Regeneration of a Grating in PMMA Inscribed by Femtosecond Laser Bessel Beam

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We demonstrate recovery of diffraction efficiency of the grating in PMMA inscribed by direct femtosecond laser writing. We investigated time-lapse measurements of the diffraction efficiency of volume gratings in PMMA after inscription with femtosecond Bessel beam. Use of the side-plane as an incident plane for diffraction measurements enables control of the volume grating thickness by the translation length of the sample. Hence optically thick volume gratings can be made. Bessel beam with an axicon lens produces a larger modification length, and is favorable for diffraction measurements. The first-order diffraction efficiency of the grating begins to increase after fabrication. Volume Bragg gratings with the thickness of 2 mm reached diffraction efficiency of approximately 90% after three days (the readout wavelength was 633 nm). Diffraction efficiency decays afterwards on the time scale of tens-of-days. Interestingly, a recovery of the grating and its diffraction efficiency was observed after 100 days. A glass-rubber relaxation under an internal strain and oxidation of laser-produced dangling bonds can explain the observation.

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1. Introduction

Femtosecond (fs-)lasers allow the fabrication of integrated photonic devices in three dimensions. Translation of a focal in a three-dimensional (3D) pattern inscribes modifications with micrometer-order precision over large macroscopic footprint required for practical use of devices [1-3]. Femtosecond laser micromachining is attractive for low volume production of widely diverse products. Fabrication of volume gratings in polymer materials has been demonstrated using fs-lasers [4-11]. The diffraction efficiency is decreasing in time as Ye *et al*. demonstrated [11]. First, the diffraction efficiency of the fabricated grating with thickness of 1-4 mm increased monotonically with time; e.g., the diffraction efficiency of the fabricated grating 5-7-mm-thick reach a maximum from hours to days after inscription and then decreased monotonically with time [11]. New phenomenon of a self-regeneration of the fabricated grating in PMMA after 100 days was observed [12] when Gaussian beam was used. With a Gaussian–Bessel beam generated with an axicon lens a longer modification inside transparent materials [13-16] can be made. Ultra-short Bessel beams were introduced for inner volume modification for stealth dicing and laser cutting applications [17-22]. In those studies [17-22], it was been demonstrated that opening of a cylindrical hole is due to compression with no ablation debris. This was confirmed by shadowgraphy and hole formation in PMMA using Bessel fs-laser pulses [23]. In this paper, we present time-lapse measurements of the diffraction efficiency of volume gratings in PMMA after inscription by femtosecond Bessel beam. When the side plane is used as an incident plane for diffraction measurements, the volume grating thickness can be controlled by the sample translation length. Volume Bragg gratings with a period of 10 μm exhibit diffraction efficiency of approx. 90% after three days; readout wavelength was 633 nm. Diffraction efficiency of the fabricated gratings reached the maximum after several days, then decreased with time before the recovery after 100 days.

2. Volume grating thickness control in transparent materials

Figure 1 shows schematics of femtosecond laser inscription and readout of a grating. The writing laser pulses propagate along *z* axis. Conventionally, the measurement beam for diffraction efficiency is incident on a plane (*xy* plane), which is perpendicular to the laser inscription axis (Fig. 1(a)). When we used the side plane (*xz* plane) as an incident plane for diffraction measurements (Fig.1(b)), the volume grating thickness during inscription can be controlled by the translation length along the *y* axis. The incident plane size in volume gratings is varied by the number of modification lines along the *x* axis and modification length. Using a Bessel beam generated by an axicon lens a larger modification length is produced; it is used for an incident plane in diffraction measurements.

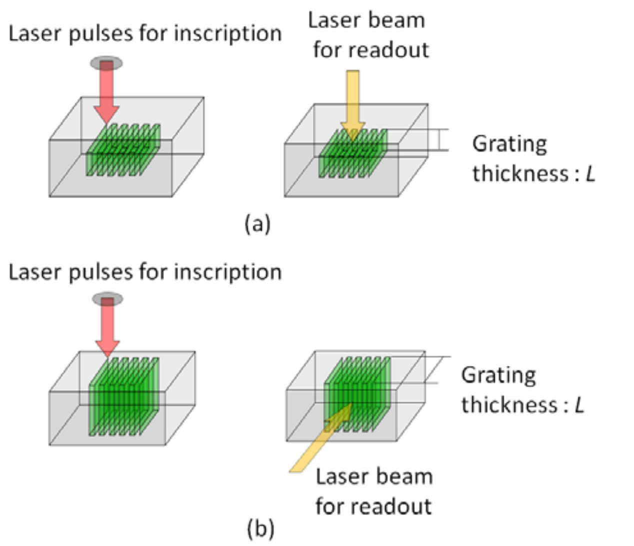

Fig. 1. Schematic setup for fs-laser irradiation and diffraction measurement. (a) The measurement beam for diffraction efficiency is incident on a plane that is perpendicular to the laser inscription axis. In this case, the volume grating thickness is controlled by the modification length. (b) The side plane (*xz* plane) is used as an incident plane for diffraction measurements.

3. Experimental setup

Fabrication of gratings was performed using an ultrafast Ti:sapphire laser-amplified system that produced 100 fs pulses of 800 nm light at a pulse repetition frequency of 1 kHz (Spectra-physics, Spitfire Pro). The laser pulse energy was attenuated by a half-wave plate in front of a polarizer. The sample dimensions are 30 mm × 10 mm × 3 mm (*x*×*y*×*z*); PMMA was commercially acquired (ACRYLITE#000, Mitsubishi Rayon, Ltd.) with a refractive index n =1.491 and the glass transition temperature T_g = 120°C. The four sample sides were polished for inscription by fs-laser pulses and for the measurement of the diffraction efficiency by a He-Ne laser beam. The sample was mounted on computer-controlled translation stage (XPS-100; Newport Corp.).

The Gaussian beam (6 mm at $1/e^2$-level) from the Ti:sapphire amplified laser passed through the axicon lens to form Bessel beam. The axicon lens with cone angle of 170° transformed the Gaussian laser beam to Bessel beam. The distance between the axicon and the sample surface was set as 15 mm [16]. The sample was translated perpendicularly to the laser beam propagation direction at a constant speed of 0.5 mm/s. Volume gratings with a period of 10 µm were fabricated. We fabricated volume gratings with thicknesses of 0.5 mm, 1 mm, 1.5 mm, 2 mm, 2.5 mm, and 3 mm at the energy of 0.2 mJ/pulse. The energy was measured before the axicon lens.

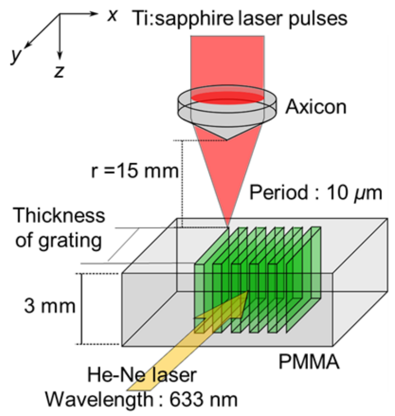

Fig. 2. Schematic diagram showing volume grating fabrication in PMMA with an axicon and readout by a He-Ne laser from side. The sample was translated perpendicularly to the laser beam propagation direction at constant speed of 0.5 mm/s. The grating period is 10 µm.

The diffraction efficiency was measured using a He-Ne laser at 632.8 nm wavelength. The He-Ne laser beam was incident upon the volume grating at the Bragg diffraction angle. The intensity of each diffracted beam was measured using a power meter. We estimated the first-order Bragg diffraction efficiency (η_1) as the ratio of the first-order diffracted beam intensity to the sum of all diffracted beam intensities, which corresponds the transmitted intensity through the sample. The sample absorption and Fresnel reflection losses were eliminated by definition. In the measurements of the diffraction efficiency, the collimated output of a He-Ne laser with the Gaussian beam (1 mm diameter) was focused with a 200 mm focal length lens. The focal spot diameter is approximately 160 µm.

4. Results

Figure 3 presents a top view of an optical image of a volume grating fabricated in PMMA using a fs-Bessel beam. The diameter of refractive-index-change region was several micrometers as recognizable in the optical microscope image. The region of filamentary modification was 3 mm long from a side view. Ablation was not observed at the entrance or exit surface and all the grating was inscribed inside the inner volume of PMMA. When femtosecond laser pulses are focused in PMMA with an axicon, the filamentary modification along the z axis is not homogeneous [15].

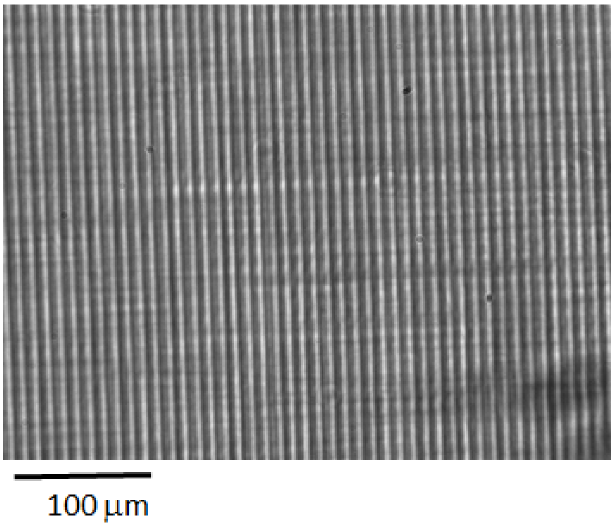

Fig. 3. Optical top-view image of volume gratings (*xy* plane) inscribed in PMMA by fs-Bessel beam with a period of 10 µm.

In order to investigate the relationship between diffraction efficiency and the thickness of the gratings, diffraction efficiency was measured three days after the fabrication of gratings. Figure 4 shows the first-order diffraction efficiency against the grating thickness. The maximum diffraction efficiency of 96% was obtained for the 2 mm thickness. The maximum diffraction efficiency in experimental measurements did not reach 100%. The difference was attributed to grating imperfections.

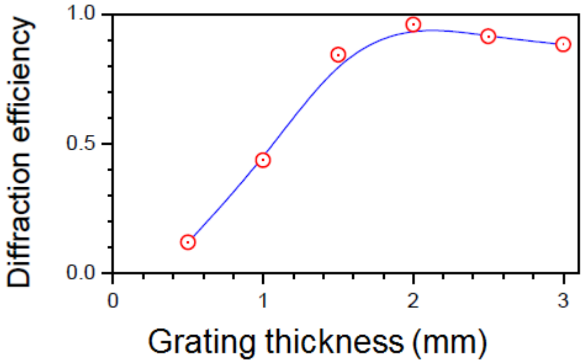

Fig. 4. Diffraction efficiency vs. grating thickness. The incident wavelength was 633 nm at the Bragg condition. The diffraction efficiency was measured three days after the fabrication of gratings.

The first-order Bragg diffraction efficiency (η_1) is related with the refractive index change, Δn, as [24]

$$\eta_1 = \sin^2\left(\frac{\pi \Delta n L}{\lambda \cos\theta_B}\right), \tag{1}$$

where θ_B is the Bragg angle. We compared experimental measurements of the fabricated grating by fs-Bessel beam and numerical results of the diffraction efficiency. The Bragg angle of 1.81 deg was used in the Bragg condition, which is expressed as

$$m\frac{\lambda}{n} = 2d\sin\theta_B, \tag{2}$$

where d stands for the grating period (d =10 µm), n represents the refractive index of material (n = 1.491), and m is the integer. By fitting the refractive index change, diffraction efficiency was calculated using Eq. (1). The refractive index change was estimated as 1.7×10^{-4}. Refractive index changes in PMMA by femtosecond laser pulses was reported to be $\Delta n = 5 \times 10^{-4}$ [4] and 0.6×10^{-4} [6]. Our results were of the same order of magnitude and coincided with previous reports.

The refractive index change induced by fs-laser pulses in PMMA is dependent on the time after fabrication [11,12]. Figure 5 shows the time evolution of the diffraction efficiency after inscription till 200 days. The initial η_1 increased during the first 3 days and reached maximum. Then, η_1 decreased till approximately 100 days. Then, the recovery of grating, i.e. regeneration of grating occurred, and the η_1 increased after 100 days. At the thickness of 2.0 mm, the initial η_1 =81 % and increased up to 96 % in 3 days. Then, η_1 decreased to 23 % after 100 days with regeneration reached 45 % after 200 days. These phenomena agree with the regeneration of grating fabricated by femtosecond laser under a single lens focusing of Gaussian fs-laser pulses [12].

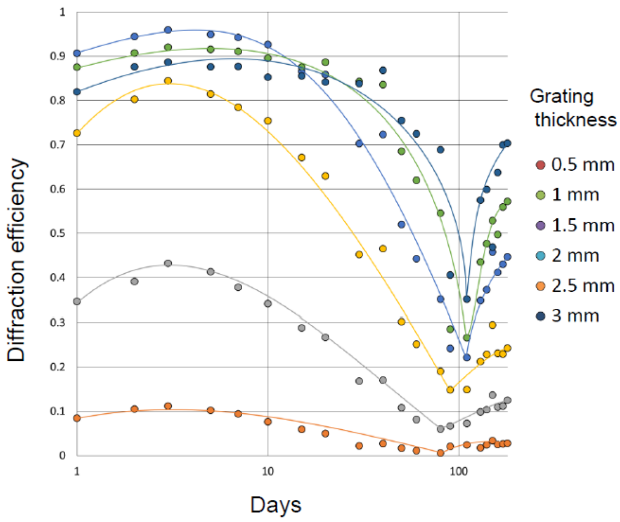

Fig. 5. Time dependence of diffraction efficiency of the volume grating after inscription by an axicon lens. The energy was 0.2 mJ/pulse.

After inscription of the grating, refractive index change decrease first in the laser-irradiated area. Femtosecond laser irradiation leads to melting of PMMA. As the melted material is resolidified, contraction of the volume causes an increase in refractive index towards the center of laser written area [25]. Next, the monomer diffusion occurs, which decreased refractive index change till 100 days. Finally, refractive index change increased again. Polymeric relaxation of the side bands (β-relaxation) and a terminal flow at longer time scales (α-relaxation) is also a candidate for the temporal evolution of the diffraction efficiency [26]. A longer time is required to relax initial stress and this occurs via slow α-relaxation responsible for mass transport. The time constant of the α-relaxation is tens-of-days and is consistent with regeneration times observed in our experiments. The recovery of the grating is called self-regeneration. After 100 days, α-relaxation of the polymer structure and oxidation of the dangling bonds are possible mechanisms for regeneration; however, the exact mechanism and its control in polymers needs further studies. The regeneration of gratings was found in glass optical fiber [27,28] and in PMMA [12]. The exact mechanism for recovery of the grating needs further studies and future work.

We utilized a low-repetition-rate laser to write the grating. Ultrashort laser micromachining in PMMA has been demonstrated including the fabrication of waveguide devices [29,30] and grating [31]. In reports of grating fabrication by using high-repetition-rate lasers, high-NA objectives were used. This limits the length of modification. It is necessary to increase the diffraction efficiency of vBragg gratings, one controls the grating thickness. Combination of the axicon lens and the objective lens will be future works by the use of high-repetition-rate lasers.

5. Conclusion and outlook

We have reported temporal evolution of volume gratings in PMMA with an axicon. Volume gratings with a period of 10 μm were fabricated using femtosecond Bessel beam with an axicon inside PMMA. Diffraction efficiency was found to be approx. 90% using the side surface as an incident plane of a readout beam at the thickness of 2 mm by varying the translation length of the sample after three days of inscription in PMMA. Time-lapse measurements of the diffraction efficiency showed initial increase of the diffraction efficiency followed by a slow decrease. Then, refractive index change increased again and recovery of grating occurred. The observation is attributed to α-relaxation of polymers. The combination of direct fabrication of volume gratings using femtosecond Bessel laser pulses and readout of diffraction pattern from the side can produce high-efficiency volume grating structures in widely diverse transparent materials.

Acknowledgments

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