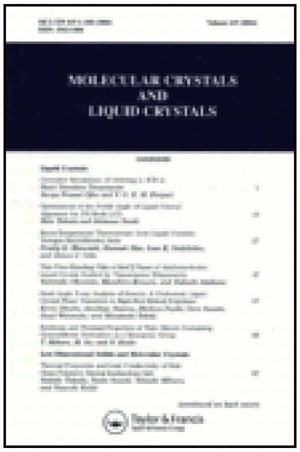

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Recent Progress in Dye-Polymers Systems for Polarization Holographic Optical Elements Development

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RECENT PROGRESS IN DYE-POLYMERS SYSTEMS FOR POLARIZATION HOLOGRAPHIC OPTICAL ELEMENTS DEVELOPMENT

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Abstract Experimental studies have been conducted with a versatile two recording beams (488 nm) arrangement that permits to investigate the real-time responses (633 nm) of studied polymeric films for a wide range of interbeam angles. Using this new automated spatial frequency analyser designed at Université Laval, dye/polymer systems can be characterized for both transmission and reflection holography. Moreover many applications involving real-time holography and four-wave mixing techniques are easily performed with this special apparatus. Most of our characterization studies have considered polarization holography. Results for thin solid films made from "azo dye colored polyvinyl alcohol (PVA)" and "Yellow dichromated PVA films" will be discussed. Photoreaction mechanisms involved in these recording processes and some modern applications of our erasable or/and permanent films are presented.

INTRODUCTION

Today's research in optical fields needs real-time recording materials of excellent performance and high spatial frequency response. Presently, the future of linear and non-linear holography is strongly dependent upon new holographic recording thin films. Also, most limitations associated with today's techniques are attributed to non-optimum recording materials. Due to their excellent responses, gratings with high spatial frequency up to 7500 mm^{-1} can be recorded, photopolymers and photocrosslinking materials seem to be good candidates to overcome some limitations. Dichromated gelatin films demonstrated excellent properties for permanent recordings but they are humidity sensitive; also they need a special chemical processing. Presently modern holographic works need erasable and real-time recording materials of low cost like organic and polymeric systems. In that way, our dye/polymer systems offer some new possibilities[1]. In this work we consider a comparison between holographic properties of two kinds of PVA films of 30 μm in thickness. The first ones are dichromated PVA films (noted DC-PVA) which record volume hologram gratings by a photocrosslinking process when ammonium dichromate (6,7%) is added during the preparation of solutions. The preparation techniques have been indicated elsewhere[2,3]. The second material is coloured PVA films obtained by adding an azo-dye, the MORDANT YELLOW 3R; the preparation technique for M.Y. 3R/PVA films was introduced in a previous paper[1]. Those organic films may record volume hologram gratings by a photoisomerization process[1] over the σ bond in the Azo chemical group (-N=N-). The two kinds of studied films can give polarization holograms[1,4]; but the present work considers the recordings obtained for right and left circularly polarized (O O) beams (488 nm) and the real-time reading (633 nm) with a plane wave having a circular polarization state similar to the reference beam used during the recording process.

EXPERIMENTAL ARRANGEMENT

Our versatile experimental arrangement[5] allows to study holographic characterization. Essentially, volume transmission polarization holograms were recorded with a simple two-beam set-up using the blue line of an Ar^+ laser (488nm). As illustrated in Fig. 1, we operated a very good fringe stabilization system using the two equal path length recording beams transmitted by the studied films. This

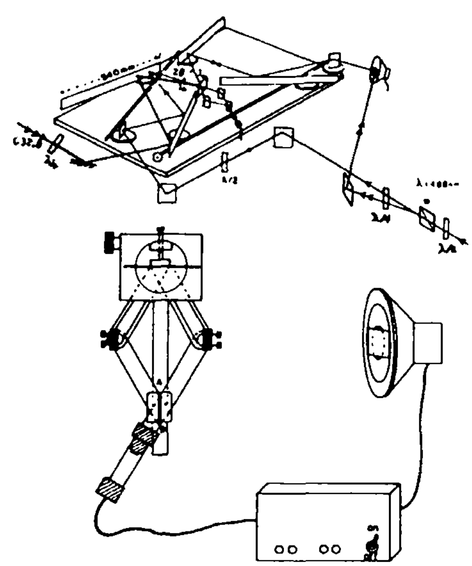


FIGURE 1 Basic set-up

special system was working during the recording process; also at the reconstruction process, when the object beam was closed, the single blue reference beam and a suitable polarized He-Ne laser (632.8 nm) beam were used in agreement with characterization experiments. These two different reconstructing beams were previously aligned at their respective Bragg angle. Consequently the hologram reading was performed by the red beam and the blue one was inducing an erasure process. Such a process gives a new CIS-TRANS equilibrium in exposed sections of azo-dye/PVA films. For DC-PVA films the basic set-up was used in conjunction with a reflection stabilization system as illustrated in fig. 2. The interbeam angles

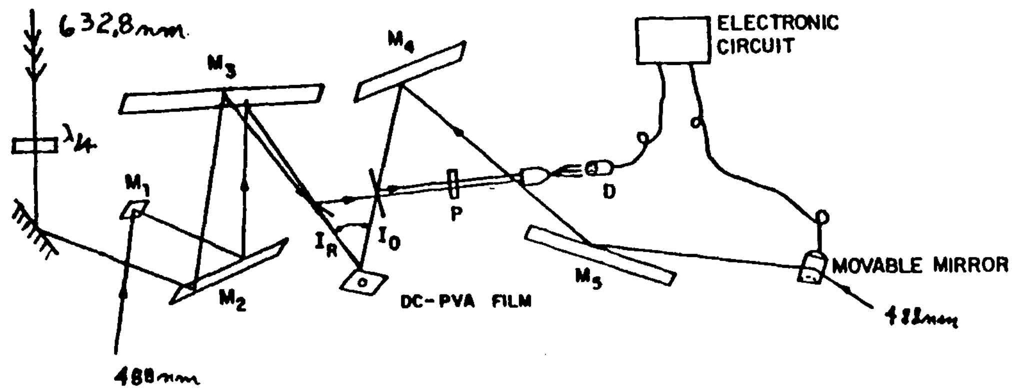


FIGURE 2 Reflection statibilization system

were continuously varied from 14° to 154° for the present study.

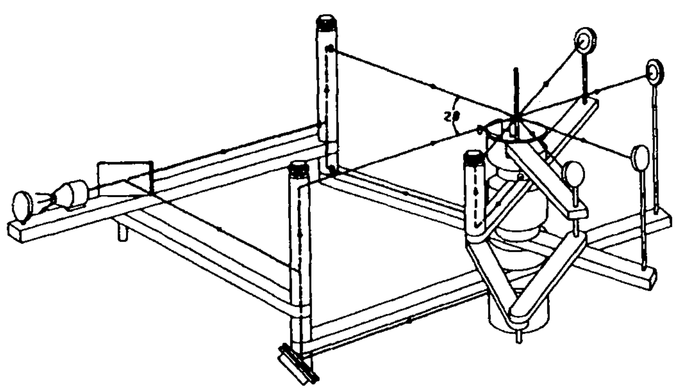
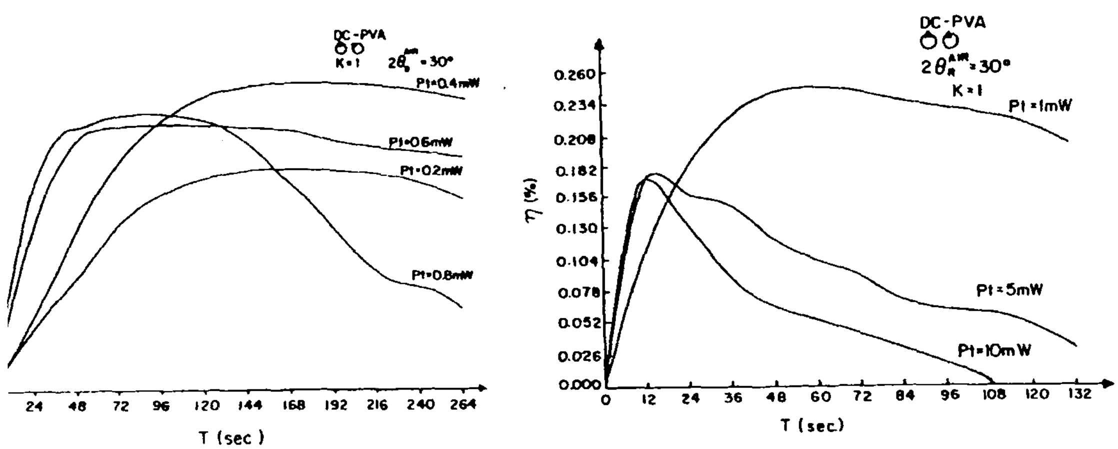


FIGURE 3 New experimental arrangement

Presently we are performing many tests on a new designed arrangement[6] illustrated in fig. 3. This more versatile apparatus permits to continuously change the interbeam angle starting from 5° to 170°.

HOLOGRAPHIC SENSITOMETRY RESULTS FOR DC-PVA FILMS

In the characterization experiments the interbeam angle (2θ) was kept to 30° and the corresponding spatial frequency was 1061 cycles/mm. We operated our basic apparatus spatial frequencies (ranging from 500 to 4000 cycles/mm) to investigate the modulation transfert function (M.T.F.) of studied PVA films. We point out that two kinds of PVA studied films in this work do not require any chemical development. Figures 4 and 5 show the dependence of real-time diffraction efficiency (or the grating growth) with respect the total power of the two blue (488 nm) recording beams for DC-PVA films. The displacement of the maximum peak is well illustrated. Following these curves, the maximum diffraction efficiency (DE) values are obtained for an exposure time changing from 50 to 150s when the total laser power is less than 1 mW and becomes about 12s if the power is over 5 mW. Consequently those DC-PVA films do not obey to the reciprocity law governing the basic behaviour of silver holographic plates. However an exposure about 200 to 400 mJ/cm^2



FIGURES 4 and 5 The real-time diffraction efficiency response versus the total power of recording beams used for DC-PVA films. The beams were of equal power (K = 1).

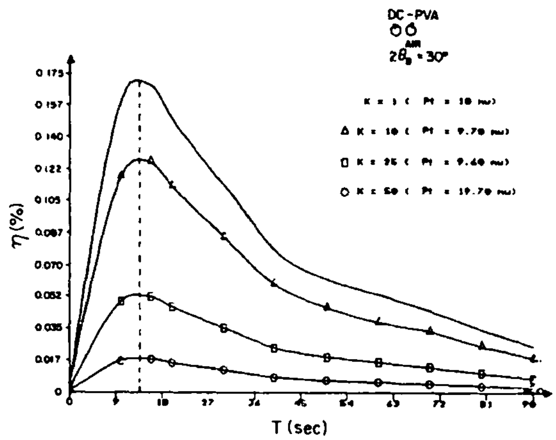


FIGURE 6 The DE dependence on the beam rate K for DC-PVA films.

give the best results. Figure 6 illustrates how the grating growthis affected by the power beam ratio K($K = P_R/P_O$) of the two recording beams. As expected the maximum DE value is obtained for K values near 1. Also the DE level changes slowly when K increases. Those last results permits to consider many engineering applications for DC-PVA films when circular polarization holograms would be recorded with two beams having an high K ratio. Figure 7 represents the spatial frequency responses of DC-PVA films for right and left circular polarizations. Those results have been obtained with two equal power recording beams ($P_R = P_O = 0.5$ mW) which lighted the DC-PVA films during 60 seconds. This M.T.F. curve is nearly uniform between 1500 and 3700 cycles/mm. The corresponding frequency bandwidth is about 2500 cycles/mm. That kind of spatial frequency response may be explained by short lengths of photocrosslinked polymer chains. When the spatial frequency overcomes 3700 cycles/mm, the photocrosslinking mechanism is less efficient because it implies very short chain lengths required to record an hologram fringes period that is lesser than 0,27 μm. So the DE value decreases when the interbeam angle increases. On the other side,

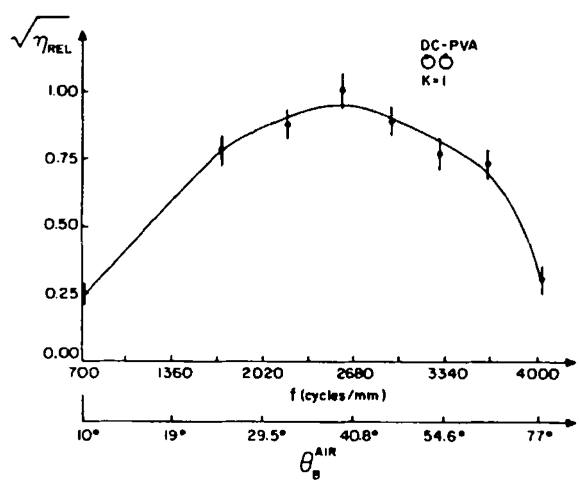


FIGURE 7 M.T.F. curve for DC-PVA films

whenever spatial frequency is less than 1000 cycles/mm, the chain formation is more important near the front surface of the DC-PVA films. Consequently the recorded holographic fringes did not penetrate the full depth of these DC-PVA films; as a result, the recorded hologram begins to look like a volume hologram which owns a thin grating component. So, the DE level decreases rapidly when the interbeam angles approach the minimum values and the M.T.F. response diminushes readily.

HOLOGRAPHIC SENSITOMETRY RESULTS FOR M.Y. 3R/PVA FILMS

Present experimental results were obtained with two orthogonal left and right polarized beams. The kinetic recording (240 sec.) and erasing (180 sec.) processes under orthogonal circularly polarized (OO) illumination are shown in Fig. 8 for the present azo-dye/PVA films. In those characterization experiments the interbeam angle (2θ) was kept at 30°. As illustrated in Fig. 8, the growth rate of the holographic grating ($d\eta/dt$) increases with the power ($P_R = P_O$) of the two beams. We point out that the linear recording process occurs just for the first 15 seconds of laser illumination; after that the diffraction efficiency begins to saturate. We observed that full erasing process was obtained when we stopped the laser illumination after 15 to 20 seconds. If a laser light exposure takes more than 30 seconds we did not observe full erasing process as it can be seen in Fig. 8. Therefore these azo-dye/PVA films demonstrated a weak memory effect. The best real-time diffraction efficiencies are about 0.3%. If corrections for reflections and absorption are taken in account this maximum diffraction efficiency value changes from 1.2 to 1,7%. This weak diffraction efficiency level occured when the power of the two writing beams were grater than 5mW (18mW/cm^2) in the present case for MORDANT YELLOW 3R/PVA films as indicated in Fig. 8. Maximum diffraction efficiency level (0.27%) was respectively obtained with an exposure of 300 mJ/cm^2.

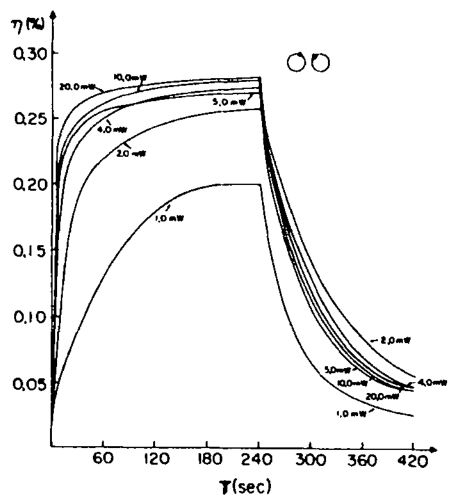


FIGURE 8 Kinetic grating growth and erasing in M.Y. 3R/PVA films.

In a second experiment we investigated the diffraction efficiency level with respect of the beam ratio K used as a parameter. The total power of the two recording polarized beams was kept at 15 mW (53.1 mW/cm^2) and the K ratio varied from 1 to 100. Linear holographic response occured in the first 15 seconds and maximum response was achieved for K ratio near 1. The results are illustrated in Fig. 9 suggest that polarization holographic recording of 3-D objects would be possibly achieved even though a depolarization action induced by such subjects; consequently these 3-D recordings would be assumed by components of parallel linear polarization, crossed linear and circular polarizations as noted in previous papers[1,4].

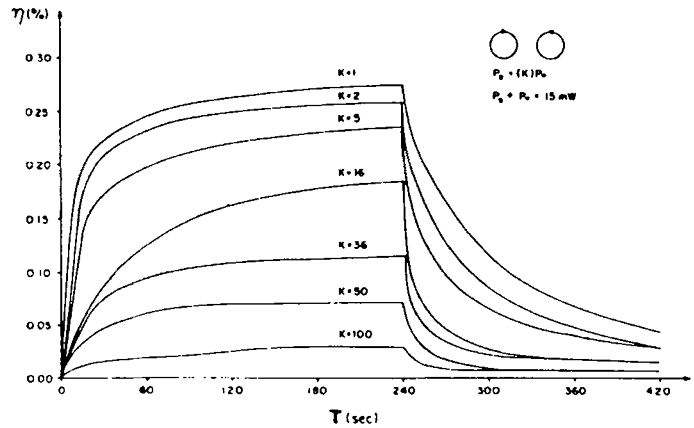


FIGURE 9 Kinetic grating growth with respect to the power beam ration (K)

To complete the present characterization study, the spatial frequency response of those azo-dye/PVA films have been investigated for three polarizations states of the two recording beams. The corresponding modulation transfer function (MTF) curves are presented in Fig. 10. Those M.R. 3R/PVA films give uniform spatial frequency responses between 500 cycles/mm and 3300 cycles/mm for volume transmission polarization recorded holograms. The spatial frequency bandwidth reaches 3500 cycles/mm. When these studied azo-dye/PVA films do not received a thermal treatment before our characterization experiments, they demonstrated a low diffraction efficiency level and they were completely erasable. Such a spatial uniform response for three states of polarization allows to investigate many useful applications like recording of polarization holographic optical elements (HOE) usable in real-time or in permanent recording mode (obtained with thermal treatment which leads to memory effect).

APPLICATIONS

Basic applications involving low diffraction efficiency holograms are critical because they imply low reconstruction beam intensity and many experimental drawbacks. However these azo-dye/PVA films can record real-time holograms and they also produce permanent

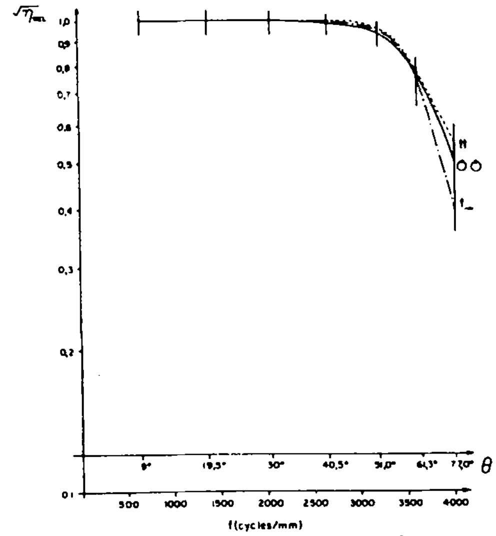


FIGURE 10 The M.T.F. of M.Y. 3R/PVA films with respect the polarization states of the two recording beams.

recordings. Consequently many monitored experiments may be improved as explained in this part of the paper. Presently many optical processing experiments are in progress for DC-PVA films which are not erasable; then we shall point out most interesting applications for azo-dye/PVA films.

1. Laboratory vibrations detection

Our spatial frequency analyser set-up was placed over special optical granite table. Sometimes residual laboratory vibrations do not permit to record holographic gratings. In order to detect these laboratory vibrations, in real-time, a simple detection technique is to record a volume transmission hologram in our low cost azo-dyes/PVA films as indicated in Fig. 11. From a working set-up and using two beamsplitters we sampled those two blue beams, R and O; these beams wrote a single hologram and a third one red beam (T) was reading, in real-time such a hologram, when it was aligned to Bragg angle. Then a time-dependent reconstruction beam is generated and strikes a sensitive photodiode. As azo-dye/PVA films are polarization sensitive films we can work following one of the three studied modes as required in many holographic studies. Laboratory vibrations of different kinds may be easily studied when this optical system is coupled to a good amplifier and simple laboratory oscilloscope or recorder even though the reconstructed red signal has a weak value. Fig. 12 shows two curves; the lower line represents the diffraction efficiency of real-time recording when disturbed by laboratory vibrations and the upper one is for an undisturbed holographic grating recording obtained when our fringe stabilization system was working. These kinds of vibrations can easily detected with azo-dye/PVA films when blue Ar^+ laser lines (488nm, 458nm) and low power He-Ne (632.8nm) are used. However we can work with a most effective blue line (442nm) from a He-Cd laser because M.Y. 3R/PVA films have a large absorption band in blue part of the visible spectrum. If we work with green laser lines (514,5nm, 532nm) and a low power He-Ne (543nm), the azo-dye/PVA systems that would be used are METHYL ORANGE/PVA and ACIDIFIED METHYL RED/PVA films which they have been studied in a previous paper[1]. Consequently our azo-dye/PVA films are used as real-time sensitive holographic optical element. As illustrated in Fig. 12 we used a Mordant Yellow 3R/PVA film thermally treated (90°C, 60 min.); so, we pointed that the maximum diffraction efficiency was increased

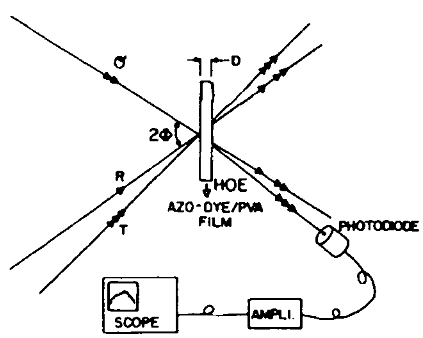


FIGURE 11 Real-time laboratory vibrations detector.

by factor 6. If corrections for reflections and absorption are taken in account this factor becomes 20. This simple holographic technique represent a high sensivity method that allows useful vibrations detection in any optical holography laboratory since the required set-up is simple and the low cost azo-dyes/PVA films are easy to prepare and reusable many thousands times. This vibration detection technique becomes interesting method in many applications where high mechanical stability is required. So we readily detected sound waves like human voices and air motions in our laboratory. A limiting application is to use the present technique in earthquake detection.

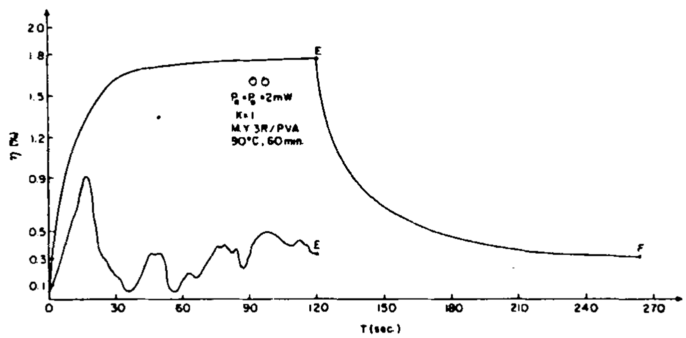


FIGURE 12 Laboratory vibrations by using a real-time recording obtained with M.Y. 3R/PVA films.

<u>2. Stability control by hologram real-time interferometry</u> (HOE)

As in the first application, we sampled two beams R and O from making an interbeam angle 2ψ from a working holographic set-up. Now we must solve the stability problem. One possible solution is to build up a HOE element that gives two parallel coherent beams at its output surface when two angularly separated beams (2ψ) strike the input surface. To make this kind of HOE, we shall take into account the memory effect of azo-dyes/PVA films. Before using those films, they were thermally treated at 90°C for 60 minutes; such a treatment gives permanent holograms having a high diffraction efficiency as pointed out in a previous paper[7]. After that, our HOE was built as following: 1° an azo-dye/PVA film is placed on the plateholder of our stabilized spatial frequency analyser 2° a first recording was made with two polarized blue beams r and O as indicated in Fig. 13A (exposure time was $t_1 = 120$ seconds) 3° a symmetric hologram recording was performed with two blue beams r and R as illustrated in Fig. 13B; the second exposure time was $t_2 = \sqrt{2}\, t_1$ or 170 seconds as required for sequentially volume multiplex holograms[8] 4° then, the two recorded permanent gratings generate two coherent parallel beams producing large fringes imaged on a screen (Fig. 13C) when the two R and O beams strike this HOE which does not need any development. 5° After that, this HOE was placed in the real set-up where two beams R and O intersect at an angle 2ψ. The produced fringes are imaged on an optical sensor which drives a movable mirror mount as shown in Fig. 13C. Consequently we can easily prepare simple HOEs with our spatial frequency analyser set-up and azo-dyes/PVA films. Those HOEs may serve as optical circuit to simplify the stabilization set-up for many different optical tables. A drawback is the laser light level involved; however electronic stabilizer

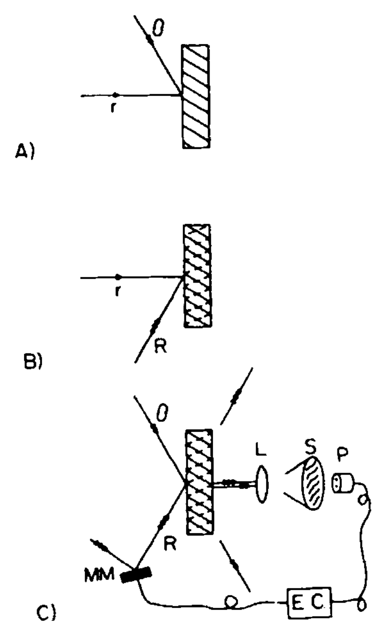


FIGURE 13 HOE construction and utilization as a beamsplitter-combiner

systems required a low fringe intensity profile, then this problem is not serious one. In our experimental test we used methyl orange/PVA films. The diffraction efficiency value of the two sequentially multiplex grating was respectively 0.05% and 0.04%.
Quite interesting was the feasibility of this HOE with MORDANT YELLOW 3R/PVA films; so we obtained better stability and diffraction efficiencies of 0.05% and 0.12% for each of two gratings and less experimental adjustments.

3. Movement measurement by hologram interferometry

Now suppose that we are interested in translation, rotation or any slow movements of a 3-D objects. In order to determine that kind of movement amplitude we fix a small mirror M on this object and we used the two-beam stabilized set-up as illustrated in Fig. 14A which illustrated the principle of the first application as reported in Fig. 12. A second possibility is to prepare a special azo/PVA plate made from two layers. First layer of thickness D_1 was a MORDANT YELLOW 3R/PVA film thermally treated. After such a treatment a second layer, D_2 in thickness was deposited over the first one; methyl orange/PVA material has been used as the second layer. When this special recording plate is placed in the stabilized apparatus, a first permanent recording has been obtained.
Then we used our set-up without any stabilization system; conse-

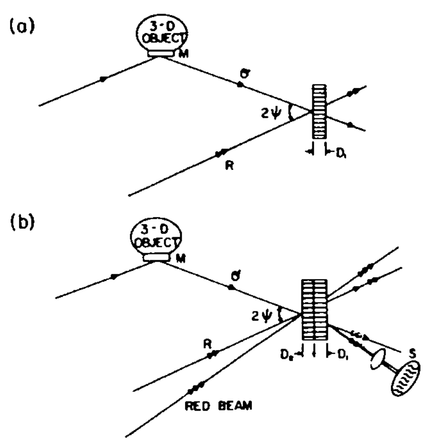


FIGURE 14 Real-time interferometric measurement of small mechanical movement by using a two layers Azo-Dye/PVA plate.

quently, we can easily observe real-time fringes produced by interference effects between a real-time grating (D_2) with a permanent grating (D_1) whenever a red beam aligned to Bragg angle is used (Fig. 14B). Visual or camera inspection becomes easily usable in the screen plane(s). This simple technique allows to detect small translations, rotations, vibrations and temperature effects. Present applications involved construction simple HOEs which can be readily adapted to more specific needs. The basic results are the possibilities of real-time and permanent recordings with azo dyes/PVA LAYERS that greatly simplifies the recording techniques for special experimental arrangements.

CONCLUSIONS

In this paper we presented some progress in holographic characterization of M.Y. 3R/PVA films which are polarization volume photosensitive recording materials. Those films have a uniform spatial frequency response between 500 and 3300 cycles/mm and a 3500 cycles/mm spatial frequency bandwidth. However they allow weak diffraction efficiency holograms but they are erasable and reusable. MORDANT YELLOW 3R/PVA films were found to be more appropriate since its rise time is shorter than two others azo/PVA films previously studied[1]. So, three interesting real-time application techniques were performed with our azo-dyes/PVA films. Moreover, the DC-PVA films studied in the first part of this paper are good candidates for real-time applications and optical processing experiments but those films are not erasable. Presently we are studying many tests to enhance the diffraction efficiency level for the two kinds of coloured PVA films characterized in this paper.

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