Ultrafast double-pulse ablation of fused silica

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Ultrafast pump-probe experiments were used to study high-intensity ultrafast pulse-ablation dynamics in fused silica. Two laser pulses with varied time delay and pulse energy were used to irradiate fused silica samples and observe the transient reflectivity and transmissivity of the probe pulse. It was seen that the probe reflectivity initially increased due to the formation of free-electron plasma and then dropped to a low value within a period of about 10 ps caused by a rapid structural change at the surface. The time-resolved measurements of reflectivity and transmissivity were also related to atomic force microscopy measurements of the depth of the laser-ablated hole. It was seen that the depth peaked at zero delay between the pulses and decreased within a period of about 1 ps as the temporal separation between the pulses was increased caused by the screening by the plasma produced by the first pulse. When the temporal separation is about 100 ps or longer, evidence for melting and resolidification during double-pulse ablation was also observed in the form of ridges at the circumference of the ablated holes. © *2005 American Institute of Physics*.
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In recent years, the use of ultrafast lasers for machining dielectrics has attracted much interest. This is mainly due to the unique nature of femtosecond pulses in that they possess very high peak intensities, which cause nonlinear absorption of the photons even in wide band-gap dielectrics.[1,2] The nonlinear absorption of photons leads to the creation of highly excited free-electron plasma, which has been observed in time-resolved reflectivity[3] and shadowgraph imaging[4] experiments. The dynamics of this free-electron plasma has been studied by several groups. Relating the plasma dynamics to the transient material removal process in dielectric materials remains an area to investigate.

In this letter, we perform femtosecond pump-probe experiments at intensities sufficient for ablation. The goal is to relate the plasma dynamics in double-pulse machining to the transient material removal process and the ablated feature size by varying the pulse separation time from zero to hundreds of picoseconds. In the double-pulse ablation of crystalline silicon,[5] it was shown that some enhancement in ablated volume could be achieved if the two pulses were separated by a period of 10 ps. Using ultrafast pulse trains to machine different materials has also received some attention.[6,7] It has been shown that double- and triple-pulse sequences synthesized using a pulse shaper lowered the optical breakdown threshold of dielectrics, and some degree of enhancement in machining quality could be achieved.[8]

Our experimental setup incorporated a typical pump-probe geometry with orthogonally polarized, collinear pump and probe pulses incident normally upon 1-mm-thick fused silica samples (Corning 7980). The samples were cleaned with methanol and acetone prior to ablation and all the experiments were carried out in air at atmospheric pressure. The ultrafast pulses used in the experiment are produced by a Spectra-Physics Spitfire regenerative amplifier system and have a pulsewidth of 90 fs [full width at half maximum (FWHM)] and a maximum energy of about 1 mJ. The delay between the pulses can be adjusted with the precision of a few femtoseconds, and the energy of both pump and probe pulses is varied using half-wave-plate–polarizer combinations. Temporal overlap between the pump and the probe was achieved by measuring the two-photon absorption signal in a GaP photodiode. A 0.28 NA long working distance Mitutoyo objective was used to focus the pulses to a 4-μm-diam. spot size on the sample surface. Another 0.5 NA collecting objective was used to gather the transmitted light, which was then measured with a fast silicon photodiode. The normally reflected light was collected by the focusing objective and sent back to another silicon photodiode through a beam splitter to measure the reflectivity. The experiments were all carried out in single-shot mode so that each laser pulse hit a fresh spot on the sample. An electronic shutter (Uniblitz LS6T2) was placed in the input beam to admit a single pulse from the 0.5-kHz output train of the amplifier. An imaging system consisting of a charge-coupled device (CCD) camera and a white-light source were used along with the focusing objective to image the surface of the sample to ensure that all data were collected under similar focusing conditions. Appropriate polarizers and 800-nm bandpass filters were placed in front of the photodiodes to block the pump beam and plasma light.

Time-resolved measurements of reflectivity and transmissivity were carried out and the results are shown in Fig. 1. A weak, vertically polarized probe at an intensity of 1.1×10^{13} W/cm^2 and strong, horizontally polarized pump pulses at four intensities varying from 2.2×10^{13} W/cm^2 to 13.4×10^{13} W/cm^2 were considered. Optical damage was observed to begin at the pump intensity of 5.5×10^{13} W/cm^2. The values plotted are normalized with respect to the initial value at time $t=-1$ ps. Negative delays correspond to the cases where the probe pulse arrives before the pump and is incident on an unperturbed sample. Each point in the plot corresponds to an average over five data

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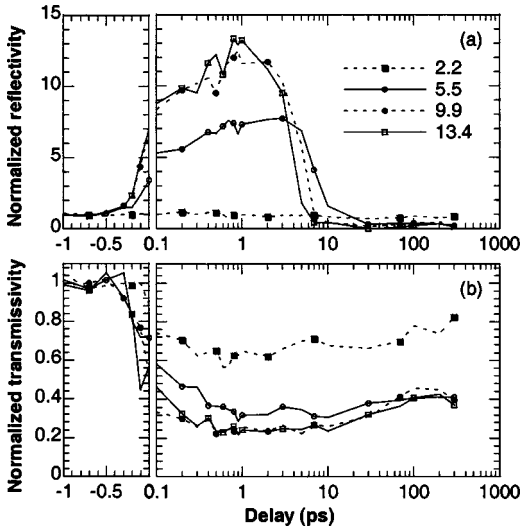

FIG. 1. Time-resolved (a) reflectivity and (b) transmissivity of a 1.1 $\times 10^{13}$ W/cm^2 probe pulse at four different values of pump intensity ($\times 10^{13}$ W/cm^2).

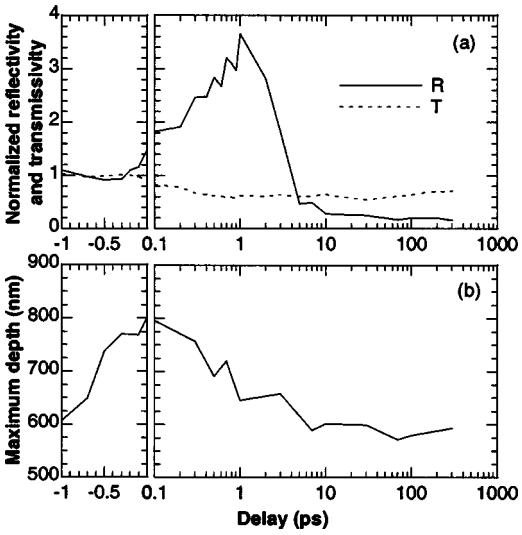

FIG. 2. Time-resolved measurements of (a) reflectivity and transmissivity, and (b) corresponding maximum hole depths in fused silica for equal pump and probe pulse intensities of 1.3×10^{14} W/cm^2.

points with the standard deviation being 10–15 %. For clarity, the error bars corresponding to the standard deviation are not shown. From Fig. 1, it is seen that there is a sharp drop in transmissivity that begins around zero delay, followed by a slow increase over a time period on the order of 10–100 ps. For those laser intensities causing ablation, the reflectivity shows a rapid rise and stays constant for about 2–3 ps and then drops sharply.

The rise in reflectivity and the fall in transmissivity can be readily explained by the creation of free-electron plasma whose density increases with increasing pump intensity. For the case of the weakest pump pulse at 2.2×10^{13} W/cm^2, there is a drop in the transmissivity of the probe due to absorption in the weak plasma created by the pump. This drop lasts longer than 100 ps, which corresponds to the lifetime of the plasma. However, there is no noticeable reflectivity increase because the free-electron density generated by the pump pulse with the intensity of 2.2×10^{13} W/cm^2 is still well below the critical density.[9] For higher intensities, the reflectivity first increases and then decays within a few picoseconds, reaching a value much below the initial reflectivity of the undisturbed sample, which is caused by structural damage at the sample surface. It is also seen from Fig. 1(a) that the reflectivity decay begins slightly earlier as the pump intensity is increased, indicating that damage occurs faster at higher intensities. Time-resolved measurements of the scattering of a probe pulse from fused silica[10] have shown that the scattering signal rises after a delay of about 3 ps, which matches with the onset of decay in our reflectivity signal.

Figure 2 shows the results for double-pulse ablation, where the orthogonally polarized pump and probe pulses have equal intensities of 1.3×10^{14} W/cm^2, and the delay between the two pulses varies from −1 to 300 ps. The corresponding values for the maximum depth of the ablated hole measured with an atomic force microscope (AFM) are shown in Fig. 2(b). Each data point is averaged over five measurements, with a standard deviation of about 5–10 %. The ablation depth is the highest when the two laser pulses overlap, and is roughly the same as that of a single pulse with double the intensity. The ablation depth decreases when the separation between the two pulses is increased, which is related to the interaction between the second pulse and the plasma formed by the first pulse. The formation of the plasma by the first pulse has two effects on the second pulse: it increases the absorption of the second pulse, which may lead to stronger ablation, but on the other hand, its high reflectivity blocks the second pulse from entering the target that leads to less ablation. From the experimental results, it is seen that the latter plays a bigger role as the ablation depth decreases with the increase of the delay time between the two pulses. When the delay time is further increased to longer than a few picoseconds, the reflection of the second pulse by the plasma formed by the first pulse decreases due to scattering. This scattering also reduces the coupling of the second pulse into the target, resulting in less ablation.

It is also seen from Fig. 2(a) that the variation in transmissivity and reflectivity for the case of two strong pulses is less than what was shown in Fig. 1 when a weak probe pulse was used. The smaller peak reflectivity in Fig. 2(a) is due to the fact that, as the probe intensity is increased, the free-electron plasma scatters the probe pulse more strongly, whereas the measurement apparatus only captures the specularly reflected light. On the other hand, the apparent difference in the minimum transmissivity is merely an artifact that the signals are being normalized with respect to the initial transmissivity at negative delays. The initial transmissivity value decreases with increasing intensity since, as the laser intensity increases, more laser energy is absorbed by the free electrons generated by the pulse itself. The decrease of the initial transmissivity value causes a relative upward shift of the transmissivity curve.

AFM images of the double-pulse machining shown in Fig. 3 reveal an interesting phenomenon. At time delays of the order of 100 fs or less, the ablated hole is just a clean crater as seen in Fig. 3(a). As the delay between the pulses is increased, there is no significant difference except a decrease in the ablated volume as expected from the trend in the hole-depth measurements shown in Fig. 2(b). This is the case at the 10-ps delay shown in Fig. 3(b). As the delay further increases, a ridge appears around the ablated hole. This is clearly seen in the AFM pictures in Figs. 3(c) and 3(d) at delays of 100 and 300 ps, respectively. The cross-sectional

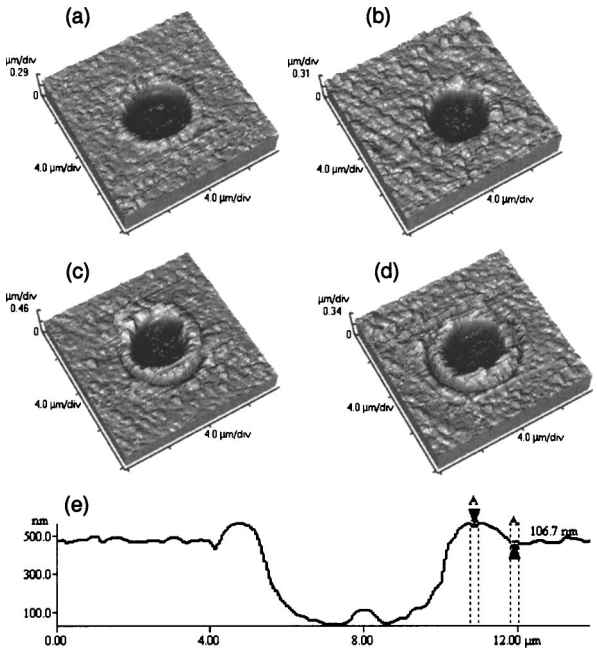

FIG. 3. AFM images of holes ablated with two pulses at 5.5×10^{14} W/cm^2 separated by (a) 100 fs, (b) 10 ps, (c) 100 ps, (d) 300 ps; (e) depth profile of the hole shown in (c).

profile of the hole in Fig. 3(c) is shown in Fig. 3(e). It is seen that the height of the ridge is of the order of 100 nm. For these longer delay cases, the first pulse causes the fused silica to melt and the second pulse is incident upon this molten material, causing it to flow outward and finally resolidify in the ringlike pattern seen in the figure. Although melting could occur much earlier than 100 ps, the melt depth could be too small to form a clearly visible ridge if the second pulse follows the first pulse closely (e.g., a few picoseconds). Figure 3(d), which shows the crater caused by two pulses separated by 300 ps, indicates that the melt duration can be longer than 300 ps. Experiments with longer delays were not carried out due to the limitation of the length of the delay stage. Similar thermal-fluid mechanisms of material modification and melting duration of hundreds of picoseconds have been observed in experiments using 10-ps pulses.[11]

In summary, the pump-probe experiments revealed the plasma formation in fused silica during ultrafast laser irradiation. It was found that the creation of the free-electron plasma by the first pulse and the delay between the first and the second pulses determined the coupling between the second pulse and the fused silica sample, as well as the results of double-pulse ablation. The total ablated volume decreased with the increase of the delay time due to the screening by the plasma formed by the first pulse. AFM imaging revealed the presence of a molten phase during the ablation process, which can last for several hundreds of picoseconds. The understanding of the fundamental energy absorption mechanism and relevant time scales could help in the design of better pulse-train separations to optimize the machining process.

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