ANGULAR DEPENDENCE OF DIFFRACTION EFFICIENCY OF A DYNAMIC HOLOGRAM IN A REVERSIBLE PHOTOCHROMATIC MEDIUM

V.V. Ivakhnik, V.I. Nikonov, E.V. Shilnikova
Samara State University, Russia

Abstract

Angular characteristics of a dynamic hologram, recorded by a "fan" of plane waves in a reversible photochromatic medium, are studied under various correlations between the intensities of these waves. It is shown that the maximum increase of the relative diffraction efficiency with the increase of the angle between the reference and object waves is observed for thin holograms. There is such a PCM thickness beginning from which the relative dependence of the hologram diffraction efficiency upon the angle ceases to depend upon the PCM thickness, reaching some maximum distribution. Coincidence is shown of the angular characteristics of the holograms, recorded under the condition that the reference wave intensity is much greater than the object wave intensity, and of the holograms, recorded by a "fan" of plane waves of equal intensity. The hologram diffraction efficiency change is studied under the readout wave deviation from the Bragg angle.

The reversibility of photochemical transformations in a number of photochromatic media (PCM) allows them to be used as non-linear media for a dynamic hologram recording [1-4]. In the works, accomplished up to now, on studying the dynamic hologram diffraction efficiency (HDE) in reversible PCM it has been considered that hologram recording is carried out by two plane waves, the surfaces of equal intensity of the recorded interference grating being perpendicular to the photochromatic layer faces [5-6]. In the present work the angular HDE dependence is investigated, knowing which, one can estimate the possibility of recording in such media the holograms of complex spatially modulated fields.

1. Hologram recording

Let us consider a "two-level" PCM model which is traditional for holography. A photochromatic medium consists of photochromatic particles which can be either in a form A, or in a form B. Transition of a photochromatic particle from the form A into the form B takes place under the action of radiation at the wavelength λ_1. The reverse transition of the photochromatic particle is carried out either under the action of radiation at the wavelength λ_2 or spontaneously, owing to thermal processes.

Let us designate by n_A, n_B the concentration of photochromatic particles in the forms A and B, respectively. We shall assume that non-photochromatic products are absent, that is the total concentration of photochromatic particles $n_0 = n_A + n_B = \text{const}$. Then the kinetic equation describing the time variation of photochromatic particles concentration in the form A is

$$\frac{dn_A}{dt} = -w_A n_A + w_B \left(n_0 - n_A\right), \qquad (1)$$

where w_A and w_B – the probabilities of photochromatic particles transition from the form A into the form B and from the form B into the form A, respectively. If the transition from one form into another is carried out under the influence of radiation, then $w_A = \eta I_A$, $w_B = \xi I_B$. Here I_A and I_B – the intensities of radiations at the wavelengths λ_1 and λ_2, ξ and η - the constants determining the speed of photochromatic particles transition from the form A into the form B and back.

Let a "fan" of plane waves at the wavelength λ_1 be incident recording the hologram on a PCM layer, located between planes $z = 0$ and $z = L$ (fig. 1). The wave equation describing the propagation of these waves in the PCM is

$$\left(\nabla^2 + k^2 - ik\alpha\right)A = 0\,, \qquad (2)$$

where $A = \sum_{j=-N}^{N} A_j(z)\exp\left(-i\vec{k}_j\vec{r}\right) + \text{c. c.}$; A_j and $\vec{k}_j$ – the amplitudes and the wave vectors of the waves recording a hologram; $\alpha = \beta_1 n_A$ – the absorption coefficient; β_1 – the radiation absorption cross section at the wavelength λ_1; $k^2 = k_j^2$; $\vec{r}\{x,z\}$ – the radius vector.

In the case of recording a stationary hologram, for the steady-state recording regime, the equation (2) breaks up into a system of coupled differential equations describing the intensity change of the waves recording the hologram

$$\frac{k_{jz}}{k}\frac{dI_j}{dz}\left\{1 + \frac{\eta}{w_B}\sum_{i=-N}^{N} I_i\right\} + I_j\left[\frac{\eta}{w_B}\sum_{\substack{i=-N \\ i\neq j}}^{N}\frac{k_{iz}}{k}\frac{dI_i}{dz} + n_0\beta_1\right] = 0\,, \qquad (3)$$

$$j = -N \div N\,,$$

where $I_j = A_j A_j^*$; k_{jz} – the projection of the wave vector $\vec{k}_j$ onto the Z - axis.

If the reverse transition of photochromatic particles from the form B to the form A takes place owing to the action of heat, then $w_B(\vec{r}) = \text{const}$. If the reverse transition of photochromatic particles from the form B to the form A takes place as the result of the PCM being influenced by the radiation of the wavelength λ_2, propagating along the Z - axis, then the transition probability change of photochromatic particles from the form B to the form A across the PCM thickness is described by the equation

$$\frac{dw_B}{dz}\left\{w_B+\eta\sum_{j=-N}^{N}I_j\right\}+n_0\beta_2\eta w_B\sum_{j=-N}^{N}I_j=0\,. \quad (4)$$

Here β_2 – the radiation absorption cross section at the wavelength λ_2.

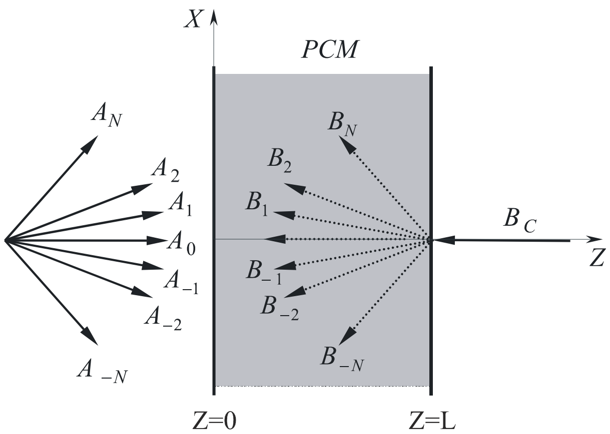

Fig. 1. Hologram recording – readout scheme

2. Hologram read-out

Let us read out the hologram by a plane wave at the wavelength λ_1 propagating along the Z - axis to meet the recording wave with a wave vector $\vec{k}_0$. We assume that the propagation of the reading and restored waves in the hologram does not change the absorption coefficient distribution across the PCM thickness.

Let us again make use of the wave equation (2). We shall replace the amplitude A by the amplitude $B=\sum_{j=-N}^{N}B_j(z)\exp\left(-i\vec{k}'_j\vec{r}\right)+c.c.$ (B_j and $\vec{k}'_j$ – amplitudes and wave vectors of the reading wave $(j=0)$ and the waves restored from the hologram $(j\neq 0)$). Taking into account, that $k'_{jz}=-k_{jz}$, we receive a system of coupled equations describing the variation of amplitudes of the reading and restored waves across the hologram thickness

$$\frac{k_{jz}}{k}\frac{dB_j}{dz}\left\{1+\frac{\eta}{w_B}\sum_{i=-N}^{N}I_i\right\}+\frac{\eta}{w_B}\sum_{\substack{i=-N\\ i\neq j}}^{N}\frac{k_{iz}}{k}\frac{dB_i}{dz}\sqrt{I_iI_j}-$$

$$-\frac{1}{2}n_0\beta_1B_j=0, \quad (5)$$

$$j=-N\div N\,.$$

The equation (5) can be generalized for the case when the propagation direction of the reading wave deviates from the Bragg angle by the $\delta\theta$ angle

$$-\frac{dB_0}{dz}\left(1+\frac{\eta}{w_B}\sum_{i=-N}^{N}I_i\right)+\frac{\eta}{w_B}\sum_{\substack{i=-N,\\ i\neq 0}}^{N}\frac{k''_{iz}}{k}\frac{dB_i}{dz}\sqrt{I_0I_{-i}}+$$

$$+\frac{1}{2}n_0\beta_1B_0+in_0\beta_1\frac{\eta}{w_B}\sum_{\substack{i=-N,\\ i\neq 0}}^{N}B_i\Gamma_i\sqrt{I_0I_{-i}}=0,$$

$$\frac{k''_{jz}}{k}\frac{dB_j}{dz}\left(1+\frac{\eta}{w_B}\sum_{i=-N}^{N}I_i\right)+\frac{\eta}{w_B}\sum_{\substack{i=-N,\\ i\neq j}}^{N}\frac{k''_i}{k}\frac{dB_i}{dz}\sqrt{I_iI_{-j}}+ \quad (6)$$

$$+\frac{1}{2}n_0\beta_1B_j+in_0\beta_1B_j\Gamma_j\left\{1+\frac{\eta}{w_B}\sum_{j=-N}^{N}I_j\right\}=0,$$

$$j=-N\div N.$$

Here $\Gamma_j=\frac{k^2-k''^2_j}{2n_0\beta_1}=\tilde{\Gamma}\sin j\theta_0$, $\tilde{\Gamma}=\frac{k\delta\theta}{n_0\beta_1}$, θ_0 – the angle between the waves recording the hologram, $\vec{k}''_j=\vec{k}'_0-\left(\vec{k}_0-\vec{k}_{-j}\right)$.

The systems of equations (3) - (5) are to be added by the following boundary conditions: for the waves recording the hologram

$$I_j(z=0)=I_{j0}\,,$$

for the reading wave

$$B_0(z=L)=1\,,$$

for the waves restored from the hologram

$$B_j(z=L)=0\,.$$

3. Discussion

While analysing the recording of the dynamic hologram in a reversible photochromatic medium, we shall consider 2 cases.

3.1. The intensity of one recording wave (of a zero-wave) is much greater than the intensities of other waves ($I_0 >> I_j$)

Under such an approach it is necessary to take into account only the gratings that arise at the zero-wave interference with other waves. The zero-wave acts as the reference wave, and the j-th wave as the object wave.

The system of equations (3) comes to two equations: the first one describes the zero-wave intensity change, the second – the waves propagating at the 0 - angle to the zero-wave:

$$\begin{cases}\dfrac{dI_0}{dz}\left\{1+\dfrac{\eta}{w_B}I_0\right\}+n_0\beta_1I_0=0\\[2ex] \dfrac{k_z}{k}\dfrac{dI}{dz}\left\{1+\dfrac{\eta}{w_B}I_0\right\}+I_j\left\{n_0\beta_1+\dfrac{\eta}{w_B}\dfrac{dI_0}{dz}\right\}=0\end{cases} \quad (7)$$

Here $I=I_j$, $k_z=k_{jz}$.

Neglecting the exhaustion of the reading wave, the system of equations (6) will take the form

$$\frac{dB_0}{dz}\left(1+\frac{\eta}{w_B}I_0\right)-\frac{1}{2}n_0\beta_1 B_0=0,$$

$$\frac{k_z''}{k}\frac{B'}{dz}\left(1+\frac{\eta}{w_B}I_0\right)+\frac{\eta}{w_B}\frac{dB_0}{dz}\sqrt{I\ I_0}+\frac{1}{2}n_0\beta_1 B'+ \quad (8)$$

$$+in_0\beta_1 B'\Gamma\left(1+\frac{\eta}{w_B}I_0\right)=0,$$

where $B' = B_j$, $k_z'' = k_{jz}''$.

The numerical analysis of the systems of equations (7) and (8), taking into account (4) and the boundary conditions, shows that depending upon the correlation of probabilities of photochromatic particles transition from the form A to the form B and vice versa on the front face of the photochromatic layer

$$\left(M=\frac{\eta\sum\limits_{j=-N}^{j=N}I_j(z=0)}{w_B(z=0)}\right) \text{ at } \delta\theta=0$$

with the growth of the PCM thickness the diffraction efficiency of the hologram ($h = B'B'^*$) either first increases and then decreases ($M < M_{cr} = \frac{\beta_1 k}{\beta_2 k_z}$) (fig. 2, curve 1) or increases, reaching a constant level ($M > M_{cr}$) (fig. 2, curve 2). At $\delta\theta \neq 0$ and a fixed angle between the reference and the object waves with the PCM thickness growth we observe the oscillating character of the hologram diffraction efficiency change (fig. 2, curve 3). For $M > M_{cr}$ with the PCM thickness growth the oscillation amplitude decreases and HDE again reaches a constant level, the value of which depends on parameter $\tilde{\Gamma}$.

The relation given in fig.2 and subsequent figures correspond to dynamic hologram recording in a reversible photochromatic medium, consisting of a phochromatic particle of 10 nm in size. The hologram recording is carried out by a 0.44 mcm wavelength radiation. The hologram is erased by a 0.53 mcm wavelength radiation.

Fig. 3 illustrates the dependence of the relative diffraction efficiency of the hologram $\left(\tilde{h}=\frac{h(\theta)}{h(\theta\to 0)}\right)$ upon the angle between the reference and the object waves at the fixed values of parameter M and various hologram thicknesses. The greatest variation of the relative HDE upon the angle Θ is observed for thin holograms ($L \to 0$). At $M < M_{crN} = \frac{\beta_1 k_{Nz}}{\beta_2 k}$ with the increase of the angle the relative HDE grows with any PCM thickness. At $M > \beta_1/\beta_2$ the increase of the relative hologram diffraction efficiency depending upon the angle is observed only in thin holograms. With the hologram thickness increasing at $M > \beta_1/\beta_2$, the relative HDE first ceases to change with the angle, and then begins to decrease with the increase of the angle. There is such a PCM thickness beginning from which the relative dependence of HDE upon the angle θ ceases to depend upon the PCM thickness, reaching some limiting distribution. The existence of this limiting distribution cannot be accounted for by assuming that the increase of the angle θ results in the increase of the effective thickness of the hologram $L_{eff} = L/\cos\theta$.

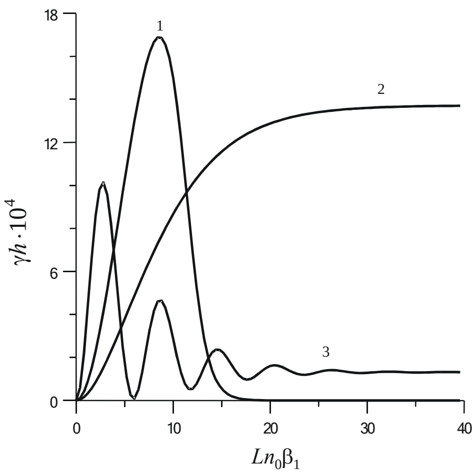

Fig. 2. Hologram diffraction efficiency dependence upon PCM thickness at $\theta = 15^\circ$ *,* $\beta_2/\beta_1 = 0.2$ *;* $\gamma = 1$ *,* $M = 4$ *,* $\tilde{\Gamma} = 0$ *(1);* $\gamma = 1$ *,* $M = 8$ *,* $\tilde{\Gamma} = 0$ *(2);* $\gamma = 15$ *,* $M = 8$ *,* $\tilde{\Gamma} = 4$ *(3)*

Under the fixed PCM thickness the deviation from the Bragg angle of the reading wave results, first of all in the intensity diminution of the waves falling on the PCM at large angles. Therefore, with the growth of parameter $\tilde{\Gamma}$ the change speed of the relative hologram efficiency depending upon the angle θ first decreases reaching zero and then, having changed the sign, starts to increase (fig. 4). To characterize the dynamic hologram, the diffraction efficiency of which decreases with the increase of the angle between the reference and object waves, we shall introduce the notion of a band of angles. Within the band of angles the HDE changes from the maximum value to half the maximum value. The width of the band of angles $(\Delta\theta)$ is determined from the condition

The numerical analysis of (7-9), taking info account (4) and the boundary conditions, demonstrates that the width of the band of angles is inversely proportional to the parameter describing the deviation of the reading wave from the Bragg angle

$$\Delta\theta \sim 1/\tilde{\Gamma} .$$

Thus, changing the propagation direction of the reading wave, it is possible to controllably change the width of the hologram band of angles.

$$B(\theta = \Delta\theta)B^*(\theta = \Delta\theta) = \frac{1}{2}B(\theta = 0)B^*(\theta = 0). \quad (9)$$

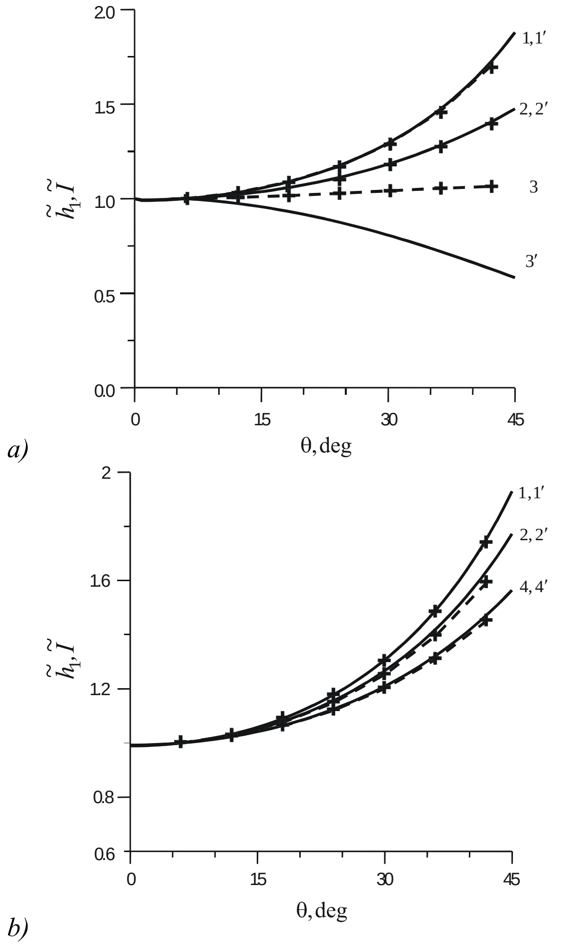

Fig. 3. Angular dependence of the relative intensity of the waves, restored from the hologram at $\beta_2/\beta_1 = 0.2$ and $M = 4$ (a), 8 (b) $Ln_0\beta_1 = 1\,(1, 1')$; $Ln_0\beta_1 = 5\,(2, 2')$; $Ln_0\beta_1 = 15\,(3, 3')$; $Ln_0\beta_1 = 25\,(4, 4')$

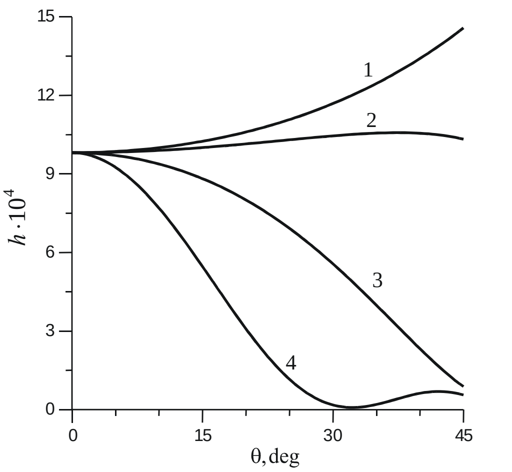

Fig. 4. Angular dependence of the hologram diffraction efficiency, taking into account the deviation from the Bragg angle at $M = 4$, $Ln_0\beta_1 = 5$, $\beta_2/\beta_1 = 0.2$. $\tilde{\Gamma} = 0$ (1), $\tilde{\Gamma} = 0.4$ (2), $\tilde{\Gamma} = 1$ (3), $\tilde{\Gamma} = 2$ (4)

3.2. The intensity of all the waves recording the hologram is equal on the front face of the PCM ($I_0 = I_j$)

In this case taken into account are all the gratings arising under interference of all the waves recording the hologram. Let us consider the intensity dependence of the waves restored from the hologram upon the direction of restoration. Suppose that the total intensity of the waves recording the hologram does not change with varying the number of the waves.

The analysis of the systems of equations (3) - (5) demonstrates, that under a fixed PCM thickness and a fixed value of parameter M there is a-dependence-upon-the-angle-curve of the relative change of the intensity of the waves restored from the hologram, $\left(\tilde{I}(\theta) = \dfrac{I_j h(\theta = \theta_0)}{I_1 h(\theta \to 0)}\right)$, the discrete values of the relative intensity of the waves restored from the hologram under certain angles exactly superimposing upon the curve. The shape of this curve does not depend on the number of the waves and on the angle between the waves recording the holograms.

The comparison of the curves $\tilde{h}(\theta)$ and $\tilde{I}(\theta)$ shows that these curves coincide well under any hologram thickness for the values of parameter $M < M_{crN}$ (fig. 3a) and when considering thin holograms for the values of parameter $M > \beta_1/\beta_2$ (fig. 3b). With the PCM thickness growing at $M > \beta_1/\beta_2$, the relative HDE becomes less in the case of hologram recording by waves of equal intensity than the relative HDE recorded under the same parameters by waves of different intensity.

Under the fixed total intensity of the waves recording the hologram the increase of their number results in modulation depth diminution of the interference pattern and, as a consequence of it, in modulation depth diminution of the grating being recorded. The gratings arising under the interference of the j-th and the k-th waves ($j, k \neq 0$) additionally diminish the modulation depth of the interference pattern. Thus, the modulation depths practically coincide of photochromatic particles concentration gratings under the diffraction on which there arise some waves propagating under the angle θ to the reading wave in the cases of $I_0 >> I_j$ and $I_0 = I_j$. It is this fact that accounts for the coincidence of the curves $\tilde{h}(\theta)$ and $\tilde{I}(\theta)$.

4. Conclusion

1. The greatest increase of the relative HDE with the angle θ is shown to be observed for thin holograms. It is established that there is a PCM thickness beginning with which the relative HDE dependence upon the angle ceases to depend upon the PCM thickness reaching some limiting distribution.

2. The angular dependencies of the relative HDE at $I_0 >> I_j$ and $I_0 = I_j$ agree well under any thickness of the hologram for the values of parameter $M < M_{crN}$ and in the case of considering thin holograms for the values of parameter $M > \beta_1 / \beta_2$.

3. It is shown that by changing the propagation direction of the reading wave it is possible to controllably change the angular selectivity of a dynamic hologram in a reversible PCM.

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