Refractive index modulation in media with saturable photosensitivity

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Abstract: Permanent change of the refractive index of photo-thermo-refractive glass can be achieved by exposure to ultraviolet light followed by thermal development. The dependence of the change on the dosage is nonlinear because the photochemical process of photosensitivity involves saturation. With a holographic recording of Bragg gratings inside glass, a sinusoidally modulated dosage is imprinted into distorted modulation of the refractive index. We investigate this distorted modulation and derive analytical expressions for the amplitudes of its spatial harmonics.

1. Introduction

Photosensitive materials such as photo-thermo-refractive (PTR) glass [JL1] have become the material of choice over the past twenty years for the manufacturing of resonant diffractive optical elements such as volume Bragg gratings (VBGs) [JL2,JL2a]. They have a wide range of applications that include the improvement of the performance of high-power lasers [JL3,JL3a]. For uses ranging from mode-locking to beam combining [JL4,JL4a], they have proven to be efficient components and can withstand very high energies. Despite the maturity of this technology, achieving high diffraction efficiency remains a challenge, which has been addressed by improving both the glass technology and the recording process. The fabrication process of VBGs begins with the initial ultraviolet (UV) exposure of a glass blank with periodically modulated intensity produced by a holographic recording. Subsequent thermal development converts the exposure pattern into refractive index modulation (RIM). The typical amplitude of RIM required for high reflectivity of a narrow-band VBG is a few hundred ppm (part per million), and it is developed in the regime of the linear photosensitivity of glass. The photosensitivity saturates at a high exposure dosage, as it follows a rational type function, and the refractive index change (RIC) reaches its maximum value of approximately two thousand ppm [Lumeau13induced]. Although the maximum RIC of one thousand ppm can be achieved in uniformly exposed PTR glass, changes of twice this magnitude can be observed in VBGs. While the origin of these larger changes remains unclear, the long-term investigation on theses glasses has shown that in the first approximation, the kinetics of the RIC in VBGs is similar to that of uniformly exposed PTR glass with an amplitude twice as large. A discussion of these processes and the associated mechanisms can be found in the literature [LumeauPTRReview].

Among the various types of VBGs, chirped Bragg gratings (CBG) with linearly varying modulation periods are used to stretch and compress short laser pulses with a relatively wide spectral range [JL6]. In this case, the RIM amplitude should be a maximum since the monochromatic components of the pulse spectrum are reflected over a short length of matched resonant conditions inside the CBG. Based on experimental observations, it has been shown that the RIM amplitude of VBGs with constant period does not increase monotonically with dosage, but instead decreases slowly after the maximum value is reached [Lumeau13diffraction]. Similar effects will happen in CBGs and will limit the maximum diffraction efficiency that can achieved in these components and will have direct impact in chirped pulse amplification systems. In this report, we analyze this effect caused by the specifics of the formation of an imprinted periodic pattern of a spatially modulated refractive index. There are parasitic side effects that cause a decrease in the effective amplitude of the RIM with the increase of the dosage. An example is the reduction of the visibility of the dosage interference fringes over a long exposure time due to instabilities in the setup for holographic recording. Another is the increase of material absorption in overexposed glass specimens that leads to the degradation of the quality of the fabricated Bragg gratings. However, these effects will not be considered here. Our objective is to determine the optimum value of the dosage that provides the maximum amplitude of effective Bragg modulation of the refractive index.

2. Holographic recording of Bragg grating in photo-thermo-refractive glass

In the scheme of the holographic recording of VBGs, a flat-top laser beam with a wavelength λ_{UV} is split into two recording beams of the same intensity I_0, which overlap at the mutual angle 2α and form an interference pattern with a spatial period Λ. The exposure of a glass specimen to this interference pattern for a time duration T produces spatial modulation of the UV-dosage $D(z)$:

$$D(z) = D_0\left(1-\cos(Qz)\right), \quad D_0 = 2I_0 T\cos\alpha, \quad Q = \frac{2\pi}{\Lambda}, \quad \Lambda = \frac{\lambda_{\mathrm{UV}}}{2\sin\alpha}, \tag{1}$$

where Q is the Bragg wavevector, and D_0 is the amplitude of the dosage modulation [JL7??].

Figure 1 illustrates the holographic recording scheme and the modulation of the dosage in one direction.

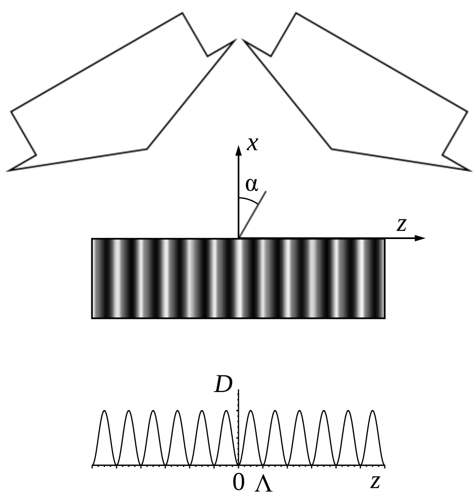

Fig. 1. Holographic recording of Bragg grating with two overlapping coherent UV waves. Modulation of dosage.

The dependence of the RIC Δn on the UV dose D can be approximated using a rational function with good accuracy [Lumeau13induced] as follows:

$$\Delta n(D) = -\nu\frac{D}{D+H}: \quad \Delta n(D=H) = -\tfrac{1}{2}\nu, \quad \begin{matrix} D \ll H: & \Delta n(D) \approx -\nu\, D/H\,, \\ D \gg H: & \Delta n(D) \approx -\nu + \nu\, H/D. \end{matrix} \tag{2}$$

The characteristic parameters of this function are its asymptotic value ν and specific dose H, setting the RIC equal to half of ν. The typical value for ν is 2000 ppm. The minus sign describes the negative photosensitivity of the PTR glass observed in experiments. This indicates that the refractive index decreases in areas of the glass exposed to UV radiation. At small dosages, $D << H$, the RIC depends linearly on the dosage as $\Delta n(D) \approx -\nu D/H$, according to Eq. (2). The actual value of the dose parameter H depends on the procedural parameters of the subsequent thermal development of a glass specimen [Lumeau13induced]. The permanent RIC then becomes imprinted in the PTR glass. Typically, $H = 0.2$ J/cm^2.

For the holographic recording of the Bragg gratings, the dose $D(z)$ is spatially modulated with a period Λ, as shown in Eq. (1). Modulation of the dosage leads to a corresponding periodic modulation of the refractive index according to Eq. (2) as:

$$n(z) = n_0 + \Delta n\big(D(z)\big). \quad (3)$$

Here, n_0 is the background refractive index of the unexposed glass.

On the left side of Figure 2, the saturation curve for Eq. (2) is shown with the dimensionless values $\Delta n/\nu$ and D/H plotted on the vertical and horizontal lines, respectively. On the right side, the corresponding dimensionless distorted modulations of the refractive index over one period Λ are shown for four dosage modulation amplitudes D_0 expressed through the characteristic dosage H as $D_0 = H/4$, H, $4H$, $16H$.

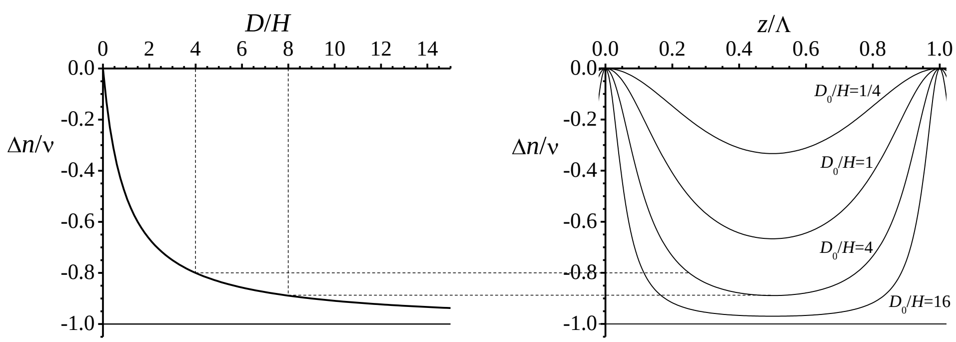

Fig. 2. Dependence of the RIC on dosage and one period of the RIM for different values of D_0/H.

The profiles of the modulation of the refractive index becomes more distorted, away from a cosine shape with the increase of the amplitude of dosage D_0. This indicates that spatial harmonics of higher orders emerge in the modulation profiles.

3. Amplitudes of the harmonics of the refractive index modulation

The RIC modulated with Λ-periodicity can be represented by a series of harmonics as:

$$\Delta n\big(D(z)\big) = -\nu \frac{D(z)}{D(z)+H} = \Delta n_0(D_0) + n_1(D_0)\cos(Qz) + n_2(D_0)\cos(2Qz) + \mathrm{K} \quad (4)$$

For the rational functional dependence of Δn on D in Eq. (1), we derived analytical expressions for the first three dose-dependent coefficients in Eq. (4). The background refractive index shift Δn_0, the amplitude of the Bragg modulation n_1, and the amplitude of the emerged second harmonics are obtained as follows:

$$\Delta n_0(D_0) = \Lambda^{-1}\int_0^{\Lambda} \Delta n(D(z))dz = \nu\left(\frac{1}{\sqrt{1+2u}} - 1\right) = -u + O(u^2) < 0, \quad u = \frac{D_0}{H},$$
$$n_1(D_0) = 2\Lambda^{-1}\int_0^{\Lambda} \Delta n(D(z))\cos(Qz)dz = \frac{2\nu}{u}\left(\frac{1+u}{\sqrt{1+2u}} - 1\right) = u + O(u^2) > 0, \quad (5)$$
$$n_2(D_0) = 2\Lambda^{-1}\int_0^{\Lambda} \Delta n(D(z))\cos(2Qz)dz = 2\nu\left(\frac{1+4/u+2/u^2}{\sqrt{1+2u}} - \frac{2}{u} - \frac{2}{u^2}\right) = \tfrac{1}{2}u^2 + O(u^3) > 0.$$

For a small modulated dosage, the background refractive index shift Δn_0 and the amplitude of the Bragg modulation exhibit linear dependence, whereas the amplitude of the second harmonics exhibits a quadratic growth. Figure 3 shows $-\Delta n_0$, n_1, and n_2, depending on the dimensionless parameter of the dosage $u = D_0/H$.

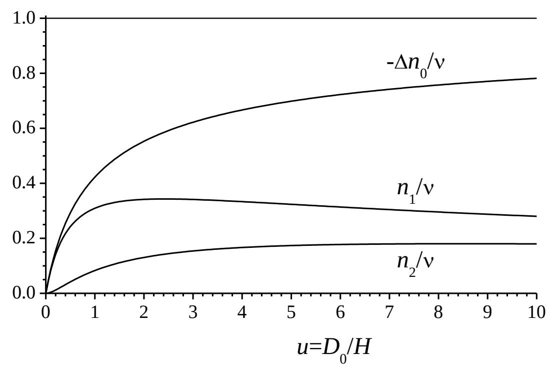

Fig. 3. Dosage dependence of the first expansion coefficients in Eq. (4).

The maximum value of n_1 and the required dosage can be found analytically from Eq. (5):

$$dn_1/du = 0 \quad \to \quad D_{n1,\max} = (1+\sqrt{2})H \approx 2.41H, \quad n_{1,\max} = (6-4\sqrt{2})\nu \approx 0.343\nu. \quad (6)$$

It was determined that $n_{1,\max}$ reaches approximately 1/3 of the maximum value ν for the saturation law represented by Eq. (2), see Fig. 3. The corresponding required dosage is equal to approximately 2.5 times H, according to Eq. (6).

The simple rational function $\Delta n(D)$ in Eq. (2) that is used to describe the multiphysics process of the saturation of photosensitivity was found to be very efficient for the accurate fitting of a wide range of experimental data. However, most studies of photosensitivity generally implement the well-known exponential functional dependence since it can approximate the saturation curve in the range of practical values of the dosage. Different functional types of saturation curves have been investigated numerically in [Kaim15].

The exponential functional dependence of the RIC on the dosage is as follows:

$$\Delta n_{\mathrm{E}}(D) = -\nu\left(1 - \exp(-D/E)\right):$$
$$\Delta n_{\mathrm{E}}(D = H_{\mathrm{E}}) = -\tfrac{1}{2}\nu, \quad H_{\mathrm{E}} = E\ln 2; \quad D << E: \quad \Delta n_{\mathrm{E}}(D) \approx -\nu\, D/E. \quad (7)$$

This curve of negative photosensitivity has the same saturation value –ν. The RIC equal to half of this value is reached at $D = H_{\mathrm{E}}$.

The periodically modulated dosage from Eq. (1) will produce the corresponding distorted modulation of the refractive index, similar to Eq. (4)

$$\Delta n_{\mathrm{E}}\left(D(z)\right) = -\nu\left(1 - e^{-D(z)/E}\right) = \Delta n_{\mathrm{E}0}(D_0) + n_{\mathrm{E}1}(D_0)\cos(Qz) + n_{\mathrm{E}2}(D_0)\cos(2Qz) + \mathrm{K} \quad (8)$$

In this case, the coefficients of the expansion are the following

$$\begin{aligned} \Delta n_{\mathrm{E}0}(D_0) &= \nu\left(-1+e^{-s}I_0(s)\right) = -s+O(s^2)<0, \quad s = D_0/E, \\ n_{\mathrm{E}1}(D_0) &= 2\nu e^{-s}I_1(s) = s+O(s^2)>0, \\ n_{\mathrm{E}2}(D_0) &= 2\nu e^{-s}I_2(s) = \tfrac{1}{4}s^2+O(s^3)>0. \end{aligned} \tag{9}$$

Here, $I_m(s)$ is the modified Bessel function of integer order m.

The derived expansion coefficients have dependencies that are very similar to those shown in Fig. 3. The amplitude of the Bragg modulation n_{E1} reaches its maximum at a dosage of $D_{n\mathrm{E1,max}}$

$$dn_{\mathrm{E}1}/ds = 0 \quad \rightarrow \quad D_{n\mathrm{E1,max}} \approx 1.545E \approx 2.23H_{\mathrm{E}}, \quad n_{\mathrm{E1,max}} \approx 0.438\nu. \tag{10}$$

This achievable value of the Bragg modulation amplitude is slightly higher than that derived for the case of rational functional dependence.

To conclude, the derived analytical results presented in Eqs. (5,6) and (9,10) provide a clear understanding of the dependencies of amplitudes of the harmonics of the RIM on the dosage.

4. Summary

The periodic profile of the modulation of refractive index developed at high exposure dosages is distorted from cosine one because of the saturability of the photosensitivity. We derived analytical expressions for the efficient amplitudes of refractive index modulation depending on the dosage. The effective value of the modulation determines the resonant Bragg reflection of incident laser beams, and its maximum value is achieved at a specific dosage. The obtained results are beneficial for the implementation of appropriate exposure regimes during the holographic recording of Bragg gratings.

Disclosures

The authors declare no conflicts of interest.

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