Laser micro/nano fabrication in glass with tunable-focus particle lens array

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Abstract: Based on medium-tuned optical field enhancement effect around a self-assembled particle-lens array (PLA) irradiated with a femtosecond (fs) laser source, we demonstrated that high-precision periodical array of micro/nano-structures can be readily fabricated on glass surface or inside glass in large areas in parallel without any cracks or debris. The technique has potential for rapid fabrication of three-dimensional structures in multiple layers inside glass.

OCIS codes: (220.4610) Optical fabrication; (350.3850) Materials processing.

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1. Introduction

Due to its outstanding mechanical, chemical and optical properties, glass has widespread applications in novel device packaging, optical communication and micro-technologies such as micro-optics and biomedical devices. In general, glass is hard, brittle and non-conducive. It is a high challenge to process glass by most conventional machining techniques. In recent years, laser microprocessing has arisen as an attractive approach in glass engineering. Before the availability of femtosecond (fs) laser, high power UV and CO_2 lasers had been generally used for glass processing, since glass has very low linear absorption rate in the visible wavelength range. The mechanism of bulk damage by these lasers involves heating of conduction band electrons by the incident radiation and transferring of this energy to the lattice. Damage occurs via the conventional thermal process. The build up of heat-affected zone (HAZ) and thermal stress often lead to glass cracking, which greatly limits the applications.

The appearance of fs laser source shed new lights on glass processing with lasers. Due to the short pulse duration, the time for laser energy deposition is generally shorter than the electron-lattice relaxation time, leading to minimized HAZ and improved spatial resolution. Furthermore, it has been found that fs infrared laser irradiation of glass can result in the modification of the refractive index at the focal point inside the glasses, which paves the way for the fabrication of optical components, such as waveguide, splitter and coupler inside glass [1]. Other techniques, such as laser-induced plasma-assisted ablation and laser induced backside etching, have been used for processing transparent glasses. It should be noted, however, most previous studies utilize a single focus spot for fabrication, and the structures have to be built up dot by dot in a serial sequence. To improve the efficiency, laser processing using micro-lens array (MLA) or particle-lens array (PLA) have been demonstrated in recent years [2-4]. Both MLA and PLA can convert a single laser beam into a multiplicity of enhanced optical spots in parallel at focus, and thus increase processing efficiency. Unlike fabrication of conventional MLA by complicated processes such as photolithography and resist reflow, PLA can be easily prepared through self-assembly process by applying the colloidal solution of small particles onto the sample surface [5, 6]. After water evaporates, the PLA can stay firmly on the sample surface because of the adhesion forces [7]. The adhesion is strong enough to hold the particles on surface even the PLA was subject to a water flow. This makes it possible to carry out laser processing using PLA in different liquid media rather than air/vacuum. Very recently, we have demonstrated that by combining fs laser radiation with PLA (micron-sized particles) in air environment, hexagonal array of holes with typical lateral size ranging from 200 nm to 400 nm and depth ~150 nm can be generated on glass surface [8, 9]. In those studies, the PLA was in direct contact with glass surface (referred to as CPLA technique) and their focuses are within the near-field distance (~λ) away from the contacting points. Due to the contacting nature, introducing of the liquid media into CPLA unavoidably leads to the simultaneous immersion of the focusing lens and samples inside liquid environment. This is different from the general cases of laser processing of materials in liquid media as they were carried out in far-field (lens-sample distance d>> λ) and only the samples were immersed in liquid. In this paper, theoretical results on the tuning effect of a liquid media on the focusing properties of PLA and a single particle are presented. Experimental evidences of large-area hexagonal nano-height ring-bumps and convex-bumps fabricated on glass surface are then given. The possibility on utilizing the technique for multi-layer micro/nano-fabrication inside glass is discussed.

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2. Theory

Although it was discovered nearly a century ago, Mie theory continues to play a crucial role in describing the optical properties of small particles [10]. To illustrate the basic physics of focus tuning effect by a liquid medium, Mie theory modeling of optical near field around a single particle has been carried out. Figure 1 shows the Mie modeling results of the cross-sectional view of the normalized local field distribution ($|E|^2$) underneath a single 5.0 µm particle in (a) air and (b) immersed in water medium, under an x-polarized plane wave excitation at 800 nm. As it can be seen in Fig. 1(a), the focus point of a 5.0 µm particle in air is located at position z/a=1.1, which is close (subwavelenth distance) to the particle surface. Such enhanced field was associated with optical cavity resonance inside transparent particles and can be thought as the inference of incident beam and evanescent waves presented in the vicinity regions of

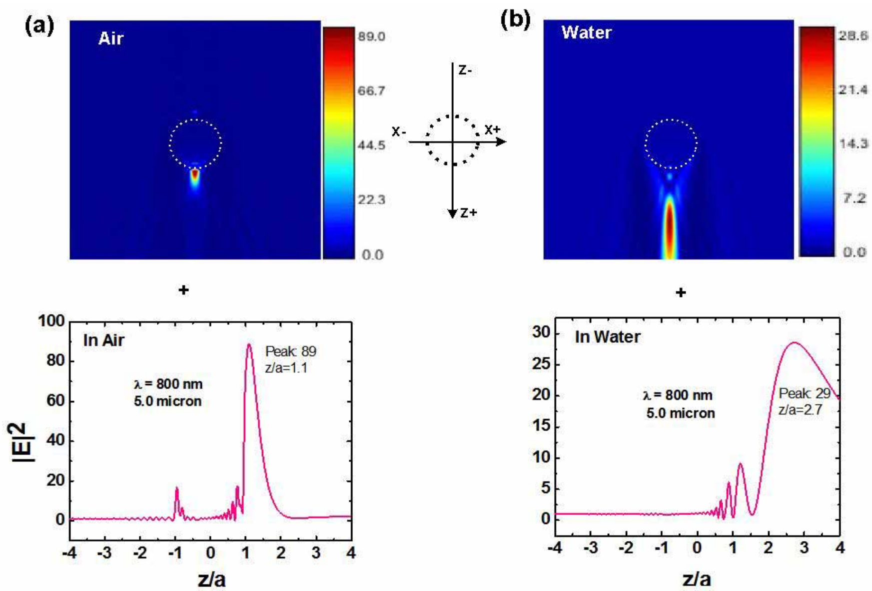

Fig. 1. Cross-sectional view of the normalized local field distribution ($|E|^2$) underneath a single 5.0 µm particle in (a) air and (b) immersed in water medium. The incident laser (λ=800 nm) beam is linearly polarized along x-axis and propagates along z-axis.

the particles [11], i.e., in the optical near-field region. As a near-field characteristics, the field enhancement within range from z/a=1.1 to z/a=2.2 in Fig. 1(a) decays almost exponentially. It means for efficient surface patterning the sample must be kept in near-field contact with particles. As such, PLA tends to be removed after single pulse irradiation due to substrate thermal expansion or ablative force. On the other hand, the focusing properties of particle changes dramatically when a surrounding medium is used. As it can be seen from Fig. 1(b), the presence of water medium greatly extends the focal length, from z/a=1.1 to z/a=2.7, as compared to Fig. 1(a). In this case, PLA tends to remain on surface after multiple laser shots. Meanwhile, the field enhancement decays from focus point much slowly compared with that in Fig. 1(a) for air medium, resulting in significant increase of the focus depth. It is beneficial for laser processing of materials in terms of fabricating higher-aspect-ratio structures into substrates with multiple shots, since more laser energy could be coupled into substrate when focus depth extends.

The focusing properties of a PLA, however, are affected by other factors that Mie theory does not take into account. These include the presence of neighbouring particles with different degree of aggregations, different substrate materials and complex (anisotropy) surrounding media. Over the last 30 years, several computational techniques that are capable of dealing

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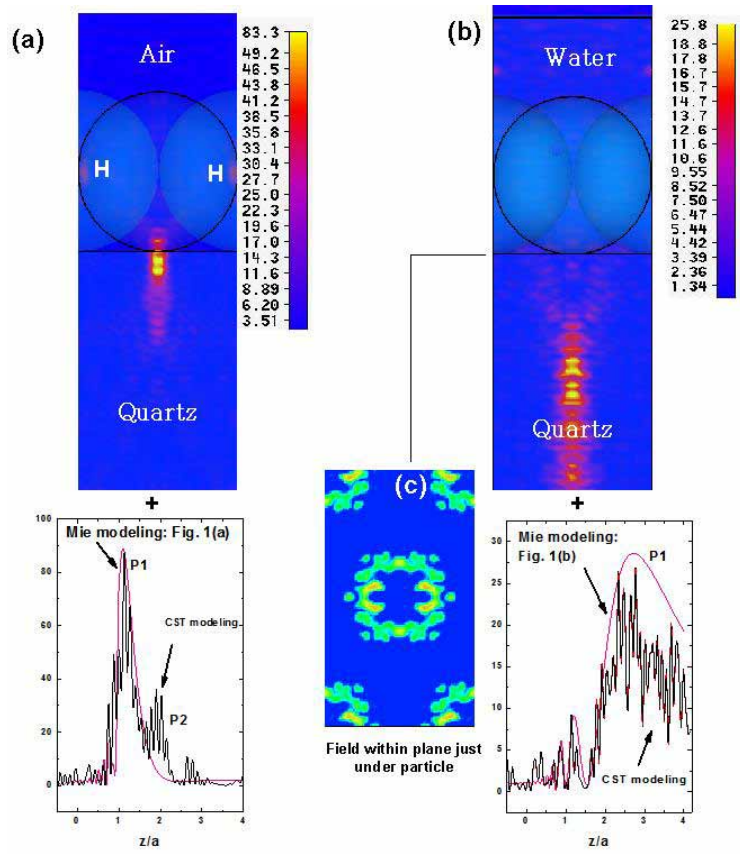

Fig. 2. Cross-sectional view of normalized local field distribution ($|E|^2$) underneath a hexagonal array of 5.0 μm particles deposited on Quartz substrate (a) in air and (b) immersed in water. The incident beam has a wavelength of 800 nm and is polarized along horizontal-direction. The refractive index of quartz and spheres are same as 1.45332, and 1.326 for water. (c) the electric field on substrate surface just under the particles.

with such cases have been developed. It includes semi-analytical methods like multiple multi-pole (MMP) technique [12], discrete dipole approximation (DDA) [13] and pure numerical methods such as finite element method (FEM) [14] , finite difference time domain (FDTD) technique [15] and Finite Integral Technique (FIT) [16]. In this paper, a commercial FIT software package (CST Microwave Studio 2006 [17]) was used for the field analyses of PLA in contact with glass surface. The structure was represented with a unit cell combined with periodical boundary conditions within the substrate plane. The mesh density was set as λ/10. A plane wave was incident perpendicular onto the sample surface, with an open boundary applied in this direction. Figure 2 shows the corresponding calculation results of cross-sectional views of normalized local field distribution ($|E|^2$) underneath a hexagonal array of 5.0 μm particles deposited on quartz substrate (a) in air and (b) immersed in water. The incident beam has a wavelength of 800 nm and is polarized along horizontal-direction. Compared with the single particle case as in Fig. 1, it is found that the fields in Fig. 2 become oscillating while the focus point (highest enhancement peak) remains almost the same. Meanwhile, the amplitudes of the fields within the focus regions were generally smaller than those in Fig. 1. The appearance of the tail of the focus (see P2 in Fig. 2(a)) extends the focal length which could increase energy coupling into deeper substrate regime (this situation remains same for water medium). As pointed out in a previous paper [18], there is an outgoing energy flow from particle within the hexagonal array to those particles sitted on the edges of the array, which results in the decrease of fields under the particles. This flow is clearly higher in air medium instead of water medium, as evidenced by the stronger coupling spots (marked as H-points) in Fig. 2(a). Multiple reflections of energy flows between the particles and the substrates could result in more laser energy flowing into the substrate. The final fields presented in Figs. 2(a) and 2(b) could be thought as the competing results of these two effects. In Fig. 2(c), it was shown that the electric field distribution on substrate surface just beneath the particles manifests a ring-shaped profile.

3. Experimental

A 1.0-mm-thick fused silica was used as the sample in experiment. It was cleaned with warmed acetone in an ultrasonic bath for 10 min followed by rinsing in IPA and DI water for 5 min. It was then immersed in 30% Nitric acid solution for 24 hours to make the glass surface hydrophilic. The sample was finally re-rinsed with DI water and dried by pure N2 gas. The spherical silica particles (Bangs Laboratories) with diameter of 5.0 μm were applied on the quartz surface after the original suspension had been diluted with DI water. The substrate was kept still until all the water had been evaporated. As a result, a reasonable uniform monolayer hexagonal array in a area of ~0.5 cm^2 was formed on the glass surface in our experiment. Before laser irradiation, the glass sample was fixed onto the bottom of a small glass vessel mounted on a three-axis stage. DI water was then added into the vessel to completely immerse the sample and the PLA.

A Ti: Sapphire regenerative amplifier system (Coherent Libra) was used as the light source (wavelength λ=800 nm, pulse duration τ=110 fs, maximum pulse energy 1.0 mJ and repetition rate variable from 1 to 1000 Hz). The nominal Gaussian beam diameter ($1/e^2$, TEM_{00} mode, M_2<1.5) at the laser exiting window is ~ 5 mm. The laser beam (linearly polarized) was incident normally on the sample surface without any focusing. The pulse energy was controlled by the Neutral Density Filters inserted in the optical path. The laser was operated in the manual mode and only a single pulse was used for each processing. The sample after processing was characterized by optical microscopy (OM) and atomic force microscopy (AFM; Vecco CP2).

Figure 3 shows the AFM images of hexagonal array of two different types of bumps formed on the quartz surface immersed inside water after a single fs laser pulse irradiation (central region of the Gaussian laser spot) at a pulse energy of 0.20 mJ (fluence:1.0 mJ/cm^2) and 0.88 mJ (fluence:4.5 mJ/cm^2) through self-assembled 5.0 µm PLA, respectively. As was shown in Fig. 2(b), the primary focus of water-immersed PLA is inside the glass bulk. In the fs time scale, the laser energy was simultaneously coupled into the glass bulk under PLA through nonlinear multiphoton absorption processes. Within each focal volume inside glass, the energy is highly confined and it pushes the matter into a state of extreme non-equilibrium. This can lead to material properties modification or the generation of micro-voids (micro-explosion) inside glass [19]. Through OM examination of samples processed at different laser energies, we confirmed that hexagonal array of patterns do exist inside glass as evidenced by the appearance of dark-contrast dot array inside glass. These in-bulk dark-contrast arrays were observed for all the samples including those processed at low pulse energies without visible patterns induced on the top surface. Ring-bumps were the first structures that can be seen on glass surface as field enhancement factor within the surface plane reaches some critical level (see Fig. 2(c)). Since there are no any field enhancements at particle-glass contacting points, the convex bumps in Fig. 3(b) can only be possible to be induced by the in-bulk micro-explosion process, which pushes the materials on the top of focal volume towards the top surface. This assumption was confirmed by different heights of convex-bumps observed at different laser energies. At a pulse energy of 0.88 mJ, the bump height is around 60 nm (Fig. 3(c)). There is no cracks and debris observed around these bumps. The roughness of the bump surface and original glass surface are almost same, indicating bumps were formed due to inner material expansion towards surface instead of violent ablation taking processing on surfaces. As a Gaussian beam was used in experiments, the bumps height are gradually decreasing toward the edges of laser spot. To improve the uniformity, a top-hat beam source should be

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applied. This can be done by inserting a homogenizer in the optical path. The fabricated high-quality convex-bumps are ideal for low flying height control in hard disk industry.

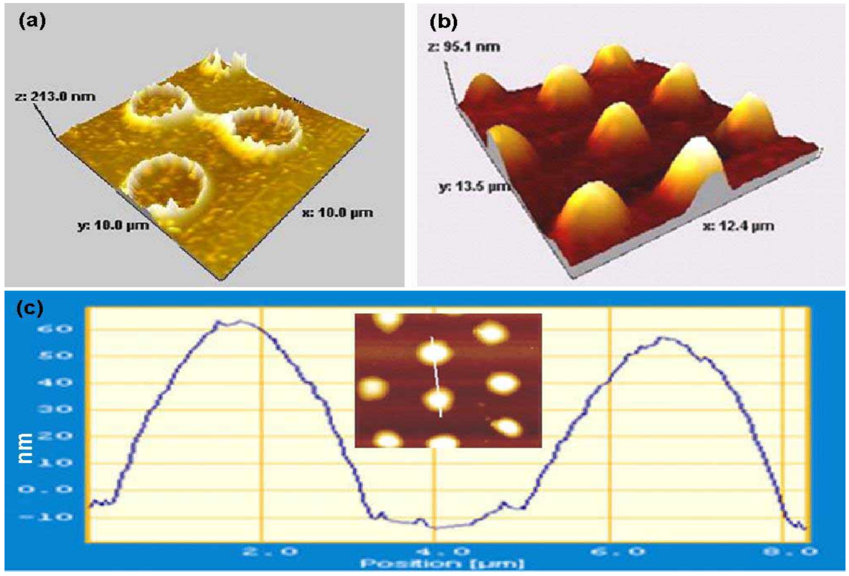

Fig. 3. Atomic Force Microscopy (AFM) images of hexagonal array of (a) ring-bumps and (b) convex bumps generated on quartz surface immersed inside water after a single femtosecond laser pulse irradiation at a pulse energy of 0.20 mJ and 0.88 mJ through a self-assembled 5.0 µm SiO_2 particle array, respectively.

As a final note, the developed technique here is possible to be extended to generate multi-layers of hexagonal array inside transparent glass materials. The focus position for different layers can be easily and precisely controlled by a liquid medium with controllable refractive index. Furthermore, if an angular-incident laser beam is applied [20], one can fabricate multi-layers of complex shaped patterns inside the glass bulk.

6. Summary

We have presented a simple and efficient technique to fabricate periodical micro/nano-structures array (hexagonal) on glass surface or inside glass. Due to the presence of water medium, the multiple focusing spots of the microsphere array illuminated by fs laser were tuned to positions beneath the surface, and focal length is greatly extended. Depending on the laser fluence, it has demonstrated that different micro/nano-structures such as nano-height ring-bumps and convex bumps can be fabricated on glass surface in large area without cracks and debris. We also discussed the possibility to extend present technique to fabricating multi-layered three dimensional micro/nano-structures inside the bulk.

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