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Spatial frequency dependence of the nonlinear characteristics of bleached silver-halide holographic materials

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Abstract

Complex nonlinear characteristics of a holographic material, i.e., square root of the diffraction efficiency as function of bias exposure, fringe visibility and spatial frequency of plane wave phase holograms recorded in silver halide emulsion have been determined for the first time. Analytical functions were fitted to the measured data and the results were checked using coupled wave theory. These characteristics can be used in the design and evaluation of holographic optical elements.

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Keywords: Holography; Holographic materials; Silver halides; Diffraction theory

1. Introduction

Macroscopic characteristics describing holographic materials are determined by the physical and chemical processes taking place at a microscopic level during and after exposure and during processing. The two principal (but not independent) characteristics of holographic recording materials are the modulation transfer function (MTF) describing the finite spatial resolving power of the material and the Lin-curves (square root of diffraction efficiency of plane wave holograms as function of the bias exposure and the fringe visibility), describing the nonlinearity of the holographic recording.

A great number of valuable papers have been published on the theory of holography, especially in the beginnings (the 1960s and 1970s). However, there remained a gap between the theoretical results and practical holography. A few of the most important papers dealing with the effects of film MTF and nonlinearity will be enumerated in the following.

The first attempts to describe the effect of the MTF of the recording material were based on assuming certain fictious masks or pupils thought to be placed in the object beam at the recording step. It was Van Ligten who first suggested the use of such a

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hypothetical masks [1]. Similar methods have been suggested by Friesem et al. [2], Lukosz [3], Urbach and Meier [4], Kozma and Zelenka [5] and Jannson [6]. However, these papers did not conclude to practically applicable results. Lukosz [3] suggested to make the amplitude transmittance of the above mentioned fictious mask equal to the MTF but he did not give an explicit form of the transmittance. Jannson [6], similarly to the greatest part of experimentalists, assumed a rectangular MTF. However, this usual approximation gives false results, the image quality is usually considerably underestimated. Besides the review paper of Urbach and Meier [4], good summaries of this topic can be found in monographs on holography [7–9].

Nonlinearity of holographic recording materials has an ample literature, too [10–20]. The authors of these works applied binomial and polynomial expansions of various forms to the nonlinear transmittance – exposure function of the recording material. For example the ghost images have been analysed be Tischer [15] on the basis of the expansion of the above function on a set of Chebyshev polynomials. Expansion into Fourier series is described in [16]. Experimental results have been reported by Friesem and Zelenka [10], Goodman and Knight [12] and Denisyuk et al. [16]. Due to approximations applied, none of the above works could give exact methods with universal applicability.

2. Theory

2.1. Modulation transfer function of the recording material

A model of holographic recording, based on the numerical calculation of the double Fresnel–Kirchhoff integral, was developed by the author. The effects of the MTF of the recording material were included in the model, too [21,22].

The author concluded that a function of the Fermi–Dirac form is a very good approximation to the square root of the MTF of a silver halide recording material. If we denote the square root of the diffraction efficiency by σ, it takes on the following form:

$$\sigma = \frac{1}{\exp\left(\frac{v-v_0}{c}\right)+1}, \tag{1}$$

where $v = v(\xi)$ is the local spatial frequency, v_0 is the resolution limit, and c describes the slope of the curve.

Multiplying the integrand of the double Fresnel–Kirchhoff integral [21] by σ we obtain the complex amplitude of the reconstructed image for a recording material of finite resolving power

$$\underline{V}(x,z) = \int_{\xi_1}^{\xi_2}\int_{u_1}^{u_2} \underline{U}(u)\frac{\cos\theta\cos\rho}{r_1 r_2}\frac{1}{\exp\left(\frac{v-v_0}{c}\right)+1} \times \exp(\mathrm{i}k(r_1-r_2))\,\mathrm{d}u\,\mathrm{d}\xi. \tag{2}$$

Note that $v = v(\xi)$, i.e., it refers to the local spatial frequency of the holographic grating at point ξ. It is determined by the angle of incidence of the reference and the object waves.

Some holographic recording materials, such as thermoplastics and photopolymers, have band-limited MTF. The MTF of these materials is usually asymmetrical, but in certain cases even a symmetrical one like a Gaussian function gives a good fit. Results on the extension of the present model to photothermoplastic recording materials were published by the author [23].

2.2. Nonlinear characteristics (Lin-curves) of the recording material

Nonlinear characteristics of silver halide [24] and photothermoplastic [25] holographic recording materials were also included in the diffraction model of holography developed by the author.

The following analytical function was fitted to the measured square root of diffraction efficiency against bias exposure and fringe visibility $[\sigma(E_0, V)]$ curves:

$$\sigma(E_0,V) = f(E_0)(1-\mathrm{e}^{-V})\exp\left\{-\frac{[V-V_0(E_0)]^2}{w^2(E_0)}\right\}, \tag{3}$$

where σ is the square root of the diffraction efficiency, E_0 is the bias exposure, V is the visibility of the interference fringes and $f(E_0)$, $V_0(E_0)$ and $w(E_0)$ are parameter functions of the following form:

$$\mathrm{Par}(E_0) = ci_{01}\left(\frac{1}{e^{\frac{ci_{11}-E_0}{ci_{12}}}+1}+ci_{13}\right)\times\left(\frac{1}{e^{\frac{E_0-ci_{21}}{ci_{22}}}+1}+ci_{23}\right)\times\left(\frac{1}{e^{\frac{ci_{31}-E_0}{ci_{32}}}+1}+ci_{33}\right), \quad (4)$$

where "Par" stands for f, V_0 and w, and c_{ixx} represent three sets of constants ($i = f, V_0, w$).

Combination of Eqs. (1) and (3) yields the full nonlinear characteristics, $\sigma(E_0, V, v)$, of the recording material.

This function was constructed in an empirical way, but it describes well practically all kinds of holographic materials. It gives an excellent fit to the measured Lin-curves of absorption holograms recorded in Kodak 649f spectroscopic emulsions [24,26], in Agfa-Gevaert 10E70 [26] and Agfa-Gevaert 8E75HD emulsion (the same that was used in the present work) [27]. The same empirical function was fitted successfully to Lin-curves of phase holograms recorded in Agfa-Gevaert 8E75HD emulsion, using a number of processing schemes (various combinations of developers and bleaching agents) [28,29]. Besides of the silver halide emulsions this empirical model gives good fits to the measured Lin-curves of photo-thermoplastic holographic materials, too [25]. Lin-curves of other holographic materials have also been measured and fitted by the author, using the same function, and the results will be published in another paper. In reality it is the modulation of the refractive index, Δn that can be related directly to the physical parameters of the recording of phase holograms. Several physical models of the recording of absorption or bleached holograms were developed [2,30–32]. However, no systematic measurements of the $\Delta n(E_0, V, v)$ characteristics of the holographic materials have been performed. Preliminary results on such measurements have been published by the author [33]. Once the $\Delta n(E_0, V, v)$ characteristics of the holographic material are measured and fitted using an adequate physical model, Lin-curves can be derived using coupled wave theory. However, one has to take into account higher-order harmonics in the grating profile and the finite lateral extent of the local plane-wave gratings used for the derivation of the Lin-curves.

Substituting $\sigma(E_0, V, v)$ into Eq. (2) makes it possible to take into account the two most important material characteristics in the evaluation of the reconstructed image.

In the previous papers of the author it was assumed that $\sigma(E_0, V, v)$ is separable, i.e., $\sigma(E_0, V, v) = \sigma_1(E_0, V)\sigma_2(v)$. This may hold true by all probability for photothermoplastic materials [25], but the present experiments showed that in case of silver halide materials this function is not separable.

The aim of the present work was to measure the Lin-curves of a silver halide recording material (Agfa-Gevaert 8E75HD) in a wide range of spatial frequencies, and try to determine the dependence of these characteristics on the recording conditions.

3. Experimental

3.1. Recording of the holograms

Four series of plane wave holograms were recorded in Agfa-Gevaert 8E75HD plates with a helium–neon laser operating at 632.8 nm. The spatial frequencies of the gratings were $v = 800$, 1200, 1600 and 2000 lp/mm.

Holograms at five values of fringe visibility, namely at $V = 0.2$, 0.4, 0.6, 0.8 and 1.0 were recorded. Twelve holograms at exposures ranging from 10 μJ/cm^2 to 1.3 mJ/cm^2 were recorded at each visibility. Care was taken to maintain the same intensity (400 μJ/cm$^2 \pm 1\%$) at recording to minimize the effect of reciprocity failure.

Holograms have been developed with AAC developer (3 min at 20 °C) and bleached in an R-9 solvent bleach without a fixation step.

The composition of the AAC developer was the following:

Ascorbic acid	18 g/l
Sodium carbonate	120 g/l

The composition of the R-9 bleach was as follows:

Sulphuric acid (96–98%, purissimum)	10 ml/l
Potassium dichromate	2 g/l

Bleaching was terminated 1 min after the plate became transparent.

Diffraction efficiency of each hologram was measured. Corrections were made both in the exposure values and in the efficiencies for Fresnel reflection losses.

3.2. Results

Measured values of the square root of the diffraction efficiency as a function of bias exposure and fringe visibility and the $\sigma(E_0, V)$ curves fitted to them using Eqs. (3) and (4) for all the four grating spatial frequencies are shown in Figs. 1–4.

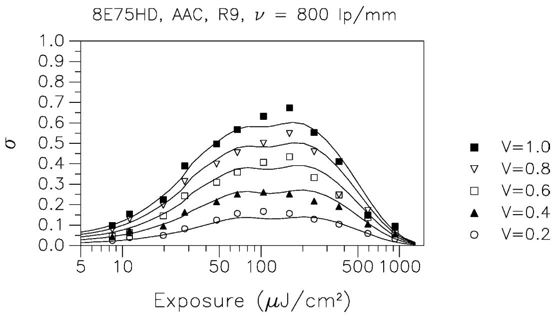

Fig. 1. $\sigma(E_0, V)$ curves of Agfa 8E75HD, $\nu = 800$ lp/mm.

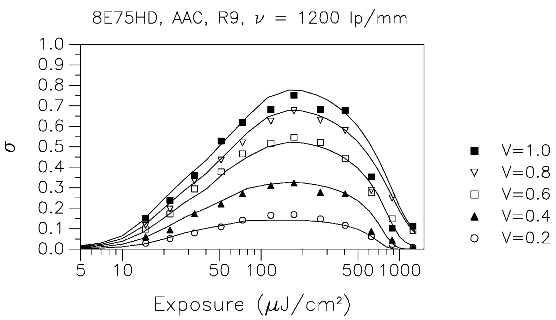

Fig. 2. $\sigma(E_0, V)$ curves of Agfa 8E75HD, $\nu = 1200$ lp/mm.

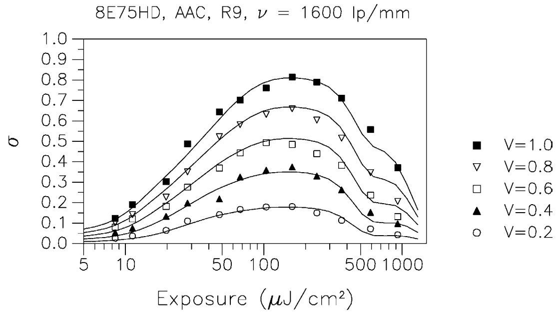

Fig. 3. $\sigma(E_0, V)$ curves of Agfa 8E75HD, $\nu = 1600$ lp/mm.

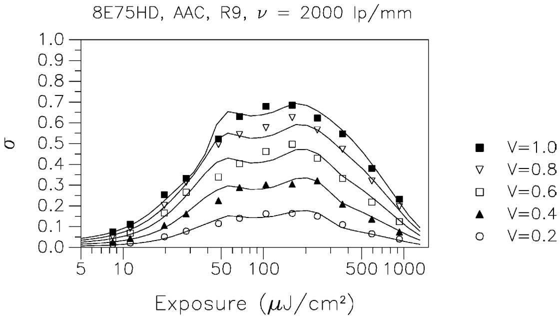

Fig. 4. $\sigma(E_0, V)$ curves of Agfa 8E75HD, $\nu = 2000$ lp/mm.

Parameters of the fitted analytical functions are shown in Tables 1–4.

As it can see from Figs. 1–4, measured data could be fitted quite well with this process of fitting 27 parameters in two steps at each spatial frequency. To have an idea of the spatial frequency dependence of σ, square root of the diffraction efficiency as a function of bias exposure at two

Table 1
Parameters of the fitted $\sigma(E_0, V)$ function, $\nu = 800$ lp/mm

Indices (xx)	c_{fxx}	$c_{(V_0)xx}$	c_{wxx}
01	1.7	3.0	3.0
11	28	62	55
12	26	19	12
13	0	0.18	0.22
21	285	60	50
22	240	60	30
23	0	0.3	0.6
31	10.8	28	28
32	6.5	−1.8	−1.8
33	0	1.24	1.34

Table 2
Parameters of the fitted $\sigma(E_0, V)$ function, $v = 1200$ lp/mm

Indices (xx)	c_{fxx}	$c_{(V_0)xx}$	c_{wxx}
01	1.5	3.0	3.0
11	38	72	20.5
12	32	2.5	3.0
13	0	0.21	0.95
21	675	53.5	700
22	300	2.8	120
23	0	0.15	0.27
31	12.5	28	4
32	3.2	−1.8	−21
33	0	1.64	0.14

Table 3
Parameters of the fitted $\sigma(E_0, V)$ function, $v = 1600$ lp/mm

Indices (xx)	c_{fxx}	$c_{(V_0)xx}$	c_{wxx}
01	1.7	3.0	3.0
11	30	24	24
12	25	15	7.5
13	0	0.2	0.2
21	800	320	370
22	260	400	80
23	0	0.4	0.45
31	6.88	9	9
32	5.9	−2.5	−2.5
33	0	0.68	0.68

Table 4
Parameters of the fitted $\sigma(E_0, V)$ function, $v = 2000$ lp/mm

Indices (xx)	c_{fxx}	$c_{(V_0)xx}$	c_{wxx}
01	1.5	3.0	3.0
11	38	24	34
12	19	15	7.5
13	0	0.2	0.2
21	250	200	350
22	190	110	45
23	0	0.4	0.36
31	6.88	9	9
32	5.9	−2.5	−2.5
33	0	0.68	0.68

fixed fringe visibilities ($V = 0.2$ and 0.8), and with the grating spatial frequency as a parameter are presented in Figs. 5 and 6. These figures show that the dependence of σ on the grating spatial frequency, v, cannot be separated from the dependence on bias exposure and fringe visibility.

As, according to the theory of volume gratings, the greatest part of the recorded holograms is in

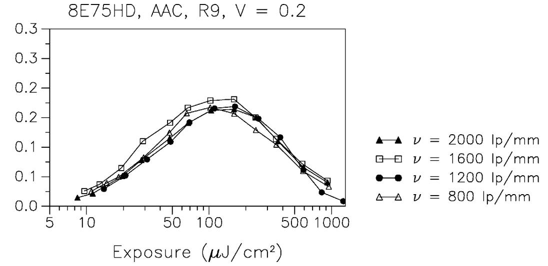

Fig. 5. Square root of the measured diffraction efficiency vs. exposure at $V = 0.2$, v is parameter.

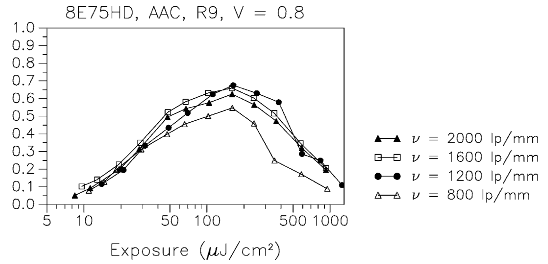

Fig. 6. Square root of the measured diffraction efficiency vs. exposure at $V = 0.8$, v is parameter.

the intermediate or in the Bragg regimes, coupled wave theory for Bragg gratings [34] can be used for the determination of the diffraction efficiency. So one can use the following equation [35] to determine the dependence of σ on v:

$$\sigma(v) = \left| \sin \frac{\pi n_1 d}{\lambda \cos\left[\arcsin(v\lambda/2)\right]} \right|, \tag{5}$$

where v is the spatial frequency of the grating, n_1 is the modulation of the refractive index, d is the hologram thickness and λ is the wavelength of the light.

Eq. (5) was fitted to the measured values of σ at three values of the bias exposure: $E_0 = 48$, 160 and 590 $\mu J/cm^2$. The parameters searched for were the amplitude of refractive index modulation, n_1, and the thickness of the hologram, d. Fitting the above function to both the "raw" measured data and the already fitted (by Eqs. (3) and (4)) "smooth" values of σ have been performed. Using the fitted values of σ resulted in bad correlations, sometimes down to $R = 0.2$ or 0.1, due to the small systematic errors introduced by the complicated fitting

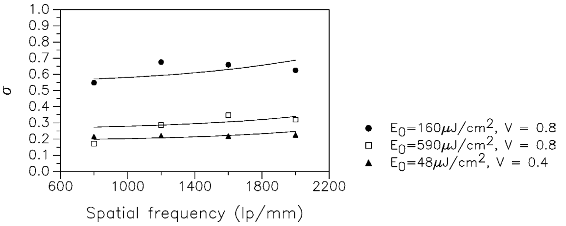

Fig. 7. Square root of the diffraction efficiency vs. spatial frequency of the grating at fixed bias exposure and fringe visibility (both indicated in the graph). Lines are curves fitted using Eq. (5).

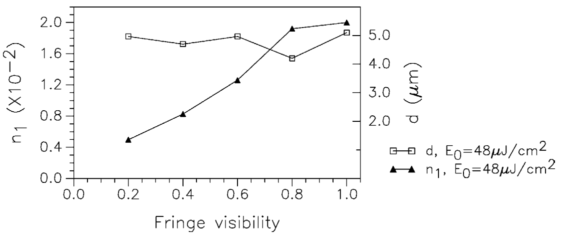

Fig. 8. Fitted values of the amplitude of refractive index modulation and hologram thickness vs. fringe visibility at a fixed value of bias exposure.

process. Using the measured data gave better correlations, always above 0.7, typically around 0.95. Results using measured values of σ are shown in Fig. 7. (σ vs. v with E_0 and V as parameters).

Values of n_1 and d, obtained by fitting Eq. (5) to the experimental data are shown in Fig. 8. These curves corroborate well the consistency of our results, since both the absolute values of n_1 and its quasi-linear dependence on fringe visibility, as well as the constant value of about 5 μm of the deduced hologram thickness correspond very well to the same data obtained either via direct measurement of the phase profile of the gratings [33] or via measuring their angular selectivity.

4. Conclusion

As far as the author can know, the first ever attempt was made to determine the *full nonlinear characteristics*, i.e., the $\sigma(E_0, V, v)$ function of a holographic recording material. A great number of plane wave holograms (240) were recorded, processed and reconstructed under very strict experimental conditions. Full ranges of exposures and fringe visibilities (at recording) were covered, as well as a reasonably wide range of grating spatial frequency. The analytical function, proposed by the author for the description of the nonlinear characteristics of holographic recording materials, proved to be adequate for fitting all the measured characteristics. It was found that the $\sigma(E_0, V, v)$ function is not separable; it depends on grating spatial frequency in a rather complicated way. However, the formula derived by Kogelnik for the diffraction efficiency of volume transmission phase gratings proved to yield consistent results for the amplitude of the refractive index modulation and the hologram thickness. So dependence of the square root of the diffraction efficiency on the grating spatial frequency is given by this formula, i.e., σ is proportional to the absolute value of the sine of the reciprocal of the cosine of the arcus sine of v (Eq. (5)).

Nonetheless, one has to take into account that, depending on the kind of processing we are using, both the refractive index modulation and the hologram thickness can vary with the grating spatial frequency. But, if we know (from direct measurements) the $\Delta n(E_0, V, v)$ function of the material [33] we can calculate the full nonlinear characteristics using the Kogelnik formula.

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