RELIEF HOLOGRAMS FORMATION AND REPLICATION IN HARDENED DICHROMATED PVA FILMS

By B. Smolińska

Institute of Physics, Warsaw Technical University*

(Received January 26, 1971)

The application of photosensitive dichromated polivinylalcohole layers in holography is described in this paper. Phase holograms with good diffraction efficiency has been achieved in direct exposure with Ar lasers light in ordinary ho ographic arrangement as as well as in replication process from amplitude holograms. Relief holograms recording in photopolymers is described in simple mode. The role of the relief thickness for diffraction efficiency is there emphasized.

Photochemical properties of organic polymers can be utilized to record relief holograms. The solvatation abbility of the polymers changes during exposure. The solubility of illuminated polymer may increase or decrease, depending on the nature of the photochemical reaction. For example in vinyl polymers the light causes a break up of double bonds what leads to a growth of the network structure or futher polimerization, and in the case of hydrophyllic polymers such as gelatine the hydrophyllic groups become blocked up. After exposure and further development in a suitable solvent a relief film remains which corresponds to the light field intensity on the polymer surface. There is need to remark that for the holographic process it is unimportant whether the exposed or the unexposed parts form the relief hologram.

The recording process occurs according to the photochemical rule

$$D = C \cdot I^{\gamma} \tag{1}$$

where D is the thickness of the polymer layer after exposure and development and C and γ are constants characteristic of the given photosensitive material. The nonlinearity of the function $\ln D = f$ ($\ln I$) can be neglected, because photopolymers have characteristics with long linear slope.

In the holographic process the light intensity distribution in the plane of the hologram corresponds with diffraction patterns formed by interference of the reference wave A_{\star} .

^{*} Address: Instytut Fizyki, Politechnika Warszawska, Warszawa, Koszykowa 75, Poland.

with the bundle of waves A_0 coming from the object

$$I_{H} = |A_{r} \exp ikz + A_{0} \exp i \varphi|^{2}. \tag{2}$$

Both amplitude A_0 and phase φ depend on the coordinates of the object points and hologram points.

After exposure and development the obtained hologram constitutes a subtly folded transparent polymer layer. The density and curvity of the folds depend on the phase differences between the reference wave and waves coming from various points of the object. The height D of the folds depends on the amplitude of the light field I_H

$$D = C|A_r \exp ikz + A_0 \exp i\varphi|^{2\gamma}. \tag{3}$$

Such a relief hologram acts as a dielectric grating. During reconstruction an arbitrary light beam A_p falls on the relief hologram and is diffracted on the folded structure. In the surrounding of one fold the wave front is changed very rapidly. For example in the case of an ordinary hologram the refractive index n varies from $n_{\rm air}=1$ to $n_{\rm polymer}\cong 1.5$ over a distance of about 10^{-6} cm.

The relief hologram can be described by the complex transmittance

$$F = C \exp i\Phi \tag{4}$$

there, C is the amplitude transmittance which is equal to 1 for white objects and φ is the phase transmittance involved by an object with alternating optical density. In the case of a relief hologram φ is equal to the phase change of a wave traversing a polymer layer of thickness D and refractive index n

$$\Phi(x, y) = k \cdot D(x, y) [n-1]. \tag{5}$$

For small values of φ we can expand F into the series (1) getting

$$F(x, y) \cong 1 + i\Phi \cong 1 + ik(n-1)D(x, y). \tag{6}$$

The light field A_i behind the hologram is given by the Gabor equation

$$A_i = A_p F. (7)$$

Using only the imaginary part of F and Eq. (1) and (3) we obtain the light field behind the hologram in the simplest case of a true recording ($\gamma = 1$)

$$\begin{split} A_i &= A_p \exp ik'z \left\{ Ck(n-1) \left(Ar^2 + A_0^2 \right) + \right. \\ &+ Ck(n-1) \left. A_r A_0^* \exp i(kz - \varphi) + \right. \\ &+ Ck(n-1) \left. A_r^* A_0 \exp i(\varphi - kz) \right\}. \end{split} \tag{8}$$

The first therm of Eq. (8) represents the zero-order beam, the next two terms describe waves carrying information about the object. Those are responsible for the reconstruction

of the real and virtual images of the object. The intensity distribution in both of the beams is the same as in the object wave.

$$I_i = A_p^2 C^2 k^2 (n-1)^2 A_r^2 A_0^2 \cong I_0. \tag{9}$$

This description neglects the volume recording because the experiments described in this paper primarily deal with the so called surface phase holograms. Also F is not expanded into a series of Bessel functions here as is suggested in the literature [2,3]. This description is limited to simple equations which represent the experimental results obtained in this paper quite closely.

Experimental

The first investigations on relief holograms concerned hardened dichromate gelatine films [4,5] and acrylic polymers [6,7]. This paper is a continuation of work [8] on the application of dichromated polyvinyl alcohol (PVA) films for holography. Illumination of a PVA layer causes photoreduction of Cr⁶ to Cr³ and there follows a blockade of hydrophyllic groups in the PVA. In this way the exposed parts of the polymer become insoluble in water, while the unexposed parts can be removed by development in water. The remaining polymer film gives the relief hologram.

PVA films of thicknesses of about 0.6 mm are prepared by coating glass blancks of uniform thickness with 4% dichromate polyvinyl alcohol in water solution. The layer is whirled in a centrifuge (47 revs/min) and thus dried. The preservation time for the sensitive material is limited to 48 hours for dry films and one month for the solution. After that

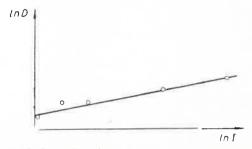


Fig. 1. Dependence between the thickness D of the relief and the exposure I for Cr hardened PVA film used in this work (in logarithmic coordinates)

irreversible changes, such as dark reduction of Cr⁶ in the presence of organic compound occurs in the material. The obtained layers may have a thickness from several to some hundreds of nanometers depending on the concentration of the solution and centrifuge velocity.

The characteristic curve of thickness *D versus* exposure for used photolayer is shown in Fig. 1. It is nearly linear. The spectral sensitivity of layer corresponds with its absorption curve shown in Fig. 2.

Photosensitive layers obtained in the manner described above are successfully applied in the holographic process. Simple two-dimensional objects are recorded on PVA films

in ordinary two beams arrangement employing Ar laser ($\lambda = 488$ nm). Reconstructions from these holograms are correct and bright. The diffraction efficiency reaches 20% but photosensitivity is less and amounts to about 1000 mJ/cm². However, the sensitivity can be enhanced by addying sensitizers (for example eosine). The advantage of photopolymers is

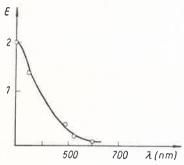


Fig. 2. Spectral absorption of the used photopolymer

their theoretical molecular resolving power. Holograms obtained in our laboratory are recorded in PVA films. Their spatial frequency exceeds 1200 lines/mm. The surface of the holograms exhibits delicate riffled structure shown in 1000 × magnification in Fig. 3.

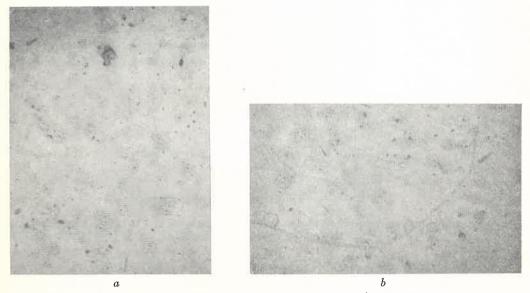


Fig. 3. Surface of relief hologram in 1000 magnification

Relief hologram replicas

Because of their lower photosensitivity, linear characteristic and high resolving power, PVA layers seem to be especially convenient for hologram replication. Replicas from various holograms are successfully achieved with the use of PVA photolayers. Holograms have been recorded on ORWO MIKRAT and KODAK MR plates as well. Only replication

from holograms recorded with lower optical density and flat contrast proves difficult. Special care has to be taken in order to assure the best contact between photographic plate and photolayer. In our laboratory replication is carried out in vacuum contact with the use of a point source of light.

Images reconstructed from replicas obtained in this way show no differences in comparison with images obtained from genuine holograms. One such image of two screws seen above a real bar through a transparent hologram replica is shown in Fig. 4.



Fig. 4. Reconstruction from a hologram relief replica made in PVA

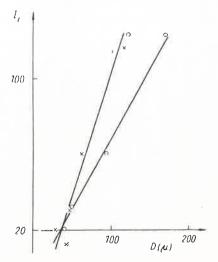


Fig. 5. Dependence between maximum thickness D of the hologram relief replica and intensity I of the diffracted wave

Hologram replication by use of photopolymers enables transformation of amplitude diffraction objects into phase diffraction objects. In this way the diffraction efficiency of a hologram can be enhanced through replication. It turns out that the relief thickness is the most important factor for getting relief holograms with great diffraction efficiency.

Hologram relief replicas are purely surface diffraction objects without any volume diffraction effects. This is why these objects are especially convenient for investigations on the connection between relief structure and diffraction effects. For this reason the dependence between maximum thickness of the relief D and brightness of the diffracted wave I is investigated in this paper. Experiments are made with a series of replicas featuring various thicknesses produced from two various hologram. The intensity of the diffracted wave is measured by an SeTe photoelement placed in the imaging beam wave. The thickness of the relief is measured with a PZO KS microscope with phase contrast in 200 and 400 magnification. The scatter of results is large and amounts to 20%, so the measurements have to be repeated many times. Two resultant curves for two different holograms are shown in Fig. 5. The dependence I = f(D) is found to be linear, what is in good agreement with Eq. (7) and (9) where $I_i \sim A_0^2$ and $D \sim A_0^2$.

Obviously many other photopolymers may be used in order to obtain phase holograms. Studies on phase holograms made in various materials will be carried out in our laboratory.

Finally I wish to thank Professor B. Karczewski for many stimulating conversations and advices. Miss L. Zuławnik is thanked for preparing the replicas.

REFERENCES

- [1] M. Born, E. Wolf, Principles of Optics, Pergamon Press, 1959, p. 424.
- [2] H. Kiemle, D. Röss, Einführung in die Holographie, Akad. V. 1969.
- [3] H. Kogelnik, J. Amer. Opt. Soc., 57, 431 (1967).
- [4] T. A. Schankoff, Appl. Optics, 7, 2101 (1968).
- [5] L. H. Lin, Appl. Optics, 8, 963 (1969).
- [6] D. H. Close, A. D. Jacobsen, J. O. Margerum, R. G. Brault, F. J. Mc Clung, Appl. Phys. Letters, 14, 159 (1969).
- [7] J. A. Jennej, J. Amer. Opt. Soc., 9, 1155 (1970).
- [8] B. Smolińska, IV Ogólnopolska Konferencja Radiospektroskopii i Elektroniki Kwantowej, Poznań 1970, in Polish.