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Transition of Femtosecond Laser Ablation Mechanism for Sodalime Glass Caused by Photoinduced Defects

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Femtosecond laser ablation mechanism was systematically investigated on sodalime glass in ambient conditions. The ablation crater diameter was measured for varying numbers of laser pulses as for varying well as the laser fluence. The analysis of the results with a one dimensional spatial Gaussian fluence distribution reveals that the inherent ablation mechanism has been altered from a multi-photon process to a single photon excitation due to defect sites that have been accumulated by successive laser pulses. Furthermore, the transition between the two regimes was found to be a function of both the laser fluence and the number of laser shots.

OCIS codes : 120.0120, 220.0220, 320.7130, 140.3330, 220.4000.

I. INTRODUCTION

The progress in ultrafast laser systems in generation and amplification with good reliability in output stability draws much attention to many areas relating to microfabrication processes [1–3]. The applications include micro structuring of three-dimensional features, micro-scale components, micro cutting and linking for the repair of photomasks for integrated circuits, and even a refractive surgery of the human eye [1–7]. The pulse duration with $\sim$ 100 fs is much shorter than the vibronic relaxation time-constant ($\tau \sim$ ps) for an ordinary substrate. Because the vibronic relaxation to the lattice phonon is the major relaxation route of the electronic energy deposited upon photoexcitation, by using the ultrafast laser pulse one can noticeably shorten the diameter of the heat-affected zone compared to the long pulsed laser micro processing [8–10]. Applying near infrared (NIR) ultrafast laser pulses rather than UV-vis long laser pulses to the material can deposite the laser energy into smaller volumes by multiphoton nonlinear optical absorption [11]. These advantages of the ultrafast laser micromachining over traditional laser fabrication method allow us to fabricate microstructures in the glass substrates with a resolution high enough to micromachine a valuable microstructure by using inexpensive commercially available sodalime glass.

The absorption of radiation from a rather long-pulsed laser leads to melting and then sputtering the material, which may not only contaminate the surrounding area but also produce micro-cracks to an extended area much larger than the spot size. Other adverse effects are damage to adjacent structures, delamination, formation of recast material, and poor shot-to-shot reproducibility. If laser energy is deposited at a time scale much shorter than both the heat transport and the electron-phonon coupling, the light-matter interaction process is essentially frozen in time [12,13]. The affected zone altered from solid to vapor phase and to plasma formation almost instantaneously.

Venkatakrishnan et al. [10] reported that holes with a diameter less than 200 nm could be drilled upon focusing the ultrafast laser with 500 fs pulse duration to a spot size of 1,700 nm on metallic thin film. Very recently, nanopatterning on the gold thin films with sub-10 nm resolution [14] was achieved using an AFM tip made of silicon through a local field enhancement effect. The reported observation of higher spatial resolution compared to laser spot size has been frequently explained in terms of the existence of a sharp ablation threshold fluence for either one- or multi-photon excitation as well as a negligible thermal diffusion length compared to the optical penetration depth [15]. Therefore, it is important to develop a precision measurement method for the ablation threshold fluence for interesting materials in order to micro- and nanomachine with a high resolution [16]. Furthermore, it is prerequisite to protect eye and skin from laser damage with an increasing spread of this ultrafast laser in industrial fields [17]. Very recently, Kruger et al. [18] reported that the threshold fluence of protection filters (Schott BG18 and BG36) to the ultrafast laser pulse at 800 nm decreases with decreasing pulse duration and increasing the number of pulses.

Concerning the ablation process with ultrafast lasers, the existence of two different ablation regimes has been reported in the ablation process of metal [19,20] as well as dielectrics [21–23]. Nolte et al. [19] investigated the ablation depth per pulse on copper substrate as a function of incident laser fluence with 150 fs laser pulses. Based on the one-dimensional two-temperature diffusion model, the two semi-logarithmic dependences of the ablated depth on the laser fluence are explained in terms of the optical penetration depth and the electronic heat conduction. At lower fluence, the ablation depth per pulse, L, should be described by the expression $L = \alpha^{-1} \ln(F/ F^{\alpha}_{th})$ [8,24,25] where α^{-1} is the optical penetration depth and F^{α}_{th} is the threshold laser fluence when the optical penetration depth is much larger than the heat penetration depth. At higher laser fluence, the different logarithmic dependence $L = l \ln(F/ F^{l}_{th})$, where l and F^{l}_{th} are the heat penetration depth and higher-threshold value, respectively.

In this work, we report the results of a femtosecond laser ablation process on sodalime glass in ambient conditions. In order to have deeper understanding on the effects of photo excitation with multiple pulses on the ablation mechanism of the dielectrics, we have investigated the dependence of the crater diameter on the laser fluence and the number of pulses.

II. EXPERIMENTAL

The detailed description of the *fs* Ti: sapphire laser system employed in this work was given elsewhere [26]. The femtosecond laser was generated by an amplified high power Ti: sapphire laser output (810 nm, 150 fs at 1 kHz). This fundamental output was used for the current micromachining experiments without any spatial filter and refining optics. The final output energy of 400 μJ/pulse is kept to be constant unless specified. The sample, 76 mm × 26 mm slide glass substrate with 1 mm thickness, was purchased from Knittel Glaser (Germany) and mounted on a precision 3-axis translation stage without any further cleaning. We have found no prominent improvements on the micromachining due to additional cleaning with ordinary solvent.

Fig. 1 shows the schematic diagram of the experimental setup. The fundamental output from the femtosecond laser was delivered to the galvanometer scanner (Scanlab AG, Germany), which is digitally controlled by an interface board (Scanlab AG, Germany)

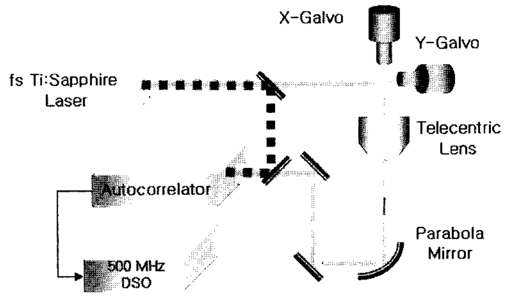

FIG. 1. Schematic diagram of the experimental set up for femtosecond laser ablation with an autocorrelation measurement.

and IBM compatible PC. An optical shutter with time constant less than 1 msec, which was synchronized to the scanner, controls the laser exposure. The variable neutral density filter (Sigma Koich, Japan) is used for controlling the laser power. Just before the input port of the scanner, the diameter of laser beam was fixed to be 4 mm. A telecentric lens (Sill Optics, Germany), of which focal length is 85 mm, was employed to focus the laser beam on the substrate surface. All the surfaces of the lens are anti-reflection coated at the wavelength of 810 nm. The diffraction limited spot size at focus is given by

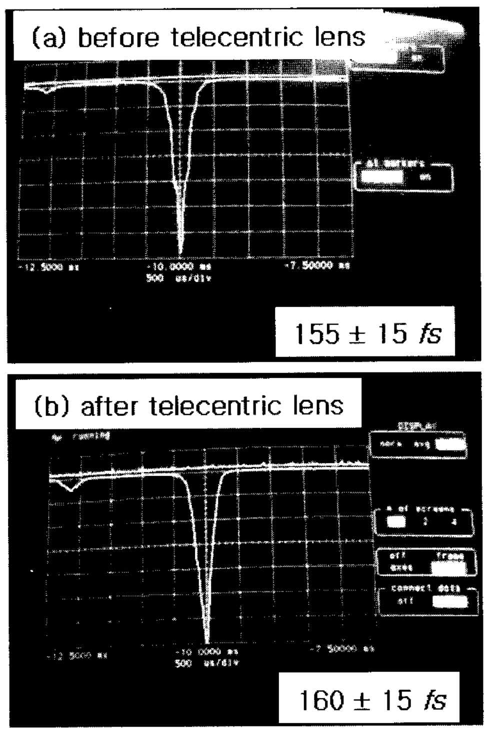

FIG. 2. Photograph of the temporal profiles, measured by 500 MHz digital storage oscilloscopes, of an autocorrelation trace of the laser pulse before (a) and after (b) being focused by telecentric lens.

$$D_{min} = 4F\lambda/\pi D \quad (1)$$

where F is focal length, λ is the wavelength and D is the incident beam diameter. The current optics used in this work results in the laser spot size of 21.6 μm in diameter (by calculation). In general, the actual spot size is bigger than the calculated value due to misalignment and deficiencies of focusing optics.

Determining whether the pulse width on the glass substrate altered due to the presence of rather thick focusing optics is crucial to investigating the interaction between an ultrafast pulse and matter. Fig. 1 shows the schematic diagram for a measurement setup of an autocorrelation trace of the laser pulse. The pulse width of 160 fs observed after the thick optics was almost equal to that of the fundamental output of the current laser system, of which pulse width was found to be about 150 fs (Fig. 2). With altering the beam position on the telecentric lens, no apparent change in pulse width was found.

III. RESULTS AND DISCUSSION

Fig. 3 shows the typical images taken by an optical microscope with varying the laser fluence from 4.8 to 51 J/cm^2. The beam spot size was kept to be 26 μm in diameter. The laser beam was scanned with four different speeds of 2, 4, 8, and 13 mm/s. The averaged shot numbers for each scanning speed could be estimated to be 13, 6.5, 3.25, and 2, respectively. For lower laser fluence, a circular spot for each laser pulse can be clearly observed on the glass substrate under higher scanning speeds of 8 and 13 mm/s. Meanwhile, the circular pattern gradually disappears with decreasing scanning speed and increasing laser fluence. Further increase in the laser fluence to more than 12 J/cm^2 results in abrupt changes in the morphology of the lines micro-machined with the scanning speed of 2 mm/s. With the aid of optical microscopy, the crater-like morphology was observed around the edges of the patterned line. Also, the image of the micromachined area shows numbers of micro-sized bumps in the center of the line in addition to the evidence of photo induced darkening.

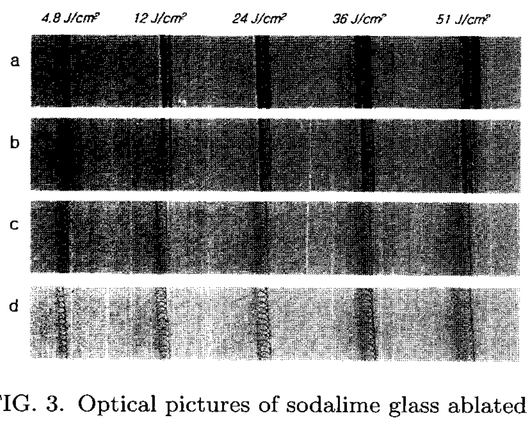

FIG. 3. Optical pictures of sodalime glass ablated with four different scanning speeds of 2 (a), 4 (b), 8 (c), and 13 mm/sec (d). In each scanning speed, the laser fluence was changed from 4.8 to 51 J/cm^2.

Many different methods to determine the ablation threshold fluence have been reported for the ultrafast laser micromachining of dielectrics [19,27–29]. Applying the laser pulses with one dimensional Gaussian beam profile, a relation between crater diameter, D, and maximum laser fluence, F_0, can be derived according to

$$D^2 = 2\omega^2 \ln(F_0/F_{th}) \quad (2)$$

where F_{th} and ω denotes the ablation threshold fluence and the $1/e^2$ Gaussian beam radius, respectively. In our experimental configuration, the width of the ablated lines could be supposed as the crater diameter.

An optical microscope is used to measure the crater diameter at five different positions of the ablated zone. Fig. 4 shows the squared crater diameter, D^2, measured with varying laser fluence at the four different scanning speeds. Two different semi-logarithmic dependences can be clearly identified. With lower laser fluence, the slope and the intercept for all the curves are invariant with changing the scanning speed. A fit of the semi-logarithmic plot of D^2 versus F_0 to the experimental data gives ω =12 μm and F_{th}= 2.3 J/cm^2.

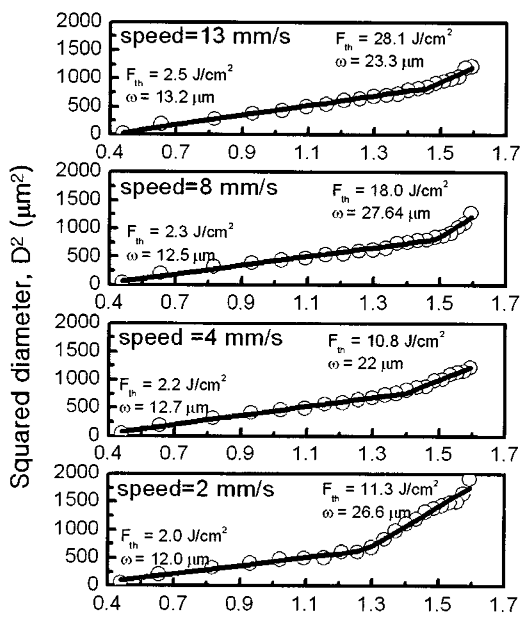

FIG. 4. Dependence of squared crater diameter, D^2 on laser fluence. Laser scanning speeds are 2, 4, 8, and 13 mm/sec (d), respectively. Open circle indicates experimental observation and solid line is the fitted one by the linear regression with Eq. (2) in text.

This value is almost identical to the reported ones for barium aluminum borosilicate glass [29] and color glass filters like Schott BG18 and BG36 [10].

With increasing the laser fluence, a second semi-logarithmic dependence of D^2 on the laser fluence appears. This regime is characterized by a larger ablated zone and higher threshold fluence. The fit of a semi-logarithmic plot of D^2 versus F_0 to the experimental data gives ω =26 μm, which is almost constant with changing the scanning speed. Meanwhile, the threshold laser fluence decreases with increasing the number of shots resulting from lowering the scanning speed. The crater diameter is strongly dependent on the number of shots in this regime. Very recently, Kruger et al. [18] also reported that the dependence of crater diameters on the laser fluence observed for two different color glass filters deviate from the linear regression based on Eq. (2) at higher fluence. They have explained the deviation in terms of the variation in the spatial beam profile from an ideal Gaussian one. If this is the case, the crater diameter should be constant regardless of the variation of the number of laser pulses. As mentioned before, however, the value of the crater diameter is dependent on the number of shots, which is a function of the scanning speed.

Meanwhile, the observation of two different ablation regimes could be explained in terms of the transition of either the optical properties of sodalime glass or transient substrate temperature due to multi-pulse excitation with rather high repetition rate of 1 kHz on the same zone of the substrate. If the latter plays a major role in the ablation process under the current experimental condition, the changes of scanning procedure should alter the crater diameter as well as the morphology of the ablated zone even though one keeps the total number of shots constant. With keeping the laser fluence constant, the crater diameter was measured with a simultaneous variation of the scanning speed from 1 mm/sec to 6 mm/sec and the number of scans from 6 to 1. The total number of the laser pulses for each experiment could be estimated by multiplying the number of scans and averaged numbers of shots per scan. Fig. 5 shows the dependence of the crater diameters on the total number of shots obtained with the laser fluence of 3, 5, 10 J/cm^2. Because the spot size was about 31 μm in diameter, the total number of shots was changed from 5 to 31. With higher total number of shots, we observed the crater-like morphology, micro-sized bumps, and photo induced darkening around the microablated region. (not shown) Fig. 5 reveals that the crater diameter is only dependent on the total number of laser pulses. This observation led us to conclude that the temperature rising effect due to a 1 kHz laser excitation is not the main cause of the transition between the two different ablation regimes.

The sodalime glass has an absorption band edge at

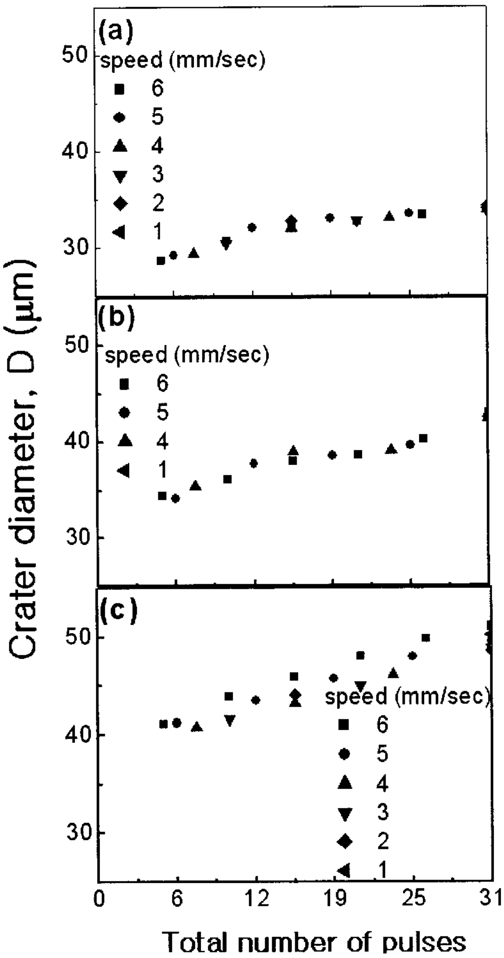

FIG. 5. Dependence of crater diameter on the total number of shots. The laser fluences are 3 (a), 5 (b), and 10 J/cm^2 (c), respectively. The spot size of 31 μm was kept to be constant.

the wavelength of 350 nm. With NIR laser (λ = 810 nm), three photons of the fundamental output need to cross the band gap of the glass. With lower laser fluence and lower total number of shots, the multiphoton process is the major route to deposit the laser energy to the substrate. The energy should result in an electron-hole plasma formed by multiphoton seeded avalanche multiplication of electrons, which is localized transversely as well as longitudinally. Therefore, the crater diameter in this regime might be related to the beam profile governed by the multiphoton absorption process. With increasing both the laser fluence and the total number of shots, however, the accumulation of photoinduced defect sites should alter the optical properties. The increase of the number of defects, due to the increase of either laser fluence or the total number of shots, has to be considered as an additional channel for photon absorption. In fact, the optical image shown in Fig. 3 exhibits apparent photodarkening around the ablated zone with higher laser fluence. The effect of the one-photon absorption at 810 nm due to the photoinduced defects is not negligible at a certain level of the defect density. The laser energy, which is absorbed by one photon excitation due to the defects, should extend the ablation zone with increasing either the laser fluence or the number of laser pulses. And then, this change eventually alters the slope as well as the intercepts of the semi-logarithmic plot of the squared diameter versus laser fluence.

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