Ultrafast Laser Polishing for Optical Fabrication

Lauren L. Taylor
llt9077@rit.edu

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Ultrafast Laser Polishing for Optical Fabrication

by

Lauren L. Taylor

A dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Chester F. Carlson Center for Imaging Science College of Science Rochester Institute of Technology

July 31, 2019

Signature of the Author

Accepted by

Coordinator, Ph.D. Degree Program

Date
The Ph.D. Degree Dissertation of Lauren L. Taylor has been examined and approved by the dissertation committee as satisfactory for the dissertation required for the Ph.D. degree in Imaging Science.

Dr. Jie Qiao, Dissertation Advisor

Dr. David Ross, External Chair

Prof. Richard Hailstone

Dr. Zoran Ninkov

Dr. John Lambropoulos
Title of Dissertation:
Ultrafast Laser Polishing for Optical Fabrication

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Ultrafast Laser Polishing for Optical Fabrication

by
Lauren L. Taylor

Submitted to the
Chester F. Carlson Center for Imaging Science
in partial fulfillment of the requirements
for the Doctor of Philosophy Degree
at the Rochester Institute of Technology

Abstract

Next-generation imaging systems for consumer electronics, AR/VR, and space telescopes require weight, size, and cost reduction while maintaining high optical performance. Freeform optics with rotationally asymmetric surface geometries condense the tasks of several spherical optics onto a single element. They are currently fabricated by ultraprecision sub-aperture tools like diamond turning and magnetorheological finishing, but the final surfaces contain mid-spatial-frequency tool marks and form errors which fall outside optical tolerances. Therefore, there remains a need for disruptive tools to generate optic-quality freeform surfaces.

This thesis work investigates a high-precision, flexible, non-contact methodology for optics polishing using femtosecond ultrafast lasers. Femtosecond lasers enable ablation-based material removal on substrates with widely different optical properties owing to their high GW-TW/cm² peak intensities. For laser-based polishing, it is imperative to precisely remove material while minimizing the onset of detrimental thermal and structural surface artifacts such as melting and oxidation. However, controlling the laser interaction is a non-trivial task due to the competing influence of nonthermal melting, ablation, electron/lattice thermalization, heat accumulation, and
thermal melting phenomena occurring on femtosecond to microsecond timescales.

Femtosecond laser-material interaction was investigated from the fundamental theoretical and experimental standpoints to determine a methodology for optic-quality polishing of optical / photonic materials. Numerical heat accumulation and two-temperature models were constructed to simulate femtosecond laser processing and predict material-specific laser parameter combinations capable of achieving ablation with controlled thermal impact. A tunable femtosecond laser polishing system was established. Polishing of germanium substrates was successfully demonstrated using the model-determined laser parameters, achieving controllable material removal while maintaining optical surface quality. The established polishing technique opens a viable path for sub-aperture, optic quality finishing of optical / photonic materials, capable of scaling up to address complex polishing tasks towards freeform optics fabrication.
Dedicated to my parents,
for their constant love and support
in the pursuit of all my endeavors
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• Dr. Aaron Schweinsberg (assisting the initial numerical implementation of a heat accumulation model)

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Publications

This dissertation work has enabled the publication of two first-author, peer-reviewed research papers. A third manuscript has recently been submitted for peer review:


In the body of this dissertation, excerpts from these peer-reviewed publications may be reproduced in part or in whole with permission from the publisher. Some slight modifications and/or elaborations may have been incorporated to improve context and clarity of the excerpt as part of the larger dissertation. For the sake of transparency, footnotes are provided at the beginning of each Chapter to reference to any of the peer-reviewed publications whose text may be incorporated as part of the Chapter body.
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Chapter 1

Introduction

1.1 Motivation

The next generation of imaging systems for applications like consumer electronics, augmented and virtual reality, and space-based telescopes require advanced optical design strategies to reduce the system footprint and weight while maintaining high optical performance [4, 5]. One such strategy is the integration of freeform optics having complex, rotationally asymmetric surface geometries to condense the task of a system of spherical optics into a single element [6]. To glean the full advantage of these novel optical elements, a standardized process chain for deterministic fabrication is required. This need has provoked the investigation of disruptive techniques to manufacture sophisticated freeform surfaces to optical tolerances.

State-of-the-art ultraprecision forming and finishing tools for freeform optics include deterministic micro-grinding, diamond turning, raster milling, magnetorheological finishing, and ion-beam figuring. These techniques have advanced sub-aperture material removal strategies and flexible tool positioning capabilities which cater to fabricating rotationally asymmetric parts and small, complex surface features [6–9]. However, the
sub-aperture material removal strategies leave behind detrimental mid-spatial frequency tool marks on the millimeter scale, and the complexity of sample-tool alignment leads to surface form errors [10, 11]. Contact-based polishing methods also generate significant waste and have long lead times for freeform parts which are disadvantageous for high-volume manufacturing. Therefore, there remains a need for alternative forming, finishing, and post-processing tools for fabricating freeform optics.

1.2 Lasers in Optics and Photonics Fabrication

Over the last decade, high power lasers have become an increasingly popular non-conventional tool for optics- and photonics-related fabrication tasks including cutting, welding, drilling, surface treatment, figuring, and bulk material modification applications [12–14]. The machining capability of lasers surmounts from the ability to enable a material to absorb sufficient incident photon energy to induce breakdown or phase change in a material to permanently modify its optical, electronic, thermal, or structural properties.

The mechanism and capability for breakdown relies on both the material and laser characteristics. During laser irradiation, a fraction of the laser incident photons will be absorbed by the material over time, where the extent of the energy absorption is controlled by the wavelength-specific reflectivity and optical properties of the material. In the simplest case, when the material is opaque to the wavelength of the laser beam, laser energy will be linearly absorbed (e.g. one photon can be absorbed by one electron to promote the electron to an excited state). For this case, the distribution of the laser intensity, $I$, transmitted through the material along the direction of laser propagation, $z$, can be described according to [15]:

$$\frac{\partial I}{\partial z} = -\alpha I \propto \frac{\partial E}{\partial z}$$  (1.1)
Chapter 1. Introduction

Here, intensity falls off according to the linear absorption coefficient $\alpha$, and is directly proportional to the distribution of transmitted laser energy, $E$. Note that laser “intensity” in Eq. 1.1 is consistent with the radiometric quantity of “irradiance”, both having units of [W/cm$^2$]. However, the term “intensity” will be maintained in this dissertation for consistency with laser-related terminology.

Absorption of energy during laser irradiation increases the surface temperature of the material. If a sufficient amount of laser energy is absorbed, the local surface temperature can reach the melting point. Once enough energy has been supplied to overcome the material-specific enthalpy of fusion, the surface becomes molten. If the surface temperature continues to rise to the vaporization point and supplies enough additional energy to overcome the enthalpy of vaporization, this portion of the laser-affected surface region can change to a gaseous state and particles can be directly carried away, or “ablated”, from the material. The onset and extent of laser-induced material breakdown and phase change is directly related to the rate at which energy is absorbed.

By carefully selecting the laser source (wavelength, mode of operation, etc.) and the irradiation parameters (intensity and irradiation time), the laser interaction can be controlled to enable high-precision laser-based machining. Additional machining advantages can be achieved by integrating lasers with high-precision, flexible, and controllable beam delivery systems based on fiber and/or free-space propagation. For example, integrating lasers with high-speed translation stages and galvanometer beam scanning systems can enable dynamic control of the laser focal position and high processing speeds [16] enabling direct access to small, complex surface features, process scaling for large-scale machining applications, and the capability to implement dynamic beam control routines for complex processing tasks. Laser processing is also nearly waste-free process in contrast to conventional machine tools which generate significant waste from material removal and the use of slurries, fluids, and lubricants.
The attributes of laser processing have allowed lasers to be specifically targeted as a methodology for additive manufacturing, surface patterning and cleaning for industrial and medical applications, post-processing additively manufactured surfaces, optics polishing, waveguide writing, and substrate welding for photonics integrated circuits [17–25]. The flexible beam delivery and tunable control of material breakdown have directly positioned lasers as a tool for high-precision optics and photonics fabrication tasks, including polishing.

1.2.1 Laser Polishing

Laser-based polishing is carried out by controlled scanning of a focused laser beam along the material surface. A general schematic of a raster-based laser polishing process is shown in Fig. 1.1. The predominant polishing strategy utilizes continuous wave and/or short pulsed lasers to melt and re-flow a layer of the material surface to reduce and/or eliminate rough surface asperities. Polishing

![Figure 1.1: General schematic of the laser polishing process carried out by raster-scanning a focused laser along the surface of a substrate [26]. Here, $\beta$ is the angle of incidence, $R$ is the laser spot radius, $v_{scan}$ is the scanning velocity, and $d_y$ is the center-to-center distance between raster lines.](image)

The laser polishing process is highly complex and tunable, as it considers many laser
parameters and properties (power/energy, repetition rate, wavelength, polarization, mode of operation, etc.) and incidence characteristics (angle of incidence, laser spot size, scan speed, raster-line spacing). The combined impact of the selected process parameters determines the precision, efficiency, and quality of the processing task. Parameter impacts must be carefully balanced to evoke an appropriate material response to ensure polishing can be achieved.

To date, polishing has been performed using both continuous-wave (CW) and pulsed lasers. Continuous-wave lasers make use of gas, dye, or solid-state gain media and are constantly pumped to continuously emit light [27]. When employing a CW laser for polishing, the continuous nature of the irradiation induces steady linear energy absorption and, over time, the substrate accumulates heat until reaching the melting point. Once the surface melts, the material begins to flow and surface tension works to smooth-out the original surface asperities [28]. The surface solidifies as the laser beam is scanned along the substrate, leaving behind a smoothed surface.

Melt-based CW laser polishing has been implemented for a variety of surface processing applications. These lasers have been used to polish both additively manufactured and conventionally machined metals, capable of achieving up to a 90% reduction in roughness in comparison to the original surface [21,29–32]. They have also been investigated in optics-related fabrication applications including in-situ healing of laser damage for high-energy laser beam delivery systems and as a smoothing step in a laser-based fabrication chain for freeform optics [33,34]. Examples of glass polishing using CW lasers are shown in Fig. 1.2.
The thermal nature of CW laser polishing can affect the flexibility and effectiveness of the polishing process, and the lead-time for surface generation. First, CW laser polishing relies on linear absorption of laser energy, requiring the wavelength of the incident beam to precisely match the optical absorption characteristics of the material substrate. This prohibits the development of a stand-alone CW laser polishing system which can address processing of different materials. Second, the extensive energy absorption required to melt the surface leads to large areas of sub-surface damage and thermal-gradient-induced stresses which can affect the optical performance and structural integrity of the finished optical element [22]. The significant thermal penetration results in sub-melt-pool heat affected zones on the order of a few hundred microns [32, 36–38], and the steep thermal gradients can lead to micro-crack formation. Third, melt-based polishing induces surface ripples resulting from re-solidification of the melt-front which are affected by the laser scan speed, fluence, and polishing line overlap [29, 39]. These ripples induce waviness artifacts in the resulting polished surface which require removal via corrective post-processing steps such as high-precision laser ablation [40]. This increases the complexity and time of the polishing process.

To meet the need for improved precision and flexibility, pulsed lasers have also been investigated for laser polishing. In contrast to CW lasers, pulsed lasers operate by emitting packets of energy at a controlled repetition rate [27]. When irradiating a material, the
periodic nature of the energy deposition allows time for the material to cool between incident laser pulses. In polishing, this reduces the overall thermal impact of the irradiation process in comparison to CW lasers. Pulsed lasers can also offer improved processing efficiencies due to the high peak powers which can be achieved by each pulse. For a pulsed laser, the peak power of a single pulse is calculated as:

$$P_{\text{peak}} = \frac{E_p}{\tau_p}$$  \hspace{1cm} (1.2)

Here, $E_p$ is the laser pulse energy and $\tau_p$ is the temporal width of the laser pulse. For a CW laser, the peak power corresponds to its measured operational power, which is comparable to the average power of a pulsed laser with frequency, $f_{\text{rep}}$.

$$P_{\text{peak}, \text{CW}} \approx P_{\text{avg}, \text{pulsed}} = E_p \cdot f_{\text{rep}} $$  \hspace{1cm} (1.3)

For a CW laser and a pulsed laser with the same measured power, the peak power of the pulsed laser will be significantly higher. For example, if a CW laser and a 10-ns pulsed laser with 10-kHz repetition rate both operate with 10 W of power, the pulsed laser has a peak power of 100 MW, whereas the CW laser has a peak power of 10 W.

The peak power of the laser determines its intensity, which drives the magnitude and extent of the material modification (e.g. ablation vs. melting). For a gaussian laser beam with a $1/e^2$ radius of $w_0$, the peak intensity is calculated as:

$$I_{\text{peak}} = \frac{2 \cdot P_{\text{peak}}}{\pi \cdot w_0^2} = \frac{2 \cdot E_p}{\pi \cdot w_0^2 \cdot \tau_p}$$  \hspace{1cm} (1.4)

Equation 1.4 shows that both the peak power and intensity of the pulse increase as the temporal width of a laser pulse decreases. Laser polishing reported thus far is normally carried out using pulse widths on the millisecond to nanosecond order. In the nanosecond regime, the achieved laser intensities are high enough to evoke both melting and ablation
using a single laser pulse [41], improving the efficiency of the material removal process. Pulsed lasers also improve processing quality and precision because the short duration of the laser interaction causes fast material removal and resolidification on the pulse timescales, improving the spatial selectivity of the laser interaction [41].

Pulsed lasers down to the nanosecond timescale have been used to polish various metals to nanometer-order roughness [42, 43]. Microfabricated nickel samples have been polished using 300-ns pulsed Nd:YAG lasers, achieving a $7 \times$ improvement over the original surface roughness (reduced from the order of 100-200 nm down to the order of 15-60 nm.) [28]. A similar polishing method using a 1.5-µs 1070-nm fiber laser showed the capability to smooth titanium alloy (Ti-6Al-4V), reducing the average surface roughness from 172 to 47 nanometers [37]. The discrete nature of pulsed laser-material interaction reduces the extent of sub-surface damage, achieving sub-melt-pool heat affected zones with depths ranging from 2.5 to 13.5 µm [36, 37], significantly smaller than for CW laser polishing.

Although ablation-based material removal can be achieved by nanosecond pulses to increase the processing efficiency and reduce its thermal impact, material removal cannot be decoupled from thermal melting because the duration of the laser-material interaction is on the same timescale as heat diffusion [44]. Additionally, the standard pulsed-laser polishing process requires linear energy absorption as the achieved intensities for millisecond to nanosecond laser pulses are not high enough to evoke significant multiphoton absorption. Therefore, the wavelength of the laser needs to match the linear optical absorption characteristics of the material to be processed. To improve the precision and flexibility of laser polishing, a new strategy for high-precision, material-flexible, laser-based material removal which can achieve smooth surfaces with minimized heat-affected zones is desired.
1.2.2 Advantages of Ultrafast Lasers

One solution for improving the precision of laser-based polishing is to reduce the spatial and temporal localization of incident laser pulses and achieve high intensities. Ultrafast lasers have pulse widths on the picosecond ($10^{-12}$ s) to femtosecond ($10^{-15}$) order and can generate high intensities up to the order of GW-TW/cm$^2$ depending upon the energy and size of the incident laser pulse [45]. They can enable high-precision ablation-based material removal with nearly negligible thermal impact because the timescales for the onset of material breakdown (fs-ns) in comparison to the timescales for thermal melting, heat diffusion, and heat accumulation (ns-µs) are decoupled [44]. The achieved intensities of ultrafast lasers can evoke nonlinear absorption, enabling processing of both opaque and transparent materials and eliminating the need for matching laser wavelength and material optical properties. The short interaction times act to confine the laser material interaction to a small region within the focal volume, achieving spatially-selective high precision machining towards applications including processing of small complex surface features (e.g., freeform optics). A comparison of the characteristics of CW- and pulsed-laser / material interaction down to the ultrafast regime is given in Fig. 1.3 [46]. A timeline detailing the onset of different physical phenomena during ultrafast laser-material interaction is shown in Fig. 1.4 [44].
Figure 1.3: Comparison of processing mechanisms for CW and pulsed laser polishing from [46]. Polishing via CW laser radiation is a highly thermal process resulting in large sub-surface heat-affected zones. The periodic nature of nanosecond laser-material interaction can reduce the thermal impact of the process, but high laser pulse energies enabling ablation send large shockwaves through the material. Ultrafast lasers can enable ablation-based material removal with mitigated thermal impact and reduced shockwave production due to the short pulse duration and small pulse energies.

Figure 1.4: Timeline of physical phenomena occurring during ultrafast laser-material interaction from [44].

The unprecedented material removal capabilities of ultrafast lasers have enabled diverse
applications in optics and photonics fabrication. Femtosecond lasers, in particular, have been exploited to process various materials including glasses, semiconductors and metals for a variety of different optics-related applications. They can be used to fabricate optical structures on silicon, including micro-lens arrays and diffractive optical elements [47,48], or to write waveguides and gratings in dielectric materials to suit applications like monolithic laser fabrication [49–51]. Self-assembly of nanoparticles and nanostructures can be initiated by femtosecond lasers to achieve birefringence and dichroism in both glass and silicon or to enhance the absorption capability of photovoltaics materials [52–54]. Surface processing using femtosecond laser radiation can enable damage-free delamination and patterning of silicon oxide, catering to photonics and photovoltaics applications [55, 56], and can be used as a tool for medical implant patterning and industrial marking applications [39]. Femtosecond lasers can also be used to weld glasses and glass to silicon for microelectronic applications or to cut glass for consumer electronics [25,57,58].

With regard to polishing, picosecond lasers have been used for melt-based polishing of metals achieving surface roughness down to the sub-micron level [59,60]. Numerical and qualitative experimental studies (Fig. 1.5) comparing nano-, pico-, and femto-second laser ablation have shown that decreasing the pulse width to the ultrafast regime can improve the processing precision towards smoother surfaces [41,61]. Further studies of ultrafast laser-based surface polishing are limited and, to our best knowledge, optic-quality ablation-based femtosecond laser polishing has not been demonstrated.
Chapter 1. Introduction

Figure 1.5: Processing with 1.4 ns, 540 ps, 50 ps and 10 ps laser pulses (left to right) on copper from [61]. Decreasing the pulse width improves the surface quality.

1.3 Research Goals and Objectives

This thesis work aims to develop a non-contact polishing methodology towards eliminating the waste, cost, and lead-time associated with freeform optics fabrication by exploiting the machining advantages of ultrafast lasers to achieve high precision polishing of optics. It focuses on polishing of semiconductor optical substrates owing to their wide imaging- and photonics-related applications in the visible and near infrared wavelength regimes. Ultrafast laser material interaction is investigated from the fundamental theoretical and experimental standpoints to determine a strategy and laser parameters to achieve controllable, repeatable polishing (e.g. high-precision material removal with optic surface quality). The overall technical approach for developing a methodology for laser polishing is presented in Fig. 1.6. The first four objectives stand as the pillars for establishing the polishing methodology, requiring some iteration to optimize the strategy for a given material. The last objective acts as the extension of this project towards future work.
Chapter 1. Introduction

1.4 Structure of Dissertation

The following Chapters of this dissertation describe the methodology, modeling, and experimental results for achieving femtosecond-laser-based polishing using the technical approach in Fig. 1.6. Chapter 2 details an experimental and numerical investigation of ultrafast laser-based material removal which lays the framework for the strategic approach to achieving laser polishing. Chapter 3 describes the implementation of a constructed Heat Accumulation Model to investigate laser parameters to control the overall surface temperature rise during laser-based material processing to mitigate the onset of thermal effects. Chapter 4 describes the construction of a Two-Temperature model of femtosecond laser/material interaction to establish a methodology for offline investigation of laser parameters which can induce ablation-based breakdown and melting. Chapter 5 describes the construction of a flexible, tunable laser ablation system and an experimental investigation of the impact of laser parameters on the quality and efficiency of laser processing. Chapter 6 presents a strategy to predict and experimentally validate optimal laser polishing parameters and demonstrates successful, controllable
femtosecond laser-based optic-quality polishing.
Chapter 2

Investigation of Ultrafast Laser-Based Material Removal

This Chapter\(^\text{†}\) presents an investigation of ultrafast laser-based material removal which acts as a stepping stone towards defining the technical approach and research tasks required to achieve laser polishing.

2.1 Experiments

2.1.1 Setup

A laser ablation system (Fig. 2.1) consisting of a femtosecond laser, a focusing lens, and a three-axis motorized translation stage was constructed to carry out ultrafast laser processing experiments.

\(^\text{†}\)The body of this Chapter includes relevant excerpts (in part or in whole) from an original, first-author publication: L. L. Taylor, J. Qiao, and J. Qiao, “Optimization of femtosecond laser processing of silicon via numerical modeling,” *Optical Materials Express*, 6(9), 2016 [1]. Slight modifications and/or elaborations may have been included to improve context and clarity as part of the larger dissertation.
The laser source is a 400-fs pulse width, 1030-nm wavelength Ytterbium fiber laser (Satsuma, Amplitude Systèmes). A system of fold mirrors was used to raise the beam and focus it downwards onto the sample. The achieved focal spot diameter was approximately 70 µm, determined by multi-shot, stationary point processing tests. Samples were mounted and scanned under the focused laser beam using three motorized translation axes with 50 mm of travel, capable of scan speeds up to 2.8 mm/s (MTS50-Z8, Thorlabs).

Initial laser processing experiments were performed on various materials including metals (aluminum, additively manufactured Ti-6Al-4V/maraging tool steel), glasses (fused silica, soda-lime glass), ceramic (Al₂O₃), and semiconductors (silicon, silicon carbide). However, this dissertation will focus on semiconductor optical substrates owing to their use in a wide variety of near- to mid-infrared imaging, detector, and consumer electronics applications which can be directly benefited by freeform optics [62,63].

To assess the capability for material removal, processing was investigated at the fundamental operating frequencies of the laser: 500 kHz, 1 MHz, and 2 MHz. The pulse energy was adjusted to maintain an average laser power of 20 W for all processing experiments. Investigations were carried out using scan speeds from 0.1 to 2.5 mm/s to enable different pulse overlaps during scanning.
2.1.2 Results

The processing experiments on semiconductor substrates were measured using laser scanning confocal microscopy (Keyence VK-X210).

Figure 2.2: Laser processing of silicon (left) and silicon carbide (right) showing effective material removal. Pileup alongside the ablated trenches is hypothesized to result from high temperatures generated during processing.

Figure 2.2 shows the surface height maps and the corresponding average line profiles of
laser-processed silicon and silicon carbide using 10 µJ pulses, a repetition rate of 2 MHz, and a scanning speed of 1.5 mm/s. An ablation depth on the order of 10 µm was achieved for Si and an ablation depth larger than 15 µm was achieved for SiC. The width of the ablated area is on the order of 20 µm for both materials. These results demonstrate the effectiveness of femtosecond laser ablation for material removal tasks. However, for both substrates, material pileup is evident adjacent to the ablated lines.

Further inspection of the line profiles shows that the material pileup ranges from 5 to 20 µm in height and that the pileup height is asymmetric about the ablated line. The uneven pileup height is attributed to an asymmetric intensity distribution at the focal plane resulting from alignment-induced aberrations. The silicon line profile shows pileup with respective heights of 8 µm and 20 µm on the left and right sides greater than the ablated material depth. This suggests that the true ablation depth is larger than is seen via surface profilometry, potentially due to the steep aspect ratio of the processed region.

A cross-section analysis revealed that the surface voids on silicon are approximately 50 µm in depth and the laser-affected region, which includes additional sub-surface voids, extends to a depth on the order of 100 µm. Material ejected from both surface and sub-surface voids likely contributed to the height of the pileup.

Energy-dispersive X-ray spectroscopy (EDS) was used to determine the elemental compositions of laser-processed and unprocessed silicon surface regions. Figure 2.3 shows normalized spectra for both conditions. The silicon carbide surface composition was not able to be evaluated due to high sample charging in the electron microscope.
Chapter 2. Investigation of Ultrafast Laser-Based Material Removal

Figure 2.3: Normalized silicon EDS spectra collected for material pileup and unprocessed surface regions. The region of material pileup shows increased oxygen and carbon content.

Figure 2.3 shows a significant increase of both oxygen (8.4:1) and carbon (6.5:1) when comparing the elemental composition of the pileup to the unprocessed region. Increased oxygen content indicates oxide growth at the location of material pileup. Oxidation in silicon occurs over the temperature range of 973 – 1573 K [64], indicating that high surface temperatures were reached during femtosecond laser processing. Ablation of material is evidence that vaporization occurred during material processing. The interaction of the vulnerable material surface during ablation and the vaporized material with hydrocarbon pollutants and carbon dioxide in the surrounding air resulted in carbon-doping of the laser-affected region [65]. Re-condensation of ablated surface material and melting may also play a role in pileup formation.

2.1.3 Discussion

The experimental study of femtosecond laser processing of Si and SiC demonstrates that femtosecond lasers are capable of effective material removal. However, the initial processing experiments yielded permanent material pileup and oxide growth detrimental to achieving a polished surface. In surface processing experiments, over-exposure to femtosecond laser
radiation via un-optimized laser parameters can lead to blistering, oxidation, melting, and cracking [66–69], suggesting that the experimentally-investigated set of laser parameters was not suitable for high-precision processing. Simulations of femtosecond laser/material interaction have shown that these phenomena result from high temperatures generated during processing [69–71].

In order to formulate a strategy to control laser parameters to achieve polishing, it is critical to understand the range of surface temperatures achieved during the initial polishing experiments and the significance of this range in comparison to critical temperature thresholds (i.e. melting, oxidation, etc.). Because it is challenging to probe and monitor temperatures during femtosecond laser polishing due to the ultrafast timescale of the laser interaction, the thermal impact is initially investigated via numerical modeling.

2.2 Numerical modeling of surface-temperature evolution during laser surface processing

2.2.1 Modeling approach

A femtosecond laser pulse causes a large density of photons to strike the surface of a material on a nearly instantaneous timescale. These photons can be absorbed to promote valence-band electrons to the conduction band (excited state). Photon energy can be linearly absorbed if it is higher than the bandgap energy of the material, or can be nonlinearly absorbed via multiphoton processes if the photon density is high enough [70]. Femtosecond laser irradiation generates a large density of excited electrons, which can lead to ablation (vaporization-based material removal) if the laser fluence is above the ablation threshold [72]. During and immediately after the pulse, energy from the excited electron system is transferred to the material lattice to cause heating and/or melting [73]. The induced surface temperature dissipates over time as heat diffuses through the bulk of the material.
In multi-pulse processing, heat can accumulate if the time for heat diffusion between pulses is restricted, enabling the onset and accumulation of thermal phenomena such as melting and oxide growth.

Models based on classical definitions of heat sources and energy transfer have been developed to simulate heating in ultrafast laser processing [68, 69, 71]. These models approximate the magnitude of the pulse-induced temperature rise without the need for subatomic simulation of energy absorption, electron plasma density, and electron/lattice heat exchange as in two-temperature models [74–76] and without the requirement of high-capacity computation platforms [77]. This modeling approach was adopted and a numerical model was constructed to predict the evolution of surface temperature during femtosecond laser processing of silicon, based upon the modeling theory presented by Bauer et al. [69]. The model assumes that the femtosecond laser pulse acts as an instantaneous heat source because the temporal pulse width is orders of magnitude smaller than the nanosecond to microsecond timescale required for normal heat diffusion [44, 69]. It acts as a first approximation of ultrafast laser heating; it does not consider electronic-level phenomena occurring within the timescale of the pulse or melt/vapor phase transitions. Additional phenomena which impact heat generation and ablation, but are outside the scope of this first-approximation model of heating during ultrafast laser processing, include free-carrier absorption of photons [78], the effect of the generated plasma temperature on surface-heating [79], decreased absorption of laser light in multi-pulse ablation caused by the plasma and ablated particles [54], change in surface roughness [80], and volume changes in material resulting from ablation/melting which affect heat transfer [81]. However, these omissions do not impede the initial modeling goals of (1) evaluating the thermal impact of the experimental laser processing in Section 2.1.2 and (2) establishing a methodology for offline evaluation and prediction of laser parameters to minimize heat accumulation and the onset of detrimental thermal effects.
A more detailed numerical model which addresses electron-level phenomena on the femtosecond timescale is detailed in Chapter 4.

2.2.2 Constructing a Heat Accumulation Model

When a femtosecond laser pulse impinges on a material, the amount of deposited laser pulse energy directly increases the energy of the surface according to the fluence distribution of the incident pulse [69]. The heat-inducing volumetric surface energy density, $\Omega$, deposited by the $n^{th}$ laser pulse centered at $(x_n, y_n)$ is described by Eq. 2.1:

$$\Omega = \frac{2 \cdot A \cdot E_{\text{pulse}}}{\pi \cdot w_o^2} e^{-\frac{2((x-x_n)^2+(y-y_n)^2)}{w_o^2}} \cdot \delta(z). \quad (2.1)$$

Here, $E_{\text{pulse}}$ is the energy of the incident Gaussian pulse with waist, $w_o$ and the factor of 2 in the numerator accounts for heat being deposited into a half-space, rather than into the infinite space required for classical, analytical temperature distribution calculations [82]. In Eq. 2.1, $A$ is the fraction of incident energy remaining in the surface after ablation (hereafter referred to as the “operational absorption coefficient”). A reference value of 0.8 [83] was used for $A$ to accommodate the ground finish of samples and the steep rise in energy absorption seen in experimental, above-fluence-threshold multi-pulse femtosecond laser processing of silicon. This value considers a range of fluences up to 1 J/cm$^2$ for multi-pulse irradiation at 800-nm [83]. However, $A$ does not directly account for the wavelength-dependent energy penetration depth. Therefore, Eq. 2.1 incorporates the Dirac delta function, $\delta(z)$, with units of inverse meters, to ensure that energy is only deposited within a controlled layer of the material at its surface, $z = 0$. For our in-house femtosecond laser system, experimentally achievable wavelengths correspond to the fundamental (1030 nm), second harmonic (515 nm) and third harmonic (343 nm) operational frequencies of Ytterbium. To accommodate the three wavelengths while maintaining a reasonable...
computational load, the model assumes that all heat-inducing energy is absorbed into a 1.5-µm thick surface layer. For 515 nm and 343 nm wavelengths, the linear absorption depth is less than this thickness. For 1030 nm wavelength light at low surface temperatures, nonlinear absorption dominates and the absorption depth may exceed the layer thickness [84]. Therefore, it is possible that the heat-inducing energy density predicted in Eq. 2.1 may be higher than what is achieved during experimental implementation.

Multiplying $\Omega$ by the discrete volume of a surface element gives the element-specific increase in surface energy. The instantaneous rise in temperature associated with this surface energy increase is calculated using Eq. 2.2 [69]:

$$T = \frac{E}{\rho \cdot c \cdot \Delta V}. \quad (2.2)$$

The surface temperature is directly proportional to its energy, $E$, and inversely proportional to the material density, $\rho$ (2.329 g/cm$^2$, [85]), the temperature-dependent specific heat capacity, $c$, and the discrete volume of a matrix element, $\Delta V$.

### 2.2.2.1 Heat diffusion

The induced surface temperature decreases over time as heat diffuses throughout the material bulk. The analytical heat conduction equation in Eq. 2.2 links the temporal and spatial rates of temperature change:

$$\rho \cdot c \cdot \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(k \cdot \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(k \cdot \frac{\partial T}{\partial y}\right) + \frac{\partial}{\partial z}\left(k \cdot \frac{\partial T}{\partial z}\right). \quad (2.3)$$

This relation depends upon three material properties: density, specific heat capacity, and thermal conductivity ($k$). The temperature dependencies of thermal conductivity and specific heat capacity were taken into account using an explicit central finite difference solution to the heat conduction equation for a 5-ns time step.
Because the size of the focal spot in laser processing experiments is spatially-constrained to the micron-scale, the Gaussian-distributed energy absorption will induce steep surface temperature gradients. A steeper temperature gradient allows faster heat diffusion, which can enable a surface irradiated with a higher fluence to quickly achieve consistent heat diffusion performance with a surface irradiated using a lower fluence. Although Eq. 2.1 may initially lead to over-prediction of the laser-induced surface temperature rise, the heat diffusion process allows the model to remain valid to predict heat accumulation during processing.

2.2.2.2 Temperature dependence of silicon properties

Both the thermal conductivity and specific heat capacity of silicon are temperature-dependent [86,87]. Figure 2.4 shows that the specific heat capacity increases and thermal conductivity decreases as temperature rises. A higher specific heat capacity requires increasingly more heat to change the temperature of silicon by a unit Kelvin. Therefore, the instantaneous temperature rise caused by a femtosecond laser pulse will decrease in magnitude as the surface temperature rises. A decrease in thermal conductivity with higher surface temperature causes silicon to act as a worse heat conductor. This reduces the rate of heat diffusion, limiting the amount of surface temperature decay between incident pulses.
Figure 2.4: Temperature dependencies of the thermal conductivity and specific heat capacity of silicon. As temperature is increased from 273 to 1685 K (silicon melting point), thermal conductivity decreases by approximately 150 W/(m·K) [86], while specific heat capacity increases by approximately 350 J/(kg·K) [87].

2.2.3 Simulating initial experimental laser processing of Silicon

The temperature evolution in silicon was simulated using the mathematical methods described in Subsection 2.1 for the experimental laser processing conditions presented in Section 2.1.2: 10 µJ pulse energy, 2 MHz repetition rate, and 1.5 mm/s scanning speed. (Because energy deposition by a femtosecond pulse is considered instantaneous in comparison to heat diffusion, pulse duration is not a parameter in the numerical model.)
Figure 2.5: Evolution of the maximum temperature of silicon over time due to an incident femtosecond laser pulse train. The initial temperature is 293 K. Local temperature maxima arise due to pulse energy deposition and local temperature minima result from heat diffusion after the pulse. $T_o$ denotes the oxidation temperature threshold for silicon (973 K [64]) and $T_m$ corresponds to the melting temperature (1685 K).

Figure 2.5 shows that the predicted maximum surface temperatures for the experimental processing conditions are consistently higher than the oxidation threshold of silicon after four pulses and fully exceed the melting temperature after ten pulses. The maximum temperatures achieved during laser processing will occur at the surface of silicon at spatial locations corresponding to peak energy deposition by the most recently incident pulse. The maximum temperature continues to increase past the melting point with the incidence of more pulses. The predicted temperatures infer the onset of thermal phenomena for the experimental processing conditions, in agreement with the material analysis presented in Section 2.1.2. This also supports the need for a set of laser parameters which minimizes heat accumulation to avoid the melting and oxidation thresholds to mitigate the onset of detrimental thermal effects during polishing. However, this requires a full understanding of the individual and combined impact of laser parameters to carefully balance the thermal impacts.
2.3 Conclusion

In this Chapter, the effectiveness of femtosecond laser surface processing of silicon was experimentally demonstrated. However, the experimental processing yielded detrimental surface artifacts including material pileup and confirmed oxide growth along the ablated track due to high induced surface temperatures. The heating mechanism in femtosecond laser processing of silicon was studied via a constructed numerical Heat Accumulation Model, capable of predicting the surface temperatures achieved during laser processing. The simulated surface temperature for the experimental processing conditions confirmed the onset of oxidation and the potential for material melting, showing the need for an improved strategy to achieve precision material removal for polishing.

For femtosecond laser-based polishing, it is imperative to remove material precisely while controlling the onset of thermal and structural effects detrimental to achieving a smooth surface. Processing in inert gas, liquid, and vacuum environments has been shown to enhance the quality of surface processing \cite{88,89}, and can potentially mitigate oxide growth, but this strategy reduces the utility of femtosecond lasers in batch processing, lengthens product lead-times, and is unsuitable for large-scale applications. Therefore, a method for controlling the surface quality for processing in air must be achieved to maintain the utility of ultrafast laser processing for various applications. This can be achieved by determining a set of optimal laser parameters for material removal (e.g., repetition rate, scan speed, and fluence) specific to the material to be polished. This is a non-trivial task due to the competing influence of different interaction phenomena such as nonthermal melting, ablation, electron-lattice heat transfer, heat accumulation, and thermal melting/oxidation \cite{44}. It is challenging to experimentally determine a set of optimum parameters to effectively balance these mechanisms to achieve polishing, as the laser parameters can be tuned in a near-continuous fashion on many femtosecond laser systems and their impact is material-specific.
Chapter 2. Investigation of Ultrafast Laser-Based Material Removal

The goal of this thesis work is to investigate a strategy for laser polishing in air by investigating laser/material interaction from the fundamental theoretical and experimental standpoints. To effectively control laser parameters to achieve polishing, several critical questions must be answered:

1. How do individual and combined laser parameters impact the thermal and material removal characteristics of laser polishing?

2. How does varying the laser parameters experimentally affect the surface quality?

3. What constitutes an optimal set of laser polishing parameters and how can one be predicted?

4. Can optimal laser parameters be determined for smooth processing?

5. Can polishing be controlled?

The investigations in the following Chapters aim to answer the posed questions towards determining a strategy to achieve femtosecond laser-based polishing of semiconductor optical substrates.
Chapter 3

Predicting Heat Accumulation During Laser Processing

In order to determine a set of laser parameters for polishing which can minimize the onset of thermal effects, it is imperative to understand the mechanism for heat accumulation and the material’s thermal response to different laser parameters. This Chapter† investigates the heat accumulation mechanism, minimization of heat accumulation via determining optimal laser parameters, and the ability to achieve uniform thermal processing conditions by controlling equilibration time between laser-induced temperature rise and heat dissipation.

3.1 Heat accumulation mechanism

A femtosecond laser deposits multiple pulses onto the surface of a material at times and locations dictated by the laser repetition rate and sample scan speed. The amount of energy absorbed at a given surface location over the course of processing is related to the scan speed of the sample. The laser repetition rate affects the amount of heat diffusion occurring between incident pulses, where low repetition rates are required for the surface to cool to its initial temperature following the incidence of a pulse. Figure 3.1 shows how

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†The body of this Chapter includes relevant excerpts (in part or in whole) from an original, first-author publication: L. L. Taylor, J. Qiao, and J. Qiao, “Optimization of femtosecond laser processing of silicon via numerical modeling,” Optical Materials Express, 6(9), 2016 [1]. Slight modifications and/or elaborations may have been included to improve context and clarity as part of the larger dissertation.
multi-pulse processing at a repetition rate of 500 kHz (10 µJ pulse energy, 1 m/s scan speed, 70 µm focal spot) leads to heat accumulation.

Each incident laser pulse radially heats a region of the surface, including locations along the processing path. Figure 3.1 shows that each incident pulse plays a role in raising the surface temperature at locations farther along the scan path in the time leading up to maximum energy deposition at those locations. Heating along the processing path drives the rise in the local base surface temperature over time, as seen in the simulation of experimental processing conditions in Chapter 2. Pulses incident after maximum energy deposition continue to heat previously processed locations in accordance with the Gaussian pulse energy distribution and through heat diffusion, increasing the likelihood for the onset of thermal phenomena. As time increases and the beam moves farther along the scan path, the energy deposited by an incident laser pulse becomes insignificant and the surface temperature relaxes back towards the initial material temperature.
3.1.1 Temperature equilibration

The change in the thermal behavior of silicon during femtosecond laser processing enables equilibration of the laser-induced surface temperature rise and its decay due to heat diffusion. Figure 3.2 shows that the magnitude of the induced temperature rise decreases as more pulses are deposited. This is due to the rise in silicon specific heat capacity with increasing surface temperature. The amount of heat dissipation between pulses increases with the initial incidence of laser pulses because the temperature gradient along the processing path is still steep and silicon acts as a better heat conductor. As more pulses are deposited, the overall surface temperature begins to rise, causing the thermal gradient induced by a single laser pulse to become more gradual. As a result, the heat-conduction performance of silicon worsens, causing less heat to be dissipated by each pulse. Figure 3.2 demonstrates that, as the number of pulses increases, the magnitude of the temperature rise and the magnitude of the temperature decay eventually reach equilibrium, allowing the surface to settle to the same temperature after each pulse (defined as the “equilibrium temperature”).
3.2 Investigating the thermal impact of individual laser processing parameters

The heat accumulation process is affected by the combined impact of spatial and temporal characteristics of femtosecond laser energy deposition. The local temperature maxima are proportional to the peak Gaussian laser pulse fluence. The amount of heat diffusion is controlled by the temporal spacing of the laser pulses. The extent of heat accumulation and the ability to achieve temperature settling are affected by the pulse overlap (PO), calculated as a percentage of the focal spot diameter:

\[
PO = 100 \cdot \left(1 - \frac{v}{(f_{\text{rep}} \cdot D)}\right),
\]

where \(v\) is the scan speed, \(f_{\text{rep}}\) is the repetition rate, and \(D\) is the 1/e^2 focal spot diameter. The Heat Accumulation Model described in Chapter 2 can be used to control heat accumulation by determining an optimal combination of laser pulse energy, focal spot diameter, repetition rate, and sample scan speed. Combinations which enable an equilibrium temperature below the oxidation threshold and local temperature maxima below the melting temperature are considered optimal. The
following numerical studies consider parameter combinations using pulse energies, repetition rates, and focal spot sizes achievable by our in-house femtosecond laser system and scan speeds up to 4 m/s, attainable by commercial galvanometer laser scanning systems.

### 3.2.1 Repetition rate

The sensitivity of heat accumulation to repetition rate was investigated first, as it affects both the temporal exposure to pulses and the pulse overlap. The focal spot diameter, sample scan speed, and pulse energy were respectively fixed at 70 µm, 1.0 m/s, and 10 µJ. The maximum temperature evolutions predicted for repetition rates of 1 MHz, 500 kHz, and 100 kHz are shown in Fig. 3.3. The key simulation results are summarized in Table 3.1.

![Figure 3.3: Maximum temperature evolutions for (a) 1 MHz, (b) 500 kHz, and (c) 100 kHz repetition rates.](image-url)
Table 3.1: Simulation Results for Repetition Rate Sensitivity Study

<table>
<thead>
<tr>
<th>Figure panel</th>
<th>Repetition rate</th>
<th>Local temperature minimum at 30 μs</th>
<th>Predominant thermal regime</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>1 MHz</td>
<td>1200 K</td>
<td>Oxidation</td>
</tr>
<tr>
<td>(b)</td>
<td>500 kHz</td>
<td>483 K</td>
<td>Below oxidation</td>
</tr>
<tr>
<td>(c)</td>
<td>100 kHz</td>
<td>303 K</td>
<td>Initial thermal state</td>
</tr>
</tbody>
</table>

Figure 3.3(a) shows that a 1 MHz repetition rate leads to processing within the oxidation regime with the potential for melting. Figure 3.3(b) shows that, for a repetition rate of 500 kHz, a $2 \times$ increase in diffusion time is achieved, allowing mitigated heat accumulation and processing below the oxide regime. Decreasing the repetition rate to 100 kHz, with diffusion time increased by $5 \times$, allows minimal heat accumulation in the silicon surface, as shown in Fig. 3.3(c). Changing the repetition rate from 1 MHz to 500 to 100 kHz respectively reduces the pulse overlap from 98.5% to 97.1% to 85%, minimizing the exposure of a specific surface region to incident laser pulses.

The repetition rate drives the overall rise in temperature during femtosecond laser processing. Although the 100 kHz repetition rate offers minimal heat accumulation, the 500 kHz repetition rate is used as the fixed parameter in the following numerical investigations so that the sensitivity of heat accumulation to parameter variation can be readily seen.

3.2.2 Scan speed

The scan speed can control the exposure of a surface region to incident femtosecond laser pulses by changing the spatial pulse overlap. Figure 3.4 shows the temperature evolutions at the spatial location corresponding to the center of the 26th incident laser pulse for different scan speeds. The repetition rate, pulse energy, and focal spot diameter were respectively fixed at 500 kHz, 10 μJ, and 70 μm. Figure 3.4(a) shows that, for a scan speed of 1 m/s, the temperature evolution at the fixed location is affected by every pulse incident within 100 μs. Figure 3.4(b) shows that, when increasing the scan speed to 4 m/s, the heating
contribution from non-local pulses is negligible, reducing the time over which that specific
location is heated. This is because increasing the scan speed decreases the pulse overlap
from 97.1% to 88.6% of the focal spot diameter.

Figure 3.4: Surface temperature evolution over time at the spatial location of incidence of the 26th incident
laser pulse along the scan direction for (a) 1 m/s and (b) 4 m/s.

The maximum temperature evolutions for the scan speeds in Fig. 3.4 are shown in
Fig. 3.5. Increasing the scan speed enables faster equilibration of the maximum surface
temperature. After 40 µs, the 1 m/s case achieves a local minimum temperature of 496 K
without reaching equilibrium, while the 4 m/s case achieves an equilibrium temperature of
450 K after 30 µs. The total reduction in heat accumulation for this 4× speed increase is 46
K. The 4 m/s scan speed allows the silicon surface to maintain uniform thermal conditions
throughout processing which will control the onset of thermal phenomena. Therefore, a 4
m/s scan speed is considered optimal.
3.2.3 Laser fluence

Because laser fluence is directly proportional to the laser pulse energy and inversely proportional to the laser spot size, it impacts both the magnitude of peak energy deposition and the surface area affected by an incident pulse. The fluence also determines whether ablation can occur and the corresponding material removal rate [16, 90]. Although heat accumulation must be mitigated in processing, the fluence must be above the ablation threshold (≈0.2 J/cm$^2$ for 515-nm processing and ≈0.45 J/cm$^2$ for 1030-nm processing of silicon [2, 72, 84]) to remove material. To remain above the ablation threshold fluences for silicon, we investigate the sensitivity of heat accumulation to fluence by increasing its value. Specifically, a 2× increase in peak Gaussian laser fluence from 0.52 J/cm$^2$ to 1.04 J/cm$^2$, as shown in Fig. 3.6, was tested for changing either the pulse energy or the focal spot diameter. Fluence was calculated as $F_{\text{laser}} = \frac{(2 \cdot E_{\text{pulse}})}{\pi \cdot w_o^2}$, where $E_{\text{pulse}}$ is the laser pulse energy and $w_o$ is the 1/e$^2$ focal spot radius. For the initial fluence of 0.52 J/cm$^2$, a 10 µJ pulse energy and a 35 µm focal spot radius were assumed. Scanning speed and repetition rate were respectively...
fixed at 1 m/s and 500 kHz. Table 3.2 summarizes the processing parameter combinations used to achieve the simulation results in Fig. 3.6.

Table 3.2: Simulation Results for Fluence Sensitivity Study

<table>
<thead>
<tr>
<th>Figure panel</th>
<th>Fluence</th>
<th>Pulse energy</th>
<th>Focal spot size</th>
<th>Local temperature minimum at 30 μs</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>0.52 J/cm²</td>
<td>10 μJ</td>
<td>70 μm</td>
<td>483 K</td>
</tr>
<tr>
<td>(b)</td>
<td>1.04 J/cm²</td>
<td>10 μJ</td>
<td>49.5 μm</td>
<td>617 K</td>
</tr>
<tr>
<td>(c)</td>
<td>1.04 J/cm²</td>
<td>20 μJ</td>
<td>70 μm</td>
<td>1053 K</td>
</tr>
</tbody>
</table>

Figure 3.6 shows that increasing the fluence by changing either the focal spot size or the pulse energy leads to different amounts of heat accumulation. When increasing the pulse fluence from 0.52 J/cm² to 1.04 J/cm² via focal spot reduction, respectively shown in Figs. 3.6(a) and (b), the rise in the local minimum after 30 μs is 134 K, while increasing the fluence by doubling the pulse energy, as shown in Fig. 3.6(c), leads to a rise of 570 K. The smaller focal spot size constrains the pulse-affected surface region and slightly reduces the pulse overlap from 97.1% to 95.9%. Reducing the focal spot size also enables
Chapter 3. Predicting Heat Accumulation During Laser Processing

steeper temperature gradients, helping to mitigate heat accumulation by improving the heat conduction response of the silicon surface, as discussed in Section 3.1.1.

Increasing the laser fluence to achieve effective ablation can be performed with a minimal rise in heat accumulation by reducing the focal spot size of the beam. Decreasing the fluence to minimize heat accumulation is most effective when reducing the pulse energy, assuming that the ablation threshold is still exceeded. Because a fluence of 0.52 J/cm² offers cooler thermal conditions and exceeds the ablation threshold for silicon, a focal spot size of 70 µm and pulse energy of 10 µJ are considered optimal.

3.3 Discussion

Sensitivity tests showed that the repetition rate plays the most significant role in heat accumulation in femtosecond laser processing of silicon. Changing the repetition rate from 1 MHz to 500 kHz doubled the heat diffusion time and reduced the pulse overlap, causing the local temperature minimum after 30 µs to be reduced by ~700 K. Further reduction of the repetition rate to 100 kHz allowed the surface temperature to decay to its initial temperature between pulses. The repetition rate dictates the extent of heat accumulation in processing and plays the largest role in eliminating the onset of thermal artifacts.

The scan speed can be increased to reduce the heat accumulation effect in femtosecond laser processing by reducing the overlap of incident pulses. Comparing the minimization of heat accumulation for a 4× increase in scan speed (46 K temperature difference after 30 µs) and the 5× reduction in repetition rate (180 K temperature difference after 30 µs for 500 kHz → 100 kHz) showed that increased diffusion time is the main contributor to temperature reduction. Significant minimization of heat accumulation via increased scan speed is restricted by the physical limit of mechanical translation and optical scan speeds and by the pulse overlap required to achieve uniform material removal. Nevertheless, the scan speed plays the largest role in attaining uniform thermal conditions during processing:
a scan speed of 4 m/s enabled temperature equilibration after 30 µs, whereas equilibrium was not predicted for 1 m/s.

The laser pulse fluence must be high enough to enable material removal, but low enough to eliminate unnecessary heat accumulation in processing. When increasing the laser fluence by a factor of 2, it was found reducing the focal spot enabled lower heat accumulation than increasing pulse energy, owing to the reduced beam footprint and pulse overlap. This indirectly shows that reducing the pulse energy provides a more drastic reduction in surface heating than changing the focal spot size. It additionally shows that increasing the ablation efficiency while maintaining lower heat accumulation can be attained by reducing the focal spot size.

The predicted set of optimized processing conditions determined by heat accumulation sensitivity studies includes a 500 kHz repetition rate, a 4 m/s scan speed, a 10 µJ pulse energy, and a 70 µm focal spot diameter. This combination allows equilibrium to be reached below the oxide threshold and for the local temperature maxima to remain below the melting temperature of silicon. With these optimized laser parameters, effective, uniform ablation is expected with mitigated thermal artifacts.

3.4 Conclusion

In this Chapter, the sensitivity of heat accumulation to femtosecond laser and scanning parameters was studied to mitigate detrimental thermal artifacts. The repetition rate was found to drive the overall magnitude of heat accumulation in processing and the scan speed was found to enable uniform thermal processing conditions. A reduction in laser pulse energy was found to enable better mitigation of heat accumulation than an increase of the focal spot size. A set of optimized processing parameters was predicted using the results of the sensitivity study to mitigate heating and the onset of thermal effects in processing. The presented method can be readily used to predict deterministic, optimal
processing parameters for non-thermal femtosecond laser processing of other materials.
Chapter 4

Predicting Laser-Induced Material Breakdown and Phase Change

This Chapter† describes the construction of a Two-Temperature Model to investigate the fundamental mechanism for femtosecond laser / material interaction. The model enables the prediction of ablation and the individual and combined impact of laser parameters on the onset of thermal effects and heat accumulation.

4.1 Modeling approach

The heat accumulation model developed in the preceding Chapters is adequate for exploration of laser parameters to minimize heat accumulation. However, it does not consider the electron-level interaction phenomena occurring on the timescale of the laser pulse and must approximate the initial wavelength-dependent energy absorption and subsequent temperature rise. The model also cannot predict/account for ablation, requiring the onset of breakdown to be confirmed via experiments. Therefore, a more comprehensive modeling method is required to gain a full understanding of laser/material

†The body of this chapter includes relevant excerpts (in part or in whole) from original, first-author publications: (1) L. L. Taylor, R. E. Scott, and J. Qiao, “Integrating Two Temperature and Classical Heat Accumulation Models to Predict Femtosecond Laser Processing of Silicon,” Optical Materials Express, 8(3), 2018 [2] and (2) L. L. Taylor et al., “Femtosecond Laser Polishing of Germanium towards Freeform Optics Fabrication,” Submitted to Optical Materials Express, July 2019. Slight modifications and/or elaborations may have been included to improve context and clarity as part of the larger dissertation.
interaction towards determining optimal laser parameters for polishing.

The overall goal of the modeling work in this dissertation is to establish a method to determine laser parameters which enable non-thermal ablation and/or thermal melting and establish a path for polishing-specific parameter optimization. The modeling method aims to fulfill two sub-goals: (1) predict initial laser parameters which enable non-thermal ablation, (2) predict the impact of laser parameters on heat accumulation and determine parameter combinations to avoid the onset of detrimental melting, oxidation, and material pileup. This method needs to combine accuracy and computational efficiency, be suitable for multi-pulse processing scenarios, have the capability to predict and evaluate the onset of material breakdown and phase change, and alleviate the need for time-consuming, iterative, experimental investigations of optimized laser-parameter combinations. To do this, the modeling approach must accommodate the full laser interaction by considering phenomena occurring on the femtosecond through microsecond timescales.

To achieve the modeling goals, a three-dimensional two-temperature model (TTM) was constructed to simulate and investigate multi-pulse femtosecond laser processing on the femtosecond timescale. The TTM can be used to investigate the impact of laser parameters, e.g., fluence, repetition rate and scanning speed, on the onset of material breakdown, pulse-induced surface temperature rise, and heat accumulation. It can also be extended to simulate laser/material interaction on other semiconductor materials and to improve the accuracy efficiency, and flexibility of the Heat Accumulation Model from Chapter 2.

4.2 Constructing a Two-Temperature Model

To achieve the first modeling goal of predicting initial laser parameters enabling non-thermal ablation, a TTM simulating femtosecond laser-silicon interaction was constructed guided by the works of Chen et al. and Van Driel [91, 92], using MATLAB® (MATLAB R2017a). The TTM simulates four major phenomena to describe ultrafast
laser-material interaction on the pulse timescale, respectively represented by Eqs. 4.1-4.4: complex absorption of energy (4.1), free-carrier electron generation (4.2), carrier temperature change (4.3), and lattice heating (4.4). The temperature, intensity, and wavelength dependencies of each phenomenon are considered, demonstrating improved simulation accuracy over the classical Heat Accumulation Model in Chapters 2-3.

During ultrafast laser-silicon interaction, laser energy is absorbed via linear, nonlinear, and free-carrier absorption mechanisms depending upon the laser intensity and the photon energy in relation to the material bandgap (1.12 eV for silicon). The spatial distribution of intensity in the material bulk is calculated as \[91\]:

\[
\frac{\partial I}{\partial z} = -\alpha I - \beta I^2 - \Theta N_c I \tag{4.1}
\]

In Eq. 4.1, \(I\) is the laser intensity which is a function of material depth, \(z\), and time, \(t\); \(\alpha\), \(\beta\), and \(\Theta\) are the respective single-photon, two-photon, and free-carrier absorption cross section coefficients; and \(N_c\) is the number density of the free-carrier electron system. The absorption of energy drives the generation of free carriers (Eq. 4.2), the carrier-system temperature (Eq. 4.3), and the rise in temperature of the material lattice (Eq. 4.4) \[92\].

\[
\frac{\partial N_c}{\partial t} = \frac{\alpha I}{h \nu} + \frac{\beta I^2}{2h \nu} + \delta N_c - \gamma N_c^3 - \nabla \cdot \vec{J} \tag{4.2}
\]

Equation 4.2 describes the change in carrier density over time. The carrier density increases due to linear and nonlinear absorption of photon energy, \(h \nu\), and impact ionization via collisions of carriers with valence-band electrons at a rate of \(\delta\). The carrier density is decreased by Auger recombination of electrons with valence-band holes, at a rate of \(\gamma\), which dominates the electron-hole recombination process when the density of carriers is high (\(>10^{18} \text{ cm}^{-3}\), achievable during ultrafast laser/Si interaction \[91\]). Auger recombination and impact ionization are three-body processes (electron, electron-hole
pair) which respectively describe the reduction and generation of the number of free carriers, shown schematically in Fig. 4.1. The number density is also impacted by the flow of carriers according to current density, $\bar{J}$.

![Figure 4.1: Schematic descriptions of impact ionization and Auger recombination. Black arrows show collisions/recombinations, gray dotted arrows show electron “movement” and red arrows show energy transfer. Impact ionization: (1) the three-body problem starts with a conduction-band (CB) free-carrier and a valence-band (VB) electron-hole pair (e+h), (2) the free-carrier collides with and transfers energy to the VB-electron, (3) the free carrier loses energy and settles at the CB-minimum, the VB-electron is promoted to the CB-minimum, and a VB-hole remains. Auger recombination: (1) the three-body starting point is the same as the end-result of impact ionization, (2) a CB-electron and VB-hole recombine and the energy emitted is absorbed by the second CB-electron, (3) the CB-electron gains energy to become a free carrier and a VB electron-hole pair is generated.](image)

Equation 4.2 can be used to predict the onset of ablation-based material breakdown. Non-thermal ablation occurs when $N_c$ reaches the critical density for silicon breakdown (6.9 x 10^{20} \text{ cm}^{-3} for 1030-nm wavelength laser pulses [84]) while the bulk lattice temperature remains below the melting point (1687 K [92]). The resulting carrier system temperature follows as [92]:

\[
T_c = \frac{2E_i}{k_B N_c} \left( \frac{v_f}{v_e} \right) \left( \frac{m_e}{m_i} \right) \left( \frac{m_i}{m_c} \right)
\]
\[
C_{eh} \frac{\partial T_c}{\partial t} = \alpha I + \beta I^2 + \Theta N_c I \\
- \frac{C_{eh}}{\tau} (T_c - T_l) - \nabla \cdot \mathbf{W} \\
- \frac{\partial N_c}{\partial t} \cdot (E_g + 3k_b T_c) - \frac{\partial E_g}{\partial t} \cdot N_c
\] (4.3)

Equation 4.3 shows that the carrier system temperature, \(T_c\), is raised by linear, nonlinear, and free-carrier energy absorption. The temperature is decreased by coupling of thermal energy to the material lattice (with temperature, \(T_l\)) at a rate of \(C_{eh}/\tau\), where \(C_{eh}\) is the heat capacity specific to electron-hole pairs and \(\tau\) is the electron relaxation time. Carrier temperature is also impacted by ambipolar diffusion of electrons, with a current of \(\mathbf{W}\), and changes in both the carrier kinetic energy and the material bandgap energy. Here, \(E_g\) is the silicon bandgap energy and \(k_b\) is the Boltzmann constant.

\[
\rho C_l \frac{\partial T_l}{\partial t} = \frac{C_{eh}}{\tau} (T_c - T_l) + \nabla \cdot (\kappa_l \nabla T_l)
\] (4.4)

The interaction of the dense free-carrier electron system with the material results in heating of the material lattice. Equation 4.4 shows that the temperature of the material lattice, \(T_l\), is a function of both thermal energy coupling from the carrier system and heat conduction through the material bulk. In Eq. 4.4, \(\rho\) is the silicon lattice density, \(C_l\) is the specific heat capacity, and \(\kappa_l\) is the thermal conductivity.

The coefficient values used by the TTM and the associated references are given in Table 4.1 and initial/boundary conditions are adapted from reference [92]. Full derivation of the TTM rate equations and an exploration of influence parameters can be found in references [91], [92], and [84].

For each simulation, the initial carrier number density was set to \(N_c = 10^{12} \text{ cm}^{-3}\) and both initial lattice and carrier temperatures were set to 300 K [91, 92]. Spatial and
temporal sampling are simulation-dependent parameters, generally on the respective sub-micron and femtosecond orders, and were selected to avoid steep temperature and carrier density gradients and to ensure complete interactions.
### Chapter 4. Predicting Laser-Induced Material Breakdown and Phase Change

#### Table 4.1: Key parameters for the Si TTM

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bandgap Energy</td>
<td>$E_g$</td>
<td>$1.16 - 7.02 \times 10^{-4} \cdot T_l^2 / (T_l + 1108) - 1.5 \times 10^{-8} \cdot N^{1/3}$</td>
<td>eV</td>
<td>[91,92]</td>
</tr>
<tr>
<td>Reflectivity</td>
<td>$R$</td>
<td>$0.37 + (5 \times 10^{-5}) \cdot (T_l - T_{room})$</td>
<td>- - -</td>
<td>[92]</td>
</tr>
<tr>
<td>Linear absorption coefficient</td>
<td>$\alpha$</td>
<td>$\begin{align*} 515 \text{ nm:} &amp; \quad 5.02 \times 10^{3} \cdot \exp\left(\frac{\eta}{T_{room}}\right) \ 1030 \text{ nm:} &amp; \quad -58.95 + 0.6226 \cdot T_l - 2.309 \times 10^{-3} \cdot T_l^2 + 3.186 \times 10^{-6} \cdot T_l^3 + 9.967 \times 10^{-10} \cdot T_l^4 - 1.409 \times 10^{-13} T_l^5 \end{align*}$</td>
<td>cm$^{-1}$</td>
<td>[84]</td>
</tr>
<tr>
<td>Two-photon absorption coefficient</td>
<td>$\beta$</td>
<td>$\begin{align*} 515 \text{ nm:} &amp; \quad 1.8 \times 10^{-12} \ 1030 \text{ nm:} &amp; \quad 1.5 \times 10^{-9} \end{align*}$</td>
<td>cm/W</td>
<td>[84]</td>
</tr>
<tr>
<td>Free-carrier absorption cross-section</td>
<td>$\Theta$</td>
<td>$\begin{align*} 515 \text{ nm:} &amp; \quad 1.2 \times 10^{-22} \cdot \frac{\eta}{T_{room}} \ 1030 \text{ nm:} &amp; \quad 5.1 \times 10^{-18} \cdot \frac{\eta}{T_{room}} \end{align*}$</td>
<td>cm$^2$</td>
<td>[84]</td>
</tr>
<tr>
<td>Impact ionization coefficient</td>
<td>$\delta$</td>
<td>$3.6 \times 10^{10} \cdot \exp\left(-1.5 \frac{E_g}{k_b T_e}\right)$</td>
<td>s$^{-1}$</td>
<td>[91,92]</td>
</tr>
<tr>
<td>Auger recombination coefficient</td>
<td>$\gamma$</td>
<td>$3.8 \times 10^{-31}$</td>
<td>cm$^6$/s</td>
<td>[91,92]</td>
</tr>
<tr>
<td>Ambipolar diffusion coefficient†</td>
<td>$D$</td>
<td>$18 \cdot \frac{T_{room}}{T_l}$</td>
<td>cm$^2$/s</td>
<td>[91,92]</td>
</tr>
<tr>
<td>Electron relaxation time</td>
<td>$\tau$</td>
<td>$240 \cdot (1 + \frac{N}{6 \times 10^{20}})$</td>
<td>fs</td>
<td>[92]</td>
</tr>
<tr>
<td>Electronic heat capacity</td>
<td>$C_{eh}$</td>
<td>$3N_c k_b$</td>
<td>J/(cm$^3$K)</td>
<td>[91,92]</td>
</tr>
<tr>
<td>Lattice heat capacity</td>
<td>$C_l$</td>
<td>$1.978 + 3.54 \times 10^{-4} \cdot T_l - 3.68 \cdot T_l^{-2}$</td>
<td>J/(cm$^3$K)</td>
<td>[91,92]</td>
</tr>
<tr>
<td>Lattice thermal conductivity</td>
<td>$\kappa_l$</td>
<td>$1585 \cdot T_l^{-1.23}$</td>
<td>W/(cm-K)</td>
<td>[91,92]</td>
</tr>
<tr>
<td>Density</td>
<td>$\rho$</td>
<td>$2.329$</td>
<td>g/cm$^3$</td>
<td>[85]</td>
</tr>
</tbody>
</table>

†Related to $J, W$
4.3 Simulating femtosecond laser/silicon interaction

4.3.1 Predicting the onset of material breakdown

The TTM is used to simulate the impact of laser fluence on the achieved maximum free-carrier number density to determine the threshold for achieving non-thermal ablation. Figure 4.2 shows an example of the TTM prediction of free-carrier electron density, carrier temperature, and lattice temperature for laser-silicon interaction using a fluence above the determined ablation threshold. The carrier energy and temperature rise as free carriers are generated. The peak number density and the peak temperature of the free-carrier system are achieved 0.3 ps after the arrival of the peak pulse intensity. Over time, carrier energy is transferred to the material lattice via collisional interaction [92], causing the lattice temperature to rise and eventually thermalize (equilibrate) with the decaying carrier temperature. The lattice temperature rises to 95% of the equilibrium temperature (847 K) just two picoseconds after the arrival of the peak pulse intensity. For this set of laser parameters, non-thermal ablation is predicted to occur because the peak electron number density exceeds the critical electron density for ablation while the lattice temperature remains below the silicon melting point (1687 K [92]).
Chapter 4. Predicting Laser-Induced Material Breakdown and Phase Change

Figure 4.2: Predicted carrier number density, carrier temperature, and lattice temperature with respective maxima of $9.3 \times 10^{20} \text{ cm}^{-3}$, $2.1 \times 10^{4} \text{ K}$, and $847 \text{ K}$ for a TTM simulation at the laser fluence of $0.53 \text{ J/cm}^2$. The vertical line marks the time of peak pulse intensity at 0 ps.

A numerical sensitivity study of the achieved maximum number density of free-carrier electrons to laser fluence was carried out to predict the onset of material breakdown.

Figure 4.3: TTM-simulated maximum electron number density versus laser fluence for laser-silicon interaction (300-fs pulse width, 1030-nm wavelength). The dashed horizontal line marks the critical density above which ablation occurs ($6.9 \times 10^{20} \text{ cm}^{-3}$ [84]).

Figure 4.3 shows that the maximum free-carrier number density increases with increasing laser fluence. The determined fluence threshold for achieving ablation is $\sim0.45 \text{ J/cm}^2$ at which the carrier density is equal to the critical value of $6.9 \times 10^{20} \text{ cm}^{-3}$. Therefore, the TTM is capable of predicting both the onset of ablation and the
temperature rise in the material lattice to assess and control laser fluence for achieving non-thermal ablation.

4.3.1.1 Experimental validation

The TTM-predicted single-pulse ablation threshold of silicon was experimentally validated via study of ablation sensitivity to laser fluence. An ultrafast laser (Satsuma HP3, Amplitude Systmes) producing 300-fs laser pulses at 1030 nm was focused to a 75-µm (1/e² diameter) spot. The laser fluence was varied by changing the pulse energy. The area of each ablation-generated crater was measured using an optical profiler (Zygo NewView 600).

Figure 4.4: (a) Ablated crater area on silicon versus laser pulse fluence. The ablation threshold was determined to be 0.43 J/cm², corresponding to the x-intercept of the regression. (b) Surface height map of single-shot laser ablation of silicon at 0.65 J/cm² fluence with depth and width on the orders of 10 nm and 35 µm, respectively.

Figure 4.4(a) shows that the ablated crater area increases logarithmically with increasing laser fluence. Figure 4.4(b) is an example of the surface profile of an ablated crater. The ablation threshold fluence for silicon processing at 1030 nm was determined
to be 0.43 J/cm² by fitting Eq. 4.5 to the experimental crater area data in Fig. 4.4(a) [84,93].

\[
A_{ablated} = \frac{\pi \cdot w_o^2}{2} \cdot \ln\left(\frac{F_{laser}}{F_{th}}\right)
\]  (4.5)

In Eq. 4.5, \(w_o\) is the \(1/e^2\) radius of the focused Gaussian beam, \(F_{laser}\) is the peak laser fluence, and \(F_{threshold}\) is the ablation threshold fluence. The determined ablation threshold is consistent with the TTM prediction of 0.45 J/cm² and agrees with the numerical and experimental predictions reported by Thorstensen and Foss [84], validating the capability of the TTM to predict ablation. This result indirectly validates the TTM’s ability to accurately predict lattice temperature because the number density of carriers, which determines the onset of ablation, also influences the carrier system temperature which controls lattice heating.

### 4.3.2 Predicting heat accumulation during multi-pulse processing

The second part of the two-fold modeling goal is to predict the impact of laser parameters on heat accumulation and determine parameter combinations to avoid onset of detrimental melting, oxidation, and material pileup during non-thermal ablation processes. To predict heat accumulation, the TTM was extended to three spatial dimensions to simulate multi-pulse laser-material interaction for area processing – advancing the applicability of existing one-dimensional two-temperature models [91, 92]. In comparison to the Heat Accumulation Model in Chapters 2-3, the TTM offers versatile simulation of heat accumulation for different laser-parameter combinations because it considers the interaction effects of laser wavelength, pulse width, and fluence and can be adjusted to simulate different materials.

The three-dimensional TTM (hereafter simply referred to as “TTM”) calculates laser interaction from irradiation through thermalization of carrier and lattice system
temperatures. The Gaussian-distributed laser pulse intensity is divided into spatially discrete values which serve as inputs to computationally parallel one-dimensional TTM simulations. Following interaction, in the time between laser pulses, heat diffusion is calculated throughout the entire material bulk. Immediately prior to the next pulse-incidence, the laser beam is spatially displaced relative to its previous location according to the laser repetition rate and scanning speed. This procedure is repeated to simulate each subsequent laser pulse.

The TTM and the Heat Accumulation Model from Chapters 2 and 3 were compared in simulating laser interaction using the predicted optimal laser parameters for polishing Si (see Ch. 3) to evaluate the accuracy improvement by the TTM’s additional simulation parameter and phenomena considerations.

Figure 4.5 shows that the classical Heat Accumulation Model prediction of surface temperature evolution is consistent with the TTM prediction for a 350-fs pulse width, a 515-nm wavelength, a 0.52 J/cm² peak laser fluence, a 500-kHz repetition rate, and a 4 m/s laser scanning speed.

Figure 4.5: Surface temperature versus time predicted by the classical Heat Accumulation Model (dotted curve) and TTM (solid curve; from Fig. 3). Models consistently predict settling temperatures for the same set of simulation parameters (515-nm wavelength, 350-fs pulse width, 0.52 J/cm² peak fluence, 500 kHz repetition rate, and 4 m/s laser scanning speed).
m/s scanning speed. The settling temperatures predicted by the classical Heat Accumulation Model differ by only 20 K from the TTM-predicted values, but the laser-induced surface temperature rises are approximately 300 K higher due to the use of the operational absorption coefficient ($A = 0.8$, refer to Ch. 2).

### 4.4 Extension of the Two-Temperature Model and its integration with the Heat Accumulation Model

#### 4.4.1 Simulating other semiconductor materials

The TTM was adapted to model scanning-based femtosecond laser processing of germanium (Ge). Adapting the TTM for Ge required identification and integration of material properties to effectively account for its electronic and thermal behaviors and re-derivation of certain equations to accommodate these changes; the original numerical algorithm was maintained. Therefore, this section only describes relevant TTM modifications to simulate Ge. The key parameters and coefficients for all equations are detailed in Table 4.2.

When a laser pulse is incident on Ge, a fraction of the energy is reflected away and the remainder of the energy is absorbed by the bulk.

$$\frac{dI}{dz} = -\alpha I - \Theta N_c I$$

Equation 4.6 describes the fall-off of intensity along the direction of laser propagation due to energy absorption. When irradiating Ge with 1030-nm light, linear absorption ($\alpha$) dominates since the photon energy is much higher than the material bandgap ($E_p = 1.2$ eV; $E_g = 0.8$ eV). Energy absorption drives the number density of generated free-carrier electrons, $N_c$, orders of magnitude above its initial, intrinsic value of $10^{13}$ cm$^{-3}$, so free-carrier absorption ($\Theta N_c$) also plays a role. The impact of two-photon absorption is negligible when $E_p > E_g$, therefore, it is not considered in this model. This demonstrates
a significant difference between the absorption mechanisms during femtosecond laser interaction with Ge and Si when using near-infrared wavelengths.

\[
\frac{\partial N_c}{\partial t} = \frac{\alpha I}{E_p} - \gamma N_c^3 - \nabla \cdot \vec{J}
\]  

Equation 4.6 shows that the number density of free-carrier electrons is increased by energy absorption and decreased by Auger recombination and ambipolar diffusion. Impact ionization is not considered because it is negligible in comparison to the dominant effect of linear absorption [94].

\[
C_{e-h} \frac{\partial T_c}{\partial t} = \alpha I + \Theta N_c I
\]

\[
- \frac{C_{e-h}}{\tau} (T_c - T_i) - \nabla \cdot \vec{W}
\]

\[
- \frac{\partial N_c}{\partial t} \cdot (E_g + 3k_b T_c) - \frac{\partial E_g}{\partial t} \cdot N_c
\]

Equation 4.7 describes the corresponding temperature of the carrier system. The equation is consistent with that of the Si TTM, but excludes contributions from two-photon absorption which is negligible for NIR processing of Ge.

\[
C_l \frac{\partial T_l}{\partial t} = \frac{C_{e-h}}{\tau} (T_c - T_i) + \nabla \cdot (\kappa_l \nabla T_l)
\]

Equation 4.9 describes the evolution of the lattice temperature throughout the laser-material interaction process and is directly consistent with lattice temperature equation for the Si TTM.
An exploration of the influence of the free-carrier absorption cross section and electron relaxation time on the simulation results was conducted to address the wide range of values reported in the literature [91, 92, 96, 97]. For each of the investigated influence parameter values, the behavior of the TTM-predicted electron number density, electron temperature, and lattice temperature were compared to the results of molecular dynamics (MD) simulations of femtosecond laser/Ge interaction [96]. The free-carrier absorption cross section and electron relaxation time in Table 4.2 were selected for the model because they enable the number density and temperatures to rise and fall to similar orders of magnitude on consistent timescales with the MD simulations.

In the TTM simulations, the laser pulse width and wavelength are 300 fs and 1030 nm,

---

Table 4.2: Key parameters for the Ge TTM

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bandgap Energy</td>
<td>$E_g$</td>
<td>$0.803 - 3.9 \times 10^{-4} \cdot T_l$</td>
<td>eV</td>
<td>[92]</td>
</tr>
<tr>
<td>Reflectivity</td>
<td>$R$</td>
<td>0.39</td>
<td>- - -</td>
<td>[94]</td>
</tr>
<tr>
<td>Linear absorption coefficient</td>
<td>$\alpha$</td>
<td>$1.4 \times 10^4 \cdot (1 + \frac{T_l}{2000})$</td>
<td>cm$^{-1}$</td>
<td>[94]</td>
</tr>
<tr>
<td>Free-carrier absorption cross-section</td>
<td>$\Theta$</td>
<td>$6.6 \times 10^{-20}$</td>
<td>cm$^2$</td>
<td>[84,95]</td>
</tr>
<tr>
<td>Auger recombination coefficient</td>
<td>$\gamma$</td>
<td>$2.0 \times 10^{-31}$</td>
<td>cm$^6$/s</td>
<td>[92,94]</td>
</tr>
<tr>
<td>Ambipolar diffusion coefficient†</td>
<td>$D$</td>
<td>$65 \cdot \left(\frac{T_l}{T_{room}}\right)^{-1.5}$</td>
<td>cm$^2$/s</td>
<td>[92,94]</td>
</tr>
<tr>
<td>Electron relaxation time</td>
<td>$\tau$</td>
<td>$400 \cdot (1 + \left(\frac{N}{10^{21}}\right)^2)$</td>
<td>fs</td>
<td>[96]</td>
</tr>
<tr>
<td>Electronic heat capacity</td>
<td>$C_{e-h}$</td>
<td>$3N_c k_b$</td>
<td>J/(cm$^3$·K)</td>
<td>[92]</td>
</tr>
<tr>
<td>Lattice heat capacity</td>
<td>$C_l$</td>
<td>$1.7 \cdot (1 + \frac{T_l}{6000})$</td>
<td>J/(cm$^3$·K)</td>
<td>[92,94]</td>
</tr>
<tr>
<td>Lattice thermal conductivity</td>
<td>$\kappa_l$</td>
<td>$675 \cdot T_l^{-1.23}$</td>
<td>W/(cm·K)</td>
<td>[92]</td>
</tr>
<tr>
<td>Density</td>
<td>$\rho$</td>
<td>5.323</td>
<td>g/cm$^2$</td>
<td>[85]</td>
</tr>
</tbody>
</table>

†Related to $J, W$
reflecting the fundamental operation state of the in-house femtosecond laser processing system. The initial Ge temperature was set to 300 K and the initial carrier density was set to $10^{13}$ cm$^{-3}$ to mimic the experimental processing environment and sample properties.

4.4.1.1 Experimental validation of the Ge TTM

Experiments were performed on Ge substrates with $<111>$ crystal orientation and 1-nm root mean square (RMS) surface roughness, cleaned with isopropanol and/or methanol before irradiation. The $1/e^2$ radius of the laser focal spot in this experiment is 30 µm.

![Figure 4.6: Sensitivity of Ge ablated crater area to single-shot laser fluence.](image)

An experimental sensitivity study of laser fluence on ablation was carried out using single-shot laser pulses. Figure 4.6(a) shows the impact of laser fluence on the resulting area of ablation craters as measured by a Zygo NewView interferometric microscope. The relationship between the crater area and the laser fluence is given in Eq. 4.5, based on [2,93]. Fitting this relationship to the experimental data yielded an ablation threshold of approximately 0.2 J/cm$^2$ for Ge.

The pulse-induced free-carrier electron number density, carrier-system temperature, and lattice temperature were simulated at a laser fluence of 0.23 J/cm$^2$ near the ablation
threshold.

Figure 4.7 shows that for processing Ge near the ablation threshold, the electron number density increases from an initial value of $10^{13}$ cm$^{-3}$ to the order of $10^{21}$ cm$^{-3}$ in less than one picosecond after the arrival of the peak intensity. This density is characteristic of the onset of material breakdown in semiconductors, indicating the potential onset of Ge ablation [92, 98]. Figure 4.7 also demonstrates that the generation of free-carrier electrons causes the electron-system temperature to rise to over 16,000 K. Over time, this thermal energy from the electron system is transferred to the material lattice via collisional interaction, causing the lattice temperature to rise to nearly 1400 K which exceeds the Ge melting temperature of 1213 K [85], significantly different than the phenomena for Si. This validates the accuracy of the Ge TTM, showing that the TTM is suitable for offline investigation of laser parameters for polishing different materials.

4.4.1.2 Comparing the laser interaction mechanism for Silicon and Germanium

Figure 4.8 compares the temporal evolutions of carrier density, carrier temperature, lattice
temperature, and absorbed energy density on the material surface during interaction of a 1030-nm, 300-fs laser pulse with Ge or Si. The fluence of the simulated laser beam was set to be slightly above the experimentally-determined ablation threshold fluence for each material (~0.2 J/cm² for Ge and ~0.4 J/cm² for Si). These fluences were selected so that the material-specific interaction phenomena can be directly compared.

Figure 4.8: Top: Comparison of the temporal TTM-simulated number density of free-carrier electrons ($N_e$), carrier-system temperature ($T_e$), and lattice temperature ($T_l$) at the location of peak intensity for near-ablation-threshold fluence irradiation of (a) Ge (0.23 J/cm²) and (b) Si (0.45 J/cm²). The dashed horizontal lines correspond to the material-specific melting points. Bottom: Comparison of the contributions from linear, nonlinear, and free-carrier absorption during femtosecond laser irradiation of (a) Ge and (b) Si. All times are relative to the arrival of the peak pulse intensity at 0 ps.
Figures 4.8(a) and (b) show that the material-specific behavior of carrier density, carrier temperature, and lattice temperature are generally similar for laser interaction using fluences near the ablation threshold. For both Ge and Si, the free-carrier density reaches a peak slightly after the incidence of the peak laser intensity at 0 ps. The rise in electron density causes the electron temperature to rise and reach its maximum value within 2 ps after the arrival of the peak pulse intensity. Energy from the electron system is transferred to the material lattice at material-specific coupling rates, however, thermalization occurs on similar timescales for the different materials (e.g., 3-5 ps). Although the general trends of the carrier density/temperature and lattice temperature are similar, three key differences exist: (1) the order of magnitude of the achieved peak electron density, (2) the characteristic evolution of the electron temperature over time, and (3) the temperature to which the material lattice rises in relation to the melting point.

The difference in the number of generated carriers results from the fundamental differences in energy absorption between the materials. Figure 4.8(c) and (d) compare the contributions of linear, two-photon, and free-carrier absorption mechanisms during laser irradiation of Ge and Si, respectively. Figure 4.8(c) shows that for Ge, linear absorption dominates the energy absorption process, as NIR photons at 1030-nm have a photon energy of 1.2 eV, significantly higher than the Ge bandgap of 0.8 eV. For Si, the incident photon energy is only slightly above the bandgap at 1.12 eV. Fig. 4.8(d) shows that for Si, laser energy is initially linearly absorbed, but intensity-dependent two-photon and free-carrier absorption processes are able to dominate the absorption process as the laser pulse intensity ramps up towards its maximum value at 0 ps.

When linear energy absorption dominates the interaction, the ratio of absorbed photons to generated excited electrons is one-to-one. When nonlinear absorption phenomena dominate the interaction, this ratio changes: two-photon absorption requires
two photons to excite one electron and free-carrier absorption does not directly generate any new excited electrons (it only increases the energy of existing electrons). Hence, the increased generation of free-carrier electrons in the Ge simulation results from the dominant linear absorption process.

The energy absorption mechanism and the density of the free carrier systems during femtosecond laser irradiation of Ge and Si influences the characteristic evolution of electron temperature over time. At an early stage in the Ge irradiation process prior to incidence of the peak intensity laser intensity, a local maximum / “shoulder” in carrier temperature can be seen. This peak occurs because the heat capacity of the generated carrier system is still low, due to the lower carrier density and rate of energy absorption at this early irradiation time [96]. For Si, a slight inflection of the carrier system temperature can be seen just prior to the arrival of the peak laser intensity, indicating the potential onset of a local maximum. However, at this irradiation time, the laser intensity is closer to its maximum value and the number density of electrons is significantly higher in comparison to the time of arrival of the local maximum in the Ge carrier temperature evolution. Therefore, the carrier system temperature continues to rise to its maximum value.

A slight difference in the maximum value of the carrier temperature can be seen for Ge and Si, resulting from the fact that nonlinear absorption processes dominate for Si near the time for incidence of the peak pulse intensity. In comparison to linear energy absorption, two-photon and free-carrier absorption processes contribute more significantly to increasing the carrier-system energy and, hence, its temperature. The delayed arrival of the carrier temperature maximum in Ge results from the continued, high linear absorption as the laser pulse intensity returns back to zero.

Although the peak Si carrier temperature is higher than that of Ge, the Ge thermalization occurs on a slightly slower timescale owing to the material-specific electron relaxation time and the higher number density of generated carriers. The higher
number of carriers also increases the potential for energy coupling to the material lattice, explaining the higher resulting lattice temperature $\sim 1400$ K (above the melting point) for Ge versus $\sim 900$ K (below the melting point) for Si.

The meticulous nature of the TTM simulation allows offline investigation of the differences between ultrafast laser interaction with different materials, showing the value and accuracy of the model for predicting the individual and combined impact of laser parameters on material breakdown and heat accumulation.

4.4.2 Extending the Heat Accumulation Model to the general case

As standalone models, neither the TTM nor the Heat Accumulation Model is ideal for simulating multi-pulse interaction. The TTM is unsuitable for extension to area laser processing simulations due to lengthy processing times. The time required to complete each TTM simulation depends on its spatial and temporal discretization, the number of voxels comprising the simulated material lattice, and the number of timesteps required to achieve thermalization and complete heat diffusion. For example, the five-pulse TTM simulation shown in Fig. 4.5 takes approximately 3.5 hours ($\sim 0.7$ hours per pulse) to complete when using 2-μm sampling of the input laser intensity, sub-micron/fs-order spatial/temporal sampling for the parallel one-dimensional TTM simulations, and using cubic-micron-order voxels/nanosecond timesteps for heat diffusion calculations. Therefore, a more time-efficient method is required to simulate heat accumulation during multi-pulse laser-material interaction for area processing because the TTM simulation time is unsuitable for simulating hundreds to thousands of laser pulses.

In contrast to the TTM, the Heat Accumulation Model can provide increased simulation efficiency due to the reduced complexity of the model. However, the Heat Accumulation Model is case-specific to laser processing conditions and material properties, requiring a priori determination of the input absorption coefficient. To extend
the time-efficient Heat Accumulation Model to the general case, an offline method is required to determine the absorption coefficient across all potential combinations of laser processing parameters and for various materials since it is time-consuming and impractical to determine experimentally.

4.4.2.1 Improving the accuracy of the Silicon Heat Accumulation Model

To overcome the trade-off between model versatility and time-efficiency and enable a methodology to scale up simulations for offline investigation of line and area configuration processing, the TTM can be used to improve the efficiency of the Heat Accumulation Model. The accuracy of the TTM can be integrated into the Heat Accumulation Model via the following strategy: (1) use the TTM to numerically determine an operational absorption coefficient describing the lattice energy after thermalization and (2) feed this coefficient to the Heat Accumulation Model in place of the operational absorption coefficient, $A$, to improve its accuracy. This integrated modeling methodology eliminates the need for time-consuming, iterative experimental calibration of $A$ and allows simulations to maintain the meticulous nature of a TTM simulation since all TTM-simulated physical phenomena and in-situ changes in optical/material properties are inherently incorporated into the absorption coefficient determination. The method also enables simulation times comparable to those of the classical Heat Accumulation Model since the multi-pulse TTM simulations for coefficient determination can be performed offline and fed back, as needed, as inputs to the Heat Accumulation Model.

The TTM can be used to predict an operational absorption coefficient via Eq. 4.10, which determines the ratio of the energy, $E_a$, absorbed by the material lattice once thermalization is achieved and the energy of a single laser pulse, $E_p$.

$$\eta = \frac{E_a}{E_p} \quad (4.10)$$
Absorbed energy, $E_a$, is calculated as the difference between the lattice energy $E_l$ in its thermalized and pre-irradiation states. The energy in each of these states can be calculated by rearranging Eq. 2.2 as $E_l = \rho \cdot V \cdot C_l(T_l) \cdot T_l$.

The average absorption coefficient for the five-pulse TTM simulation was found to be $\bar{\eta} = 0.59$. This coefficient was fed to the classical Heat Accumulation Model in place of $A$ in Eq. 2.1 to re-calculate the heat accumulation using the same laser parameters as in Fig. 4.5.

Figure 4.9 shows that the simulation of heat accumulation using the improved, integrated modeling method consistently predicts surface temperatures with the TTM (differing by less than 65 K) whereas the original Heat Accumulation Model over-predicted temperature rises by up to 300 K. The simulation durations of the integrated modeling method are equivalent to the original Heat Accumulation Model ($< 4$ min. for five pulses). The surface temperature agreement and the maintained time-efficiency show that the integrated modeling method enables accurate predictions of
heat accumulation and is suitable for simulating multi-pulse laser-silicon interaction. However, it is important to recognize that the integrated modeling method is capable of predicting optimal laser parameters to control heat accumulation, while only the TTM is capable of predicting laser fluences for achieving non-thermal ablation.

4.4.2.2 Adapting the Heat Accumulation Model for Germanium

The strategy to achieve simultaneous accuracy and efficiency in predicting surface temperature evolution by using the TTM to improve the Heat Accumulation Model can enable extension to accommodate different laser parameters. Hence, the Heat Accumulation model can be adapted to simulate laser interaction with other semiconductor materials including Ge.

Extending the TTM for Ge in Section 4.4.1.2 showed that for 1030 nm light, linear absorption dominates the energy-absorption process and that impact ionization is negligible. This means that the key proponent of electron generation during laser/Ge interaction is linear absorption and that all energy which is not reflected away has the potential to contribute to increasing the energy of the material lattice. Therefore, the Ge residual energy coefficient in the Heat Accumulation Model from Chapter 2 can be simplified to

$$A = (1 - R)$$

where $R$ is the reflectivity of Ge (0.39 for 1030-nm light, refer to Table 4.2). Because light is absorbed linearly, the penetration depth can be calculated as the inverse of the linear absorption coefficient in Table 4.2. Over the range of temperatures for which the material remains solid (e.g. 300 K to 1213 K), the predicted energy penetration depth ranges from 0.62 to 0.44 $\mu$m, decreasing with increasing lattice temperature. As such, the Heat Accumulation Model’s computational efficiency driven assumption that full energy absorption occurs within a 1.5 $\mu$m thick surface layer holds for simulating 1030-nm processing of Ge.

The Ge TTM enabled sufficient understanding of laser/material interaction process to enable conversion of the Heat Accumulation Model to accurately and efficiently predict
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femtosecond laser processing of Ge. The reflectivity, lattice heat capacity, and thermal conductivity of Ge in Table 4.2 were used to replace the appropriate parameters in the Heat Accumulation Model equations described in Chapter 2. The behaviors of the Ge heat capacity and thermal conductivity are shown in Fig. 4.10.

Figure 4.10: Temperature dependence of conductivity and specific heat capacity of germanium. As temperature is increased from 273 to 1213 K (germanium melting point), thermal conductivity decreases by approximately 60 W/(mK) [92], while specific heat capacity increases by approximately 40 J/(kgK) [92].

The Ge Heat Accumulation Model was used to simulate laser processing using an initial set of laser parameters: 0.7 J/cm², 500 kHz repetition rate, and 1 m/s scanning speed, guided by the range of parameters determined to achieve minimized heat accumulation via the numerical sensitivity study of laser parameters in Chapter 3. Figure 4.11 compares the Heat-Accumulation-Model-predicted surface temperature evolution on Ge and Si when simulating the same processing conditions.
Figure 4.11: Comparison of heat accumulation modeling of Ge (left) and Si (right) using the same laser parameters: 0.7 J/cm², 500 kHz repetition rate, and 1 m/s scanning speed.

Figure 4.11 shows that, the Heat Accumulation Model predicts that a fluence of 0.7 J/cm² will induce higher surface temperature rises on Ge in comparison to Si. This results from the difference in the dominating energy absorption mechanisms and because the specific heat capacity for Ge is significantly lower than that of Si (compare Fig. 4.10 and Fig. 2.4). Because the thermal conductivity of Ge is lower, Ge cannot diffuse heat as well as Si. Therefore, Ge has a higher propensity for heat accumulation than Si when processing both materials with the same set of laser parameters. Although the Heat Accumulation Model can be used to investigate the thermal behavior of Ge and predict the impact of various laser parameters during femtosecond laser processing, it is still imperative that the numerical investigations be supplemented by experiments. This way, the onset and extent of ablation and thermal effects can be understood and used to guide the selection of optimal laser parameters to achieve laser polishing.
4.5 Conclusion

In this Chapter, a TTM was constructed to enable accurate prediction of non-thermal ablation and heat accumulation during multi-pulse femtosecond laser/Si interaction. The Si TTM was effectively extended to simulate laser interaction with Ge to provide a strategy for offline assessment of the impact of laser parameters for processing different materials. Therefore, the TTM can be used as a tool to predict optimal laser parameters for polishing.

The TTM was found to have additional utility in improving the accuracy of the classical Heat Accumulation Model from Chapter 2 to incorporate the impact of laser parameters which are not directly considered by the algorithm (e.g., wavelength) and to extend it to the general case to simulate different materials. The integrated modeling method overcomes the trade-off between model versatility and time-efficiency, demonstrating a path for model reduction and time-efficient simulation of multi-pulse ultrafast laser processing.
Chapter 5

Experimentally Investigating the Impact of Laser Parameters on Processing Efficiency and Quality

In order to determine effective laser parameter combinations for polishing, the thermal and ablation impacts of individual and combined laser parameters must be understood. Although the TTM and Heat Accumulation Models can predict these impacts, it is critical to experimentally validate the processing. For space-selective laser-based polishing, it is also important to experimentally investigate the spatial characteristics of ablation during point and line processing, the generation and redeposition of ablated material, and the resulting surface roughness. These three factors are critical for determining the effectiveness, efficiency, and controllability of laser-based material removal and determining an experimentally-validated strategy for laser polishing. This Chapter discusses the construction of a flexible laser ablation system for polishing and experimental sensitivity studies to evaluate the individual and combined impact of laser parameters on material removal and surface quality.
5.1 Construction of a femtosecond laser ablation system

The laser-based material removal experiments and numerical modeling studies in Chapters 2-4 demonstrated the need for tunable laser processing parameters to control the laser interaction mechanism and the onset of ablation and thermal effects towards polishing. To enable adequate laser polishing investigations, a femtosecond Ytterbium fiber laser-based ablation system with controllable repetition rate, pulse energy, focal spot size, and scanning speed was constructed (Fig 5.1).

![Figure 5.1: Updated femtosecond laser ablation system for polishing experiments consisting of (A) a femtosecond laser from Amplitude Systèmes, (B) a beam expansion, attenuation, and power monitoring system, (C) a high speed, three-axis optical laser scanner, and (D) a sample positioning stage.]

The Ytterbium fiber laser source (Satsuma HP3, Amplitude Systèmes; Fig. 5.1 A) generates laser pulses with tunable energy up to 40 µJ. It can operate in single-shot and repetitive-shot modes with repetition rates up to 2 MHz, enabling a maximum average power of up to 50 W. The laser also has tunable pulse width from ~300 fs to 10 ps, and can operate at NIR (1030 nm) and visible (515 nm) wavelengths. For polishing investigations, the pulse width is set to its smallest value, and the operational wavelength is fixed at 1030 nm to accommodate the AR coatings of downstream optical hardware for beam monitoring/adjustment and laser scanning. Upon exiting the laser housing, the laser is directed through a beam control module (LS-Shape, Lasea, Fig. 5.1 B). The device contains
a variable attenuator and photodiode-based power sampling unit to dynamically control and monitor the laser power/energy during laser processing. It also contains a manual variable beam expander unit which can be used to adjust the minimum-achievable focal spot size. After exiting the beam expansion/monitoring device, the laser is directed upwards and into a high speed, three-axis optical galvanometer-based laser scanner (LS-Scan, Lasea, Fig. 5.1 C), coupled to an f-theta focusing lens. The achieved $1/e^2$ focal spot diameter for the system is $60-75$ µm. The scanning unit has the capability to perform up to $\pm 8$ mm of beam defocus by optically adjusting the focal spot height. The defocus can be further increased by adjusting the height of the sample in relation to the beam focus. The scan mirrors and f-theta lens enable a lateral processing area of approximately $100$ mm $\times$ $100$ mm over which the laser can remain in focus. The galvanometer scan mirrors can enable high processing speeds up to $4$ m/s. The laser polishing system is also equipped with a sample fixturing and positioning stage (Fig. 5.1 D) to ensure accurate and adjustable alignment of various samples to the laser system.

To enable dynamic control of the laser polishing system, the laser, beam adjustment, and scanning hardware in Fig. 5.1 were aligned and electronically integrated. An electronic circuit was built to power, control, and integrate “Bridge” and “Laser Head Interface” printed circuit boards which respectively drive the beam control module and the laser scan head. The “Bridge” board was wired to communicate with and drive a power-sampling photodiode and power meter, a flip mirror to control beam pointing towards module output or into the power meter, a mechanical shutter for full beam attenuation, and pellicle-based variable beam attenuator. The “Laser Head Interface” board was wired to power the laser scan head and to enable synchronized operation of the laser and scan head by sending $5V$ output signals to correctly trigger and/or gate the laser emission for specific marking jobs. The scanner was calibrated to ensure accurate spatial precision in laser marking and to establish timing delays to improve the laser emission response to avoid hot spots.
from laser dwell. Laser-based processing routines were generated using the Newson Rhothr
Script Executor (environment for writing and executing .dll-library scripts for marking [99])
and/or the Lasea Kyla Control Software (CAD/G-Code programming environment [100]).

5.2 Evaluating the individual impact of laser parameters on material removal

Line-configuration laser ablation experiments were conducted to investigate the impact of
individual and combined laser parameters on Ge material removal and surface quality. The
individual impacts of laser fluence, repetition rate, and scan speed were considered.

Experiments were carried out on Ge substrates with $<111>$ crystal orientation
and $\sim$1-nm RMS surface roughness, cleaned with isopropanol and/or methanol before
irradiation. Laser fluences above the Ge ablation threshold for 1030-nm processing (0.2
J/cm$^2$) were selected for investigation and fluence was tuned by adjusting the pulse energy
rather than by changing the focal spot size. A matrix of laser parameters was determined
to reduce the number of experiments to investigate material removal and surface quality,
guided by the numerical sensitivity study on heat accumulation in Chapter 3. The laser
parameter matrix is given in Table 5.1. To investigate the individual impact of a laser
parameter on material removal, one parameter was varied at a time while the two others
were held fixed.
5.2.1 Laser fluence

5.2.1.1 Material removal

The sensitivity of Ge material removal to laser fluence was investigated for a fixed repetition rate of 250 kHz and a fixed scan speed of 1.5 m/s. The average removal profiles over a 300-µm scan length are shown in Fig. 5.2.
Figure 5.2: Average material removal profiles and volumes for processing Ge using different laser fluences. Profiles are averaged over a 300-μm scan length. The repetition rate was fixed at 250 kHz and the scan speed was fixed at 1.5 m/s.

Figure 5.2 demonstrates that changing the laser fluence can control the amount of material removal while maintaining the general shape of removal track. When increasing the pulse fluence by a factor of five, the material removal volume increases by a factor of 6.5, demonstrating nearly linear control of the material removal. However, further investigation at fluences closer to the ablation threshold is required to draw a complete conclusion on the linearity of material removal. It is also anticipated that the removal volume can be further increased by using different combinations of repetition rates and scanning speeds to increase the amount of energy deposited per unit area, per unit time.

Figure 5.2 also shows that pulse fluence has the capability to control the spatial-selectivity of the ablation process. The width of the laser processing track at the material surface is directly related to the spatial extent of the laser beam which is above the threshold fluence. The relationship between crater area and laser fluence for the single-shot ablation threshold studies in Chapter 4 can be generalized to describe line-configuration processing: \( W^2 = \frac{w_o^2}{2} \cdot (\ln(F_{\text{laser}}) - \ln(F_{\text{th}})) \), where \( W \) is the surface-width of the ablated track, \( w_o \) is the 1/e² width of the laser beam (33 μm in this
study), and \( F_{\text{laser}} \) and \( F_{\text{th}} \) are the respective laser and threshold fluences [84,93].

Figure 5.3: Surface-width of ablation track versus pulse fluence for line-configuration processing using the same laser parameters as for Fig. 5.2.

Figure 5.3 shows that the squared surface-width of the ablated track follows the defined logarithmic trend. Setting \( y \) to zero in the fitting equation allows the ablation threshold fluence to be estimated, yielding a value of \( \sim 0.16 \text{ J/cm}^2 \) which is consistent with the ablation threshold fluence of \( 0.2 \text{ J/cm}^2 \) determined in Chapter 4. The slightly lower ablation threshold for area processing may be due to incubation effects from multiple laser pulses [101].

Overall, laser fluence is capable of controlling the space-selectivity of the laser pulse and scaling up the material removal volume towards accommodating different optical fabrication tasks.

5.2.1.2 Surface quality

Although tunable material removal is highly desirable for polishing applications, it is also critical to determine the capability for laser parameters to generate a smooth surface. Images of the laser fluence sensitivity study on Ge were collected using scanning electron
microscopy (SEM). Figure 5.4 shows images of processing using three different laser fluences.

Figure 5.4: SEM images of laser processing using laser fluences of 0.35, 0.82, and 1.75 J/cm$^2$. The repetition rate was fixed at 250 kHz and the scan speed was fixed at 1.5 m/s.

In each of the processed tracks in the SEM images in Fig. 5.4, the laser-affected region contains two key features: (1) a smooth, modified outer edge and (2) a rough central track. The width of the rough track increases according to the spatial area of the incident beam that is above the ablation threshold. When increasing the laser fluence, the size of the rough features in the center of the ablated track also increase, potentially exacerbated by thermal effects owing to the higher incident fluence. A Zygo New View interferometric microscope was used to measure the central roughness of the ablated lines. For the 0.35 J/cm$^2$ the average RMS roughness within a 20 µm × 300 µm area was found to be on the order of 213 nm, significantly rougher than the ∼1 nm roughness of the original surface. Increasing the fluence to 0.82 and 1.40 J/cm$^2$ respectively increased the RMS roughness to 283 and 392 nm. Although increasing the fluence can significantly improve the material removal volume, it can be detrimental to the resulting surface quality. This study demonstrates the need to carefully control the laser fluence to minimize the roughness of polishing. In order to achieve polishing, it is important to further investigate the material removal and surface quality impact of laser fluences closer to the ablation threshold.
5.2.2 Repetition rate and scan speed

5.2.2.1 Material removal

The sensitivity of Ge material removal to both repetition rate and scanning speed was investigated for a fixed fluence of 0.47 J/cm$^2$. The secondary fixed parameters (repetition rate or scan speed) were carefully selected to ensure the same achieved pulse overlaps so that the studies could be directly compared. For the repetition rate study, the scan speed was fixed at 2.5 m/s and for the scan speed study, the repetition rate was fixed at 250 kHz. The average removal profiles for both sensitivity studies over a 300-μm scan length are shown in Fig. 5.5.
Figure 5.5 shows that repetition rate and scan speed are both capable of controlling the material removal volume while maintaining a relatively consistent ablation surface-width. The increased removal depths result from reducing the time between pulse energy deposition and/or the spatial overlap of the laser pulses. The removal width remains relatively consistent across the different parameter combinations because changing the repetition rate and scan speed does not affect the spatial extent of the laser beam which exceeds the ablation threshold, as was the case in the laser fluence study.

Figure 5.5 also shows that the relationship between material removal volume and laser
repetition rate / scan speed differs. For repetition rate, the material removal volume is controlled by changing the temporal deposition of the laser pulses, which directly impacts the amount of energy deposited per second. Repetition rate controls the removal volume in a nearly linear fashion since energy deposition is proportional to material removal volume: increasing the repetition rate by a factor of 4 (from 150 to 625 kHz) increases the volume removal by a factor of 3. For scan speed, the achieved material removal volume is controlled by changing the spatial deposition of the laser pulses, which only impacts the energy deposited per unit area during line-configuration processing. Hence, the material removal volume is inversely proportional to the scan speed. To directly compare the material removal controllability from changing the repetition rate and scan speed, Fig. 5.6 compares the material removal volumes and rates corresponding to the achieved pulse overlaps.

Figure 5.6: Material removal depth versus pulse overlap.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Repetition rate (kHz)</th>
<th>Scan speed (m/s)</th>
<th>Pulse energy (µJ)</th>
<th>Spot size (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scan speed</td>
<td>250</td>
<td>4, 3, 1.5, 1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rep. rate</td>
<td>150, 200, 400, 625</td>
<td>2.5</td>
<td>8</td>
<td>66</td>
</tr>
</tbody>
</table>

Figure 5.6 shows that the impact of pulse overlaps achieved by controlling the scan speed or repetition rate achieve consistent volume removals, demonstrating the potential capability for scaling material removal volume while maintaining space-selectivity during
processing. However, only varying the repetition rate is capable of scaling up the material removal rate since it increases both energy deposited per unit area and per unit time.

5.2.2.2 Surface quality

Since changing the laser repetition rate and scan speed affect the pulse overlap achieved during line-configuration processing, it is critical to evaluate the combined impact of these parameters on processing uniformity and surface quality. Figures 5.7 and 5.8 respectively show SEM images of lines processed using high scan speeds and low repetition rates achieving pulse overlaps of 76, 82, and 85% of the $1/e^2$ focal spot diameter of the laser beam (66 µm in this study).

Figure 5.7: SEM images of laser processing using scan speeds of 4.0, 3.0 and 2.5 m/s achieving respective pulse overlaps of 76, 82, and 85%. The laser fluence was fixed at 0.47 J/cm² and the repetition rate was fixed at 250 kHz. Some debris is evident in the images, e.g., “peanut-shaped” dust particle in the leftmost image and oil mark in the rightmost image.
Figure 5.8: SEM images of laser processing using repetition rates of 150, 200, and 250 kHz achieving respective pulse overlaps of 76, 82, and 85%. The fluence was fixed at 0.47 J/cm$^2$ and the scan speed was fixed at 2.5 m/s.

Figures 5.7 and 5.8 show that processing with pulse overlaps of 76% and 82% via low repetition rates and high scan speeds affects the uniformity of the processing: large circular / ripple artifacts can be seen in the central rough ablated track and the edge uniformity of the rough central track is perturbed. When increasing the pulse overlap to 85% for both parameters, the central ablated region of the processing track becomes uniform.

When processing at higher pulse overlaps, the thermal impact of processing can become a concern, owing to the increased energy deposited per unit area and/or unit time. Figures 5.9 and 5.10 respectively show SEM images of lines processed using low scan speeds and high repetition rates achieving pulse overlaps of 91, 94, and 97% of the $1/e^2$ focal spot diameter of the laser beam (66 μm in this study).
Chapter 5. Impact of Laser Parameters on Processing Efficiency and Quality

Figure 5.9: SEM images of laser processing using scan speeds of 1.5, 1.0, and 0.5 m/s achieving pulse overlaps of 91, 94, and 97%. The laser fluence was fixed at 0.47 J/cm$^2$ and the repetition rate was fixed at 250 kHz.

Figure 5.10: SEM images of laser processing using repetition rates of 400, 625, and 1250 kHz achieving pulse overlaps of 91, 94, and 97%. The laser fluence was fixed at 0.47 J/cm$^2$ and the scan speed was fixed at 2.5 m/s.

Figures 5.9 and 5.10 consistently show that increasing the pulse overlap to 91 and 94% results in an increase in the size of the rough features in the center of the ablated track. However, when processing with a pulse overlap of 97%, the resulting surface quality is drastically different for the high repetition rate case. Figure 5.10 shows that increasing the repetition rate to 1250 kHz eliminates the two-feature processing track (e.g. smooth outside tracks and rough central structure). Instead, a smooth processing track is generated because the high overlap and quasi-continuous laser irradiation process.
enable the onset of significant melting during the ablation process. This is further evidenced by the semicircular ripple features in the center of the ablated track resulting from the resolidification of molten material. Figure 5.9 shows that processing with a 97% pulse overlap achieved by changing the scan speed still produces the two-feature track, indicating that the transition to the melting regime is more significantly impacted by increasing the energy deposited per unit time, rather than the energy deposited per unit area.

The numerical Heat Accumulation Model of femtosecond laser / Ge interaction was used to investigate the surface temperatures achieved by processing at high pulse overlaps. Figure 5.11 compares numerical heat accumulation modeling simulations of the Ge surface temperature response to processing using 94% and 97% pulse overlaps for different laser repetition rates and scan speeds.

Figure 5.11: Numerical modeling of high-pulse-overlap processing achieved using (a) high repetition rates of 625 kHz and 1250 KHz (94% and 97% overlaps), and (b) a low scan speed of 0.5 m/s (97% overlap). For the repetition rate study, fluence was fixed at 0.47 J/cm$^2$ and the scan speed was fixed at 2.5 m/s scan speed. For the scan speed study, fluence was fixed at 0.47 J/cm$^2$ and the repetition rate was fixed at 250 kHz.
Figure 5.10(a) shows that, for processing with a 625 kHz repetition rate (94% overlap), the surface temperature achieved prior to each incident pulse approaches the melting point over time, confirming the fact that high repetition rates can induce high thermal impact processing conditions. For 1250 kHz (97% overlap), higher heat accumulation is predicted, driving the surface temperature above the melting point after just five laser pulses. The predicted high thermal processing condition for the 1250 kHz case is consistent with the melting-dominated processing result in Fig. 5.11.

Fig. 5.10(b) shows that reduced thermal impact is predicted for the low scan speed case of 0.5 m/s, even though its combination with a 250 kHz repetition rate enables the same 97% overlap as for the highest repetition rate case. In comparison to the 1250 kHz high repetition rate case, the 250 kHz repetition rate in this study allows five times longer for the material surface to cool between pulses. Therefore, the energy deposited per unit time (e.g., repetition rate) most significantly drives the onset of thermal processing conditions.

Although the 1250 kHz repetition rate case achieves a pseudo-polished result, the achieved high thermal impact processing conditions are not desirable for laser polishing as they can lead to material pileup, oxidation, and sub-surface damage. Additionally, the surface roughness in the center of this smooth track is on the order of 20-50 nm RMS and cannot meet the sub-nanometer optical tolerances for surface quality.

5.3 Evaluating the combined impact of laser parameters on controllable material removal

To investigate the overall controllability of the laser polishing process, a metric was defined which accounts for the combined impact of laser parameters – total deposited energy per unit distance along the scan direction (J/m):

\[
\frac{J}{m} = \frac{\text{Repetition Rate} \cdot \text{Pulse Energy}}{\text{Scan Speed}}
\]  

(5.1)
Chapter 5. Impact of Laser Parameters on Processing Efficiency and Quality

The metric in Eq. 5.1 is directly related to material volume. Therefore, it indicates that the repetition rate and pulse energy (laser fluence) have direct proportionality with material removal volume, while scan speed follows inverse proportionality. The relationship between material removal volume and the defined metric for sensitivity study parameter combinations from the previous sections is shown in Fig. 5.12.

![Graph showing material removal volume versus total deposited energy](image)

<table>
<thead>
<tr>
<th>Varied parameter</th>
<th>Repetition rate (kHz)</th>
<th>Scan speed (m/s)</th>
<th>Pulse energy (µJ)</th>
<th>Spot size (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Repetition rate</td>
<td>150, 200, 400, 625</td>
<td>2.5</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>Pulse energy</td>
<td>250</td>
<td>1.5</td>
<td>6, 8, 10, 14, 18, 24</td>
<td>66</td>
</tr>
<tr>
<td>Scan speed</td>
<td>250</td>
<td>4, 3, 2.5, 1.5, 1.0, 0.5</td>
<td>8</td>
<td></td>
</tr>
</tbody>
</table>

Figure 5.12: Removal volume versus total deposited energy along a 300-micron line-scan length. The parameters used to determine the energy deposited per unit length are summarized in the table.

Although different combinations of laser parameters were used to process Ge, Fig. 5.12 shows that the achieved material removal volumes follow a consistent linear trend with increasing deposited energy per unit scan length. This validates the established metric as a strategy to consider the combined impact of laser parameters on material removal. The metric can act as a means for determining combinations of laser parameters to accommodate different processing tasks and scale-up the material removal. It also demonstrates a path towards achieving deterministic ultrafast laser processing.
5.4 Conclusion

In this Chapter, a flexible laser ablation system with tunable fluence, repetition rate, and scan speed was constructed to carry out laser polishing experiments and investigate the impact of laser parameters on material removal and surface quality. Experimental sensitivity studies on the individual impact of laser parameters on processing drew several key conclusions:

- Laser fluence
  1. Controls material removal volume in a near-linear fashion
  2. Drives space-selectivity of laser/material interaction
  3. Significantly impacts the achieved surface roughness

- Repetition rate
  1. Controls material removal volume in a near-linear fashion while maintaining space-selectivity of the laser/material interaction
  2. Directly impacts the material removal rate and processing uniformity
  3. Drives the onset of melting due to control of surface cooling time

- Scan speed
  1. Inversely controls material removal volume while maintaining space-selectivity of the laser/material interaction
  2. Directly impacts processing uniformity
  3. Can improve the ablation efficiency while maintaining low thermal impact if paired with an appropriate repetition rate
Although flexible processing was demonstrated, the SEM images of the processed surfaces in this study revealed that high amounts of ablation led to increasingly rough surfaces. Processing at low pulse overlaps led to increasingly nonuniform processing, while processing at high pulse overlaps led to the onset of thermal melting effects. Because rough surface features, nonuniform material removal, and high thermal impact processing conditions are unsuitable for achieving a polished surface, a new approach is needed to control laser parameters to achieve polishing. A potential approach for determining laser polishing parameters is established as follows:

1. Reduce the laser fluence to control the onset of rough surface structures
2. Adjust the repetition rate to allow sufficient time for the surface to cool between pulses to minimize the thermal impact of processing
3. Balance the scan speed and selected repetition rate to control the pulse overlap and achieve uniform processing edges and material removal depths

This approach will require both numerical and experimental investigations of the impact of laser parameters on material removal and surface quality, and necessitates a strategy for extending to area-based polishing.
Chapter 6

Establishing and Implementing a Methodology for Laser Polishing

The numerical and experimental investigations of femtosecond laser / material interaction in the preceding chapters revealed the importance of precisely controlling laser parameters to balance the competing mechanisms of ablation, surface heating, heat diffusion, and thermal effects to achieve high precision material removal. This Chapter† presents a strategy to achieve laser polishing based on the integrated numerical and experimental approach defined in Chapter 5, outlined in Table 6.1:

†The body of this chapter includes relevant excerpts (in part or in whole) from an original, first-author publication: L. L. Taylor et al., “Femtosecond Laser Polishing of Germanium towards Freeform Optics Fabrication,” Submitted to Optical Materials Express, July 2019. Slight modifications and/or elaborations may have been included to improve context and clarity as part of the larger dissertation.
### 6.1 Numerical prediction of optimal laser polishing parameters

#### 6.1.1 Laser fluence

The Two-Temperature Model (TTM) formulated in Chapter 4 was first used to investigate the stand-alone impact of fluence during femtosecond laser / Ge interaction, as it can independently drive the onset of ablation and heating of the material surface. The pulse-induced free-carrier electron number density, carrier-system temperature, and
Chapter 6. Establishing and Implementing a Methodology for Laser Polishing

Lattice temperature were simulated to predict the potential onset of ablation and melting at different fluences. In the TTM simulations, the laser pulse width and wavelength are 300 fs and 1030 nm, reflecting the fundamental operation state of our in-house femtosecond laser processing system. The initial Ge temperature was set to 300 K and the initial carrier density was set to $10^{13}$ cm$^{-3}$ to mimic the experimental processing environment and sample properties.

Figure 6.1: TTM-simulated number density of free-carrier electrons ($N_c$), carrier-system temperature ($T_c$), and lattice temperature ($T_l$) at the location of peak intensity for incident pulse fluences of (a) 0.44 J/cm$^2$, (b) 0.37 J/cm$^2$, and (c) 0.22 J/cm$^2$. Times are relative to the arrival of the peak pulse intensity at 0 ps. In all plots, the dashed horizontal line corresponds to the Ge melting point at 1213 K [85].

Figure 6.1 compares the TTM results for three different peak fluences: (a) 0.44 J/cm$^2$ (approximately $2 \times$ the Ge ablation threshold determined in Chapter 4), (b) 0.37 J/cm$^2$, and (c) 0.22 J/cm$^2$ (near the Ge ablation threshold). For all fluence cases, the electron number density increases from an initial value of $10^{13}$ cm$^{-3}$ to the order of $10^{21}$ cm$^{-3}$ in less than one picosecond after the arrival of the peak intensity. This density is characteristic of the onset of material breakdown in semiconductors, indicating the potential onset of ablation for all three cases [92, 98]. The generation of free-carrier electrons causes the electron-system temperature to rise to 33,000 K, 29,000 K, and 16,000 K for respective fluences of 0.44 J/cm$^2$, 0.37 J/cm$^2$, and 0.22 J/cm$^2$. The higher
electron temperatures achieved in 6.1(a,b) allow stronger coupling of thermal energy to the material lattice, causing the lattice temperature to rise to above 2000 K for the higher two fluences, versus \( \sim 1400 \) K for the lowest fluence. For all investigated fluences, the predicted surface temperatures exceed the Ge melting temperature of 1213 K [85]. The TTM does not simulate phase change, so only the onset of melting is predicted.

![Figure 6.2: Dissipation of surface temperature following laser irradiation and electron/lattice thermalization.](image)

Figure 6.2 compares heat diffusion behavior on the nanosecond timescale post electron/lattice thermalization. The surface temperature induced by the 0.22 J/cm\(^2\) fluence is predicted to dissipate to below the melting point an order of magnitude faster than for 0.37 J/cm\(^2\) (4 ns vs. 40 ns). Heat diffusion controls the expansion of molten material, indicating that reducing laser fluence can minimize the time over which detrimental thermal melting may occur [44]. Therefore, a fluence near 0.22 J/cm\(^2\) is expected to induce ablation while controlling the extent of thermal effects.

Although the TTM can predict the surface temperature rise in the material lattice, it does not simulate solid/liquid phase change. Therefore, the surface temperature prediction
is treated only as an indication of thermal energy transfer to the lattice. In order for the laser to thermally melt the material surface, enough energy must be supplied after the melting point has been reached to overcome the enthalpy of fusion required for solid-liquid phase change (36.94 kJ/mol for Ge [85]). In a TTM Ge lattice voxel with dimensions of 2 m x 2 m x 5 nm, 50 pJ of energy would need to be supplied to induce full melting. The supplied energy can be calculated as $\Delta E = (T_{TTM} - T_{melt}) \cdot c_v V$, where $T_{TTM}$ is the TTM-predicted voxel temperature, $V$ is the voxel volume, $T_{melt}$ is the melting temperature, and $c_v$ is the volumetric specific heat capacity [1]. For the 0.22 J/cm$^2$ simulation in Fig. 6.1, the energy supplied to the surface voxel after the melting point is reached is just 9 pJ, less than 20% of the energy required for full melting. Hence, we predict only the onset of thermal melting at the surface, and that the melt depth is constrained to the single-digit nanometer order.

6.1.2 Repetition rate and scanning speed

To ensure that the reduced thermal impact achieved by controlling the laser fluence can be maintained during scanning-based laser polishing, the impacts of the laser fluence, repetition rate, and scan speed must be carefully balanced. To minimize the extent of heat accumulation, sufficient time must be allotted for heat diffusion to ensure that the surface can return near to its initial temperature prior to the next pulse incidence. The selected scan speed and repetition rate must control the temporal and spatial deposition of laser pulses to accommodate the material heat capacity and thermal conductivity [1]. However, various combinations of repetition rate and scan speeds can be potentially suitable for achieving low thermal impact processing, as seen in Chapter 3.

The repetition rate and scan speed of the in-house femtosecond laser ablation system can be nearly continuously tuned up to 2 MHz and 4 m/s. To avoid unbounded iterative numerical investigations, the numerical sensitivity study on Si from Chapter 3 and the
experimental study of Ge material removal / numerical sensitivity study on Ge heat accumulation from Chapter 5 were taken into consideration to narrow the range of parameters to consider for laser processing experiments. A survey of literature on laser micropolishing and ultrafast laser surface processing was also performed to investigate pulse overlaps for efficient and smooth processing. The investigated overlaps for high-quality surface processing correspond to 50-97.5% of the focal spot diameter, with improved smoothing occurring for overlaps above 75% [16, 59]. This is consistent with the pulse overlap of approximately 85% which demonstrated uniform material removal in the experimental sensitivity study in Chapter 5.

To address the numerical and experimental considerations for parameter combinations, a 250-kHz repetition rate and a 1-m/s scan speed were initially selected as parameters for line-configuration processing. This set of laser parameters adheres to the numerically-determined parameter ranges for low thermal impact processing Chapters 3-5 [1]. It also enables a pulse overlap of 93% of the 1/e² focal spot diameter of the in-house femtosecond laser processing system (∼60 μm).

Figure 6.3: Predicted surface temperature evolution for processing with potentially optimal laser polishing parameters: a 0.22-J/cm² fluence, a 250-kHz repetition rate, and a 1-m/s scan speed. The maximum surface temperature corresponds to the location of peak fluence of the immediate-past pulse. (b) Predicted base surface temperature achieved prior to the next laser pulse for processing with an extended number of laser pulses corresponding to a 500-m scan length.
The TTM was used to evaluate the capability of the identified laser parameters to control heat accumulation and the onset of thermal effects for polishing. Figure 6.3(a) shows that for each laser pulse, the surface temperature rises and then dissipates due to heat diffusion in the time between laser pulses. As more pulses are deposited, heat continues to accumulate until the thermal energy deposited by each laser pulse and the amount of heat dissipated between pulses reach equilibrium, controlled by the Ge thermal properties (specific heat capacity, thermal conductivity) [1]. Figure 6.3(b) shows that base temperature achieved following each laser pulse begins to settle after 25 pulses at a temperature of just 345 K. Between 25 and 125 pulses, the base temperature is predicted to rise by only ten Kelvin to a value of 355 K. This demonstrates the capability for the selected laser parameters to produce controlled thermal processing conditions with minimal heat accumulation while producing a pulse overlap in the regime for uniform processing. Therefore, we find the combination of a 0.22-J/cm\(^2\) laser fluence, a 250-kHz repetition rate, and a 1-m/s scan speed as a set of potential parameters for femtosecond laser-based polishing.

### 6.2 Experimental validation of predicted laser polishing parameters

Point and line processing experiments were carried out to evaluate the TTM-predicted laser parameters for femtosecond laser polishing of Ge. Experiments were performed on Ge substrates with <1 1 1> crystal orientation and ~1-nm RMS surface roughness, cleaned with isopropanol and/or methanol before irradiation.
Figure 6.4: Optical micrographs of multi-pulse point processing with up to ten pulses using different laser fluences: \(0.44 \text{ J/cm}^2\) (left) and \(0.22 \text{ J/cm}^2\) (right).
Figure 6.4 compares stationary, multi-pulse point processing using two different fluences – 0.44 J/cm$^2$ and 0.22 J/cm$^2$ – to validate the TTM investigations in Section 6.1.1. The higher laser fluence produces rough central features like melt-induced ripples or nucleated gas bubbles [72], while the near-threshold fluence results in a smooth, central processed region. To investigate the potential accumulation of rough surface features during multi-pulse laser interaction, up to ten pulses were used to irradiate the Ge surface in the same spatial location. To isolate the impact of laser fluence from repetition rate and heat accumulation effects, the incidence of laser pulses was separated by two seconds. Figure 6.4 shows that increasing the number of incident laser pulses results in an increasingly rough surface for the higher fluence, but that processed region produced by the near-threshold fluence remains smooth. This confirms the TTM-prediction that the 0.22 J/cm$^2$ fluence has the potential for high-precision, smooth, ablation-based material removal.

Line-configuration processing was carried out to examine the combined effectiveness of the TTM-investigated fluence, repetition rate, and scan speed towards polishing Ge.

Figure 6.5: Optical micrographs of line processing using a 1 m/s scan speed and a 250 kHz repetition rate for two laser fluences: (a) 0.37 J/cm$^2$, (b) 0.22 J/cm$^2$. The processed region corresponds to the 30-50 m wide bright track and the structures therein.

Figure 6.5(a) shows that using a higher fluence of 0.37 J/cm$^2$ for a fixed 250-kHz
repetition rate and 1-m/s scan speed produces rough structures in the center of the processed track. The structures have periodicity on the order of the laser wavelength resulting from interference between the incident electric field and the dense ionic plasma generated by the laser pulses [102], and are potentially exacerbated by the thermal impact of processing at higher fluences. Figure 6.5(b) shows that by reducing the laser fluence to 0.22 J/cm$^2$, the predicted optimal parameters generate a uniformly colored processing track with the potential onset of surface smoothing, evidenced by “blurring” of the scratches which passed through the processing track.

The experimental results for point and line processing confirm the model prediction that selecting a laser fluence near the ablation threshold and a repetition rate and scanning speed to minimize heat accumulation is a strategy towards achieving smooth femtosecond laser polishing.

6.3 Laser polishing of Germanium

Femtosecond laser polishing experiments were carried out using the experimentally validated set of laser parameters for smooth processing in Fig. 6.5(a). A strategy to generate overlapping lines of processing was devised to polish a region of the Ge surface ($\sim$0.5 mm $\times$ 0.5 mm). Lines were marked unidirectionally and the line overlap was initially set to 75% of the 1/e$^2$ laser focal spot diameter to maintain processing efficiency [16]. A completed scan over the defined polishing area is referred to as a “polishing pass.”

Figure 6.6 compares optical micrographs of an unprocessed (control) Ge surface and a laser-polished Ge surface generated using 20 polishing passes. The control surface (Fig. 6.6(a)) contains defects including scratches and discoloration which are not evident in the laser-polished surface (Fig. 6.6(b)). An image of the edge of the laser processed region (Fig. 6.6(c)) directly demonstrates the capability of laser polishing for removing the surface
defects, showing that the devised laser polishing strategy can effectively improve the Ge surface quality with high precision towards optical polishing.
Figure 6.6: Comparison of optical micrographs within (a) unprocessed (b) 20-pass laser-polished Ge surface regions. The circular hole features at the top of (b) are defects from the original surface, and the dark spots in the center of the image result from debris in the microscope optical train. (c) Edge between unprocessed (top-half) and laser polished (bottom-half) surface region demonstrating effective removal of defects. The height of each image is approximately 100 $\mu$m.
Figure 6.7: Surface height map of full, 20-pass laser-polished Ge surface area.

Figure 6.7(c) shows a surface height map of the 20-pass laser-polished area and the surrounding unprocessed surface. The depth of material removal in the polished area is 6 nm. The RMS roughness in the center of the laser-polished area is 0.95 nm and the RMS roughness in the surrounding area is 0.99 nm. Figure 6.8 zooms in to compare the average line profiles 200 μm × 300 μm evaluation areas for 20-pass polished and unpolished Ge both across and along the ablation line direction. The profiles demonstrate consistent peak-to-valley surface features on the order of ± 2 nm and sub-nm RMS surface roughness. The 20-pass polishing result shows that the devised laser polishing strategy effectively removes surface defects without degrading the sub-nanometer optic-quality surface roughness, revealing the capability of femtosecond laser polishing for high-precision material removal tasks.
Chapter 6. Establishing and Implementing a Methodology for Laser Polishing

6.3.1 Controllability of material removal

The controllability of material removal by femtosecond laser polishing was investigated by varying the number of polishing passes used in the scan strategy. For each polishing pass, the unidirectional laser scan direction and the experiment-specific line overlap were maintained.

Figure 6.9 shows surface height maps for processing using 15 to 100 laser polishing passes. For polishing with 50 passes, the central defect corresponds to a deep void on the Ge surface which existed prior to implementing polishing.

Figure 6.9 shows surface height maps for processing using 15 to 100 laser polishing...
Chapter 6. Establishing and Implementing a Methodology for Laser Polishing

passes. The slightly cylindrical material removal profile results from laser irradiation during scan mirror acceleration, resulting in an increased laser pulse overlap at the start and end of the line marking. This cylindrical removal profile has since been corrected by implementing a strategy to suppress the laser emission during beam acceleration. A shallow, high spatial frequency ripple can be seen in the surface profiles of the polished regions. This ripple results from the fact that each polishing pass has the same scan configuration, causing the laser to perform line-configuration processing in the same spatial locations for each pass. The ripple periodicity corresponds to the spatial separation of the lines in the devised scan configuration (∼15 µm). To mitigate this ripple, a strategy was devised to “dither” the polishing passes by slightly shifting the start-point of each incident polishing pass by selecting a random offset from a uniformly-distributed set of integers between zero and the specified line spacing.

Figure 6.10 shows that the central material removal depth increases from 4 nm to approximately 30 nm when increasing the number of polishing passes from 15 to 100. The removal depth is nearly linearly controlled by the number of passes because each polishing
pass deposits the same amount of laser energy onto the material surface. For each polishing experiment, the optic quality surface with $\sim 1$ nm RMS roughness is maintained during polishing. Hence, increasing the number of polishing passes is a strategy for controlling and scaling up material removal during polishing.

In similar fashion to the line-processing sensitivity study in Chapter 5, a relationship can be determined between the removal depth and the amount of energy deposited per unit area during polishing. This metric considers the combined impact of laser parameters and processing parameters, calculated as:

$$\frac{J}{\text{mm}^2} = \frac{\text{Repetition Rate} \cdot \text{Pulse Energy}}{\text{Scan Speed}} \cdot \frac{\# \text{ Drawn Lines} \cdot \# \text{ Polishing Passes}}{\text{Length of Processing in Overlap Direction}}$$

Figure 6.11 shows the relationship between material removal depth and total deposited energy resulting from various combinations of line overlap and number of polishing passes.

![Figure 6.11: Material removal depth versus total deposited energy varied by polishing with: (▲) 10 passes / scan-line overlap of 60 to 90% of the laser spot diameter, (♦) 100 passes / scan-line overlap of 60 to 75%, and (●) 5 to 20 passes / 75% scan-line overlap.](image)

The removal depth can be linearly controlled by changing the total energy deposited per unit area regardless of the combination of laser parameters used for processing. This
shows that total deposited energy is a metric by which laser parameter combinations can be determined to maximize the processing efficiency, accommodate larger-scale polishing tasks, and achieve dynamic control of material removal for extension to complex freeform surface geometries.

6.4 Discussion

6.4.1 Polishing mechanism

The physical mechanism for achieving smooth polishing is attributed to high precision laser ablation with controlled thermal impact. Ablation is predicted because the 0.22 J/cm$^2$ laser fluence is at the experimentally-determined ablation threshold for Ge and the TTM-predicted number density rises to $10^{21}$ cm$^{-3}$ (Fig. 1), characteristic of the onset of material breakdown in semiconductor materials [92, 103]. We expect that the onset of melting may also contribute to the smoothing mechanism since the TTM predicts that the 0.22 J/cm$^2$ fluence drives the surface temperature to slightly exceed the Ge melting point. However, only the onset of thermal melting, controlled to the nanometer order, is expected due to the small magnitude of the temperature rise / short time spent above the melting point. This high-precision melt-depth is a significant improvement over continuous-wave and micro/nanosecond-pulsed laser-based polishing strategies which generate melt / heat-affected zones with depths up to tens of micrometers [36].

Other laser-induced-breakdown phenomena could also play a role in laser polishing. For example, nonthermal melting and laser annealing are phenomena which cause lattice ordering/disordering in single-crystal semiconductors [44, 104, 105], potentially contributing to smoothing the Ge surface. These phenomena can occur once approximately ten percent of the valence band electrons have been promoted to the conduction band, signified by electron densities in the range of $10^{21}$-$10^{22}$ cm$^{-3}$ [92, 103, 104, 106], consistent with the TTM-
predicted electron densities in Section 6.1.1. However, the TTM cannot independently assess or differentiate these phenomena from ablation-based material removal.

6.4.2 Comparison to conventional ultraprecision polishing techniques

The material removal rates for the femtosecond laser polishing experiments in Fig. 6.10 are on the order of $10^{-4}$ mm$^3$/min, comparable to certain ion-beam figuring processes with small beam sizes used in final finishing of freeform optics [107,108]. Using the metric of total deposited energy, we can determine an optimal combination of laser spot size, line overlap, scan speed, and repetition rate to improve the femtosecond laser polishing material removal rate to compete with faster ultraprecision final-finishing techniques like magnetorheological finishing. However, at the current state, femtosecond laser polishing stands as a viable approach to final finishing of complex surface features owing to the controllable nature of material removal, the spatial selectivity, and the flexibility for carrying out complex tool paths using the galvanometer laser scanner.

6.5 Conclusion

In this Chapter, a strategy for polishing of Ge was established to precisely remove material while maintaining optical surface quality. The TTM of scanning femtosecond laser processing was used to successfully determine an experimentally-validated set of laser polishing parameters which produces controlled ablation and minimized thermal effects on the Ge surface. For the first time, to our knowledge, femtosecond-laser-based polishing of Ge with controllable material removal and optic surface quality with $\sim$1 nm roughness was achieved. A metric based on the total deposited energy from combined processing parameters was established to scale-up the material removal towards larger polishing tasks and non-flat surfaces. The controllable material removal with high spatial
precision, geometrical flexibility, and optic-quality surface roughness position femtosecond-laser-based polishing as an unprecedented ultraprecision non-contact polishing technique, showing promise for freeform optics fabrication applications.
Chapter 7

Conclusion

This dissertation worked to establish a novel methodology for femtosecond-laser-based polishing of optical substrates to address the need for high-precision, high-flexibility techniques for forming and finishing freeform surfaces. Ultrafast lasers were specifically investigated due to their unprecedented capability for high-precision ablation-based material removal with controlled thermal impact and flexible, space-selective interaction.

To establish a methodology for laser polishing, the laser material interaction mechanism was studied from both the fundamental theoretical and experimental standpoints. A Heat Accumulation Model was built to investigate the range of temperatures achieved during laser polishing experiments, the thermal impact of laser repetition rate, scan speed, and fluence, and laser parameters to minimize heat accumulation. A Two-Temperature Model was built to investigate laser/material interaction on the timescale of the ultrafast laser pulse so that complex absorption, free-carrier electron generation, and electron/lattice thermalization phenomena could be simulated to enable prediction of material breakdown, surface temperature rise, and the onset of phase change during processing. The model was extended to predict laser interaction with both Silicon and Germanium. It was also used to extend the Heat Accumulation Model to the general case for efficient and accurate
A flexible laser ablation system based on a 300-fs, 1030-nm Ytterbium fiber laser was established to enable tunable investigation of the impact of laser parameters on polishing, guided by the numerical investigations. Experiments were conducted to investigate the individual and combined impact of laser repetition rate, scan speed, and fluence on material removal. Tuning the laser parameters enabled controllable material removal and a metric based on total deposited energy was established to enable offline prediction of the combined impact of laser parameters to achieve specific material removal characteristics. The experimental sensitivity studies were used to guide the selection of laser parameters to potentially achieve smooth polishing. Optimal laser parameters for polishing were first investigated using the Two-Temperature Model and experimentally validated via point and line polishing tests. The predicted set of optimal laser parameters enabled smooth processing and defect removal on Germanium. A raster-based scan strategy was devised to extend polishing to an area configuration. For the first time, femtosecond laser-based polishing of Germanium was achieved. The polishing strategy enabled controllable material removal depths up to the order of 30 nm while maintaining sub-nanometer optic surface quality. The initial removal rates were found to compete with ultraprecision final finishing techniques like ion beam figuring.

The devised femtosecond laser-based polishing strategy shows promise as a tool for high-precision optical polishing tasks including forming and finishing of complex freeform optical surfaces. The demonstrated controllability, repeatability, and maintained optical surface quality shows the viability for extending the process to non-flat surface geometries, other materials including glasses, and large-scale polishing applications.
7.1 Future Work

Extension of the developed polishing techniques to non-flat surfaces and other materials will be investigated to prove the applicability of laser polishing for spherical, aspherical, and freeform surface geometries. To meet this need, experiments are currently underway to investigate the capability of femtosecond laser polishing to process tilted plano-surfaces and substrates with higher surface roughness, and to remove mid-spatial-frequency tool marks. Investigations on the impact of polarization and beam shaping for improving processing efficiency and surface roughness are also underway. The laser polishing process will be extended to spherical and freeform surfaces and to other materials such as silicon carbide and/or glass (desirable for next-generation optical elements and photonics devices). Extending to non-flat surfaces will require extensive investigation of dynamic tool paths, online laser parameter control, and in-situ process monitoring/system feedback. To extend to other materials, the polishing methodology devised in this dissertation (both numerical and experimental) can be implemented to determine optimal laser parameters for polishing.
References


References


