Silica-based microstructures on nonplanar substrates by femtosecond laser-induced nonlinear lithography

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Abstract. We developed a technique for the formation of nonplanar surfaces of inorganic optical materials by a combined process of nonlinear lithography and plasma etching. This technique can be used to fabricate structures even on non-flat substrates, which is difficult using current semiconductor technology. Three-dimensional patterns were written directly inside a positive-tone photoresist using femtosecond laser-induced nonlinear optical absorption. The patterns were then transferred to underlying nonplanar substrates by the ion beam etching technique. For the lithographic process, we obtained a minimum feature size of 900 nm, which is below the diffraction limit. We demonstrated the fabrication of silica-based hybrid diffractive-refractive lenses. Fresnel zone plates with smooth surfaces were obtained on convex microlenses. When a 633-nm-wavelength He-Ne laser was coupled normally to the hybrid lens, the primary focal length was measured as 630 µm. This hybridization shifted the focal length by 200 µm, which agreed with the theoretical value. Our process is useful for the precise fabrication of nonplanar structures based on inorganic materials.

1. Introduction

Micro-optical elements are becoming increasing important for compact and functional components in optical imaging, optical pickup systems and so on. Hybrid elements have received particular attention because they are useful for the creation of new photonic functions. For example, multifocal and achromatic lenses can be obtained by combining refractive and diffractive lenses because of the opposite wavelength dispersion between both elements. To fabricate hybrid structures, microfabrication on nonplanar substrates is required. However, it is difficult to obtain such structures using current semiconductor technology because of the difficulty in achieving a uniform resist coating. The fabrication of polymeric diffractive elements on bulk convex lenses was demonstrated by maskless ultraviolet laser lithography [3]. However, specially modified stage systems are needed. A spray-coating technique was also reported to obtain a uniform resist thickness, even on nonplanar structures. The spatial resolution is several tens of micrometers [4]. In these techniques, complex exposure systems are required because single-photon processes are employed.

Nonlinear optical processes such as multiphoton absorption and tunneling ionization occur only around the focal volume when femtosecond laser pulses are tightly focused into transparent materials.
Consequently, polymerization, densification and an increase in refractive index can be induced directly inside materials. By utilizing such nonlinear optical processes, three-dimensional (3-D) microsculptures of resin including photonic crystals and so on, have been formed using photosensitive resins such as SCR-500 and SU-8 [5-7]. Direct laser writing of 3-D waveguides inside glass materials has also been reported [8].

In this paper, we report femtosecond laser lithography-assisted micromachining (FLAM), which is a combined process of nonlinear lithography and subsequent plasma etching for fabricating structures onto nonplanar substrates. Silica-based diffractive-refractive hybrid microlenses were obtained using FLAM.

2. Experimental

We used a femtosecond fiber laser for this process. The wavelength, pulse duration and repetition rate were 780 nm, 68 fs and 50 MHz, respectively. The laser beam was focused by an objective lens with a numerical aperture (NA) of 0.5. The focal spot diameter and focal depth, as measured by the knife-edge method, were 2.6 and 14 µm, respectively. Silica glass plates of 1 mm thickness and silica microlens arrays were used. The diameter, height, curvature radius and focal length of each individual lens were 240, 18.9, 380, and 830 µm, respectively. We used a chemically amplified positive-tone photoresist PMER P-CA1000PM (Tokyo Ohka Kogyo Co., LTD.). Silica was etched by electron cyclotron resonance (ECR) plasma using CHF3 gas.

3. Results and discussion

3.1. Femtosecond laser lithography-assisted micromachining

Figure 1 schematically shows the FLAM process for the fabrication on nonplanar structures. We begin with spin-coating of the positive tone resist PMER onto nonplanar substrates. Then, the patterns are directly written inside the resist, using femtosecond laser-induced nonlinear optical absorption. After post-exposure baking and development, the patterns are transferred to the underlying nonplanar substrates by plasma etching. Subsequently, we obtain nonplanar microstructures of inorganic materials after removal of the residual resist. Unlike previously reported femtosecond laser microsculptures, a positive-tone resist is desirable for use in FLAM because unwanted plasma damage can be avoided to the surroundings.

In the standard semiconductor process, ultraviolet light is absorbed from the resist surfaces. Therefore, a uniform resist coating is required to obtain well-defined patterns. When the resist is coated upon nonplanar substrates, the resist thickness varies from area to area mainly because of surface tension. As a result, it is difficult to form precise structures even with highly accurate laser control. In contrast, we can directly expose the internal region of the resist using the nonlinear process in FLAM. Unlike standard lithography, the pattern width is mainly determined by the diameters of the region where the nonlinear process occurs, and does not depend on the exposed position in the resist. Therefore, after plasma etching, we obtain precise inorganic structures even on nonplanar substrates.

![Figure 1. A schematic illustration of FLAM on nonplanar substrates.](image-url)
3.2. Nonlinear lithographic processes

Cross sectional patterns of resists on silica glass plates are shown in Figure 2(a). The focal position for writing each pattern is indicated in the figure. The substrate surface is defined as the origin of the $z$-axis, which is the optical axis. These patterns were formed without laser scanning the laser spot along the $z$-axis. The average laser power and writing speed were 18 mW and 150 $\mu$m/s, respectively. It is apparent that the patterns were formed directly inside the resist under the condition that the focal position was at $z = -3.5$ $\mu$m. PMER is transparent at wavelengths $\lambda > 480$ nm. This suggests that multiphoton absorption is responsible for the absorption at a laser wavelength of 780 nm.

The dependence of $z$-positions of the laser focal spot on pattern heights is shown in Figure 2(b). The dashed line indicates the resist thickness on the substrate. As shown in the inset, the pattern height and width were determined as the groove depth and surface width, respectively. We scanned the laser spot inside a resist of 13 $\mu$m thickness at a writing speed of 100 $\mu$m/s. The pattern widths were approximately 2–3 $\mu$m. The pattern heights decreased linearly with the focal position. The disagreement of the trend near the resist surface is most likely due to the shift in the focal spot, which originated from the difference in the refractive index between air and the resist. For our exposure setup, a minimum width of 900 nm was obtained at a power of 21 mW and writing speed of 1500 $\mu$m/s.

3.3. Hybrid lenses

To obtain silica-based diffractive-refractive hybrid microlenses, binary Fresnel lenses were fabricated on convex microlenses. The focal length of the hybrid lens can be designed using eq. (1) [9]:

$$f_h = \frac{1}{f_h} + \frac{1}{f_m} + \frac{1}{f_F}$$

where $f_h$, $f_r$, and $f_m$ are the primary focal lengths of a hybrid lens, a Fresnel lens, and a convex lens, respectively. From eq. (1) the focal length of a hybrid lens should be 618 $\mu$m when a Fresnel lens with a focal length of 2420 $\mu$m is formed on the convex lens. The radius $r_m$ of the $m$-th zone of the binary Fresnel lens can be expressed by $r_m = \sqrt{mf_r \lambda}$ for an incident wavelength of 633 nm. Figure 3 schematically shows the laser writing procedure of the hybrid lenses. The average laser power and writing speed were 20 mW and 300 $\mu$m/s, respectively. Fresnel lenses consist of a series of concentric rings of different radii. The separation between adjacent rings ($\Delta r$) was 2.0 $\mu$m, which was comparable in size to the diameter of the spot. To penetrate a thick resist (> 20 $\mu$m), multiple patterns of the Fresnel lens, centered on the same axis ($z$-axis), were written at different heights in the resist. The

![Figure 2](image_url)
layer spacing was 3.0 µm, which was smaller than the height of the photo-modified region. Figure 4 shows SEM images of a silica-based diffractive-refractive hybrid microlens after pattern transfer. We obtained well-defined silica structures with smooth surfaces on curved substrates. The ECR power, bias voltage, and gas pressure were 100 W, 700 V and 2.0 × 10⁻² Pa, respectively. The etching depth was approximately 1 µm. The primary focal length was 630 µm when a 633 nm wavelength He-Ne laser light was coupled normally to the hybrid lens. This hybridization shifted the focal length by 200 µm, which is consistent with the theoretical value of 212 µm. In the lithographic process, the patterns were written with $dr = 2.0$ µm. This relatively large spacing made it different to an ideal Fresnel lens structure resulting in a diffraction angle shift. An improvement in the FLAM process can result in the fabrication of more functional devices with 3-D surfaces on inorganic materials.

4. Conclusion

We report on FLAM, which is a combined process of nonlinear lithography and plasma etching, for the fabrication of nonplanar structures on inorganic materials. The fabrication of silica-based diffractive-refractive hybrid microlenses was demonstrated. The silica Fresnel lenses were created on 18.9 µm-thick convex lenses. This hybridization shifted the focal spot by 200 µm, which agreed with the theoretical value. Improvement of the FLAM process can lead to the fabrication of more functional devices with 3-D surfaces on inorganic materials.

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