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Angular responses of the first and second diffracted orders in transmission diffraction grating recorded on photopolymer material

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Abstract: Some of the theoretical models in the literature describing the mechanism of hologram formation in photopolymer materials predict the existence of higher harmonics in the Fourier expansion of the recorded refractive index. Nevertheless, quantitative information is only obtained for the first harmonic of the refractive index using Kogelnik’s Coupled Wave Theory. In this work we apply the Rigorous Coupled Wave Theory to demonstrate that when recording phase diffraction gratings in PVA/acrylamide photopolymer materials, a second order grating is also recorded in the hologram even when the material is exposed to a sinusoidal interference pattern. The influence of this second order grating on the efficiency of the first order for replay at the first on-Bragg angular replay condition is studied and the size of the 2nd harmonic examined.

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References and links
1. Introduction

Photopolymers are attractive materials for the recording of volume phase gratings. The advantages of these materials are basically that they do not require wet processing, their stability and their relatively low cost. In addition, these materials allow obtaining high diffraction efficiencies (if Fresnel losses are taken into account up to 100% can be obtained). The search for optimum chemical formulations to improve the quality of the final polymer materials has yielded a great deal of experimental work, which can be found in the extensive literature [1,2]. Nonetheless, although the general picture is understood, there is a lack of quantitative data that clarify the mechanism of hologram formation inside photopolymer materials. Only a few attempts have been made to provide clear information of what is going on inside the polymer material [3-9]. Among these publications, the diffusion based model proposed by Sheridan et al. [9-11] appears to provide a useful description of the mechanism of hologram formation. For instance, it predicts the cut-off of diffraction efficiency for high spatial frequencies. This cut-off had been observed experimentally in photopolymer materials, but apart from Sheridan’s model none of the diffusion models available in the literature appear to predict this behaviour. This model and almost the great majority of the others, predict the existence of higher harmonics in the refractive index recorded on the material. However, with exception of the approximate analysis of Zhao et al. [12], there is no quantitative information regarding diffraction by these higher harmonics.

The lack of quantitative data with respect to higher harmonics in the refractive index is basically due to the fact that the material models are usually checked using Kogelnik’s Coupled Wave Theory [13]. By fitting the temporal evolution response (growth curve) of the diffraction efficiency at first on-Bragg angular replay condition, information about the first harmonic of the refractive index can be obtained. But, to obtain information about the higher harmonics in the refractive index the Rigorous Coupled Wave Theory (RCW) is needed. A difficulty associated with the original method proposed by Moharam and Gaylord [14] to solve the coupled wave equations, is that for high values of the grating thickness numerical unstabilities emerge. This issue originally prevented researchers using the RCW method for
high thickness polymer materials. However this problem was overcome by the same authors with the formulation of a stable and efficient implementation of the RCW analysis of binary gratings [15], which can also be extended to deal with unslanted gratings. This numerical reformulation allows the retention of high values of thickness and many orders exact boundary conditions, and thus provides the theoretical framework to check the validity of the material models proposed in the literature.

In this work we make use of our implementation of the RCW method to study the influence of the second Fourier component of the refractive index on the efficiency of the first order grating at first on-Bragg angular replay condition. It is clearly demonstrated that a second order Fourier component of the refractive index is formed in phase diffraction gratings recorded in a PVA/acrylamide photopolymer material. Fitting manually the angular response, centered at second on-Bragg angular replay condition, of the diffraction efficiency of the second order, quantitative information about the second Fourier component of the refractive index stored in the grating is also obtained.

2. Theoretical diffraction efficiency

In this section we briefly review the RCW for the case of lossless sinusoidal transmission diffraction grating. For more details see Ref. [15].

In this case the Fourier expansion of the relative permittivity in the hologram is expressed as:

\[ \varepsilon(x, z) = \sum_{h} \varepsilon_h \exp[jhK \cdot r] \]  

(1)

\( \varepsilon_h \) is the \( h \)th Fourier component of the relative permittivity in the grating region and \( K \) is the grating vector, the magnitude of which is related to the grating period, \( \Lambda \), as follows:

\[ |K| = \frac{2\pi}{\Lambda} \]  

(2)

In the unslanted geometry expression (1) can be converted into:

\[ \varepsilon(x) = \sum_{h} \varepsilon_h \exp[jhKx] \]  

(3)

where it is assumed that the periodic relative permittivity varies in the \( x \) direction, parallel to the grating boundaries. The waves are assumed to propagate in the \( xz \) plane, with the electric field polarized in the \( y \) direction, so that TE polarization is studied.

It is further assumed here that the hologram (medium 2) is embedded between two homogeneous media, 1 and 3. If a plane wave impinges onto the hologram from medium 1, the electric field in mediums 1 and 3 will be expressed as:

\[ E_i = \exp\{- j(k_{i0}x + k_{i0}z)\} + \sum_{j} R_j \exp\{- j(k_{j0}x - k_{j0}z)\} \]  

(4)

\[ E_i = \sum_{j} T_j \exp\{- j(k_{j0}x - k_{j0}(z-d))\} \]  

(5)

we define \( k_{i0} = k_0 \varepsilon_i^{1/2} \sin \theta \), \( k_{j0} = k_0 \varepsilon_j^{1/2} \cos \theta \), \( k_0 = 2\pi/\lambda_0 \) where \( \theta \) is the angle of incidence in medium 1, \( \lambda_0 \) is the free-space wavelength, \( \varepsilon_i \) is the relative permittivity of medium 1, \( R_i \) is the amplitude of the \( i \)-th order backward-diffracted wave and \( T_i \) is the amplitude of the \( i \)-th order forward-diffracted wave. \( k_{j0} \) is determined from the vector Floquet condition:

\[ k_{j0} = k_{i0} - iK \]  

(6)

where the \( z \) components of the propagation vectors for the \( i \)-th orders are:

\[ k_{j0} = (k_{j0}^2 \varepsilon_i - k_{j0}^2)^{1/2} \quad 1 = 1,3 \]  

(7)

In the grating region the tangential electric (\( y \) component) and magnetic (\( x \) component) fields may be expressed as:

\[ E_{zj} = \sum_{j} S_{zj}(z) \exp(-jk_{j0}x) \]  

(8)
\[ H_{z_1} = - j \left( \frac{\varepsilon_r}{\mu_r} \right)^{1/2} \sum U_{i_1}(z) \exp(-jk_{_1}x) \]  

(9)

By substituting Eqs. (8) and (9) with Eq. (3) into Maxwell’s equations the following set of first order coupled equations can be derived:

\[ \frac{\partial S_u}{\partial z} = k_0 U_u \]  

(10)

\[ \frac{\partial U_u}{\partial z} = \left( \frac{k_0}{k} \right)^2 S_u - k_0 \sum_p \varepsilon_{(p)} S_{ip} \]  

(11)

The amplitudes \( S_u \) and \( U_u \), can be obtained after solving Eqs. (10) and (11) by using the formalism described in Ref. [15]. The 2xN arbitrary constants that arise from the 2xN coupled Eqs. (10), (11), where N is the number of orders retained, can be obtained by matching the tangential electric and magnetic field components at the two boundaries. Imposing the boundary conditions at the 1-2 and 2-3 interfaces permits the amplitudes of the backward waves, \( R_i \), and of the forward waves, \( T_i \) to be calculated.

The diffraction efficiencies for the different orders are expressed as:

\[ \eta_i^r = R_i R_i^* \Re \left( \frac{k_{ro}}{k_{ro}} \right) \]  

(12)

\[ \eta_i^i = T_i T_i^* \Re \left( \frac{k_{ro}}{k_{ro}} \right) \]  

(13)

3. Influence of the second Fourier component of the refractive index on the efficiency of the first order at first on-Bragg angular replay condition.

In this section we will study the influence of the second order grating in the efficiency of the first order at first on-Bragg angular replay condition. We will make use of the implemented RCW, retaining 21 orders in the calculations, 0, ±1, ±2, ... This study will allow us to understand under which conditions is it possible to disregard the effects of this component completely.

We will suppose, therefore that a relative permittivity of the form indicated by equation (14) is stored in the hologram.

\[ \varepsilon(x) = \varepsilon_r + \varepsilon_1 \cos[Kx] + \varepsilon_2 \cos[2Kx] \]  

(14)

To investigate whether the second order term in Eq. (14) can be disregarded from the calculations of the diffraction efficiency of the first order at the first on-Bragg angular replay condition we define the relative error as:

\[ \text{Error} = \left| \frac{\eta_{(1)} - \eta_{(2)}}{\eta_{(2)}} \right| \]  

(15)

where \( \eta_{(2)} \) is the efficiency of the first order when both \( \varepsilon_1 \) and \( \varepsilon_2 \) are retained in the calculations and \( \eta_{(1)} \) is the efficiency when only \( \varepsilon_1 \) is retained. The evolution of the error as a function the grating strength will be investigated, where the grating strength is defined as

\[ \nu = \frac{\pi \varepsilon d}{2 \varepsilon_0^{1/2} \cos \theta} \]  

(16)

d is the thickness of the polymer material, which varies from [0 µm, 80 µm]. The range of the grating strength was restricted to [0, 3π/4]. The value of \( \nu_1 \) was chosen in such a way as to ensure that maximum diffraction efficiency, \( \nu = \pi/2 \), occurs at \( d = 45 \) µm, thus over-modulation, \( \nu > \pi/2 \) is also investigated.
To take into account effects at the boundaries a realistic plate made of polymeric material and glass was considered. A unit plane wave was assumed incident on the polymer material from air, and the forward diffracted orders coming from the hologram medium propagate inside glass. To include the new boundary, glass-air (medium 3-4), Fresnel losses were taken into account between the glass substrate and air. The refractive indexes considered in the theoretical simulations were: $n_{a}=1$, $n_{m}=1.59$, $n_{g}=1.54$ for air, polymeric medium and glass, respectively.

Figure 1 shows the relative error calculated using RCW as a function of the grating strength for transmission diffraction gratings presenting different spatial frequencies, 500, 700, 1000 and 1500 lines/mm. The curves presented in each figure correspond to different values of the ratio $\varepsilon_2/\varepsilon_1$: 1/2, 1/4 and 1/8. It is clear from the figures that whenever the spatial frequency is increased the relative error decreases. For low spatial frequencies, 500 and 700 lines/mm, the relative error is significant for low values of the grating strength. This is due to the fact that for low spatial frequencies multi-wave diffraction occurs [16,17] and the influence of $\varepsilon_2$ is potentially high. Nonetheless, for spatial frequencies higher than 1000 lines/mm the relative error is always below a 3%, and mainly below 1% when the ratio $\varepsilon_2/\varepsilon_1 < 1/8$, in the range $[0 \ 3\pi/4]$. Therefore, for spatial frequencies higher than 1000 lines/mm and ratios $\varepsilon_2/\varepsilon_1 < 1/2$, the influence of the second harmonic is very low on diffraction efficiency of the first order for on-Bragg replay.

![Graphs showing relative error as a function of grating strength for different spatial frequencies and ratios](image)

Fig. 1. Relative error of the first order efficiency, at first Bragg angle condition, for transmission diffraction gratings of different spatial frequencies for different values of the ratio $\varepsilon_2/\varepsilon_1$: 1/2, 1/4 and 1/8.
4. Angular responses of first and second orders for unslanted transmission gratings recorded on PVA/Acrylamide photopolymer materials

In this section we will show the angular responses of the first order, centered at first on-Bragg angular replay condition, and second order angular responses, centered at second on-Bragg angular replay condition, for transmission gratings recorded on PVA/acrylamide photopolymer material. The importance of RCW will be demonstrated here. The ratios of $\epsilon_2/\epsilon_1$ are demonstrated to be of the order of $\sim 1/7$ for the diffraction gratings studied here. This is a clear demonstration that a non-perfect sinusoidal pattern is recorded in the polymeric material and that a significant second order grating is recorded in it.

The holograms were recorded on PVA/acrylamide photopolymer material using an Argon laser of wavelength 514 nm. The intensity was of 6 mW/cm$^2$. The beams impinged on the material symmetric to the normal to the recording material, with a beam ratio of 1:1, so unslanted transmission gratings were recorded. The gratings were recorded with two different spatial frequencies: 545 lines/mm and 1125 lines/mm, in order to observe the effect of the spatial frequency on the efficiency of the second order.

To prepare the material a method similar to that described in other papers was used [18-21]. A polymerization solution was prepared using polyvinylalcohol 18-88 (PVA) provided by Fluka as a binder, acrylamide (AA) as monomer and yellowish eosin as dye.

Fig. 2. Angular responses of the first and second order efficiency for a transmission diffraction grating recorded on PVA/acrylamide photopolymer material with a spatial frequency of 545 lines/mm and a thickness of 73 $\mu$m.

Fig. 3. Angular responses of the first and second order efficiency for a transmission diffraction grating recorded on PVA/acrylamide photopolymer material with a spatial frequency of 545 lines/mm and a thickness of 105 $\mu$m.
Figure 2 shows the efficiencies of the first and second order as a function of the reconstruction angle (in air), for a transmission grating recorded on the polymeric material with a spatial frequency of 545 lines/mm. The experimental data are represented as dots, whereas the theoretical simulation is represented as a continuous line. The data obtained from the theoretical simulation are given in Table 1 for the three gratings studied. The ratio $\varepsilon_2/\varepsilon_1 \sim 1/7$ in this case. Good agreement between the theoretical curve and the experimental data can be seen for Fig. 2(a), whereas the experimental data for the second order take slightly higher values than the corresponding theoretical curve. This is possibly due to attenuation of the periodic profile in the grating depth, the effect of which is seen in the smoothed side lobe structure. Figure 3 shows the first and second order angular responses for a transmission diffraction efficiency recorded with the same spatial frequency as that in Fig. 2, 545 lines/mm, but for a higher thickness. The thickness for the transmission grating of Fig. 2 was 73 mm, whereas the thickness for the transmission grating of Fig. 3 was 105 mm. The estimated value of the ratio $\varepsilon_2/\varepsilon_1 \sim 1/9$ for this case.

Table 1. Parameters of the theoretical simulations for transmission diffraction gratings recorded on PVA/Acrylamide photopolymers

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<th>Figure 2</th>
<th>Figure 3</th>
<th>Figure 4</th>
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<tr>
<td>$f$ (lines/mm)</td>
<td>545</td>
<td>545</td>
<td>1125</td>
</tr>
<tr>
<td>$\varepsilon_1$</td>
<td>0.0261</td>
<td>0.0248</td>
<td>0.0356</td>
</tr>
<tr>
<td>$\varepsilon_2$</td>
<td>0.0037</td>
<td>0.0028</td>
<td>0.0049</td>
</tr>
<tr>
<td>$d$ (µm)</td>
<td>73</td>
<td>105</td>
<td>33</td>
</tr>
</tbody>
</table>

Finally, the results corresponding to a transmission grating recorded with a spatial frequency of 1125 lines/mm are also presented. Figure 4 shows the angular responses of the efficiency of the first and second order, respectively. The thickness of this grating was measured as 31 µm, which is lower than in the previous cases so the attenuation effects observed in Fig. 3 are not so important. The ratio $\varepsilon_2/\varepsilon_1$ was of $\sim 1/7$ in this case.

From the results presented in this work, it is clear that the study of the angular responses of the second order centered at the second on-Bragg angular replay condition is useful in
obtaining information about the refractive index second Fourier component. By doing this, the predictions of the different diffusion based theoretical models can be compared. For instance, here we also present the ratio $\varepsilon_2/\varepsilon_1$ predicted by models presented in Ref. [9], model I, and model of Ref. [11], model II, for transmission diffraction gratings with two different spatial frequencies, 545 and 1125 lines/mm. Model I correspond to a non-local diffusion based model called “Nonlocal-response diffusion model”. The difference between this model and all diffusion based models presented in the literature is that in this model a non-local response of the material to explain the growth of the chains of photopolymer away from their initiation point was introduced. In model I the rate of polymerization was assumed to depend linearly with the intensity pattern stored, whereas in model II a square root dependence was assumed. It should be commented that model II has demonstrated to be more accurate to describe the growth curves of the diffraction efficiency as a function of time for different transmission gratings analyzed in Ref. [2]. The value of the parameter $\sqrt{\sigma}$, which characterizes the length scale over which the nonlocal effect is significant, was chosen to be of 100 nm for the theoretical simulations [2]. Table 2 shows different values of $\varepsilon_2/\varepsilon_1$ for different values of the parameter $R$, defined in Ref. [9], for a transmission grating of 545 lines/mm, and Table 3 shows the values for a transmission grating of 1125 lines/mm.

Table 2. Values of $\varepsilon_2/\varepsilon_1$ for a transmission grating with a spatial frequency of 545 lines/mm predicted by the diffusion models presented in Refs. [9,11].

<table>
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<th>$R$</th>
<th>Model I</th>
<th>Model II</th>
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<tr>
<td>0.1</td>
<td>1.419</td>
<td>1.671</td>
</tr>
<tr>
<td>1</td>
<td>0.331</td>
<td>0.516</td>
</tr>
<tr>
<td>10</td>
<td>0.039</td>
<td>0.208</td>
</tr>
</tbody>
</table>

Table 3. Values of $\varepsilon_2/\varepsilon_1$ for a transmission grating with a spatial frequency of 1125 lines/mm predicted by the diffusion models presented in Refs. [9,11].

<table>
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<th>$R$</th>
<th>Model I</th>
<th>Model II</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>0.969</td>
<td>1.152</td>
</tr>
<tr>
<td>1</td>
<td>0.169</td>
<td>0.276</td>
</tr>
<tr>
<td>10</td>
<td>0.018</td>
<td>0.114</td>
</tr>
</tbody>
</table>

From Tables 2 and 3 it is interesting to note that for increasing values of $R$, the grating profile stored in the hologram gets more sinusoidal (lower values of $\varepsilon_2/\varepsilon_1$). This is in agreement with a recent work by S. Wu and W. N. Glytsis [22]. In this work the one-dimensional nonlocal diffusion equation of Ref. [9] is solved using a finite-difference time-domain method, so that all the harmonic components of the polymer and monomer concentration are retained. As they show, the higher the values of $R$ and $\sigma$, the more the polymer concentration profile resembles a sinusoidal pattern. On the other hand, it should be inferred that the particular value of $R$ presented by the PVA/acrylamide material studied, if only model II is used, is closer to 10 for the particular ratios of $\varepsilon_2/\varepsilon_1$ obtained in this work.

5. Conclusions

The existence of second Fourier components in the relative dielectric permittivity has been studied in the particular case of transmission diffraction gratings recorded in PVA/acrylamide photopolymers. It has been demonstrated that a ratio of $\sim1/7$ exists between the second and
first order Fourier components of the dielectric permittivity which are stored in the diffraction gratings studied. The effect of the second order Fourier component in the first order efficiency at first Bragg angle condition has also been investigated. It has been demonstrated that, especially for on-Bragg angular replay condition of high diffraction efficiency not over-modulated gratings, for spatial frequencies of over 1000 lines/mm and ratios $\varepsilon_2/\varepsilon_1 < 1/4$ the second order Fourier component can be disregarded from the calculations. The results presented demonstrate that the study of the angular responses of second orders may give important information about the kind of profiles recorded in photopolymer materials. These results must be considered in the context of the different material models that describe the mechanism of hologram formation in photopolymer materials.

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