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Qualitative Analysis of Bleached Holographic Diffraction Grating

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With nonhardening fixer, dichromated bleacher and alcohol drying, the diffraction efficiency of over 71% has been achieved for holographic phase gratings in silver halide emulsion. The swollen emulsion of Agfa 8E75 HD film is identified by scanning electron microscope (SEM) after chemical processing. Dichromated bleacher and rapid dehydration using alcohol drying make a strong modulation so that diffraction efficiency is increased over 20%. The principal characteristic parameters in coupled wave theory are investigated and new modified parameter values are presented by computer simulation. Controlling the emulsion thickness has an important role as a potential source for high diffraction efficiency.

I. INTRODUCTION

The ideal holographic recording materials should have a high resolution, a linear trasfer characteristics, low noise and high sensitivity well matched to available laser wavelength. Among the recording materials for holography, silver halide photographic emulsions are the most convenient for phase hologram due to therr high sensitivity and ease of handling. In addition, they can be dye sensitized so that their spectral sensitivity matches the most commonly used laser wavelengths[1].

While the results that can be achieved using silver halide emulsions will not approach the efficiency obtainable with dichromated gelatin or photoresist, it has until recently been the shortcomings of its processing rather than of the materials themselves[2]. Diffraction efficiency of bleached phase hologram has been markedly greater in the case of transmission grating rather than reflection grating.

Holographic diffraction gratings formed by interference patterns have been investigated as holographic optical elements (HOE_S) for several years. In this case, a hologram is an optical element modifying the path of electromagnetic radiation rather than storing the image. The most important problem for practical applications of HOE_S is the diffraction efficiency, especially in case of diffraction grating. This paper is to describe and discuss the experimental results on the transmission gratings recorded in Agfa 8E75 HD leading to the achievement of over 71% efficiency. New modified parameter values in Kogelnik's coupled wave theory are presented by computer simulation due to swollen emulsion.

II. BACKGROUND

Application of photographic bleaching agents permits converting density differences into phase differences. It is apparent that most research has been preoccupied with the bleacher as the major stumbling block. The problem has been eased significantly with the introduction of the ferric nitrate bleacher and of the p-benzoquinon, reagents now widely recommended[3]. And the efficiency of bleached holograms is related to the polarizability of the molecules of the converted silver compound[4]. Apart from the diffraction efficiency of such phase holograms, scattering problems are also of importance. For scattering problems, recent researches have favoured reversal bleach processes in which the fixing bath is omitted and the developed silver is dissolved away, leaving a phase hologram made up of the undeveloped silver halide crystals.

It is shown that the diffraction efficiency of bleached photographic holograms depends very much on the types of bleacher and drying procedures. The influence of drying procedures was presented by Burchhardt and Doherty as well as, more recently, by Hariharan, who showed that soaking a bleached hologram in alcohol prior to drying gave improved diffraction efficiency[5-9].

The thickness of the processed photographic emulsion layer is usually about 15% less than its original thickness, mainly because of the removal of the unexposed silver halide gratings during fixing. The effects of emulsion thickness shrinkage are most noticeable in volume reflection holograms, since the fringe planes run almost parallel to the surface of the emulsion[12]. Because of emulsion shrinkage, amplitude holograms recorded in photographic emulsion also exhibit phase modulation which can modify the modulation trasfer function (MTF). This phase modulation property is mainly due to a surface relief structure arising from local tanning (hardening) of the gelatin by the oxidation vicinity of reduced silver[3]. The formation of such a relief image can be avoided by the use of a developer with a high sulphite content. The use of a tanning fixer as well as a tanning bleacher also helps to minimize emulsion shrinkage. But the use of a nonhardening fixer and alcohol drying causes the emulsion thickness swollen.

III. EXPERIMENT AND DISCUSSION

Two polarized waves incident at angles θ_1 and θ_2 as in Fig. 1. form a interference pattern with spatial frequency

$$f = 1/\wedge = 1/\lambda \ (\sin\theta_1 - \sin\theta_2) \quad (1)$$

where $\wedge$ is the grating period and λ is the wavelength of He-Ne laser.

Collimated waves of equal intensity are used to record transmission grating with exposures ranging from 10 $\mu J/cm^2$ to 500 $\mu J/cm^2$. For high efficiency optical ele-

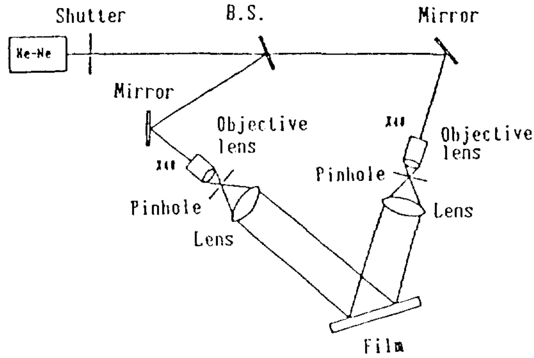


Fig. 1. Recording system for holographic diffraction grating.

ments to be obtained, the spatial frequency must be increased until all but one diffracted order is evanescent. This condition is characterized by the Q facter,

$$Q = 2\pi\lambda d / n_0 \wedge^2 \quad (2)$$

which is related with grating period $\wedge$, thickness d and refractive index of the emulsion n_0. Small values of $Q (Q<1)$ correspond to thin gratings, while large values of $Q (Q \gg 1)$ correspond to volume gratings. However, this condition is not always adequate. According to Moharam et al.,[11] Bragg diffraction regimes follow the curve

$$Q'/\Phi = 20 \quad (3)$$

where $Q' = Q/\cos\theta$, θ being the angle of incident within the grating, and $\Phi = \pi n_1 d/\lambda\cos\theta$ the modulation parameter n_1 being the refractive index modulation. For this experiments, $\theta_1 = 25°$ and $\theta_2 = -25°$, so f=1335 lines/mm, $Q \gg 1$, $Q'/\Phi > 20$, it is considered as volume type.

The diffraction efficiency of grating depends very much on the chemical processing, so that the choices of developer, bleacher and drying methods are important. The processing schedules for these experiments are summarized in Table 1., 2.

The diffraction efficiency η is defined as the ratio

$$\eta = I_1/I_0 \quad (4)$$

Table 1. Chemical processing

Process	Time
1. Exposure	
2. Develop (Kodak D-19)	4 min
3. Rinse (Distilled water)	1 min
4. Fix (Kodak Rapid Fixer Sol. A)	4 min
5. Rinse (Distilled water)	1 min
6. Bleach (Table 2)	
7. Rinse (Distilled water)	10 min
8. Ethyl alcohol	2 min
9. Isopropyl alcohol	4 min
10. Drying (20°C and <50%RH)	

Table 2. Bleacher formula

Bleacher	
Potassium dichromate	1.8 g
potassium bromide	4.0 g
Sulfuric acid	1.0 m*l*
Distilled water to make	1.0 *l*

where I_1 is a power in the first diffracted order at the Bragg angle and I_0 is a incident power. Because reflection loss at the surface is inevitable, intrinsic diffraction efficiency η_i only in view of diffraction is defined as

$$\eta_i = I_1/(I_0 - I_r) \tag{5}$$

where I_r is a reflected power at the surfaces. The other diffracted orders power is negligible which could be taken as the losses due to absorption and scattering in the emulsion.

The diffraction efficiencies obtained as a function of exposure are shown in Fig. 2. The peak diffraction efficiencies of $\eta=71\%$ and $\eta_i=81\%$ are demonstrated at an exposure of 180 μJ/cm^2.

Fig. 3. shows curves plotted for the efficiency η_i, one of which is dried directly after final washing, while the other is soaked in alcohol before drying. For uniform drying, it has been suggested that the film be rinsed in a solution of ethanol, then washed twice in isopropanol[5-9]. The rapid dehydration of the swollen gelatin using alcohol drying introduces physical stresses that result in cracks (voids) along in the dielectric emulsion.

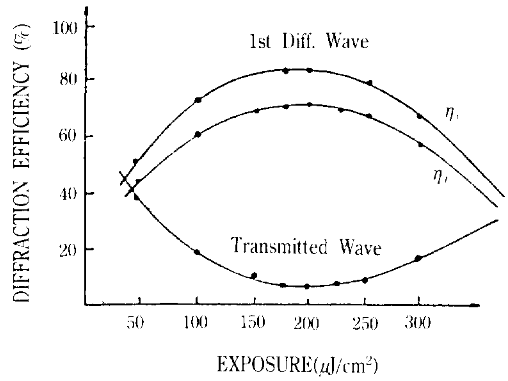


Fig. 2. Diffraction efficiencies vs exposure.

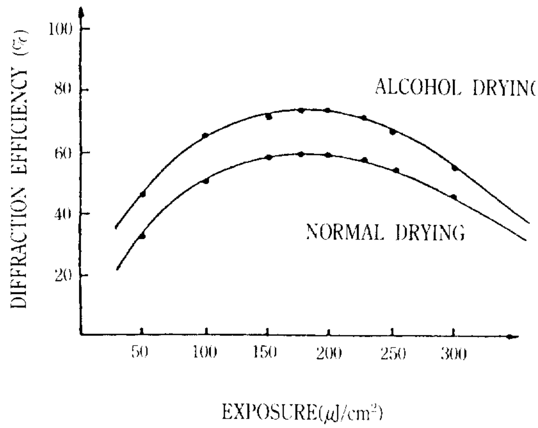
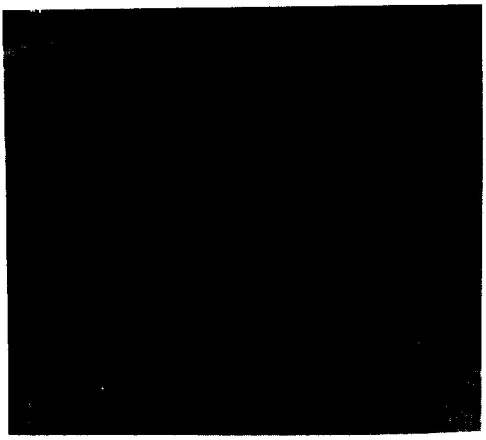


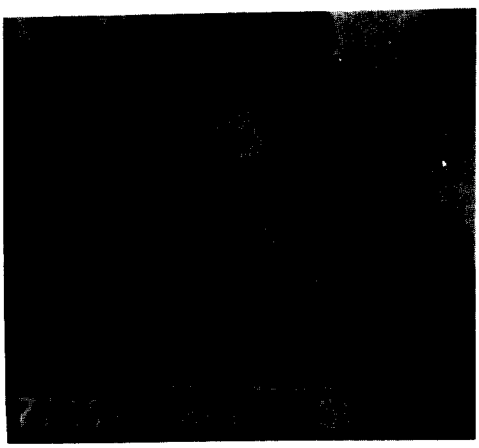
Fig. 3. Diffraction efficiency increasement due to alcohol drying.

Soaking the emulsion in alcohol after chemical processing in this experiment has a modulation mechanism similar to that used to alcohol modulation in dichromated gelatin (DCG)[12]. Dichromated cross-linkage and air void by rapid dehydration increase refractive index modulation. Photo 1. is the cross-sectional view of swollen gelatin by SEM photography. It is also found that the alcohol processed holographic grating replayed at an angle larger than the recording angle because of a swollen emulsion[2].

This swollen effect, thickness 7 μm (original thickness 5 μm) , is newly identified by SEM photography. And modification of parameters in coupled-wave theory is required with respect to thickness variation.



(a) emulsion on the film substrate



(b) Magnified emulsion

Photo. 1. Cross view of swollen gelatin by SEM.

IV. MODIFICATION OF PARAMETER VALUES

The optical grating modulation by periodic structure is constructed in the dielectric permittivity (or equivalently refractive index) or in the conductivity (or equivalently absorption). Also grating may be of the surface-relief type with periodic variation in the surface of a dielectric material[13].

Diffraction of electromagnetic waves by spatially periodic media may be analyzed by numerous methods and with a wide variety of possible assumptions. The most common methods of analyzing grating diffraction are the coupled-wave theory and the modal approach[14, 14]. In their full rigorous forms, these formulations are completely equivalent, but merely alternative of representing the electromagnetic fields inside the grating.

Relevant differential equations have been derived and solved, if necessary by numerical methods. Unfortunately it is the lack of thorough analysis of even simple optical elements fabricated in practical materials and the empirical state of materials research. There would sits to be no simple way to test a structure of such small detail as a recorded holographic grating to determine its material parameters at optical frequencies other than by measuring its diffraction performance. Although the theory of Kogelnik is limited to the description of two diffraction orders, it might be expected that if the grating is in volume type, it would form a good approximation to rigorous analysis close at the Bragg angle.

The equations of zero and first diffracted order, $|S_0|^2$ and $|S_1|^2$ are simplified for the case of Bragg condition as follows.

$$|S_0|^2 = \exp(-2\alpha d/\cos\theta_0)\cos^2(\kappa d//\cos\theta_0) \quad (6)$$

$$|S_1|^2 = \exp(-2\alpha d/\cos\theta_0)\sin^2(\kappa d//\cos\theta_0) \quad (7)$$

$$\kappa = \beta\varepsilon_1/(4\varepsilon_0) \quad (8)$$

$$\alpha = \beta\varepsilon_0''/(2\varepsilon_0),\ \beta = 2\pi(\varepsilon_0)^{1/2}/\lambda \quad (9)$$

d : emulsion thickness

ε_0 : real part of dielectric constant of emulsion

ε_0'' : imaginary part of dielectric constant of emulsion

ε_1 : modulation amplitude of dielectric constant of emulsion

Equation(7) shows that at Bragg incidence efficiency is likely to depend on product of two terms, one representing absorption constant α and one representing the coupling constant κ. Both α and κ depend on exposure as shown in Fig. 2. The material parameters allowed by Kogelnik model are the hologram thickness d, refractive index $(\varepsilon_0)^{1/2}$, imaginary part of dielectric constant ε_0'', phase modulation ratio $\varepsilon_1/\varepsilon_0$, and absorption modulation ratio $\varepsilon_0''/\varepsilon_0$. All other parameters, such as grating period and slant angle, were determined by the recording geometry and knowledge of material physical parameters themselves.

Fig. 4. shows the prediction of diffraction efficiency for coupled wave model in Agfa 8E56 hologram plate simulated by R. R. Syms and L. Solymer[16]. It is known that a hologram formed in bleached photographic emulsion will undergo shrinkage. Emulsion thickness is considered by determination of the hologram thickness as 4.6 $\mu m \sim 4.7\ \mu m$ a shrinkage of 8% from that of a raw 8E56 plate, $5 \mu m$. That shrinkage is caused by removal of unexposed material during processing. However the experimental result in photo 1 is on the contrary to the general notion of emulsion shrinkage.

Fig. 5. shows the prediction of diffraction efficiency with exposure in Agfa 8E75 HD hologram film which uses the different thickness values. Variations of emulsion thickness deeply influence on the diffraction efficiency of bleached phase grating. It is also well known that the average refractive index of a processed hologram formed in bleached photographic emulsion is lower than that of the unexposed film[17]. The refractive index determined for the hologram in Fig. 5. is 1.59 against 1.62 for a raw 8E75 film. The change is caused by local removal of the high refractive index material silver bromide. The maximum transmission at the Bragg angle is 78% if no grating is recorded. Incident angle 25° to the boundary surface gives 15° refraction angle by Snell's law. The zero order transmission of 78% may be mainly attributed to absorption, and thus it might be put $\exp(-2\alpha d/\cos\theta_0) \cong 0.78$ *giving* $\alpha d = 0.12$ at Bragg angle $\theta_0 = 15°$. Of this 78% available transmission power, the first diffraction power 71% is diffracted neglecting any higher orders. Thus it might be $\sin^2(\kappa d/\cos\theta_0) = 0.71/0.78$ giving $\kappa d = 1.265$ using eq. (7). From eq. (8) and eq. (9), $\varepsilon_1/\varepsilon_0$ is calculated as follows:

$$\frac{\varepsilon_1}{\varepsilon_0} = \frac{4\pi}{\beta} = \frac{4\times 1.26/d}{\beta} = \frac{4\times 1.26/d}{2\pi/\lambda(\varepsilon_0)^{1/2}}$$

At $d = 7\ \mu m$, $(\varepsilon_0)^{1/2} = 1.59$ and $\lambda = 0.6328\ \mu m$, dielectric modulation $\varepsilon_1/\varepsilon_0$ is 0.0458. After final chemical processing, swollen emulsion thickness $d = 7\ \mu m$ is identified by SEM photo 1.

Fig. 6. can be plotted by these parameter values and shows the predictions of diffraction efficiency with different dielectric modulation values. Theoretical Bragg angle 15° in the emulsion interior of Fig. 6. is refracted to 25° in the emulsion exterior by Snell's law. That

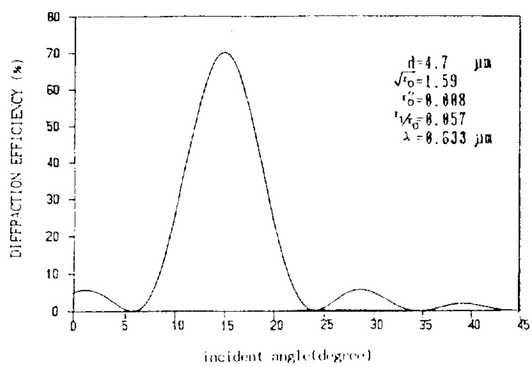


Fig. 4. Predictions of diffraction efficiency by R. Syms and L. Solymer.

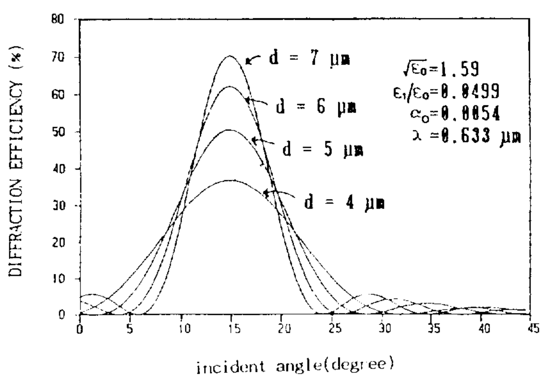


Fig. 5. Predictions of diffraction efficiency vs diffrent emulsion thickness.

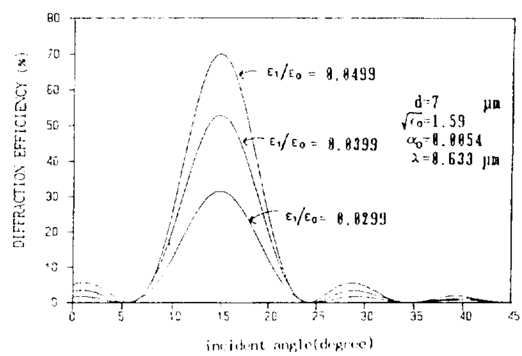


Fig. 6. Predictions of diffraction efficiency vs various dielectric modulation values.

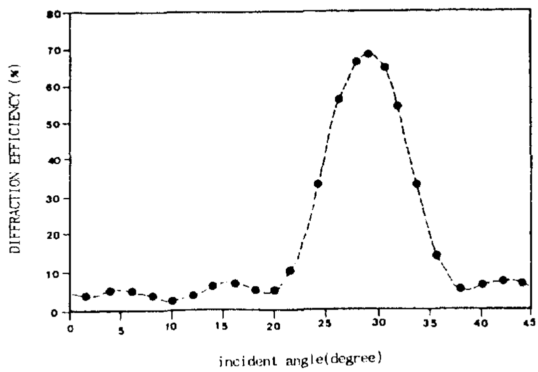


Fig. 7. Diffraction efficiency vs incident angle.

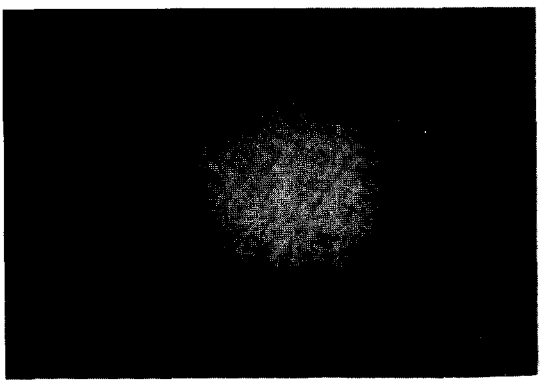


Photo. 2. Spurious interference pattern by boundary reflections.

is, incident beams are recorded at each 25° normal to the boundary surface. But in this experiment Bragg angle is shifted to 28° due to swollen emulsion depicted in Fig. 7. These parameter values should be verified by various precise methods through further research.

Photo 2. shows the presence of a spurious noise generated by reflection from two boundaries and scatter from the silver bromide grains, which lowers diffraction efficiency. If the refractive index of the substrate is matched to that of the emulsion or if the recording system is set up within the index matching liquid, the spurious noise can be reduced.

V. CONCLUSION

Phase volume holograms of diffraction efficiency over 71% can be formed in silver halide of over 1200 lines/mm at an exposure of 180 μJ/cm^2. Transmisson holographic grating are recorded in the Agfa 8E75 HD emulsion, subsequently processed with a nonhardening fixer, a patassium dichromated bleacher and an alcohol drying. The diffraction efficiency of this grating is increased over 20% due to strong modulation using alcohol drying.

High diffraction mechanism is a localized refraction index modulation by cross-linkage of Cr^{+3} ion and emulsion thickness variation by alcohol drying process. The swollen emulsion is newly identified by SEM photography. The modification of parameter values in coupled wave theory is carried out by computer simulation. Consequently, controlling the emulsion thickness has an important role as a potential source of additional modulation leading to higher efficiency.

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홀로그래피 위상형 회절격자의 정성적 해석

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홀로 그래피 은염 감광 물질에 위상형 회절격차를 기록하여 71%가 넘는 회절 효율을 얻었다. 화학적 처리가 끝난 후 전자 주사 현미경(SEM)을 통해 Agfa 8E75 감광유제의 부풀은 모습을 밝혔다. 비경화 정착액, 중크롬산 표백액과 알코올에 의한 급속 건조가 강한 변조효과를 가져와 회절 효율이 20% 이상 증가하였다. Kogelnik 결합파 이론의 중요한 특성 파라미터들에 대해 검토하였으며, 두께 변화에 따른 새로운 파라미터 값을 컴퓨터 시뮬레이션을 통해 제시하였다. 결론적으로 감광유제의 정밀한 두께 제어가 고회절 효율을 얻기 위한 잠재적인 원천으로 작용함을 보였다.