# Picosecond time-space holographic interferograms stored by persistent spectral hole burning

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A novel approach to interferometric processing of ultrafast optical signals is demonstrated by using hologram storage in persistent spectral hole burning media. Multiple time-space holographic images of ultrafast scenes are interferometrically compared by using read-out with a frequency-tunable narrow band laser. The feasibility of simultaneous detection of spatial and spectral phase distortions of picosecond wavefronts is demonstrated.

## 1. Introduction

Time and space domain holography in persistent spectral hole burning (PHB) media [1,2] has been recently used to store and process temporally short and spectrally polychromatic optical signals with duration from about a nanosecond to less than 100 fs [3].

It has been suggested [4] that time-space holograms can be applied for the purpose of holographic interferometry of ultrashort transient scenes occurring on pico- and subpicosecond time scales. To conventional holographic interferometry [5], which is based on conventional holographic image storage techniques [6], such short time scales pose serious problems: the extremely short coherence lengths of ultrashort pulses (e.g. coherence length of a 100 fs laser pulse is about 30  $\mu$ m) turns the hologram storage and consequently the formation of interference fringes between the stored-in holographic images into an impractical task.

To overcome this difficulty ref. [4] suggests to use the ability of the PHB media to store and reproduce not only the spatial wavefronts but also the spectral structure of ultrafast images. In the method of time-space holography the stored-in spectral structure is Fourier-related to the temporal dependence of the ultrafast image. To get a complete holographic recording of a time-space image the spectral width of

the signal should not exceed the inhomogeneous absorption bandwidth of the storage medium (typically  $150-300 \text{ cm}^{-1}$ ) and the overall duration of the signal in time-domain should be less than the phase relaxation time  $T_2$  of the PHB-active impurity molecules (for low-temperature organic systems  $T_2$  is typically about 1 ns) [2].

In the present paper we carry out experiments to verify the feasibility of storage of double exposure holographic interferograms of picosecond time and space domain images in persistent spectral hole burning media. We show that by applying a spectrally narrow band read-out of multiple time-space holographic images stored in one PHB hologram an interference pattern is produced between the initially temporally separated holographic images. We also show that time-space holographic interferograms can be used to investigate the spectral dependence of interferometric phase shifts not observably by conventional methods.

### 2. Experimental

In the present experiment we apply two different dye lasers: to record holograms we use a picosecond dye laser which is synchronously pumped by a modelocked Ar ion laser, and to read the holograms we use a narrow band cw dye laser pumped by a cw Ar ion laser. The output pulses of the picosecond laser have a duration of 3–4 ps (measured by a noncollinear autocorrelator) and a spectral width of 10–15 cm<sup>-1</sup>. The linewidth of the narrow band laser is 0.1 cm<sup>-1</sup> and the frequency of the laser can be scanned continuously in an interval of 4.5 cm<sup>-1</sup> by turning an intracavity etalon. The wavelength of the narrow band laser is adjusted to match the wavelength of the picosecond laser. The wavelength and the linewidth measurements are carried out with a MDR-4 monochromator, a Burleigh WA-10L wavemeter and a Coherent 240 spectrum analyzer.

The PHB hologram storage media which we use is similar to that applied in previous experiments [2,3] and is prepared from polystyrene by adding impurity molecules of octaethylporphine (OEP) or protoprophyrine (PrP) at a concentration of  $10^{-3}$ – $10^{-4}$  mol/l. Typical dimensions of the hologram plates are 2 cm across and 4–5 mm in thickness. During the experiments the hologram plates are immersed in superliquid helium at temperature 2 K. The wavelength interval which can be used to record holograms is 617–620 nm for OEP-doped media and 619–624 nm for PrP-doped media. The transmission of the holograms before the write-in exposure varies in these wavelength intervals between 0.3% and 2%.

A scheme of the optical arrangement of the experiment is presented in fig. 1. A semitransparent mirror splits the output beam of the picosecond laser into a reference beam and a signal beam which then intersect at an angle of 10° at the hologram plate positioned inside a cryostat with plain optical windows. The length of the beam path is shorter for the reference arm so that at the hologram the signal pulses are delayed with respect to the reference pulses for 187 ps. The delay is shortened to 47 ps when a 7 cm thick glass block is inserted into the reference arm.

At the hologram the reference beam has a nearly plain wavefront. The wavefront of the signal beam is modified by inserting into the beam before it enters the cryostat different optical elements serving as model samples. Lenses are used to collimate the signal beam. A video camera views the image of the sample and displays it on the screen of a TV monitor. A synchroscan streak camera is used to measure the time domain response of the holograms and to monitor the delays between the write-in pulses.

### 3. Experimental procedure and discussion of results

The first step in our experiment is to store in one hologram two images of an ultrafast scene. To model the scene we use a calibration bar scale engraved on a thin glass plate which is illuminated by a passing-through picosecond laser beam as described in the previous section. The intensity of the write-in beams is 0.1 mW/cm<sup>-1</sup> and the exposure needed to store a high contrast image is 10 mJ/cm<sup>2</sup> per unit [cm<sup>-1</sup>] frequency interval. The corresponding exposure time is on the order of several minutes during which the PHB effect of about 10<sup>10</sup> reference and signal pulse pairs is accumulated.

Note that to store a hologram in the PHB media the reference and the signal pulse need not to coincide in time [1,2]. The hologram is stored if the delay between the reference and the signal does not exceed the value of the phase relaxation time  $T_2$  of the storage media (under present conditions  $T_2$  is about 0.6 ns). It is also required that the reference pulse precedes in time with respect to the signal pulse because the reversed temporal order of the write-in pulses results in writing a conjugated hologram [2].

For the first exposure the delay between the reference and the signal is set to 187 ps. For the second exposure the delay is changed to 47 ps by inserting the glass block delay line into the reference beam. For the second exposure the angle between the write-in beams is also slightly altered. Further in this paper we demonstrate that while the changing of the angle between the beams brings about finite fringes in the spatial domain, changing of the delay between the reference and the signal pulse creates finite interference fringes in the temporal frequency domain [4].

After completing the two successive exposures the signal arm is blocked and a stack of neutral density filters is inserted into the reference arm in order to avoid destruction of the hologram during the following read-out procedures.

The resulting hologram is an analog of a pulsed double exposure holographic interferogram [5] comprising two images of a scene illuminated by picosecond pulses at different time moments. However, because the PHB hologram records not only the images but also their temporal delays, the usual procedure of the read-out of time-space holograms with short laser pulses [1,2] does not give the desired in-

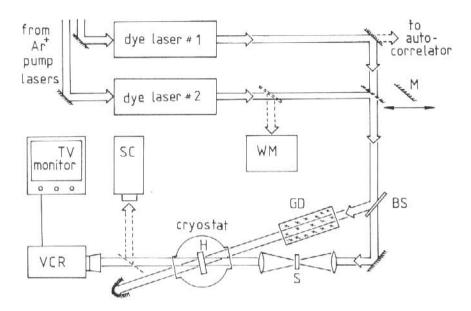


Fig. 1. The experimental set-up. Dye laser #1 is a picosecond Rh6G laser synchronously pumped by a mode-locked Ar ion laser; dye laser #2 is a narrow band CW Rh6G or DCM laser pumped by a cw Ar ion laser; WM: wavemeter and spectrum analyser; SC: synchroscan streak camera; VCR: video camera and video cassette recorder; BS: semitransparent mirror; M: mirror used to switch between the picosecond and the narrow band laser beams; H: hologram plate; GD: thick glass block optical delay; S: sample creating model images.

terferogram. Indeed, the two signal light pulses which are recalled from the hologram are separated in time by an interval that greatly exceeds the coherence time of the picosecond laser pulses and therefore cannot produce mutual interference in the usual understanding of this term (see, for example ref. [8]). This is illustrated in fig. 2 where the image recalled from the double exposure hologram by picosecond readout is reproduced. Fig. 3 presents the corresponding time domain streak camera recording of the double exposure hologram output. Although the hologram comprises high contrast images of both picosecond scenes, no interference between them can take place due to the 140 ps temporal delay between the signals (the coherence time of the pulses estimated from their spectral width is about 1 ps).

Our next step is to block the output of the picosecond laser and to illuminate the hologram with the attenuated beam of the narrow band cw dye laser. The frequency  $\nu_L$  of the cw laser output is tuned to the wavelength of the recorded double exposure hologram. The resulting holographic image displays high contrast interference fringes and is reproduced in fig. 4.

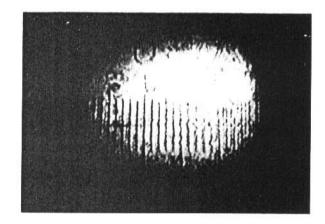


Fig. 2. Double exposure holographic image recalled by illuminating the hologram with attenuated picosecond laser beam. In the experiment the images are first recorded on a videocassette and later photographed from the TV monitor.

The reason why the interference fringes are observed is correctly understood if we consider, first that the PHB hologram actually comprises many holographic images which correspond to different time domain frequency components of the ultrash-

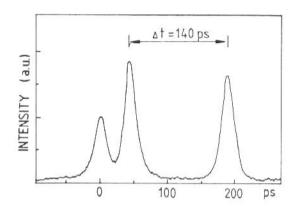
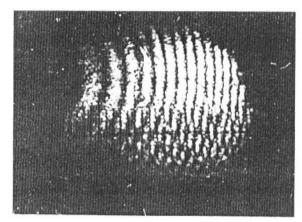


Fig. 3. Streak camera recording of the time-domain signal at the output of the double exposure hologram illuminated by the attenuated reference beam from the picosecond laser. The signal at zero-delay corresponds to the passed-though read-out pulse. The pulses at the delays of 47 ps and 187 ps correspond to the images recorded at the second and at the first exposure. The delay  $\Delta t$  corresponds to the delay interval of the glass block.

ort signal and which are recorded at different optical frequencies in the PHB media [1,2]. Secondly, as the PHB hologram can be regarded simply as a linear filter, the coherence of the read-out laser determines also the coherence properties of the recalled signal. The coherence time of the narrow band laser is about 0.3 ns, i.e. almost twice the temporal delay between the two stored images. Consequently, the narrow band read-out reproduces only the corresponding  $\nu_{\rm L}$  frequency components of both of the stored images. At the output of the hologram the two signals are mutually coherent and therefore give the observed spatial interference fringe pattern.

The fringe pattern corresponding to a certain readout frequency  $\nu_L$  can be interpreted in the same way as in the case of conventional pulsed holographic interferograms [5]. However, by analysing the fringe patterns recalled at different read-out frequencies it is possible to get additional information about the ultrashort signals such as temporal and frequency domain amplitude and phase differences between the two scenes [4].

In the case of the two recorded scenes having identical temporal (and spectral) amplitudes the  $\nu_{\rm L}$ -dependence of the interferogram is trivial: the phase of the fringes has strictly linear dependence on the frequency  $\nu_{\rm L}$  [4]. This situation is illustrated in fig. 4a,



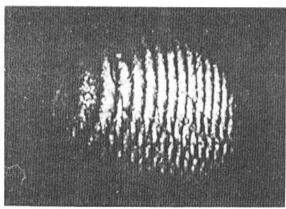
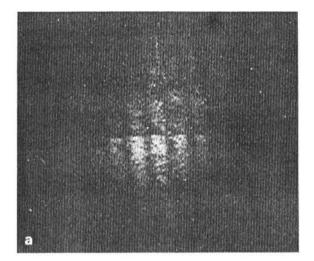


Fig. 4. Double exposure interferogram recalled from the same hologram as in fig. 3 by narrow band read-out. The curved shape of the fringes is due to the uneven end surfaces of the glass block delay line. The frequency dependence of the phase of the spatial fringes is strictly linear and changes by  $+\pi$  ( $-\pi$ ) when  $\nu_{\rm L}$  is increased (decreased) by 0.15 cm<sup>-1</sup>; (a)  $\nu_{\rm L} = 16051.45$  cm<sup>-1</sup>; (b)  $\nu_{\rm L} = 16051.30$  cm<sup>-1</sup>.

b. Scanning of  $v_{\rm L}$  in either direction produces a corresponding continuous phase shift of the whole fringe pattern; reversing of the scanning direction reverses also the direction of the phase shift. By scanning  $v_{\rm L}$  in a range of 4.5 cm<sup>-1</sup> at every given point of the image an alternation of about 16 interference minima and maxima is observed whish agrees with the value of the frequency domain fringe period estimated from the delay (140 ps) between the two signals.

Fig. 5 illustrates a situation where the time-space interferogram reveals small frequency domain phase distortions of a model picosecond signal. The upper



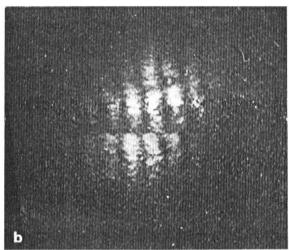


Fig. 5. Holographic interferogram displaying a nonlinear frequency dependence of the fringe pattern. The image above the dark horizontal line corresponds to a reflection coated parallel glass plate acting as a thin Fabry-Perot etalon. The lower part of the image serves as a reference. When the read-out laser frequency is changed from  $\nu_L = 16601.0 \text{ cm}^{-1}$  (a) to  $\nu_L = 16604.0 \text{ cm}^{-1}$  (b) a  $\frac{1}{4}\pi$  phase shift is observed between the upper and the lower part of the interferogram.

part of the image in fig. 5 corresponds to a reflective-coated parallel glass plate (a thin Fabry-Perot etalon) which is inserted into the signal beam for the first exposure. The fraction of the write-in signal beam that passes through the etalon experiences due to the dispersion caused by the optical element a small frequency-dependent nonlinear phase shift. As a result the frequency dependence of the phase of the

interferogram fringe pattern in the upper part of the image is no more strictly linear. The nonlinearity of the phase shift of the fringes is observed by comparing it to the lower part of the interferogram which corresponds to an undistorted image (identical to that presented in fig. 4), i.e. a strictly linear phase shift. By scanning  $\nu_{\rm L}$  in a range of 3.0 cm<sup>-1</sup> a phase shift on the order of  $\frac{3}{4}\pi$  is observed between the upper and the lower part of the image.

In principle, by this method ultrafast transient dispersion changes can be measured with an interferometric precision even if the phase shift varies spatially over the cross section of the beam of the probing laser. Note that in the present experiment, however, the observed phase shift is a trivial consequence of the stationary dispersive properties of the Fabry-Perot etalon.

In conclusion we have demonstrated picosecond holographic interferograms stored by persistent hole burning in low-temperature impurity media. This method presents a unique possibility to combine the usual spatial interferometry with the time- and spectral domain processing techniques of ultrafast optical signals and extends, as we believe, the range of possible applications of holographic interferometry.

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