Spatially resolved measurement of femtosecond laser induced refractive index changes in transparent materials

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We present a practical method to determine femtosecond laser induced refractive index changes in transparent materials. Based on an iterative Fourier transform algorithm, this technique spatially resolves the refractive index of complex structures by combining the dimensions of the modified region with the corresponding phase change extracted from far-field intensity measurements. This approach is used to characterize optical waveguides written by a femtosecond laser in borosilicate glass. © 2012 Optical Society of America

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Femtosecond laser direct writing (FLDW) has become a common tool to fabricate three-dimensional photonic devices in transparent materials [1,2]. Recent investigations show its potential for creating volumetric diffractive optical elements (DOEs) [3–5] capable of multiplexing spatial and spectral information. However, for efficient design of those elements, accurate characterization of the amount and the spatial distribution of the laser induced refractive index change (Δn) is essential. Furthermore, for each application, different writing parameters may need to be adapted and optimized so that a practical and flexible refractive index measurement technique is required for the customized fabrication of DOEs.

It is common to deduce Δn from measurements of the diffraction efficiency of a grating [6] or from the NA of a written waveguide [7]. Both approaches are practical since they are fast and do not require special analysis equipment. However, they are limited in scope since they are based on approximations, neither providing data for complex structures nor any spatial information. Other methods, which investigate, for instance, the reflectivity of a volume Bragg grating [8] or the mode profile of a waveguide [9], yield better accuracy as well as good spatial resolution, but are limited to special structures. Refractive near-field profilometry [1] or selective chemical etching [10] techniques provide a high sensitivity ($\Delta n = 10^{-5}$ relative and 10^{-4} absolute) and a high spatial resolution ($\sim 0.5\ \mu$m). However, they are costly and require additional sample preparation. To our knowledge, at the present time there is no practical, fast method to spatially resolve femtosecond laser induced refractive index changes with high measurement accuracy.

In this Letter, we present a novel solution to this need. When a given FLDW structure is illuminated with a laser beam of wavelength λ [Fig. 1(a)], the interaction can be described using a thin element approximation (TEA). Within this approximation, diffraction effects during the light propagation through the structure can be neglected. This approach can be applied because the femtosecond laser interaction induces only a small change in the refractive index over spatial features with sizes large compared to the wavelength λ [11]. By using TEA, the value of the field following the structure $u_{\mathrm{out}}(x, y, z_0)$ is obtained through multiplication of the incoming field $u_{\mathrm{in}}(x, y, z_0)$ with a transfer function $t(x, y)$. For a nonabsorbent structure, this gives

$$u_{\mathrm{out}}(x, y, z_0) = u_{\mathrm{in}}(x, y, z_0) \cdot t(x, y) = u_{\mathrm{in}}(x, y, z_0) \cdot \exp[i \Delta\phi(x, y)], \qquad (1)$$

where $\Delta\varphi(x, y)$ is the induced phase change and is related to the refractive index structure $n(x, y, z)$ by

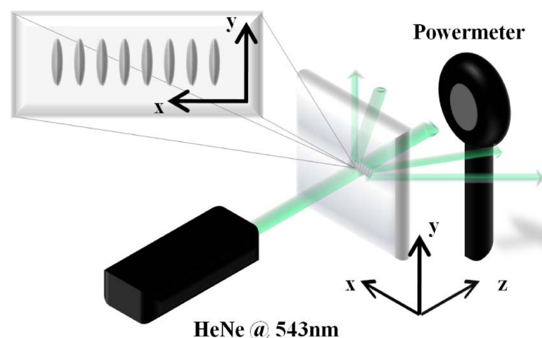

Fig. 1. (Color online) Experimental setup to measure the far-field intensity pattern caused by an induced refractive index change distribution replicated in x direction.

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$$\Delta\phi(x,y) = \frac{2\pi}{\lambda}\int_0^d \Delta n(x,y,z)\mathrm{d}z = \frac{2\pi}{\lambda}\cdot \Delta n_{\mathrm{eff}}(x,y)\cdot d. \quad (2)$$

Here, $\Delta n_{\mathrm{eff}}(x,y)$ is an effective refractive index change averaged in the propagation direction over the structure depth d. From Eq. (2), $\Delta n_{\mathrm{eff}}(x,y)$ can be determined with a knowledge of $\Delta\varphi(x,y)$ and d. The structure depth d can be deduced through microscopy of the modified region, while the induced phase change can be obtained using the Fourier relation between the element plane and the far-field distribution. Since the phase information of the far field is usually not accessible, it is not possible to reconstruct $\Delta\varphi(x,y)$ directly. In our approach an iterative Fourier transform algorithm (IFTA) [12] is applied, capable of retrieving the $\Delta\varphi(x,y)$ distribution numerically from a measurement of the far-field intensity. In general, single FLDW features cause only weak scattering and therefore marginal diffraction efficiency. It is thus advantageous to fabricate multiple replica separated with a known periodicity (p_x, p_y). Because of the Fourier relation, it leads to a discretely sampled diffraction pattern in the far field. The intensity of the corresponding diffraction orders can then easily be measured with a power meter (see Fig. 1). Eventually, the implementation into the numerical algorithm yields the refractive index distribution $\Delta n_{\mathrm{eff}}(x,y)$ averaged over the individual replica. A uniform feature replication is consequently essential in order to avoid artifacts in the reconstruction. The lateral resolution of $\Delta n_{\mathrm{eff}}(x,y)$ is limited by the number of measured orders $N\times M$ by

$$(\Delta x_{\min}, \Delta y_{\min}) \simeq \frac{2(p_x, p_y)}{(N-1, M-1)}. \quad (3)$$

To validate this concept, we have first applied it to a one-dimensional surface profile having a known refractive index distribution. The surface structure chosen consisted of a top hat profile repeated periodically to form a binary grating. This had a period of 20 μm and a duty cycle of 0.55, emulating feature sizes similar to FLDW structures. These structures were fabricated by electron beam lithography. A 950 PMMA A5 layer with a thickness of approximately 500 nm and a refractive index of 1.49 at 543 nm wavelength was spun on a sapphire glass substrate. The layer was patterned using a Leica EBPG 5000+ electron beam. To obtain an accurate reference of the refractive index distribution, the resulting surface profile was measured using an atomic force microscope capable of resolving the grating height up to a few nanometers. The surface profile, shown for one grating line in Fig. 2 (black line), was characterized by a grating height of (547 ± 4) nm and a line width of (10.8 ± 0.6) μm. Applying the Fourier method to this sample, the grating was illuminated with a 543 nm He–Ne laser. The resulting intensities in the different diffraction orders were measured using a power meter. The measured distribution was inserted into the IFTA, producing the reconstructed phase change shown in Fig. 2 (blue dashed line). This phase distribution directly matches the measured surface profile. The width of phase change (11.0 ± 0.8) μm correlates well with the AFM data. Furthermore, when the value of the phase change

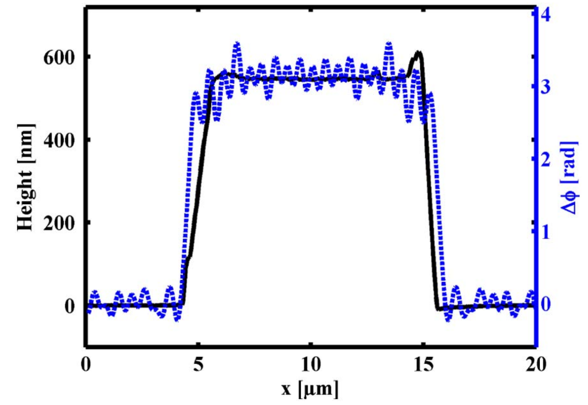

Fig. 2. (Color online) 950 PMMA A5 surface profile for one grating line measured with an AFM (solid curve) in comparison with the reconstructed phase change (dashed curve) using the Fourier reconstruction method.

(3.11 ± 0.15) rad, determined from Fig. 2, is inserted into Eq. (2), and combined with the measured grating height, it leads to a calculated change in refractive index of (0.49 ± 0.03). This agrees exactly with the difference in refractive index between that of air and of the PMMA used to fabricate the surface grating structure.

In a second demonstration of this new approach to measure Δn, FLDW structures in borosilicate glass (Corning EAGLE 2000) were analyzed. The structures comprised a parallel array of linear waveguides written into a bulk borosilicate glass sample forming a planar, one-dimensional grating. The grating lines separated with a 20 μm period were written using an amplified 1045 nm fiber laser (IMRA America μJewel D-400-VR) with pulse energies of $\sim$1.5 μJ at a repetition rate of 200 kHz, focused with a 0.4 NA objective, 225 μm below the glass surface with a translation speed of 1 mm/s.

The dimensions of the refractive index changed region, shown in Fig. 3, were measured with an optical transmission microscope. The cross section in Fig. 3(b) shows a total depth d of 40 μm and a maximum width of 10 μm. Additionally, smaller subfeatures within the modified region, such as the 2 μm wide center area, can also be observed. The varying brightness is caused by dissimilar scattering and deflection in different parts of the modified area. This observation indicates an inhomogeneous refractive index distribution within each waveguide [13]. The intensity of the far-field diffraction pattern was measured, where 43 separated orders could be distinguished.

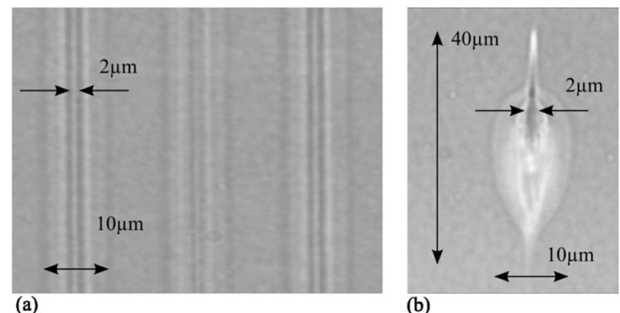

Fig. 3. Microscopic image of femtosecond-laser induced refractive index change. (a) Top view (x,y) of three grating lines and (b) cross section (x,y) of single grating line.

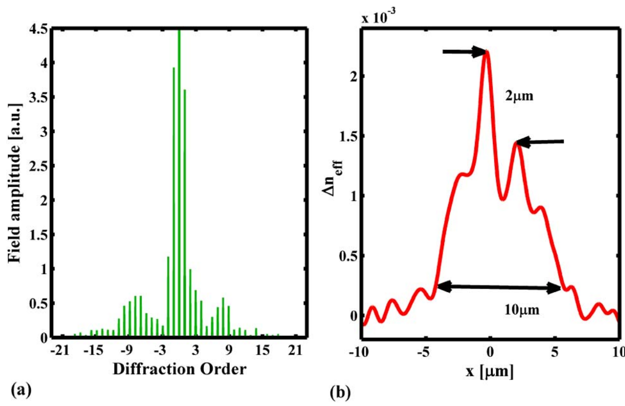

Fig. 4. (Color online) Refractive index change determination of a repeated waveguide written in borosilicate glass with a femtosecond-laser. (a) Far-field amplitude of resulting diffraction pattern using an He–Ne laser at 543 nm. (b) Determined effective refractive index change distribution, which represents an averaged induced waveguide.

The corresponding field amplitudes are shown in Fig. 4(a). In addition to the diffraction efficiency of approximately 3%, a slight asymmetry in the distribution was noticed. Since the grating was adjusted perpendicular to the incident laser beam, this indicated that the modified area itself was not perfectly symmetric. The phase change was reconstructed using the IFTA, and by applying Eq. (2), the distribution of the induced effective was determined. Here we assumed that an overall positive Δn was produced, following Eaton *et al.* [13] for similar writing parameters. Figure 4(b) shows a well-separated peak that corresponds to an average grating line with a main feature size of 10 μm matching well the optical microscope measurements in Fig. 3. According to Eq. (3), a lateral resolution of $\sim$1.0 μm was obtained. A maximum effective refractive index change of $(2.2 \pm 0.1) \times 10^{-3}$ was determined, which is of the same order as the results presented in [14]. In addition, a reduced effective refractive index modification of $(0.9 \pm 0.1) \times 10^{-3}$ in the central part of the modification was detected. Its width of 2 μm agreed well with the width of the dark subarea in the center of the cross section in Fig. 3(b). This could be referred to a region where a negative refractive index modification was induced. Measurements by Eaton *et al.* [14] showed a similar phenomenon in borosilicate glass.

Finally as a direct comparison, the grating method used by Mailis *et al.* [6] to estimate Δn was applied to the structures used here. This approach gave a value of $\sim 0.8 \times 10^{-3}$, which is close to the refractive index change in the reduced central region but less than that of the overall complex structure. The principal difference between the grating method and the Fourier reconstruction approach is the assumption of a simple sinusoidal refractive index distribution. Consequently, the grating method only provides a lateral average over the whole structure. The Fourier method we describe here is thus of added value in resolving fine differences in the refractive index change.

In conclusion, we have demonstrated a practical method of measuring femtosecond laser induced refractive index changes to an accuracy of 10^{-4} using an iterative Fourier transform technique that renders an accurate estimation of the refractive index profile with high spatial resolution across small micrometer-scale structures. No additional sample preparation is required. This method should find application in the fabrication of many two- and three-dimensional laser written refractive index structures in transparent media, including the fabrication of efficient DOEs, fiber Bragg gratings, and other photonic structures.

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