Modulation spectra of volume holograms recorded in additively colored fluorite crystals

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Abstract. Additively colored fluorite crystals are of considerable interest as a holographic recording medium. Due to very high stability as well as potentially large refractive index modulation, it is possible to use holograms recorded in these crystals as high quality diffractive elements and photonic-crystalline structures. Therefore, it is important to be able to correctly and accurately analyze such structures. Spectral dependencies of absorption coefficient and refractive index profiles are the most precious characteristics in this regard. Unfortunately, they are also the most elusive ones. In this study the required parameters were obtained by a method based on the analysis of selectivity hypercontours (combined spectral and angular dependencies of diffraction efficiency) using the Kogelnik’s coupled wave theory in wide optical spectral range. Hologram profile was reconstructed from first three spatial harmonics. The results were crosschecked with monochromatic analysis to prove the validity of proposed technique.

1. Introduction
In the modern world holography has serious relevance in terms of its applications. Volume holograms can be used as narrowband spectral selectors, holographic prisms, beam combiners, holographic memory elements, and, if provided refractive index modulation is sufficient, as photonic-crystalline structures. A holographic medium for such applications must possess certain qualities. First of all, it should be stable enough to remain for a long time and persist a strong energy impact. Also, it must have the variability on the modulation of the refractive index along the spectrum. It should be also possible to make a sample thick enough for low-frequency applications. Finally, in the case of large physical thickness that ensures really high selectivity, moderation of refractive index modulation may be sufficient (about $10^{-5}$ for narrowband spectral needs and from $10^{-3}$ for photonic crystals production).

Additively colored crystals of calcium fluoride ($CaF_2$), in which holograms are recorded through phototransformation and photoinduced displacement of color centers, matches those requirements because of very large thickness and extreme stability. The ability of tuning refractive index and absorbance modulation by means of nondestructive post-exposure conversion of the centers makes $CaF_2$ one of the most promising photosensitive media. Moreover, last studies indicate the possibility of forming metal holographic structure in $CaF_2$ if concentration of color centers is sufficient, thus pointing out the chance to achieve a significant modulation of refraction [1].
Certainly, careful and accurate analysis of hologram properties is necessary for such high-precision applications. From this standpoint, the hologram profiles - spatial distributions of its optical parameters across the grating - are of significant value. The profiles can be obtained by means of optical microscopy or reconstructed basing on coupled-wave analysis of angular selectivity contours [2, 3]. Despite obtaining hologram profiles for a single wavelength is not tough, same task for wide spectral range, which is so necessary for the completeness of the study, has not yet acquired an optimal realization [4, 5]. This work describes the technique capable of obtaining rather complete angular-spectral-spatial dependencies of volume holograms parameters.

2. Methods

2.1. Coupled Wave Theory Application

Since the aim of the work is to obtain information on the profiles of holograms, it is necessary to choose the correct method for their analysis. Coupled wave theory approximation approach has proved its accuracy and reliability in volume hologram analysis as it provides inartificial access to both amplitude and phase modulation components as well as phase shift between them [6, 7]. Coupled wave theory calculates angular and spectral diffraction response from hologram properties. Therefore, it can be applied to determine hologram properties from spectral and angular selectivity contours by means of an approximation procedure.

The first mathematical expressions of coupled wave theory were presented by Hervig Kogelnik in 1969 [8], and they were developed rigorously since then [9, 10]. The amplitude-phase nature of the hologram is revealed in the Borrmann effect [11] (zeroth order diffraction enlargement at the Bragg angle for pure amplitude holograms, and shifted with respect to the Bragg angle - for mixed ones [12, 13]) which can be best seen on model angular selectivity contours (figure 1).

![Figure 1](image)

Figure 1. The shape of all simulated 1st order (dashed) selectivity contours looks the same; information about amplitude-phase nature of hologram can be extracted from 0th order (solid).

2.2. Contours Measurement Technique

The main experimental technique broadly used for recording angular selectivity of volume holograms is generally based on their rotation with respect to a collimated light beam [6, 12, 13, 14, 15] and broadly used for last decades. Sufficiently low beam divergence of about 0.1 mrad can be achieved by means of a collimator and a coherent light source. To study spectral selectivity, either wavelength-tunable or broadband source with a monochromator or spectrum analyzer become necessary. Tunable or broadband lasers (such as supercontinuum generators) are not always affordable, while proper collimation of non-laser white-light source may be challenging.
Reading a grating with a beam whose divergence exceeds the grating’s angular selectivity requires complicated deconvolution procedure at the stage of data analysis, which makes the approach fully impracticable. Therefore, despite dramatic loss of light energy, proper collimation of incoherent source seems to be the most digestible option. Since key information on hologram properties can be extracted from zero order diffraction, it is sufficient to measure only the transmitted light intensity.

2.3. Experimental Realization

Hologram in question was recorded with CW DPSS Nd:YAG laser (532 nm) in an additively colored fluorite (CaF$_2$) crystal at 190°C with spatial period of 4.55 µm. For measuring both spectral and angular selectivity contours a bright white-light LED (CREE XM-L T6) was employed as a light source. Its light was collimated with two sequential telescopic beam expanders to bring the divergence down to about 0.1 mrad. The beam passed through the hologram under incidence angle varied with a rotation stage. Transmittance spectra were recorded at each angular step with a matrix spectrometer (Ocean Optics USB4000-UV-VIS) and, divided by LED emission spectrum, finally were combined into 3D surfaces of zeros order diffraction efficiency plotted vs. angle and wavelength - spectral-angular (spectrangular) selectivity contour (hypercontour) (figure 2).

![Figure 2](image_url)

Figure 2. Three ”valleys” with lifted edges at hologram zero order diffraction hypercontour manifest nonlinear phase-amplitude recording.

The hypercontour was approximated by coupled wave theory expressions as a series of angular selectivity contours along the spectrum. In this work we use the formulae previously discussed in [16]. Angular selectivity contours shape at different wavelengths (figure 3) shows how the hologram appears a mixed one with absorptive and refractive contributions in counterphase (a) and in-phase (c), purely amplitude (b), and almost phase one (d), depending on reading wavelength. The depicted contours almost coincide with the simulated model ones in figure 1.
Figure 3. Approximation of angular selectivity contours for the hologram allowed to determine the values of absorption and refraction modulation amplitudes (figure 4).

3. Results

3.1. Hypercontour Analysis

Fitting with the expressions of the coupled wave theory was consistently applied to each angular selectivity contour. Therefore, refractive index and absorption coefficient modulation amplitudes were determined for the 1st spatial harmonic of the hologram along the studied spectrum with 1 nm-step (figure 4).

Figure 4. Spectral dependencies of 1st harmonics of absorbance and refraction modulation determined as selectivity hypercontour fit-parameters confirm previous observations in figure 3.
3.2. Profile Reconstruction

Since each diffraction order corresponds to a certain spatial harmonic of a volume hologram, the refractive index and absorption coefficient amplitudes of corresponding harmonics can be obtained by approximation of zero order diffraction contour in the vicinity of the given diffraction order. Therefore, spectra of amplitudes were obtained for the first three spatial harmonics whereupon hologram profile at different wavelengths was reconstructed by partial Fourier synthesis (figure 5).

![Figure 5](image_url)

**Figure 5.** Spatial profiles of hologram absorbance and refraction reconstructed as a sum of three first harmonics demonstrate how the phase and amplitude contributions change with wavelength.

3.3. Results Crosscheck

To confirm validity of the proposed technique, the particular angular selectivity contours were re-measured using laser radiation. For this crosscheck two different laser sources were used: CW DPSS Nd:YAG laser at 532 nm where hologram is almost amplitude and CW He-Ne laser at 633 nm where hologram looks an in-phase mixed one with predominant phase contribution.

![Figure 6](image_url)

**Figure 6.** The coincidence of selectivity contours measured with monochromatic (laser) light and obtained with white light (LED) source confirms the validity of proposed technique.
4. Conclusions
In the course the study, experimental setup and approximation algorithm for automated acquisition and analysis of volume hologram selectivity hypercontours were designed. An exemplary transmission hologram in $\text{CaF}_2$ was analyzed by means of this algorithm, so that the spectral distributions of the parameters were obtained. The hologram demonstrated change of phase-amplitude nature along the spectrum as well as evidence of nonlinear recording which was revealed in the appearance of higher diffraction orders. Series of hologram profiles with wavelength scan were reconstructed from first three spatial harmonics of the hologram.

As an approval of the method, crosscheck measurements of monochromatic angular selectivity contours were made by means of laser sources at two different wavelengths. Therefore, the proposed spectral-angular (spectrangular) technique allows fast and accurate determination of spectral characteristics of materials and holographic structures recorded in them. The method is a promising tool for analysis of elementary holograms, and it deserves further development to be applied to more complex holographic structures, such as holographic photonic crystals.

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