Experimental evidence of mixed gratings with a phase difference between the phase and amplitude grating in volume holograms

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Abstract: The Coupled Wave Theory of Kogelnik has given a well-established basis for the comprehension of how light propagates inside a hologram. This theory gives an accurate approximation for the diffraction efficiency of volume phase holograms and volume absorption holograms as well. Mixed holograms (phase and absorption) have been also treated from the point of view of this theory. For instance, Guibelalde theoretically described the diffraction efficiency of out of phase mixed volume gratings. In this work we will show that when using fixation-free rehalogenating bleaches, out of phase mixed volume gratings can be recorded on the hologram at high exposures. This is due to the oxidation products of the developer and the bleaching agent. The effects described theoretically for out of phase mixed volume hologram gratings are experimentally observed.

OCIS Codes: (090.0090) Holography, (090.7330) Volume holographic gratings, (090.2900) Holographic recording materials.

References and links

1. L. Solymar and D. J. Cooke, *Volume Holography and Volume Gratings*, (Academic, London 1981).
2. R. R. A. Syms, *Practical Volume Holography*, (Clarendon, Oxford 1990).
3. H. Kogelnik, "Coupled wave theory for thick hologram gratings," Bell Sys. Tech. J. **48**, 2909-47 (1969).
4. A. Beléndez, T. Beléndez, C. Neipp and I. Pascual, "Determination of the refractive index and thickness of holographic silver halide materials by the use of polarized reflectances," Appl. Opt. **41**, 6802-08 (2002).
5. E. Guibelalde, "Coupled wave analysis for out-of-phase mixed thick hologram gratings," Opt. Quantum Electron. **16**, 173-178 (1984).
6. N. J. Phillips, A. A. Ward, R. Cullen and D. Porter, "Advances in holographic bleaches," Phot. Sci. Eng. **24**, 120-4 (1980).
7. B. J. Chang and C. D. Leonard, "Dichromated gelatin for the fabrication of holographic optical elements," Appl. Opt. **18**, 2407-17 (1979).
8. K. S. Pennington and J. S. Harper, "Techniques for producing low-noise, improved efficiency holograms," Appl. Opt. **9**, 1643-50 (1970).
9. H. I. Bjelkhagen, *Silver-Halide Recording Materials*, (Springer, Berlin 1995).
10. J. Crespo, A. Fimia and J. A. Quintana, "Fixation-free methods in bleached reflection holography," Appl. Opt. **25**, 1642-5 (1986).
11. P. Hariharan and C. M. Chidley, "Rehalogenating bleaches for photographic phase holograms: the influence of halide type and concentration on diffraction efficiency and scattering," Appl. Opt. **26**, 3895-8 (1987).
12. P. Hariharan, "Rehalogenating bleaches for photographic phase holograms 3: Mechanism of material transfer," Appl. Opt. **29**, 2983-5 (1990).
13. C. Neipp, I. Pascual and A. Beléndez, "Over modulation effects in fixation-free rehalogenating bleached holograms," Appl. Opt. **40**, 3402-3408 (2001).
14. C. Neipp, I. Pascual and A. Beléndez, "Optimization of a fixation-free rehalogenating bleach for BB-640 holographic emulsion," J. Mod. Opt. **47**, 1671-1679 (2000).
15. C. Neipp, I. Pascual and A. Beléndez, "Fixation-free rehalogenating bleached reflection holograms recorded on BB-640 plates," Opt. Commun. **182**, 107-114 (2000).

16. M.Ulibarrena, M. J. Méndez, L. Carretero, R. Madrigal and A. Fimia, "Comparison of direct, rehalogenating, and solvent bleaching processes with BB-640 plates," Appl. Opt. **41**, 4120-4123 (2002).
17. C. Neipp, C. Pascual and A. Beléndez, "Mixed phase-amplitude holographic gratings recorded in bleached silver halide materials," J. Phys. D **35**, 957-967 (2002).
18. L. Carretero, R. F. Madrigal, A. Fimia, S. Blaya and A. Beléndez, "Study of angular responses of mixed amplitude-phase holographic gratings: shifted Borrmann effect," Opt. Lett. **26**, 786-7888 (2001).
19. R. F. Madrigal, L. Carretero, S. Blaya, M. Ulibarrena, A. Beléndez y A. Fimia, "Diffraction efficiency of unbleached phase and amplitude holograms as a function of volume fraction of metallic silver," Opt. Commun. **201**, 279-282 (2002).
20. C. Neipp, I. Pascual and A. Beléndez, "Theoretical and experimental analysis of overmodulation effects in volume holograms recorded on Bb-640 emulsions," J. Opt. A 3, 504-513 (2002).

1. Introduction

Several theories have treated the propagation of light inside a hologram. A good review of them is made in the books of Solymar and Cooke [1] and Syms [2]. The Coupled Wave Theory of Kogelnik [3] has the advantage over many theories in that it is easy to understand from the physical point of view. This theory can be applied to volume holograms. A volume hologram is defined to be when the thickness of the recording medium is of the same order as the fringe spacing. The distinction between thick and thin holograms can be made according to a non-dimensional parameter, Q, defined as follows:

$$Q = \frac{2\pi\lambda d}{n\Lambda^2} \tag{1}$$

where λ is the wavelength in air, and where d and n are the thickness and the refractive index, respectively. When $Q < 1$ the hologram is considered thin, whereas when $Q > 10$ the hologram is thick. The Coupled Wave Theory gives good results for $Q > 10$. The experiments in this work were done by using BB-640 plates, $n \sim 1.62$, $d \sim 7$ µm [4]. For holograms recorded on these plates with a spatial frequency of 1200 lines/mm the value of the parameter Q was found to be ~ 29, so the Coupled Wave Theory is applicable in this case.

The main assumption made by the Coupled Wave Theory is that only two orders propagate inside the hologram: the first order (+1) and the zero order (0). With this assumption the electric field inside the hologram results from the superposition of the electric fields of the two orders. By solving Helmholtz equation, two differential equations are obtained related by the coupling constant. An analytical approximation for the diffraction and transmission efficiencies are then obtained for phase and absorption holograms and also for mixed gratings. For mixed gratings, in the case in which the absorption constant modulation and the refractive index modulation are in phase, the diffraction efficiency can be calculated as the contribution of two different terms separately, one term due to the phase grating and the other one due to the absorption one. In this case, the influence of the absorption term was demonstrated to be low (3.6%). An out of phase theoretical analysis was made by Guibelalde [5] demonstrating that the diffraction efficiency may show strong dependence of dephasing.

As Guibelalde says the study made could be important for the case of ferroelectric materials where the dielectric grating and the absorption grating may have different phases. We will show that out-of-phase mixed gratings are stored in bleached holograms recorded on photographic emulsions, giving an experimental counterpart of the theoretical analysis. The effects of the absorption modulation and the dephasing in the angular response of the transmittance for mixed gratings are also studied.

2. Theoretical model

2.1. Solution of the wave equation

When a sinusoidal interference pattern of light is recorded on a photosensitive medium the electro-optic properties of the medium vary in an harmonic way:

$$n = n_0 + n_1 \cos(\boldsymbol{Kr}) \tag{2}$$

$$\alpha = \alpha_0 + \alpha_1 \cos(\boldsymbol{Kr} + \varphi) \tag{3}$$

where n is the refractive index of the medium at any point, $\boldsymbol{r}$, n_0 is the average refractive index and n_1 is the refractive index modulation. On the other hand α is the absorption constant at any point, α_0 is the average absorption constant and α_1 is the absorption constant modulation. $\boldsymbol{K}$ is the grating vector, which is perpendicular to the fringes recorded in the medium. It can be seen that a phase difference between the refractive index and the absorption constant is allowed [5].

In order to obtain an expression for the diffraction and transmission efficiencies for a volume phase transmission grating a similar treatment to that made by Guibelalde is done. By supposing that only two orders propagate inside the emulsion [3], +1 order, S, and zero order, R, and considering slowly coupling between the diffracted wave and the transmitted wave one can finally obtain the solutions of the coupled wave equations for the diffracted and transmitted wave amplitudes:

$$R(z) = -\frac{1}{c_r(\gamma_1 - \gamma_2)}[-(c_r\gamma_2 + \alpha_0)\exp(\gamma_1 z) + (c_r\gamma_1 + \alpha_0)\exp(\gamma_2 z)] \tag{4}$$

$$S(z) = -\frac{j\chi_2}{c_s(\gamma_1 - \gamma_2)}[\exp(\gamma_1 z) - \exp(\gamma_2 z)] \tag{5}$$

where:

$$\gamma_{1,2} = -\frac{1}{2}\left[\frac{\alpha_0}{c_r} + \frac{\alpha_0}{c_s} + j\frac{\vartheta}{c_s}\right] \pm \frac{1}{2}\left[\left(\frac{\alpha_0}{c_r} - \frac{\alpha_0}{c_s} - j\frac{\vartheta}{c_s}\right)^2 - 4\frac{\chi_1\chi_2}{c_r c_s}\right]^{1/2} \tag{6}$$

where $c_r = \cos\theta_r$ and $c_s = \cos\theta_d$, θ_r and θ_d are the angles that the transmitted and diffracted propagation vectors form with the normal of the hologram, ϑ is the off-Bragg parameter [1], and:

$$\chi_1 = \frac{\pi n_1}{\lambda} - j\exp(-j\varphi)\frac{\alpha_1}{2} \tag{7}$$

$$\chi_2 = \frac{\pi n_1}{\lambda} - j\exp(j\varphi)\frac{\alpha_1}{2} \tag{8}$$

The diffraction efficiency can then be obtained as:

$$\eta = \frac{|c_s|}{c_r} S(d)S(d)^* \tag{9}$$

and the transmission efficiency as:

$$\tau = R(d)R(d)^* \tag{10}$$

Because photographic emulsions are composed of small silver halide grains suspended in gelatin, light entering the hologram scatters inside it. Therefore the effects of scattering due to the silver halide grains must be taken into account. To do this, equation (10) was transformed into the following:

$$\tau = \exp(-\alpha_s d) \cdot RR^* \tag{11}$$

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where α_s takes into account the effects of scattering.

Figure 1 shows the transmittance (obtained from equation (10)) as a function of the angle for volume holographic diffraction gratings with different values of the dephasing φ between the refractive index modulation and the absorption modulation. The computer simulation was done with the following parameters: d = 8 µm, n_1 = 0.05 α_0 = 0.03 µm^{-1} α_1 = 0.02 µm^{-1} and φ took the values: 0, π/2, π.

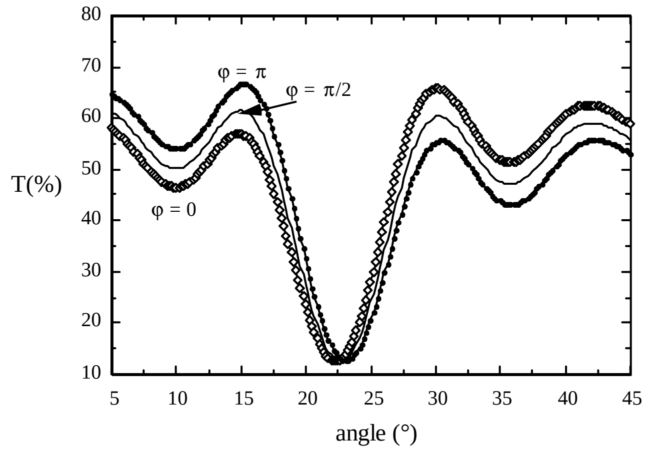

Fig. 1. Transmission efficiency as a function of the angle for different values of the phase difference, φ. n_1 = 0.05, d = 8 µm, α_0 = 0.03 µm^{-1} and α_1 = 0.03 µm^{-1}.

2.2. Diffraction efficiency at the Bragg condition.

When the Bragg condition is satisfied, $\vartheta = 0$, the amplitude of the diffracted wave takes the form:

$$S(d) = -j\exp\left(-\frac{\alpha_0 d}{c_r}\right)\left(a - je^{j\varphi}b\right)\frac{sin\left[\sqrt{(a - je^{j\varphi}b)\cdot(a - bje^{-j\varphi})}\right]}{\sqrt{(a - bje^{j\varphi})\cdot(a - bje^{-j\varphi})}} \quad (12)$$

where:

$$a = \frac{d\pi n_1}{\lambda c_r} \quad (13)$$

$$b = \frac{\alpha_1 d}{2c_r} \quad (14)$$

In the case in which $\varphi = 0$ (there is no phase difference between the refractive index modulation and the absorption modulation):

$$S(d) = -j\left(\frac{c_r}{c_s}\right)^{1/2}\exp\left(-\frac{\alpha_0 d}{c_r}\right)sin(a - jb) \quad (15)$$

$$\eta = \exp\left(-\frac{2\alpha_0 d}{c_r}\right)\cdot\left[sin^2(a) + sinh^2(b)\right] \quad (16)$$

Expression (16) is obtained from Kogelnik's Coupled Wave Theory [3] in the case of mixed gratings (no phase difference assumed). This expression indicates that when there is no phase difference between the refractive index modulation and the absorption coefficient, the diffraction efficiency is the sum of two contributions, one from the phase grating and the other from the absorption grating. The part corresponding to the modulation of the absorption coefficient hardly influences the final diffraction efficiency: the absorption effects are taken into account in the value of the exponential and the contribution to diffraction efficiency is reflected in the refractive index modulation. Nevertheless, Guibelalde [5] demonstrated that the effects of modulation of the absorption constant are significant when there is a phase difference between the refractive index modulation and the modulation of the absorption constant. Figure 2 shows the diffraction efficiency at the Bragg condition as a function of the phase difference φ, for mixed diffraction gratings and different values of modulation of the absorption constant α_1. It can be seen that as α_1 increases, so does the influence of the absorption grating.

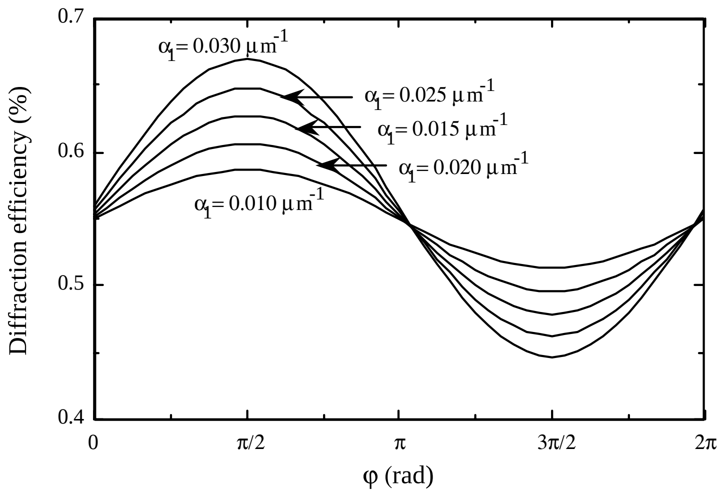

Fig. 2. Diffraction efficiency as a function of the phase difference, φ, between the refractive index and the absorption constant for different values of the absorption constant modulation, α_1, n_1 = 0.030 and α_0 = 0.030 μm^{-1}.

3. Fixation-free bleached holograms

In order to obtain phase holograms, several methods are described in the literature [6-8]. For instance, bleached silver halide emulsions have long been used as a medium for recording volume phase holograms because they offer several attractive advantages [9]. These advantages include a relatively high sensitivity, ease of processing of the results, improved chemical processing, their availability as commercial films and the repeatability of the results. One of the drawbacks of using conventional or reversal bleaches is that the emulsion shrinks after the procedure because material is removed from the emulsion. This problem was solved by using fixation-free bleaching processes [10-12]. In this method, after exposure, the silver halide grains in the exposed zones are converted into silver grains in a reduction process: the development. Subsequently, using a rehalogenating bleaching bath, the silver grains are converted back into silver halide grains in an oxidation process. During the bleach bath there is a transfer of material from the exposed to the unexposed zones [12] and as a result the silver-halide grains in the non-exposed zones increase in size. The refractive index modulation is a result of the difference in size of the silver halide grains in the exposed and non-exposed zones. This diffusion process is particularly influenced by the halide concentration in the bleach solution [11]. Although fixation-free rehalogenating bleaches are generally known for creating pure phase holograms [9, 13-16], the oxidation products of the bleach can give rise to an absorption modulation at high values of exposure and high concentrations of potassium bromide in the bleach bath. Experimental confirmation of this fact can be found in reference [17]. In this reference, mixed gratings without dephasing between the amplitude and phase gratings were analyzed. By fitting the theoretical function of transmittance to the experimental data, quantitative information about the absorption constant and absorption constant modulation was obtained. The effects of the absorption modulation on the angular response of transmittance were studied. It is interesting to note that in this study we recorded mixed-gratings using an experimental procedure which is usually used to produce phase gratings. In a recent study Carretero et al [18, 19] experimentally investigated the response of transmittance in mixed holograms recorded using procedures which typically produce absorption holograms.

4. Experimental

Figure 3 shows the experimental set-up used in the study. Experiments were carried out with BB-640 red sensitive silver-halide plates. Unslanted holographic transmission gratings were recorded on the emulsions by the interference of two collimated beams from a He-Ne laser (633 nm). The beam ratio was 1:1, and the polarization plane was normal to the plane of incidence. The angle (in air) between the normal of the plate and each of the incident beams was θ_0 = 22.5º, and so the interbeam angle was $2\theta_0$ = 45º. With this arrangement, the spatial frequency of the diffraction gratings was calculated as 1200 lines/mm. Holographic plates were mounted upon a motorized rotation stage, which was connected to a personal computer by an IEEE-488 interface. The rotation device had a resolution of 0.001º.

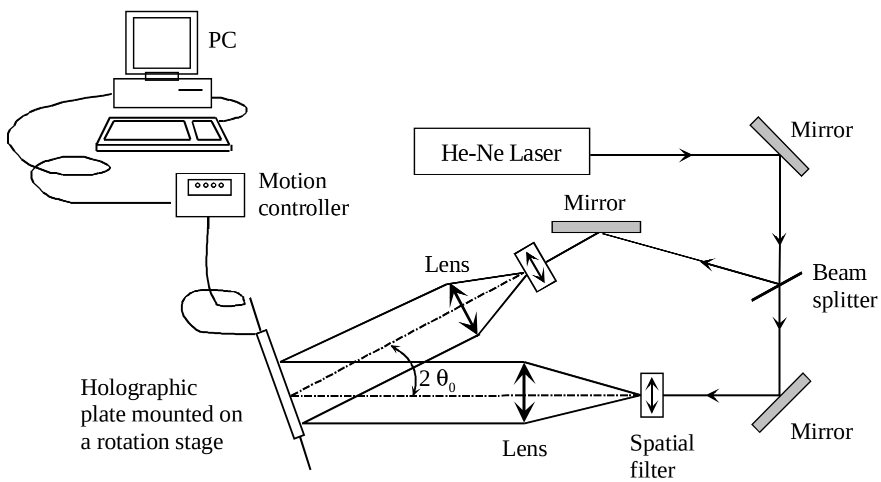

Fig. 3. Experimental set-up.

After exposure the plates underwent the schedule procedure in Table I. In order to obtain phase holograms the plates were bleached with a modified version of the R-10 bleach (Table II). The bleach bath solution is composed of two different solutions: A and B. The oxidizer is contained in solution A, whereas the potassium bromide is contained in solution B. To obtain the bleach solution, 1 part of A is mixed with 10 parts of distilled water and X parts of B. The ratio X = B/A indicates the relation between the potassium bromide concentration and the oxidizer concentration (potassium dichromate). In previous articles the B/A ratio was optimized in order to take into account different experimental conditions [14, 15]. The holograms studied in this paper were bleached with a B/A ratio of 240.

In order to obtain the transmittance as a function of the angle of reconstruction we placed the plates on a rotating stage. The polarization plane of the transmitted beam was kept perpendicular to the plane of incidence and the transmittance was calculated as the ratio of the transmitted beam intensity to the incident power. In order to take into account Fresnel losses the expression was multiplied by the same factor as the diffraction efficiency.

Table I. Schedule procedure

1.- Develop (20°C) (2 min in CW-C2 developer)
2.- Rinse in running water 1 min
3.- Bleach for 1 min after the plate has cleared
4.- Rinse in running water 5 min
5.- Dry at room temperature

Table II. Bleach bath composition (modified version of R-10)

Solution A	
Potassium dichromate	20 g
Sulfuric acid	15 ml
Distilled water	1 l
Solution B	
Potassium bromide	100 g
Distilled water	1 l

(*Just before use, mix 1 part of A with 10 parts of distilled water and add X parts of B*)

5. Results and discussion

At first we will explain the mechanism which could yield to the creation of out of phase mixed volume gratings when fixation-free rehalogenating techniques are used. As commented in Section 3 a refractive index modulation is stored in the hologram as a consequence of the differences in size of the silver halide grains in the exposed and non-exposed zones, n_{r1}. The refractive index created by this mechanism, n_r, has the form:

$$n_r = n_0 + n_{r1} \cos(\boldsymbol{Kr} + \pi) \qquad (17)$$

The phase π indicates the dephasing between the refractive index, n_r, and the interference pattern of light recorded in the hologram. This phase difference is due to the fact that the diffusion of silver halide grains takes place from the exposed to the unexposed zones. On the other hand, if bleaches with potassium dichromate are used in the procedure, another refractive index modulation, n_{h1}, could be stored in the hologram due to the differences in the degree of hardening between the exposed and non-exposed zones [13]. The refractive index created by this mechanism, n_h, has the form:

$$n_h = n_0 + n_{h1} \cos(\boldsymbol{Kr}) \qquad (18)$$

Note that there is no dephasing now. This is because the hardening effect occurs when the ion Cr^{+6} is chemically reduced to Cr^{+3} during the bleaching bath [13]. This action takes place in the exposed zones, therefore the refractive index is in phase with the interference pattern recorded.

Finally, as explained in Section 3, the oxidation products of the bleach could create an absorption modulation, α_1, which has the form:

$$\alpha = \alpha_0 + \alpha_1 \cos(\boldsymbol{Kr}) \qquad (19)$$

There is no dephasing between the absorption modulation and the intereference pattern because the oxidation process takes place in the exposed zones.

From equations (17) and (18) the refractive index stored in the hologram can be found as:

$$n = n_0 + n_1 \cos(\boldsymbol{Kr} + \varphi) \qquad (20)$$

where n_1 and φ will depend on n_{r1} and n_{h1}. φ indicates the dephasing between the absorption and the refractive index modulation.

We will comment now how the phase difference, φ, will be reflected in an asymmetry in the angular response of the diffraction and transmission efficiency if the grating is reconstructed in one or another Bragg angle. Let's analyze the volume diffraction grating of Fig. 4, where a phase difference, φ, between the amplitude and refractive index maxima is allowed. It can be seen that the phase difference is φ for beam 1 and $(2\pi-\varphi)$ for beam 2. From Fig. 2 it can be inferred that this relation between the dephasings means different values of the diffraction efficiency if the grating is reconstructed in one or other Bragg angle.

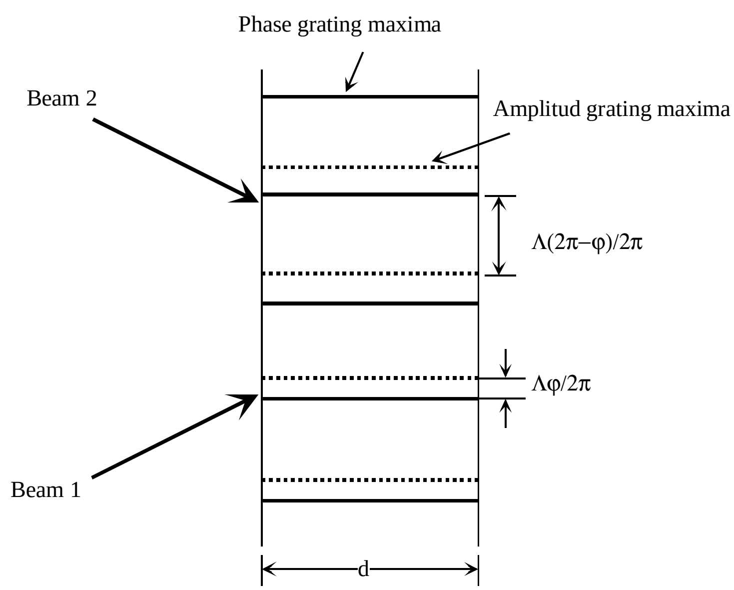

Fig. 4. Unslanted mixed diffraction gratings.

Figures 5 and 6 show the angular response of the transmittance for phase transmission gratings recorded on BB-640 emulsion using fixation free bleaching techniques. The circles represent the experimental data, whereas the continuous line represents the theoretical function of transmission efficiency, obtained from equation (11). Some important aspects must be commented. It has been explained that in order to create a phase difference, φ, between the amplitude and refractive index modulations a hardening of the gelatin must occur during the bleach bath. Nonetheless, the hardening action due to chemical reduction of ion Cr^{+6} is low, therefore only for high exposures, where this hardening effect is increased, out of phase volume gratings were obtained. For instance the exposure of volume grating in Fig. 5 was of 2500 $\mu J/cm^2$ and grating of Figure 6 was exposed to a value of 3900 $\mu J/cm^2$. The fact that the diffraction gratings were recorded at high exposures implies on one hand that the diffusion process becomes more chaotic and on the other that higher order terms could appear. This is why the theoretical function does not exactly reproduce the experimental data. Nevertheless the general behavior of the experimental transmittance curves is quite well described by the theoretical simulation. For instance, the asymmetry commented if the reconstruction is made near one or other Bragg angle is clearly observed, red arrows. This asymmetry is, in fact, only understood if a phase difference φ, between the refractive index and absorption modulation is allowed.

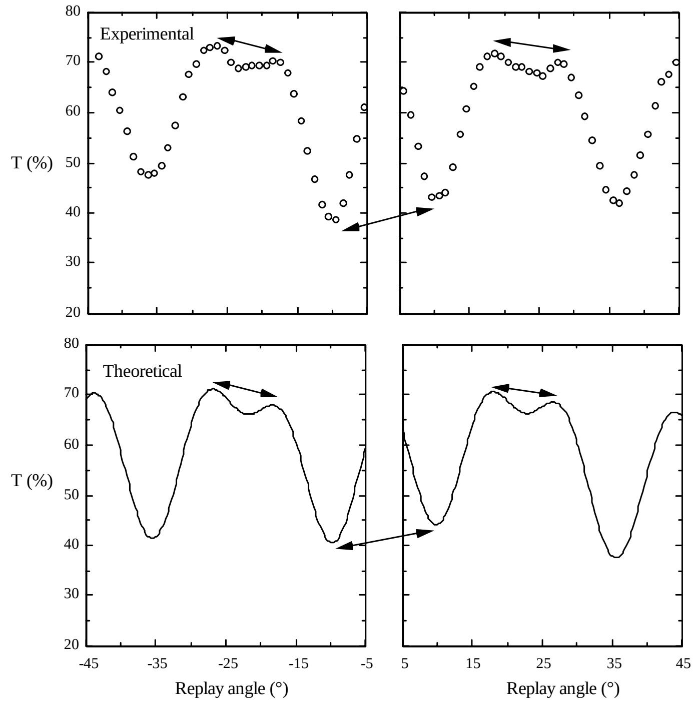

Fig. 5.- Transmittance as a function of the angle for a mixed diffraction grating. Parameters: n_1 = 0.085, d = 6.8 μm, α_0 = 0.017 μm^{-1}, α_1 = 0.012 μm^{-1}, α_s = 0.018 μm^{-1}, φ = 0.25 rad for $\theta \in$ [-45°,0°], φ = 4.31 rad for $\theta \in$ [0°,45°].

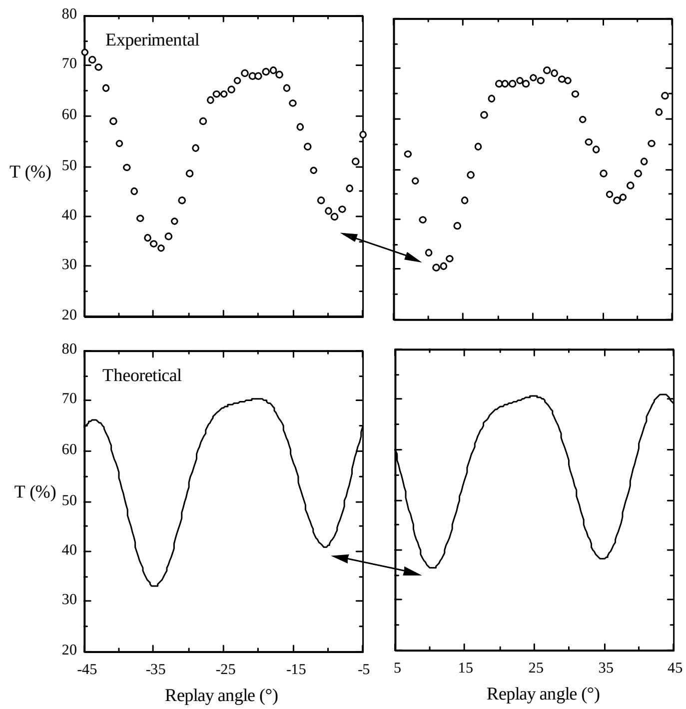

Fig. 6.- Transmittance as a function of the angle for a mixed diffraction grating. Parameters: n_1 = 0.091, d = 6.8 μm, α_0 = 0.019 μm^{-1}, α_1 = 0.014 μm^{-1}, α_s = 0.014 μm^{-1}, φ = 4.19 rad for $\theta \in$ [-45°,0°], φ = 0.13 rad for $\theta \in$ [0°,45°].

Another interesting feature of the transmittance curves shown in Fig. 5 and 6 is that the values of the refractive index modulation used in the computer simulations are really high: $n_1 = 0.085$ for grating of Fig. 5 and $n_1 = 0.091$ for grating of Fig. 6. In a recent article [20] it was analyzed how these high values of the refractive index modulation yield to a family of theoretical curves of the angular response of the transmittance. One can see that these curves shown in Fig. 5 and 6 resemble those of reference [20], but are slightly modulated by the influence of the absorption modulation and the dephasing φ.

Finally it must be remarked that the theoretical functions shown in Fig. 5 and 6 correctly predict the experimental data, thereby validating the theoretical model.

6. Conclusions

In this article, experimental evidence of mixed (phase-amplitude) gratings with a phase difference between the refractive and the amplitude modulation is given. Fixation-free bleached transmission gratings were recorded on BB-640 plates and the transmittance as a function of the angle was measured experimentally. The theoretical function of the transmittance was fitted to the experimental data and good agreement between the theoretical model and the experimental data was found. The results presented confirm the applicability of Guibelalde's model, based on Kogelnik's Coupled Wave Theory, to the case of mixed gratings with dephasing.

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