

Total internal reflection holographic recording in very thin films

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Abstract. We present a theoretical background and experimental verification of holographic recording in nano-sized films. The interference pattern is created by a total internal reflected (TIR) reference wave and normally incident plane one. Due to a surface-propagating evanescent wave, the holographic recording is stored in 39 nm arsenic trisulfide film. The exposure and modulation dependence on the diffraction efficiency is investigated. The angular and wavelength selectivity of the thin TIR hologram is shown to be similar to that of thick Bragg gratings.

Keywords: Total internal reflection, holography, thin films

1. Introduction

Total internal reflection (TIR) was applied in holography by Nassenstein 30 years ago [1]. In his recording arrangement the incidence angle of the reference wave is greater than the critical one, determined by the refractive index ratio of the recording and the first, optically denser, medium. In fact, because of the weak light absorption by the photosensitive optical storage medium, we have a typical case of an attenuated TIR, where the critical angle is not strictly defined. Soon afterwards, Bringdahl [2] also used TIR-created evanescent waves for holographic recordings. The main feature of these holograms is that the interference pattern is stored in a small part of the recording medium, compared with the penetration of the TIR wave. In 1967 Stetson [3] proposed another TIR holographic recording method in which the reference wave undergoes TIR between air and a thick silver halide emulsion recording medium, i.e. the latter acts as an optically denser one, in which two sets of Bragg-type gratings are stored. The diffraction efficiency (DE) of these TIR holograms can be analysed by the Kogelnik's coupled-wave theory [4].

Another TIR holographic method was proposed by Lin [5]. His 'edge-illuminated' holograms are similar to Stetson's, but the TIR reference wave undergoes multiple TIR in the glass substrate that acts as a multimode waveguide. These holograms were later called 'substrate-mode' holograms [6, 7].

The evanescent part of the guided wave in optical thin-film waveguides is used as a reference wave in holographic recording, first proposed by Japanese scientists Suhara *et al* [8, 9] in 1976, and two months later by Lukosz and

Wuthrich [10]. These holograms are known as 'waveguide holograms', where dichromated gelatin has been used as a thick optical storage medium.

Usually in TIR holographic recording the photosensitive film's thickness is up to 10 μm , but the surface-propagated evanescent wave enables a holographic recording in very thin films. Waveguide holographic recording in 250 nm As_2S_3 was reported in [8]. Later, we succeeded in TIR holographic recording using a 70 nm thick As_2S_3 film [11]. This optical storage medium is suitable for such holograms, because its large photoinduced refractive index changes [12].

In this paper we first report a theoretical background of a holographic TIR recording in very thin photosensitive films on the basis of Harrick's thin film 'effective thickness' approach [13], in which the refractive index of the optical storage medium does not determine the critical angle value. In this case the film's refractive index can be much higher than the TIR prism refractive index. The TIR holographic recording in a 39 nm thick As_2S_3 film has been performed. The maximum DE obtained is 0.15% at 250 mJ cm^{-2} exposure. The holographic characteristics such as the recording intensity's ratio dependence, exposure time and angular dependence on DE are investigated. The angular shift observed in red light reconstruction is also the same as in thick Bragg gratings.

2. Theoretical background

In order to elucidate the TIR holographic recording, let us consider the interference between a normally incident plane wave and a reflected one, falling at the angle φ to the interface between the first medium with refractive index n_1 and air

$n_3 = 1$. Total reflection takes place when the incidence angle φ is greater than the critical one, determined as

$$\varphi_c = \sin^{-1}(n_3/n_1). \quad (1)$$

The TIR reflected wave, propagating along the axis X on the surface $Z = 0$ for s-polarization is described by

$$u_s(x, z) = a_s(z) \exp(2i\pi x \sin \varphi / \lambda_1) \quad (2)$$

where the amplitude is $a_s(z) = \frac{2 \cos \varphi a_{e0}}{(1-n_1^{-2})^{1/2}} \exp(-z/z_0)$, a_{e0} is the incident wave amplitude, $\varphi > \varphi_c = \sin^{-1}(1/n_1)$, λ_1 is the wavelength in the first medium, and $z_0 = \lambda_1/2\pi(\sin^2 \varphi - \sin^2 \varphi_c)^{1/2}$.

z_0 is a characteristic length of the TIR wave, where its amplitude is e^{-1} times smaller than at the surface $Z = 0$ and is usually called 'penetration depth', but this is true only for s-polarization near the critical angle. In fact, the penetration of the TIR wave depends on the polarization and near the critical angle is greater for a p-polarized wave, which has been established theoretically and verified experimentally in optical TIR tunnelling investigations.

The object plane wave is

$$u_p(z) = a_{p0} \exp(2i\pi z / \lambda_1) \quad (3)$$

with the initial amplitude a_{p0} .

The interference term I_{12} can be written as

$$I_{12}(x, z) = |u_p|^2 + |u_s|^2 + 2|u_p||u_s| \cos[2\pi(x \sin \varphi - z)/\lambda_1]$$

or

$$I_{12}(x, z) = I_p + I_s + 2(I_p I_s)^{1/2} \cos \Phi. \quad (4)$$

This interference pattern can be stored in the second recording medium, with a complex refractive index $n_2^* = n_2(1 + i\kappa_2)$, absorption coefficient $\alpha = 4\pi\kappa_2/\lambda_2$ and thickness d .

Following Harrick [13] we can introduce the so-called 'effective thickness' d_e in order to describe the optical field $I_{12}(x, z)$ interaction with the recording medium:

$$d_e = \frac{4n_1 n_2 d \cos \varphi}{(n_1^2 - 1)} = qd. \quad (5)$$

The conditions for this are

$$\begin{aligned} \alpha d \ll 1 \text{ (weak absorption)} \quad \text{and} \\ d \ll z_0 \text{ (very thin film)}. \end{aligned} \quad (6)$$

It is interesting to note that in this case the critical angle does not depend on the recording film's refractive index which can be higher than that of the first medium, thus widening the list of the optical storage media.

For the intensity I_p and I_s we have

$$I_p = I_{p0}(1 - \alpha d) \quad \text{and} \quad I_s = I_{s0}(1 - \alpha d_e). \quad (7)$$

The thin film's condition $d \ll z_0$ in the case of grazing incident angle $\varphi \approx \pi/2$ (corresponding to the minimal characteristic length z_0) is

$$2\pi d / \lambda_1 \ll \frac{n_1}{(n_1^2 - 1)^{1/2}}. \quad (8)$$

For the two extreme cases of GaP- $n_1 = 3.5$ and NaF- $n_1 = 1.33$, we have $2\pi d / \lambda_1 \ll 1.04$ and $2\pi d / \lambda_1 \ll 1.52$, respectively. Harrick noted that $2\pi d / \lambda_1 \ll 0.1$ and $\alpha d < 0.1$ ensure an accuracy of several per cent for TIR spectroscopy use.

Taking into account these conditions, from (4), (5) and (7) after some transformation we obtain

$$\begin{aligned} I_{12} = I_{12}(\alpha = 0) - \alpha d [(I_{p0} + q I_{s0}) \\ + (I_{p0} I_{s0})^{1/2} (q + 1) \cos(2\pi x \sin \varphi / \lambda_1)]. \end{aligned} \quad (9)$$

The grating period Λ over X is $\Lambda = \lambda_1 / \sin \varphi$.

Introducing the intensity ratio at the surface $Z = 0$: $I_{p0}/I_{s0} = m^2$, relation (9) can be written as

$$\begin{aligned} I_{12} = I_{12}(\alpha = 0) \\ - \alpha d I_{s0} [(q + m)(m + 1) \cos(2\pi x \sin \varphi / \lambda_1)]. \end{aligned} \quad (10)$$

3. Experiment and discussion

The experimental samples are prepared in a production line for linear encoder gratings developed in the Central Laboratory of Photoprocesses at the Bulgarian Academy of Sciences. Thin layers are obtained by vacuum deposition of high purity As_2S_3 onto linearly moving glass substrates in vacuum plant at residual pressure of the order of 6×10^{-4} Pa.

The specially constructed evaporation source for As_2S_3 ensures the uniform thickness of the deposited film on the whole substrate surface. The deposition rate 0.1 nm s^{-1} is controlled and maintained constant during the evaporation. The film thickness is 39 nm, measured by Talystep profilograph, Code 112/1037 M-S, produced by Rank Taylor Hobson Ltd.

The optical set-up for the TIR holographic recording is shown in figure 1. As a light source we use an Ar⁺-laser ILA-120 (Karl Zeiss Jena), generating at 488 nm. With the adjustable beam splitter the initial beam of 20 mW cm^{-2} intensity is divided into two beams with equal intensity. The used TIR prism is made of light crown K-8 with $n_1 = 1.52$ for 488 nm wavelength. The incident angle φ is 45° when the critical angle is 41.1° . The calculated characteristic length z_0 is 196 nm and the grating period Λ is 454 nm. Contact microscope oil has been used for index matching. The oil also makes it possible for films to slide along the reflection wall of the TIR prism and obtain up to 10 TIR holograms on a $2 \times 4 \text{ cm}^2$ film area.

The DE with 5% accuracy is measured continuously using 0.5 mW He-Ne laser at 633 nm and a powermeter 'Ealing' 910. The intensity ratio m^2 between the two beams is changed with grey filters. The incident angle of the monitoring red beam is $15^\circ \pm 1^\circ$ in air to achieve maximum DE, which is different from the recording normal incidence position. Using the Bragg relation $2d \sin \varphi = \lambda$, we have $\Delta\varphi = \Delta\lambda/2d = 0.16$. The measured value in the TIR prism is 0.17 ± 0.02 which is in good agreement with the calculation, proving the close analogy between the TIR surface and thick grating recording.

The exposure time is varied in the 0.5–20 s interval, which corresponds to 10–400 mJ cm^{-2} exposure. Figure 2 shows the diffraction efficiency dependence on exposure. The best result, 0.15%, is obtained at 250 mJ cm^{-2} ,

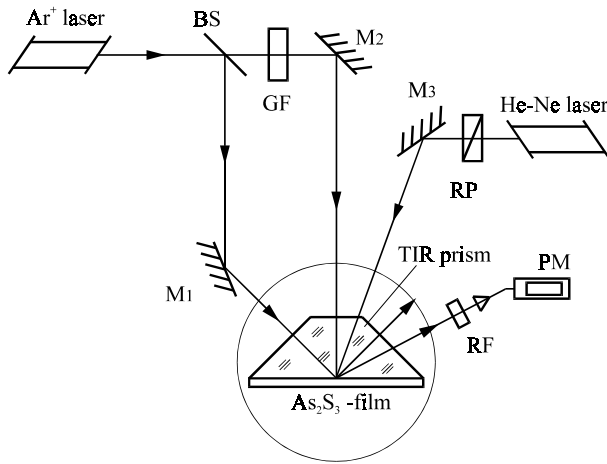


Figure 1. Experimental arrangement for TIR holographic recording: BS, beam splitter; M_i , mirrors; GF, grey filter; RP, rotator of polarization; PM, powermeter; RF, red filter.

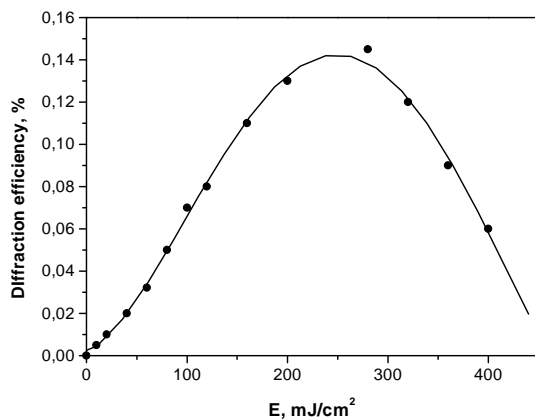


Figure 2. Exposure dependence of the TIR diffraction efficiency.

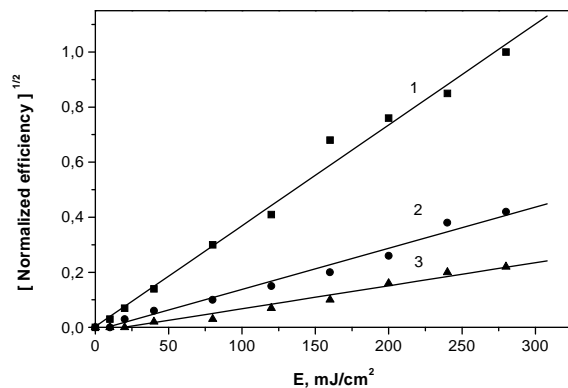


Figure 3. Intensity ratio dependence of the TIR diffraction efficiency: 1, $m = 1$; 2, $m = \frac{1}{2}$; 3, $m = \frac{1}{5}$.

which is comparable to thick grating exposure. A possible explanation of the observed DE behaviour is given by the Kogelnik's coupled-wave theory, by which the amplitude of the reconstructed wave is proportional to the sine of the refractive index modulation and after reaching maximum phase ($\pi/2$) the efficiency is diminishing.

The intensity ratio is 1, which corresponds to the maximal visibility of the interference pattern. Diminishing

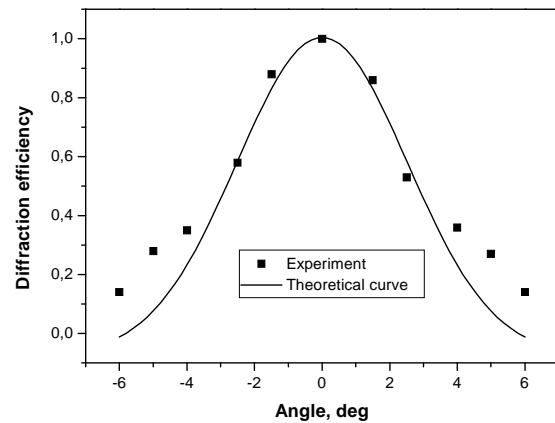


Figure 4. Angular dependence of TIR diffraction efficiency: reconstruction with object-wave copy.

that ratio, the DE is also diminished, as shown in figure 3, where normalized DE is proved to be a linear function of exposure.

Another measurement proving once again the close analogy between thick Bragg grating and TIR hologram recorded in very thin, nano-sized films is an angular selectivity investigation. Figure 4 demonstrates the angular dependence of the normalized DE. The theoretical curve is calculated for a substrate-mode hologram [14] with thickness $5.3 \mu\text{m}$. The squares represent the experimental values, measured with a copy of the object plane wave, illustrating a relatively good agreement between thick Bragg grating theory and the very thin TIR surface hologram.

4. Conclusion

A holographic recording in a very thin photosensitive film has been reported. The interference between a surface-propagating evanescent wave and a plane wave create a TIR hologram with properties similar to a thick Bragg grating: wavelength selectivity and sharp angular dependence. A very interesting feature of this TIR hologram is the critical angle independence of the optical storage medium refractive index.

Despite the relatively low DE value, a good signal-to-noise ratio makes this TIR holographic recording suitable for grating technique application in nano-scale surface investigations.

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