Optics Communications 86 (1991) 7–13
North-Holland

OPTICS
COMMUNICATIONS

Holography in frequency selective media: hologram phase and causality

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Received 11 April 1991

Diffraction properties of holograms stored in persistent spectral hole burning media are shown to be directly related to the causality principle and are studied experimentally by using a frequency-tunable arrow band laser. Amplification (erasing) of certain diffraction orders that are allowed (forbidden) by the causality principle is done by manipulating the spectral and phase properties of the holograms. The possibility to minimize cross-talk between holograms stored at different frequencies is also demonstrated.

1. Introduction

Persistent spectral hole burning (PSHB) [1–3] opens new prospects for high density optical information storage [4–6] and processing [7]. By using a narrow-band laser, which is tunable over the full width of an inhomogeneously broadened PSHB-active absorption band, narrow spectral dips or "holes" can be burned and detected allowing an increase in the ultimate density of optical data storage by 4–6 orders of magnitude. Even fully parallel data processing seems possible [7].

Holography is one of the most promising practical techniques for implementing frequency-selective PSHB optical data storage [8]. In the holographic approach, hole burning is carried out with two crossed coherent laser beams (object and reference beam) which bleach a narrow frequency domain hole in the inhomogeneously broadened absorption band and record, at the same time, a spatial holographic fringe pattern. During the read out of the hologram diffraction occurs from this spatial fringe pattern, and the holographic image is reconstructed but only if the read out laser frequency coincides with the spectral range of the burned hole [9–12].

It follows from the dispersion relations that a hole burnt in the inhomogeneously broadened absorption spectrum is always accompanied by a corresponding change in the index of refraction. The variation of the refractive index with frequency has a tendency to extend further than the corresponding absorption index change. If two or more holograms are recorded at adjacent frequencies, the refractive index gratings accompanying each of the absorption gratings, may cause interaction and cross-talk between holographic signals recorded at different frequencies [10].

It is easy to evaluate the diffraction properties (and also to control the interaction) if the number of holographic holes burnt in the spectrum of a PSHB media is not very large. High-density data storage and processing applications imply, however, the recording and reproduction of complicated frequency and space domain holographic patterns. In this case special measures are needed to keep the interaction between different frequencies as small as possible.

One possibility to cut back the cross-talk is to burn in the holograms sufficiently far apart from each other in the frequency coordinate. Such a zero-order approach is unacceptable because it does not make good use of the storage potential of the PSHB media (a large portion of the frequency domain recording space will be wasted). Another way to avoid interaction and to suppress the accumulation of a spectrally non-selective background scattering signal, is to use an alternating relative phase (phase difference of π) when recording holograms that are neighbors in the frequency domain [10]. In this case, however, the holograms stored at different frequencies should have similar spatial structure and contrast.

In the present paper we demonstrate a new technique of manipulating the diffraction properties of frequency-selective holograms which is based upon general causality principle considerations.

Let us note that the effect of refractive index gratings consists, in the first approximation, in the redistribution of the efficiency of diffraction between the positive and the negative orders. If a PSHB hologram is written in a thin polymer film with a narrow band laser at one fixed frequency so that the sample comprises only one grating with a narrow spectral width of $\Delta\omega_{\rm holo} \sim \Gamma_{\rm hom}$, where $\Gamma_{\rm hom}$ is the minimum (homogeneous) hole width, then the hologram diffracts light symmetrically into the positive and the negative diffraction orders. When a second hologram is added at an adjacent frequency, an asymmetry between the holographic signals detected in opposite diffraction orders is observed [10,12].

If a PSHB hologram is produced with a spectrally broad light source, e.g., short laser pulses, with a spectral width large in comparison to the homogeneous hole width ($\Delta\omega_{\rm laser} \gg \Gamma_{\rm hom}$), and, during the recording of the hologram, there is a time delay $|\tau| \gg (\Delta\omega_{\rm laser})^{-1}$ between the writing beams which is due to an optical pathlength difference of d ($d=\tau c$, where c is the speed of light), then the holographic signal reproduced from the hologram will appear either only the positive or only in the negative (conjugated) orders of diffraction depending on the temporal ordering of the pulses [13–18]. Note that the phase of the hologram recorded in this way depends on the frequency as $\phi(\omega) = d\omega/c = \omega\tau$. By using this technique holographic reconstruction of femtosecond signals covering a spectral range of about 150 $\rm cm^{-1}$ has been carried out without any observable interaction or cross-talk between the spectral components [19].

The diffraction properties of non-monochromatic hole burning holograms can be summarized in a "causality condition" by saying that, in general, the response of the hologram detected in opposite diffraction orders is asymmetric and depends on which of the writing beams represents the "past", and which corresponds to the "future". Note that to the "causality condition" to apply one needs to satisfy $\Delta\omega_{\rm holo} > \Gamma_{\rm hom}$, rather than the more stringent condition $\Delta\omega_{\rm holo} \gg \Gamma_{\rm hom}$ which has been valid in pulsed time-domain experiments [13–17]. In other words, if a hole burning hologram occupies an interval in the frequency domain that is broader than the homogeneous hole width, and if the spatial phase of the hologram is made to vary linearly with the frequency ω, the diffraction of the hologram contributes either only to the positive or to the negative diffraction orders, depending on the sign of the phase shift $\phi(\omega)$. If there is now an arbitrary number of holograms at adjacent frequencies, each hologram stored in such a way that it gives only positive or only negative diffraction orders, there will be no further redistribution of the diffraction efficiency between the opposite orders, even if the holograms have quite different spatial structures and contrasts. It follows now immediately that the interaction between the holograms can be completely eliminated. Of course, in the case of a thick hologram, care has to be taken also that the observed diffraction order which is allowed by the "causality condition", also meets the requirements of phase matching.

In the present paper we investigate experimentally the above mentioned causality property of PSHB holograms. We vary the spectral profile of a burned-in, narrow-band hologram using a tunable signal mode dye laser and compare the efficiency of holographic image reproduction when a time delay is introduced into each of the writing beams.

2. Experimental procedure and discussion

The PSHB material we use to record holograms is polyvinylbutyral doped with chlorin at a concentration of the impurity molecules of 10^{-3} M. The hologram sample is a 90 μm thick film and is positioned inside an optical bath cryostat (Oxford MD10). The PSHB active inhomogeneous absorption band has a maximum at the wavelength of 633 nm with a width of 5 nm (130 $\rm cm^{-1}$), the homogeneous hole width at 1.7 K is $\sim$300 MHz (0.01 $\rm cm^{-1}$).

A lay-out of the experimental set up is depicted in fig. 1. The frequency-tunable single mode dye laser (CR 599-21, scan range 30 GHz) and the signal detection system we use in this experiment are the same as described earlier [10]. A thick parallel-polished glass block ($l = 10$ cm) – the optical delay – is positioned either in the reference or in the object arm

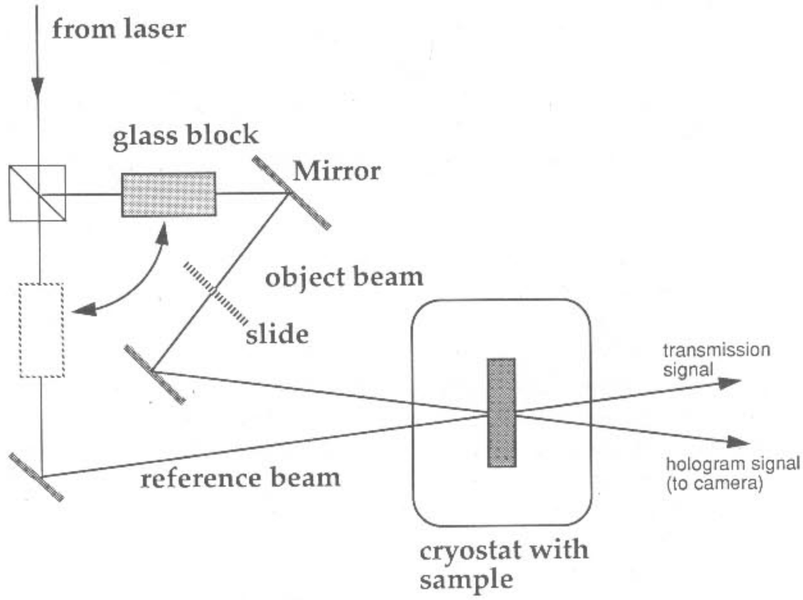

Fig. 1. Optical lay-out of the hologram recording scheme. The glass block can be positioned either in the object beam (positive delay) or in the reference beam (negative delay). Without the glass block the optical pathlengths of the reference and of the object beams are equal within an error of 1 mm.

of the "interferometer". Without the glass block the optical pathlength of the reference and of the object beams are roughly equal within a precision of about 1 mm. The absolute delay value $|\tau|$ between the writing beams is related to the thickness of the block l as $|\tau| = l(n-1)/c$, where n is the glass refractive index. The delay is considered to be positive if the block is positioned in the object arm or negative if the glass block is in the reference arm.

According to the discussion presented above the inverse value of the delay $|\tau|^{-1}$ has to be smaller than the frequency range where the hologram is stored. On the other hand, the delay should not be larger than the inverse value of the homogeneous hole width, otherwise the spectral response of the PSHB media is unable to record the fringe pattern and the contrast of the hologram will vanish. In our experiment $|\tau| \sim 170$ ps which corresponds to $|\tau|^{-1} \sim 6$ GHz ($0.2\ \mathrm{cm}^{-1}$). The spectral width of the holograms we burn is equal to the 30 GHz ($1\ \mathrm{cm}^{-1}$) scan range of the dye laser. The homogeneous hole width is, as already mentioned, $\Gamma_{\mathrm{hom}} \sim 300$ MHz ($0.01\ \mathrm{cm}^{-1}$).

The experimental procedure consists first in writing of a narrow hologram with a spectral width of $\Delta\omega_1 \sim \Gamma_{\mathrm{hom}} < |\tau|^{-1}$ and, afterwards over-writing this hologram with a second hologram with a broader spectral width of $\Delta\omega_2 > |\tau|^{-1}$. Due to its narrow spectral width the hologram resulting from the first exposure is not subject to the "causality condition", while the second exposure increases the overall spectral width of the recorded holograms and switches this condition "on".

Note that changing the sign of the delay does not affect the signal from the spectrally narrow hologram because the diffraction from one (thin) spectral grating is always symmetrical with respect to the positive and negative orders. However, for the spectrally broad hologram, changing the sign of the delay means changing the sign of the allowed diffraction order.

During the read-out of the hologram we block the object beam and attenuate the reference beam by a factor of 4 by inserting a neutral density filter. In addition, the diffracted signal is separated from the passed-through read-out reference beam by focussing through a pinhole before the signal hits the detector. To measure the transmission spectrum of the sample we use a second photomultiplier which monitors the intensity of the passed-through read-out reference beam. The recalled images are recorded by using a CCD video camera.

For technical reasons we are not yet able to detect the holographic signals simultaneously from both, positive and negative orders of diffraction. To overcome this difficulty we first position the delay in the object beam (positive delay) and detect the signal diffracted in the direction of the object beam (+1 order). Secondly, after the read out of the hologram is completed, we change the laser frequency by a few wavenumbers and store a new hologram in a "virgin" spectral region, but now with the delay positioned in the reference arm (negative delay).

For the first exposure (narrow hologram) we keep the laser frequency fixed in the center of the dye laser scanning range. The position of the optical delay is "positive". Typical recording exposure time is 25 s with an average intensity of the writing beams of about 100 $\mu W/cm^2$. The object image in the first exposure is a slide with vertical stripes.

Fig. 2 presents the frequency dependence of the transmitted (a) and the diffracted (b) signals measured after the first exposure. Fig. 3a shows the corresponding image which is reconstructed from the hologram when the laser is tuned to frequency of the narrow hole.

During the second exposure the frequency of the laser is scanned at a constant rate over the 30 GHz frequency interval. In order to achieve an average

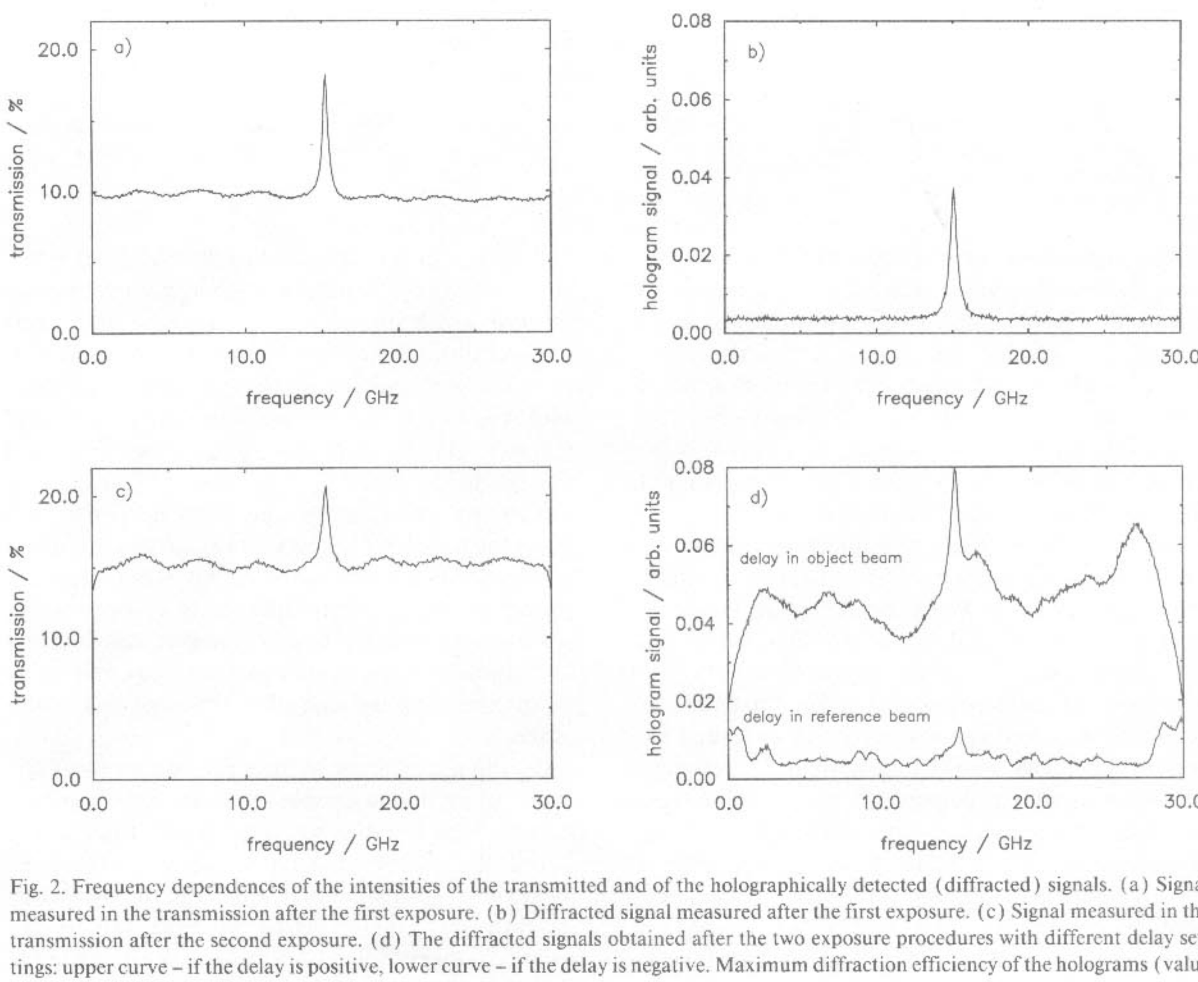

Fig. 2. Frequency dependences of the intensities of the transmitted and of the holographically detected (diffracted) signals. (a) Signal measured in the transmission after the first exposure. (b) Diffracted signal measured after the first exposure. (c) Signal measured in the transmission after the second exposure. (d) The diffracted signals obtained after the two exposure procedures with different delay settings: upper curve – if the delay is positive, lower curve – if the delay is negative. Maximum diffraction efficiency of the holograms (value of the signal relative to the directly transmitted beam) is about 5%.

hologram contrast comparable to that in the first exposure (the second hologram is spread out over a much broader frequency range) we increase the burning time to 8.5 min. The second exposure records an image of a slide with horizontal stripes. The resulting transmission spectrum of the double-exposure hologram is presented in fig. 2c.

The second exposure does not have a noticeable influence on the first narrow spectral region where the hole has been already burnt by the first exposure. This can be observed by comparing the sample transmission spectra before and after the second exposure. The maximum depth of the narrow hole measured right after the first exposure (fig. 2a) corresponds to an increase in the transmission by a factor of 2, while the bleaching effect due to the second exposure (fig. 2c) is smaller (average transmission increases only by 60%). Furthermore, the second exposure increases the maximum transmission only by 15% at the position of the narrow hole. This may be due to partial saturation of the hole depth. For this reason the effects observed in this experiment are associated mainly with the variation of the index of refraction rather than with direct absorption coefficient changes. In the following discussion of the interaction between the holograms we are going to ignore the direct overwriting of the holograms.

Fig. 2d (solid line) shows the holographic signal measured after the second exposure. In the frequency domain two regions with different diffraction behavior can be observed. At frequencies which are far from the narrow hole region (and also far away from the cut-off edges of the laser frequency scan) the diffraction efficiency of the hologram is more or less constant which corresponds to a spectrally uniform bleaching during the second exposure. The image reconstructed within this frequency range (first positive order) is presented in fig. 3b.

We note that the peak intensity of the diffracted signal at the position of the narrow hole has increased as a result of the second exposure by a factor of two. This increase takes place in spite of the fact that the additional hole burning at this particular frequency is negligible as we have shown above. The amplification of the diffraction signal is even more evident in the holographic image reproduced in fig. 3c. At the locations where the two sets of orthogonal stripes overlap the signal is much brighter than could result by just adding the amplitudes of the two images.

To explain this unusual amplification effect let us notice that by burning a spectrally broad hologram on the top of a spectrally narrow hologram we actually cancel the diffraction in the -1 order. Consequently, those sections of the first image (vertical stripes) which overlap with the second image (horizontal stripes) are cancelled from the -1 order (not detected in the present configuration) and instead add constructively to the signal amplitude observed in the direction of the $+1$ order.

To measure the -1 diffraction order we switch the sign of the delay by positioning the optical delay in the reference beam. The image and the spectral profiles of the first (spectrally narrow) hologram are identical to those described for the $+1$ order. The holographic signal measured after the second exposure is different, however, and is also presented in fig. 2d (broken line). The diffraction efficiency of the spectrally broad hologram is much lower than in the previous case. Note also that the holographic signal increases towards the edges of the laser scan region where the spectral profile changes abruptly (such oscillations would not occur if the hologram had a smooth frequency-domain envelope). The image recalled after double exposure at the narrow hole position is shown in fig. 3d. Note that at the image positions where the two sets of orthogonal stripes overlap the intensity of the reconstructed vertical stripes is strongly reduced. The relative brightness of the second image (horizontal stripes) remains low everywhere. The explanation for this effect follows directly from the discussion above: The second exposure does not give any strong diffracted signal in the observed direction and also cancels the signal from the first hologram at the positions where the two images overlap.

The "causality condition" can also be used to perform certain parallel image processing operations. Indeed, if we take as two arbitrary images the first and the second slides in our experiment, then, in the terms of Boolean logic, the procedure used in obtaining an image of fig. 3b is the equivalent of a logic "AND" operation, and the procedure resulting in an image analogous to that shown in fig. 2c is an equivalent of an "ANTICOINCIDENCE FUNCTION".

We also pointed out that the minimum spectral

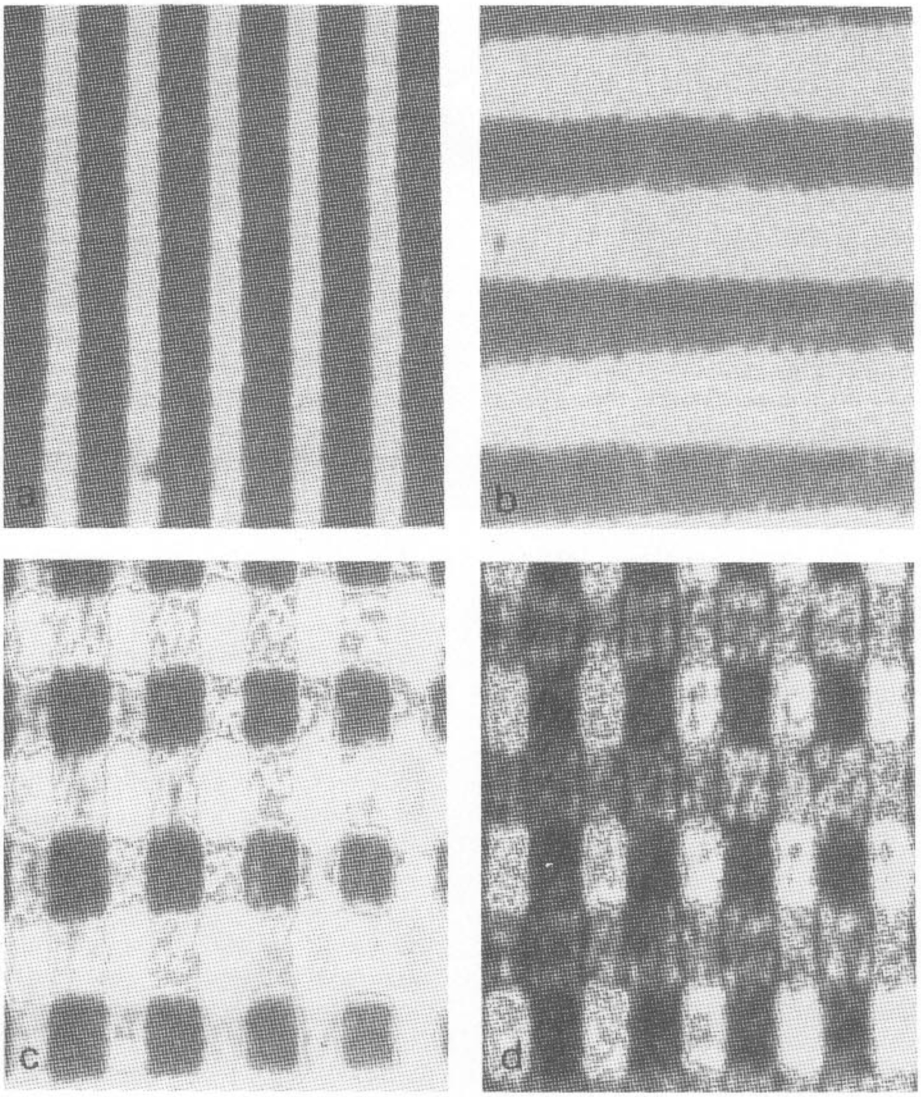

Fig. 3. Holographically reconstructed object images after different write-in procedures and read out at different laser frequencies. (a) After the storage of the first exposure, the frequency of the read-out reference beam is at the maximum of the spectrally narrow hologram. (b) After the storage of the second exposure, read-out laser frequency is tuned several hole widths off from the narrow hologram position. (c) After the same double write-in exposure as in (positive delay) but the read-out laser tuned to the position of the spectrally narrow hologram. (d) After the double exposure storage procedure repeated at a different wavelength with the reversed (negative) delay value, read-out frequency corresponds to the maximum of the narrow hologram.

width of PSHB holograms at which the "causality condition" can still have an effect upon the diffraction properties is about twice the value of the minimum (homogeneous) hole width. This means that by applying the hologram storage method discussed in this paper the cross-talk between the different holograms in spectrally selective media can be eliminated by sacrificing approximately one half of the total spectral storage capacity. On the other hand, if we consider that the amplitude diffraction efficiency of the holograms (thin as well as thick holograms) is in this case higher on an average by a factor of two, then the actual loss in storage (and processing) capacity will be less of a disadvantage.

In conclusion, we have shown that the cancellation of certain diffraction orders from spectrally selective holograms, previously demonstrated for time-domain holograms, applies also in the case of holograms synthesized by a tunable single frequency cw laser beam. On this basis we have suggested a way to reduce cross-talk between holograms recorded at different frequencies and have discussed the possibility of implementing parallel logic operations with holographic images. The experimental results presented here show that the hologram phase is the key to a field of exciting experiments in hole burning holography.

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