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S Sainov

1113 Sofia, PO Box 95, Central Laboratory of Optical Storage and Processing of Information,
Bulgarian Academy of Sciences, Bulgaria

E-mail: `ssainov@optics.bas.bg`

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Abstract. The theoretical background of holographic recording in very thin layers is given. Interference of two surface evanescent waves is used to achieve high and low grating spatial frequency. Another important recording case with surface reference and the usual (homogeneous) object wave is also elucidated. The calculations are made on the base of the effective thickness, introduced by N Harrick for internal reflection spectroscopy of very thin layers. The possible applications of this type of holographic recording in surface investigations are discussed.

1. Introduction

Holographic recording by interference of inhomogeneous surface evanescent waves was demonstrated by H Nassenstein in 1968 [1], where he used total internal light reflection for evanescent-wave creation. Nassenstein called them ‘surface’ waves, emphasizing propagation along the boundary surface between the recording and optically dense medium. A profound review of that holographic method was given by O Bryngdahl [2], where the most interesting features were discussed. The recorded hologram has characteristics similar to a Bragg reflection grating: sharp angular selectivity and maximum diffraction efficiency at the critical angle. A very interesting feature is the possibility of optical information recording in the thin layer near the surface of the recording medium [3], comparable with the penetration depth of the evanescent optical field, but the thickness of the recording can be diminished considerably. The reason for that is the result obtained by C Carniglia *et al* [4], where dye molecules are excited by surface evanescent waves. The total thickness of the contained dye molecules layers is about 5 nm. Another important work is that of N Harrick [5, 6], where the total internal reflection spectroscopy of very thin films is considered.

Our treatment of surface holographic recording is based on the Harrick effective thickness approach, see [6, p 41], that is the case of weak absorption of light by the optically rarer (recording) medium. Thus we can take into account that part of the interfering optical field that is responsible for the surface holographic recording.

2. Surface-wave interference in very thin layers

Let us consider the following three cases of holographic grating recording, shown in figure 1:

(a) homogeneous (3)–surface-wave (1) interference;
(b) surface-wave (1)–surface-wave (2′) interference (one way propagation) and

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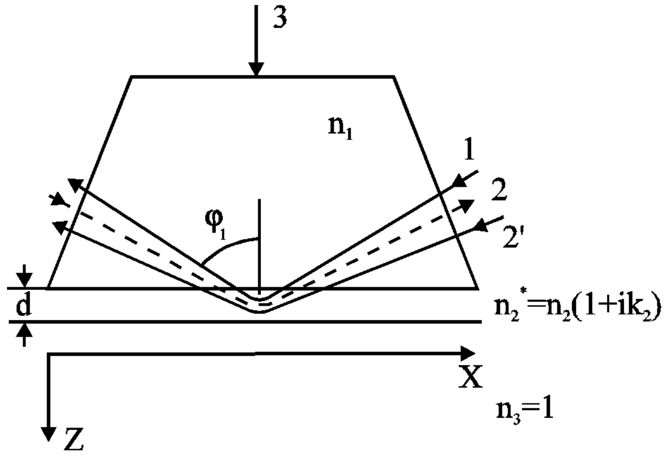

Figure 1. Optical set-up for surface holographic recording.

(c) surface-wave (1)–surface-wave (2) interference (contrapropagating case).

The homogeneous wave (3) is described by

$$\boldsymbol{u}_h = \boldsymbol{a}_h \exp(\mathrm{i}k_h z)$$

and inhomogeneous waves (1), (2) and (2′) by

$$\boldsymbol{u}_s = \boldsymbol{a}_s \exp(\mathrm{i}k_s x)$$

where $\boldsymbol{a}_h$ and $\boldsymbol{a}_s$ are the corresponding amplitudes and $k_h = 2\pi n_2/\lambda_0$; $k_s = 2\pi n_1 \sin\varphi_i/\lambda_0$, where n_i are the corresponding refractive indices and φ_i the angles of incidence; λ_0 is the wavelength in the air.

The interference terms corresponding to the interaction between the waves shown in figure 1 are:

$$\begin{aligned}
&\text{(a) } I_{h,s} = 2a_h a_s \cos[2\pi(xn_1 \sin\varphi_1 - zn_2)/\lambda_0]\\
&\text{(b) } I_{s1,s2'} = 2a_{s1}a_{s2'} \cos[2\pi x n_1(\sin\varphi_1 - \sin\varphi_{2'})/\lambda_0]\\
&\text{(c) } I_{s1,s2} = 2a_{s1}, a_{s2} \cos[2\pi x n_1(\sin\varphi_1 + \sin\varphi 2)/\lambda_0].
\end{aligned} \tag{1}$$

If we take into account small thickness compared to λ_0/n_2, there will be no intensity change $I_{h,s}$ in the Z-direction. In the case when $2\pi n_2 d/\lambda_0 < 0.1$ we can write:

$$\begin{aligned}
&\text{(a) } I_{h,s} = 2a_h a_s \cos(2\pi x n_1 \sin\varphi_1/\lambda_0)\\
&\text{(b) } I_{s1,s2'} = 2a_{s1}a_{s2'} \cos(4\pi x n_1 \cos\varphi_1/\lambda_0)\\
&\text{(c) } I_{s1,s2} = 2a_{s1}a_{s2} \cos(4\pi x n_1 \sin\varphi_1/\lambda_0)
\end{aligned} \tag{2}$$

where we use the following relations taking place in the experiments:

$$\begin{aligned}
&\varphi_2 = \varphi_1 + \delta \qquad \delta \ll 1\\
&\varphi_{2'} = \varphi_2 + \delta_1 \qquad \delta_1 \ll 1\\
&\varphi_1 > \sin(n_2/n_1) = \sin n = \sin\varphi_c.
\end{aligned} \tag{3}$$

Case (b) corresponds to the large grating period in the X-direction and usually is less interesting. It can be noticed that the period in (c) is half that in (a), describing surface-plane or focused-wave interaction.

For the wave's amplitudes for s polarization, following [6] we have:

$$\begin{aligned} a_h &= a_{00}(1 - 0.5\alpha d) \\ a_{s1} &= a_{01}(1 - 0.5\alpha d_{e1}) \\ a_{s2} &= a_{02}(1 - 0.5\alpha d_{e2}) \end{aligned} \tag{4}$$

where

$$\begin{aligned} &\alpha = 4\pi n_2 k_2/\lambda_0 \text{ is an absorption coefficient} \\ &d_{ei} = 4n_1 n_2 d \cos\varphi_i/(n_1^2 - 1) \text{ is an effective thickness.} \end{aligned} \tag{5}$$

Accepting that all initial amplitudes a_{0i} are unity at the boundary surface dividing the media with different index of refraction, from (1), (4) and (5) we have:

$$\begin{aligned} I_{h,s} &= 2[1 - \alpha d(n_1^2 - 1 + 4n_1 n_2 \cos\varphi_1)/2(n_1^2 - 1)] \cos(2\pi x n_1 \sin\varphi_1/\lambda_0) \\ I_{s1,s2'} &= 2[1 - \alpha d 8 n_1 n_2 \cos\varphi_1/(n_1^2 - 1)] \cos(4\pi x n_1 \cos\varphi_1/\lambda_0) \\ I_{s1,s2} &= 2[1 - \alpha d 8 n_1 n_2 \cos\varphi_1/(n_1^2 - 1)] \cos(4\pi x n_1 \sin\varphi_1/\lambda_0). \end{aligned} \tag{6}$$

The equations for effective thickness, according to [6] are good to a few per cent, when $2\pi n_1 d/\lambda_0 < 0.1$ and $k_2 < 0.1$. It can be noticed that they are in good accordance with (2) since $2 < n_1 < 3$ and $1.5 < n_2 < 2$. For $\lambda_0 = 325$ nm and $n_1 \approx 2.2$, we have $d < 3$ nm.

3. Experiment

Holographic recording with a surface reference wave according to case (a) is performed with an Ar^+ laser at 488 nm. As a recording medium a 29 nm thin As_2S_3 film has been used. The sublimation and evaporation of As_2S_3 is allowed without local overheating, thus preventing the undesirable effects of thermal decomposition. A deposition rate of 0.1 nm s^{-1} has been achieved at an evaporation temperature of 240 °C and residual pressure $2\text{–}4 \times 10^{-4}$ Pa [7].

The exposure time is varied between 0.5 and 20 s at beam intensities of 10 mW cm^{-2} each. The refractive index n_1 of the total internal reflection prism is 1.52 at 488 nm. The incidence angle of the reference wave is 45° that determines a grating period of 454 nm.

The diffraction efficiency has been measured with a low power He–Ne laser at 633 nm. In figure 2 the exposure dependence on the diffraction efficiency is demonstrated. The maximum value of 0.04% has been obtained at 150 mJ cm^{-2} exposure.

4. Conclusions

Despite the weak absorption part in nanoscale recording materials, it is possible to store a holographic interference pattern. Our preliminary investigations have shown that a recorded thin surface grating behaves like a thick Bragg-type one: with sharp angular sensitivity, strong polarization dependence and a spectral selectivity. The most interesting feature of this holographic recording is the recording medium has higher refractive index—2.5—than the total internal reflection prism. The critical angle—41.1°—is defined by the air/prism refractive index ratio. Another suitable recording medium is an azo-dye sensitized Langmuir–Blodgett layer.

The possible applications of this holographic recording are:

(1) submicron lithography;
(2) surface diffusion investigations by forced Rayleigh scattering;
(3) surface phase-change investigations;
(4) nanoscale diffractive optics.

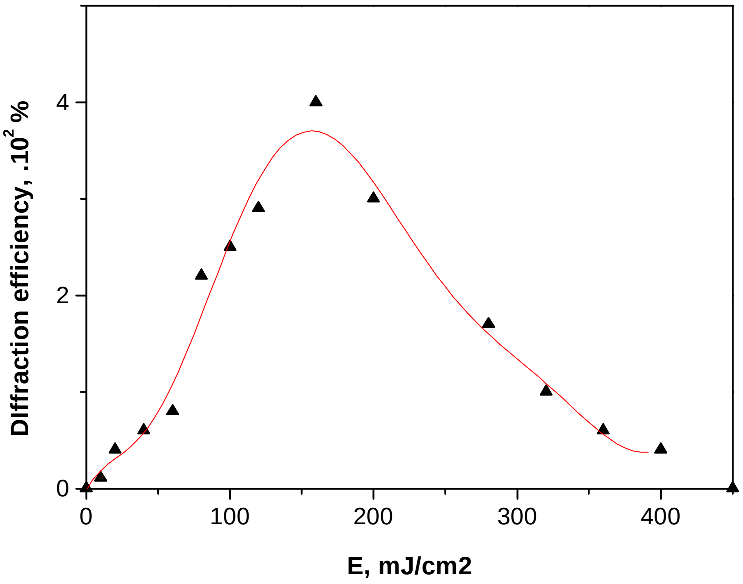

Figure 2. Diffraction efficiency as a function of exposure.

This type of holographic recording is also related to near-field optics and is connected with the super-resolution problem in optics, dealing with visualization of very fine surface structures.

Acknowledgments

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