

Femtosecond-pulse Laser Interactions with Materials
: Fundamentals and Applications for Non-thermal Ultra-precise Machining

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To my family, especially my parents

ABSTRACT *

Femtosecond-pulse Laser Interactions with Materials

: Fundamentals and Applications for Non-thermal Ultra-precise Machining

The intense femtosecond pulsed laser interaction with materials is a subject of practical interest as well as material science interest. Significant advancements in solid state lasers and the chirped pulse amplification technique using Ti:sapphire laser media have led to the challenging phase of this research works. The application fields include high-intensity physics, femtosecond pulsed laser machining, high order harmonics generation, and so on. The femtosecond laser machining is one of the most promising technologies among the femtosecond laser applications because machining physics is drastically different from those of conventional nanosecond and longer pulsed laser machining.

In this dissertation, the first, the motivation, background and previous studies at the introduction of this dissertation were presented. The second, femtosecond laser absorption and ablation were theoretically described. The ablation of metals by femtosecond laser is theoretically described using two-temperature model. Femtosecond laser ablation is expressed by physics theory that takes account of electron contribution calculated by a two-temperature model. The ablation rate and

the thickness of heat-affected zone are estimated theoretically. The third, machining in various materials was studied experimentally.

In the experimental first works, the internal modification in transparent materials including glass and flexible polymer (PDMS) using femtosecond laser pulses irradiation has been studied.

The first, optical diffraction gratings embedded in BK-7 glass by low-density plasma formation using a femtosecond laser was studied. In this work, the optical embedded diffraction gratings with the internal refractive index modification in BK-7 glass plates using low-density plasma formation excited by a high-intensity femtosecond (130 fs) Ti: sapphire laser ($\lambda_p=790$ nm) was demonstrated and studied. The refractive index modifications with diameters ranging from 400 nm to 4 μ m were photoinduced after plasma formation occurred upon irradiation with peak intensities of more than 1×10^{13} W/cm². The graded refractive index profile was fabricated to be a symmetric around from the center of the point at which low-density plasma occurred. The maximum refractive index change (Δn) was estimated to be 1.5×10^{-2} . The several optical embedded gratings in BK-7 glass plate were demonstrated with refractive index modification induced by the scanning of low-density plasma formation.

The second, internal modification in transparent materials using plasma formation induced by a femtosecond laser was studied. In this research, the fabrication of internal diffraction gratings with photoinduced refractive index modification in transparent materials was demonstrated using low-density plasma formation excited by a femtosecond (130 fs) Ti: sapphire laser ($\lambda_p = 800$ nm). The

refractive index modifications with diameters ranging from 1 μm to 3 μm were photoinduced after plasma formation occurred upon irradiation with peak intensities of more than $2.0 \times 10^{13} \text{ W/cm}^2$. The graded refractive index profile was fabricated to be a symmetric around from the center of the point at which low-density plasma occurred.

The third, flexible gratings fabricated in polymeric plate using femtosecond laser irradiation were studied. In this research, flexible gratings embedded in PDMS (poly-dimethylsiloxane) were fabricated using femtosecond laser pulses. Photo-induced gratings in flexible PDMS plate were directly written by a high-intensity femtosecond (130 fs) Ti: Sapphire laser ($\lambda_p = 800 \text{ nm}$). Refractive index modifications with 4 μm diameters were photo-induced after irradiation of the femtosecond pulses with peak intensities of more than $1 \times 10^{11} \text{ W/cm}^2$. The graded refractive index profile was fabricated to be symmetric around the center of the focal point. The diffraction efficiency of the grating samples is measured by a He-Ne laser. The maximum value of refractive index change (Δn) in the laser-modified regions was estimated to be approximately 3.17×10^{-3} .

In the experimental second works, vibration assisted femtosecond laser machining effect on metal was studied. In this work, I demonstrate a novel approach to improve laser machining quality on metals by vibrating the optical objective lens with a frequency (of 500Hz) and various displacements (0~16.5 μm) during a femtosecond laser machining process. The laser used in this experiment is an amplified Ti:sapphire fs laser system that generates 100 fs pulses having an energy of 3.5 mJ/pulse with a 5 kHz repetition rate at a central wavelength of 790 nm. It is

found that both the wall surface finish of the machined structures and the aspect ratio obtained using the frequency vibration assisted laser machining are improved, compared to those derived via laser machining without vibration assistance. This is the first report of low frequency vibration of an optical objective lens in the femtosecond laser machining process being exploited to obtain significantly improved surface roughness of machined side walls and increased aspect ratios.

In the experimental final works on this dissertation, laser cleaning effect on the wafer using plasma shockwave and plasma filament excited by femtosecond laser irradiation.

The first, removal of nanoparticles from a silicon wafer using plasma shockwaves excited with a femtosecond laser was studied. In this work, experiments on the cleaning effect of 100nm-sized polystyrene latex (PSL) particles on silicon wafers using plasma shockwaves excited via a femtosecond (130 fs) Ti:Sapphire laser ($\lambda_p=790$ nm) are reported. The removal efficiency depended on the gap distance between the focused laser beam point and the silicon wafer surface; however some cases exhibited damaged surfaces due to the excessive laser intensity. The cleaning efficiency was strongly dependent on the gap distance between the plasma formation point and the surface. The removal efficiency of the nanoparticles reached 95% without surface damage when the gap distance was 150 μm .

The second, removal of nanoparticles on silicon wafer using a self-channeled plasma filament was studied. In this research, The effective removal of nanoparticles from a silicon wafer surface was demonstrated using the self-channeled plasma filament excited by a femtosecond (130 fs) Ti:Sapphire laser

($\lambda_p=790$ nm). The photoinduced self-channeled plasma filament in air reached a length of approximately 110-130 mm from the first focal spot with diameters ranging from 40 to 50 μm at input intensities of more than 1.0×10^{14} W/cm^2 . By the scan of wafer using the X-Y-Z stage during self-channeled plasma filament, the removal variation of nanoparticles on surface was observed in situ before and after plasma filament occurred. The cleaning efficiency was strongly dependent on the gap distance between the plasma filament and the surface. The removal efficiency of nanoparticles reached to 96% with no damage to the surface when the gap was 150 μm .

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Chapter 1. Introduction

1.1 Femtosecond laser machining

Material processing with pulsed lasers has been an intensive research topic since the invention of the laser in 1960. Nowadays, the laser is used as an efficient and qualified tool in many industrial processes such as heavy industrial cutting, hardening, and welding. In the micro- and nano- fabrication industry, however, the laser has not yet become a universal instrument. In general, one needs a special laser for a particular microstructuring application. Excimer lasers are used, for example, for the micromachining of ceramics and polymers, and ND:YAG-lasers are used for microdrilling and marking. Moreover, for several applications, particularly for the precise microstructuring of metallic materials, the use of lasers with pulse durations in the range of nanoseconds to microseconds is limited due to thermal or mechanical damage (melting, formation of burr and cracks, changes in the morphology, etc.). These limitations have stimulated widespread research activities to minimize collateral damage and thermal diffusion out of the irradiated area by using ultrashort laser pulses, including investigations on the ablation of dielectrics (e.g., Küper and Stuke, 1989; Du et al., 1994; Pronko et al., 1995; Stuart et al., 1995, 1996; Varel et al., 1996; Ashkenasi et al., 1998; Lenzner et al., 1998; von der Linde and Sokolowaki-Tintek, 2000) and metals (e.g., Preuss et al., 1995;

Krüger and Kautek, 1995; Momma et al., 1996; Nolte et al., 1997; Feuerhake et al., 1998; Wellershoff et al., 1999) as well as attempts to produce submicrometer structures (Pronko et al., 1995; Simon and Ihlemann, 1996).

The femtosecond laser seems to be an excellent and universal tool for microfabrication. Metals, semiconductors, dielectrics, polymers, etc., transparent and opaque materials (hard and fragile), can be microstructured with femtosecond pulses so that no postprocessing is required. Furthermore, the surrounding areas are not affected or damaged, which opens the possibility for new fields of application.

Femtosecond laser micromachining is a process by which pulses from a femtosecond laser are used to induce micrometer-sized structures on the surface or in the bulk of solid materials. The most common laser system used for micromachining is the Ti:sapphire laser. This type of laser uses a titanium-doped sapphire crystal as the gain medium, and has a center wavelength near 800 nm. Other fiber-based and solid-state femtosecond lasers have recently been developed that are also good sources for micromachining. The study of the interaction of femtosecond pulses from a Ti:sapphire laser with absorptive materials and transparent materials has been extensive. Ablation has been demonstrated in both absorptive and transparent materials, and bulk modification, manifesting itself primarily as a refractive index change, has been widely shown in transparent materials.

Micromachining in absorptive materials has been applied to hole drilling, removal of surface defects, and photolithographic mask repair [1-5]. Bulk refractive index modifications in transparent materials have been utilized primarily for the

fabrication of optical waveguides and waveguide-based photonic devices [6-12]. In addition, micromachining in transparent materials has found applications in the fabrication of microfluidic channels [13-20].

1.2 Generation of intense, ultrashort light pulses

Most of the high peak-power, ultrafast laser systems today, make use of two techniques developed in 1980's and 1990's. One of them, *Chirped Pulse Amplification* (CPA) was developed earlier and was used in microwave (radar) applications [Cook, 1960], [Strickland & Mourou, 1985]. Though the other technique, *Kerr-lens modelocking* (KLM) [Spence *et al.*, 1991] was developed later, a reverse-chronological description would be appropriate here as KLM forms the first stage of the laser system. Kerr-lens modelocking is normally found in Titanium-doped Sapphire (Ti:Sapphire) oscillators. The large bandwidth (~ 400 nm) and reasonably good thermal conductivity make this crystal ideal for ultrafast oscillators [Moulton, 1982].

Chopping laser light in to short pulses began as soon as the laser was invented, by the invention of modelocking [DiDomenico, 1964], [Yariv, 1965]. The shortest pulses generated even today, use this method. A detailed description of modelocking and its history are not relevant to this thesis and the reader is directed to [Smith, 1970], [Brabec and Krausz, 2000] for this purpose. A laser cavity can have several longitudinal modes (the allowed frequencies in a cavity), but with

completely unrelated phases. Modelocking, as the word suggests, involves locking a large number of longitudinal modes of a laser in phase. In a modelocked laser, the electric fields associated with the different modes add constructively at one point and destructively elsewhere to create a short spike of light. Mathematically, the Fourier transform of a large number of frequency components with same phase gives an infinite series of short pulses in time [Rulliere, 1998]. In a laser cavity, these modes are equally spaced (with a spacing depending on the cavity length). The electric field distribution with N such modes in phase (considered to be zero, for convenience) can be written as

$$E(t) = \sum_{n=1}^{N-1} E_n e^{i\omega_0 + n\Delta\omega t} \propto \frac{e^{N\Delta\omega t} - 1}{e^{i\Delta\omega t} - 1} e^{i\omega_0 t}$$

where ω_0 is the central frequency and $\Delta\omega$ is the mode spacing. This appears as a carrier wave with frequency ω_0 , modulated by the function with exponents, in time-domain. The laser intensity is given by

$$I(t) = |E(t)|^2 \propto \frac{\sin^2(N\Delta\omega t/2)}{\sin^2(\Delta\omega t/2)}$$

This is a series of pulses with width inversely proportional to the number of modes that are locked in phase or the mode spacing. The concept of modelocking is easier said than done. One has to employ nontrivial methods to lock the phases of

different modes, an assumption we started with. Till the advent of KLM, this was achieved in two ways (a) active modelocking (acousto-optic modulator to create a frequency modulation to lock the modes) and (b) passive modelocking (using saturable absorbers to create a time-dependent loss in the cavity). The underlying principle is to strengthen the strongest of field fluctuations, at the expense of the weaker ones eventually leading to the concentration of the whole laser energy in a pulse. A large number of lasers still operate on these mechanisms, usually in the picosecond regime. The limiting factor is the response time of these methods – the time in which one can induce changes in the cavity.

The Kerr-lens modelocking is a self-locking process - the modes are locked automatically, in most cases without any special effort like installing active and passive modulators in the laser cavity. It was accidentally discovered in a Ti: Sapphire oscillator [Spence *et al.*, 1991][Keller *et al.*, 1991] . This happens due to the nonlinear property of the Ti: Sapphire crystal – an intensity dependent refractive index. As a high intensity pulse propagates through it, the crystal acts as a lens (Kerr effect) since the refractive index is intensity dependent (self-focusing). Modelocked-short pulsed-operation is more susceptible to this because of the higher intensity generated by short pulses, as compared to the case of continuous-wave (CW) operation (random phases, multimodes). Thus, short pulses propagate with smaller beam radii. Now, by placing a variable aperture in the cavity, one can select the short pulses because it selectively introduces losses to the CW mode. This implies that a stable operation is possible only in modelocked condition – any spike in the cavity, intentional or nonintentional, switches the laser from CW to

modelocked operation (for an excellent pictorial representation of various modelocking procedures see [Keller, 2003]). Our femtosecond oscillator is a typical Kerr-lens modelocked laser. Since no extra medium is involved, the effect is instantaneous, which enables such systems to generate pulses as short as 5 fs [Pshenichnikov *et al.*, 1998]. Note that, since every optical element in the laser cavity introduce dispersion, the phase relation between various wavelength components have to be maintained using prisms or chirped-dielectric mirrors, to compensate for the cavity dispersion.

However, a normal femtosecond oscillator produces only a few nanojoules per pulse. To reach the high peak powers mentioned in the introductory paragraph, they need to be amplified by several orders of magnitude. The main limitation in earlier attempts towards this end (see, for instance, [Köechnner, 1996]), was the catastrophic intensity-dependent phase shifts as well as optical damage [Perry & Mourou, 1994]. Expanding the beam spatially, in order to reduce the intensity, would demand large-sized optical components – a very cost-prohibitive method. This limitation has been overcome by the ingenious concept of chirped pulse amplification [Strickland & Mourou, 1985], [Maine *et al.*, 1988], the principle of which is sketched in Figure 1.1.

In this method, the low energy, femtosecond pulses from the oscillator are stretched in time using a stretcher (with grating-mirror combination in our system [Pessot *et al.*, 1987]). The stretcher introduces a frequency dependent-phase to all frequency components in the pulse. *Chirp*, is the time-dependent instantaneous frequency of the pulse and arises because of the phase modulation upon the pulse.

In a stretched pulse, the frequency components are no longer in phase. Care has to be taken in choosing the optical elements of the stretcher such that the bandwidth is not lost. The pulse is amplified by several orders using normally a combination of regenerative and multipass amplifiers. The intensity of a ‘stretched pulse’ is low enough not to cause non-linear effects in the optical elements in the amplifier. The amplified pulse is compressed back to its original duration (ideally) using a grating compressor [Pessot *et al.*, 1987], [Strickland & Mourou, 1985]. The effectiveness of compression, however, is limited by the gain-narrowing [Siegman, 1986], [Perry *et al.*, 1990] in the regenerative amplifier and the un-compensated dispersion elements added to the pulse during the amplification process and invariably, an effective compression to the original bandwidth-limited pulse duration can be achieved only with more than one compression units. Nonetheless, this method is rather ubiquitous now – almost all high-power ultrashort systems make use of CPA.

1.3 General laser requirements/setup

Femtosecond micromachining has primarily been done with laser systems using titanium-doped sapphire crystals as the lasing medium. Typical Ti:sapphire laser oscillators have megahertz repetition rates and output pulses on the order of 50 to 100 femtoseconds, with pulse energies of tens of nanojoules. Higher energy laser oscillators are becoming available, but in order to achieve pulse energies in the

microjoule range, it is necessary to amplify the output from the laser oscillator. The amplification reduces the repetition rate to the kilohertz range, which affects the laser interaction with materials.

Micromachining in transparent materials is done using both amplified lasers and oscillator-only systems. In both cases, the laser beam is focused into the sample using a microscope objective. For laser oscillator micromachining, with only nanojoules of energy per pulse, a higher numerical aperture microscope objective is required to ensure intensities high enough to microstructure the sample. Amplified systems output much higher energies than oscillators, meaning tight focusing is not as crucial to reach the same intensity. Therefore lower numerical aperture objectives can be used for micromachining with amplified lasers. The microscope objective is held fixed, while the sample is placed on a movable three-axis stage. The stage is translated with respect to the laser beam to create continuous structures on the surface or in the bulk of the sample. Figure 1.2 shows a schematic of a typical micromachining setup.

The micromachining setup for absorptive materials is very similar, typically employing a microscope objective for focusing the laser beam onto the sample surface. This ensures the fabrication of small features. For applications that require larger features, like hole drilling, long focal length lenses are often used. Although many aspects of the laser/material interaction may differ, the setups for transparent material microstructuring and absorptive material ablation are essentially the same.

Laser parameters and material properties have a significant effect on the interaction and therefore on the results of the micromachining process. Pulse energy,

repetition rate, wavelength, and pulse duration are several laser properties that play a role in the absorption of laser pulses by solid materials. Specific properties of materials, such as bandgap and thermal properties determine whether micromachining with femtosecond pulses is possible and how the resulting structures look and behave. The processes by which energy from the laser is absorbed by a material and the mechanisms by which the material responds are the key to understanding the micromachining process.

1.4 Organization of dissertation

This dissertation has been set out in seven chapters covering the experimental three key topic areas excluding this chapter. I investigated a diverse range of scientific and engineering concepts in this dissertation, including: internal modification in transparent materials such as glass and flexible polymers, laser ablation at material surface, vibration-assisted hybrid machining, and laser cleaning using plasma shockwave and plasma filament excited by femtosecond laser irradiation. Since these three topics are quite different in nature they are each set-out within self contained chapters that deal with the background physics, the experimental equipment, procedures and the results. In this way any future readers of this dissertation with a particular interest in one or other of these subject areas may refer to the appropriate chapter alone. The content of the chapters is arranged as follows;

Chapter 1 is prepared to provide a general introduction to the fundamentals of generation of intense, ultrashort light pulses and femtosecond lasers processing.

Chapter 2 provides overview of interactions induced in materials with femtosecond laser pulses and general introduction to the fundamentals of femtosecond lasers processing. Material modification in transparent materials and ablation on metals with femtosecond laser pulses irradiation and experimental consideration of femtosecond laser machining are included.

Chapter 3 looks at laser machining of transparent materials including glasses and flexible polymeric materials from both fundamental and fabrication perspectives. In this chapter, we report the experimental results of plasma-induced bulk modification in optical planar plates of BK-7 glass using low-density plasma formation excited by a tightly focused femtosecond laser. In these experiments, optical planar plates composed of BK-7, for use in optical communications, lasers and optical sensors are used to study the optical properties of the area of low-density plasma-induced refractive index modification. And the experimental results of plasma-induced bulk modification in optical planar silica plates using low-density plasma formation excited by a tightly focused femtosecond laser was demonstrated. In addition, we report experimental results of a direct femtosecond laser writing technique to inscribe periodic arrays of permanent refractive index modifications in a PDMS polymer using a low-density plasma formation excited by a tightly focused femtosecond laser. These results and the proposed process may be a useful tool for designing optically transparent flexible components in polymer plates with refractive index modification induced by low-intensity femtosecond

laser pulses.

Chapter 4 looks at the material processing at surface using femtosecond laser pulses for vibration assisted femtosecond laser machining on metal. In this chapter, we have demonstrated the effectiveness of vibration of an objective lens on the reduction of burrs at the entrance of machined hole, improvement of the machined wall surface in micro/nano machining, and increasing the aspect ratio for machined metal. Compared to conventional femtosecond laser machining currently used in practical applications, the results in this experiment demonstrate the potential of utilizing a femtosecond laser for controlling the surface roughness and machined depth in fabrication work in various fields such as semiconductors, displays, the bio-medical industry, and so on. Future work will be also needed focusing on more parametric investigations to explore the optimal laser and vibration parameters with respect to various materials.

Chapter 5 looks at the experimental results of trials involving the effective removal of nanosized florescent particles from the surface of a silicon wafer using plasma shockwaves formed by a high-intensity femtosecond laser. We investigated the removal characteristics of nanosized particles during the interactions between plasma shockwaves and nanoparticles on a silicon surface. In second works, we report the experimental results of the effective removal of nanosized florescent nanoparticles from the surface of a silicon wafer using a self-channeled plasma filament excited by a high-intensity femtosecond laser. We studied the removal of polystyrene latex (PSL) particles 100 nm in size using plasma shockwaves laterally induced by a self-channeled plasma filament. This novel technique can be an

effective removal tool considering the large area it can clean due to the long-distance plasma filament, the high removal efficiency, the good cleaning speed, and the lack of any additional damage to the substrate by the propagating self-channeled plasma filament passed from focal point.

Chapter 6 concludes the dissertation with a summary of the key findings and femtosecond laser machining.

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