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Near-field enhanced femtosecond laser nano-drilling of glass substrate

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Abstract

Particle mask assisted near-field enhanced femtosecond laser nano-drilling of transparent glass substrate was demonstrated in this paper. A particle mask was fabricated by self-assembly of spherical 1 μm silica particles on the substrate surface. Then the samples were exposed to femtosecond laser (800 nm, 100 fs) and characterized by field emission scanning electron microscope (FESEM) and atomic force microscope (AFM). The nano-hole array was found on the glass surface. The hole sizes were measured from 200 to 300 nm with an average depth of 150 nm and increased with laser fluence. Non-linear triple-photon absorption and near-field enhancement were the main mechanisms of the nano-feature formation. Calculations based on Mie theory shows an agreement with experiment results. More debris, however, was found at high laser fluence. This can be attributed to the explosion of silica particles because the focusing point is inside the 1 μm particle. The simulation predicts that the focusing point will move outside the particle if the particle size increases. The experiment performed under 6.84 μm silica particles verified that no debris was formed. And for all the samples, no cracks were found on the substrate surface because of ultra-short pulse width of femtosecond laser. This method has potential applications in nano-patterning of transparent glass substrate for nano-structure device fabrication.

Keywords: Glass substrate; Nano-drilling; Multi-photon absorption; Femtosecond laser; Near-field effect

1. Introduction

Transparent glass substrate is the most common commercial glass with low cost. It possesses a large variety of applications in optic devices and optical communications when doped with other elements or compounds. For example, rare-earth doped soda-lime glass can be used in medical diagnostics, laser glasses, undersea optical communication, integrated-optical devices and optical data storage [1,2] where periodical nano-structures on glass substrate are necessary to obtain required optical properties. It is, however, a high challenge for glass processing to get nano-structures due to its hard, brittle, non-conductive and other inert properties. Recently, there is a growing interest in developing nano-processing to fabricate nanoscale structures on glass substrate. It leads to many investigations on the sophisticated fabrication methods, such as nanoimprinting [3], electron beam lithography [4], and chemical etching. Femtosecond laser nano-machining is an attractive approach in the glass engineering with the advantages of elimination of collateral damage in dielectrics, reduction of heat-affected zones, and the ability to create sub-diffraction limited target regions [5]. Femtosecond laser can also provide an extremely high peak power intensity for non-linear multi-photon absorption. Furthermore, non-linear absorption of femtosecond laser irradiation can result in an increase in the refractive index at the focal point inside the glass, which is potentially effective to produce structures in nano-scale feature size [6]. As simultaneous absorption of n photons is proportional to the nth power of laser fluence, the changes in optical properties can be confined inside a tiny region with its sub-micron dimensions. In this paper, nano-crater formation by femtosecond laser irradiation was studied. By depositing a monolayer of microspheres on the substrate surface as a particle mask and utilizing femtosecond laser as light source, nanoscale holes on the glass surface can be created [7]. It is proposed that non-linear absorption and optical near-field enhancement play the important roles in the nano-crater formation on the glass substrate.

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2. Experimental

Soda-lime glass substrate (1.1 mm in thickness) was cleaned with acetone in an ultrasonic bath for 5 min followed by rinsing in DI water and then dried by N_2 gas (purity 99.999%). The spherical silica particles (Duke, $n = 1.6$, 10% size deviation) with the diameter of 1 μm were applied on the glass substrate after the suspension had been diluted with DI water. The substrate was kept still until all the water was evaporated. As a result, a silica particle monolayer array was formed on the substrate surface. A Ti:Sapphire (Spectra Physics Tsunami, Mode 3960) and a regenerative amplifier (Spectra Physics Spitfire, Mode 9769A) were used as the light source (central wavelength $\lambda = 800$ nm, pulse duration $\tau = 100$ fs and repetition rate from 1 to 1000 Hz). The laser was focused by a 20× microscope objective lens mounted on a Z stage. The laser spot size irradiated on the surface is about 5.4 μm. The laser fluence is in the range from 17.5 to 61.3 J/cm^2. The laser was incident normally on the substrate surface. After the exposure, the sample was characterized by AFM (Digital Instruments D3000) and FESEM (Hitachi S-4100). For SEM imaging, samples were coated with a thin Au film by a thermal evaporator before the observation.

3. Results and discussion

Fig. 1a shows a typical nano-hole array created on the glass substrate after a femtosecond laser shot at a laser fluence of 35 J/cm^2. As can be seen, the silica particles on the glass surface were removed after the laser irradiation. Meanwhile, debris were observed around the nano-crater. They are hard to remove by conventional ultrasonic cleaning method due to strong adhesion. The cross section profile measurement by AFM is shown in Fig. 1b. The full width at half maximum (FWHM) is about 250 nm and average depth of nano-features is around 150 nm. The size of melting zone ranges from 380 to 650 nm.

In order to explain these results, the interaction between ultrashort laser and transparent glass materials has to be analyzed. Experiments have shown that the nature of the ablation process depends upon the transfer of energy from the laser pulse to the system of electrons on target, then from the electrons to the lattices of the bulk materials. For transparent media, laser ablation is proceeded by the production of a dense laser induced plasma [8,9], which transforms the transparent media into an absorbing media. However, multi-photon absorption often takes place for the ultrashort laser pulse. The n-photon absorption is proportional to the nth power of the intensity [10]. Considering the electronic bandgap of the glass (5 eV) and the laser photon energy (1.55 eV), it is a triple-photon absorption process. This effect significantly narrows the processed area. As a result, with femtosecond laser, the processing area can be confined at the central part of optical axis [11]. Furthermore, femtosecond laser-induced non-linear effect gives rise to the change of glass optical properties which affect the propagation of incident light [6,12]. Then laser beam was self-focused in a much smaller area, which leads to the advantages of smaller spot size and extremely high laser fluence to form nanoscale feature size on the glass surface.

Besides these effects, the near-field enhancement due to light scattering by the small spherical transparent particle plays another important role. The distribution of laser energy around spherical particles can be calculated from Mie [13]. In Fig. 2, the energy distribution, $I = S_z$, within the incident plane (xz-plane) is shown for silica particle (refraction index $n = 1.6$, particle diameter $2a = 1$ μm) on glass surface under 800 nm laser irradiation. At the interface between particle and glass surface, laser energy is localized within a small region, both inside and outside the particle. The focused spot size was about 300 nm. This theoretical value is in a good agreement with experimental measurement. It should be noted that the silica particles in our experiments have a size deviation of 10%, which corresponds to a variation

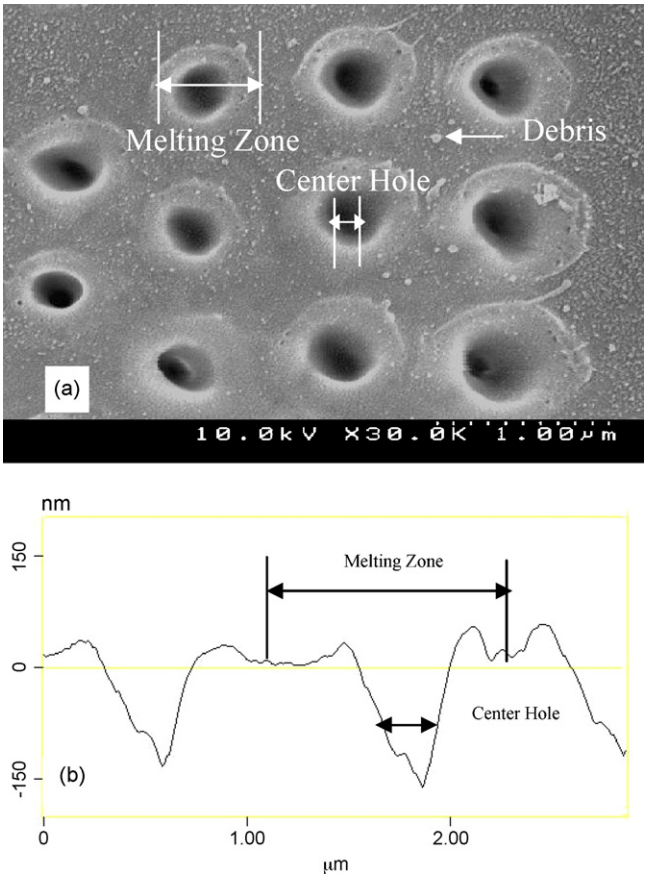

Fig. 1. (a) SEM image of nano-craters formed under self-assembly 1.0 μm silica particles on glass surface by 800 nm, 100 fs laser single pulse irradiation at a laser fluence of 35 J/cm^2 and (b) AFM image of cross section view of the nano-craters.

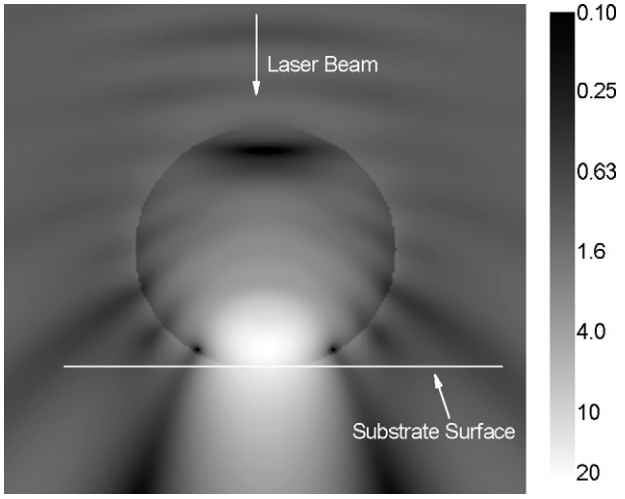

Fig. 2. Contour plot of calculated laser energy distribution, $I = S_z$, within incident plane under a 1.0 μm silica particle based on Mie theory, where laser is irradiated normally on the particle and the refractive index of particle is 1.6.

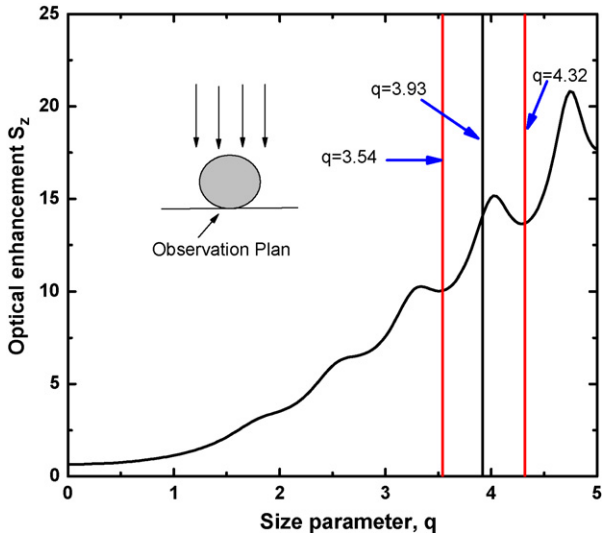

Fig. 3. Variation of the optical near-field enhancement under the particle as a function of particle size parameter q, $q = 2\pi a/\lambda$, where a is the radius of the particle. The silica particle is considered as a non-absorbing material of the laser light with a refractive index of 1.6.

in size parameter q ($q = 2\pi a/\lambda$, where a is the radius of particle) from 3.54 to 4.32. Fig. 3 presents the optical near-field enhancement under the particle as a function of q. As can be seen, the enhancement is about 10 times for $q = 3.54$ ($2a = 902$ nm) and 15 times for $q = 4.12$ ($2a = 1050$ nm), which shows an oscillation [14]. It is clear that the enhancement factor affects the formation of nano-crater even under the same laser fluence irradiation, and this could be used to explain why the nano-craters shown in Fig. 1a have different sizes in the nano-crater diameter.

It is observed that the edges of these nano-structures are free of cracks. This can be attributed to the ultra-short laser pulse. For a long pulse duration laser, the formation of cracks caused mainly by laser associated high temperature and high pressure [8]. The processing with ultra-short laser pulses essentially eliminates heat flow to surrounding material due to two-temperature model where the hot electron gas causes ablation of material, while the lattice stays cool at a significantly lower temperature. Therefore, thermally induced substrate cracking is prevented.

To further investigate the effect of laser fluence on the formation of the nano-craters, the experiments at different laser fluences were carried out. Fig. 4 is the relationship between the sizes of center-hole and melting zone as the functions of laser fluence. Both center-hole and melting zone sizes increase with laser fluence while the size of melting zone increases faster than that of center-hole. At high laser fluence, the melting zone can be as large as the particle size of 1 μm while the diameter of center-hole remains at about 300 nm. When the laser fluence increases further to 61.3 J/cm^2, the whole area was damaged. In contrast, the glass substrate without the particle mask was irradiated at the same laser fluence and no damage was observed. It proves that near-field enhancement was the main mechanism for the nano-feature formation.

Meanwhile, much debris was observed when increasing the laser fluence. Some of them was resulted from the high fluence laser ablation of the glass substrate. Fig. 5 is the normalized

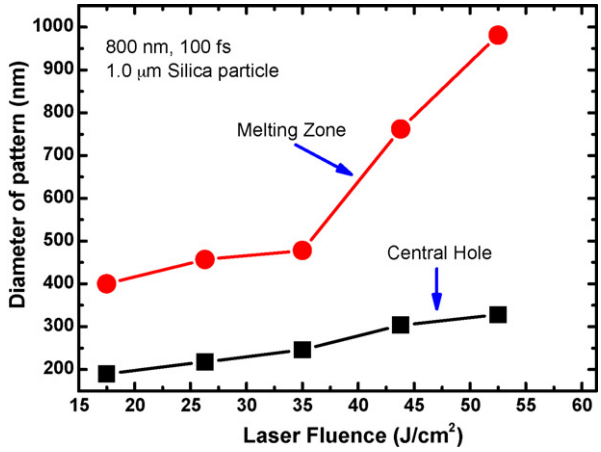

Fig. 4. Average size of melting zone and central hole as the functions of laser fluence under 1 μm particle mask exposed to 800 nm, 100 fs pulse duration femtosecond laser surface.

poynting intensity distribution along z axis under a silica particle ($a = 0.5$ μm, $n = 1.6$) by 800 nm femtosecond laser irradiation based on Mie theory. It indicates that the maximum enhancement (focus point) is located in the interior of the particle. Hence, some of the ablation debris comes from the exploded silica particles due to that highly localized field inside the sphere. For comparison, silica particles with the diameter of 6.84 μm (Bangs Laboratories, Inc.) were used to check whether there would be debris or not under bigger size particles. Theoretical calculation predicts that the focusing point moves outward as the size of particle increases. Fig. 6 is the normalized poynting intensity distribution along z axis under a silica particle ($a = 3.42$ μm, $n = 1.6$) by 800 nm femtosecond laser irradiation based on Mie theory. The focus point was found at the interface between particle and substrate surface. Fig. 7 presents the image of substrate surface after the femtosecond laser irradiation. The craters formed hexagonally closed-packed array on the glass surface with the spacing of 6.84 μm which is the diameter of particles applied. No debris was found except the particles in the center ejected from other location which were in their integrity. The profile of the crater

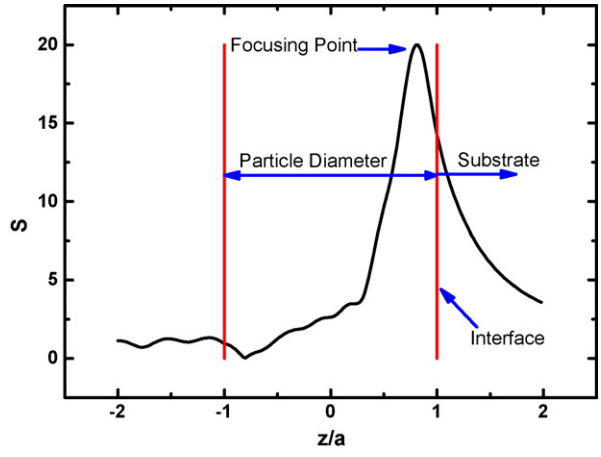

Fig. 5. Normalized poynting intensity distribution along z axis under a silica particle ($a = 0.5$ μm) by 800 nm femtosecond laser irradiation based on Mie theory.

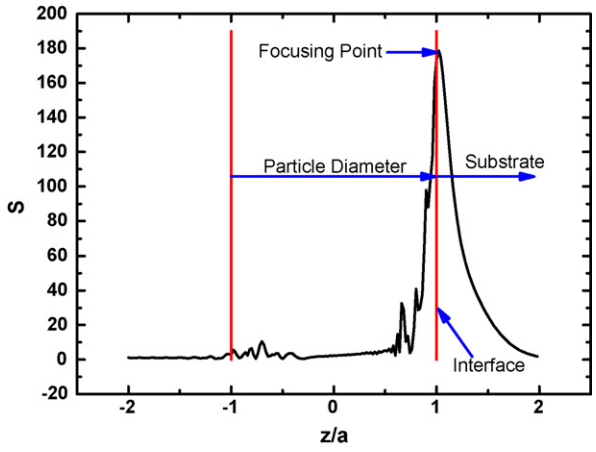

Fig. 6. Normalized poynting intensity distribution along z axis under a silica particle ($a = 3.42\ \mu m$) by 800 nm femtosecond laser irradiation based on Mie theory.

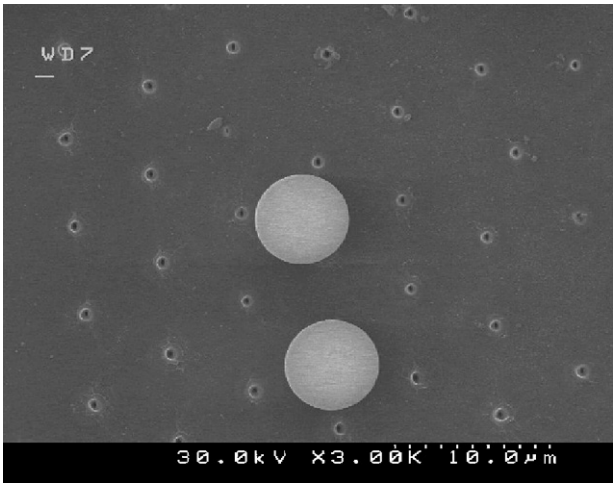

Fig. 7. SEM image of substrate surface after femtosecond laser (100 fs, 800 nm) irradiation of self-assembly 6.84 μm silica particles.

measured by AFM indicates that average diameter of craters is 400 nm which is 100 nm bigger than those formed under 1 μm particle mask while the depth remains the same at about 150 nm.

4. Conclusions

Near-field enhanced nano-drilling of transparent glass substrate by 800 nm femtosecond laser was investigated. Nano-craters were created on the glass surface after laser irradiation of self-assembly 1 μm silica particles mask. No cracks were found at the edges of produced nano-structures due to two-temperature non-equilibrium for electrons and lattices. Nano-structure formation can be attributed to two main reasons: (1) triple-photon absorption arising from the interaction between femtosecond laser pulse and transparent glass substrate, and (2) optical near-field enhancement due to light scattering by the transparent silica particles. At a low fluence, feature sizes were found from 200 to 300 nm with an average depth of 150 nm. While more debris was formed at a high fluence. When using 6.84 μm particles as mask, particles are in their integrity which verifies that the location of focus point with the highest enhancement depends on the size of particle. This nano-drilling method provides a potential approach to achieve no crack nano-fabrication of glass substrate.

Acknowledgements

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