

## Internal modification in transparent hybrid germanium-silica plates using plasma formation induced by a femtosecond laser

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**Abstract:** The fabrication of an internal diffraction grating with photoinduced refractive index modification in planar hybrid germanium-silica plates was demonstrated using low-density plasma formation excited by a high-intensity femtosecond (150 fs) Ti: sapphire laser ( $\lambda_p=790$  nm). The refractive index modifications with diameters ranging from 400 nm to 3  $\mu$ m were photoinduced after plasma formation occurred upon irradiation with peak intensities of more than  $2 \times 10^{13}$  W/cm<sup>2</sup>. The graded refractive index profile was fabricated to be a symmetric around from the center of the point at which low-density plasma occurred.

**Key words:** diffraction grating; refractive index modification; plasma formation; hybrid germanium-silica plate; femtosecond laser

### 1 Introduction

Several types of optical components based on hybrid germanium-silica glass have been developed in the fields of optical communications, medicine and optical sensors because of their high transmission of UV to near-IR wavelength. Among them, much attention has been paid to gratings as powerful tools for lasers, amplifiers with high optical wavelength division efficiency, and sensitive wavelength interferometers in optical waveguide sensors. In particular, an optical diffraction grating, which is a fundamental optical component used to periodically modulate the phase or amplitude of incident waves, has been expected to be a useful device for wavelength division of propagated multi-wavelength beams into a single-wavelength beam in a multi-wavelength network system [1].

In the meantime, the interaction between ultrashort, high-intensity laser light and transparent materials has become major concern since the advent of high-intensity femtosecond lasers [2–5]. The structural bulk modification of dielectrics by tight focusing of

femtosecond laser pulses was recently demonstrated [6]. Also, a large increase ( $>10^{-2}$ ) in refractive index was obtained in a modified region of glass by irradiation with femtosecond laser pulses. The infrared photosensitivity of transparent materials allows the fabrication of three-dimensional photonic structures or devices through translation of the sample with respect to the focal point. Although the physical mechanisms responsible for infrared photosensitivity are still under investigation, this technique has been applied to three-dimensional optical storage, waveguides in a wide variety of glasses, couplers, and photonic crystals [7,8].

Although several experiments of plasma formation and bulk modification in solids using high-intensity femtosecond laser pulses have been reported, the relationship between plasma formation and structural change induced in the solid has not been explained in detail [9–13]. To fabricate an optical transparent diffraction grating in a transparent planar plate, the interaction between plasma formation and a planar plate with hybrid germanium-silica were used, which enables to fabricate the periodical structural change of the refractive index along the germanium-silica plate.

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In this work, the experimental results of plasma-induced bulk modification in optical planar hybrid germanium-silica plates were reported using low-density plasma formation excited by a tightly focused femtosecond laser.

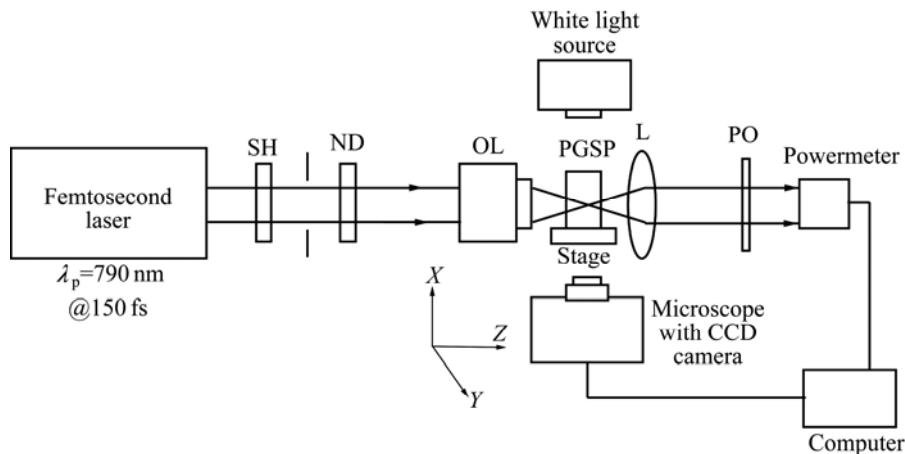
## 2 Experimental

The schematic diagram of the experimental setup for plasma-induced refractive index modification is shown in Fig. 1. The irradiation laser used in the experiment is a Ti:sapphire oscillator-amplifier laser system ( $\lambda_p=790$  nm) based on the chirped pulse amplification technique with a 150 fs pulse duration, 1 W average output power, and 1 kHz repetition rate. The linearly polarized laser beam with a Gaussian profile is focused tightly onto the planar hybrid germanium-silica plate through an objective lens (X60, N.A.: 0.85). The planar hybrid germanium-silica plate is a commercially available fused one from the Tokyo Taisei Glass Co. The dimensions of hybrid germanium-silica plate are 15 mm×25 mm× 4 mm. The six sides of the hybrid germanium-silica plate were optically polished for in situ observation. The sample (planar hybrid germanium-silica plate) is set on the  $X-Y-Z$  stage with space resolution of 50 nm to be scanned. The energy of the incident beam irradiating the planar hybrid germanium-silica plate is controlled using neutral density (ND) filters that are inserted between the laser and focusing objective lens. The power transmitted through the planar hybrid germanium-silica plate is recorded using the optical powermeter connected to the computer.

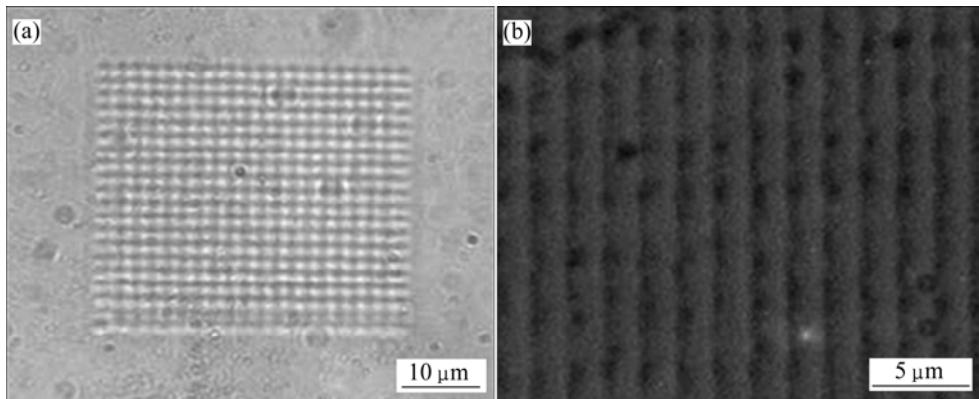
## 3 Results and discussion

By scanning the planar hybrid germanium-silica plate, using the optical  $X-Y-Z$  stages during laser irradiation, periodically arrayed structures of modified bulk were fabricated with irradiation at different intensities (Fig. 2). The depth of such periodically arrayed structures was 1 mm from the irradiated surface of the hybrid germanium-silica substrate (sample). Periodic structures with refractive index modification were fabricated due to low-density plasma formation ( $n_c < 1.79 \times 10^{27} \text{ m}^{-3}$ ) upon single-shot irradiation at  $5 \times 10^{13} \text{ W/cm}^2$  (Fig. 2 (a)). The diameter of structures with induced refractive index modification was approximately 2  $\mu\text{m}$ . The pitch between such structures is 20  $\mu\text{m}$ . Periodic structures with optical cracks were also fabricated due to solid-density plasma formation ( $n_c > 1.79 \times 10^{27} \text{ m}^{-3}$ ) upon single-shot irradiation at  $4 \times 10^{14} \text{ W/cm}^2$  (Fig. 2(b)). The diameter of structures with cracks is 8–13  $\mu\text{m}$ . The pitch between optical cracks is 20  $\mu\text{m}$ . Neither optical damage nor laser ablation on the surface of the hybrid germanium-silica substrate after laser irradiation was observed in Figs. 2(a) and (b). It is inferred that low-density plasma formation by tightly focused femtosecond beams would be useful for fabricating internal gratings with refractive index modification in hybrid germanium-silica plates.

From the electron spin resonance (ESR) spectroscopic measurement, the variation of the concentration of induced defects was measured in modified regions (Fig. 3). No defect is observed in the region of no plasma formation upon irradiation at  $6 \times 10^{12} \text{ W/cm}^2$  (Fig. 3(a)). However, the defect of SiE' is firstly



**Fig. 1** Schematic diagram of experimental setup for laser-induced plasma formation and refractive index modification in optical hybrid germanium-silica plates (SH—Shutter; ND—Neutral density filter; OL—Objective lens; PGSP—Planar germanium-silica plate; L—Lens, PO—Polarization plate)

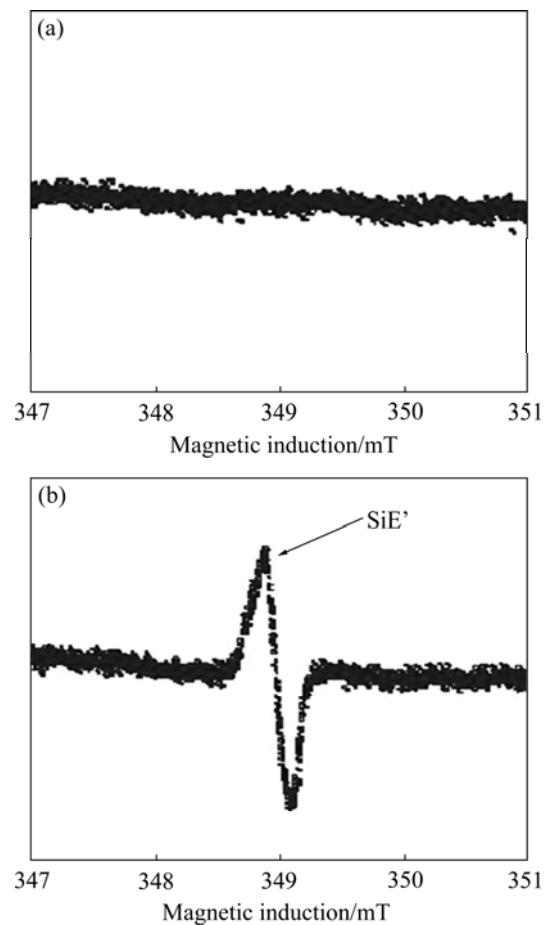


**Fig. 2** Two types of internal gratings with refractive index modification by scanning planar hybrid germanium-silica plate using optical stage: (a) Dot structure; (b) Linear structure

observed in the region of plasma formation upon irradiation at  $1.2 \times 10^{14} \text{ W/cm}^2$  (Fig. 3(b)). These results indicate that defects of  $\text{SiE}'$  are induced where plasma formation occurs. At high intensities, the concentrations of  $\text{SiE}'$  are significantly increased in refractive index modification areas. To the plasma formation, the concentration of  $\text{SiE}'$  ( $\text{E}'$  center) is significantly increased in refractive-index modified regions (Fig. 3(b)). The measured ESR signals in the modified regions agree well with the absorption spectrum of  $\text{SiE}'$ , which indicates unpaired electron in a dangling, tetrahedral ( $\text{sp}^3$ ) orbit of a single silicon bound to three oxygen atoms in the glass. Defects other than  $\text{E}'$  centers are not observed in refractive index modification.

In order to measure the profile of refractive-index change by bulk modification induced by low-density plasma formation, perpendicular Fresnel reflection was employed. This technique, as developed by Eickhoff and Weidel, relies on the Fresnel relation between the refractive index and reflectivity of the materials.

A linearly polarized He–Ne laser is incident upon a  $10\times$  beam expander and is focused onto the surface of the area of bulk refractive index modification on a polished hybrid germanium-silica plate by a  $20\times$  microscope objective. The minimum spot diameter of the focused beam on the surface of the optical hybrid germanium-silica plate is approximately 2  $\mu\text{m}$ . The opposite surface of the planar hybrid germanium-silica plate was coupled with the matching oil ( $n=1.457$ ) to prevent Fresnel reflection from the opposite surface of the optical hybrid germanium-silica plate. The thickness of the optical hybrid germanium-silica plate does not affect the incorporated measurement of the refractive index modification. The He–Ne laser is widely used as the probe light source in the measurement of refractive index by the Fresnel reflection method because of the stability of its mode and frequency. The reflected beam was detected by a photodiode and its signal was recorded



**Fig. 3** ESR spectroscopic measurement in a planar hybrid germanium-silica plate after laser irradiation of single pulses with various input intensities ( $\text{SiE}'$  center is paramagnetic defects): (a)  $6 \times 10^{12} \text{ W/cm}^2$ ; (b)  $1.2 \times 10^{14} \text{ W/cm}^2$  in plasma-induced refractive index modification

in the computer through a lock-in amplifier. The refractive index profile was obtained by parabolic fitting. The refractive index profile of the area of bulk modification at the intensity of  $8 \times 10^{13} \text{ W/cm}^2$  clearly shows that bulk modification resulted in a graded

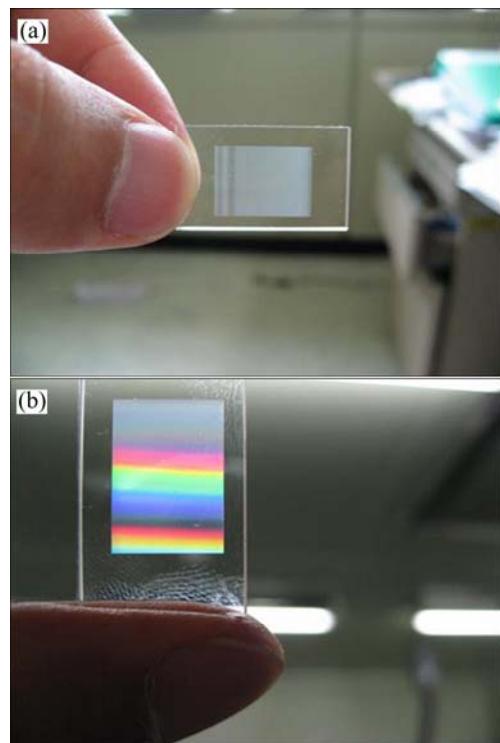
refractive index profile in an optical hybrid germanium-silica plate in comparison with the refractive index profile of the unmodified area of optical hybrid germanium-silica plate. From the measured refractive index profile of induced modification, it is found that the induced refractive index modification exhibits a graded refractive index profile symmetric about point at which the low-density plasma occurs.

Based on the diffraction efficiency of Kogelnik's coupled mode theory, photoinduced refractive index changes upon irradiation at various intensities were measured. The measured minimum value of refractive index change is  $3.5 \times 10^{-3}$  for irradiation at  $2 \times 10^{13} \text{ W/cm}^2$ . The maximum value of refractive index change is estimated to be  $1.6 \times 10^{-2}$ . As the irradiated intensity was increased, the value of refractive index change increases linearly within the range from  $3.5 \times 10^{-3}$  to  $1.6 \times 10^{-2}$  at intensities higher than  $2.0 \times 10^{13} \text{ W/cm}^2$ . The measured error of refractive index change is less than 15%.

By scanning planar hybrid germanium-silica plates using the optical  $X$ - $Y$ - $Z$  stage during laser irradiation, two types of structures of the internal grating in planar hybrid germanium-silica plates were obtained. Both a dot structure and a linear structure were fabricated using low-density plasma formation induced by irradiation of a tightly focused femtosecond laser pulse. The structure of the internal grating can be controlled using both the shutter and optical  $X$ - $Y$ - $Z$  stage during laser irradiation.

By scanning a planar hybrid germanium-silica plate using the optical  $X$ - $Y$ - $Z$  stage, the internal diffraction grating in a planar hybrid germanium-silica plate was induced by irradiation of tightly focused femtosecond laser. The linear periodically arrayed structure with refractive index modification was employed. Figure 4(b) shows the diffraction images of internal grating with refractive index modification fabricated using a white light source. The planar hybrid germanium-silica plate was scanned at the speed of 1 mm/s using the optical  $X$ - $Y$ - $Z$  stage. The laser pulse of 5 J/pulse was irradiated onto the hybrid germanium-silica plate. The pitch of the internal grating was 2  $\mu\text{m}$  (500/mm). The refractive index change of the areas of induced bulk modification is  $4.0 \times 10^{-3}$ , which is a relatively low change of the refractive index.

In this experiment, plasma formation and structural transformation were demonstrated in the regime of refractive index modification in optical transparent planar plates composed of hybrid germanium-silica at irradiation intensities lower than the threshold of optical breakdown,  $1.5 \times 10^{14} \text{ W/cm}^2$ . The underlying process of refractive index modification has been explained in related studies as the thermalization of the excited electrons due to multiphonon emission, leading to



**Fig. 4** Diffraction images of fabricated internal diffraction grating with refractive index modification using no reference light (a) and white light source (b)

permanent photoinduced structural transformations [14]. The results of ESR spectroscopic measurement show that the concentration of defects is increased around the modified regions. The formation of color centers changes both the absorption coefficient and density of the medium, leading to the increase of the refractive index. Similar procedures of positive variation of the refractive index due to the generation of conduction electrons were reported in studies of plasma in solids.

Although the details of the physical mechanisms responsible for infrared photosensitivity in the femtosecond regime are still under investigation, low-density plasma formation can easily induce refractive index modification with defects in transparent material. The diffraction images of the output beam transmitted through the periodically arrayed internal structures show that refractive index modification results in an internal grating structure. Plasma-induced refractive index modification in transparent bulk materials will be a useful technique for the design of optical devices with limited refractive index change for applications, such as optical sensors and optical communications.

#### 4 Conclusions

A new method of fabricating internal diffraction gratings with bulk refractive index modification is

demonstrated in planar hybrid germanium-silica plates using low-density plasma formation induced by a tightly focused femtosecond laser. Low-density plasma-induced refractive index modification would be a useful technique for the design of optical transmission diffraction gratings for such application as optical sensors and optical communications.

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