Improved spatial frequency response in silver halide sensitized gelatin holograms

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ABSTRACT

We report what we believe to be the best results obtained to date with regard to the spatial frequency response of silver halide sensitized gelatin (SHSG) holograms. A very high diffraction efficiency, as high as 91% (after allowing for reflection), and an almost flat spatial frequency response between 800 lines/mm and 2800 lines/mm have been achieved using the new BB-640 plates manufactured by Holographic Recording Technologies. The results are compared with those for gratings recorded in the familiar Agfa 8E75 HD emulsion under the same recording and processing conditions. Theoretical results obtained using Buschmann’s weakly scattering model are also compared. Our investigations reveal that high quality SHSG transmission holograms with high spatial frequencies and an almost flat spatial frequency response may be obtained using the new BB-640 plates.

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KEY WORDS: Holography, holographic recording materials, photographic emulsions, photosensitive processing
1. Introduction

The spatial frequency response of the holographic recording material used to produce holographic optical elements (HOE) is one of the most important factors to be taken into account. The obtention of high-efficiency low-noise HOEs depends on the spatial frequency response of the material used for recording the hologram. Dichromated gelatin (DCG) is a well-known recording material with high efficiency at high-frequencies. DCG has so little scattering and absorption that it is almost indistinguishable from clear glass plates. Nevertheless, it has low spectral and exposure sensitivities compared with silver halide emulsions. Silver halide sensitized gelatin (SHSG) has proven to be a good alternative to DCG in the production of transmission holograms. This technique was initially developed by Pennington et al. [1], although the technique, as used today, is the result of the work done by Graver et al. [2] on the one hand, and Chang and Winick [3] on the other. SHSG has the sensitometric and spectral response of silver halide emulsions and the high diffraction efficiency and low noise of DCG. However, one of the main problems involved in the SHSG processing technique is that the spatial frequency response is dependent on the resolution of the photographic emulsion used [4], which is limited by the size of the silver halide crystals. Ferrante [5] has studied this aspect closely and has reported that SHSG suffers from severe roll-off of the modulation transfer function (MTF), finding a significant decrease in MTF when the spatial frequency gets to 2000 lines/mm.

In this paper we demonstrate that these shortcomings are not insurmountable since we have achieved substantial improvement in diffraction efficiency and MTF for BB-640 emulsion from Holographic Recording Technologies GmbH, Germany. Transmission holographic gratings were recorded using the SHSG process, which together with the sensitometric and spectral advantages of standard photographic emulsions has an excellent spatial frequency response between 800 and 2800 lines/mm. A very high diffraction efficiency of 84% (after allowing for Fresnel losses) was obtained for transmission gratings with a high spatial frequency of ~2800 lines/mm. The key to obtaining these results is to use photographic emulsions with small grain sizes such as the emulsion of the BB-640 plates. The comparison of the results obtained using ultra-fine-grained (BB-640) and fine-grained (Agfa 8E75 HD) holographic emulsions are presented and discussed.

BB-640 emulsions have a low grain size between 20 nm and 25 nm [6]. The mean
grain size for the coated Agfa 8E75 HD material is of about 44 nm [7, 8]. However, the sensitivity of BB-640 plates is lower than that of Agfa 8E75 HD plates. In addition, BB-640 emulsions are hardened to a high degree [6] and their gelatin is too hard to be used directly for recording SHSG holograms. We solved the problem of the high degree of the initial hardening of the gelatin emulsion by soaking the unexposed plates in a weak fixer. As a result, all the plates employed in this study were pretreated by immersion in a solution of distilled water with a sodium sulfite concentration of 1% and an urea concentration of 5% (by weight) for 10 minutes at 20°C. The plates were then rinsed in running water for 1 minute and dried (20°C and 60% RH) for 24 hours. This pretreatment produces not only a softening of the gelatin of the BB-640 emulsion, but also an increase in the sensitivity by a factor of ~2.6 and a significant increase in diffraction efficiency.

2. Experiment

To determine the frequency response of SHSG, experiments were carried out with BB-640 plates and Agfa 8E75 HD plates in order to compare the results obtained. Two collimated beams of equal intensity from a He-Ne laser ($\lambda = 632.8$ nm) were used to record a series of unslanted transmission gratings. The two beams interfered to give unslanted fringes with a spatial frequency:

$$f = 2\sin \theta / \lambda$$

(1)

where $2\theta$ is the interbeam angle in air. Gratings were recorded for a range of exposures at interbeam angles $2\theta$ (in air) of 29.4°, 45.3°, 60.2°, 81.6°, 99.0° and 122.2°, yielding spatial frequencies of 802, 1217, 1585, 2065, 2403 and 2767 lines/mm (fringe spacings between 1.25 and 0.36 μm). Four holographic gratings were recorded on each plate for different values of exposure, and two plates were employed for each spatial frequency and type of emulsion. Therefore, experiments for each spatial frequency involved exposure of four plates. Two of these plates corresponded to BB-640 emulsion and the other two plates to Agfa 8E75 HD emulsion. The exposed plates were processed according to the processing schedule shown in Table I. The process that we followed was similar to that of reference 9 but with the temperature of the bleaching bath modified to 70°C. We also included hypersensitization of the plates before exposure, an increase in the time of washing of the
plates after developing and bleaching (in order to eliminate residual chemicals in the layer),
and we dried the processed plates in a dissecator at low relativite humidity for 24 hours
instead of using a vacuum chamber. The developer used in these experiments was Kodak D-
19, a nontanning developer. However it is important to note that oxidation products of this
developer, which have a local tanning action [10], also contribute to the formation of the
latent image [9]. For BB-640 and Agfa 8E75 HD plates diffraction efficiency increases
significantly using hypersensitized plates and a bleach-bath temperature of 70°C. For Agfa
8E75 HD plates this high temperature produces an increase in scattering, however for BB-
640 plates scattering hardly changes when the bleach-bath temperature is increased.

3. Results and discussion

Measurements were made of the diffraction efficiency of these gratings. The
diffraction efficiency η was calculated using the equation:

\[ \eta = \frac{I_1}{I_0} \]  

(2)

where \( I_1 \) is the power in the first diffracted order at Bragg angle and \( I_0 \) is the incident power.
Due to the large difference in the replay angle (\( \theta \) takes values from 14.7° to 61.1°) for the
spatial frequency interval considered, the boundary reflection losses are substantially
different for each replay angle. Therefore an appropriate correction factor must be applied to
achieve a meaningful comparison. All measurements of diffraction efficiency were made in
air and were corrected in order to take into account reflection losses at the boundaries of air-
to-emulsion, emulsion-to-glass, and glass-to-air, using Fresnel coefficients. Both corrected
and uncorrected results for peak diffraction efficiency at Bragg angle as a function of the
spatial frequency are plotted in Fig. 1, where the results for BB-640 and Agfa 8E75 HD
plates are compared. As can be seen from this figure, the maximum diffraction efficiency
obtained for Agfa plates is 83% (73% if we do not consider Fresnel losses) for a spatial
frequency of 802 lines/mm, and falls rapidly when the spatial frequency is above this value.
For a spatial frequency of ~2800 lines/mm, the diffraction efficiency achieved is ~40%. For
BB-640 emulsion, the diffraction efficiency has a higher peak value of 91% for 1585
lines/mm and starts to drop even at a spatial frequency of > 1600 lines/mm. Any further
increase in the spatial frequency results in a slow fall in the diffraction efficiency and a value of 84% was obtained for transmission holographic gratings with a spatial frequency of ~2800 lines/mm. The essential cause of the rapid fall in the diffraction efficiency observed with Agfa plates as the spatial frequency is increased above a critical value is due to the MTF of the photographic emulsion which falls rapidly at high spatial frequencies when the grain size of the emulsion is large [5, 11]. We think that an improved spatial frequency response is obtained for BB-640 plates due to their smaller grain size. In order to confirm this statement, in Fig. 2 we have compared the normalized diffraction efficiency obtained using the experimental data with that calculated theoretically as a function of spatial frequency. We have normalized the diffraction efficiency using the maximum value obtained and shown in Fig. 1 (91% and 83% for BB-640 and Agfa 8E75HD plates, respectively). Theoretical curves correspond to the weakly scattering model proposed by Buschmann [12] and have been obtained using data shown in references 5 and 11. Comparison between theoretical and experimental data indicates greatly reduced SHSG performance at high spatial frequencies for Agfa plates but not for BB-640 plates. For Agfa 8E75 HD emulsion at > 1600 lines/mm, the normalized diffraction efficiency approaches that predicted for a 65 nm mean grain size. The D-19 developer can give a higher diffraction efficiency for relatively low spatial frequencies. This is due to the fact that the oxidation products coming from the hydroquinone, which favour the final differential hardening at low frequencies, turn out to have a negative action for high resolution values [13]. However, for BB-640 plates the normalized diffraction efficiency approaches that predicted for a 25 nm mean grain size, and the spatial frequency response obtained is much better than in the case of Agfa 8E75 HD plates. These results show that it is possible to improve the results obtained for D-19 developer at high frequencies by working with emulsions with smaller grain sizes.

Finally, Fig. 3 shows the comparison between the spatial frequency response for SHSG transmission holographic gratings developed with D-19 developer for Agfa plates (graphs obtained from references 13 and 14) and our results obtained for BB-640 plates. Diffraction efficiencies are corrected taking into account Fresnel losses. As can be seen from this figure, the spatial frequency response for BB-640 plates is the best that has been reported for SHSG holograms to date.
4. Conclusions

In conclusion, our results have demonstrated the possibility of recording high quality transmission holograms in SHSG with a very good spatial frequency response using the novel BB-640 emulsion. At interbeam angles between 29.4° and 122.2° a minimum decrease in the diffraction efficiency has been obtained. The diffraction efficiency for BB-640 plates is greater than 83% (after allowing for reflections) for spatial frequencies between 800 and 2800 lines/mm. These results show that BB-640 plates are a potential replacement for Agfa 8E75 HD material for SHSG holograms when Agfa holographic materials will no longer be produced. The comparison between the experimental results presented in this paper with other results published previously, reveals a significant improvement in spatial frequency response of SHSG transmission holograms. The small size of silver halide grains in BB-640 emulsion, besides increasing the spatial frequency response, enhances the quality of the holograms achieving higher efficiencies and lower values of losses due to absorption and scatter. Finally, the information presented in this paper with regard to the spatial frequency response of BB-640 plates might be useful to researchers working in the areas of display holograms, holographic optical elements and holographic data storage. It is also reasonable to expect that BB-640 plates will offer new possibilities for recording high spatial frequency transmission and reflection holograms in SHSG with diffraction efficiencies higher than the efficiencies reported at the present time for this type of hologram.

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REFERENCES


FIGURE CAPTIONS

Fig. 1.- Corrected and uncorrected results for peak diffraction efficiency of SHSG transmission gratings as a function of spatial frequency for BB-640 and Agfa 8E75 HD plates.

Fig. 2.- Normalized peak diffraction efficiency of SHSG transmission gratings as a function of spatial frequency for BB-640 and Agfa 8E75 HD plates and theoretical curves for mean grain sizes of 25 and 65 nm obtained using Buschmann’s weakly scattering model.

Fig. 3.- Peak diffraction efficiency of SHSG transmission gratings as a function of spatial frequency for BB-640 and the results for Agfa 8E75 HD plates shown in references 13 and 14.

TABLES

Table I.- Processing schedule.
TABLE I

A. Hypersensitization
1. Soak plates in 1% sodium sulfite and 5% urea solution for 10 min at 20°C.
2. Rinse in running water for 1 min.
3. Dry for 24 h at 20°C and RH = 60%.

B. Exposure

C. Development
1. Develop in D-19 for 5 min.
2. Rinse in running water for 2 min.
3. Bleach for 30 s after the plate has cleared at 70°C.
4. Rinse in running water for 2 min.
5. Soak in nontanning fixer F-24 for 2 min.
6. Wash in running water for 10 min.
8. Dehydrate in 50% isopropanol for 3 min.
9. Dehydrate in 90% isopropanol for 3 min.
10. Dehydrate in 100% isopropanol for 3 min.
11. Dry in dissecator for 24 h at 20°C and RH < 20%.

All solutions are at 20°C except the bleaching step.

Bleach formula

Solution A: Ammonium dichromate 20 g
Sulfuric acid 14 mL
Distilled water 1 L

Solution B: Potassium bromide 92 g
Distilled water 1 L

Just before use, mix one part A with ten parts distilled water, then add 30 parts B.
FIG. 1

Peak diffraction efficiency (%)

Spatial frequency (lines/mm)

BB-640
- uncorrected
- corrected

Agfa 8E75 HD
- uncorrected
- corrected