Nanograting Bragg responses of femtosecond laser written optical waveguides in fused silica glass

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Abstract: Multiple Bragg nanograting stop bands are reported for the first time in single and multi-mode optical waveguides generated by femtosecond laser direct writing in bulk fused silica glass. The stop bands observed in the guided broadband light spectra originated with the orthogonal alignment of volume nanogratings co-generated with the waveguides. Rapid shifting of stop bands across the near UV and visible spectrum was sensitively controlled by laser exposure and sample scanning direction. Bragg periods anticipated from the observed stop bands concurred with the nanograting structural pitches revealed by scanning electron microscopy. The spectroscopic characterization of nanogratings along macroscopic-scale (12.5 mm long) waveguide sections constitutes a non-destructive, convenient and sensitive approach to examine long-range order and uniformity of the self-organized periodic structures that may assist to unravel the laser-glass interaction physics of nanograting formation.

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References and links


1. Introduction

The formation of ordered ripples on surfaces irradiated with intense laser light was discovered shortly after the invention of the laser [1]. Such laser induced periodic surface structures (LIPSSs) [2] constitute a universal phenomenon observed in a wide range of materials such as semiconductors, metals and dielectrics when subjected to intense cw or pulsed laser exposure [2–7]. Several decades later, a similar phenomenon represented by the formation of volume nanogratings was reported inside transparent fused silica glass when irradiated with a tightly focused femtosecond laser beam [8]. Like LIPSSs, volume nanogratings possess periods (~100 to 300 nm) scaling with $\lambda_d/2n$, where $\lambda_d$ is the laser wavelength and $n$ the material refractive index. The volume nanogratings are invariably aligned orthogonal to the writing laser polarization and can be formed in the laser focal volume [8–11] extending over long lengths [12,13] by scanning the glass sample during laser irradiation.
Laser-generated volume nanogratings impose a strong form birefringence that has underpinned new approaches in developing birefringent optical waveguides [14,15], polarization selective diffractive optical elements [16], buried optical waveplates [17], rewritable data recordings [12,18] and micro/nanofluidic channels generated with preferential chemical etching [19–23]. While volume nanogratings offer such intriguing prospects for creating highly functional three-dimensional photonic and optofluidic microsystems, insight into the femtosecond laser-glass interaction physics underlying nanograting formation is not yet adequate, centering mainly on the two proposed models of plasmon and light wave interference [8] and plasmononic self-organization [9,12]. These fundamental studies have relied principally on morphological characterization of the volume nanogratings, encompassing tools such as backscattering [8] or secondary [9–11] electron scanning electron microscopy (SEM), Auger electron spectroscopy [8], or atomic force microscopy [12]. All these methods are destructive and require precise grinding and polishing of the glass sample into the nanograting volume, which frequently is followed with chemical etching to generate a nanograting surface morphology that enhances the characterization contrast. Alternatively, Kazansky and associates applied optical reflection spectroscopy to examine an assembly of femtosecond laser modification tracks inside fused silica glass. Two reflection bands around 460 nm and 835 nm were reported and inferred as Bragg reflections from a self-organized periodic refractive index modification nanostructure [24] that was later verified by SEM imaging [8].

In this paper we introduce a new diagnostic approach to spectrally characterize volume nanogratings when formed into optical waveguides where we take advantage of the high localization of the probing guided mode(s) over the nanograting volume. This non-destructive spectroscopic approach enables definitive and highly sensitive optical characterization to examine the long-range coherence and spectral shifting of the self-organized nanograting structures when formed over varying laser writing conditions. Such spectral characterization may improve our understanding of the laser interaction physics and advance the development of photonic and micro/nano-fluidic devices based on such embedded nanograting structures.

2. Fabrication and characterization

An amplified femtosecond fiber laser (IMRA America, µJewel D-400-VR) with 1045-nm wavelength and 1-MHz repetition rate was frequency-doubled to 523-nm wavelength with 200-fs pulse duration to write the nanograting embedded optical waveguides in fused silica glass (Corning 7980, 12.5 mm wide and 1 mm thick). Laser modification tracks were formed by transverse scanning across the whole sample width with the beam focused at a 150-μm depth via a 0.55-NA aspheric lens (New Focus, 40×, 5722-A-H). The objective was to simultaneously form strong and uniform nanograting tracks and strong optical waveguides in the visible spectrum. This combination was found possible for laser exposure in the range of 30 to 150-nJ pulse energy and 0.1 to 1.2-mm/s scanning speed. In order to observe Bragg stop bands, the laser polarization was aligned along the sample scanning direction to orient the nanograting planes perpendicular to the waveguide. The scanning directional dependence potentially arising from the quill or non-reciprocal writing effect [25–27] was also examined by forming waveguides along the orthogonal ± X and ± Y sample scanning directions. These effects have been associated with pulse front tilt [25,28] coming from a non-ideally aligned laser compressor, which aligns here with the ± Y sample scanning direction.

Figure 1 illustrates the schematic arrangement for the spectral characterization of volume nanograting-embedded waveguides. The output of a multiple-LED light source (Mightex Systems, FCS-0001-000) was coupled into an optical fiber (Corning SMF-28) and butt-coupled to the entrance facet of the laser modification tracks while a second optical fiber (Corning SMF-28) butt-coupled to the opposite facet collected the transmitted light for recording transmission spectra in an optical spectrum analyzer (Ando, AQ-6315A). All transmission spectra were normalized to the source spectrum recorded by direct fiber-to-fiber
coupling. Index matching oil was used at all fiber couplings to reduce Fresnel loss. Bragg reflection spectra could not be recorded with a free space beamsplitter arrangement due to the low spectral brightness (~20 dBm/nm) and high insertion loss (~40 dB) for coupling the multi-mode source light into the present single or low-mode waveguides.

![Figure 1. Schematic arrangement for transmission spectral characterization of nanograting embedded waveguides written in fused silica glass.](image)

3. Results and discussions

For increasing laser exposure in the 30 to 150-nJ pulse energy and 1.2 to 0.1-mm/s scanning speed ranges, the laser modification tracks showed increasing refractive index contrast under an optical microscope. However, nanograting periods anticipated in the ~100 to 250 nm range were below the present optical resolution limit (> ~500 nm). Waveguiding in both the visible (635 nm) and infrared (1550 nm) spectrum was observed from tracks formed above an exposure threshold of ~30 nJ.

Figure 2 shows the normalized transmission spectra of waveguides formed with –Y (a-d, black traces), + Y (d, blue trace), + X (e-h, black traces), and –X (h, blue trace) directional scanning at 0.25-mm/s (a-c, e-g) and 0.1-mm/s (d, h) speed and 75-nJ (a, e), 50-nJ (b, d, f, h), and 30-nJ (c, g) laser pulse energy. For –Y-scanning, a moderately strong (~10 dB) and broad (~50 nm at 3-dB bandwidth) stop band peaked at 480 nm and classified as the primary band (Fig. 2(c), faint red bar highlighted) is clearly seen for the waveguide formed with the lowest 30-nJ pulse energy and 0.25-mm/s scanning speed. At a higher exposure of 50 nJ and the same scanning speed, two weaker (~5 dB) stop bands were observed (Fig. 2(b)) at 540 and 730 nm, classified as the secondary (cyan bar) and tertiary (green bar) stop bands, respectively. These bands have similar 3-dB bandwidth (~50 nm), and are seen to red-shift with decreasing scanning speed (0.1 mm/s) to 570 and 770 nm (Fig. 2(d)), respectively, with a 2.5-fold slower scanning speed of 0.1 mm/s. Although waveguides were strongly guiding when formed at 75-nJ or higher pulse energy, Bragg stop bands could not be definitively identified (Fig. 2(a)), possibly owing to extremely broad stop bands that arise from a less coherent organization of the nano-periodic structures. The observed spectra showed a waveguide insertion loss of 8 to 15 dB near λ = 800 nm wavelength, which generally followed the expected 1/λ^4 scaling of Rayleigh scattering loss that was represented by the red dashed lines calculated for each spectrum. These scattering loss baselines then revealed highly asymmetric stop bands existing for all the observed Bragg resonances in Fig. 2.

For waveguides formed along the orthogonal + X direction, only a single similarly broad and weak (~3 dB) Bragg resonance, classified as a tertiary stop band (green bar), was observed at 690 nm for 50-nJ pulse energy and 0.25-mm/s scanning speed (Fig. 2(f)). This stop band likewise was found to red-shift with decreasing scanning speed (0.1 mm/s) to 730 nm (Fig. 2(h)). No Bragg stop bands were discernible at both 75-nJ and 30-nJ pulse energy as seen in Figs. 2(e) and 2(g). Further, the insertion loss of the waveguide formed at 30-nJ pulse...
energy (Fig. 2(g)) deviated from the $1/\lambda^4$ Rayleigh scaling above $\sim650$ nm, implying a weak laser refractive index modification at this low laser exposure.

A comparison of spectra from waveguides formed along the $\pm X$ (Fig. 2(h)) and $\pm Y$ (Fig. 2(d)) directions, with identical laser pulse energy (50 nJ) and scanning speed (0.1 mm/s), showed an insignificant difference in loss or the position of the secondary and tertiary stop bands for reversal along the $Y$ direction (Fig. 2(d)). In contrast, the $\pm X$ direction reversal
yielded a ~20-nm stop band shift and a large difference (~2 to 15 dB) in the waveguide insertion loss. These band shifting trends were observed over a broad range of laser exposure conditions, as summarized in Fig. 3, where all discernible Bragg stop band positions found in the 30 to 50 nJ processing window were recorded as a function of sample scanning speed. All primary, secondary and tertiary stop bands blue-shifted with faster scanning speed or effectively less laser exposure, regardless of the ±X or ±Y scanning directions. However, for the case of tertiary bands, the stop band positions were much more sensitive (Δλ_B ≈ 50-100 nm) to orthogonal changes (X↔Y) in scanning direction than simply to reversal (±X or ±Y) of scanning direction (Δλ_B < 20 nm) over a large scanning speed range from 0.1 to 1.2 mm/s.

Fig. 3. Bragg stop band peak wavelength observed as a function of sample scanning speed from nanograting waveguides formed at the labeled pulse energy and + (red) or – (black) scanning directions.

To definitively connect the Bragg spectral response with the underlying nanograting structure formed inside the glass, samples were ground and polished to cross longitudinally at ~1° tilt angle with the laser modified tracks and thus reveal the nanograting structure across the whole waveguide depth when followed with chemical etching (1% HF for 2 minutes). The SEM images in Fig. 4 were selected from within the waveguide to reveal the highest contrast and best ordered nanograting structure for writing conditions of 75-nJ (a), 50-nJ (b) and 30-nJ (c) pulse energy and 0.25-mm/s scanning speed along –Y scanning direction, and of 50-nJ pulse energy and 0.25-mm/s scanning speed along the +X scanning direction (d). For –Y scanning, high-contrast nanograting structure is clearly visible, showing a widening (from ~0.5 μm to ~1.5 μm) but less ordered structure developed for pulse energy rising from 30 to 75 nJ. At 75 nJ (Fig. 4(a)), the grating planes became curved, discontinuous and less ordered, thus possessing a broader distribution of spatial frequencies that may be associated with an overly broadened and weakened Bragg stop band that could not be definitively identified in Fig. 2(a). A better ordered nanograting structure was observed at 50 nJ (Fig. 4(b)), where discernible alignments of ~125-nm and ~145-nm periodic components were illustrated by the green and red side scale, respectively, predicting Bragg stop bands at ~365 and 423 nm (λ_B = 2n_eff Λ), respectively, which are outside or at the short wavelength limit of spectral sensitivity (Fig. 2(b)). An effective refractive index of n_eff = 1.46 was inferred for the waveguides by
scaling \( n_{\text{eff}} \) values measured at \( \sim 1.55 \, \mu m \) from Bragg grating waveguides [29] to the visible spectrum with Sellmeier’s equation. There is also discernible alignment of the grating lines on \( \sim 165\text{-nm periodicity} \) (yellow side scale) for the 30-nJ pulse energy case in Fig. 4(c) for which an inferred Bragg stop band of 482 nm was found to closely match the observed primary stop band in Fig. 2(c). Figures 4(b) and 4(d) highlighted the significant morphological change in simply shifting from \(-Y\) to \(+X\) scanning direction, for otherwise identical exposure of 50-nJ pulse energy and 0.25-mm/s scanning speed. The \(+X\) direction scanning yielded a wider waveguide with shorter grating period (\( \Lambda \approx 118 \) nm) predicting a blue-shifted primary stop band to exist at 345 nm that was outside the current detectable spectral range.

For a better quantitative correlation of the Bragg spectral response with the nanograting structure, spatial frequency (\( f \)) spectra were generated by fast Fourier transform (FFT) calculation from the grayscale profiles of the SEM nanograting waveguide images (Fig. 4). One example of such FFT spectra was plotted in Fig. 2(b) (blue dashed trace) to match the wavelength scale (\( \lambda = 2n_{\text{eff}}/f \)) of the transmission spectrum (black trace) recorded from nanograting waveguides formed along \(-Y\)-scanning direction at 50-nJ pulse energy and 0.25-mm/s scanning speed. The strong single \( \sim 425 \) nm FFT peak matches with a deeply attenuating optical band seen below \( \sim 470 \) nm, indicating a primary stop band (illustrated by the faint red bar in Fig. 2(b)). However, the random overlay of multi-period nanogratings and short-range (inferred long-range as well) disorder as seen in the SEM images (Fig. 4) also manifested in FFT spectra that were frequently too noisy to allow definitive assignments of the weak secondary and tertiary Bragg stop bands at all the exposure conditions. The maximum FFT peak was seen to vary from \( \sim 425 \) nm for the 50-nJ and 0.25-mm/s waveguides to \( \sim 485 \) nm for the 30-nJ and 0.25-mm/s waveguides (Fig. 2(c)), which infers blue shift of the primary stop band with increased writing laser pulse energy. Coinciding with the much shorter nanograting period (Fig. 4(d)) for \( \pm X\)-scanning waveguides, no definitive FFT peaks were found in the visible spectral range. The observed stop bands for \( \pm X\)-scanning waveguides are thus assigned as the tertiary stop bands blue-shifted relative to the \( \pm Y\)-scanning nanograting stop bands assuming the same trend as for the primary stop bands. The secondary and tertiary stop bands for \( \pm Y\)-scanning waveguides as well as the tertiary stop bands for \( \pm X\)-scanning waveguides are thus assigned as the tertiary stop bands blue-shifted relative to the \( \pm Y\)-scanning nanograting stop bands assuming the same trend as for the primary stop bands.
waveguides distinctly resolved in the lower loss spectral range ($\lambda > \sim 500$ nm) of the waveguides were likely associated with nanograting components ($\Lambda = 190$ to 270 nm) appearing hidden as weaker overtone periods obscured by the primary periodic structures identified by SEM as shown in Fig. 4.

The observation of multiple discernible Bragg stop bands presents a view of the self-organization of nano-structures with limited coherence that can be tuned by the laser pulse energy, scanning speed and scanning direction. The assigned stop bands manifest blue-shifting with increasing sample scanning speed or increasing laser pulse energy. A larger structural disorder seen by SEM followed with an observed broadening of the stop band when waveguides were formed above 50-nJ pulse energy. Primary and secondary stop bands are further predicted to exist in the UV spectrum for $\pm X$ scanning that lie outside the strong guiding region of our present waveguides and below the current characterization spectral limit. Overall, the multi-band observations here are consistent with the existence of both a strong and weak Bragg reflection band reported by Kazansky and associates at 460 and 835 nm, respectively, which were connected with fundamental and higher periodicity nanograting components [24].

By associating a decreasing scanning speed with increasing electron density and temperature, the light and plasmon wave interference model [8] concurs with the present trend of stop bands red-shifting with decreasing sample scanning speed (Fig. 3), but is in contradiction with the blue-shifts observed with increasing laser pulse energy. Further, the widely shifting stop bands found here cannot follow the nanoplasmonic self-organization model [9,12] that predicts no dependence of nanograting period with laser exposure.

The stop band positions were found to shift only slightly ($\Delta \lambda_B < 20$ nm) in Fig. 3 with reversal of scanning directions ( $\pm X$ or $\pm Y$). For the tertiary band, larger band shifts were observed for the $\pm X$ directions, which are expected to have little pulse front tilt contribution in contrast with $\pm Y$ directional changes. Further, much stronger band shifts of 50-100 nm were observed (Fig. 3) for 90° directional changes ( $\pm X \leftrightarrow \pm Y$). These observations suggest that factors other than pulse front tilt, such as asymmetric beam shaping, may be contributing more significantly to a new type of orthogonal quill effect.

The spectral profile found for all Bragg stop bands were strongly skewed to longer wavelength, indicating that waveguide radiation mode coupling to shorter wavelengths was not a key factor for the asymmetric spectral band shape. The broad ~50-nm wide stop bands are therefore expected to follow a wide and skewed distribution of nanograting periods with limited long range coherence of $\Delta \Lambda = \pm 10$ nm that provided only moderately strong (~10 dB) stop bands in contrast with ~40 dB strong and ~0.1 nm narrow stop bands found in precisely ordered fiber Bragg gratings [30]. Further study to examine the underlying mechanisms of forming such bulk nanostructures in glass is necessary to improve the order of their self-organization and possibly enable stronger and narrower Bragg responses.

4. Summary

In conclusion, volume nanogratings in fused silica glass were characterized for the first time by means of transmission spectroscopy through optical waveguides comprising the nanogratings that were formed simultaneously with the waveguides. Multiple Bragg stop bands were observed in the visible spectrum for waveguides formed in a narrow laser exposure window (30 to 50-nJ laser pulse energy). Rapid shifting of all stop bands was found with changes of writing laser pulse energy, scanning speed and direction, including a new orthogonal quill effect. The spectroscopic approach reported here offers convenient, non-destructive and highly sensitive characterization of volume nanogratings, which, with further laser exposure optimization, may lead to formation of more coherently self-organized nanogratings for applications such as optical sensing, filtering and phase control through optical circuits.
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