論文の内容の要旨

Investigations of the optical breakdown characteristics of dielectric materials induced by ultrashort laser pulses

(超短パルスレーザーによる誘電体破壊特性の探索)

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It has been over 50 years since the invention of the first laser, which opened the doors to the exploration of physical phenomena under unprecedented high intensity electric fields. The optical breakdown of dielectric materials is one such phenomenon which became observable. The process has attracted much attention from industry in recent years as an alternative to traditional machining techniques. By utilizing breakdown and ablation processes, the laser has developed as a versatile tool to realize precise, free-form materials processing.

Despite the increased application, there is still much to be uncovered regarding the basic physics of the process. The fundamental process of material breakdown involves phenomena rooted in a wide range of academic fields, from optics and condensed matter physics, to thermodynamics and mechanics. The theories involved must describe interactions evolving over multiple timescales in a non-equilibrium, non-perturbative regime. Experimental exploration is markedly difficult as well, for it requires the accurate measurement of the properties of breaking materials at ultrafast timescales. As such, despite decades of research, there is much left unknown about the physics governing the breakdown process. However, such elucidation is important for the continued development of industrial technology, and will serve to improve our understanding of physics in this extreme parameter regime.

In this dissertation, we focus on experimentally characterizing the breakdown characteristics of dielectric materials by ultrashort laser pulses. We focus mainly on morphological methods. Whereas the morphology is one of the first features studied in conventional analysis, it is also one of the most important result of ablation. The full information gained from morphology measurements has hardly been utilized in the literature. We show in this thesis how careful analysis of morphological data can shed light upon the processes involved in ablation both in the single and multiple-pulse regime.

<u>(i) Single Pulse Crater Analysis of Dielectric Materials</u>

Traditional analysis of ablation morphology have mainly extracted a single representative value from a three-dimensional profile, such as the diameter of an ablation region, or the depth of the crater developed. However, far more information is contained within a single crater, where a full two-dimensional space of parameters is available for analysis.

In order to fully utilize this data, we develop the *fluence mapping* technique. It involves spatially correlating the local beam fluence (pulse energy/unit area) profile with the local ablated volume. To do this, we use a home-made small-pixel beam profiler, and correlate this data to the morphology gained from laser scanning microscopy, as shown in Figure 1(a). We can then create a histogram of the local crater depths as a function of the local fluence (Figure 1(b)). This analysis presents several advantages to traditional measurements, such as being able to accurately analyze data gained from notably non-Gaussian pulses, as well as the efficient extraction of threshold fluences. Fundamentally, it allows for the evaluation of the *locality* of the ablation phenomena.

As an application where we can fully utilize the robustness of this method to systematic errors induced by non-ideal spatial profiles, we measure the ablation due to

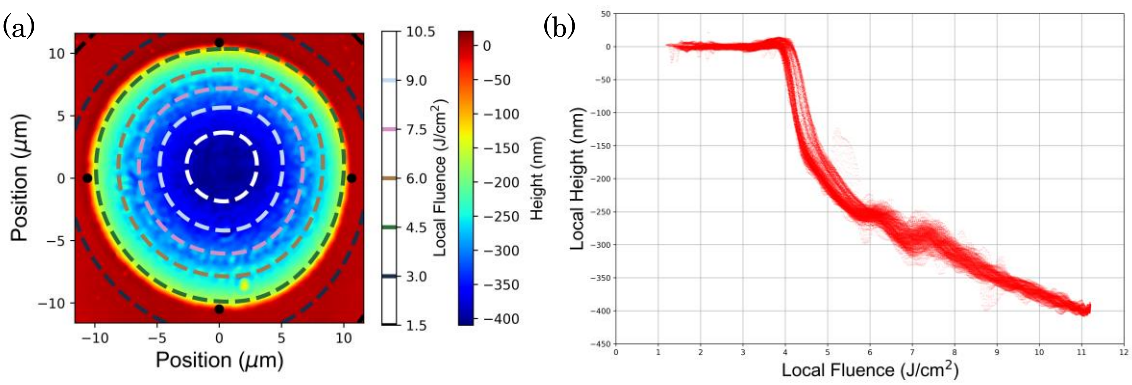

Figure 1: (a) The beam profile (contour line) is spatially correlated with the crater profile. (b) A histogram of the data is created, allowing us to correlate the relationship between the local fluence and the local ablated profile.

a high-power optical parametric amplifier, a light source where the beam spatial profile is known to systematically vary between wavelengths. As representative samples, we measure the wavelength dependent ablation thresholds of two materials: TiO_2 (rutile) and Al_2O_3 (sapphire). We focus on solving a fundamental open question on ablation: whether the multiphoton nature of the initial excitation is imprinted on the final ablation thresholds. For both materials, and for the first time in the literature for a single pulse, we succeed in observing a step-like behavior in the threshold due to changes in the lowest-order allowed multiphoton transition. We analyze this data and discuss the implication they have on the traditional rate-equation approach to modeling the electronic excitation process in ablation.

<u>(ii) Exploration of Multiple-Pulse Ablation Characteristics</u>

While uncovering the fundamental processes occurring during single-pulse ablation is important from a fundamental perspective, there is a large gap between this situation and the actual processing conditions seen in applications. In general, a multitude of pulses are irradiated onto an evolving material surface. In order to address this gap, in the latter half of the dissertation, we focus on two intrinsically multiple-pulse features in ablation which must be qualified in order to properly model more realistic situations.

In the first half of this part, we address the problem of damage incubation, where the formation of defects within the material alters the light-absorption characteristics for later pulses. This effect can cause macroscopic changes in the way that material is ablated from the surface, as seen in Figure 2(a). This can be seen prominently in the laser grooving of sapphire, where pulses are scanned along the material surface. We

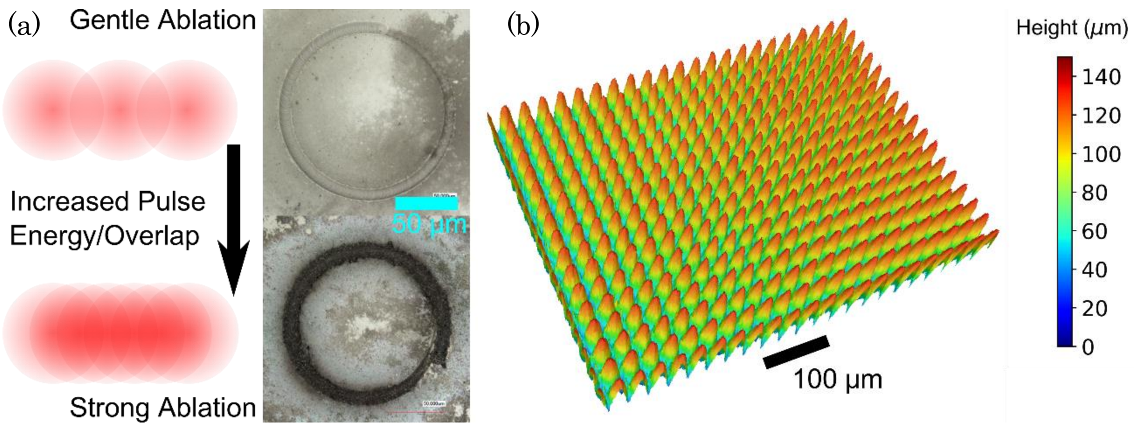

Figure 2: (a) The surface of sapphire at different pulse overlaps, where the damage incubation effect can be seen to cause large qualitative differences in the morphology. (b) Fabricated moth-eye structures utilizing the saturating property of ablated grooves

create an empirical model for the light absorption in this scenario, and derive simple scaling relationships to predict the material morphology for varying pulse energies and spacing.

In the second half, we address the effect of the changing surface angles relative to the incident pulse. The angling behavior seen in grooving results in an eventual saturation of depth, due to a decreasing projected fluence and optical transmissivity. We then show how this saturation trend can be utilized to create uniform needle-like protrusions on a sample surface. We use this to create moth-eye anti-reflection structures on the surface of silicon, shown in Figure 2(b). We demonstrate its performance with standard terahertz time-domain spectroscopy measurements.