowing calibrations at higher temperature and
in an inert atmosphere to reduce the risk of oxidation. This oven will be described later in the chapter dedicated to characterization of the submicron heater. In this chapter the procedure to obtain $\alpha$ using the standard oven will explain.

The visualization of the bubble formation was possible thanks to a pulsed image acquisition system synchronized with the bubble generation system. Digital image processing technique allowed a quantitative comparison of the bubble size.

This chapter describes the details of the experimental set-up. The first section is dedicated to the bubble generation set-up, the second section introduces the instruments used to acquire and to record the voltage drop in the heater and the current supplied. Next, the system for calibration is introduced. Finally, the image acquisition system is described.

### 3.1 Bubble generation set-up

For the bubble generation a current pulse was supplied to the thin film heater. The chip with the heater is plugged into a standard connector (Flexprint, Alphenol, Compona AG, Switzerland). The connector is permanently fixed onto a holder supporting the BNC-female connection between the chip and the instrument. Soldered cables join the corresponding poles of the flexprint connector to the BNC connector. The pulse generator output is connected with a coaxial cable to the BNC-female on the chip holder.

The thin film heater is heated by a current pulse provided by a pulse generator HP 8114A, (Hewlett Packard AG, Switzerland); Table 3.1 summarizes the main features of the pulse generator. A very fast settling time is necessary to characterize the fast thermal transient we want to study. For this pulse generator, and the amplitudes used, the settling time is 100 ns only representing a small part of the transient we would like to study.

In some experiments it was convenient to externally trigger the pulse generator. This was done by connecting one of the channels of a digital delay and pulse generator (Model DG535, Stanford Research Systems, Roper Scientific, Munich, Germany) to the trigger input connector of the pulse generator.
3.2 Fast Resistive Thermometry

The resistive temperature measurement was performed using the heater itself as a temperature sensor. Knowing the induced resistance change we measured the average temperature change by applying the formula:

\[ T(t) = T_o + \frac{R(t) - R_o}{R_o \cdot \alpha} \]  

(3.1)

where \( R_o \) is the nominal resistance measured at the reference temperature, \( T_o \), and \( \alpha \) is the temperature coefficient of resistance. This method is known as resistive thermometry.

To obtain the temperature of the heater we needed to measure the resistance of the
Experimental set-up

Figure 3.2: Set-up for microbubble generation.

The resistance ($R(t)$) was measured using the four-point method. This method uses four electrodes to measure resistance, two electrodes to supply the current and the other two to measure the voltage drop. The four-points method eliminates the measurement errors due to the cable resistance, thin film electrode resistance, and the contact resistance between the connector and the thin film electrode. Since high impedance voltmeters draw little current, the voltage drop across the two voltage sensing electrodes, due to electrode, cable and contact resistances, is very small compared to the voltage drop generated on the heater. The instantaneous electrical resistance of the heater $R(t)$, can be obtained by Ohm’s law as:

$$R(t) = \frac{V(t)}{I(t)} \quad (3.2)$$

We calculated the average temperature increase of the heater by measuring the voltage drop along the heater together with the current inducing the voltage drop.

The voltage drop over the heater was monitored by a passive differential probe pair, (DXC200, LeCroy, Switzerland) which was connected to a differential amplifier (DA1822A, LeCroy, Switzerland) and monitored using a storage oscilloscope (LC334, LeCroy, Switzerland). The current supplied to the heater through the wider electrical connections was also monitored using a current probe (APO15, LeCroy, Switzerland). The current probe
is based on the Hall effect and therefore did not require the breaking of the circuit. The signals were stored in a PC using an oscilloscope interface software (ScopeXplorer 2.2, LeCroy, Switzerland) via GPIB communication. The traces from the oscilloscope were read with a Matlab program and fed into Eq. 3.1 and 3.2 to calculate and to plot temperature evolution.

In order to obtain the temperature of the heater using eq. 3.1, it is necessary to know the value of the thermal coefficient of resistance, \( \alpha \) for the fabricated heaters. Despite the heater being made of platinum (a well characterized material), in general the measurement of the \( \alpha \) value for the thin film is necessary. The procedure used to measured it will be described in the next section and it is what will be refereed to as the ‘calibration’.

### 3.3 Set-up for Heater Calibration

The heater was calibrated measuring the resistance of a given specimen at different ambient temperatures. A test device specially designed for calibration purposes and fabricated on the same wafer as the heaters was placed inside the oven. The test device was a platinum film of 100 \( \mu m \) width and 10 mm length. It is represented in Fig. 3.3. The layout shows the four contact pads used for the four-point resistance measurement. The thickness and quality of the platinum layer for the heater and for the specimen prepared for the calibration was the same since all were fabricated on the same wafer. The resistance of the calibration device is then 100 times higher than the resistance of a square heater. The higher resistance value of this device compared to the heater allows using a lower sensing current and thus a more accurate resistance measurement.

### Sheet resistance

The sheet resistance is typically used to characterize thin film conductors. The sheet resistance of a layer with electrical resistivity, \( \rho \), and thickness, \( t \), is given by their ratio:

\[
R_\square = \frac{\rho}{t} \quad (3.3)
\]
It is commonly given in Ω/□. The resistance of a rectangular piece of material with length L and width W equals the product of the sheet resistance and the number of squares, where the number of squares equals the length divided by the width:

\[ R = R_\square \frac{L}{W} \]  

(3.4)

The calibration device was also used to calculate the sheet resistance of the deposited platinum film.

![Device fabricated for calibration. The resistance is 100 times the Platinum thin film sheet resistance](image)

**Figure 3.3**: Device fabricated for calibration. The resistance is 100 times the Platinum thin film sheet resistance

**Calibration using commercial oven**

To perform the calibration the device was heated up and its resistance was measured at different ambient temperatures. The chip device was placed inside a metallic box (see Fig.3.4). The function of this box is to maintain a stable temperature in the vicinity of the device. The box is a stainless steel block with a recess to allocate the calibration device and the connector (see Fig.3.4). The connector was made by gluing four spring contacts to a piece of glass. This glass is held in its position with two small magnetic elements glued to the isolated part of the spring contacts. The magnetic elements fixed and pressed together the spring contacts against the contact pads in the chip, providing a good electric contact between the spring contact tip and the thin film. A cover is then screwed on the top of the box. A groove at one side of the box allows an outlet for the cables. At the same side of the box a hole below the recess is made for placing a thermometer. The box with the screwed cover is then placed in the oven (Model FB1300, Thermolyne, ISMATEC SA, Glattbrugg, Switzerland) for the calibration.
3.3 Set-up for Heater Calibration

The oven had an opening in the back wall to provide access for the precision thermometer (PTM3040, Tectron AG, Wald, Switzerland) and the cables. The electrical resistance of the test device was measured by a data acquisition unit (HP 3852A, Hewlett Packard AG, Switzerland) connected to a PC. The sensing current was in the range of a few mA. The induced heating produced by the sensing current for these measurements was below the uncertainty of the measurement. The uncertainty of the measurement is 0.1 °C (0.3Ω). The electrical resistance was initially 175±0.3 Ω. The resistance increase was measured for several temperatures up to a maximum allowable temperature of 110 °C. The temperature coefficient of resistance (α) measured for the fabricated devices is 2.0 \times 10^{-3} \text{ K}^{-1}. No drift due to thermal cycling was found when the device was previously annealed for 1 hour at 150°C in a vacuum oven and then 2 hours at 500 °C in normal oven (non vacuum).
3.4 Pulsed image acquisition system

The working principle of this imaging technique consists of illuminating the process to be observed with a pulsed light source of short duration. The light can be pulsed to illuminate a certain "moment" after triggering the process. To achieve this, two pulses with a certain delay between them are needed. The first one triggers the pulse generator that starts the heating pulse and the second one triggers the flash lamp. The camera and the frame grabber are triggered at the same moment than the pulse generator.

The phenomena under investigation can be repeated by applying repeated pulses and by allowing enough time between the pulses so that the chip recovers to the initial conditions.

If the delay between the trigger signals is set exactly to the same value the sequence of images seen is always the same, getting the impression of a frozen process. If the delay is slightly increased then the sequence of images seen gives the impression of a slowly moving process.

The image acquisition system is composed of:

- Nanosecond flash light source and driver, (Nanolite KL-K, High Speed Photo-
Systeme, Germany). The lamp is mounted on a home-made lamp housing and fixed to the reflected-light port on the microscope.

- A CCD camera (JAI M10, Stemmer Imaging GmbH, Germany)
- IC-PCI Frame Grabber, (Stemmer Imaging GmbH, Germany)
- A computer with image acquisition software (Optimas v6.2, Stemmer Imaging GmbH, Germany)
- Digital delay and pulse generator (DG535, Stanford Research Systems, Roper Scientific, Munich, Germany)
- PC with the software for data acquisition (LabView ver.8, National Instruments) and GPIB card to control the acquisition of series of images.
Experimental set-up

The digital delay and pulse generator (DG 535) triggers the HP pulse generator and the image acquisition system simultaneously and with a certain delay the flash lamp. The DG 535 is controlled via GPIB by a Labview program. This program can record series of images. A sketch of the visualization set-up is depicted in Fig. 3.6. The dashed connections from the DG 535 to the pulse generator, flash lamp, camera and frame grabber represent the signals to synchronize the heating process and the image acquisition. Since the flash lamp has a jittering of 0.2 \( \mu s \) there is a limitation in the temporal resolution of our system. Still this system allowed the acquisition of series of excellent images showing the bubble growth and collapse.

### Nanolite Lamp KL-L main features:

<table>
<thead>
<tr>
<th>Feature</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rise Time 10 % - 90 %</td>
<td>2.1 ± 0.2 ns</td>
</tr>
<tr>
<td>Decay Time 90 ±- 1/e</td>
<td>11.3 ± 0.5 ns</td>
</tr>
<tr>
<td>Flash duration (nominal)</td>
<td>18 ns</td>
</tr>
<tr>
<td>Energy per flash</td>
<td>25 mJ</td>
</tr>
<tr>
<td>Max. Frequency</td>
<td>10kHz</td>
</tr>
</tbody>
</table>

Table 3.2: Characteristics of the flash lamp

### Nanolite Driver main features:

<table>
<thead>
<tr>
<th>Feature</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flash time lag</td>
<td>&lt; 6 ( \mu s )</td>
</tr>
<tr>
<td>Radiation time</td>
<td>typ. 35 ns, max 60 ns</td>
</tr>
<tr>
<td>External trigger</td>
<td>TTL (&gt;5 ( \mu s ) &gt;3 V)</td>
</tr>
</tbody>
</table>

Table 3.3: Characteristics of the Nanolite driver
Chapter 4

Microfabrication

To investigate explosive boiling taking place inside a microcavity a specially designed chip was fabricated. The chip needed to fulfill the following requirements:

1. Include an integrated microheater that can withstand high thermal and mechanical stresses.

2. The microheater should be in contact with a liquid and the liquid should be confined within a microcavity.

3. At least one of the walls of the microcavity should allow optical access to the liquid.

4. The microheater itself should be used as a temperature sensor measure the temperature of the liquid in contact with it.

The heater is a small electrical resistor made of either a conductive or a semiconductive material. The fluid, in contact with the heater, is heated and, if the temperature of the liquid is high enough, vaporization is expected to occur. The maximum temperature at which the liquid may exist is of special interest for practical but also for academic purposes. The heating process is concentrated in a small area that changes its temperature very rapidly. To measure the temperature evolution during the transient, it is necessary to use an integrated temperature sensor. In most cases, dealing with the investigation of explosive boiling, the heater itself is used as the temperature sensor. The visualization of
the vapor formation is also a goal of this investigation. Therefore, at least one side of the microcavity should allow optical access.

The microdevice was fabricated using the ETH microfabrication facilities. There are many technologies available nowadays to ‘build’ a microdevice as the previously described. Some of these technologies will be described in this chapter together with the detailed description of the fabrication of the microdevice used in the present work.

The chapter follows the order of the fabrication flow (see Fig. 4.2): Heater fabrication (1), microcavity fabrication (2), heater and microcavity bonding (3), fluidic interconnection (4) and electrical packaging (5). In each of these steps, an overview of some of the available fabrication techniques preceeds the details of the fabrication process.

### 4.1 Integrated Heaters in MEMS

When a current is applied to an electrical resistance heat is dissipated due to the Joule effect. This heat induces an increase of the temperature of the heater at a rate that depends on the current intensity, the electrical resistivity of the material, the size of the heater and the thermal properties of the heater and the substrate. The microheater dissipates heat to the surrounding media including the substrate. Although any integrated
A circuit dissipates heat due to the Joule effect we will refer to a resistor designed to produce a localized increase of temperature as a heater. Integrated heaters in microdevices assume a wide variety of functionalities:

- Measure flow velocity in fluidic channels, as in the device presented by Lammerink et al. (1993).
II. PDMS Micromolding

1. Mold preparation
   1.a) SU-8
   1.b) Mask 3
   1.c) PDMS liquid

2. Micromolding

3. Release

4. Cut and drill holes

III. Aligning and bonding

- Elevate the temperature of a contained medium whose expansion will deform the container for hydrostatic actuation for MEMS as in the device presented by Mutzenich et al. (2004)
- Produce phase change to generate thrust as in the microthruster investigated for
satellite applications. (Chu et al. (2006); Maurya et al. (2005); Rossi et al. (2006)).

- Deposit small droplets onto a substrate on demand.

The last application for drop on-demand deposition, can be found in about 70% of current ink jet printers. The technology was named Thermal Ink Jet (TIJ) by Hewlett -Packard engineers. In this technology a microheater is in contact with the ink which is ejected through a nozzle. Several microheaters are typically arranged in a matrix. When one of the microheaters is actuated, the ink in contact with the heater vaporizes and ejects a droplet of liquid though the nozzle opposed to the heater.

As these examples show, there are many different applications of MEMS utilizing microheaters. In these cases, the function of the microheater is, in principle, the same: producing an increase of temperature. However, depending on the application, it is subject to very different requirements. When the microheater is used to induce a superheated state in a liquid, leading to explosive boiling, it needs to endure high thermal stresses. Moreover, it should not deteriorate or melt under operation. The bubble formation and collapse also generate cavitation-like stresses that may deteriorate the surface affecting the performance of the device.

In this section we will explore briefly the different possibilities that exist in order to fabricate heaters integrated in MEMS device. Special attention will be given to heaters used to generate explosive boiling.

**Heater Fabrication**

A microheater is, in general, composed of a patterned conductive material deposited on a substrate. Most designs include an insulation or protective layers covering the conductive structure. As an example, the heaters manufactured by Hewlett-Packard Corporation are square elements 64.5 $\mu m$ wide. First, a layer of 1.6 $\mu m$ thick Silicon dioxide ($\text{SiO}_2$) is deposited on the Silicon substrate to reduce rapid heat loss into the substrate. Next, the heater material, consisting of a 0.2 $\mu m$ layer of a tantalum/aluminum alloy, is deposited on the $\text{SiO}_2$ layer. 0.5 $\mu m$ of gold is deposited and patterned on top of it to provide electrical contacts. A 0.5 $\mu m$ thick tantalum layer is deposited on the top to protect the
heater from the bubble collapse impact. The approximate resistance value of the element is 15 Ω.

Avedisian et al. (1999) used a heater similar to the Hewlett-Packard commercial heater without the protective layer to measure the bubble nucleation temperature. Although these heaters withstand operation at high frequencies, they exhibit only a weak dependence of the resistivity on the temperature. This means that very sophisticated amplification circuitry was needed to measure the boiling temperature.

Also non-metallic materials have been used as heaters, polysilicon has been used as heater material in the blinking bubble pump presented in Yin and Prosperetti (2005) and in the research on bubble nucleation by Lin’s team, Lin (1998) and by Hong’s team, Hong et al. (2004). Polysilicon is an isotropic form of silicon. It can be deposited on the top of a wafer substrate and its electrical properties can be tuned by doping it with the suitable ions to increase its conductivity. It exhibits a rather linear relationship between its resistance and temperature which makes it ideal for temperature sensing. The fabrication in this case starts with the growth of a SiO$_2$ layer of about 0.5 µm to 1.5 µm. This layer serves as a thermal barrier as well as an electric insulation. A layer of Low Pressure Chemical Vapor Deposited (LPCVD) polysilicon doped with phosphorous 0.36 µm thick is deposited and patterned by plasma dry etching. Additionally, a protective layer of silicon nitride is then deposited to prevent mechanical damage. Some of the latest published investigations on explosive vaporization use heaters made of polysilicon, Okuyama et al. (2004) Lin (1998).

Highly oriented Chemical Vapor Deposited (CVD) diamond films on a silicon substrate have also been used as thermal actuators to eject liquid. Diamond films seem to have a great potential in MEMS applications and particularly as thermal actuators: diamond films have high thermal conductivities, low heat capacities, low thermal expansion coefficients and high mechanical and thermal stabilities. This makes them a good candidate to be used to fabricate microheaters which can operate under highly dynamic, thermal stresses, Gluche et al. (1998) Kohn et al. (1999).

Another material widely used in microheaters is platinum; it was also applied to investigate explosive vaporization. Iida and Okuyama used a platinum heater without a protective layer, Iida et al. (1994). Platinum is well suited as a resistive thermometer,
making it the favored material for commercial temperature sensors, \cite{Kim2001}. Platinum has good thermal response (linearly increase of resistivity with temperature). It has mechanical strength and stability meaning that it does not require an extra protective layer. Therefore we opted for using platinum for the thin-film heaters on a Pyrex glass or insulated silicon substrate. The next part of the section gives the details of the fabrication process.

**Mask production**

Photolithography is the most widely used technique to transfer copies of a master pattern onto thin films on a silicon wafer \cite{Madou2002}. The process consists of several steps: First a wafer is coated with a photoresist layer, (a photoresist is a light-sensitive material). Next, the wafer is exposed to light through a mask containing opaque/transparent areas so that only some areas are exposed to light. Finally the wafer is rinsed in a developing solution which removes the unexposed areas of photoresist leaving parts of bare substrate and photoresist-coated substrate. We used photolithography to transfer the pattern to the metallic film using a process known as lift-off which will be explained later.

The first step in the microfabrication process was to prepare the mask used in the photolithography. It required considering the steps in the fabrication process. In the process flow, represented in Fig. 4.2 all the steps of the process have been summarized. The fabrication of the heater needed two photolithography steps, the first mask (mask 1, in 4.2 (b)) defined the shapes of the platinum structures, and the second mask, (mask 2 in 4.2 (h)) was used to open the silicon nitride layer over the contacts. For the preparation of the fluidic part of the chip an third mask was used, (mask 3 in 4.3 (b)).

For the preparation of the masks we used a software for mask production (CLEWIN). This software was used for designing the different layers corresponding to the different fabrication steps and for generating a file with a format that is compatible with the mask producer. There are different technologies for mask production and they affect strongly the quality of the photolithography, especially the minimum features size. For the two mask processes of the heater fabrication, we used a chromium mask (DeltaMask V.O.F., Enschede, The Netherlands) to achieve a feature size of 4 /\mu m. For the third mask the
minimum feature size was not critical and therefore it was fabricated using transparent foil.

**Platinum sputtering and lift-off**

‘Lift-off’ is a method for patterning metallic films. It is generally used to pattern films which are not easily patterned directly by etching such as platinum films. The method is sketched in Fig. 4.4. First, the substrate is coated with a layer of photoresist, 4.4(a). Next, the photoresist is patterned using photolithography, (Fig. 4.4 (b)). Then, the film, usually a metal, is blanket-deposited over the entire substrate, covering both the bare and the photoresist-coated substrate (Fig. 4.4 (c)). During the actual lifting-off, the photoresist, under the metallic film, is removed with solvent, taking the film with it, and leaving only the film which was deposited directly on the substrate, (Fig. 4.4(d)).

![Figure 4.4: Lift-off process](image)

For this work two different substrates were selected: a) a 100 mm, double side polished, 300 µm thick silicon wafer with a 1.5 µm thick layer of thermally grown silicon dioxide, and b) 100 mm, 500 µm thick Pyrex glass wafer. Before starting the process, the wafers were cleaned in oxygen plasma for 10 minutes at 300W (TecPla 200 Plasma System).

The fabrication started by dehydrating the wafers for 5 minutes on a hotplate at 170 °C prior to spin-coating 5 ml of Lift Off Resist (LOR, Microchem, Germany) at 3000 r.p.m. for 40 sec. The LOR layer was soft baked for 5 minutes on a 170 °C hot
plate. This LOR is insensitive to light and thus is removed isotropically, generating a reentrant sidewall profile that favors the lift-off. Then, the positive resist Microposit 1828 (Shipley, USA) was spun at 4000 r.p.m. for 40 seconds and soft baked at 115 °C for 2 minutes. The photoresist layer is then ready to be exposed. Exposure time was set to 11 seconds. The exposed regions were dissolved dipping the wafer into Microposit Developer concentrate mixed with water (1:1) for 4 minutes and 30 seconds with agitation. Any possible photoresist residues were removed in oxygen plasma for 1 minute at 200 W. The resulting patterned photoresist coating is depicted in Fig. 4.5(a), showing a region of the wafer centered on one of the heaters.

![Coating](image1.png) ![Substrate](image2.png)

(a) (b)

Figure 4.5: (a) Photo of photoresist structure after photolithography and development. The area in the center is clear of photoresist and is where the platinum will remain. (b) Photo of the platinum structure (white regions) onto the glass substrate (black in the picture)

The 100 nm thick platinum layer, forming the heaters, was then deposited by sputtering. Sputtering is a process in which chemically inert atoms, in this case argon, are ionized into a plasma. The ions are accelerated onto a target by an electric field so that atoms from the target are knocked out (sputtering process) and reach the substrate. Sputtering takes place in a low-pressure gas environment. An adhesion layer of 5 nm TiW was deposited first, followed by a 100-nm platinum layer. During deposition the substrate can become hot due to the impact of the atoms of the deposited material onto the substrate. To reduce the risk of melting the photoresist, the platinum is deposited in two 15-second stages and allowing 20 minutes between them for cooling down the substrate.
Figure 4.6: Photograph of the microfabricated heater. The heater (100×100 µm²) is in the middle. Two connections for voltage-drop measurements (voltage taps) are located on the top left corner and bottom right corner of the heater. Large electric contacts to supply the heating pulse extend out of the image.

The platinum layer was subsequently patterned by removing the underlying photoresist (lift-off). The wafer was submerged in undiluted remover 1165 (Microchem, Germany) at 60°C for several hours with agitation. The platinum structure after the lift-off step can be seen in Fig. 4.5 (b).

**Silicon nitride deposition**

In Chemical Vapor Deposition (CVD), the constituents of a vapor phase react at a hot surface to deposit a solid film. Since several activation barriers need to be surmounted to arrive at this end product, some energy source is required (Madou 2002). In the Plasma Enhanced Chemical Vapor Deposition, PECVD a Radio Frequency (RF) induced plasma,
transfers energy into the reactant gases allowing the substrate to remain at moderate temperatures.

We used PECVD to deposit a 200-nm-thick protective layer of silicon nitride on top of the platinum structures. This layer serves as an electrical insulator. Therefore we had to open some metallic areas to establish an electrical contact with the microheater. This was done by dry etching the nitride layer after a second photolithography step.

**Dry etching**

The wafer with the silicon nitride layer on the top was electrically insulated. We had to open selected regions to establish electrical contact with the microheater. A layer of photoresist (Microposit 1828, Microchem, Germany) was deposited and patterned on the silicon nitride to protect the regions where the nitride layer should remain; the photoresist was spined onto the wafer at 4000 r.p.m. for 40 seconds and soft baked for 2 minutes at 115°C. The second mask was then aligned to the wafer with the structured platinum and exposed for 40 seconds. Microposit Developer was used to dissolve the exposed photoresist. The wafer was then ready for etching the holes in the areas free of photoresist. The 200 nm thick layer of silicon nitride was subsequently etched away using Reactive Ion Etching, (RIE). This step opened the electric contact pads. The remaining photoresist was dissolved.

The last step was to cut the chips out of the wafer, which was done using a wafer saw.

Fig. 4.6 is a microphotograph of the heater device: The heater (100×100 µm²) is in the middle of the image. The large electric contacts to supply the heating pulse extend out of the image. Two connections for voltage-drop measurement (voltage taps) are located in the top left and bottom right corner of the heater. This first version of the chip included an independent temperature sensor (meander shape) and two opened electrodes to sense conductivity thought the liquid. The idea of the secondary temperature sensor was to detect the initial temperature of the substrate and the liquid. The conductive electrodes were included to detect the formation of vapor inside the chamber. It was found however that the read-out circuit needed to obtain useful measurement out of these sensor were too complicated to be covered in the present work.
4.2 Fabrication of the microfluidic chip

Review of available methods to fabricate microfluidic devices

The standard approach to the fabrication of microfluidic devices consists of etching the channels into a substrate, usually silicon, quartz or glass. The substrate is first coated with a material resistant to the etching agent and then the coating is patterned with the geometry of the channels. Then an etching process can be used to remove the bare regions of the substrate. There are many etching methods, for example, when using silicon we can chose between wet etching and dry etching. Wet etching using KOH as etching agent produces anisotropic profiles along crystallographic planes. Alternatively dry etching can be applied allowing vertical walls as in Reactive Ion Etching (RIE), in RIE-Inductively Couples Plasma (RIE-ICP) or Deep Reactive Ion Etching (DRIE) systems. For glass substrates the etching agent usually is hydrofluoric acid. The channels obtained by this method have rounded walls. The top of this microfluidic structures is sealed by bonding a wafer on the top of the substrate containing the channels. Anodic bonding and direct silicon bonding are two frequent bonding methods used to seal microfluidic channels made of silicon and glass substrates. Pure glass or quartz devices are typically sealed by fusing bonding.

A slightly different approach consists of coating the substrate with a photopatternable structural material and then defining the fluidic channels by photolithography. Then, instead of etching the channels on the substrate, the top sealing wafer is bonded onto the patterned coating containing the fluidic channels. The fluidic structures (channels and cavities) are made in an intermediate layer between two bonded wafers. The intermediate layer is used both as structural material and as bonding agent. Many devices were fabricated over the last years using this approach; [Blanco et al. (2004); Agirregabiria et al. (2005)] used SU8 as an intermediate layer. They obtained vertical walls up to 60 µm high. Similarly, fluidic channels can be designed on both wafers to form more complex fluidic structures. A complex structure using several patterned SU8 layers can be generated by depositing a layer of Kapton (a polyimide film) onto the substrate and underneath the photoresist. The substrate can then be released after bonding, [Agirregabiria et al. (2005)] produced complex 3D fluidic structures by several subsequent bond and release steps.
With this second approach, the integration of a microheater with the fluidic channel is simpler than with the first approach. The reason is that the intermediate layer will cover any irregularity on the substrate, including for example the thin metallic microheater patterned on the top of it. By contrast the bonding methods used to seal channels etched onto silicon and glass (anodic bonding or fusion bonding) are effective only between completely flat surfaces.

Fabrication of microfluidic devices on PDMS

A different approach consists in making the fluidic channels in a layer of poly(dimethylsiloxane), (PDMS), bonded onto a substrate. This method was proposed by Xia and Whitesides (1998) and consist of several steps: mold fabrication, PDMS casting, peel off and bonding. The mold is usually made by either patterning a structurally hard photoresist, or etching a substrate. The mold consists of the negative pattern of the fluidic channels. The PDMS is then casted onto the silicon surface where it assumes a shape which is the inverse of the mold. After curing, the PDMS layer can be removed from the silicon and then sealed to a smooth glass or silicon substrate. The master, involving expensive microfabrication processes, needs to be made only once and the low cost casting process can be repeated several times obtaining exactly the same fluidic structure making it very appealing for mass production and prototyping in research, (Ng et al., 2002; Duffy et al., 1998). It was used for forming membranes in chips with integrated pumps, valves and channels. Many applications of this fabrication method arise in biochemical analysis such as the device proposed in Su and Lin (2004).

For the device used in this investigation we opted for fabricating the channels by micromolding of PDMS. PDMS is transparent, facilitating optical access to the microchannels and it allows to simplify the bonding process and the fluidic interconnection. We will describe now the details of the fabrication of the fluidic part of the chip: first we describe the fabrication of the mold, later the micromolding itself.
Mask fabrication and fabrication of the mold

Mask fabrication

The fabrication of the microcavity and the channels involved only one mask that was used to pattern a layer of SU8 by photolithographic technique. The structured SU8 layer was used as mold for PDMS micromolding. High-quality prints on foil were used as the mask with a resolution of 12700 dpi (Selba S.A., Versoix, Switzerland). The printed foil was cut and taped onto a 2.5 mm thick 5 inch side square glass support.

Micromold preparation

The mold with the micro cavity and channels was made by patterning a layer of a photoresist material (SU8-50, Microchem, USA) onto a polished silicon wafer. The silicon wafer was cleaned first and dehydrated on a hotplate at 200 °C for 30 min. Then the wafer was coated with SU8-50 using a 2-stages- coating process: 1. 400 rpm with 200 rpm/sec, for 15 seconds and 2. 2000 rpm with 300 rpm/s for 35 seconds). After waiting till the photoresist layer leveled (15 min) to reduce edge beads, the photoresist was soft baked. The goal of the soft baking step was to evaporate the solvent and to densify the film. Then, the photoresist was exposed through a mask with the shape of the cavity and channels. After the exposure, the wafer was baked to cross-link the resin (post-exposure bake). The development was the final step in the process of fabrication of the mold. It revealed the negative shape of the microcavity and the channels.

Micromolding of PDMS

Once the mold was ready, we could proceed to the micromolding step. We used the commercial product Sylgard 186, (Suter-Kunststoffe AG, Switzerland). It consists of two parts: the prepolymer and the cross linking agent (the hardener). We first mixed well the PDMS reagents 1:10. The mixture was then degassed for 30 minutes.

The thickness of the PDMS layer could be controlled by weighing the PDMS poured onto the wafer. Thinner layers of PDMS could be obtained by spin coating the PDMS
4.3 Bonding

Sealing of PDMS channels is much simpler than sealing channels that are made in glass or silicon because high temperatures, pressures and high voltages are not required. PDMS
Figure 4.8: Photograph of the assembled chip. The heater (100×100 \( \mu m^2 \)) is in the middle. The bigger square marked with dashed line is the top view of the cavity (200 \( \mu m \) side square). The channels to fill the cavity with liquid are 50 \( \mu m \) wide and 7 millimeters long so that the inlet/outlet (out of the image) can be accessed when the device is under the microscope.

Channels can be sealed irreversibly to glass, silicon or silicon nitride by exposing both, the surface of PDMS and the surface of the substrate, to an air or oxygen plasma. Oxidization using a plasma produces silanol groups on PDMS and -OH-containing functional groups on the other materials; these polar groups form covalent -O-Si-O-bonds with oxidized PDMS when these surfaces are brought into contact.

The surfaces were first cleaned and then activated for 30 seconds in an oxygen plasma (TecPla 200 Plasma System) at 100 Watt. Without touching the surface to be bonded the heater was aligned to the centre of the cavity before putting them into contact. This was made on a test-probe station where the micromanipulators were replaced by a support made of polyacrylate where the PDMS could be fixed upside down during alignment.
4.4 Fluidic Interconnection

The heater was located on the XYZ-stage under the microscope and the PDMS cover was held upside down under the microscope and on the center of the field of view. The two parts are aligned by carefully positioning the XY-stage so that the heater was centered with respect to the cavity and then the Z-position was adjusted until both parts were in contact. Fig. 4.8 shows the final assembly.

4.4 Fluidic Interconnection

It was already mentioned briefly how a simple hole drilled with a needle tip through the PDMS chip allowed to fill in the cavities. However, when the channels are made in silicon, in glass or in and intermediate layer between two substrates, providing the necessary fluidic connection between the microchannel and an external fluidic system is a not trivial issue. A brief summary of different techniques to connect the microchannels with the outside fluidic system is summarized next.

The more straightforward method to connect microchannels to an external tube consists in building relatively large input and output regions into the microfluidic structure where capillary tubes can be adapted. The tubes can then be glued to the port. This method requires manual work and is difficult to be done at wafer level making it not suited for mass production. Tsai and Lin (2001) integrated a film of biaxially-oriented polyethylene terephthalate (boPET) polyester (Mylar film) between the inlet port and the capillary tube to seal the interconnection and they proposed a mass production version of the method in which the sealant layer is coated onto the inlet region. Other fluidic interconnections are based on micromachined inlet/outlet ports. Fluidic interconnection can be achieved by using micromachined silicon couplers to fit fussed silica capillaries. This couplers are microfabricated by wet etching and Deep Reactive Ion Etching (DRIE), (Meng et al., 2001). The same concept was also used to achieve in-plane connections (Gonzalez et al., 1998; Gray et al., 2002). Interlocking structures using notched cylinder/hole type interconnections were also proposed using DRIE of silicon or SU8 photopatterned structures (Gray et al., 2004). Other authors found solutions based on the use of a rubber o-ring to couple glass capillary tubes and silicon chips. The trenches to position the o-ring are micromachined by DRIE (Yao et al., 2000). Silicon/plastic couplers where used in
There is not a universal solution for fluidic interconnections suited for most microfluidic devices but many developments go in the direction of finding a sort of standard microfluidic connector (Morishima et al., 2004).

Fluidic interconnections in PDMS-based microfluidic devices is much simpler. Drilling holes through the PDMS-based fluidic device is relatively easy and can be made with a needle. Also, because of its flexibility it is possible to fit tubes into the drilled holes that are sealed without any gasket material (Saarela et al., 2006).

### 4.5 Electric packaging

We designed the chip to connect it to a standard connector with 26 poles, (Flexprint, Alphenol, Compona AG, Switzerland), see Fig. 4.9. Based on the information from the producer, we selected the electric layout: The contacts were placed on one side of the chip, with 1 mm distance between them and they were 0.7 mm wide and 3 mm long (see Fig. 4.10). Two poles were connected to each side of the wide conductors that converge on the much narrower microheater. Each one of the two voltage taps was connected to one pole. Four poles were dedicated to a four-point resistive measurement of an auxiliary meander-type resistance. One pole each was connected to the conductivity sensors. Twelve poles were needed in total for each heater device. Two microheaters were fitted in each chip. We could fabricate ten chips out of a 100 mm diameter wafer. The layout of the platinum thin film of one chip is shown in Fig. 4.10.

It was observed that this particular design was prone to generate strong temperature gradients during the pulse heating experiments. The reason was the asymmetric convergence of the electrodes supplying the heater. The electrical current passing through the conductor can be imagined as a ‘flow’ of electrons trying to pass through the narrow portion of the conductor. We simulated the electrical response of the actual layout with a Finite Element Analysis package (QuickField). The electrical current intensity obtained when a current of 1 A is applied, is shown in Fig. 4.11(a). The current intensity in the areas close to the 90° is considerable higher than in the rest of the heater. For comparison the second symmetric design is also simulated and the result can be seen in 4.11(b). Therefore in the second generation of heater devices we used a more symmetric design.
Figure 4.9: Assembled chip consisting of two test devices (right) and the standard connector used to establish the electrical connection (left). Holes to access the fluidic channels are punched onto the PDMS cover prior to bonding.

The second generation of chips were designed to fit an eight poles connector. The auxiliary meander shape temperature sensor was eliminated from the design. A photo of the connector and the second version of the chip can be seen in Fig. 4.13. The layout of the chip is also shown in Fig. 4.12.
Figure 4.10: Chip layout: the contacts are designed to fit the 26 contacts of a commercial flexprint connector. In the close up the two big connectors converge asymmetrically on the narrow region (the microheater). The microheater is 100 µm wide.

Figure 4.11: Current intensity distribution. Simulation neglecting thermal effects. Layout corresponding to the first version (a) of heaters and to the second version (b).
Figure 4.12: Chip Layout of the second generation of fabricated devices. The layout was modified to have a more symmetric electrical current distribution and the secondary temperature sensor was eliminated.

Figure 4.13: Assembled chip (second version) and a standard connector used to establish the electrical connection (left).
Chapter 5

Temperature measurements

The temperature traces measured during pulsed heating are described and discussed in the present chapter. In the first section, the result of the calibration will be shown and discussed and the resistance temperature measurement method will be illustrated with real current and voltage traces measured during a pulsed heating experiment. When the microheater is subjected to pulsed heating, its temperature increases at a rate that is related to the current amplitude of the applied pulse. If the heater is loaded with water and the electric pulse is sufficiently intense, the temperature evolution trace presented an inflection or kink towards an increase of slope. This kink in the curve coincides with the instant at which the microheater is entirely covered with vapor and thus it is associated with the temperature at which explosive vaporization takes place. The temperature at which the kink takes place will be called the nucleation temperature and the ratio between the corresponding increase of temperature and the time lag after heating onset will be called the rate of temperature rise.

The influence of current amplitude on the average nucleation temperature and on the rate of temperature rise will be analyzed. After that some results for the composed device (heater plus fluidic cover) will be shown first letting open the inlet/outlet ports and afterwards closing them.
5.1 Method

The resistance of a metal depends on temperature. This property is applied to measure temperature in resistance temperature detectors (RTD). Platinum is widely used as temperature sensor because of its linear dependence of resistance with temperature, also it is stable and it withstands very high temperatures making it very suitable for industrial applications. The problem of measuring the temperature of the microheater during pulsed heating is solved by measuring the instantaneous electrical resistance increase of the microheater.

The thermal coefficient of resistance

For the experiments performed here we rely on the ability of the microheater to measure the temperature of the liquid layer in contact with the microheater surface. By measuring the change in resistance of the microheater we can calculate the increase of temperature. If the heater is thin enough, gradients in the thickness can be negligible. Gradients along the length and width of the heater may thought not be so small. Therefore it is important to note that we will obtain the average temperature over the microheater surface. We assume that the temperature of the microheater is equal to the temperature of the liquid in contact with it.

The calibration of the system is crucial to have correct temperature values. The calibration was made using a special thin film resistor (100 μm × 10 mm) fabricated on the same wafer than the microheaters used for the experiments. Details of the calibration specimen and the calibration procedure are given in the previous chapter (set-up). The result of the calibration is depicted in Fig. 7.6. The resistance of the calibration device is initially 194 Ω, \( (R_0) \). The unitless resistance increase was plotted against the temperature. A linear fit of the represented points was used to calculate the temperature coefficient of resistance, \( (\alpha) \), that is the slope of the linear fit, in this case \( \alpha = 0.00199 \ K^{-1} \). The error in the calculation of \( \alpha \) was taken as the statistical error in calculating the slope of the trend line, (see Fig. 7.6). This value of \( \alpha \) is lower than the value for bulk platinum \( (\alpha =0.003925 \ based \ on \ ITS-90) \). Other groups (Zhang et al., 2005; Kim et al., 2001) obtained values for the thermal coefficient of resistance of platinum films. Those values
are substantially lower than that associated to bulk platinum too. This difference in properties may be associated with differences in the crystalline structure and with the presence of the underlaying chromium adhesion layer.

The calibration device was used to measure the sheet resistance of the metallic layer. The sheet resistance, $R_s$ is a parameter used to characterize thin films of deposited metals which resistivity is a strong function of the film thickness. The sheet resistance is defined as the resistivity divided by the film thickness $d$, $(R_s = \rho/d)$. For a thin-film sample of length $L$, width $W$ and resistance $R$, the sheet resistance can be calculated as the resistance divided by the number of squares (or $L/W$). Strictly speaking, the unit for sheet resistance is $\Omega$ (since $L/W$ is unitless). To avoid confusion between $R$ and $R_s$, however, the sheet resistance is specified in unit of $\Omega$ ‘ohms per square’. The $L/W$ ratio can be thought of as the number of unit squares (of any size) of material in the resistor. Since the structure used for calibration is $10 \text{ mm length} \times 100 \mu\text{m}$ wide, (see picture in chapter set-up), we can obtain the sheet resistance that is: $1.94 \ \Omega$. This value is consistent with similar microfabricated platinum heaters found in the literature (Kim et al., 2001).
Resistance thermometry

In this section, an example using real traces from one of the experiments illustrates the calculation of the temperature using resistance thermometry. Fig. 5.2 shows in (a), the current pulse supplied to the microheater, (acquired by the current probe, APO15, LeCroy) and in (b), the induced voltage drop signal (conditioned by the differential amplifier, DA1822, LeCroy). The instantaneous electrical resistance of the heater $R(t)$, is obtained by dividing the measured voltage difference $V(t)$ by the monitored current $I(t)$ according to the Ohm’s law: \( R(t) = \frac{V(t)}{I(t)} \). The obtained $R(t)$ values are shown in Fig. 5.2 (c), where also the initial resistance of the heater ($R_o$) is plotted for comparison (dashed line). The change in resistance of the heater is related to the change in temperature:

\[
T(t) = T_o + \frac{V(t)}{I(t)} - R_o \cdot R_o \cdot \alpha
\]  

(5.1)

Where $R_o$ is the nominal resistance measured at the reference temperature, $T_o$ and $\alpha$ is the temperature coefficient of resistance.

Detecting nucleation temperature

This section illustrates with an example how the voltage drop signal acquired during pulsed heating can be used to detect boiling phenomena. An identical electric current pulse was supplied to a microheater with and without water on it. The Fig. 5.3 (a) shows the voltage drop monitored for the experiment with the dry heater (solid line) and for the experiment with the microheater loaded with a drop of water. The current supplied was 0.4 A. The voltage drop measured for the dry heater (solid line) is higher than for the case in which the microheater is loaded with water (dashed line). The voltage traces are converted to temperature and the result is shown in Fig. 5.3 (b) for both, the dry experiment and the experiment with the microheater loaded with water.

In the dry experiment the temperature reaches values up to 400°C, while in the experiment performed with the heater loaded with water the temperatures are lower. Moreover, a change on the slope and a kink or inflection in the curve can be observed lagging 13 µs the heating onset. Visualization of the phenomena showed that the kink corresponds with the moment when most of the microheater is covered with vapor.
5.2 Measurement results

Figure 5.2: Signals acquired: (a) electrical current pulse, (b) voltage drop, (c) calculated resistance of the microheater and (d) temperature.

5.2 Measurement results: Effect of the current pulse amplitude on the heating process

The influence of the current amplitude was studied by supplying different current pulses to a microheater device. For the experiments presented in this section, a square microheater of 50 \( \mu m \) side without the fluidic part was used. The initial resistance of the heater was 3.4 \( \Omega \). The heater was loaded with DI water. The voltage and the current traces were measured for the duration of the heating pulse and the average temperature was calculated using Eq.5.1. Fig.5.4 summarizes the results: The calculated average temperature increase of the microheater is plotted against the time for the respective heating pulse durations. To avoid the rapid deterioration of the heater, the pulse length was selected to stop shortly after having a vapor layer covering the entire surface of the microheater. The amplitude
Temperature measurements

Figure 5.3: Voltage traces (a) and temperature traces (b) for the same microheater without liquid (solid line), and loaded with water (dashed line).

of the current pulses (heating pulses) supplied in each experiment is shown in the legend. The pulses supplied range from 0.47 A amplitude and 16 μs duration to 0.7 A amplitude and 5 μs duration.

The initial heat generation rate in each of these experiments can be calculated from the initial resistance of the microheater and the current supplied during the experiment using the Joule’s law (the heat generated by an electrical current passing by a resistor is proportional to its resistance and to the square of the current passed through it, $R_o \cdot I^2$). In this experiments it ranges from: 0.75 W to 1.81 W. If we divide the power generated by the surface area of the heater, heat fluxes of the order of 600 MW/m² are obtained. We need to consider, however, that only a fraction of the total heat generated is transferred to the liquid. Since the mass of the microheater is very small the fraction of heat accumulated in the microheater is negligible and the part that is not transferred to the liquid is conducted through the substrate.

The rate of temperature rise obtained during these experiments was in the order of $10^7$ to $10^8$ K·s⁻¹. Higher current amplitude pulses induced higher rates of temperature rise. All the curves exhibit a change of the trend or kink in the temperature evolution. The kink is in some cases not obvious and we used numerical methods based on moving average and differential analysis to detect the point where the change in slope takes place, (marked with × in Fig.5.4). The time delay between the start of the pulse, (heating
5.2 Measurement results

Figure 5.4: Average temperature increase induced in a square platinum heater of 50 × 50 µm². The initial resistance of the heater (measured at ambient temperature) is 3.4 Ω.

onset), and the kink (representing the time when the vaporization of the layer of water in contact with the heater takes place) shortens with the increase of current amplitude: it is 11 µs for a current pulse of 0.47 A, 4.5 µs for a current amplitude of 0.62 A and even shorter, 2.7 µs for 0.7 A current amplitude.
Temperature measurements

Rate of temperature rise

The temperatures plotted as a function of time in Fig. 5.4 show, at the beginning an exponential increase of the temperature with time (characteristic of a lumped parameters thermal problem); the temperature increases at a faster rate close to the origin and the slope of the curve decreases and stays approximately linear until the kink on the curve indicates that the heater is covered with vapor. Fig. 5.5 shows an example of temperature trace and it illustrates the parameters that we will later use to discuss the results. The rate of temperature rise is the temperature increase measured on the heater corresponding to the kink in the curve, divided by the time at the position of the kink, (the slope $\beta$ in Fig. 5.5). The nucleation temperature was calculated as the temperature measured on the microheater at the position of the kink in the temperature trace, ($T_b$ in Fig. 5.5).

![Figure 5.5: Example of temperature trace with a mark ($\times$) in the position of the kink. The rate of temperature rise is the slope of the line ($\beta$) passing by ($T_o, t_o$) and ($T_b, t_b$).](image)

Error Analysis

The error in the calculation of the temperature can be obtained from the expression in Eq. 5.1, calculating the partial error induced by each variable appearing in this expression.
5.2 Measurement results

\[ \Delta T = \Delta T_o + \left| \frac{1}{R_o \cdot \alpha} \right| \cdot \Delta R + \left| \frac{R}{R_o^2 \cdot \alpha} \right| \cdot \Delta R_o + \left| \frac{R - R_o}{R_o \alpha^2} \right| \cdot \Delta \alpha \]  

(5.2)

Where \( \Delta T_o, \Delta R_o, \Delta \alpha \), represent the uncertainty measuring \( T_o, R_o \) and \( \alpha \) respectively, and \( \Delta R \) is the error associated with the measurement of the resistance. Since the resistance of the microheater, \( R \), is calculated from the expression: \( R = \frac{V}{I} \) we can obtain its uncertainty, \( \Delta R \) from the measured values of current, \( I \), and voltage, \( V \), as:

\[ \Delta R = \left| \frac{1}{I} \right| \cdot \Delta V + \left| \frac{V}{I^2} \right| \cdot \Delta I \]  

(5.3)

Using the uncertainty associated to our measurement devices; \( \Delta T_o = 1^\circ C, \Delta R_o = 0.02\Omega, \Delta \alpha = 10^{-5}K^{-1}, \Delta V = 10^{-2}V \) and \( \Delta I/I = 2 \cdot 10^{-4} \). The maximum uncertainty obtained was 6\(^\circ\)C; 2\(^\circ\)C due to the error measuring \( R \), 2\(^\circ\)C due to errors associated with the measurement of \( R_o \) and less than 1\(^\circ\)C due to the calibration errors.

<table>
<thead>
<tr>
<th>Rate of temp.rise [K·s(^{-1})]</th>
<th>Pulse length [( \mu )s]</th>
<th>Nucleation temp. [(^\circ)C]</th>
<th>Total power sup. [W]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.56 ·10(^7)</td>
<td>16</td>
<td>213.1</td>
<td>0.7</td>
</tr>
<tr>
<td>2.2 ·10(^7)</td>
<td>11.5</td>
<td>230</td>
<td>0.9</td>
</tr>
<tr>
<td>3.2 ·10(^7)</td>
<td>8</td>
<td>236</td>
<td>1.14</td>
</tr>
<tr>
<td>3.9 ·10(^7)</td>
<td>7.2</td>
<td>249</td>
<td>1.26</td>
</tr>
<tr>
<td>5 ·10(^7)</td>
<td>5.5</td>
<td>252</td>
<td>1.47</td>
</tr>
<tr>
<td>7.1 ·10(^7)</td>
<td>4.2</td>
<td>273.0</td>
<td>1.68</td>
</tr>
</tbody>
</table>

Table 5.1: Experimental data gathered for different heating pulses

Table 5.1 summarizes the rate of temperature rise and the nucleation temperature obtained for a series of experiments with different current pulses. The values of nucleation temperature are plotted against the rate of temperature rise in Fig.5.6. The nucleation temperature increases with the rate of temperature rise. The maximum nucleation temperature obtained in the present experiment is 272 \(^\circ\)C corresponding to a rate of temperature rise of 70 ·10\(^6\) K·s\(^{-1}\) and a total power input of 1.68 W. the data scattering was of ± 6 \(^\circ\)C. It accounts for the errors in measuring temperature and also in finding the kink in the curve. We did not reach a maximum as in the experiments performed by Glod et al. (2002). They found that for a rate of temperature rise above 60 ·10\(^6\) K·s\(^{-1}\)
the nucleation temperature reach an asymptotic maximum value of 301°C. This value is close to the theoretical maximum superheat attainable in water at atmospheric pressure. The average nucleation temperature measured in the present experiments are quite below the maximum limit or superheat.

Some considerations may help to explain the lower temperatures measured in this experiments. First, the geometry of the thin film heater used for the present experiments induces temperature gradients along the heater surface decreasing the average temperature measured. By contrast, the experiments presented in Glod et al. (2002) used a platinum wire where the temperature is expected to be rather homogenous. Second, in the present experiments only half of the area of the microheater (neglecting the borders) is in contact with the liquid, (the other half is in contact with the substrate). Therefore it is less sensitive to the lower heat transfer associated with the formation of vapor on top of the heater. This makes the identification of the kink in the curve difficult. And third, the substrate may influence the heat transfer problem: once the vapor starts to form, the amount of heat transferred to the fluid is reduced so that the microheater suddenly gets hotter and, as a consequence, more heat is transferred to the substrate than when there is liquid onto the microheater. That may have an influence in the relation between the
rate of temperature rise and the nucleation temperature obtained.

<table>
<thead>
<tr>
<th>Rate of temp. rise $[K \cdot s^{-1}]$</th>
<th>Heater</th>
<th>Nucleation temp. $[^\circ C]$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>71·$10^6$</td>
<td>$50 \times 50 \mu m^2$ Pt thin film</td>
<td>273</td>
<td>present study</td>
</tr>
<tr>
<td>86·$10^6$</td>
<td>Pt wire ($\varnothing = 10\mu m$)</td>
<td>303</td>
<td>Glod et al. (2002)</td>
</tr>
<tr>
<td>1·$10^6$</td>
<td>Pt wire ($\varnothing = 20\mu m$)</td>
<td>302</td>
<td>Skripov and Pavlov (1970)</td>
</tr>
<tr>
<td>10·$10^6$</td>
<td>Pt wire ($\varnothing = 25\mu m$)</td>
<td>290</td>
<td>Derewnicki (1985)</td>
</tr>
<tr>
<td>93·$10^6$</td>
<td>Pt film heater</td>
<td>295</td>
<td>Iida et al. (1994)</td>
</tr>
<tr>
<td>250·$10^6$</td>
<td>Ta-Al film heater</td>
<td>283</td>
<td>Avedisian et al. (1999)</td>
</tr>
</tbody>
</table>

Table 5.2: Comparison of maximum nucleation temperature measured for water at atmospheric pressure

The table contains a list of the maximum nucleation temperature experimentally obtained for water at atmospheric pressure using different heater configurations. From the values listed there, it can be seen that in general lower values are obtained using thin film heater configuration even at very high rates of temperature rise.

5.3 Effect of the current amplitude for an enclosed system

The temperature evolution of the microheater under pulsed heating was investigated for the confined-liquid system. In this case the microheater device is composed of the heater plus a PDMS chip (with embedded fluidic microchannels and a cavity) bonded onto it. The liquid is not loaded onto the heater but filled in through the inlet port drilled through the PDMS. The general trend of the temperature evolution measured in this case does not differ from the case described in the previous section (heater loaded with water).

Two situation were measured with the composed device (confined system), with open access to the cavity (ports) and with closed inlet/outlet ports. The holes (ports) were closed with stabs fitting the diameter of the holes (0.8 mm diameter). The confinement of the liquid was expected to elevate the nucleation temperature. This was shown analytically by Carey, (Carey, 2000). He predicted that the confinement should induce a
Figure 5.7: Average temperature increase induced in a square platinum heater of 50 × 50 μm². The initial resistance of the heater (measured at ambient temperature) is 3.4 Ω.

We could not observe any difference in the measured temperature trace for the open and the closed system. The reason why we do not observed a corresponding increase in the measured values of nucleation temperature may be that the enclosure is too flexible. In that case the increase in pressure may be too small to significatively modify the nucleation temperature.

### 5.4 Influence of the initial resistance

The actual resistance of the fabricated microheaters differed among different specimens. Since we have square heaters, the resistance of each microheater was expected to be about 1.94 Ω, \( R_s = 1.94\Omega/\square \). However, the resistance of the microheaters was 3.5 ± 1 Ω. The main reason for these high values of resistance is that the electrodes used to sense the...
voltage drop define a resistance that is slightly longer than the width, (not 1 square but
1.3 squares, see Fig. 5.8). Another reason for this high value of the resistance and also for
the dispersion of the measured values could be the non homogeneity of the thickness of the
platinum layer. Also the fabrication process is susceptible to generate slight differences
in the dimension of the specimen that could account for slight bias in the final resistance
of the device.

Figure 5.8: Dimensions of the fabricated microheater

Comparison between microheaters with different initial resistance should consider the
total power input as controlling parameter rather than the amplitude of the current pulse.
An example of this issue is illustrated in Fig. 5.9 Two different heaters of initial resis-
tance, $R_{o1} = 3.72 \, \Omega$ and $R_{o2} = 3.4 \, \Omega$ were used to illustrate the influence of the initial
resistance on the effect of pulsed heating. The temperature traces obtained for three
different experimental conditions are plotted for both specimens in Fig. 5.9 (a). The
current amplitude supplied in each case is indicated next to the temperature trace. For
each experiment with the first microheater ($R_{o1}$), the current amplitude was tuned so that
the two temperature evolution curves are identical, meaning that they lead to the same
rate of temperature rise and same nucleation temperature. The current pulses supplied
for the experiments are depicted in 5.9 (c), and the voltage traces in 5.9 (d). We also
plotted the instantaneous total power input (Fig. 5.9 (b)), which shows identical values
for the traces corresponding to both microheaters in each one of the three current levels.

In view of these results we can conclude that for two different microheater devices of
different initial resistances, a similar temperature evolution can be induced when supplying
a similar initial power (rather than similar current amplitude).
5.5 Conclusions

The fabricated platinum microheater subjected to pulsed heating reached high temperatures in few microseconds. The rates of temperature rise obtained were as high as \(70 \times 10^6 K \cdot s^{-1}\). The inflection in the temperature evolution curve can be used to identify the time and temperature associated with the formation of vapor on the heater. As observed by Glod et al. (2002); Skripov (1974); Avedisian et al. (1999) the nucleation temperatures increase with the rate of temperature rise. The maximum nucleation temperature measured with our device is 273 °C, thus below the maximum superheat for water at atmospheric pressure. The reason for the low nucleation temperature measured in the present study may be due to microheater geometry, (strong temperature gradients on the heater surface during pulsed heater are induced by inhomogeneous current intensity distribution).
The temperature evolution of the microheater under pulsed heating was investigated for the confined-liquid system. The measurements using the confined system did not reveal differences in the temperature evolution nor in the nucleation temperature measured as predicted in previous analytical work by Carey (2000). The reason why we did not observe a corresponding increase in the obtained nucleation temperature measured may be that the enclosure is too flexible. In that case the increase in pressure may be too small to significantly modify the nucleation temperature. It was also shown that for two different microheater devices of different initial resistances, a similar temperature evolution can be induced when supplying a similar initial power (rather than similar current amplitude).
Chapter 6

Visualization results

The microbubble formation and collapse cycle takes place typically in few $\mu s$. Visualizing the process in such a time ($\mu s$) and length ($\mu m$) scale is quite important in order to understand the details of the phenomena and it is essential to efficiently control the processes involving thermal actuation.

Since explosive vaporization is quite repeatable, visualization was achieved by a stroboscopic technique. Skripov [1974] was the first who used a Xenon flash lamp to illuminate explosive vaporization in a pulse-heated wire of 20 $\mu m$ diameter. The electric pulse acting as the heating pulse triggers the flash lamp. Glod et al. [2002] used a similar method to visualize vaporization on top of a thin film heater and around thin wires. The main advantage of this method is that it is very easy to be combined with microscopy visualization; for example in Glod et al. [2002] the lamp was mounted on a home-made lamp-housing and fixed to the reflected light port on the microscope. The camera does not need to be a high speed camera what reduces the costs of the system.

The images shown in this chapter were acquired with a set-up based on flash illumination. We acquired images with a CCD camera and recorded them with a triggered frame grabber. For each heating event one picture was taken. The short duration flash is triggered to illuminate a given instant of the fast vaporization process. A sequence can be made by progressively increasing the time lag between the heating onset and the lamp trigger. The acquisition of the sequences was programmed with a Labview routine. The
heating pulses were fired with a frequency of 1 Hz to allow enough cooling down period between the heating events.

In this chapter we will see sequences of images corresponding to different experimental conditions. Explosive vaporization on the microfabricated heaters loaded with deionized water will be shown first. Next, the explosive vaporization within the PDMS microcavity will be presented. Then the effect of closing the inlet and outlet ports in the confined system will be discussed. The visualization of explosive vaporization on a bigger heater (100 $\mu$m side length) will be shown for comparison. We will conclude present the chapter discussing these results and the limitations of this visualization system.

Figure 6.1: Sequence of images showing the bubble growth and collapse on top of a heater loaded with water. The heater has 50 $\mu$m side length. $R_o = 3.4 \, \Omega$. The pulse is 0.47 A and 14 $\mu$s duration
6.1 Visualization of explosive vaporization on the microfabricated heaters

The sequence of images depicted in Fig. 6.1 shows the bubble formation and collapse on a square microheater of 50 µm side length and initial resistance, $R_o = 3.4 \, \Omega$. The current pulse (heating pulse) supplied for this experiment had an amplitude of 0.47 A and a duration of 14 µs. The rate of temperature rise in this case was $1.5 \times 10^7 \, K \cdot s^{-1}$.

![Temperature trace](image)

Figure 6.2: Temperature trace measured showing the average temperature corresponding to selected frames in Fig. 6.1

In the sequence depicted in Fig. 6.1, the first vapor bubble appeared in the frame taken at 5 µs. This first vapor bubble can be seen in the corner of the heater. The next frame at 6 µs revealed a vapor bubble in the opposing corner. This indicates the presence of gradients of temperature in the heater, probably due to its topology that induces an inhomogeneous current intensity distribution. Both sites grow towards the center of the heater.

The two vapor bubbles grow and merge in the elapsed image corresponding to 11 µs which shows the microheater almost completely covered with vapor. The shadow in the perimeter of the vapor region indicates that the bubble grows also in the perpendicular direction. The bubble reaches its maximum size in the frame corresponding to 15 µs, and then it starts to collapse. The collapse phase occurs faster than the bubble growth, lasting 6 µs (versus the 10 µs duration of the growth period).
Figure 6.3: Sequence of images showing bubble growth and collapse on top of a heater loaded with water. The heater has 50 µm side length. $R_o = 3.4 \, \Omega$. The pulse is 0.7 A and 5 µs duration.

The temperature evolution corresponding to the visualization in Fig. 6.1 is shown in Fig. 6.2. The red mark (×) indicates the kink in the temperature trace. The small marks (+) along the temperature trace indicate the time corresponding to the acquired frames shown in Fig. 6.1. The information from the visualization (Fig. 6.1) and from the temperature measurement (Fig. 6.2) can be combined. For example, the average temperature of the heater 6 µs after heating onset is 132 °C and corresponds to the frame labeled as ‘6 µs’ in Fig.6.1 (third frame). Furthermore, these results can be used to interpret the meaning of the inflection in the temperature trace. As it was already mentioned, the temperature associated with the kink is called the nucleation temperature. In this case it corresponds to a temperature of 213 °C and a time lag of 12.5 µs with respect to the heating onset. This instant is associated with a bubble topography that is
Figure 6.4: Temperature trace measured showing the average temperature corresponding to selected frames in Fig. 6.3

in between the frames labeled as 12 µs and 13 µs in Fig. 6.1

For comparison we also acquired a series of images using the same microheater device but a much more intense heating pulse (higher current amplitude). The pulse duration was reduced to avoid the burn out of the microheater. The sequence of images taken in this case is presented in Fig. 6.3. Initially vapor bubbles formed on two of the corners (see the second frame in Fig. 6.3). Then the four corners of the microheater appeared covered with vapor (at 2.5 µs). These four vapor regions in the corners grow towards the center of the heater. The heater is covered with vapor 4 µs after starting the heating pulse. The shadow in the borders indicates that the bubble grows also upwards towards the bulk fluid. The maximum bubble size was reached at 6.5 µs and then it started to collapse. In this case the duration of the collapsing process lasts approximately the same than the growth process.

The temperature evolution corresponding to this visualization set (Fig. 6.3) is shown in Fig. 6.4. The red mark (×) indicate the kink in the temperature trace. The small marks (+) along the trace indicate the time corresponding to the frames shown in Fig. 6.3. The information from the visualization (Fig. 6.3) can be complemented, as before, with the average temperature of the microheater at the time corresponding to each frame. This temperature values can be read out of the temperature trace depicted in Fig. 6.4. For the selected frames (labels in boxes) the corresponding value of the temperature is
Visualization results

written and connected with an arrow to the corresponding point in the temperature curve. The instant identified with the average nucleation temperature, (marked with a ×) can be associated with the first moment when all the heater is covered with vapor, (between frames 3.5 μs and 4 μs in Fig 6.3).

Fig 6.1 and Fig 6.3 show the effect of modifying the current intensity. The temperature of the heater increases much faster and vaporization initiates earlier and in a more homogenous way. The bubble collapse process seems to be independent from the applied current intensity lasting in both cases ∼ 6 μs.

6.2 Visualization of explosive vaporization within the PDMS microcavity

In the previous visualization experiment the liquid was deposited onto the heater. To study the effect of confining the liquid a cover with an embedded cavity was fabricated in a transparent flexible polymer, (Poly-dimethylsiloxane also know as PDMS). This fluidic cover was bonded to the glass chip and the liquid was injected into the channels and the cavity.

The resistance of the microheater bonded to the fluidic cover was 3.4 Ω, (similar to the one presented in the precedent section). The projection of the cavity can be seen from the frames in Fig. 6.5. It is a square of base area 200 μm × 200 μm.

Fig 6.5 shows a sequence of images for a heating process induced by a current pulse of 0.47 A amplitude and 15 μs duration. The process corresponds to an initial power supply of 0.75 W. Visualization was made through the PDMS. The first nucleation site can be seen in the third frame of the sequence, corresponding to 7 μs. The region covered with vapor evolved from the corners towards the center following a pattern similar to the one in Fig. 6.1. The frame correspondonging to 11 μs shows the microheater covered with vapor. The vapor bubble extends even slightly further the heater limits and it starts to collapse, (frame corresponding to 15 μs in Fig 6.5). The temperature trace corresponding to the experiment in Fig 6.5 is shown in Fig. 6.6.
6.2 Visualization of explosive vaporization within the PDMS microcavity

Figure 6.5: Sequence of images showing bubble growth and collapse inside microcavity. The current pulse is 0.47 A, 15 µs duration. The microcavity is filled with DI liquid. The initial resistance of the microheater is $R_0 = 3.4 \, \Omega$

For comparison the same system was subjected to a current pulse of higher amplitude. A sequence of images capturing the bubble formation and collapse process is shown in Fig. 6.7. The process corresponds to an initial power supply of 1.66 W. First, two bubbles appeared in the corners of the heater at 1.5 µs (third frame in Fig. 6.7). Next frame was acquired at 2.5 µs, two more bubbles appear in the opposing corners. In the frame corresponding to 3.5 µs a film of vapor was covering the whole heater. The temperature trace corresponding to the experiment in Fig. 6.7 is shown in Fig. 6.8.

We can not be sure that the liquid is not leaving the cavity as the bubble forms relieving the pressure inside the cavity. Therefore, we blocked the inlet and the outlet ports so that the liquid could not leave the cavity. The sequences in Fig. 6.11 and Fig. 6.12 show the bubble growth-collapse cycles for analogous current pulses but closing the input and outlet ports.
Figure 6.6: Temperature trace measured showing the average temperature corresponding to selected frames in Fig. 6.5.

6.3 Comparison between open and closed system

The bubble projected area divided by the area of the microhater is shown in Fig. 6.9. This plot depicts the projected area evolution for the experiments performed in the PDMS cavity (confined liquid) with open (\textasteriskcentered) and closed (\textdiamondd) ports. The data reveals that the initial phase (growing phase) is not modified by having the system closed or open. However, the duration of the collapse phase is shorter for the closed system.

6.4 Visualization on a 100 \( \mu \text{m} \) side length micro-heater

We fabricated microheaters of two different dimensions 50 \( \mu \text{m} \) and 100 \( \mu \text{m} \). Most of the experiments were carried out using the 50 \( \mu \text{m} \) side length microheater because higher rate of temperature rise could be achieved. For comparison also a sequence showing vaporization on top of a 100\( \mu \text{m} \) side length microheater is shown in Fig. 6.10.

The images are clearer because the size of the microheater allows the use of a lower magnification and thus the depth of focus is higher. Furthermore, the process is slower than the previously presented ones. The microheater is covered by vapor 17 \( \mu \text{s} \) after starting the heating pulse. Since the image is frozen by the illumination flash, the speed
Figure 6.7: Sequence of images showing bubble growth and collapse inside microcavity. The current pulse is 0.7 A, 4 \(\mu\)s duration. The microcavity is filled with DI liquid. The initial resistance of the microheater is \( R_0 = 3.4 \, \Omega \)

of the process has a direct effect on the quality of the image, (the movement during the illumination period induced a blur in the image). This is a limitation of the system in terms of temporal resolution. The fact that the flash lamp has a jittering further lowers the temporal resolution.

### 6.5 Conclusion

Visualization of explosive vaporization on top of thin film platinum microheaters were achieved by means of a pulsed image acquisition system using flash lamp illumination. We
Figure 6.8: Temperature trace measured showing the average temperature corresponding to selected frames in Fig. 6.8

Figure 6.9: Time evolution of the bubble projected area during pulsed heating induced boiling for open and closed systems

could associate the temperature measurements and the frames corresponding to selected instants, confirming that the kink in the curve used to measure the nucleation temperature coincide with the first moment when vapor covers the entire surface of the microheater. Visualization of explosive vaporization through a PDMS fluidic chip was also achieved. The confinement does not affect the bubble growth but it accelerates the bubble collapse process. Visualization of vaporization on a bigger heater (100 µm side length square) was also presented.
Figure 6.10: Sequence of images showing slow bubble growth and collapse process induced on a 100 \( \mu m \) square heater.
Figure 6.11: Sequence of images showing bubble growth and collapse inside the microcavity. The current pulse is 0.47 A, 12 µs duration. The microcavity is filled with DI liquid. The initial resistance of the microheater is $R_o = 3.4 \, \Omega$, closed system.
Figure 6.12: Sequence of images showing bubble growth and collapse inside the microcavity. The current pulse is 0.7 A, 4 µs duration. The microcavity is filled with DI liquid. The initial resistance of the microheater is $R_o = 3.4 \, \Omega$, closed system.
Chapter 7

Explosive vaporization on submicron strip heaters

7.1 Introduction

Explosive boiling is an abrupt phase change phenomenon taking place in a highly superheated liquid. A liquid is superheated when its temperature is above the saturation temperature. The liquid can become superheated if: 1) the liquid is depressurized rapidly at nearly constant temperature (Bartak, 1990) or 2) the liquid is heated to a higher temperature while its pressure is maintained. Under carefully controlled conditions (no contamination, smooth enclosure surfaces and no physical disturbances) it is possible for a liquid to be maintained in a superheated state. However, under most practical circumstances the ideal conditions cannot be met and the superheated liquid will return to its equilibrium condition through vaporization.

Explosive boiling in the microscale has been used to eject droplets of ink (ink jet printer) and, currently, works are in progress towards developing microfluidic devices actuated by thermally generated bubbles as drug delivery systems, valves and pumps. Even though it has been successfully exploited, there is a lack of complete understanding of the phenomenon.

Many researchers investigated the physics and controllability of explosive boiling in wires and in thin film heaters, (Glod et al. 2002, Avedisian et al. 1999, Skripov and
Explosive vaporization on submicron strip heaters

Despite the abundance in such researches, very little refers to submicron heaters. The only example known to the author is Deng et al. (2004) who reported bubble formation on two submicron size heaters. The heater sizes were: $0.5 \, \mu m \times 0.5 \, \mu m$ and $1 \, \mu m \times 0.5 \, \mu m$. In their work, they used pulsed heating with a fixed duration of 1.66 ms and the current was adjusted to the minimum required for nucleation within the pulse duration. In their experiments, nucleation took place after a delay of 51 $\mu s$ and 60 $\mu s$ and at heater temperatures of 245°C and 207°C respectively. The maximum bubble diameter for both cases was $2.5 \pm 0.3 \mu m$. Bubble nucleation onset was detected by a sudden change in the measured average temperature as a function of time. They achieved heating rates of the order of $10^7 K/s$ with heat transfer rate from the heater of 190.9 MW/m$^2$ and 112.4 MW/m$^2$ respectively.

The same group of authors considered the scaling effects of micro bubble actuation (Deng et al., 2005, 2006). They fabricated microheaters ranging from $150 \, \mu m \times 50 \, \mu m$ to $0.5 \, \mu m \times 0.5 \, \mu m$ out of the same wafer to minimize the differences due to the fabrication process. They observed two different vapor region types: ‘oblate vapor blank’ for the bigger heaters ($> 10 \mu m$) and ‘spherical bubble’ for the submicron heaters. They related this boiling pattern to the superheated region around the heater.

In this work we present an investigation of explosive boiling on a self-sensing heater element. The novelty of this investigation is the small dimension of the heater ($\sim 0.5 \, \mu m \times 3-6 \, \mu m$) as well as the device layout which allows for a 4-point, resistive measurement technique. We measured the average temperature increase on a submicron platinum heater subjected to electrical pulses. We observed the occurrence of boiling and the effect of current intensity on the moment at which vaporization takes place. Very high rates of temperature rise were achieved because of the small dimensions of the device, leading to the initiation of vaporization in less than 1 $\mu s$.

Additionally, high frequency temperature oscillations (MHz) were observed which are associated with rapid oscillations of the vapor layer thickness taking place on the surface of the heater.

This oscillatory phenomenon was observed previously (Avedisian et al., 1999, Osborne 1947). In the work of Avedisian et al. (1999) the heater was a thin film heater such as those used in an HP ink jet printer cartridge. Not much attention was dedicated to this
phenomenon since the author focused on the initial explosion leading to a single droplet ejection. The second work (Osborne, 1947), used a metallic wire immersed in a pool of liquid. They measured the sound generated by the boiling phenomena taking place in the wire.

We measured the frequency and amplitude of the temperature oscillations for different current intensities showing that the increase of the current intensity increases the frequency and decreases the amplitude of the temperature oscillations. To our knowledge, this is the first study of this phenomenon using thin film resistors.

Visualization of the vapor layer was also achieved using a pulsed image acquisition system synchronized with the heating pulse. Limited temporal resolution of the illumination system did not allow the visualization of the bubble size oscillations but the formation of bubbles of submicron size could be confirmed.

Possible applications of this phenomenon can be found in biomedical devices, such as an ultrasound generator for the positioning of catheters, (Merdes and Wolf, 2001; Tatar et al., 2002). Also, ultrasound can be used to drive particles on a fluid by acoustic streaming (Marmottant and Hilgenfeldt, 2004).

7.2 Heater Fabrication

The substrate for the device is a 300 $\mu$m thick silicon wafer. A silicon nitride layer 500 nm thick was grown by using the low-pressure chemical vapor deposition (LPCVD) technique. Chromium (10 nm) and platinum (100 nm) were patterned on this substrate by the standard lift-off technique: First, a layer of photoresist is defined by photolithography. After the deposition of the metal by e-beam evaporation (UNIVEX 550) on top of the exposed substrate (the nitride layer), the photoresist is stripped off in acetone to define the metal patterns.

The entire metal pattern for 4-wire construction is shown in Fig. 7.1 (a). The central region, the size of a few tenths of a micrometer, was then milled by using a focused ion beam source (FIB). A narrow central region (the heater) could be patterned with the size of half a micron since the ion beam size (typically less than 10 nm) is much smaller.
Explosive vaporization on submicron strip heaters

Figure 7.1: (a) Metal structure patterned by lift-off before the Focused Ion Beam (FIB) milling. (b) Submicron heater structure (narrow central region enclosed in dashed rectangle) fabricated by milling the initial platinum structure. The voltage drop is measured across the two small electrodes ($V_1$ and $V_2$) for localize resistance thermometry.

than the region of interest. The two small electrodes in the middle were utilized for voltage drop measurement (Fig. 7.1 (b)). A ceramic carrier (28 Lead Side Brazed package, Spectrum, US) was used to interface the microfabricated chip with the measurement equipment.

7.3 Heater Characterization

Submicron heater calibration set-up

The resistor can be used as a temperature sensor since the increase of temperature induces an increase of resistance that can be measured through the 4-point arrangement, (see in Fig. 7.1 $V_1$, $V_2$, $I_1$ and $I_2$). To obtain a temperature value out of the resistance measurement we need to measure first the temperature coefficient of resistance ($\alpha$). We measured the resistance of the heater element at several heater temperatures and obtained the slope of the linear trend relating the relative resistance increase $(R - R_o)/R_o$ to the temperature increase $T - T_o$, where $R_o$ is the initial resistance measured at the initial temperature, $T_o$. 
This section describes the calibration procedure and the calibration set-up.

Figure 7.2: Calibration oven: The test device is mounted between two plates that completely cover the chip package. The upper cover was removed for the picture in (a) in which it is shown the ceramic package containing the chip (upside down). The assembled chip holder is depicted in picture (b). The cover of the vacuum chamber on which the chip holder is mounted is shown in (c).
Explosive vaporization on submicron strip heaters

Figure 7.3: Heater calibration set-up. A chip holder mounted in a vacuum chamber is used to fix the heater temperature. The temperature of the holder is set to different levels and the change in resistance of the heater is measured.

The calibration set-up is composed of:

- A metallic vacuum chamber with a modified front cover with vacuum tight connectors and a gas inlet port for nitrogen.
- Vacuum pump connected to the vacuum chamber.
- Nitrogen bottle.
- Chip holder.
- Data acquisition unit (HP2852A, Hewlett-Packard) to monitor the temperature of the chip holder.
- Temperature control unit.
- Low level current source, (6221, Keithley) and a Nanovoltmeter (2182A, Keithley).
7.3 Heater Characterization

Typical commercial sockets do not withstand high temperatures. Therefore, we fabricated a holder that provides electrical contacts (to perform the resistance measurement) and withstands high temperatures. The chip holder is composed of three parts which can be screwed together: the upper and lower aluminium covers and a steel plate 6 mm thick located between them. Holes are drilled into the upper and lower covers into which heating elements are inserted. The steel plate was machined to allocate 14 lead-throughs with glass insulation (BC-Tech AG, Chur, Switzerland). The glass insulation was needed to electrically insulate the lead-throughs from the steel plate. The lead-throughs are positioned to match the pins of the ceramic carrier which are connected together using screws. The two external aluminium plates completely cover the chip package. A photo of the holder assembly can be seen in Fig. 7.2. The chip holder is mounted on the cover of the vacuum chamber, (7.2c). The temperature in the holder is measured by two temperature sensors, PT100, (Distrelec AG, Nünikon, Switzerland).

The temperature read out from the PT100 sensors located in the holder are acquired using the data acquisition unit (HP 3852A) and read by a PC with a Labview program via GPIB. The temperature control unit uses the PT100 readings together with the set point temperature to control the heating elements. A sketch of the calibration set up is represented in Fig.7.3.

Resistance measurement:

For the measurement of the resistance during the calibration we used a low level current source (model 6221, Keithley) and a Nanovoltmeter (model 2182A, Keithley). The applied current needs to be kept small to reduce errors associated with self heating.

Thermoelectric voltages are generated when different parts of a circuit are at different temperatures or when conductors made of dissimilar materials are joined together. When the calibration is performed, a temperature gradient is present in the circuit especially inside the oven. Because it is not possible to avoid the presence of contacts between dissimilar materials and the applied current need to be kept small to avoid self heating, the thermoelectric voltages are considerable and need to be corrected.

The thermoelectric voltages can be canceled out by using the combination of the Keithley 6221 Source and the 2182A Nanovoltmeter. The two instruments work in a
synchronized way supplying the current pulses and taking one measurement before and one during each pulse. The Keithley 6221 calculates the difference between these two measurements suppressing any constant thermoelectric offset and displays the true value of the voltage.

**I-V Characteristics**

In order to characterize the heater, we determined it current-voltage characteristic (I-V) showing the relationship between the dc current passing through the electronic device and the dc voltage across its terminals. This characterization curve is useful for two purposes: first, it shows the nonlinearity due to the temperature dependence of the resistance and second, it indicates the amplitude of the current pulse necessary to achieve a certain temperature.

According to the Ohm’s Law, for a resistor, there is a linear relationship between the applied current and the resulting electrical voltage drop. Since the applied current increases the temperature of the heater, and an increase of temperature modifies the resistance, the I-V characteristic becomes non-linear.

Different current pulses were supplied to the submicron heater and the corresponding voltage drops were measured. The pulses were 200 $\mu$s in duration to ensure that the heater reaches thermal equilibrium. This has been performed in vacuum, in air and in water.

### 7.4 Experimental set-up for explosive boiling study

The experimental set-up used to investigate explosive boiling in the submicron heater will be described in this section. We first introduce the set-up for heating the device and monitoring the temperature evolution. Next, the image acquisition system used to capture images of the submicron bubble.
Pulsed heating set-up

For the fast heating experiments a pulse generator that has a short settling time (model HP8114A, Hewlett-Packard) was used. It supplied a pulse to a circuit consisting of a 100 Ω resistor connected in series with the submicron heater (Fig 7.4). The resistor is used for measuring the current (by measuring the voltage drop across the resistor) and as a voltage divider. A differential amplifier (DA1822A, LeCroy) is connected to the terminals of the resistor. The terminals used to sense the voltage drop across the submicron heater are connected to a second differential amplifier (DA1855A, LeCroy). The two signals are monitored with an oscilloscope (LC334A, LeCroy). The oscilloscope is triggered by the heating pulse and it is set to acquire at 1 GHz. The signals are then exported to a PC via GPIB interface.

The instantaneous current through the circuit at time t, is obtained by applying the Ohm’s law:  
\[ I(t) = \frac{V}{100\Omega} / 100\Omega \]

The instantaneous temperature of the heater (spatially averaged over its surface) can be obtained from the current, \( I(t) \) and the voltage drop across the heater, \( V(t) \):

\[ T(t) = T_0 + \frac{V(t)}{I(t)} - \frac{R_o}{R_o \cdot \alpha} \]  

(7.1)

Pulsed image acquisition

The visualization technique used is based on flash lamp illumination of the region of interest at a certain ‘instant’ after triggering the process. To achieve this, two pulses with a certain delay between them were generated. The first one triggers the pulse generator that starts the heating pulse and the second one triggers the flash lamp.

The phenomenon under investigation can be repeated as long as the time between successive pulses is long enough to return to its initial conditions.

When the delay between heating onset and the flash lamp trigger is kept constant the sequence captured images seen are always the same, getting the impression of a frozen process. If the delay is increased by small increments the sequence of images seen gives the impression of a slowly moving process.

The image acquisition system is composed of:
Figure 7.4: Set-up for the explosive vaporization experiments. The 100 Ω resistor is used as a voltage divider and to measure the current.

- A microscope, (BX60, Olympus, Switzerland) equipped with a water immersion objective (LUMPlan FL40xWI, Olympus, Switzerland).
- A nanosecond flash light source and its driver, (Nanolite KL-K, High Speed Photo-Systeme, Germany). The lamp housing is fixed to the reflected light port on the microscope.
- A CCD camera (CV-M10 Progressive Scan, Stemmer Imaging GmbH, Germany)
- A frame grabber, (PX-510, Stemmer Imaging GmbH, Germany)
- A computer with image acquisition software (Optimas v6.2, Stemmer Imaging GmbH, Germany)

The signals from the digital delay/pulse generator trigger the HP pulse generator (heating pulse) and the flash lamp. The flash lamp is triggered with a given delay with respect to the heating pulse. A sketch of the visualization set-up is depicted in Fig. 7.5.
Figure 7.5: Set-up for pulsed image acquisition. The connections represented with dashed lines are the triggering signals.

The dashed lines represent the connections of synchronizing the heating pulse and the visualization system.

The holder was mounted on the microscope stage (BX60 Olympus). The visualization was made with a long working-distance, water-immersion objective (LUMPlan FL40xWI, Olympus Schweiz AG) combined with an extra 1.6× magnification resulting in a total magnification of ×64.

The light flash (lamp discharge) illuminating the region of interest occurs with a delay with respect to the trigger signal (leading edge). The delay is associated with the time needed to ionize the gasses inside the lamp and in this case was of $4.1 \pm 0.15 \mu s$. The uncertainty is due to the jittering of the flash lamp. The distortion is a marked voltage peak superposed to the signal. This peak occurs at the moment of the maximum illumination intensity and it was used to measure the discharge delay ($4.1 \, \mu s$). The jittering of the lamp (150 ns) was obtained calculating the standard deviation of the discharge delay measured from a series of acquired trigger signals.
The duration of the flash of light is 100 ns, the jittering of the lamp is about 150 ns what gives a temporal resolution of 250 ns. The bubble growth time is considered to be of the order of magnitude of the observed temperature oscillations 500 ns. Because of this we can not obtain clear images of the bubble formation process with this set-up. However the images obtained clearly confirmed the formation of vapor.

7.5 Results and discussion

Device calibration

The relationship between the electrical resistance change and the holder temperature for a representative calibration curve is shown in Fig. 7.6. The electrical resistance of the submicron heater at the corresponding temperature was measured by supplying low-amplitude current pulses (4 µA) and measuring the induced voltage drop. Short pulses of 100 µs were programmed to avoid errors induced by self-heating and the thermal offsets were canceled out by measuring before and during the applied pulses. Using a least-squared fit, the slope of $R-R_0/R_0$ to $T-T_0$, (known as the temperature coefficient of resistance, $\alpha$) was determined. The measured temperature coefficient of resistance of the device is $1.60 \pm 1 \times 10^{-5} \ K^{-1}$ ($3.8 \times 10^{-3} \ K^{-1}$ for bulk platinum). This value of the temperature coefficient of resistance is comparable to the values obtained by other authors (Kim et al., 2001; Zhang et al., 2005). The $\alpha$ value seems to depend strongly on various process parameters such as the sputtering conditions for film deposition and the heat treatment which may affect the crystalline structure of the film. Table 7.1 shows some $\alpha$ values from the literature.

I-V characteristic and temperature rise for different input powers

Fig. 7.7 shows the I-V characteristic obtained in vacuum. The solid line represents the characteristic behavior of a resistor equal to the initial resistance of the device.

The data points represented in the V-I characteristic in Fig. 7.7 can be used to obtain the values of the resistance $R = V/I$, and the temperature can be determined using the
Table 7.1: Thermal coefficient of resistance ($\alpha$) for thin film platinum

<table>
<thead>
<tr>
<th>$\alpha$ [$K^{-1}$]</th>
<th>Fabrication Method</th>
<th>Thickness</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00199</td>
<td>sputtering</td>
<td>100 nm</td>
<td>present work</td>
</tr>
<tr>
<td>0.00329</td>
<td>sputtering</td>
<td>206 nm</td>
<td>Kim et al. (2001)</td>
</tr>
<tr>
<td>0.0016</td>
<td>EBPVD</td>
<td>100 nm</td>
<td>present work</td>
</tr>
<tr>
<td>0.0014</td>
<td>EBPVD</td>
<td>28 nm</td>
<td>Zhang et al. (2005)</td>
</tr>
<tr>
<td>0.003925</td>
<td>high purity wire</td>
<td>bulk</td>
<td>-</td>
</tr>
</tbody>
</table>

Figure 7.6: Measured relative resistance change for different heater temperatures and linear fit for the calculation of the thermal coefficient of resistance, $\alpha$.

expression:

$$\Delta T = \frac{R - R_0}{R_0 \cdot \alpha}$$  \hspace{1cm} (7.2)

The input power is $V \times I$. The volume-averaged temperature rise in the heater as a function of the input power is shown in Fig. 7.8.

Because the mass of the heater is negligible the total heat transfer rate from the heater is composed of: 1. Conduction to the substrate on which the heater is fabricated. 2. Heat transfer to the liquid above the heater and 3. Radiation from the heater. The calculated
Figure 7.7: V-I characteristic of a chip of initial resistance $R_o=13.60 \, \Omega$. The line shows the characteristic of a resistor.

By neglecting radiation and considering at a given heater temperature the total heat transfer rate in vacuum ($q''_{tv}$) and the total heat transfer rate when the heater is loaded with the fluid ($q''_{tl}$), we can eliminate the heat dissipated to the substrate and write:

$$q''_l = q''_{tl} - q''_{tv}$$  \hspace{1cm} (7.3)

where $q''_l$ is the heat transfer rate to the fluid.

Using the values depicted in Fig. 7.8 and Eq. 7.3 the heat transfer rate to the liquid for a temperature of the heater of 250°C turns out to be 10% of the total electrical power input to the heater. This evaluation of the heat transfer rate to the liquid is useful to compare our results with other systems used to generate boiling as suspended metallic wires. Heat transfer rates to the fluid in our experiments are thus of the order of 700 MW/m².
7.6 Temperature measurement during pulsed heating

For the pulse heating experiments the electric pulses were supplied to the circuit sketched in Fig. 7.4. The average temperature of the heater was obtained using Eq. 7.1. The shape of the temperature curve shifts with the current amplitude of the supplied heating pulse. A representative temperature curve is depicted in Fig. 7.9. The origin in the time scale is set at the leading edge of the heating pulse (heating onset). As the temperature measurement is based on acquiring $I(t)$ and $V(t)$ we can only measure temperature during the time when the heating pulse is on.

The average temperature of the heater increases very fast at the origin (the transient) until it reaches a stable value (the pseudo-stationary temperature, $T_{ps}$). In the case represented in Fig. 7.9 the duration of the transient is 2 $\mu$s and the pseudo stationary temperature is 250°C. We define a rate of temperature rise for each experiment. This rate of temperature rise is defined as the ratio of the pseudo-steady temperature, $T_{ps}$ to
Figure 7.9: A representative temperature evolution curve showing the fast temperature increase till reaching the pseudo-stationary temperature. The kink in the temperature evolution indicates the boiling incipience.

the time duration of the transient. If the pseudo-steady temperature is high enough, after few microseconds an abrupt temperature increase is observed. In the example in Fig. 7.9 the kink occurs 6.4 µs after heating onset. This abrupt temperature increase is associated with the formation of vapor on the top of the heater, (Avedisian et al., 1999; Glod et al., 2002). When the vapor forms on top of the heater, it temporarily insulates the heater and as a consequence, the average temperature in the heater increases. We define the boiling incipience temperature $T_b$ and the boiling incipience time $t_b$ as the temperature and time at which the kink in the temperature evolution curve occurs.

After this first abrupt temperature increase (40 K temperature increase in about 0.5 µs), the temperature starts to oscillate around the pseudo-stationary temperature. The oscillation is maintained for approximately 40 µs (not shown in Fig. 7.9). More details about this oscillatory phenomenon will be provided in the following section.

Figure 7.10 shows a temperature evolution curve for an experiment with higher current amplitude (> 26 mA). In this example the rate of temperature rise is very high and a kink in the temperature curve can be observed already during the transient. This
means that the boiling incipience temperature, $T_b$ is reached before reaching the stationary temperature. For the cases following this pattern (depicted in Fig. 7.10), the pseudo-steady temperature will be defined as the value around which the temperature oscillates and the rate of temperature rise as the quotient of the boiling incipience temperature divided by the boiling incipience time.

Figures 7.11 and 7.12 depict the temperature evolution curve for different input current amplitudes. We started supplying 15.42 mA to the device so that the average temperature of the heater elevates to 114 °C, (Fig. 7.11(a)). The temperature reaches the steady state in 13 µs. No boiling was observed even for longer pulse duration (up to 300 µs). The same plot depicts the temperature measured for a current supply of 20.05 mA. The heater reaches an average temperature of 167°C but no boiling is observed in this case neither for a 150µs pulse duration.

For a current supply of 24.2 mA (Fig. 7.11(b)) the average temperature of the heater increases till 256°C. The average temperature stays constant till 72 µs. Then an abrupt increase of temperature is observed.
Explosive vaporization on submicron strip heaters

Increasing the supplied current from by 1% shortens the delay time from 70\(\mu\)s to 20\(\mu\)s (see Fig.7.11(c)). Further increasing the current supply by 1% (to 24.78 mA) reduces the boiling incipience time to 6.5 \(\mu\)s. In these tests the heating pulse duration was
7.6 Temperature measurement during pulsed heating

Figure 7.12: Temperature of the heater as a function of time for different heating pulses. Part II progressively reduced in order to minimize the stress in the heater.

In the curves shown in Fig. 7.12, boiling incipience occurs before reaching the pseudo-
Explosive vaporization on submicron strip heaters

Table 7.2: Summary of the results with water at room temperature

<table>
<thead>
<tr>
<th>Current [mA]</th>
<th>Pulse duration [$\mu$s]</th>
<th>$T_{ps}$ [$^\circ$C]</th>
<th>Rate of temp. rise [$K/s$]</th>
<th>$T_b$ [$^\circ$C]</th>
<th>$t_b$ [$\mu$s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.4</td>
<td>300</td>
<td>114</td>
<td>6.4 $\times 10^6$</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>20.05</td>
<td>150</td>
<td>167</td>
<td>14 $\times 10^6$</td>
<td>256</td>
<td>72</td>
</tr>
<tr>
<td>24.2</td>
<td>90</td>
<td>256</td>
<td>22 $\times 10^6$</td>
<td>260</td>
<td>13.6</td>
</tr>
<tr>
<td>24.49</td>
<td>40</td>
<td>260</td>
<td>27 $\times 10^6$</td>
<td>255</td>
<td>6.5</td>
</tr>
<tr>
<td>24.78</td>
<td>20</td>
<td>255</td>
<td>71 $\times 10^6$</td>
<td>252</td>
<td>2.015</td>
</tr>
<tr>
<td>25.05</td>
<td>10</td>
<td>269</td>
<td>111 $\times 10^6$</td>
<td>258</td>
<td>1.572</td>
</tr>
<tr>
<td>25.36</td>
<td>5</td>
<td>269</td>
<td>147 $\times 10^6$</td>
<td>260</td>
<td>0.91</td>
</tr>
<tr>
<td>26</td>
<td>3</td>
<td>283</td>
<td>257 $\times 10^6$</td>
<td>266</td>
<td>0.609</td>
</tr>
<tr>
<td>26.66</td>
<td>4</td>
<td>305</td>
<td>394 $\times 10^6$</td>
<td>257</td>
<td>0.436</td>
</tr>
<tr>
<td>27.43</td>
<td>4</td>
<td>327</td>
<td>530 $\times 10^6$</td>
<td>288</td>
<td>0.321</td>
</tr>
<tr>
<td>29.42</td>
<td>2</td>
<td>389</td>
<td>815 $\times 10^6$</td>
<td>313</td>
<td>0.29</td>
</tr>
<tr>
<td>30.33</td>
<td>2</td>
<td>421</td>
<td>988 $\times 10^6$</td>
<td>313</td>
<td>0.29</td>
</tr>
</tbody>
</table>

steady state. In the last two figures: (7.12(d) and (e)) the kink identifying the boiling incipience is less marked and we used the inflection of the curve to determine the boiling incipience temperature and time.

In Fig. 7.12(e) boiling was observed 0.28 $\mu$s after starting the heating. The average temperature around which the temperature oscillates increases considerably for the cases represented in Fig. 7.12(d) and (e). Table 7.2 summarizes the results shown in Fig. 7.11 and 7.12 indicating the current supplied, the duration of the pulse, the obtained pseudo-steady temperature, the rate of temperature rise and the boiling incipience temperature and time.

The initial heat transfer rate to the liquid for each case can be estimated by computing the total power supply: $R_o \times I^2$ and dividing it by the nominal dimensions of the chip (0.5 $\mu$m $\times$ 3$\mu$m). As it was discussed previously for an average temperature of 250 $^\circ$C only a 10% of the total power dissipated is available for heating the fluid (the other 90% is transferred to the substrate). This gives heat transfer rates to the liquid ranging from 530 MW/m² to 830 MW/m².
7.6 Temperature measurement during pulsed heating

Figure 7.13: Jittering effect: Three different recorded temperature for the same supplied current 24.2 mA. The kink in the curve appears at 72 µs, 59 µs and 80.5 µs respectively.

Table 7.3 contains values of boiling incipience temperature obtained by other authors. In the works by Deng et al. (2004, 2006), the boiling incipience was detected by a ‘V’ shape temperature change. The last row refers to the device used by Avedisian et al. (1999). The rate of temperature rise measured for this device was $2.5 \times 10^8$ K/s, which is one of the highest values obtained in pulsed heater experiments. In the present work a maximum rate of temperature rise of $9.88 \times 10^8$ K/s was reached. This very high value of the rate of temperature rise achieved is due to the small dimensions of this heater compared with previous studies.

Several measurements were performed for the same electrical pulse to check the repeatability. In Fig. 7.13 three different temperature evolution curves for the same heating pulse are plotted. The boiling incipience time varies several µs from case to case. Figure 7.14 shows three different curves of temperature evolution for 24.7 mA. Fig. 7.15 shows the temperature evolution for 25.3 mA. Those figures can be compared with the previous case in Fig 7.13. It is clear that this jitter effect is lower for high current levels;
Figure 7.14: Jittering effect: Three different recorded temperature for the same supplied current 24.7 mA. The kink in the curve appears at 6.5 µs, 6.8 µs and 6.05 µs respectively.

<table>
<thead>
<tr>
<th>Feature size $[\mu m^2]$</th>
<th>Pulse duration</th>
<th>$T_b$ [°C]</th>
<th>Rate of temp. $[K/s]$</th>
<th>$t_b[\mu s]$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5×0.5</td>
<td>1.66 ms</td>
<td>245</td>
<td>$16 \times 10^6$</td>
<td>51</td>
<td>Deng et al. (2004)</td>
</tr>
<tr>
<td>1×0.5</td>
<td>1.66 ms</td>
<td>207</td>
<td>$12.1 \times 10^6$</td>
<td>60</td>
<td>Deng et al. (2004)</td>
</tr>
<tr>
<td>2×1</td>
<td>1.66 ms</td>
<td>214</td>
<td>$10^6-10^7$</td>
<td>60</td>
<td>Deng et al. (2006)</td>
</tr>
<tr>
<td>65×65</td>
<td>5 µs</td>
<td>283</td>
<td>$2.5 \times 10^8$</td>
<td>0.7</td>
<td>Avedisian et al. (1999)</td>
</tr>
</tbody>
</table>

Table 7.3: Boiling incipience temperature obtained in other works with microheaters.

This means that an increase in current level makes the process more predictable. Fig. 7.16 summarizes the obtained $t_b$ for all the cases listed in table 7.2. It can be seen that for higher values of current, the time at which boiling begins is shorter and the jittering disappears.
Figure 7.15: Jittering effect: Three different recorded temperature for the same supplied current 25.3 mA. The kink in the curve appears at 1.56 $\mu$s, 1.55 $\mu$s and 1.55 $\mu$s respectively.

Figure 7.16: Jittering effect: The bar for each point represents the dispersion in the measured boiling inception time, $t_b$. 
7.7 Frequency analysis of the oscillations of the temperature evolution

In the previous cases, the temperature oscillations, once started, were maintained until the end of the pulse. In order to find out how long this oscillation exists, we performed a series of experiments by applying longer heating pulses. The general trend is shown in Fig. 7.17. After boiling incipience, the temperature starts to oscillate at high frequency (MHz) for about 40 µs. The amplitude of the oscillation decays in time but the mean temperature slowly increases reaching a final temperature slightly above the pseudo-steady temperature.

To analyze the frequencies contained in the signal, Fast Fourier Transform (FFT) was used. We extracted the data points of the three regions of the signal marked in Fig. 7.17 as A, B and C. The result of the FFT analysis is presented in Fig. 7.17 (c). The time interval considered for the analysis is indicated on the top of each plot. There exists a distinct main frequency component in each region. The main frequency component for each region is indicated in the figure.

The influence of the heating pulse amplitude on the frequency of the temperature oscillation was studied for a heater of $R_o = 16.85\Omega$. The temperature evolution for different heating pulses of sufficiently long duration are presented in Fig. 7.18. The current supply was 18.97 mA for (a, b), 19.9 mA for (c, d), 20.34 mA for (e, f) and 21.19 mA for (g, h). It can be seen in the region A, that the main frequency increases as the current amplitude increases.

7.8 Reason for bubble oscillations

The previously described oscillation does not occur when the experiments are performed without liquid neither when the current pulse is not intense enough to induce vapor formation. Because of this we conclude that the oscillation observed in the temperature evolution curve is a boiling induced phenomenon. A plausible explanation behind this phenomenon can be found by considering the difference between two bubble growth scenarios: 1. growth in initially uniform superheated liquid and 2. growth in the electrically
7.8 Reason for bubble oscillations

Figure 7.17: A temperature evolution curve recorded for a heating pulse of 70 µs duration. After the boiling incipience (first abrupt temperature increase) the temperature starts to oscillate. The oscillation stays for about 40 µs decaying in time. (b) Close-up on the oscillating part of the curve. (c) Fast fourier transform was applied to analyze the oscillation in three regions A, B and C.

heated system. A vapor bubble formed in the superheated liquid will grow absorbing heat from the surrounding liquid because the liquid temperature gradient in radial direction has a positive slope, (the heat transfer by conduction between the vapor and the liquid phase is towards the vapor phase, sustaining the growth of the bubble). In the second
Figure 7.18: Temperature of the heater as a function of time for different heating pulses and frequency analysis of the temperature oscillation in different regions of the signal scenario the vapor covers the heated surface and energy is supplied to the bubble interface by conduction through the vapor phase. The temperature gradient in the liquid has a
negative slope because the liquid away from the hot surface is colder than in the vapor. Therefore conduction heat transfer in the liquid goes in the direction of cooling down the interface (liquid side). For certain bubble sizes because the heat removal is larger than the heat input. This makes the bubble to shrink and to re-grow again[7.19]. Similar behavior was predicted in a simulation of vapor bubble growth around heated particles (Dietzel and Poulikakos 2006).

Effect of initial temperature of the liquid

The effect of the initial temperature of the liquid was investigated. To heat the liquid, the chip holder was completed with an aluminium block with a drilled hole to insert a heating element. The aluminium block is fabricated to fit underneath the ceramic carrier (see Fig. 7.20). A dc source (TTi EX344T) supplies the necessary power to the heater element. The temperatures in the aluminium block under the carrier and in the liquid above the heater were monitored using two thermocouples (Type K). The temperature of the liquid near the heater will be obtained by averaging the two thermocouple measurements. The error in determining this temperature is ±6 °C.

The same heating current (18.6 mA) was supplied for three different bulk liquid temperatures: 1) Room temperature, 2) 50°C and 3) 80°C. The results are depicted in Fig.
Figure 7.20: Photo showing the chip holder used to perform experiments at different initial temperature of the liquid.

The temperature at boiling incipience is 263.2 °C, 243.9 °C and 261.9 °C respectively. The boiling incipience takes place at 17.19, 0.75 and 0.4 µs after initiating the electrical heating, respectively. The temperature difference between the heater and the liquid at boiling incipience is 243 °C, 193°C and 180 °C respectively. The temperature oscillation previously observed at room temperature was still present at 50 °C bulk liquid temperature, but it was not observed when the liquid temperature was set to 80°C (see Fig.7.21).

This arrangement for increasing the temperature of the liquid does not allow an homogeneous liquid bulk temperature: the droplet of liquid is hot at the bottom but at the air-liquid interface the temperature of the liquid is close to the ambient temperature. We will assume that near the heater the temperature variations are negligible compared to the thermal gradients generated during pulsed heating. We believe that the limitations of the set-up do not affect the qualitative result, that is to proof that the oscillation disappears when the liquid is hotter confirming the explanation given to the observed oscillation.
Figure 7.21: Temperature evolution curve for experiments performed with three different initial liquid temperatures: room temperature, 50°C and 80°C.

(b) FFT of the temperature oscillation measured at ambient temperature. The frequency in the abscissa axis is in Hz.

(c) FFT of the temperature oscillation measured at 50 °C. The frequency in the abscissa axis is in Hz.
7.9 Submicron bubble visualization

A vapor bubble was observed on the heater by synchronizing the heating pulse with a flash lamp. The photo in Fig. 7.22 shows the heater before the heating to compare with the images of the excited heater. Fig. 7.23(a-d) shows images acquired with the heating on. We recorded together with the image the trigger signal and the heater response (voltage drop). The instant (or phase in the oscillation) to which the frozen image corresponds is indicated on the temperature evolution plot on the top of Fig. 7.23. The picture shown in Fig. 7.23(a) corresponds to an instance 1 \( \mu s \) after boiling incipience. The second image corresponds to 2 \( \mu s \) after the first perturbation (b). The third one was taken close to the end of the oscillations (d). The fourth image was taken once the oscillations stopped (e) and it shows that a bubble is located on the center of the heater. The bubble stayed till the end of the heating pulse.

![Figure 7.22: A picture of the heater before supply the heating pulse.](image)

7.10 Conclusions

A method based on ion beam milling was applied to fabricate a thin film platinum heater of 0.5 \( \mu m \times 3 \mu m \). The micro/nanofabricated structure was used as both heater and temperature sensor.

Short electrical pulses were used to increase the heater temperature in contact with liquid water. We measured voltage drop across the heater and the current supplied and calculated the instantaneous temperature (spatially averaged) of the heater. The temperature evolution showed a very fast transient followed by a temperature plateau. If the
temperature at the plateau, or pseudo-steady temperature, is high enough after a certain time a perturbation consisting in a sudden increase of temperature takes place indicating the formation of vapor on the heater. The effect of current intensity in the boiling incipience temperature and on the rate of temperature rise was studied.

An unexpected oscillation in the temperature curve was observed. The frequency of the
oscillation is in the order of MHz. It could be associated to the effect of the strong thermal gradient in the vicinity of the heater. Moreover the effect of the initial temperature of the liquid was studied showing that the oscillation disappeared when the initial temperature of the liquid is increased. This result corroborate the observations by [Osborne (1947)](https://doi.org/10.1080/00018734700101240) what confirm the fact that the temperature oscillation observed is induced by a boiling related phenomenon.

Visualization of vapor on the heater was possible using a pulsed visualization system where the frame acquisition and illumination system is synchronized with the heating pulse. The temporal resolution of the visualization system does not allow to visualize the changes associated to the temperature oscillations.
Chapter 8

Conclusions and outlook

The fabricated platinum microheater subjected to pulsed heating reach high temperatures in few microseconds. The rates of temperature rise obtained were as high as $70 \cdot 10^6 K \cdot s^{-1}$. An inflection in the temperature evolution curve can be used to identify the time and temperature associated with the formation of vapor on the top of the heater. As observed by Glod et al. (2002); Skripov (1974); Avedisian et al. (1999) the maximum nucleation temperatures increases with the rate of temperature rise. The maximum nucleation temperature measured in the present work was 273 °C, thus below the maximum superheat for water at atmospheric pressure. The low nucleation temperatures measured in the present study may be a result of the microheater geometry and structure which induce current density inhomogeneities. This current density gradients may induced temperature gradients.

The temperature evolution of the microheater under pulsed heating was investigated for the confined-liquid system. The measurements using the confined system did not reveal differences in the temperature evolution and in the nucleation temperature measured as it was predicted in the analytical work by Carey (2000). We did not observe a corresponding increase in the obtained nucleation temperature may be because the enclosure is too flexible. In that case the increase in pressure may be too small to significantly modify the nucleation temperature. It was also shown that for two different microheater devices of different initial resistances, a similar temperature evolution can be induced when supplying a similar initial power (rather than similar current amplitude).
Visualization of explosive vaporization on top of thin film platinum microheaters were achieved by means of a pulsed image acquisition system using flash lamp illumination. We could associate the temperature measurements with the images, confirming that the kink in the curve used to measure the nucleation temperature coincides with the first moment when vapor covers the entire surface of the microheater. Visualization of explosive vaporization through a PDMS fluidic chip was also achieved and the confinement did not affect the bubble growth but it accelerated the bubble collapse process.

**Explosive vaporization and microbubble oscillations on strip heaters**

A method based on Ion Beam Milling was applied to fabricate a thin film platinum heater of $0.5 \mu m \times 3\mu m$ size. The microfabricated structure can be used as both, the heater and the temperature sensor. The set-up for characterization and calibration of the heater was presented and the I-V characteristics of the heater measured. The thermal coefficient of resistance of the heater is $0.0016 \text{ K}^{-1}$.

Short electrical pulses were used to increase the heater temperature in contact with liquid water. We measured the voltage drop across the heater and the supplied current and we calculated the instantaneous temperature (spatially averaged) of the heater. The temperature evolution showed a very fast transient followed by a temperature plateau. If the temperature at the plateau, or pseudo-steady temperature, is high enough, after a certain time a perturbation consisting in a sudden increase of temperature takes place indicating the formation of vapor on the heater. The effect of current intensity in the boiling incipience temperature and on the rate of temperature rise was studied.

An unexpected oscillation in the temperature curve was observed. The frequency of the oscillation is in the order of the MHz. It could be associated with the effect of the strong thermal gradient in the vicinity of the heater. Moreover the effect of the initial temperature of the liquid was studied showing that the oscillation disappeared when the initial temperature of the liquid is increased. This result corroborates the observations by Osborne (1947) what confirms that the observed temperature oscillation is induced by a boiling related phenomenon.

Visualization of vapor on the heater was possible using a pulsed visualization system where the frame acquisition and illumination system were synchronized with the heating
The fabricated device allowed good visualization of the explosive vaporization phenomena but it had the disadvantage that the pressure inside the cavity could not be monitored.

The initial design included integrated piezoresistive elements to sense deformation of a membrane made of low stress silicon nitride (as in some commercial pressure sensors). The fluidic part of the chip had to be fabricated using silicon, and the channels and the cavity fabricated by wet etching (KOH). In the first prototypes it was observed that the membranes should be fabricated with a large area to allow the visualization of the heater but this was not feasible for mechanical integrity reasons. An other solution that could be explored is to fabricate the channels and the cavity in an intermediate layer made of SU8. The channels and cavity could then be sealed with a second layer of SU8 where the pressure sensors could be fabricated [Agirregabiria et al. 2005].

The system used to visualize the phenomena could be improved with a different illumination source. During the experiments it was clear that the duration of the light pulses (100 ns) causes blur in the quality of the images. Other authors achieved better temporal resolution (30 ns ) applying pulsed diode laser as the illumination source [Hong et al. 2004]. Avedisian et al. (2006) achieved 5 ns temporal resolution using a pulsed laser for illumination. They needed to replace the internal lenses of the microscope illumination port by coated lenses that could withstand the laser power without cracking. Especially to achieve the visualization of the vapor size oscillations postulated in the last chapter it would require pulsed laser illumination.

For the experiments using the submicron heater, it could be interesting to investigate the effect of the heater dimension on the temperature oscillation. Additionally, pressure measurements of the type presented by Glod et al. (2002) could be used to assess the potential applications of the vapor oscillation. Measuring the generated acoustic power would require a very high frequency response sensor element. Using a different light source would be important to improve the temporal resolution of the pulsed image acquisition.
system. This would allow observing the evolution of the bubble during the high frequency temperature oscillations. To assess the effect of the initial temperature of the liquid, a more sophisticated system to heat the liquid would allow a more homogeneous temperature distribution and also a better control of the initial temperature of the liquid.
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