Study of femtosecond laser pulse drilling of silicon

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2014

Jiao, L. (2014). Study of femtosecond laser pulse drilling of silicon. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/62050>

<https://doi.org/10.32657/10356/62050>

Study of Femtosecond Laser Pulse Drilling of Silicon

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A thesis submitted to the Nanyang Technological University in partial
fulfilment of the requirements for the degree of

Doctor of Philosophy

2014

Abstract

Femtosecond (fs) laser drilling has been proved to be an efficient tool to drill various materials. In the drilling process, however, there are constant needs to eliminate spatter, control the taper, increase the aspect ratio and material removal rate. In addition, it is necessary to further understand the laser material interaction mechanism. In this thesis, some new features were proposed into the femtosecond laser drilling area in order to overcome these problems.

In this study, the effect of six alcohol liquids on the fs laser drilling process was evaluated and compared. The relation between the volatility of the liquids and the material removal rate of laser drilling was found. A more volatile liquid could assist better in carrying away the debris and allow more laser energy to reach the ablation front. The material removal efficiency was increased by 40% when applying methanol as the assist liquid.

The spatial wavelength of laser induce period surface structure (LIPSS) as fabricated in ethanol was found to be 455 nm which is about 40% less than that in air (772 nm). This finding could improve the applicability of LIPSS as optical gate and catalyst support.

A systematic assessment of geometry evolution of the laser drilled through hole at different substrate temperatures was conducted. The result suggested that the entrance hole diameter was increased by 25% while the exit hole was increased by 30% when the substrate temperature was increased to 900 K. The laser drilling efficiency was also greatly increased

by elevating the substrate temperature. This high drilling efficiency was attributed to the enhanced laser energy absorption of silicon wafer and thereafter wave guiding effect. The spatter area was found, however, to be continuously decrease with increasing the substrate temperature. This study provided useful knowledge for better understanding the fs laser energy absorption of silicon wafer at higher temperature.

In order to further study the hole wall's effect on the laser beam propagation inside the micro-hole during the laser drilling, a theoretical analysis was conducted by numerically solving the time harmonic Maxwell's wave equation. The taper angle of micro-hole was varied to investigate its effect on the laser energy distribution at the bottom of the hole. The ellipse entrance hole shape and zonal structure at the hole bottom which were observed in the previous experiment were reasonably explained by using the laser intensity distribution as obtained in the present model.

Acknowledgements

Firstly, I would like to take this opportunity to express my deepest gratitude to my supervisors, Associate Professor Eddie Ng Yin-Kwee and Dr. Zheng Hongyu of Singapore Institute of Manufacturing Technology. Their invaluable technical guidance, inspiring advising and mental support helped me to overcome the difficulties and accomplish the thesis.

I would also like to thank Associate Professor Vladimir Vladimirovich Kulish and Associate Professor Leong Kai Choong. I benefitted a lot from their criticisms and advice during my confirmation defense. It was a great lesson for me.

I am also grateful to Dr. Wee Lee Mein who provided a lot of assistance at the beginning of this project. I would also like to convey my thanks to Dr. Hamid Farrokhi. I learned many useful knowledge from the discussion with him. It's a pleasure to express my thank to my friends in NTU: Kathic, Jamil, Taureze, Liu Dan, Jin Xianfei. I have received considerable help from them.

Finally, I want to show my greatest appreciation to my family for their long-term and priceless support, encouragement and understanding.

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Nomenclature

A	Heat source term (W/m^3)
A_l	Area of the focused spot (mm^2)
C_e	Heat capacity for electrons ($\text{J}/\text{K} \cdot \text{m}^3$)
C_i	Heat capacity for ions ($\text{J}/\text{K} \cdot \text{m}^3$)
c	Heat capacity ($\text{J}/\text{K} \cdot \text{m}^3$)
c_0	Optical speed (m/s)
D	Laser beam diameter (mm)
$D_{duration}$	Pulse duration (s)
d	Hole depth (μm)
$d_{entrance}$	Entrance hole diameter (μm)
d_{exit}	Exit hole diameter (μm)
E_{pulse}	Pulse energy (μJ)
$\vec{E}$	Electric field (V/m)
E_x	Component of electric field in x direction (V/m)
E_y	Component of electric field in y direction (V/m)
E_z	Component of electric field in z direction (V/m)
E_0	Peak power of the laser pulse (mW)

f	Focal length (mm)
Δf	Depth of focus (μm)
G	Electron-lattice coupling coefficient ($\text{W/K}\cdot\text{m}^3$)
$\vec{H}$	Magnetic field (A/m)
I	Laser intensity (W/cm^2)
k_0	Wave number of free space (1/m)
$k_{\text{imaginary}}$	Refractive index, imaginary part
k_e	Thermal conductivity for electrons ($\text{W/K}\cdot\text{m}$)
k_i	Thermal conductivity for ions ($\text{W/K}\cdot\text{m}$)
k	Thermal conductivity ($\text{W/K}\cdot\text{m}$)
n	Refractive index
n_1	Refractive indices of medium 1
n_2	Refractive indices of medium 2
n_{silicon}	Refractive index of silicon
$\hat{n}$	Unit normal vector
P_{average}	Average power (mW)
P_s	Saturated vapour pressure (N/m^2)
P_r	Recoil pressure (N/m^2)
$\dot{Q}_{\text{out},1}$	Outward energy flows by time averaging at boundary 1 (W)

$\dot{Q}_{out,2}$	Outward energy flows by time averaging at boundary 2 (W)
$R_{repetition}$	Repetition rate (1/s)
R	Reflectance
r_0	Gaussian beam radius (mm)
s	Energy source (W/cm ²)
$\vec{S}$	Poynting vector (W/cm ²)
T	Transmittance
T_e	Electron temperature (K)
T_i	Ion temperature (K)
t	Material thickness (μm)
w_0	Radius of the laser beam (μm)
Z_f	Position of work-piece surface in the vertical direction (mm)
ΔZ_1	Thermal expansion distance of the alumina refractory (mm)
ΔZ_2	Elevated distance for focus plane (mm)
α	Thermal diffusivity (mm ² /s)
ε_1	Complex dielectric constant of material
ε_2	Complex dielectric constant of medium
ε_0	Electric permittivity (F/m)
ε_r	Relative permittivity

η	Real part of the effective refractive index of interface for surface plasma
θ	Taper angle (degree)
θ_i	Incident angle (degree)
θ_r	Refractive angle (degree)
θ_d	Divergence angle (degree)
$\theta_{critical}$	Critical taper angle (degree)
λ	Wavelength of the laser (nm)
Λ	Period of surface ripples (nm)
μ_0	Magnetic permeability (H/m)
μ_r	Relative permeability
ρ	Density of the material (kg/m ³)
σ	Electrical conductivity (S/m)
ω	Frequency of laser (1/s)

Glossary of Terms

1-D	One Dimensional
2-D	Two Dimensional
3-D	Three Dimensional
AFM	Atomic Force Microscope
BCs	Boundary Conditions
CCD	Charged Coupled Device
CNC	Computer Numerical Control
CW	Continuous Wave
DMSO	Dimethyl Sulfoxide
DUV	Deep Ultra Violet
EDX	Energy Dispersive X-ray
EVP	Enthalpy of Vaporization
FDM	Finite Difference Method
FDTD	Finite Difference Time Domain
FEM	Finite Element Method
FS	Femtosecond
FVM	Finite Volume Method
FWHM	Full Width at Half Maximum

HAZ	Heat Affected Zone
HF	Hydrofluoric Acid
IPA	Isopropyl Alcohol
LIPSS	Laser Induced Period Surface Structure
MLI	Maximum Laser Intensity
MS	Millisecond
Nd:YAG	Neodymium-doped Yttrium Aluminum Garnet
NOP	Number of Pulses
NS	Nanosecond
PIA	Pixcavator Image Analysis
PS	Picosecond
SEM	Scanning Electron Microscopy
TAB	Tape Automated Bonding
TEM	Transverse Electromagnetic
TTM	Two Temperature Mode

Chapter 1 Introduction

1.1 Background

Since the invention of the laser in 1960s, it has found diverse applications in engineering and industry. The ultra-short laser pulses with a large amount of energy have been proved to be an efficient way of processing a wide range of materials including metals (Furusawa et al. 1999), polymers (Kumagai et al. 1994), ceramics (Ihlemann et al. 1995) and silicon (Jiao et al. 2011, Jiao et al. 2013), some of which are hard to process by the mechanical method. The main advantages of ultra-short pulses laser processing over mechanical processing have been its high precision (Chichkov et al. 1997), better repeatability (Ng and Li 2001), and small kerf width (Sudani et al. 2011) with a very narrow heat affected zone (HAZ) (Le Harzic et al. 2002).

Silicon is the typical semiconductor material used to create most integrated circuits applied in electronics industries. The nonlinear absorption (Moison et al. 1983) of the ultra-short pulse laser energy enables precise drilling of hard and brittle materials such as silicon, which is a challenging task by conventional mechanical drilling methods. In microelectronic and solar cell applications (Halbwax et al. 2008), drilling of consistent

micro-holes in silicon is an important manufacturing step. The femtosecond (fs) pulse laser has been demonstrated as a potential tool for such effective drilling applications.

1.2 Motivation

Although the process of laser machining of silicon appears to be attractive, it however has not been very successful to date. There are some challenges encountered in laser drilling of silicon.

The spatter is formed by the redeposition of molten or sublimation materials which are the by-products of laser drilling. This spatter around the entrance hole boundary is hard to avoid. The traditional post-processing cleaning technique is ineffective and costly for removing small particulates. There is a constant need to develop more efficient techniques to minimize debris formation during laser processing (Zheng and Jiang 2010).

In addition, it is true that there are many advantages of fs laser over the long pulse laser, however, the average output power of fs laser is relatively small (Schulz et al. 2013) (below 1W). This can limit the material removal rate of fs laser. It is essential to conduct some research to further improve the ablation efficiency for fs laser.

Taper shape of the micro-hole drilled by laser is commonly formed due to the Gaussian distribution of the laser energy and the following reflection of laser beam. The taper angle of micro-hole can affect the flow rate in the microfluidic devices (Martanto et al. 2005).

There are also some needs to control the hole taper to efficiently deposit the copper diffusion barrier and copper seed metal on the hole wall in semiconductor industry (Bär et al. 2002, Figueroa et al. 2005). To explore the application area of the laser drilling, it is of great importance to conduct some experiments to find a reliable technique to control the taper angle of the laser drilled hole.

Because of the electromagnetic nature of the laser and the small scale of the micro-hole, the laser beam propagation behaviour that affected significantly by the hole wall's interaction with the laser pulse is very complex and unclear so far. The in-situation experimental observation of laser beam propagation inside the micro-hole study is very challenging because of the lack of fast-response equipment. In order to better understand the laser drilling mechanism, it is necessary to conduct a numerical study to investigate the laser propagation behaviour inside the micro-hole.

1.3 Objectives and scope of the work

The objectives of this project are:

1. To find a reliable approach that can apply liquid film with thickness less than 500 μm during the laser processing, compare the spatter area, taper angle, material removal rate of the hole drilled in various liquids which have lower surface tension than water and are more volatile in nature.

2. To develop a preheating setup that can provide steady temperature for sample during the laser drilling process, evaluate the influence of the enhanced optical absorption of the laser energy at the elevated substrate temperature on the hole geometry and the spatter area around the drilled hole, investigate the laser material interaction mechanism at elevated temperature.
3. To develop a numerical model to solve Maxwell's wave equations in micro hole, vary the taper angle of hole wall to investigate the influence of multi-reflection and thereby interference of the laser beam on the laser energy distribution at the bottom of the micro hole in silicon.

The scope of this thesis is limited to the investigation of micro-hole smaller than 100 μm and micro surface structure on silicon wafer ablated by fs laser with fixed repetition rate of 1000. The percussion drilling technique is chosen due to its high drilling speed.

1.4 Organisation of the thesis

The organisation of the thesis is as follows. In Chapter 2, the fundamental knowledge of laser process material is firstly given. In addition, the laser parameters affecting the hole quality in terms of aspect ratio, taper, barreling, spatter, cracking and heat affected zone are discussed. Then the recent techniques related to laser ablation including liquid assist laser drilling, laser induced period surface structure (LIPSS), preheating laser ablation are

summarised and discussed. The limitations of the previous research for each subject are discussed. At last, the existing research about the theoretical study of laser drilling is presented.

In order to obtain a basic understanding of the fs laser setup using in this thesis, the parametric study is conducted which focuses on the effects of different laser parameters including average laser power, number of pulse, focus position, focus lens on the spatter area and geometry of the drilled hole in silicon wafer. The dependence of entrance hole diameter, exit hole diameter and spatter area on these parameters is discussed in Chapter 3.

In order to overcome the technical problems in the motivation Chapter 1.2, liquid assist method is proposed in Chapter 4. In Chapter 4 some experiments were carried out by comparing various liquid's effects on the amount of debris, taper, drilling efficiency and oxygen concentration of the hole wall. It is found that more volatile liquid can better help carrying away the debris and allow more laser energy to reach the ablation front which leading to a higher drilling efficiency.

During the liquid assisted laser drilling experiment, it is found that the liquid have some influence on the laser induce period surface structures. The LIPSS fabricated in air and in ethanol are compared. The experimental result shows the wavelength of LIPSS is as short as 455 nm which is about 40% less than that in air (772 nm). These contents can be found in Chapter 5.

Other than the liquid assist liquid laser drilling, some experiments are conducted on the temperature of substrate' effect on the taper and spatter area. These contents can be found in Chapter 6. In this research, temperature is varied from room temperature (300K) to 900K.

The result shows that the entrance hole diameter is increased by 25% and the exit hole is increased by 30% when the substrate temperature is increased to 900 K from room temperature. The laser drilling efficiency is greatly increased by elevating the temperature.

The results at different temperatures indicate that the initial hole geometry may have significant influence for drilling behaviour of the successive laser pulses.

In order to further study the material removal mechanism, numerical research is conducted on the laser drilling, which can be found in Chapter 7. The numerical model involves the hole wall's effect on the laser beam propagation inside the micro-hole by numerically solving the time harmonic Maxwell's wave equation. The numerical results show that energy intensity distribution at the hole bottom is significantly influenced by the hole wall when varying the taper angle.

Finally, this study is concluded in Chapter 8. The future research direction to extend the current works is also recommended in Chapter 8.

Chapter 2 Literature Review

2.1 Preamble

In this chapter, the background knowledge of laser drilling including various laser parameters' effects on drilling process is presented. The characterisation for hole quality is given so as to evaluate the laser drilled hole quality. The previous experimental works that focused on the improvement of hole quality are also summarised and discussed. In addition, the recently innovative laser ablation techniques including liquid assisted laser drilling, laser induced period surface structures and pre-heating laser drilling are presented. Furthermore, previous theoretical studies of laser drilling are summarised which involves the heat transfer modelling for both long pulse and short pulse laser. The modelling works related to the laser beam propagation in the drilled hole are provided. Finally, the limitations of the previous works and the research gaps for each section are also discussed.

2.2 Laser parameters that affecting the hole quality

In this section, a review of the literature on the laser drilling process is presented. In the experimental studies, the major parameters being considered include laser pulse energy, pulse duration, focus settings, assist gases, material properties. Efficient utilization of the laser depends upon proper selection and optimisation of all these factors.

2.2.1 Pulse duration and wavelength

The pulse duration ($D_{duration}$) is an important laser parameter because material removal mechanism is different with varying the pulse duration from ns to fs.

Researcher (Stuart et al. 1995) studied the pulse duration's effect on the damage threshold of dielectric material. It was found that the damage threshold decreased consistently with decreasing the pulse duration from 1 ns to 270 fs (Fig. 2.1). At the fs range, the laser irradiation can cause multi-photon ionization which was the main material removal mechanism.

Fig. 2.1 The dependence of damage threshold of fused silica and calcium fluoride on pulse duration (Stuart et al. 1995)

This led to the material damage at lower laser fluence. In the case of ns laser, however, the conventional heating and melting were responsible for the material removal which need high laser fluence.

Furthermore, previous study (Chichkov et al. 1997) proved the advantages of fs laser for precision material processing when comparing the morphology of drilled area by using fs, ps and ns laser as presented in Fig. 2.2. There was no trace of molten layer for fs laser with fluence of 0.5 J/cm^2 . However, the irregular molten trace can be found around the drilled area in the cases of nanosecond (ns) and picosecond (ps) laser.

For ns and ps lasers there are enough time for the heat to propagate into the material and to produce a large molten layer. In these cases the target material are removed both in vapour and liquid phases. On the other hand, damage caused by fs laser pulse is fundamentally different from damage caused by ps and ns laser pulses. For fs laser pulse, the timescale over which the electrons are excited is smaller than the electron phonon relaxation time (1 ps) (Gattass and Mazur 2008). Therefore, a fs laser pulse will end before the electrons can thermally excite any ions. Heat diffusion outside the focal area is minimised, thus increasing the precision of fs laser (Liu et al. 1997).

Fig. 2.2 Hole drilled on steel foil by fs (a), ps (b), and ns (c) lasers (Chichkov et al. 1997)

The wavelength of laser is another important parameter for laser drilling quality. The material optical properties such as absorptivity and reflectivity show sharp variation for lasers with different wavelength. Researchers (Kataoka et al. 2006) investigated the effect of wavelength on machining characteristics of aluminium nitride in micro-drilling using Q-switched Nd:YAG (Neodymium-doped yttrium aluminium garnet) laser. The depth of drilled hole increases with the decrease of wavelength, since photon energy and absorptivity become higher with decreasing the wavelength of laser beam. Besides, the energy loss by the plasma in laser processing decreases with decreasing the wavelength.

Therefore, shorter wavelength is more effective in order to drill the micro-hole with high aspect ratio.

2.2.2 Pulse energy and repetition rate

The pulse energy E_{pulse} is defined as the total optical energy of single laser pulse. It decides the peak power and the spot size of laser. It was observed that the hole diameter (Yokotani et al. 2005) and drilling speed (Johnson and Pfladderer 1989) continually increased with increasing the pulse energy.

The average laser power $P_{average}$ that is a function of pulse energy can be calculated by using Eq. (2.1) (LUMONICS.LTD 2000).

$$P_{average} = E_{pulse} \cdot R_{repetition} \quad (2.1)$$

where $R_{repetition}$ is the repetition rate. The $R_{repetition}$ decides both the laser output power and the time interval between the two successive pulses. The drilling speed can be increased with increasing $R_{repetition}$ at lower repetition rate range. However, when $R_{repetition}$ is so high that the time interval between pulses is larger than the plasma extinguishing time (a few microseconds), the plasma shielding (Chaleard et al. 1998) will then occur and the drilled efficiency will decrease with increasing the repetition rate (Ancona et al. 2009).

2.2.3 Beam profile and pulse shape

The intensity distribution of the laser beam is referred as the laser beam profile. A laser with stable power and low order mode i.e. Gaussian beam, is mostly employed in the drilling process which gives better drilling quality. The beam can be represented exactly by a Gaussian curve of the form as expressed in Eq. (2.2) (LUMONICS.LTD 2000).

$$I(r) = I_0 \exp(-2r^2 / w_0^2) \quad (2.2)$$

where w_0 is the radius of the laser beam at the beam waist, r is the distance from the point to center line position, I_0 is laser intensity.

The influence of fs laser pulse shape on silicon wafer micromachining was investigated in previous study (Cournoyer et al. 2014). The ns fiber laser platform that was based on master oscillator with programmable amplitude waveform was applied to modulate the temporal pulse shape. It was found that the laser with peak power appearing at the beginning of the single pulse was more efficient and resulted in a deeper hole. This phenomenon was attributed to the enhancement of silicon absorptivity at higher temperature. The higher power at the beginning of the laser pulse can lead to suddenly temperature rise which allow more laser energy absorption. This can maximise the laser ablation efficiency.

2.2.4 Assist gases

Laser drilling process depends greatly upon the selection of the type and pressure of the processing gas. Processing gas is commonly used for melt ejection, protection of optics lens and removal of plasma and vapour. However, any misuse of gas may result in a hindering effect on the hole breakthrough and hole formation. It was reported that the assist gas hindered the ejection of molten material by creating a pressure at the surface for most of the hole formation, but then assisted the ejection of the final piece of molten material from the rear of the sample (Rodden et al. 2001). Thus, the type of assist gases must be selected carefully depending on the laser set up and material properties. Previous laser drilling works on various materials involving assist gases are summarised in Table 2.1 (Low et al. 2000, Rodden et al. 2001, Khan et al. 2004, Ghoreishi and Nakhjavani 2008, Okasha et al. 2010, Webster et al. 2010, Choudhury et al. 2012, Biffi and Previtali 2013).

Table 2.1 Summary of assist gases for the laser drilling of various materials

Materials	Assist gases	Laser sources	References
Titanium	Oxygen	Nd:YAG	(Rodden et al. 2001)
Stainless steel 304	Oxygen	Nd:YAG	(Ghoreishi and Nakhjavani 2008)
Stainless steel 316L	Air or Oxygen	Nd:YAG	(Khan et al. 2004)
Nimonic 263 alloy	Oxygen, Nitrogen or argon	Nd:YAG	(Low et al. 2000)
In718 alloy	Oxygen, Nitrogen or Compressed air	Nd:YAG	(Okasha et al. 2010)
Titanium	Helium or argon	Fiber laser	(Biffi and Previtali 2013)
Stainless steel 304	Oxygen	Fiber laser	(Webster et al. 2010)
Silicon steel	Nitrogen	CO ₂	(Lei et al. 2009)
PMMA	Compressed air	CO ₂	(Choudhury et al. 2012)

The shape and size of the nozzle play an important role in gas assisted laser drilling. Khan et al. investigated the assist gases' influence on the material removal rate and drilled hole profile by applying the ns laser and micro-supersonic air and oxygen jets. (Khan et al. 2004, Khan et al. 2007). The nozzles with diameters of 200, 300 and 500 μm were used to provide the assist gases. In Khan's experiment, the profile of hole drilled with assistance of air and oxygen was compared to the condition that no process gas was used. It was found that the

highest drilling speed was obtained when there was no process gas. However, the drilling speed was found to decrease by 50% when choosing air as assist gas. In the case of oxygen assist gas, the drilling speed was reduced by 60%. It was concluded that high pressure assist gases can increase the plasma shielding effect by providing higher gas molecules density. More laser energy was shielded by the plasma and cannot reach the ablation front which lead to a decrease in drilling efficiency. It was also found that large nozzle can enhance the plasma shielding which would further decrease the drilling speed (Khan et al. 2004, Khan et al. 2007).

The assist gas also has a significant effect on the spatter. The influence of the types of assist gases on the mechanism of material ejection and removal was reported (Low et al. 2000).

The spatter bonding strength was found to be related to the inertness of the process gas used, in which a progressive increase was observed in the order of oxygen, compressed air, nitrogen and argon. It was reported that a non-uniform material removal rate and sidewall evolution in stainless steel were observed to strongly depend on assist gas (Webster et al. 2010).

Previous study (Okasha et al. 2010) presented a detailed analysis of the gas type and gas pressure effects on the drilling speed and hole quality. Oxygen, nitrogen, argon and compressed air were used in their investigation for Nd:YAG laser drilling of In718 alloy.

It was shown that despite the lower penetration rate, the total volume of material removed

was higher with oxygen assist gas due to oxide layer formation and plasma formation within the holes which increased the hole wall ablation, although drilling time with oxygen was longer than with other gases. Hole quality variations for holes under different gas type and pressure conditions were evaluated. One very interesting and significant finding was that the hole quality which achieved by laser drilling without assisting gas was better than that obtained when using argon, nitrogen or compressed air.

In another study, experiment was conducted under vacuum condition (Kataoka et al. 2006).

It was found that the drilled depth under vacuum condition was deeper than that under the atmosphere pressure condition, since the removal of debris was carried out more efficiently compared to the atmosphere pressure and assist gas supplying conditions.

2.2.5 Depth of focus and beam waist

The radius of the focus spot size is at a minimum value w_0 at the certain position along the beam axis, known as the beam waist which can be calculated by using Eq. (2.3) (Johnson and Pfladderer 1989).

$$w_0 = f \cdot \theta_d \quad (2.3)$$

where f is the focal length of lens and θ_d is the beam divergence angle as presented in Fig.

2.3. The diameter of focus increases with the increase in the focus length of the lens. Fig.

2.4 shows that the shape of the hole has changed as the distance between lens and target is

varied (Kuriyama and Ito 2003). It can be indicated that lens with small focus length should be chosen in order to obtain the minimal entrance diameter.

Fig. 2.3 Focal pattern of converging lens (Johnson and Pfladderer 1989)

As shown in Fig. 2.3, focus depth is the distance along the beam axis where the focused laser beam has approximately the same energy density. It is defined as the distance over where the spot varies by 5%. The depth of focus Δf can be estimated with Eq. (2.4) (Meyer-Arendt 1984):

$$\Delta f = \frac{2 \cdot w_0 \cdot f}{D} \quad (2.4)$$

where D is beam diameter before the lens.

Fig. 2.4 Ring drilling on glass plate by fs laser at various focus position for $f = 5 \text{ mm}$ (left) and $f = 3 \text{ mm}$ (right) apertures in air. Z_f is the position of work-piece surface in the vertical direction. Pulse energy is $165 \text{ }\mu\text{J}$ and number of pulses is 250 (Kuriyama and Ito 2003)

The variation of beam diameter with beam waist position is shown in Fig. 2.5. The position of focal plane of the laser beam significantly influence the depth and profile of laser drilled holes. Inappropriate focus position will result in a hole with poor straightness. It was reported (Yilbas and Sami 1996) that altering the beam waist positions tended to produce converging and diverging hole walls for negative and positive positions, respectively.

Fig. 2.5 Optical and knife-edge damage results for beam diameter with beam waist position (Yilbas and Sami 1996)

2.2.6 Material properties that affecting the laser drilling

Material parameters that have significant influence on the laser process are listed as follow:

2.2.6.1. Absorptivity

Material absorptivity determines how much of the energy can be absorbed by the work-piece surface. Absorptivity varies with wavelength, angle of incidence, beam polarization, temperature, surface roughness and contamination.

2.2.6.2 Material removal temperature

The material removal temperature is defined to be the temperature at which active material removal takes place. In a conventional sense, this may be viewed as the boiling, sublimation,

decomposition point of the material. For some materials, the material removal temperature is identical to the boiling point. However, most other materials melt before boiling and a substantial portion of the melt is blown away by the assist gas. A change in ambient pressure results in the change of this temperature.

2.2.6.3 Specific heat of material

The specific heat (c) determines how much energy is required per unit mass to raise the temperature of the material from the ambient temperature to removal temperature. As the specific heat increases, a larger amount of energy is needed to heat the material to a given temperature. Ceramics typically have about three times the specific heat of metals.

2.2.6.4 Thermal conductivity of material

The thermal conductivity determines how fast of the absorbed energy is dissipated via heat conduction under steady-state conditions. The Fourier's law of heat conduction is applicable in CW (Continuous Wave) or long pulse laser machining. A large value of thermal conductivity results in heating of the whole work-piece rather than rapid heating of a targeted zone. This results in a smaller depth of drilling and lower drilling speeds. Materials with low conductivity such as paper, plastic and wood are easily machined even at low powers with very high drilling speeds (Ivarson 1993). The thermal conductivity of ceramics decreases with increasing temperature. This variation needs to be included in order to accurately model the laser machining process. Some materials exhibit substantially

different conductivities in different directions causing additional uncertainties in accurate modelling of laser processing.

2.2.6.5 Thermal diffusivity of material

The thermal diffusivity α is a measure of the speed of heat propagation through the medium and is related to thermal conductivity, density and specific heat through the relation, $\alpha = k / \rho c$ where ρ is the density of the material. The effects of thermal diffusivity on the laser machining process can be deduced from arguments as given above for thermal conductivity k and specific heat c since the density of the material is usually fairly constant.

2.2.7 Different types of processing techniques in laser drilling

Laser drilling can be accomplished by several techniques as presented in Fig. 2.6 (Abeln et al. 1999). Percussion laser drilling is a direct drilling method where the successive laser pulses from the stationary laser beam are delivered to the same spot on the material. The hole can be as small as the size of focused laser beam spot in percussion drilling.

Trepanning drilling is applied to produce large hole with several hundred micro meters (Dhar et al. 2006). The cylindrical core of material is removed by rotating the laser beam in trepanning method.

Another drilling technique is helical drilling method in which the ablation front penetrates the work-piece on a helical path. Trepanning and helical drilling show some advantages in

controlling the taper of the drilled hole by using multiple axis computer numerical control (CNC) facilities. However, their drilling speeds are much slower than the percussion drilling (Li et al. 2002).

Fig. 2.6 Beam delivery processing options (Abeln et al. 1999)

2.3 Characterisation of hole quality

2.3.1 Aspect ratio

Aspect ratio of laser drilled hole is the ratio of hole depth to the average hole diameter.

There is an increasing demand for high aspect ratio micro hole in different materials for micro sensors and micro heat exchangers to obtain higher efficiency (Huang et al. 2014).

Optical system with low numerical aperture (N. A.) that can provide smaller angular divergence of laser beam is helpful to drill a high aspect ratio hole (Tokarev 2006). On the other hand, the material optical and thermal properties are of great importance to the hole

aspect ratio. It is much easier to obtain high aspect ratio on the polymer materials which have high optical absorption coefficient and low ablation threshold. It was reported (Lazare et al. 1999) that hole with aspect ratio of 500 (as shown in Fig. 2.7) can be drilled on PET (Polyethylene Terephthalate) by using KrF excimer laser even at relatively low fluence (1.8 J/cm^2). It was concluded that the multi-reflection (as presented in Fig. 2.8) on the smooth hole wall of polymer dominated the laser energy transport and was the main reason for the formation of high aspect ratio hole.

Fig. 2.7 Optical micrographs of micro holes drilled on PMMA (Poly(methyl methacrylate)) (a), PET (b), PC (Polycarbonates) (c) (Belforte and Levitt 1992)

Fig. 2.8 Laser beam multi-reflection mechanism

2.3.2 Taper

Taper in laser drilled holes is formed due to the Gaussian energy distribution of the laser beam in the initial stage of laser drilling and also results from the erosion that is caused by the ejection of melting or vaporized material when the hole goes deeper. Based on the hole diameter measurements, the taper is calculated using Eq. (2.5)

$$Taper(\theta) = \tan^{-1} \frac{d_{entrance} - d_{exit}}{2t} \quad (2.5)$$

where θ is the taper angle and t is the material thickness. Researchers (Li et al. 2002) studied the laser beam interpulse shaping's effect on the hole-taper. It was found that by linearly increasing the laser pulse energies throughout the pulse train, parallel holes with diameter around 500 μm were produced on the 2.2 mm Nimonic 263 alloy. This study also indicated that hole taper was most sensitive to focal plane position variations followed by pulse width, peak power and number of pulses.

It was reported that ANOVA results (Bandyopadhyay et al. 2005) on Nd:YAG laser drilling IN 718 showed a similar result with Li et al.'s study (Li et al. 2002). It was found that the laser power, focus position and pulse duration were the most important parameters affecting the taper. The results also suggested that the taper angle of hole drilled by pulse laser can be minimised by choosing shorter pulse duration, lower pulse energy and lower level settings of focus position.

A similar experiment (Tan 2006) was conducted on silicon by using the multi-burst pulse train. Firstly, a burst of laser pulses with smaller fluence punched the target. A through hole with smaller diameter than desired value was produced. Afterwards, the second burst of pulses with higher fluence widened the exit of hole. The micro-hole with straight hole wall and zero taper angle can be obtained. The hole drilled by the initial burst of pulses provided the space and exit for the expansion of laser induced plasma plume and vapour. Therefore, the subsequent burst of pulses can remove the material without any plasma induced deformation.

2.3.3 Spatter

The spatter that is formed by the ejection of melting droplet or resolidification of vaporized material is found not only at the entrance hole but also at the exit hole. The spatter and recast layer can be partially reduced by properly choosing laser parameters that effectively

eject melting and vaporized material. The previous studies (Yilbas 1997, Low et al. 2000, Tunna et al. 2001, Jackson and O'Neill 2003, Das and Pollock 2009) showed that low level setting of power, pulse width and wavelength can result in smaller spatter area. However, the drilling productivity is sacrificed by adjusting the laser parameters to meet the requirement of minimal spatter. In addition, it is difficult to completely clear up the spatter by applying such method. There is thus a constant need to develop more efficient techniques to minimise debris formation during laser processing (Zheng and Jiang 2010).

Hence, a great number of studies have focused on the techniques for the spatter reduction or prevention during laser drilling. These techniques included using pressurized gas (Low et al. 2000), pre-coating (Lizotte 2002), and ultrasonic techniques (Li et al. 2009).

The role of different gases on removing spatter was already summarised and discussed in Section 2.2.4. Hence, only one study (Biffi and Previtali 2013) involving an innovative gas nozzle is introduced in this section. Biffi and Previtali proposed a shielding gas nozzle which was designed to completely contact with the target material during laser drilling. The spatter area was found to decrease to one fourth of that when using traditional nozzle. The limitation of this method is that nozzle needs to be contact with the flat surface of the target material, thus the focus position is fixed.

The pre-coating is also an effective way to reduce the spatter. Researchers (Low et al. 2001) applied an antispatter composite coating on the surface of aerospace materials before laser

drilling. It was found that the coating can effectively prevent the spatter deposition due to the lower wettability of the coating. In the presence of pre-coating, the ejected molten material can hardly wet the substrate surface, thereby the amount of spatter was significantly reduced. In another study, researcher (Choo et al. 2004) introduced a simple, alternate method for collecting the debris generated in laser ablation and preventing it from redepositing on the finished surface. This involved covering the surface of the silicon wafer to be ablated with an adhesive tape. Results of the holes ablated before and after peeling off of the tape showed minimal thermal damage similar to the hole ablated by excimer laser under water and absence of redeposited molten and resolidified material from laser ablation. It was reported (Lizotte 2002) that a sacrificial water-soluble coating was applied onto the tape-automated bonding (TAB) tape before laser processing. The spatter was found to be nearly eliminated after the coating was washed by the high pressure deionized water. Additionally, a study (Zhang et al. 2009) involving a hybrid process of laser drilling assisted with jet electrochemical (NaNO_3) machining was presented. Due to the effect of electrolyte and anodic dissolution, the spatter around the hole on stainless steel drilled by Nd:YAG laser was reduced by 95% compared with laser drilling in air. Unfortunately, the annular electrochemical overcut was left surrounding the entrance surface.

Some researchers implemented post processing to remove the spatter. It was reported (Li et al. 2009) that the spatter fabricated by the fs laser on alumina wafer can be washed away in an ultrasonic bath with mixture of acetone and water.

2.3.4 Barreling

As shown in Fig. 2.9 (Tan 2006), barreling was formed at the bottom of the deep hole when laser drilled materials with large thickness. It was reported (Yilbas 1987) that the barreling was a result of the erosion of ejections inside the deep hole. It was found that barreling effect can be decreased when assist gas pressure reduced. However, the barreling problem was worsen when the material thickness increased. Quantitatively, referring to Fig. 2.10, barreling was calculated by using Eq. (2.6) (Yeo et al. 1994).

$$Barrelling = a - \frac{d_{entrance} + d_{exit}}{2} \quad (2.6)$$

where a is the maximum hole diameter at mid-span. It was reported (Tan 2006) that the barreling was observed on the hole wall for all the thickness of the hole. However, the barreling became serious when hole diameter was larger. The plasma expansion was confined by the side wall of the deep hole which resulting in the sharp temperature increase of the plasma. This can cause more material removal. In Tan's study, the barreling shape was attributed to the plasma enhanced effect by the side wall in the deep hole.

Fig. 2.9 Holes drilled with 60 pulses of $168 \mu\text{J}$ energy at a 60 kHz repetition rate (Tan 2006)

Fig. 2.10 Salient geometrical features of laser-drilled holes

2.3.5 Heat affected zone

During the laser drilling process, a considerable amount of laser energy is transferred into the substrate which would lead to permanent modification of the material properties. This can create a narrow region near the laser drilled area which undergoes intense heating process. The area where the material property is modified by the laser heating process can be defined as the heat affected zone. HAZ is produced during laser drilling in materials when the temperature rises above the critical transformation point temperature. In laser cutting, this is localized near the cutting zone. Since the HAZ is brittle, this area has a lower tolerance for cracking during bending or stress. In most cases, the HAZ can be eliminated by post-heat treating the part, but there is a risk of distortion (Hou and Komanduri 2000).

Fs laser is a reliable tool to reduce the HAZ. When fs laser is interacting with a substrate material, the ultra-short pulses cause minimal thermal diffusion and therefore produce more precise machined features with minimal HAZ (Chichkov et al. 1997). It was reported (Le Harzic et al. 2002) that experiments were conducted to compare the HAZ on aluminum sheet produced by ns and fs laser drilling. It was found that the width of HAZ was within 2 μm when applying fs laser with 2 J/cm^2 fluence. However, a 40 μm wide HAZ was induced by ns laser with 5 J/cm^2 fluence. This result provided the evident to prove that minimal HAZ can be achieved by fs laser.

2.4 Liquid assisted laser drilling techniques

2.4.1 Previous research works related to liquid assisted laser drilling

In the recent studies (Choo et al. 2004, Li and Achara 2004) a few types of liquids were studied for their roles in minimising debris and recast layer formation in laser drilling.

The dynamics of the pressure generated by the laser processing at solid-liquid interface were quantitatively investigated (Park et al. 1996). A ns pulse KrF excimer laser was applied to irradiate water on a solid surface where the water underwent rapid thermal expansion and explosive vaporization. The measurements showed that a compressional pressure wave was generated at the interface with the peak intensity of the order of 1 MPa.

The optical specular reflectance probe was applied to conduct a simultaneous monitoring of the kinetics of bubbles growth. It was observed that the bubble expansion in the superheated water can result in the enhancement of pressure generation which is helpful to blow off the melting material (Li et al. 2009).

The mechanisms of ns pulse laser induced breakdown of strongly absorbing solution were investigated in previous study (Kim et al. 1998). A modelling work was conducted to study the explosive vaporization process of liquid when applying high fluence laser pulse. It was found that in the case of short pulse laser, the breakdown of liquid was induced at low laser

fluences. During the laser induced liquid breakdown process, it was observed that there were no sharp increase in the liquid temperature. Nevertheless, when the laser heating process was rapid enough to lead to the superheating of the liquid, homogeneous bubbles nucleation would speed up. The sharp increase in the rate of homogeneous bubbles nucleation can result in the explosive vaporization which would be the main mechanism of the laser induced liquid breakdown. The experimental results also showed that strong shock wave was generated in the ambient air when the explosive vaporization happened in the liquid.

Researchers (Lu et al. 2004) studied the mechanisms of laser drilling metal plates for enhancing the efficiency and quality of laser processing in underwater. The impact generated by laser ablation increased the peak amplitude and duration due to water confinement. The first and second liquid-jet-induced impulses by cavitation bubble collapse in the presence of solid boundary, amplitudes were 12.4 and 5.2 times that of laser ablation impact in air. The conductivity of liquid was better than air for heat generation, the excessive heat was dissipated into water during laser processing for better edge profile. The efficiency of laser-drilling a metal plate in water was observed higher than that in air (Xu et al. 2004). It was shown that in air the sample was only affected by laser-induced ablation, while underwater it was impacted by not only ablation, but also two liquid-jet impulses. The material removal efficiency underwater is larger than that in air.

Laser trepanning drilling experiments were conducted on silicon under flowing water by applying 355 nm-XAVIA laser (Wee et al. 2011). It was found that the flowing water helped to wash the silicon surface and reduce the spatter area. In their study, the cleaner surface was attributed to the generation of shock wave during the bubbles expansion process. It was also found that smaller spatter area and hole diameter can be obtained when using low laser frequency, i.e. 3040 kHz, and high pulse energy. The taper angle of hole wall was found to decrease with increasing the laser frequency and the laser fluence. The laser beam scan speed had no influence on taper angle of hole drilled in air. In addition, focus position's effect on laser drilling process under water was investigated. It was found that the smaller spatter area can be obtained when the focus plane was set to be below the surface of silicon wafer as compared with the condition that laser beam was focused right on the silicon surface.

Laser processing of silicon both in air and in water was investigated using Ti: sapphire fs laser with the pulse duration from 50 fs to 24 ps (Wang et al. 2009). It was found that multiple laser pulses with relatively small pulse energy can result in high aspect ratio hole. The porous surface structures were found in the laser irradiated area in the water environment.

Experiments of laser drilling silicon were conducted the in the dimethyl sulfoxide (DMSO) and in water at various laser powers and number of pulses (Karimzadeh et al. 2009). The

silicon sample was ablated by using laser with pulse duration of 15 ns and wavelength of 532 nm. The porous structures were observed around the laser ablation area on the silicon surface. The micro-cavities that was observed at the irradiated area indicated that the micro-bubbles nucleation occurred. These micro-structures provided the evidence to prove that the dominant mechanism of material removal during laser ablation in liquids could be explosive melting expulsion when using ns pulse laser with wavelength of 532 nm. In addition, the diameters of laser damaged area on silicon were measured under various laser power and number of pulses in DMSO and water. It was found that the diameter of laser induced hole increased exponentially with laser pulse energy. The diameter of laser ablation area in DMSO is found to be smaller than that in water for the same pulse energy. The single-pulse laser ablation threshold of silicon was obtained using energy accumulation model. It was found that the laser ablation threshold of silicon was smaller when the sample was ablated in the DMSO. In their study, the multi-pulses ablation threshold was also determined by the same model. It was found that the ablation threshold became smaller with increasing the number of pulses. It was concluded that the relatively longer pulse duration of the ns laser overheated the laser induced plasma plume which was confined by the liquid. The strong shock wave induced by the confinement of plasma expansion provided an additional force which was helpful to remove the material and reduce the ablation threshold.

In another study, fs laser ablation technique was applied to machine silicon and gold samples in various environments (Besner et al. 2004). It was found that the laser ablation threshold of silicon was smaller than gold. This was attributed to the higher optical absorption coefficient of silicon at wavelength of 800 nm. The ablation threshold of silicon was found to be similar in the different environments (air, vacuum and water). Additionally, in the case of lower fluence, the environmental medium had no significant influence on the profile of the hole on silicon wafer. At high laser fluence, however, the morphology of ablation area was found to be distinct when irradiated in different mediums. The surface of hole ablated in vacuum is relatively clean and smooth. In the case of air environment, a great amount of debris around the hole was found. However, the ripples structures appears when the hole was ablated in water. The debris was found absent in the case of water. At high laser fluence, the plasma heating effect would be enhanced by higher laser power. This heating effect produced rapid bubbles generation which can clean off the debris.

2.4.2 Limitation of previous studies

A number of issues have been identified but yet to be resolved. These issues include: laser power loss due to a large portion of laser energy being absorbed by the liquid, non-circular hole geometries due to the scattered laser beam by the liquid and bubbles, and material corrosion due to the use of corrosive liquids such as acid solution. The hole drilled in liquid

usually has a larger entrance diameter than that drilled in air at the same laser setting (Ren et al. 2005, Kang and Welch 2007). In addition, the ablated surface is rougher in the case of under water than that in air. In an attempt to provide some solutions to these issues, researchers (Kang and Welch 2007) studied the effect of water thickness on laser drilling efficiency. They reported that the laser ablation efficiency increased with the decrease in the water film thickness. Due to the high surface tension of water, the minimal liquid thickness that Kang and Welch was able to apply was 0.5 mm. It is necessary to study the effect of liquid layer thinner than 0.5mm on laser drilling quality. This is also the main objective of Chapter 4 in this thesis.

2.5 Laser induced period surface structures (LIPSS)

2.5.1 Previous research works related to LIPSS

LIPSS can potentially be used for engineering surface properties such as thermionic emission which is used in variety of industrial applications (Barmina et al. 2012). Generation of periodic surface structure on different materials using different lasers has attracted lot of attention since it was first reported (Birnbaum 1965). Various research reported in the literatures used ultra-short pulse lasers to generate finer and cleaner periodic structures on a variety of materials such as semiconductors (Bonse et al. 2009), metals

(Wang and Guo 2005) and dielectrics (Wang et al. 2005). The processing parameters of ultra-short laser included wavelength, number of pulses, laser fluence, pulse duration, incident angles and polarization. These have been investigated for their effects on the spatial wavelength and depth of periodic surface structure. The periodic structures were observed as oriented perpendicularly to the polarization of incident laser radiation. The LIPSS period induced by ultra-short pulses at normal incidence was slightly less than the laser wavelength. Pulse duration has a significant impact on the features of LIPSS. Compared to fs pulse laser, the ns laser or other long pulse produced ripples with coarser and deeper structures (Hsu et al. 2007) due to its relative larger thermal effect. In addition, the wavelength of LIPSS was considerably large (Trtica et al. 2004, Khaleeq-ur-Rahman et al. 2009, Umm et al. 2012) when applying the longer pulse. Thus, the fs laser with minimal thermal effect was preferred in order to obtain a fine and orderly periodical surface structure.

It was reported that the period ripples were resulted from the optical interference of the incident laser irradiation with surface scattered waves (Bonch-Bruevich et al. 1992). The non-uniform surface energy distribution led to periodic material melting and resolidification.

2.5.2 Limitation of previous studies

The modulation of the spatial wavelength of LIPSS is still an unsolved problem. In the recent literatures, most of the experiment involving LIPSS was conducted in air. The environmental mediums effect on the generation of LIPSS is largely unknown. It cannot be found for the study that focus on the fs laser induced periodic surface generation on silicon using ethanol as replacement to air. This will hence be the main content of Chapter 5.

2.6 Pre-heating laser drilling

2.6.1 Previous research works related to pre-heating laser drilling

Researchers (Yahng et al. 2009) reported the effect of environmental temperature on the fs pulse laser ablation of various materials. The surface roughness of ablation area in silicon and stainless steel was found to decrease significantly when applying high substrate temperature. As shown in Fig. 2.11, the substrate temperature had no significant influence on the material removal efficiency of glass, however, in the case of stainless steel the laser ablation efficiency was increased by 20% when substrate temperature was elevated to 900 K. It was found that the material removal efficiency for silicon was slightly increased at 900 K. The ablation threshold for silicon was also found to be smaller at higher temperature.

This was attributed to the increase of electron density in the conduction band and narrowing of band gap for silicon at high temperature.

Fig. 2.11 AFM images of the processed glass and steel surfaces at two different substrate temperatures (300 and 900 K) (Yahng et al. 2009)

In the following research, a detailed study (Yahng and Jeoung 2011) was conducted to investigate the effect of substrate temperature on surface roughness of silicon ablated by fs laser. It was found that the surface roughness of silicon showed an opposite trend with increasing substrate temperature at different laser fluence. When laser fluence was less than damage threshold, the surface roughness increased with increasing temperature. However, the surface roughness decreased with increasing temperature for laser fluence of 0.5 and 1.0 J/cm², respectively. If the laser fluence was larger than 2.0 J/cm² the surface roughness was independent with temperature increasing. This phenomenon was attributed to the

temperature dependence optical absorption coefficient of silicon and the melting of laser-induced period surface structures at high temperatures.

Recently, the fs laser ablation threshold in silicon as function of temperature was investigated (Thorstensen and Erik Foss 2012). The temperature range was from room temperature to 593K. It was found that the ablation threshold was reduced by 43% at elevated substrate temperature when the wavelength was 1030nm. The increase of absorption coefficient at high temperature was the main cause for the reduction of ablation threshold.

2.6.2 Limitation of previous studies

From the previous studies, it can be concluded that the substrate heating is a useful technique to reduce the surface roughness and ablation threshold of the silicon during the laser machining. These experimental results also proved that the laser material interaction process can be significantly influenced by the substrate temperature because of the variation of the absorption coefficient at various temperature. However, the previous studies is limited in surface processing on the target material. The temperature's influence on laser drilling of deep through hole is however still an unexplored area. This will be the main content of the Chapter 6 in this thesis.

2.7 Theoretical studies of laser drilling

Due to the high investment costs of laser systems and the need of highly skilled operators to conduct the laser machining experiments, there is a need to model the laser interaction process to compute approximate data and evaluate relationships of laser drilling parameters which would be only achieved by a large number of time-consuming and costly precision experiments. The modelling approaches include statistical, analytical, and numerical such as Finite Element Method (FEM), Finite Difference Method (FDM) and Finite Volume Method (FVM).

A number of existing models of laser drilling-related works are tabulated and summarised in Table 2.2.

Table 2.2 Summary of previous computational models for pulsed laser interactions with various materials

Reference	Pulse Duration	Materials	Features
(Szyszko 1995)	12 ns, 38 ns	Silicon	One dimensional (1-D) heat conduction equation, finite-element method
(Anisimov and Rethfeld 1997)	1 ps	Golden	Two temperature model (TTM), melting and evaporation
(Xu et al. 1999)	6 ns	Nickel	1-D heat transfer equation
(Zhvavyi 2000)	70 ns	Silicon	1-D heat transfer equation
(Atanasov et al. 2001)	10 ns	Silicon nitride, Aluminum	1-D heat transfer equation, Forward Time Central Space (FTCS) finite difference method, plasma plume
(Quanming et al. 2002)	3 ns	Silicon	1-D heat transfer equation, evaporation, vapour plasma
(Ho and Lu 2003)	10 ns	Silicon	1-D heat transfer equation, analysis solution Plasma plume
(Li et al. 2003)	0.08 ms	Silicon	3-D heat transfer equation, evaporation FTCS FDM
(Ki and Mazumder 2005)	80 fs	Silicon	TTM
(Semak 2006)	4 ns, 200 fs	Aluminum, Copper, Steel	TTM, FDM

2.7.1 Heat transfer modelling for long pulse laser

A millisecond or nanosecond pulse laser processing of materials utilises the high power density provided by the laser beam, which is focused on the work-piece. As a result, the work-piece material experiences heating, melting, vaporization and re-solidification. Understanding the temporal evolution of the temperature field during laser process is thus one of the most significant factors in achieving a desired quality of processing. The thermal distribution and its history are required to determine the related stresses, phase transformations taking place, and the final metallurgical microstructures. Therefore, the ability to determine the thermal field has been a major aspect of most models in laser micromachining.

An analytical solution was obtained for the laser drilling silicon nitride and alumina ceramics by solving the one-dimensional heat-diffusion equation using a Laplace transformation method (Ho and Lu 2003). It was assumed that the direct phase change from solid to vapour dominated the material removal mechanism.

Figure 2.12 shows the computational domain (Ho and Lu 2003). Only the z -direction of laser material interaction was considered because the laser spot size was much larger than the optical penetration depth and heat diffusion length. Under this condition the simulation was simplified into 1-D problem.

Fig. 2.12 Laser beam and work-piece (Ho and Lu 2003)

The heat equation (2.7) has this form:

$$c\rho\frac{\partial T}{\partial t} = \frac{\partial}{\partial z}\left(k\frac{\partial T}{\partial z}\right) + S(z,t) \quad (2.7)$$

A 10 nanosecond (ns) pulse Nd:YAG laser is assumed as an energy source which is represented by $S(z,t)$.

In addition to the thermal considerations, laser material processing involves several other complex phenomena, such as the laser induced plasma which significantly affects the absorption of laser beam energy as was discussed by this study (Ho and Lu 2003). By solving Eq. (2.7), the dependence of ablation depth for each pulse on laser energy was calculated. The variation of ablation depth increased more sharply in the case of low laser energy but at slower rate when applying high laser energy. The slow increase of drill efficiency at higher laser energy can be attributed to the formation of laser induced plasma

at high laser fluence. A great portions of laser energy would be absorbed by the plasma plume which would affect the laser drilling efficiency.

The discrepancy between the experiment and simulation drilling depths occurred at certain fluences. In the simulation the value of the absorption coefficient of plasma was set to be constant (Ho and Lu 2003). However, the plasma absorbance depends on the laser intensity in the actual situation.

A 3-D modelling was developed in previous study (Li et al. 2003). The governing equation is written in terms of temperature T as shown in Eq. (2.8):

$$\frac{\partial(c\rho T)}{\partial t} = \nabla \cdot [k(T)\nabla T] + A \quad (2.8)$$

where A is the heat source term being externally injected or extracted. When the material's temperature reached the melting point and the energy of laser irradiation exceeded the melting latent heat of the material, portion of the substrate material was assumed to be removed and no longer considered. In this work, the proposed numerical model was to evaluate the evolution of the temperature field and the propagation of the hole boundary by solving the time-dependent heat diffusion equation in three dimensions (most theoretical analyses and models in the literature were based on a one-dimensional steady state assumption). The laser drilling experiment was conducted on silicon wafer and the experimental results were compared with simulation predictions. It was found that there

was a good agreement between simulation results and available experimental results (Li et al. 2003).

2.7.2 Heat transfer modelling for ultra-short pulse laser

However, phonon energy relaxation happens when the laser pulse becomes shorter. In the microscale energy transfer, $1ps$ has a special meaning as a threshold since it is about the time needed for phonon energy relaxation. If the pulse width is smaller than this threshold, the heating process is so intense that physical interactions occur in the non-thermal process which means non-equilibrium energy transport occurs between electrons and lattices.

In 1974, researchers (Anisimov 1974) introduced the two temperature mode (TTM) to explain the laser-metal interaction as shown in Fig. 2.13. It is based on the 1-D heat transfer equation and considers the interaction process of phonon-electron and electron-lattice. They developed the partial differential Eq. (2.9) and Eq. (2.10) to describe the change of temperature between the electron and the lattice:

$$C_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x} k \frac{\partial T_e}{\partial x} - G(T_e - T_i) + A(x, t) \quad (2.9)$$

$$C_i \frac{\partial T_i}{\partial t} = G(T_e - T_i) \quad (2.10)$$

where T_e and T_i are electron and ion temperatures, C_e and C_i are the volume heat capacities for electrons and ions, $A(x, t)$ is the energy source term due to laser energy dissipation into

the matter, k is the thermal conductivities of material and G is the temperature-dependent electron-lattice coupling coefficient.

In order to obtain the electron-lattice coupling coefficient, researchers (Fujimoto et al. 1984) applied fs optical pulses to observe the multiphotons and thermally enhanced photo emission from a tungsten metal surface. Experimental results suggested the presence of anomalous heating, a transient non-equilibrium temperature difference between the electrons and lattice. Pump-probe measurements indicated an electron-phonon energy relaxation time of several hundred femtoseconds.

Additionally, researcher (Schoenlein et al. 1987) applied fs laser pulses to induce non-equilibrium electronic heating in gold sheet. Measurements were conducted for various laser pulse fluences and probe photon energies. It was observed that the reflectivity line shape proved the generation of non-equilibrium electronic temperature that was cooled down by the lattice on a time scale of 2 to 3 ps.

Fig. 2.13 Fs laser interaction with metal materials (Anisimov 1974)

Fs laser pulses was applied on thin copper films and detected the electron-phonon energy transfer time was 1-4 ps (Elsayed-Ali et al. 1987). The process of electron-phonon energy transfer was time resolved using TTM and was observed to increase with decreasing of the value of coupling coefficient G . The G value was determined to be $1 \times 10^{17} \text{ W/k}\cdot\text{m}^3$.

One study (Corkum et al. 1988) used TTM to explain the experimental result of fs and ns laser processes on Copper and Molybdenum. They compared the effect of different G values on the damage threshold. The G value was determined as $2 \times 10^{16} \text{ W/k}\cdot\text{m}^3$. It was indicated that damage on metal caused by pulses with duration less than 1 ns can only be understood with a TTM.

TTM was simplified by neglecting the heat conduction term in Eq. (2.9) and obtained the analytical solution for the T_e and T_i (Momma et al. 1997). Coupling the analysis solution with Arrhenius-type equation, the qualitative behaviour of the ablation depth for fs laser pulses in the low fluence regime was reproduced rather well against the experimental results.

TTM was used to calculate the multi-pulses laser ablation thresholds for gold and nickel thin films of various thicknesses (Gudde et al. 1998). By applying the TTM, they showed that in the case of fs laser pulse the hot electrons diffusion dominated energy transport mechanism during the first picoseconds. However, energy transport for ns-laser pulses was found to be determined by the heat diffusion in the lattice.

Researchers (Wellershoff et al. 1998) used TTM to analyse the damage threshold for melting metal films by ns and fs laser. For ns laser, the TTM was simplified to the usual heat transfer equation. For fs laser, TTM was solved using numerical method. The experimental data of damage threshold was detected by measuring changes in the scattering, reflection and transmission of the incident light. They indicated that laser damage of metals, even with fs lasers, was a purely thermal process.

In the following study, researchers (Furusawa et al. 1999) did not simplify the specific heat and the thermal conductivity in TTM as temperature independent. It assumed that the phase transition occurred after equilibrium. Then an analytical solution for the final equilibrium temperature was obtained. Experiments were conducted to confirm that two different ablation regimes were found in terms of the laser fluence. The characteristic length of different ablation regimes was explained in terms of the optical surface depth and thermal diffusion length. It was determined by the peak electron temperature in the TTM.

Two temperature and hot-electron blast models are extended to investigate the deformation in metal films subjected to ultra-short laser heating (Chen et al. 2002). Two potential material removal mechanisms, namely the thermal (melting) and non-thermal (high stress), were identified. Numerical results showed that the non-thermal damage could be a dominating mechanism in ultra-short laser-material ablation. The major driving force for

the non-thermal damage was the so-called hot-electron blast force that was generated by non-equilibrium hot electrons.

All of the above research applied TTM into laser-metal interaction. In another study (Ki and Mazumder 2005), TTM was introduced into the simulation of fs laser drilling silicon.

In this study, the heat diffusion term which was usually simplified in the pioneer works was included in the governing equations as shown in Eqs. (2.11) and (2.12).

$$C_e \frac{\partial T_e}{\partial t} = \frac{\partial}{r \partial r} (r k_e \frac{\partial T_e}{\partial r}) + \frac{\partial}{\partial z} (k_e \frac{\partial T_e}{\partial z}) - G(T_e - T_i) + A(x, t) \quad (2.11)$$

$$C_i \frac{\partial T_i}{\partial t} = \frac{\partial}{r \partial r} (r k_i \frac{\partial T_i}{\partial r}) + \frac{\partial}{\partial z} (k_i \frac{\partial T_i}{\partial z}) + G(T_e - T_i) \quad (2.12)$$

where k_e and k_i are the thermal conductivities for electrons and ions, r is the radial coordinate, and the definition of others parameters is the same as that in Eqs. (2.9) and (2.10). The time histories of electron and lattice temperature distribution inside the silicon substrate were successfully simulated numerically in two dimensions. The results showed that the electron temperature can reach more than 50000 K in ultra-short time (less than 200 fs) for both flat-top and parabolic cavity targets. However, the lattice temperature only started to increase very slowly long time after the electrons were heated. Within the time period of the simulation, the lattice temperature was raised only by around 50 K.

In TTM, numerical solution turned out to be sensitive to a number of governing parameters, such as electron-phonon coefficient, thermal conductivity coefficient, optical absorption depth, which in turn may be functions of temperature, density, etc. In order to make the

prediction for damage threshold more accurate, researchers (Fedosejevs et al. 2006) discussed the value of electron thermal conductivity and electron-lattice coefficient (G). The simulation work indicated that the G value should be chosen as 3×10^{16} W/K•m³ to fit the measured ablation threshold if a more realistic thermal conductivity model was to be applied.

2.7.3 Modelling for laser beam propagation

There have been a number of simulation works using various theories that make contributions to study the hole wall's influence on the propagation of the laser beam in the micro hole drilling.

Recently, researchers (Zhang et al. 2013) introduced three functions including convergence function, coordination function and factor function to simulate the laser energy redistribution due to the multi-reflection inside the micro hole. A simple transformation of source term in the heat transfer equation was used to compensate the multi-reflection of the laser on keyhole wall and thereby simulate the dynamics formation of the keyhole during the laser drilling.

Wave guide theory was also used to simulate the attenuation of the laser pulse energy inside the cavity and calculate the drilling depth per pulse on copper (Türkoğlu et al. 2012). The model can only be applied to the low fluence regime that is close to the ablation threshold.

In addition, the wave guide theory used in their model was only suitable in explaining the wave propagation when the cavity size is much larger than the incoming laser wavelength that make it difficult to analyse the wave behaviour around the sub-micro-features or sharp corners of the micro cavities.

Ray tracing method was most commonly used (Ki et al. 2002, Cho and Na 2006, Medale et al. 2007) to consider the hole wall's influence on the laser beam propagation due to its simplicity in implementation. Ray tracing theory that relies on the approximate solution of Maxwell's equations is only valid when the size of obstacle is much larger than the wavelength of the laser beam. Thus, the accurate solution cannot be obtained when encountering complex topological features in the micro hole.

In order to fundamentally understand the laser beam propagation behaviour in the micro hole, it is essential to solve the Maxwell's equations which describe the nature of the electromagnetic wave.

Some researchers used Maxwell's equations to predict the laser propagation behaviour during the laser ablation process. In one study (Ki and Mazumder 2005), the two dimensional (2-D) axial symmetric numerical model was firstly built for simulating fs laser interaction with silicon. The model simulated the fs laser propagation in 50 μm silicon cavity by solving Maxwell's equations using finite-difference time-domain (FDTD) scheme. Their results indicated that the heating pattern by a fs laser was very complicated.

In the following work, based on former model as mentioned above, same group (Li and Ki 2006) investigated the ionization dynamics and nonlinear material response. Electron number density was calculated based on the pulse energy and pulse duration.

In recent work (Tao et al. 2011), the effect of micro-hole's size on the fs laser intensity profile near the surface of substrate was studied by solving the three dimensional (3-D) Maxwell's wave equation. It was assumed that the 1 μm micro-hole had a straight wall and boundary condition was set as perfect electric conductor. Thus, the tangential component of electric field on the wall was neglected. Incident angle's effect on reflection and absorption on the hole wall however was not included in their work.

More recently, researchers (Courtois et al. 2013) developed a complex model in which stationary analysis was conducted by numerically solving wave equation for the gaseous and dense phases. This model involved the multi-reflection of the laser beam on the hole wall that was used to predict the boundary evolution of the key hole formation during the continuous wave (CW) laser welding.

2.7.4 Limitation of previous modelling works

From the previous works, it can be concluded that it is essential to solve Maxwell's equation to fully investigate the laser propagation behaviour inside the micro-hole. Only by this way, the laser energy distribution inside the micro-hole can be fundamentally understood.

Experiment study in Chapter 6 showed that the taper angle of the hole may have significantly influence on the laser energy distribution at bottom of the hole. However, so far, modelling works that focus on the investigation of the taper angle's influence on the laser beam propagation by solving the Maxwell's equations cannot be found. This will thus be the main content of Chapter 7 in this thesis.

2.8 Summary

In this chapter, a comprehensive literature review for the laser ablation was presented. The existing problems and research gap in fs laser drilling area can be summarised as follows:

The fs laser drilling can minimise the HAZ, however, there are constant needs to control the aspect ratio and taper of the hole drilled by fs laser. Additionally, it is necessary to find a reliable and effective way to remove the spatter during the fs laser drilling.

In the liquid assisted laser ablation process, it is hard to avoid the excessive laser power loss, non-circular hole geometries and material corrosion. The hole drilled in liquid usually holds a larger entrance diameter than that drilled in air at the same laser setting. In addition, the ablated surface is rougher in the case of under water as compared to in air.

The modulation of the spatial wavelength of LIPSS is still an unsolved problem. In the recent literatures, most of the experiments involving LIPSS were conducted in air. The environmental mediums effect on the generation of LIPSS is however largely unknown.

The previous pre-heating laser drilling studies were limited in surface processing on the target material. The temperature's influence on laser drilling deep through hole is still an unexplored area.

From the previous works, it was found the hole wall can have significant influence on the laser beam propagation inside the micro-hole. However, we have not found any the modelling works that focus on the investigation of the taper angle's influence on the laser beam propagation by solving the Maxwell's equations.

These existing problems summarised above would be the main research directions of this thesis. The detailed studies for each problem would be presented in the following chapters.

Chapter 3 Parametric Study of Fs Laser Drilling in Silicon

3.1 Preamble

As discussed in Chapter 2.2, the laser parameters such as peak power, number of pulse have significant influence on the laser drilling process. The previous investigations (Low et al. 2000, Alexander et al. 2002, Ito et al. 2003, Kamlage et al. 2003, Yokotani et al. 2005) were limited to the specific experimental material and technique. So far as known, the systematical study for fs laser parameters' effect on both laser drilled hole geometry and spatter area can hardly be found. Very few (Matsumura et al. 2007, Ahn et al. 2012) laser drilling works have been carried out on a single crystal silicon with high aspect ratio without any assisting medium. In order to obtain an overall understanding for the fs laser drilling mechanism, it is necessary to conduct a series of experiments to fundamentally investigate various laser parameters' effect on fs laser drilled hole profile and spatter features.

3.2 Experimental procedure

The experimental setup was as follows: Clark MXR fs laser emitted pulse of 200 fs with linearly polarized light at a central wavelength of approximately 775nm (nominal repetition rate of 1 kHz). The mode of the equipment was set to the Gaussian. The total pulse energy

was attenuated by a rotating half wave. The average laser power after the lens was measured using a power meter. The mechanical shutter was controlled to release the desired laser on the substrate. A three-dimensional CNC stage was applied to position the specimens.

The experiments were conducted on silicon with thickness of 0.725 mm by changing the laser parameters such as laser power (200 to 600 mW), focus position (-2 to +2 mm), focus lens (75,100 mm), number of pulses (200 to 7000). Their effects on the holes quality were studied.

The spatter of specimen was removed by ultrasonic bath for 10 minutes. Thus, the entrance hole diameter ($d_{entrance}$) and circularity could be measured more accurately. The top, bottom and cross sections of the holes were examined by using Deep Ultra Violet (DUV) microscope.

3.3 Result and discussion

3.3.1 Effect of number of pulse

The drilled hole is not perfectly circular due to the spot shape. As shown in Fig. 3.1, the ellipse hole has a diameter of 75 μm in the x direction and 50 μm in the y direction. The average value of the diameter in the two directions was taken as the hole diameter.

Fig. 3.1 Top view of the laser drilling hole at Power = 400 mW, Number of pulse = 4000,

$$Z_f = -2 \text{ mm}$$

Figure 3.2 shows the $d_{entrance}$ as a function of the number of pulses at various laser power levels. The $d_{entrance}$ increases gradually with increasing number of pulses before 1000 pulses. There can be two main reasons responsible for this phenomenon. Firstly, due to the Gaussian distribution of laser energy intensity, the center of the laser spot has higher intensity than the outer rim of the spot. During the laser process, it is possible that only the intensity of central part can exceed the damage threshold of silicon. For single pulse laser ablation, the damage area can be much smaller than the laser beam spot. However, previous study reported that due to the energy accumulation effect the multi-pulse laser damage threshold is lower than the single pulse laser damage threshold (Pronko et al. 1996, Bonse

et al. 2002). This means the more pulses can lead to a material removal in some area that may not be modified by single pulse. This is one possible reason why the more laser pulse can result in a larger hole diameter. Secondly, when the hole goes deeper, more molten or solid materials (Sezer et al. 2005) are ejected from the bottom due to the recoil pressure resulting from the fast vaporization of the silicon. These hot ejections can lead to the erosion of the hole boundary (Li et al. 2002). Thus, the entrance diameter can be enlarged when the hole is drilled deeper by more laser pulses.

Fig. 3.2 Effects of number of pulses on $d_{entrance}$ at various laser powers

However, it is found that from 1000 pulses onwards, $d_{entrance}$ remains nearly as constant when applying more pulses. This is because the entrance hole diameter exceeds the beam spot size at this point. Therefore, there are no further power absorptions on the surface of the silicon since the hole entrance is large enough to avoid the contact with laser beam. Meanwhile, when the hole goes deeper, the molten or solid material can be ejected out along the hole wall which would be much straighter than that near the entrance. The change of moving direction of ejections decreases the erosion effect on the entrance. This may also be the possible reason why entrance hole diameter does not have sharp variation when applying more pulses after 1000 number of pulses.

Furthermore, it is observed that the spatter area is proportional to the number of pulses at various laser powers, as shown in Fig. 3.3. The spatter area shows a linear increase with the number of pulses until it reaches 4000 pulses. In addition, it is found that the hole is drilled through after 4000 pulses. After the breakthrough of the laser beam, the spatter area remains nearly constant as the maximum value. Simultaneously, the exit hole starts to continuously increase as shown in Fig. 3.4.

The relationship between the entrance hole, spatter area and exit hole shows that the number of pulse curve can be divided into three distinct regions. This partition is similar to previous study (Low et al. 2000) where a 400W Nd:YAG ms pulse laser was used.

In the first region, the diameter of entrance hole is sharply and continually enlarged with the increase of the initial laser pulses of 0-1000, prior to the breakthrough. A blind hole is produced and its maximum $d_{entrance}$ is determined. These initial pulses contribute to the expansion of the hole boundary on the surface of the sample, till the hole diameter reaches as the steady state. In region II, medium number of 1000-4000 pulses, laser pulses are sufficient to break through the wafer. Although the wider spatter area is deposited around the hole, $d_{entrance}$ remains nearly constant. As the number of pulses increases, the heat has sufficient time to penetrate into the work-piece which results in a greater material removal rate. In region II, as the number of pulses increases, the $d_{entrance}$ stops to increase further. However, the ejection accumulated beside the hole becomes more and suggests that the middle pulses lead to the propagation of the hole boundary in downward direction. In the third region with 4000-8000 of pulses, there is no significant variation in the spatter deposition area after the breakthrough of laser beam. However, the d_{exit} increases sharply which indicates further absorption is held at the bottom. The fact that there is no significant variation in the spatter area after the breakthrough of laser beam but d_{exit} increases sharply shows most of the ejection can be ejected out downward through the exit.

Fig. 3.3 Effects of number of pulses on spatter area diameter at various laser powers

Fig. 3.4 Effects of number of pulses on d_{exit} at various laser powers

3.3.2 Effect of focus plane position (Z_f)

The Z_f is of great importance to laser drilling because Z_f determines laser spot size at the plane of material surface. In this Section, the effect of focus position on the $d_{entrance}$ is studied with range from -2mm to +2mm. Positive, zero and negative Z_f (see Fig. 2.3) refer to focal plane position above, on, and below the front surface of the wafer, respectively.

Fig. 3.5 shows the variation of $d_{entrance}$ as a function of Z_f at different laser power levels with 4000 pulses and $f = 75$ mm (f is focus length). It is interesting that the smallest $d_{entrance}$ is obtained at $Z_f = -0.2$ mm but not at $Z_f = 0$ where the spot size on the surface plane is smallest. The present finding can provide more evidences to prove that entrance hole diameter is not only dependent on the size of laser energy radiation area at the surface but also highly related to the erosion of ejection material from the hole bottom.

The hole is not drilled through with Z_f below -0.3 mm and above 0.1 mm from the work-piece front or entrance surface. It is revealed that the poor circularity holes are drilled with enlarged entrance hole diameter and non-through hole state. In addition, the beam would be diverged at positive Z_f and its power density is reduced consequently as the depth of the hole increases. As Z_f moves below the front surface, the beam energy density under the surface becomes higher. This means the laser beam is focused inside the work-piece (Han and Pryputniewicz 2004). More energy is absorbed by the materials which make it easy to produce the through hole.

Fig. 3.5 Effects of focus position on $d_{entrance}$

Fig. 3.6 Effects of number of pulses on $d_{entrance}$ at various focus length

3.3.3 Effect of focus lens

Two focus lens, namely 75 mm and 100 mm were used to investigate the effect of the focus length on the $d_{entrance}$. The laser spot size diameter d can be estimated as 60 μm and 80 μm for each lens by Eq. (3.1)

$$d = 2.44 \frac{f\lambda}{D} (2p + l + 1) \quad (3.1)$$

where f is the focal length, λ is the wavelength of the laser, D is the beam diameter before the lens, p and l are the mode numbers, the values of p and l are zero.

Figure 3.6 shows the variation of $d_{entrance}$ at two focus lens versus the number of pulses. It is observed that the hole size is greater when $f = 100$ mm. The larger laser energy irradiation area and heavier erosion from the ejection material when choosing lens with $f = 100$ can produce a larger entrance hole and remove more materials.

Figure 3.7 shows the variation of spatter area diameters for two types of lens as a function of the number of pulses. The spatter area diameter before the breakthrough point is greater with $f = 100$ mm. It cannot be simply concluded that larger spatter area is formed because of a higher material removal volume as a result of the larger spot size at $f = 100$ mm. Low et al.'s study revealed that the spatter area size may be independent of amount of material removal. It is well known the spatter is formed by the resolidification of the molten and vaporizing material. It is possible that the size of the spatter area is determined by how far the ejection material can reach. Thus the ejection's speed at the entrance of the hole can be

of important reason for the size of spatter. In detail, spatter area size can strongly depend on the ejection's speed component in radial direction. In this study, when the lens with larger focus length is chosen, the drilled hole in the initial stage would be shallower due to the larger spot size and relatively smaller energy intensity. Compared with deep hole, the shallow hole profile can provide a larger radial component of the ejection speed. This is the possible reason why the lens with 100 mm focus length can produce larger spatter area before the breakthrough point as shown in Fig. 3.7.

The first breakthrough point appears at 3000 and 4000 pulses when $f = 100$ mm and $f = 75$ mm, respectively. The longer focus depth (Yeo et al. 1994) of laser beam when $f = 100$ mm allows higher laser intensity at the bottom when hole goes deeper. Whereas for the lens with $f = 75$, the laser beam would diverge faster at the bottom resulting in smaller energy intensity. Thus, the longer focus length makes it easier to drill through the silicon wafer as presented in Fig. 3.7.

Fig. 3.7 Effects of number of pulses on spatter area diameter at various focus lengths

Fig. 3.8 Effects of laser power on $d_{entrance}$ and d_{exit}

3.3.4 Effect of laser power

The effect of average laser power on diameter of the hole is investigated in this Section. To obtain the deepest hole, the 75 mm lens was used and the laser beam waist was set at $z = -0.2$ mm, the number of pulses was 4000 which was the minimum drilling through value. The variation of $d_{entrance}$ as a function of laser power range from 50 mW to 600 mW is shown in Fig. 3.8. Both $d_{entrance}$ and d_{exit} are proportional to the laser power. The cross section view of the through hole is presented in Fig. 3.9. As the laser power increases, energy spreads on the sample and more molten material is repelled due to the recoil pressure. Greater material removal rate is achieved and thus the larger entrance hole is produced. However, due to the divergence effect of laser beam during the drilling process, the laser energy intensity is further reduced when the depth of the hole is increased. At the power level of 200 mW, the average diameter is 23 μm and aspect ratio is 30:1. It can be observed from Fig. 3.9 that the taper angle of hole is larger when applying higher average power. At higher power, the inside wall is relatively straight except some scratches resulting from the grinding process. The formation of the straight wall is due to the erosion of melting ejection indicating that a large portion of melting material is produced at higher laser power. Although it was claimed that fs laser had minimal thermal effect, material removal mechanism may not be dominated by multi-ionization but by thermal melting at higher power and multiple pulses. In other words, the cold drilling would be replaced by the hot

drilling when fs laser is set at high power. This thermal induced material removal mechanism is usually used to explain the longer pulse laser ablation (Zweig 1991). However, even for the multi-pulse drilling of the fs laser in this study, it is known that laser-plasma interaction can be significantly increased when subsequent pluses hit plasma plume that is left by preceding pulse (Breitling et al. 2002). When the hole goes deeper, the laser induced plasma can be trapped inside the hole (Zeng et al. 2003) and absorb more laser's energy which can further increase the electron density and temperature of the plasma. The hot plasma may cause a considerable melting of the silicon wafer at high laser power (Breitling et al. 1999, 2001). The rapid plasma expanding (Breitling et al. 2004) and melting ejections resulting from plasma heating can brush the hole wall and enlarge the entrance hole that would lead to a larger taper and straighter hole wall.

Fig. 3.9 Cross section of the through hole at Power = 200, 300, 400, 600 mW

3.4 Summary

The effects of fs laser parameters on the laser drilled hole profiles and characteristics of spatter area formation in laser drilling of silicon were studied. This study provides a thorough description for the hole boundary propagation behaviour as well as the spatter area expansion when gradually increasing number of pulses. The spatter formation mechanism is discussed. It was also found that the minimal entrance hole diameter is produced when the beam waist is below the work-piece surface. This reveals that entrance

hole diameter is not only dependent on the size of laser energy radiation area at the surface but also highly related to the erosion of ejection material from the hole bottom. The taper of hole is increased with increasing the average laser power indicating the laser material removal mechanism for fs laser at high power levels is also dominated by the heating process which is similar to the longer pulse laser.

Chapter 4 Role of Volatile Liquids in Debris and Hole Taper Angle Reduction

4.1 Preamble

As discussed in Chapter 2.5.2, there are many unsolved issues and limitations in the area of liquid assisted laser drilling. It is an unexplored area for liquid layer thinner than 0.5 mm. In this chapter, the aim is to further improve laser drilling efficiency and reduce debris formation by applying a thinner liquid film with thickness smaller than 0.5 mm. Six types of volatile liquids are chosen to study their effect on spatter reduction and taper control. These liquids have lower surface tension than water and are more volatile in nature.

4.2 Experimental procedure

4.2.1 Experimental setup and laser parameters

The experimental set up of liquid assist laser drilling is shown in Fig. 4.1. The fs laser used for conducting experiments is same with Chapter 3. The laser power was set at 200 mW. The focal point of the laser beam was positioned on the silicon surface. The actual laser beam diameter on silicon surface with liquid film is estimated to be 50 μm . The laser

fluence applied at the silicon surface was 40.8 J/cm^2 . The number of pulses was fixed at 4000 for all the drilling experiments. The silicon wafer has a thickness of 0.725 mm. Because of the high efficiency of the fs pulse laser, the drilling time for through-hole in the silicon wafer can be as short as 4 seconds. The top and bottom of the laser drilled holes were examined using a DUV. The surrounding areas of both the entrance and the exit holes were determined via Pixcavator Image Analysis (PIA). The morphology mapping of laser drilled holes was examined by Scanning Electron Microscopy (SEM). Energy Dispersive X-ray (EDX) analysis was conducted to detect the oxygen level at the surface of the laser drilled area.

Fig. 4.1 Experimental set up of liquid assist laser drilling

4.2.2 Physicochemical properties of various liquids

Table 4.1 summarises the physicochemical properties of alcohols (methanol, ethanol, isopropyl alcohol (IPA), butanol and pentanol) used as assist liquids. They were selected based on their lower boiling points, surface tension and enthalpy of vaporization when compared to water (71.97 mN/m) which was used in the previous studies (Choo et al. 2004, Kang et al. 2006, Kang and Welch 2007). Deposition of thin liquid films on the silicon surface was achieved with the aid of a burette before the firing of the laser pulses as shown in Fig. 4.1. The thickness of liquid film is estimated to be 0.1 - 0.2mm by dividing the volume with the area.

Table 4.1 Physicochemical properties of the liquids used in laser drilling

Alcohols solvent						
	1	2	3	4	5	
						Water
	Methanol	Ethanol	IPA	Butanol	Pentanol	(Base reference)
Molecular formula	CH ₃ OH	C ₂ H ₅ OH	C ₃ H ₇ OH	C ₄ H ₉ OH	C ₅ H ₁₁ OH	H ₂ O
Surface tension (mN/m) (Dean 1999)	22.6	22.3	21.7	24	25	71.9
Density (g/cm ³) (Dean 1999)	0.79	0.79	0.78	0.81	0.81	1.00
Boiling Point (°C)(Dean 1999)	64.7	78.3	82.5	117.7	137	100
Enthalpy of vaporization (EVP)(kJ/mol) (Majer and Svoboda 1985)	35.3	38.6	47.4	52.3	57.1	40.65

4.3 Result and discussion

4.3.1 Evolution of liquid layer on silicon wafer

In the experiment, the layer thickness can hardly be measured directly due to it extremely small thickness. Thus the simple relation volume-dividing-area was used to estimate the

thickness as 0.1 – 0.2 mm. However, it can be guaranteed that the volatile liquid layer can stay on the surface during the laser exposure time because the laser irradiation time is as short as 4 seconds. The evolution of the most volatile liquid (methanol) layer on silicon wafer is shown in Fig. 4.2. In the initial time (before 12 s), the methanol spreads outward and reach maximum area in 12s. It should be noticed that the edge of the methanol layer is slightly higher than the center. The center area of methanol layer is reasonably flat due to the low surface tension. From 12s to 19s, there is no change in the area of methanol. After 19 s, the area of methanol shrinks from the boundary to centre quickly. Finally, it aggregates to several droplets. Normally, laser shooting is started on 12-13s and finish it on 16-17s. In this time duration the liquid layer is relatively stable.

During the laser drilling process, the liquid near the beam spot may evaporate quickly because of the heating effect by plasma. However the liquid cover an area about 600 mm² which is much larger than the spot size of laser beam (about 2000 μm²). The loss of liquid near the laser beam can be compensated by the surrounding liquid. Thus the laser liquid interaction can be successive. Additionally, the previous study reported during fs laser drilling process, the material remove of via-hole is largely contributed from the initial pulses (first 1000 pulse). Thus the lack of liquid in the later period of laser exposure may not have significant influence on the drilling process.

Fig. 4.2 Evolution of methanol layer on silicon wafer

4.3.2 Profile analysis of entrance hole and exit hole in various liquids

The DUV images of irradiated silicon wafer are presented in Figs. 4.3 and 4.4, in which the figures in left column are the front view of the hole, whereas the figures in right column are the back view. The figures show the comparison of the morphology of the laser drilling holes in air and in various types of alcohols, respectively. The amount of debris is

significantly reduced when methanol is used as the assist liquid. The exit hole diameter drilling in ethanol is 75% larger than that in air, which indicates that the material removal efficiency is higher by applying ethanol as the assist liquid. One possible reason for the highly efficient drilling is that the effect of plasma confinement (Kang et al. 2006) on laser ablation is enhanced due to the presence of liquid thin film. The previous study (Mak et al. 2011) reported that compared with drilling in air, the plasma size and duration are much reduced when drilling in water. The plasma cannot interact with the first laser pulse because the plasma is generated after several nanosecond. In this time the first laser pulse with 200 fs is already finished. However, it can interact with the following pulses which have interval of 1ms with the previous pulse. The shorter duration of plasma can reduce the overlap between the plasma and the following laser pulse. These factors weaken the plasma shielding effect, causing a stronger coupling of laser beam with the material. Thus, in the case of liquid environment the laser energy loss due to the plasma absorption is much reduced. More laser energy is hence being channeled deeper into the substrate. The material removal efficiency can be enhanced in liquid. On the other hand, the plasma is confined in the liquid, results in higher acoustic and shock waves than the dry condition (Dupont et al. 1995, Zhu et al. 2001). In addition, the homogenous vapour bubbles generation will happen as a result of the temperature increases in liquid induced by the heat conduction of target material (Kang et al. 2006). The bubble generation would be

accompanied with shock waves, which will provide an additional force to clean off the debris induced by laser ablation (Lu et al. 1999). The enhanced mechanical wave emission helps the removal of laser ablated materials. The amount of debris that forms on the silicon surface is therefore reduced.

In the case of using butanol and pentanol as the possible assist liquids, however they deteriorate the hole edge quality and cannot reduce the debris effectively due to their relatively higher surface tension which causes a thicker liquid film (0.18 - 0.21 mm for butanol and pentanol compared to 0.1 - 0.13 mm for methanol). The focus waist of the laser can be distorted by the thick liquid film due to the refraction and scattering of the laser beam by heat induced bubbles and curving surface and results in irregular hole profiles. As shown in Figs. 4.3 and 4.4, the debris around the hole is significantly reduced with the assist liquids such as methanol, ethanol and IPA. These liquids have relatively low values of surface tension as tabulated in Table 4.1.

Fig. 4.3 Entrance (left column) and exit holes (right column) drilled at the laser power of 200 mW in various liquids. Number of pulses: 4000. (a, b) in air; (c, d) in methanol; (e, f)

in ethanol;

Fig. 4.4 Entrance (left column) and exit holes (right column) drilled at the laser power of 200 mW in various liquids. Number of pulse: 4000. (a, b) in IPA; (c, d) in butanol; (e, f)

in pentanol

Figure 4.5 shows the hole diameters versus types of assist liquid used. There are no significant differences for entrance hole diameters when applying various assist liquids and drilling in the air. However, when comparing with the hole drilled in air, larger exit holes are produced with methanol, ethanol and IPA as the assist liquids. The exit hole diameter decreases with increasing the boiling point of the liquids. The taper of the hole is thus reduced as well with less carbon content as shown in Fig. 4.5. The taper is calculated as

$$Taper(\theta) = \tan^{-1} \frac{d_{entrance} - d_{exit}}{2t}$$

where θ is the taper angle and t is the material thickness.

Fig. 4.5 Hole diameter versus types of assist liquid in order of increasing boiling point

Fig. 4.6 Taper versus types of assist liquid in order of increasing boiling point

It is interesting that the plot of boiling point of the liquid versus the number of carbon atoms however shows a contrary trend with that of the exit hole diameter, indicating that the mechanism of liquid assist laser drilling has close relationship with the vaporization of the liquid during the laser drilling process. The liquid flow and vaporization are helpful to carry away the debris inside the hole which reduces the laser energy absorption by the molten material. Thus, more laser energy can be used to enlarge the exit hole with the presence of assist liquid. According to Fig. 4.6, the exit hole diameter decreases with increasing boiling point of the liquid, indicating that the mass removed by laser is increased when applying the liquid with low boiling point. Table 4.1 shows that the enthalpy of vaporization (EVP) of the various alcohols increases when the boiling point of alcohol solvents becomes higher.

The liquid with higher EVP leads to more heat loss during laser irradiation, reducing the temperature rise of the substrate, and thus likely limits the pressure generation during the bubble formation. This would account for the low drilling efficiency when applying liquid with high boiling point.

4.3.3 Chemical composition and microstructure of laser drilled hole in various liquids

Figure 4.7 shows the SEM images of entrance hole in air, methanol, ethanol and IPA. It can be found that the inside wall of the hole drilled in air is covered by a thick redeposition layer. However, almost no redeposition was observed around the holes drilled in liquids. In order to further investigate the surface chemical composition of the inside wall around the ablation edge, EDX was applied to determine the concentration of various elements. The star shape markings in the Fig. 4.7 are the points where the EDX data were collected. The EDX analysis in Fig. 4.8 (a) shows the sample drilled in air has relatively high oxygen concentration (63.9 atomic%), indicating that the silicon has been oxidised in air during the laser ablation and the silicon dioxide is formed. Figures 4.8 (b), (c), (d) show there is a significant decrease of the relative surface oxygen content of samples drilled in the liquids, suggesting that the oxidation of silicon is significantly suppressed by the liquid during the

laser ablation. The variation of silicon surface oxygen concentration with laser power levels in various liquid is plotted in Fig. 4.9. In Fig. 4.9, EDX data at six different points as presented in Fig. 4.7 are averaged for each sample. Figure 4.9 shows that the variation of laser power levels has no significant influence on the oxygen content of sample drilled in air. The atomic oxygen percentage at the sidewall of the hole drilled in air is varied from 64% to 68% which is close to the atomic oxygen percentage of silicon dioxide (SiO_2). This indicates that hole drilled in air is almost entirely covered by the SiO_2 . For the cases of drilling in liquids, the oxygen content of sample is reduced by about 58% when compared with that in air at 150 mW laser power. In laser ablation process, the liquid layer can insulate the oxygen in the air with the ablation area thus avoids the fast and massive oxidation of silicon at high temperature. However, the fs laser fluence as high as 20 J/cm^2 is sufficient to photodissociate the covalent bonds (O-H) (Yi et al. 2007, Yuan et al. 2008) of the alcohol liquids. This process can excite the (C-O) stretching vibration (Koretsky et al. 2001) which can increase the possibility of oxidation reaction of hot silicon atoms at the silicon-liquid interface. This is the possible reason why the oxygen element is observed on the surface of the hole drilled in liquids.

Moreover, the oxygen content increases gradually with increasing the laser power to 200mW. This increment is possibly due to the enlarged silicon-liquid reaction rate at higher laser power. Figure 4.9 shows that the oxygen content of sample in methanol is highest

among the three liquids. This may result from the relatively higher oxygen concentration (50 wt%) in methanol atoms as compared to the ethanol (34.8 wt%) and IPA (26.7 wt%).

Fig. 4.7 SEM image of entrance hole in air (a), methanol (b), ethanol (c), and IPA (d)

Fig. 4.8 The oxygen level detected by EDX of hole drilled in air (a), methanol (b), ethanol (c), and IPA (d)

Fig. 4.9 Variation of silicon surface oxygen concentration with laser power in various liquids

From Fig. 4.7 (b), (c) and (d), it can be observed that there are some sub-micron features in the inside wall of laser drilled hole. The detailed magnification of the Fig. 4.7 (b) is shown in Fig. 4.10. As highlighted in the white ellipse of Fig. 4.10, the parallel sub-micro structures with same spatial interval is observed. This features that are resulted from the interference between incident laser beam and surface plasmon are named laser induced period surface structures (LIPSS). In the next chapter, a detailed study is conducted to

investigate the formation of LIPSS and the liquid medium's influence on the properties of LIPSS.

Fig. 4.10 Detailed magnification of the SEM image of laser drilled hole wall in ethanol

4.4 Summary

This chapter provides a study about the role of different types of volatile liquid films in reducing debris formation during fs laser drilling of silicon. It was found that the more volatile liquids, i.e. liquids with lower boiling points and lower number of carbon atoms in the molecular formula, were more effective in reducing debris formation as well as

reducing the taper angle of the fs laser drilled holes in silicon. These liquids have low boiling points and are more volatile. Methanol, ethanol and IPA solvents are the examples of such liquids. The confinement of plasma and the strong shock waves induced by bubbles are the main reasons for the effective removal of debris. The volatile liquids also led to reduction in oxygen concentration on the inside wall of laser drilled hole.

Chapter 5 Refining Laser Induced Periodical Surface Structures with Liquid Assist

5.1 Preamble

In this chapter, experiments on silicon wafer were conducted using fs laser to generate periodic surface structures. The experiments were conducted using both air and ethanol as a medium. The spatial wavelength and cleanliness were studied. The effect of each medium on the surface structure generated was also investigated and analysed.

5.2 Material and method

The laser setup used in this chapter is the same with Chapter 3. The laser beam was focused with a focusing lens of 75 mm focal length. In current study, LIPSS was produced on polished single crystal silicon wafer. The pulse energy applied at the silicon surface was 7 μJ . The number of pulses was varied from 1 to 500. Deposition of thin liquid films on the silicon surface was achieved with the aid of a burette before the firing of the laser pulses. The thickness (assumed uniformly distributed here with relatively low surface tension) of liquid film was estimated to be 0.1 - 0.2 mm by dividing the volume with the area. The micro-structural features of the laser irradiated area were examined by scanning electron microscope (SEM) and the surface topography and cross section profile of the sample were

inspected by the atomic force microscope (AFM) in tapping mode. The probed area was $5 \times 5 \mu\text{m}^2$ for the AFM measurements.

5.3 Result and discussion

Figure 5.1 (a) shows the SEM image of a silicon surface area irradiated in air by 100 laser pulses at a fluence of 0.2 J/cm^2 which is lower than the single pulse threshold of 0.4 J/cm^2 (Ngoi et al. 2001). As shown in Fig. 5.2 (c), a magnified image of the ripple area is acquired in the center of machining spot (zone D). The observation reveals that, with spatial periods of 630 nm (less than laser wavelength of 775 nm) ripples are formed perpendicular to the direction of the polarization of the laser irradiation. These ripples are characterised by nearly parallel lines extending over the entire area despite some melting zones *i.e.* A, B, C as shown in Fig. 5.1(a), which are due to the accumulation of thermal effect from the intensive energy of hot spot in the laser beam. This hot spot is caused by the non-uniform energy delivered by the laser beam. A more uniform energy distribution can be achieved either by tuning the optics or by using improved apertures. The ripple area is covered by a large amount of debris coming from the redeposition of ejection materials. Figure 5.1(b) and Fig. 5.2(d) present the LIPSS on silicon irradiated in ethanol environment and when applied the exact same laser parameters as in the air. There are two distinct differences in the features of LIPSS between the air and ethanol environment.

(a)

(b)

Fig. 5.1 SEM image of LIPSS formed in air (a) and in ethanol (b) by the p-polarized laser beam with 100 pulses at pulse energy of 7 μ J

Fig. 5.2 Detailed magnification of the SEM image of LIPSS in air (left column) and in ethanol (right column) drilled at the laser fluence of $0.16\text{J}/\text{cm}^2$ in various number of pulses: (a), (b) 10; (c), (d) 100;

Firstly, the ripple area in liquid is much cleaner than that in air. The amount of debris is significantly reduced when ethanol is used as the assist liquid. The vapour bubbles were produced as a result of the temperature rise in the liquid induced by absorbed laser energy (Jiao et al. 2011). The bubble generation would be accompanied with shock waves, which

will provide an additional force or source to clean off the debris as induced by laser ablation (Lu et al. 1999). The enhanced mechanical wave emission helps in the removal of laser ablated materials. The amount of debris on the silicon surface is therefore reduced. According to the literature (Wee et al. 2011), the dimension of bubbles generated by fs laser was estimated below $1\mu\text{m}$. These micro-size bubbles were observed around the peripheral of the laser irradiated area (not in the laser beam path). Thus, the bubble's scatter effect of laser beam was not observed.

Secondly, as observed from Fig. 5.2, when the number of pulse is fixed at 10, the ripple period LIPSS in ethanol is 455 nm that is about 40% less than that in air (772 nm) and the laser wavelength (775 nm). Figure 5.3 shows the ripple's profile at 200 and 300 pulses. For the same pulses, it can be observed that the ripples produced in ethanol have shorter wavelength than as compared to air.

Fig. 5.3 Detailed magnification of the SEM image of LIPSS in air (left column) and in ethanol (right column) drilled at the laser fluence of $0.16\text{J}/\text{cm}^2$ in various number of pulse: (a), (b) 200; (c), (d) 300

Moreover, the AFM analyses (Fig. 5.4) of the LIPSS and the cross section profiles (Fig. 5.5) confirm the SEM observation that the spatial wavelength is reduced by 30% in ethanol environment when number of pulse is increased to 100. This result shows that the change of environment medium plays an important role in the formation of LIPSS. For normally incident p-polarized laser beam, the period of surface ripples resulting from the interference

Fig. 5.4 Topographical profiles of LIPSS in air (a) and ethanol (b) at laser pulse energy of

7 μJ

Fig. 5.5 Cross section profiles of LIPSS in air (a) and ethanol (b) at laser pulse energy of

$$7 \mu\text{J}$$

between the incident laser light and the excited surface plasmon wave is given by Eq. (5.1):

$$\Lambda = \lambda / \eta \quad (5.1)$$

where λ is the incident light wavelength, $\eta = \text{Re}[\varepsilon_1 \varepsilon_2 / (\varepsilon_1 + \varepsilon_2)]$ [10] is the real part of the effective refractive index of interface for surface plasma, ε_1 is the complex dielectric

constant of material, ε_2 is the complex dielectric constant of medium. The complex dielectric function is given as $\varepsilon = (n + ik)^2$. When ethanol is applied as medium instead of air, η is calculated to be 1.75 for $\varepsilon_1 = 13.8 + 0.059i$, $\varepsilon_2 = 3.5 + 2.1i$ (Kitahara et al. 2005). The ripples period Λ is found to be 442 nm which is rather close to the experiment result of 455 nm. When this model is applied to calculate the period of ripples formed in air, the value of spatial wavelength is 780 nm which approximates the experiment result of 772 nm with the number of pulse 10. However, the spatial wavelength decreases sharply with increasing the number of pulse to 100 as shown in Fig. 5.6. This discrepancy is because this model neglects the effect of transient increment of laser induced electron density in conduction band which will lead to an increase of real part of refractive index (Vorobyev et al. 2007). The LIPSS spatial period decreases when increasing the η according to the Eq. (5.1). Nevertheless, for the case of ethanol environment, there is no apparent change in the spatial wavelength when the number of pulse increases from 10 to 400. One possible reason for this phenomenon is that the effect of plasma confinement (Kang et al. 2006) is enhanced due to the presence of liquid thin film.

The previous study (Mak et al. 2011) reported that compared with drilling in air, the plasma size and duration are much reduced when laser irradiating in water. Thus, in the case of liquid environment the temperature rise in the ablation area due to the heat conduction from the plasma is much reduced. This process can prevent the sharp increase of electron density

in conduction bond when applying more laser pulse. Thus, the spatial wavelength can remain almost unchanged with the assistance of ethanol.

Fig. 5.6 Dependence of the LIPSS wavelength as function of the number of pulses per spot

5.4 Summary

In conclusion, laser induced periodic surface structures were generated on silicon wafer using fs laser below the single pulse damage threshold. The medium used in this study is both air and ethanol. The laser process parameters such as wavelength, number of pulse,

laser fluence were kept constant for both the mediums. The focus of the study is to analyse the spatial wavelength. It was found that as compared with LIPSS formed in the air, ethanol environment can produce cleaner surface ripples with a period of 450 nm which is far less than the wavelength of incident laser beam. The cleanliness of the surface generated using ethanol showed considerably less debris than in air. The results observed from the above investigation suggested that the medium plays a predominant role in the generation of surface structures. The bubble generation process is responsible for the significant decrease of debris on the surface. The reduction of spatial wavelength of LIPSS in ethanol environment can be attributed to the increase of real part of the effective refractive index.

Chapter 6 Effect of Substrate Heating on Hole Geometry and Spatter Area

6.1 Preamble

In laser-material interactions, the optical absorption coefficient is an important factor that determines how much laser energy is coupled into the substrate for the material ablation process. The silicon band-gap energy, as a function of temperature, decreases with increasing temperature. In other words, the optical absorption coefficient of silicon increases with temperature (Thurmond 1975). It is primarily for this reason, a heating device was developed to pre-heat the silicon substrate during the laser drilling process. As mentioned in Chapter 2.6, there are some studies which have considered the environmental temperature's influence on surface roughness of laser grooving on various materials (Yahng et al. 2009, Yahng and Jeoung 2011), laser ablation threshold of silicon for one pulse (Thorstensen and Erik Foss 2012). However, research working on the interaction of work-piece temperature and laser ablation for the case of deep through hole cannot be found. In this study, the objective is to evaluate how the enhanced optical absorption of the laser energy at the elevated substrate temperature would affect the hole geometry and the spatter area around the drilled hole.

6.2 Experimental set-up

6.2.1 Laser parameter and heater

Fig. 6.1 shows the experimental arrangement used in the present study. The detail of laser setup used in this chapter was described in Chapter 3. The configuration of heater was described in Appendix. The laser beam was focused with a focusing lens of 75 mm focal length.

Fig. 6.1 Pre-heating fs laser processing setup

In the current study, through holes were produced on polished single crystal silicon wafer which was fixed on the stainless steel block with two heaters inside. The temperature of

silicon wafer ranging from 300 K to 900 K in a step of 100 K was monitored by two calibrated thermocouples. The maximum laser fluence applied at the silicon surface was 40.8 J/cm^2 . The number of pulses was varied from 10 to 100 in a step of 10 and from 2000 to 5000 in a step of 1000 for each temperature step. In the experiment, the drilled sample underwent acid etching by hydrofluoric acid (HF) to remove the spatter around the hole and make the boundary distinguishable (as shown in Fig. 6.2). It is true that the hole profile is not regular due to the non-uniform energy delivered by the laser beam. In order to evaluate the hole parameter objectively, the image analysis software was applied to capture the edge of the hole and calculate the area of the hole by integrating the boundary line (the green line in the Fig. 6.2 and the red line in Fig. 6.3). Average diameter can be obtained from the area of the hole assuming the hole has perfect circularity.

Fig. 6.2 The profile of entrance hole with post-processing of acid etching and the edge line captured using image analysis software

List of objects				
ID	Type	Location X	Location Y	Size
☒ 1	D	205	80	3962
☒ 2	D	448	85	2196
☒ 3	D	326	75	3552
☒ 4	D	77	85	3053

Fig. 6.3 The profile of exit hole with edge capture line using image analysis software and the list of calculated area by integrating the boundary line

6.2.2 Calibration of the beam waist position at high temperature

In this experiment, the silicon wafer with size of 8mm×8mm was mounted on the stainless steel block which had two heating elements and one thermocouple embedded inside. The surface temperature of silicon wafer was measured by another thermocouple to ensure the silicon wafer can reach the intended setting temperature. In order to avoid the thermal damage to the CNC system at the high temperature, the alumina refractory block with

thickness of 100 mm was set as thermal insulator beneath the heating device. However, the thermal expansion of the alumina refractory at higher temperature can cause the displacement (ΔZ_I) of silicon wafer in the perpendicular direction which is shown in Fig 6.4. From Chapter 3, it is known that the entrance hole diameter is sensitive to Z_f . The entrance hole diameter increased from 50 μm to 60 μm when Z_f was increased by 250 μm from the beam waist position. (Fig. 3.5, Page 65) At the elevated temperature of 900 K, the silicon wafer would be elevated by 300 μm considering that the thermal expansion coefficient of alumina is $5.4 \times 10^{-6} \text{ K}^{-1}$ (Ho and Taylor 1998). Thus, the defocus of the laser beam on silicon wafer caused by the thermal expansion of the thermal insulation can be the main noise or error factor in this experiment. To offset this displacement, it is feasible to elevate focus lens controlled numerically by an arm whose positional accuracy is 2 μm . As shown in Fig. 6.4, ΔZ_2 has to be accommodated by a distance that is equivalent to ΔZ_I .

Fig. 6.4 The defocus caused by thermal expansion and offset method by elevating focus

lens

Therefore, it is essential to conduct the calibration experiment to measure the silicon wafer displacement ΔZ_l at each temperature increment step in order to ensure that the beam waist is maintained at the identical plane of silicon wafer surface. It is inappropriate to use the slide caliper rule to measure a small displacement at high temperature. Thus, the Scribing Lines method is applied to find the Z position of silicon surface at each temperature step as follows: the approximated beam waist position was calculated based on the thermal coefficient of alumina at one elevated temperature. Around the estimated beam waist, 20 lines (portion of them are shown in Fig. 6.5) were scanned at various heights at a step of

50 μm . As illustrated in Fig. 6.5, the line with minimum width among these lines is corresponding to the intended realistic beam waist. By using this method, ΔZ_I can be obtained at 300 K, 400 K, 500 K, 600 K, 700 K, 800 K and 900 K accordingly.

Fig. 6.5 Illustration of Scribing Lines method used to find the minimum beam waist

The expansion values in the vertical direction at temperatures ranging from 400 to 900 K were recorded and plotted in Fig. 6.6. The thermal expansion rate that obtained from the regression of the data in Fig. 6.6 is $6.6 \times 10^{-6} \text{ K}^{-1}$ that is close to the linear expansion coefficient of alumina that of $5.4 \times 10^{-6} \text{ K}^{-1}$. (Ho and Taylor 1998)

Fig. 6.6 Expansion of the insulator versus temperature

6.3 Result and discussion

6.3.1 Entrance hole

Figure 6.7 exhibits the front side of the micro-holes drilled by fs laser with laser fluence of 20.4 J/cm^2 at various temperatures. The diameter of the entrance hole increases by 25% at the 873 K when comparing to that at 300 K. In this study, the photon energy of 775nm laser

is below the direct band gap of silicon (3.4eV) (Jellison Jr and Modine 1982). Therefore, the main mechanism of electron excitation in the silicon is through the indirect band gap transition. In this process, the optical absorption property of silicon highly depends on the number of acoustic phonons which is a function of temperature (Pankove 1975). At elevated temperature, there are more acoustic phonons, thus there is more possibility that an acoustic phonon in the lattice and a photon from the laser irradiating can be simultaneously absorbed to create an indirect transition. As a result of a large number of electrons photoexcited from valence band into conduction band, a considerable amount of covalent bonds are destroyed (Chen et al. 2005) and, multi-ionization occurs (Gattass and Mazur 2008). Hence, at elevated temperature the pressure from the coulomb explosion (Mao et al. 2004) ejects more ion and atom clusters that account for the more material damage in the early stage of multi-pulse laser ablation. It is reasonable to conclude that the higher substrate temperature causes a larger entrance hole diameter.

Fig. 6.7 SEM images of the micro-hole (front side) drilled by fs laser at temperature 300

K (a), 400 K (b), 500 K (c), 600 K (d), 700 K (e), 800 K (f), 900 K (g)

On the other hand, it is understood that under the high temperature, due to comparatively higher optical absorption coefficient of silicon as well as the decrease of the optical penetration depth, more energy from the laser will focus in a thinner layer at the top silicon surface. This results in larger material removal rate in the vertical direction which causes a larger ablation depth. Fig. 6.8 compares the geometry of laser drilling hole at 300 K, 600 K and 900 K. In order to investigate the temperature' influence on hole geometry at very early stage of the hole formation, low number of pulses in terms of 10, 20, 30 are chosen. Figure 6.8 shows that at 900 K the depth of hole is larger than that at 300 K. The wall taper of drilled hole is smaller at elevated temperature which means the hole depth is shallower at 300 K.

Fig. 6.8 Comparison of laser drilling hole at room temperature (left column) and at 873K (right column) with number of pulse of 10(a,b), 20(c,d), 30(e,f)

Fig. 6.9 The entrance hole diameter as a function of substrate temperature at average laser power of 200 mW, 300 mW and 400 mW, respectively

Figure 6.9 represents the values of the entrance hole diameter at various laser power levels as a function of the substrate temperature from 300K to 873K. The variation of temperature below 500 K has no significant effect on the entrance hole diameter. However, when the temperature was raised to 600 K the entrance hole diameter sharply increases. It was reported (Jellison Jr and Modine 1982) that the optical absorption coefficient silicon at 694 nm has a precipitous increase around 573 K when temperature increases. This is the possible reason for sharp increase of the entrance hole diameter at 600 K.

6.3.2 Exit hole

Meanwhile it is observed that the exit hole diameter produced at the elevated temperature is larger than that under the room temperature as shown in Fig. 6.10. At the laser fluence of 20.4 J/cm^2 , the exit hole diameter drilled under the elevated temperature of 900K is more than 30% of that drilled under the 300K as shown in Fig. 6.10. It is also observed that less number of pulses are needed to penetrate the silicon thickness at higher temperature. It is known that during the percussion laser drilling, the material is removed layer by layer for each laser pulse. As mentioned above, under the elevated temperature, a less tapered hole is obtained after the irradiating of first several pulses. It was reported (Ruberto et al. 1991) that the laser drilling hole at initial stage acts as a waveguiding for the successive laser pulses. In the waveguiding process, less tapered angle wall is helpful in delivering the laser beam to the deeper position. Researchers (Zheng et al. 2007) reported that multiple reflection of following laser beam in the shallow hole cause more serious energy loss due to the absorption of the laser power by hole wall. That means at the elevated temperature, the relatively straight wall of drilled hole delivers more laser energy to the bottom of hole, resulting in the larger exit hole.

Fig. 6.10 SEM images of the micro-hole (back side) drilled by fs laser at temperatures of

(a) 300 K, (b) 400 K, (c) 500 K, (d) 600 K, (e) 700 K, (f) 800 K, (g) 900 K

Figure 6.11 represents the values of the exit hole diameter at various laser power levels as a function of the substrate temperature from 300 K to 873 K. Generally, the exit hole diameter gradually increases with increasing substrate temperature. The influence of temperature increase can be reduced due to considerable laser energy loss which comes from the multi-reflection of laser beam on the hole wall. Therefore, no sharp increase of exit hole diameter is observed.

Fig. 6.11 The exit hole diameter as a function of substrate temperature at average laser powers of 200 mW, 300 mW and 400 mW, respectively

6.3.3 Spatter area

As the ablation products, the debris is hardly avoided when silicon is machined by fs laser in air environment. In the initial stage of the laser ablation, the formation of debris originates from the atoms and clusters due to the coulomb explosion (Matsumura et al. 2005). Thereafter, the recoil pressure from the fast evaporation in the molten layer provides the energy to eject the hot atom cluster and liquid droplet. The recoil pressure (Lee and Jeong 2004) can be expressed:

$$P_r = 0.56P_s(T_s)$$

P_s is the saturated vapour pressure at T_s . In the case of elevated temperature, P_r is expected to be higher due to larger T_s . The increase of P_r results in a stronger force to push away the debris and produce a larger spatter area. However, the observation from Fig. 6.12 shows a contrary result when elevating the baseline temperature. Fig. 6.12 indicates that the spatter area is dramatically reduced by half with increasing of the substrate temperature from 300 K to 773 K at various number of pulse and power density. It is reasonable to explain the mechanism for the spatter area reduction at the elevated temperature in terms of the enhanced energy absorption. For the multi-pulses laser ablation, the laser radiation is strongly absorbed with the laser induced plasma (Kang et al. 2006). As the drilling depth grows, part of the plasma plume is trapped in the laser drilling hole (Luft et al. 1997) and become a semi-medium between the laser beam and silicon surface. In this situation, the material

removal mechanism consists of laser ablation, plasma etching and joule heating from the heat conduction of the hot plasma. During this process, coupled with the thermal energy supplied due to the substrate heating, the energy from plasma joule heating may be sufficient to melt a larger amount of the material. This would produce larger liquid droplets with increased gravity that can not be pushed so far as the small particles do, even if the recoil pressure is growing. The amorphisation and redeposition of these liquid droplets accumulates at the periphery of the hole and forms the spatter. As seen from the Fig. 6.7, the particle size in the spatter formed at 900 K is significantly larger than that formed at 300 K. The granular structure around the hole may indicate the presence of liquid material during the laser process. In case of laser drilling under the elevated temperature, coarser pillars can be found in the granular structure. This may indicate the forming of melting droplets before their resolidification.

Fig. 6.12 The spatter area as a function of substrate temperature at number of pulse

(NOP) of 2000, 3000, 4000, 5000 with various power densities

6.4 Summary

The fs laser drilling on silicon was investigated at various elevated substrate temperatures.

The results showed that the entrance hole diameter was increased by 25% and the exit hole was increased by 30% when the substrate temperature was elevated to 900 K. The laser drilling efficiency was greatly increased by elevating the temperature. This high drilling efficiency is attributed to the enhanced laser energy absorption of silicon wafer and

thereafter wave guiding effect. The spatter area was found, however, continuously decreased with increasing the substrate temperature. A large droplet trace was found around the ablation area which indicated liquid phase was increased by the joule heating from the heat conduction of the hot plasma.

Chapter 7 Theoretical Study of Hole Geometry's Influence on Laser Energy Distribution

7.1 Preamble

For percussion laser drilling, the material inside the micro hole is removed pulse by pulse. However, in the laser drilling process, the taper (Ghoreishi et al. 2002) , barreling shape (Tan 2006) and tail shape (Döring et al. 2011) of the hole can hardly be avoided owing to the power intensity distribution of the Gaussian beam and plasma expansion when the hole is going deeper. Because of the electromagnetic nature of the fs laser, the hole geometry formed by the previous laser pulses will interact with the successive pulses. The absorption, reflection, scattering, transmission at every point of the hole wall will significant influence in the following propagation behaviour of laser pulse. The various shapes of the hole wall with different angles may act like wave guide (Ruberto et al. 1991) for laser beam or just cause energy loss with absorption. Accordingly, due to the multi-reflection on the hole wall, the energy density distribution of the laser pulse inside the hole exhibits distinctive structures with Gaussian distribution at the orifice of the hole. In the later stage of the laser percussion drilling, the amount of laser energy that can reach the bottom of the hole decides the laser drilling depth. In other words, the aspect ratio and drilling efficiency of the hole

is highly related to the hole geometry at the beginning of the laser ablation. However, direct experimental study of the hole wall's influence on the laser drilling depth and laser energy propagation is rare because of the dynamic nature of the laser drilling process and the lack of in-situ characterisation tools for the micro features. Thus, it is of great importance to conduct a simulation to investigate the influence of geometrical effects on the laser beam propagation and subsequent laser ablation. In order to fundamentally understand the laser beam propagation behaviour in the micro hole, it is essential to solve the Maxwell's equations which describe the nature of the laser beam.

The limitation of previous works involving hole wall's effect on laser beam propagation is summarised and discussed in Chapter 2.7.4.

7.2 Methodology

In this chapter, the steady state analysis of energy distribution of fs pulse laser beam in the micro hole was conducted by numerically solving time harmonic Maxwell's wave equation (Eq. 7.1). FEM analysis was carried out using COMSOL Multiphysics software (COMSOL 2013). Moreover, the taper angle of hole wall was varied to investigate the influence of multi-reflection and thereby interference of the laser beam on the laser energy distribution at the bottom of the micro hole in silicon.

7.2.1 Model scheme

Figure 7.1 shows the schematic view of the model setup. The laser beam propagation inside the micro-hole was numerically simulated by solving the three dimensional time harmonic Maxwell's wave equations (Eq. (7.1)). There are two regions in computational domain, namely air domain and silicon domain representing the micro-hole in the silicon wafer. The incoming laser beam was applied at the top. The silicon electromagnetic properties used in this model are listed in Table 7.1.

Table 7.1 Electromagnetic properties of silicon at the wavelength of 775 nm

Property	Symbol	Value	Reference
Refractive index	n	3.71	(Palik 1999)
Refractive index, imaginary part	$k_{imaginary}$	0.008	(Palik 1999)
Relative permeability	μ_r	1	(COMSOL 2013)
Relative permittivity	ϵ_r	13.8	(Palik 1999)
Electric conductivity (S/m)	σ	1×10^{-12}	(Serway 1998)

Fig. 7.1 Side view of the computational domain

7.2.2 Assumptions

There are no surface charges on the inside of the micro hole.

The laser induced rejection materials and plasma gas have no interaction with the electromagnetic field.

The inside wall of the micro hole is smooth and the liquid phase layer is neglected.

All variations of electromagnetic field in time occur as sinusoidal signals.

7.2.3 Governing equation

The phasor form of time harmonic wave equation derived from the Maxwell's equations can be written as Eq. (7.1) (Courtois et al. 2013).

$$\nabla^2 \vec{E} - \mu_r k_0^2 (\epsilon_r - \frac{j\sigma}{\omega\epsilon_0}) \vec{E} = 0 \quad (7.1)$$

where $\vec{E}$ is the electric field, k_0 is the wave number of free space $k_0 = \frac{\omega}{c_0} = \omega\sqrt{\epsilon_0\mu_0}$, ϵ_0 is electric permittivity, μ_0 is magnetic permeability, c_0 is the optical speed, ω is the frequency of laser, μ_r is the relative permeability, ϵ_r is the relative permittivity, σ is the electrical conductivity.

The laser intensity I that is also called irradiance is most commonly used to describe the laser beam power flow in the laser related numerical work involving energy transfer. It can be obtained from the Poynting's theorem. The Poynting vector $\vec{S}$ stating the rate of flow of electromagnetic energy per unit area is given by Eq. (7.2) (Schwartz 1987, Fowles 1989).

$$\vec{S} = \vec{E} \times \vec{H} \quad (7.2)$$

where the magnetic field $\vec{H}$ can be obtained from Faraday's law (Eq. (7.3)).

$$\nabla \times \vec{E} = -j\omega\mu_0\vec{H} \quad (7.3)$$

The intensity is given by Eq. (7.4).

$$I = |\vec{S}| = |\vec{E} \times \vec{H}| \quad (7.4)$$

7.2.4 Laser source

In this work, a fs laser with the wavelength of 775 nm and pulse duration of 250 fs is assumed. The laser beam is assumed as linear polarized transverse electric wave which was focused by a lens at the top of computational domain. The polarization direction is along the x direction in Fig. 7.1. The electric field in phasor form describing the laser beam at the plane of the silicon surface is given by Eqs. (7.5-7.7) (Tao et al. 2011).

$$E_x = Ae^{j\omega t} \quad (7.5)$$

$$E_y = 0 \quad (7.6)$$

$$E_z = 0 \quad (7.7)$$

The amplitude is given by Eq. (7.8).

$$A = E_0 \exp\left(-\frac{x^2+y^2}{r_0^2}\right) \quad (7.8)$$

The frequency is given by Eq. (7.9).

$$\omega = \frac{2\pi}{\lambda} c \quad (7.9)$$

where E_0 is the peak power of the laser pulse, r_0 is the Gaussian beam radius, λ is the laser wavelength. E_x , E_y and E_z are the components of electric field in the x, y and z direction, respectively.

7.2.5 Boundary conditions (BCs)

Boundary B is the interface between air and silicon (Fig. 7.1). In order to predict the reflection and refraction at this boundary, the tangential component of electric field across the interface is continuous. Meanwhile, tangential component of magnetic potential across the interface is also continuous because of the assumption of zero surface charges. The BCs at boundary A can be mathematically expressed as Eq. (7.10, 7.11) (Jackson 1999) for electric and magnetic field, respectively. $\hat{n}$ is the unit normal vector. The subscripts ‘1’ & ‘2’ denote the respective air domain and silicon domain.

$$\hat{n} \times \vec{E}_1 - \hat{n} \times \vec{E}_2 = 0 \quad (7.10)$$

$$\hat{n} \times \vec{H}_1 - \hat{n} \times \vec{H}_2 = 0 \quad (7.11)$$

Boundary A is the port for the input of the laser beam. The expression for the BCs in electric field form that can be found in the Section 7.2.4 of laser source (Tao et al. 2011).

Boundary C is the port for releasing the electromagnetic field. In order to allow all the waves to penetrate this boundary and no waves to reflect back to the computational domain, the scattering boundary condition expressed in Eq. (7.12) is selected (Chen and Zhan 2007).

$$\mu_0 \hat{n} \times \vec{H} - k_0 / \mu_r \hat{n} \times (\vec{E} \times \hat{n}) = 0 \quad (7.12)$$

7.2.6 Grid generation

This study is aimed to simulate the propagation of electromagnetic field of laser in the micro scale. In order to obtain numerical stability and sufficiently high spatial resolution, the element size is chosen to be less than $\lambda/5$ in the air domain (Marburg 2002, Marburg and Schneider 2003). A finer mesh is required in the silicon domain due to the shortened wavelength when light is coming into the solid material. Thus, in the case of silicon domain, the element size is less than $\lambda/(5n_{\text{silicon}})$, where n_{silicon} is the refractive index of silicon at 775 nm. As shown in Fig. 7.2, the free tetrahedral mesh elements were constructed by COMSOL native mesh generator (COMSOL 2013). The maximum element size was adjusted to control number of elements in each domain.

Fig. 7.2 Multi-domain grid used for the 3-D model with 681542 tetrahedral elements

7.2.7 Mesh independent study

A mesh invariant study was conducted to determine the number of mesh elements that is necessary to produce acceptable numerical results which are mesh insensitive. This is crucial to guarantee that the electric field energy over the domain is modeled accurately. Nine sets of the meshes that varied from coarse to fine were examined. The total number of the mesh elements used in each set is specifically given by 66867, 151814, 223070, 315206, 389266, 472869, 570403, 681542 and 804779. The power outflow at boundary B in Fig. 7.1 is used to check for the convergence of the numerical solution. Figure 7.3 shows the results obtained from the mesh independent study. The points indicate the power outflow by time averaging at boundary B for each set of mesh. It is observed that numerical solution can be considered to achieve convergence as the number of element increases to 681542. It is thus reasonable to conclude that the numerical scheme developed in the previous section obtains mesh-independent solution when choosing mesh element of more than 681542.

Fig. 7.3 Result of mesh element independency study

7.3 Validation of Fresnel's law

In this study, the reflection and transmission at the interface is of great importance because they are the basis to describe the complicated propagation behaviour of the electromagnetic in micro-hole. Thus, it is essential to conduct a validation in a simple computational domain to evaluate the credibility of the boundary condition at the silicon\air interface with complex physics. The Fresnel equations (Hecht 2002) are used widely to predict the reflection and transmission behaviour of light at uniform planar interface. The equations

assume the interface is flat, planar, and homogeneous. The light is assumed to be plane wave.

From the Fresnel's law, the analytic form of reflectance R and transmittance T of the s-polarization light at the interface of two mediums are given by Eq. (7.13, 7.14).

$$R = \left(\frac{n_1 \cos \theta_i - n_2 \cos \theta_t}{n_1 \cos \theta_i + n_2 \cos \theta_t} \right)^2 \quad (7.13)$$

$$T = \frac{n_2 \cos \theta_t}{n_1 \cos \theta_i} \left(\frac{2n_1 \cos \theta_i}{n_1 \cos \theta_i + n_2 \cos \theta_t} \right)^2 \quad (7.14)$$

where n_1 and n_2 are the refractive indices of medium 1 (air) and 2 (silicon), θ_i and θ_t is the incident angle and refractive angle, respectively.

Fig. 7.4 Side view illustration of model to evaluate the reflectance and transmittance at the interface between air and silicon with laser beam incident angle of θ

Figure 7.4 represents the model used to determine the reflectance and transmittance at air/silicon interface with laser beam incident angle of θ . The electric field representing the laser beam is excited at the top boundary. Portion of the laser beam is reflected at interface and enters the right boundary (this boundary is defined as boundary 1). The rest of the electric field penetrates the interface and gets into the silicon domain (the air/silicon interface is defined as boundary 2). Assuming the electromagnetic field is in stationary state, the laser energy flow $I(x, y, z)$ at the boundary is obtained from the Eq. (7.5).

The outward energy flows by time averaging at boundary 1 and boundary 2 are given by Eqs. (7.15, 7.16).

$$\dot{Q}_{out,1} = \iint_{s_1} I(x, y, z) dS \quad (7.15)$$

$$\dot{Q}_{out,2} = \iint_{s_2} I(x, y, z) dS \quad (7.16)$$

The reflectance at the air/silicon interface is given by Eq. (7.17).

$$R = \frac{\dot{Q}_{out,1}}{\dot{Q}_{out,1} + \dot{Q}_{out,2}} \quad (7.17)$$

The transmittance at the air/silicon interface is given by Eq. (7.18).

$$T = \frac{\dot{Q}_{out,2}}{\dot{Q}_{out,1} + \dot{Q}_{out,2}} \quad (7.18)$$

In the simulation, the angles of interface plane were varied from 10 to 80 degree at a step of 10 degree. The numerical results of reflectance and transmittance at these angles were compared with the analytical solution obtained from Fresnel laws. The comparison between numerical and analytical solution is plotted in Fig. 7.5. In Fig. 7.5, the points obtained from

numerical solution as a function of incident angles show good agreement with the curve acquired from Fresnel equations. The maximum discrepancy is within 3%.

Fig. 7.5 Comparison of numerical and analytical solution of the reflectance and transmittance at silicon air interface

7.4 Result and discussion

In most of the previous studies (Matsunawa and Semak 1997, Semak et al. 1999, Ki et al. 2002, Nedialkov et al. 2007) involving the 3-D or 2-D axial symmetry modeling of laser ablation, the energy of laser source term is commonly considered as Gaussian distribution.

However, what exactly happens for the laser power distribution inside the hole and at the bottom surface is still an unsolved problem. In this study, electric field and magnetic field are obtained by solving the Maxwell's wave equation inside the micro-hole of silicon wafer. The laser beam intensity is calculated by using Eq. (7.4). Fig. 7.6 shows the laser intensity by time averaging (in one optical period) at the bottom of the micro-hole ($z = 0.75 \mu\text{m}$) with varying the taper angle. As shown in Fig. 7.6, the laser intensity distribution varies dramatically when the taper angle increases from 0 degree to 53 degree. The laser beam intensity pattern turns to be very complex when the taper angle is increasing to above 46 degree. Although the silicon can be considered as the transparent medium for the infrared light spectrum, the refractive index of the single crystal silicon is 3.71 (Palik 1999) at a wavelength of 775 nm which indicates that the reflection of optical wave at the air/silicon interface is much stronger than the other transparent material such as glass and polymer (Palik 1999). Thus, a considerable portion of the laser beam is reflected into the bottom by the hole wall. This result shows that reflection of the hole wall has a significant influence to the distribution of the laser energy inside the micro-hole.

Fig. 7.6 Laser intensity (W/m^2) at the bottom of the micro-hole at taper angles of 0 (a), 15 (b), 28 (c), 38 (d), 46 (e), 53 (f) degree

Figure 7.7 shows the maximum laser intensity (MLI) at the bottom of the micro-hole at various taper angles. MLI is defined as the maximum value of calculated laser intensity.

As indicated in Fig. 7.7, the MLI firstly increases slowly with increasing the taper angle until it reaches the peak point at 34 degree. Then, the MLI shows a sharp decrease when taper angle is further increased to 53 degree.

Fig. 7.7 MLI at the bottom of the micro-hole at various taper angles

The critical taper angle in terms of laser beam propagation direction can be calculated using the Eq. (7.19) which is obtained from the geometrical relation of the entrance hole diameter $d_{entrance}$, hole depth d and taper angle $\theta_{critical}$ as shown in Fig. 7.8.

$$\tan(\theta_{critical}) + \tan(2\theta_{critical}) = \frac{2d_{entrance}}{d} \quad (7.19)$$

It can be calculated that the 33 degree is the critical taper angle when $d_{entrance}$ is 3.6 μm and d is 0.75 μm which is the value obtained from our previous experiment in Chapter 6 and used in the current model. This critical value is close to 34 degree which is the peak point of the MLI curve in Fig. 7.7. Figure 7.8 illustrates the comparison of multi-reflections of the laser beam on the hole wall with taper angles of 25, 33, 40 degree, respectively.

Fig. 7.8 Illustration of multi-reflection of incoming laser beam on hole wall with a taper angle of 25, 33, 40 degree, respectively

The laser wave that interacts with hole wall for taper angle smaller than 33 degree can propagate to the bottom of the hole and contribute to energy concentration at hole bottom.

It is possible that the hole wall acts as waveguide which is helpful to deliver the laser energy to the bottom under these circumstances (Zheng et al. 2007). However, when taper angle is larger than 33 degree, a large portion of the laser energy can be reflected back to the air or

strongly absorbed by the side wall due to increased transmittance resulting from the small incident angle on the hole wall. This can lead to lower MLI as shown in Fig. 7.7.

It also can be observed from Fig. 7.6 that the laser intensity shows a ring shape when taper angles are 46 and 53 degree. In these two cases, laser intensity at center of the bottom is much lower than the surrounding ring shape area which is contrary to the assumptions in most of the previous modeling works where the laser intensity is always assumed Gaussian distribution. However, in some previous experimental studies (Dumitru et al. 2002, Chien and Gupta 2005) as shown in Fig. 7.9, the zonal structure at the bottom of the micro-hole is also evident which suggests the assumption of Gaussian energy distribution at the hole bottom may not be accurate.

Fig. 7.9 AFM images of fs laser drilled micro-hole on silicon wafer (Chien and Gupta 2005)

On the other hand, the current result of the intensity distribution at bottom surface can explain the zonal structure as observed in the previous experiment (Dumitru et al. 2002).

The ring shape distribution of the laser intensity resulted from the reflection of the hole wall can lead to a higher material removal rate not at the centre of the bottom but at the surrounding annularity area. This high material removal rate probably produces a deeper ring shaper groove at the area surrounding the center part of the bottom surface. Eventually, the zonal structure is formed at the hole bottom.

Additionally, Fig. 7.6 shows that the energy distribution inside the hole is even not axial symmetry. This is due to fact that the laser beam is a linear polarized plane wave which indicates that the component of electric field in polarized direction is stronger than that in other directions. The laser intensity distribution shows an ellipse shape that can lead to an ellipse shape of the entrance hole drilled by linear polarized laser which is used in this study. Thus, the ellipse shape of entrance hole that is experimentally observed in Chapters 3, 4, 5, 6 and other studies (Bonse et al. 2002, Choi and Grigoropoulos 2002, Venkatakrishnan et al. 2002) can be explained by the current numerical result.

Figure 7.10 and 7.11 indicate the evolution of spatial distribution of laser intensity at the x-z plane and y-z plane inside the micro-hole at various taper angles. The definitions of the x-z plane and y-z plane are illustrated in the Fig. 7.12.

Fig. 7.10 Laser intensity (W/m^2) at the y-z plane (left column) and x-z plane (right column) of the micro-hole at taper angles of 0 (a, b), 15 (c, d), 28 (e, f) degree

Fig. 7.11 Laser intensity (W/m²) at the y-z plane (left column) and x-z plane (right column) of the micro-hole at taper angles of 38 (a, b), 47 (c, d), 54 (e, f) degree

Fig. 7.12 Illustration of y-z plane (a) and x-z plane (b) in the model

As shown in Fig. 7.10 and Fig. 7.11, the laser intensity of the x-z plane and y-z plane at same taper angle are distinctive in the distribution pattern. This asymmetric laser intensity distribution in two orthogonal planes is resulted from the linear polarization (Venkatakrishnan et al. 2002) of the laser beam and the following multi-reflection of the

plane wave on the hole wall. The maximum values of the energy intensity in these two planes are however close to each other which indicates the point with maximum intensity in the 3-D space is located at the central axis of the 3-D model. In Fig. 7.13, the MLIs at y-z plane and x-z plane of the micro-hole at various taper angles are plotted. The two curves in Fig. 7.13 show a similar trend with the Fig. 7.7 that presents the MLI in the bottom surface. The peak point of the MLI at x-z and y-z planes appears at around 35 degree of the taper angle. This result indicates that there is strong correlation between the energy intensity inside the hole and outflow at the hole bottom surface. The wave-guide effect of the hole wall at smaller taper angle is helpful to focus the laser energy to the central area and elevate the MLI.

Fig. 7.13 MLI at y-z plane and x-z plane of the micro-hole at various taper angles

However, when taper angle become larger than the critical point (35 degree), a considerable portion of the energy is absorbed by the hole wall or repelled out the of the hole which lead to the decline of the MLI. In Fig. 7.10 and Fig. 7.11, it can be observed that the position of the maximum intensity point turns to be closer to the top plane when increasing the taper angle. It is reasonable to explain that the reflection waves from the hole wall at higher taper angle are more likely to focus at a higher position due to a smaller reflection angle (measured from the incoming beam direction) as shown in Fig. 7.14. The previous experimental study (Zeng et al. 2003) reported that the micro-hole wall has a strong influence on the laser induced plasma's temperature, electron density and position. In the laser material interaction process, it is difficult to avoid the plasma (Devaux et al. 1993) due to the fact that the laser energy is sufficient high to excite the electron to conduction bond and cause multi-ionization. After the generation of plasma plume near the ablation surface, the laser beam can consider to be the energy source of the heating of the plasma through the inverse bremsstrahlung (Chang and Warner 1996, Ong and Chang 1997). Thus, the maximum temperature point of plasma is mostly located at the maximum laser intensity point. The observation on the varying location of maximum laser intensity point at various taper angles is of great importance to provide valuable information to predict the status of plasma plume inside the micro hole. In Fig. 7.11 (c, d, e, f), it suggests that the higher taper angle the energy absorption by the side wall is much stronger than that at lower taper angle.

Fig. 7.14 Illustration of the effect of laser beam reflection on the focus point position with taper angles of 25, 33, 40 degree

7.5 Summary

In conclusion, theoretical result on analysis of the hole wall's effect on the laser beam propagation inside the micro-hole of silicon wafer was obtained by numerically solving the time harmonic Maxwell's wave equation. The taper angle of micro-hole was varied to investigate its effect on the laser energy distribution at the bottom of the hole. The MLI at hole bottom firstly increased with increasing taper angle, then reduced with increasing taper

angle beyonds the critical point. The formula between the critical taper angle and geometrical dimension of micro-hole was derived based on the reflection behaviour of the laser beam. The ellipse entrance hole shape and zonal structure at the hole bottom which were observed in the previous experiment have been reasonably explained by using the laser intensity distribution obtained in the present model. In addition, the laser intensity in the vertical planes was studied. The position of the maximum laser intensity point in the vertical direction plane was found to become higher with increasing the taper angle. This result can provide important information for the status of laser induced plasma which is of great significance for both experimental and theoretical studies of the laser material interaction.

Chapter 8 Conclusions and Recommendations for Future Work

In this chapter, the key conclusions which are derived from the present research work are highlighted. A brief overview of the potential research works to be undertaken and their significance is also presented to further extend the current study.

8.1 Conclusions

Although fs laser drilling has attracted significant research interests for decades, in this thesis, some new features were proposed to this area to further improve the fs laser drilling process in terms of eliminating the debris, taper controlling, refining LIPSS, and increasing material removal rate.

The role of six types of volatile liquid films in reducing debris formation during fs laser drilling of silicon was investigated. These liquids have low boiling points and are more volatile. Methanol, ethanol and IPA solvents are the examples of such liquids. It was found that more volatile liquids, i.e. liquids with lower boiling points and lower number of carbon atoms in the molecular formula, were more effective in reducing debris formation as well as reducing the taper angle of the fs laser drilled holes in silicon. The material removal

efficiency was increased by 40% when applying methanol as the assist liquid. The confinement of plasma and the strong shock waves induced by bubbles are the main reasons for the effective removal of debris. The volatile liquids also led to reduction in oxygen concentration in the wall of laser drilled hole due to the oxygen insulating effect of the liquid layer. During the laser drilling experiment, the LIPSS was found at the inside wall of the drilled hole. The microstructures of LIPSS fabricated in liquid environment were investigated. The LIPSSs were generated on silicon wafer using fs laser below the single pulse damage threshold. The media used in the study was both air and ethanol. The laser process parameters such as wavelength, number of pulse, laser fluence were kept constant for both the mediums. The focus of the study was to analyse spatial wavelength. Compared with LIPSS formed in the air, it was concluded that ethanol environment can produce cleaner surface ripples with a period of 450 nm which was far less than the wavelength of incident laser beam. The cleanliness of the surface generated using ethanol showed considerably less debris than in air. The results observed from the above investigation showed that the medium played a predominant role in the generation of surface structures. The bubble generation process was responsible for the significant decrease of debris on the surface. The reduction of spatial wavelength of LIPSS in ethanol environment could be attributed to the increase of real part of effective refractive index.

Subsequently, the fs laser drilling on silicon was investigated at various elevated substrate temperatures in air. The result suggested that the entrance hole diameter was increased by 25% and the exit hole was increased by 30% when the substrate temperature was increased to 900 K. The laser drilling efficiency was also greatly increased by elevating the temperature. This high drilling efficiency was attributed to the enhanced laser energy absorption of silicon wafer and thereafter wave guiding effect. The spatter area was found, however, to continuously decrease with increasing substrate temperature. A large droplet trace was found around the ablation area which indicated liquid phase was increased by the joule heating from the heat conduction of the hot plasma. The results at different temperatures revealed that the initial hole geometry might have significant influence for drilling behaviour of the successive laser pulses.

Finally, in order to further study the hole wall's effect on the laser beam propagation inside the micro-hole, a theoretical analysis was conducted by numerically solving the time harmonic Maxwell's wave equation. The taper angle of micro-hole was varied to investigate its effect on the laser energy distribution at the bottom of the hole. It was found that the MLI at hole bottom firstly increased with increasing taper angle, then dropped with increasing taper angle after the critical point. The formula between the critical taper angle and geometrical dimension of micro-hole was derived based on the reflection behaviour of the laser beam. The ellipse entrance hole shape and zonal structure at the hole bottom which

were observed in the previous experiment were reasonably explained by using the laser intensity distribution obtained in the present model. In addition, the laser intensity in the vertical planes was studied. The position of the maximum laser intensity point in the vertical direction was found to become higher with increasing the taper angle. This result can provide important information for the status of laser induced plasma that is of great significance for both experimental and theoretical studies of the laser material interaction.

8.2 Contribution of this thesis

In this study, for the first time, six alcohol liquids' influence on the fs laser drilling process was evaluated and compared. The relation between the volatility of the liquids and the material removal rate of laser drilling was found. More volatile liquid can better assist carrying away the debris and allow more laser energy reach the ablation front.

The spatial wavelength of LIPSS fabricated in ethanol was found to be 455 nm that is about 40% less than that in air (772 nm). This finding can improve the applicability of LIPSS as optical gate and catalyst support.

A quantitative and systematic assessment of geometry evolution of the laser drilled through hole at different substrate temperature was conducted. The laser drilling efficiency was found to be greatly increased at elevated temperature. This study provides useful

knowledge for better understanding the fs laser energy absorption of silicon wafer at higher temperature.

Theoretical results that obtain in solving the Maxwell's equations illustrate the energy intensity distribution of the laser. The relation between taper angle and MLI at the hole bottom was built. This result not only provides a reasonable explanation for ellipse and zonal shape of the hole profile that were observed in the previous experiment but also shed some information for locating the laser induced plasma inside the hole which is useful for the complex laser drilling modeling involving both the plasma absorption and heat conduction.

8.3 Current limitations and recommendations for future works

The author would like to recommend the following future directions to extend and supplement the current research work.

In the current study, the area of LIPSS was the same as the spot size of laser beam. Scanning method can be applied to obtain a larger area (5×5 mm) of uniform LIPSS. Then the water contact angle's dependence on the spatial wavelength of LIPSS is able to be studied. It is possible that the LIPSS can modify surface wettability (Wang et al. 2011) which affects the flowing resistance of microfluidic channels.

The substrate temperature was limited from 300 K to 900 K in this thesis. It is necessary to explore the temperature to the range below zero. The liquid nitrogen (boiling point 77 K), dry ice (sublimation point of 194.5 K), and ice water (273 K) can be selected as the cooling medium and used to maintain the negative temperature during the drilling process. The experiment can give a basic study for the laser absorption behaviour at different temperature below zero degree. This study can be used to compare with the result obtained at elevated temperature which may further extend the knowledge of environment temperature's influence on the fs laser ablation.

In future work, it would be interesting to simulate some cases that are in the later stage of the laser drilling, i.e. evaluation of the energy loss and energy absorption on the hole wall involving the multi-reflection at both the deeper blind hole and through hole, respectively. Finally, it is necessary to develop a relation between the intensity of electromagnetic field and heat conduction in the material. The dynamic of material removal behaviour due to suddenly temperature change can then be studied.

Publications Arising from this Thesis

Journal paper:

L. S. Jiao, S. K. Moon, E. Y. K. Ng, H. Y. Zheng, and H. S. Son, "Influence of substrate heating on hole geometry and spatter area in femtosecond laser drilling of silicon," *Applied Physics Letters*, vol. 104, pp. 1819021-4, 2014.

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Appendix

Heater Configuration

A 1: Heating element

Two cartridge heating elements (P/N: AFGB0TF010GK040, Watlow) with incoloy sheath as shown in Fig. A1 are set inside the heater block to provide heat flux for the system. The specifications of heating element are listed in Table A1.

Fig. A1 Heating element

Table A1 Specifications of heating element

Specifications	Value
Diameter	1/4"
Length	40mm
Input Voltage	240V
Power	150W

A 2: Heater Block

Size: 40mmL×40mmW×20mmH

Material: Mild Steel

Fig. A2 Dimensions of heater block

A 3: Thermocouple

Thermocouple (P/N: AFGB0TF010GK040) is set in the center hole of the heater block. Thermal couple is used to detect the temperature of the heater block and provide the feedback signal to the controller. The specifications of thermal couple are listed in table A2.

Table A2 Specifications of thermocouple

Specifications	Details
Style	Metal transition with strain relief
Sheath outer diameter	0.125"
Sheath material	316ss
Sheath length	1"
Lead wire construction	Standard fiber glass
Lead wire termination	Standard 1.5 inch split leads
Junction	Grounded

A 4: Controller

EZ-ZONE™ PM Controller (P/N: PM6CICJ-AAAAAAA, Watlow) is used to receive the signal from the thermocouple and automatically control the on and off of the heating element inside the heater block. The specifications of controller are listed in table A3.

Table A3 Specifications of controller

Specifications	Details
Primary function	PID controller
Power supply, digital I/O	100 to 240V
Output hardware	Switched dc/open collector
Functional operating range	0 to 2315 °C
Universal input	Thermocouple
Digital input	Update rate 10Hz
Digital output	Update rate 10Hz
Operator interface	Dual 4 digit, 7 segment LED displays