Laser Chem. 1986, Vol. 6, pp. 165–180
0278-6273/86/0603-0165$12.00/0

Printed in Great Britain

SPACE-TIME HOLOGRAPHY OF PICOSECOND PULSED FIELDS IN HIGHLY-SELECTIVE PHOTOCHROMIC MEDIA

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1. INTRODUCTION

The recording and reproducing of spatial and temporal properties of rapidly-changing optical signals and fields of picosecond duration are a currently central scientific and technological problem in contemporary optics, the solution of which may determine the ways of developing the future ultrahighspeed performance computer technique. The key problem is how to record and then retrieve ultrashort light pulses.

In a series of investigations[1-5], it was shown that by means of spectrally highly selective photosensitive materials, i.e. the media able to memorize the spectral composition of the absorbed light, it is possible to store and reconstruct the time behaviour of optical pulses on a hololgraphic principle.

The experiments performed on photoreactive impurity molecules in frozen matrices, where picosecond light pulses were recorded and highly efficiently (~50%) restored, show how promising such materials are in practical applications.

In the present talk (1) the main idea of holographing the time dependences of the optical signal is explaned: it is based on the phenomenon called photochemically accumulated stimulated photon echo; (2) the process of holographing time-development of motion pictures is analysed, and (3) some practical applications are discussed.

2. HOLOGRAPHY OF TIME DEPENDENCES OF THE OPTICAL FIELD IN SPECTRALLY SELECTIVE PHOTOSENSITIVE MATERIALS

In this part the following questions will be answered: (1) How to obtain a spectrally highly selective photosensitive medium; (2) what is the photochemically accumulated stimulated photon echo (PASPE), and (3) how is it possible to perform the holography of optical field time-dependences on the basis of PASPE.

To explain the essence of our method it should be noted that impurity molecules in low-temperature matrices have extremely narrow (10^{-3}-10^{-4} cm^{-1}) lines of purely electronic transition (s.c. optical analogue of the Mössbauer line)[6]. It is also known[7-9] that if such systems undergo photoinduced transformations upon resonance monochromatic excitation, the inhomogeneously broadened spectrum may develop narrow stable gaps whose widths are limited by the homogeneous width of zero-phonon lines (hole burning effect[7-9]) and shich may persist for a number of days, but also for months or even years. Thus, based on photochemical hole burning, it is possible to create a spectrally highly selective photosensitive midium.

If a train of picosecond pulses is introduced into such medium, in the absorption spectrum of the medium a narrow-band transmission grating with the "fringe spacing" $\Delta\nu = 1/\tau$ (τ - interval between the pulses) can be created, i.e. a spectral hologram[1)] in the form of the Fourier transform of the temporal shape of the train can be made.

As regards the treatment of the recording, it is important to

1) The calling of such spectral grating as hologram and the described method as a holographic one will be justified below.

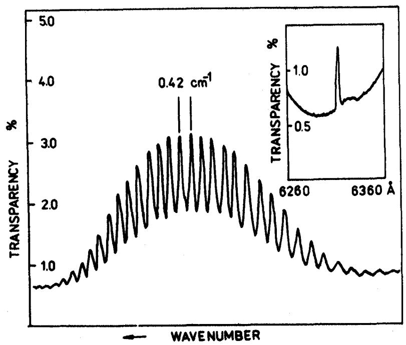

FIGURE 1 The spectral grating (hologram) obtained in sample I after burning by a train of picosecond pulses with 80 ps repetition rate. The envelope of the modulated hole corresponds to a single pulse spectrum; the fine structure is determined by the pulse interval in the train. On the right a general wiew of the hole is depicted, which is recorded with a lower spectral resolution.

keep in mind also that the width of the ingomogeneous spectrum must exceed that of the picosecond pulse spectrum and that the phase relaxation time T_2 of the operating transition must exceed the duration of the train. Transmitting a single attenuated picosecond pulse through such a hologram like linear spectral filter leads to the transformation of the power spectrum of the reading pulse, that in its turn should lead to the appearance of repetitive pulses (interval 80 ps) in the response. The experiment[2)] shows that in the response of a single attenuated probe pulse, transmitted through the transparent with a spectral grating (Figure 1), there really appear complementary emission pulses with a repetition rate corresponding to that in the burning train (Figure 2)[2,3].

It is noteworthy that the relative intensity of "echo" pulses is high - 4% of that of the passed-through probing or reading

2) The experimental set-up, data on samples and methods are presented in the appendix.

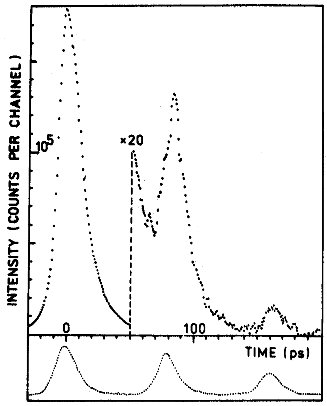

FIGURE 2 Time response of sample I after burning the spectral grating (Figure 1) by a pulse train shown below. First from the left - the probing pulse. For recording a synchroscan streak camera system was used.

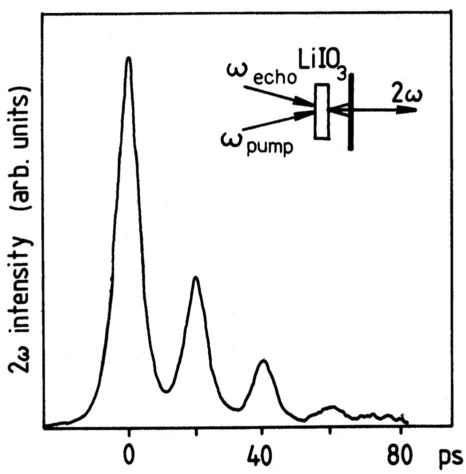

FIGURE 3 Time response of sample II after burning the spectral grating by a pulse train (20 ps intervals) detected by up-conversion. The integral intensity of the restored train is approximately equal to that of passed-through probing pulse.

pulse. Moreover, after selecting the spectrally selective photochromic medium (sample II) one managed to restore picosecond pulse trains with ~50% efficiency[4] and to observe up to six "echo" pulses (Figure 3).

To explain the nature of this phenomenon and its relation to what has been known earlier, the following must be noted. First, the fine spectral structure of the hole can be regarded as a result of interference: the coherent excitation of molecules, created by the initial reference pulse, interacts with the following ("object" or "code") part of the illumination. Analogously to common (space-domain) holography we store amplitude as well as phase relations between the spectral components of the code and reference pulse. This explains also why the interference-created spectral grating is called a hologram and the proposed method, a holographic one. Second, physically, the complementary pulses in the hologram's time response are a spontaneous coherent emission (free decay singal) of an ensemble of coherently excited dipoles with a specially prepared inhomogeneous distributuion of transition frequencies. It follows that (a) the duration of the recorded and retrieved signal is confined by the phase relaxation time 10^{-9}-10^{-7} s; (b) the phenomenon may be interpreted as a new modification of (stimulated) photon echo or photochemically accumulated stimulated photon echo (PASPE) peculiar to highly-selective photochromic media.

Differences between the phenomena originate from the differences in depopulation mechanisms of the excited state and characteristic restoration times of the absorbers ground state population (for stimulated echo the excited state lifetime T_1 of molecules is topical for PASPE, the life time of photoproducts $T_p \gtrsim$ 10 hours or maybe years). From here a number of PASPE properties useful in practical applications can be inferred:[3)] (1) it is easy to accumulate a high-contrast interference pattern in the medium even in the case of weak pulses by a multiple ($\gtrsim 10^{10}$ times in this experiment) repeating of the recording cycle during a prolonged period; (2) the processes of recording and recalling the run of picosecond events are distinguishable: the formation of PASPE is reduced to a linear filtration of the reading pulse whose intensity application time and even repetition rate do not essentially affect the relative intensity of the response pulses. High efficiency is another advantage of PASPE (see, e.g. Figures 2 and 3) A determining factor here is the modulation depth of the spectral hologram - grating, which in its turn is limited by the homogeneous linewidth ($\sim 1/T_2$) and by saturation in the process of burning.

Assume that the signal (object) pulse is of a complex structure and the reference pulse is shorter than any of the shortest features of signal pulse, then, on recording, owing to interference between the excitation pulses, a hole of a complex form appears in the absorption band, which is determined by the mutual spectra of reference and code pulse. It is the interference with sufficiently short pulse (see above), i.e., whose spectrum in the region of code pulse spectrum can be considered uniform, that allows the phases of the harmonic components of the latter to be fixed.

On transmitting a short reading pulse through such spectral hologram as a linear filter, its response reveals a pulse with a form and structure coinciding with that of the initial signal pulse, i.e. analogously to the previous simple case of a train a pulse of a complex form is retrieved.

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3) The correlation of stimulated-echo shapes with those of excitation pulses was first pointed out in[10,11], applicabilities of "ordinary" photon echo in holography is discussed in[12-15].

3. TIME-SPACE HOLOGRAPHY

In the previous chapter the recording and retrieval of only the time dependence of pulses with a plane wavefront was regarded. Now we shall consider the holography of both space and time domain properties of pulsed light fields. In the course of this observation we shall try to answer the following questions:

(1) What requirements are set to the recording process?
(2) What are the pecularities the spectral hologram reveals on recording the pulses with complex wavefront?
(3) What are the distinguishing features of the retrieval process in comparison with the common space domain holography?
(4) How the changes of the refraction index (phase grating) of the spectral hologram act?

Suppose that on recording the short reference pulse with a plane pulse front and the pulse scattered from the object scene (object pulse that is of a complex spatial and temporal structure) fall onto the spectrally selective photochromic medium at some angle (Figure 4). Such scheme (s.c. off-axis scheme) is well known in ordinary holography and it allows one to avoid a spatial overlapping of the undiffracted wave with restored images. Now, if the reference pulse is delayed with respect ot the object one, then in each medium volume element, besides the spectrum of each pulse, their mutual spectrum is traced in the form of a spectral absorption grating whose fringe spacing is determined by this delay[5]. In different sites of the hologram, in off-exis scheme, the delay between excitation pulses varies and therefore the carrier frequency of the spectral grating also changes from site to site.

It should be noted that in the spectral hologram the spatial modulation of medium transmittance is observed only in the case of narrow monochromatic radiation components. As the period of this

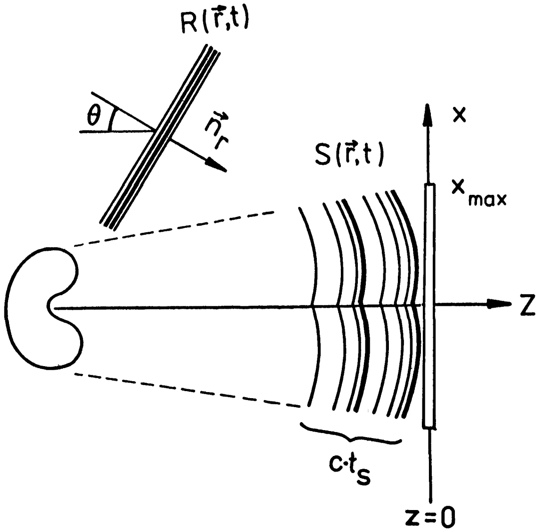

FIGURE 4 The recording scheme of a space-time hologram $R(\vec{r},t)$ is the reference pulse; $S(\vec{r},t)$ the object pulse. The photosensitive medium with the dimension $2x_{max}$ lies in the plane z = 0.

spatial modulation is determined by the frequency of the component, the interfringe spacing varies for different components, that results in the smearing of the spatial light-induced grating for white light.

However, if the object pulse will be overlapped by the reference one in time, in the region of pulse overlapping an ordinary interference pattern can be observed and as a result a spatial modulation of hologram in white light transmittance appears.

Proceeding to the process of restoring, one should note that in order to avoid the loss of information the reading pulse should be short in the same sense as the reference one, or the latter should be used for reading.

On restoring by a pulse with a flat wavefront towards the

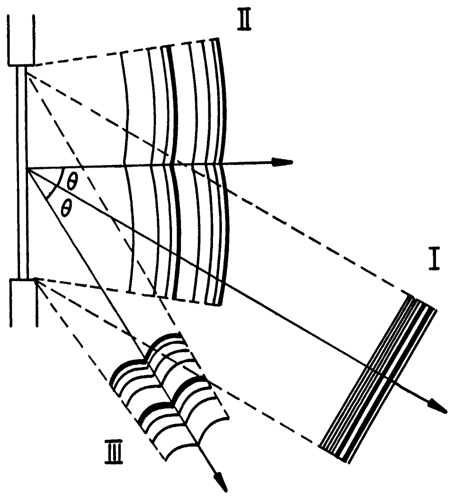

FIGURE 5 The pulses forming behind the hologram in the course of restoring process. I - the undiffracted part of the restoring pulse in $\vec{K}_R$ direction; II - the pulse in $\vec{K}_S$ direction, which forms the virtual image run; III - the pulse in $2\vec{K}_R-\vec{K}_S$ direction, which forms the real image of the events.

reference pulse ($\vec{K}_3 = \vec{K}_R$) there are two possible directions for the image-forming hologram-diffracted waves (see Figure 5): in the direction of $\vec{K}_S$ appears the virtual image of object events and in the direction of $2\vec{K}_R - \vec{K}_S$, the real one.

The most interesting feature of the spectral hologram is its ability to distinguish on restoring the "future" and the "past" in the word-for-word meaning: the virtual image of the events taking place in the object scene after the application of the reference pulse, tends to one direction, while the real image of the object events before the reference pulse, to anoter (Figure 5). In other words, depending on the sequence of the arrival of signal and reference pulses, on restoring on, one of the images is imposed prohibition.

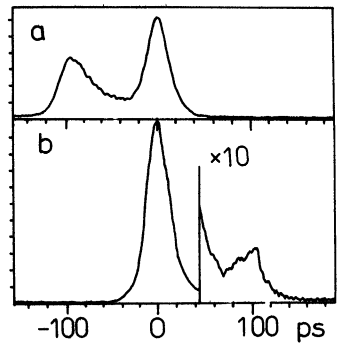

FIGURE 6 Recording and restoring the asymmetric signal pulse in the case when it is first sent to the medium. a - the asymmetric signal pulse and the delayed reference pulse; b - the restoring pulse and the restored time-reversed pulse; in Figure 5 to the latter corresponds the 3rd beam.

As became clear in the previous part already, the spectral hologram is able to memorize also the arrival time of the object pulse. This means that the replicas of the object scene scattered over such hologram are delayed with respect to the restoring pulse in accordance with the delay on recording.

Besides, the real image is formed by a wave whose front is conjugated with respect to the object wave and which has an inverse time behaviour (see Figure 6). The on-and-off switching of the directions of hologram-scattered waves or the prohibition of one of the images is conditioned by the causal behaviour of the volume element polarization, that is always ensured by the dispersion relations between the absorption coefficient and the refraction index of the medium.

If one disregards the spectral phase grating, which inevitably exists together with the amplitude grating, the restoration of both the virtual and real images is allowed, that contradicts the causality principle. More exactly, the account of the refraction index changes in the spectral hologram forbids the restoration of the image which, according to the pulse sequence, should be scattered until the arrival of the reading pulse. This is what leads to the violation of causality in the previous case.

4. SOME APPLICATIONS OF SPACE-TIME HOLOGRAPHY

High efficiency, linearity of recording and reading, and especially the longevity of storing allows the PASPE-based space-time holography to be considered promising for applicational purposes. The results of this work leave no doubt about the feasibility of the holography of space-time events of picosecond and sub-picosecond scale holographyc movie of ultrahighspeed processes. Besides, the PASPE phenomenon itself and the holographic methods based on it have a number of applications. Some of them are considered below.

1. Phase Relaxation Time Measurements

The interference of the reference and object pulse in a selective photosensitive medium are limited by the phase relaxation time T_2 of the excited state of the impurity. The longer the interval between the pulses is, the larger is the number of the excited molecules which have forgotten the phase of the first excitation pulse before the arrival of the second one. This leads to the decrease of the modulation depth of the spectral hologram, that in its turn leads to the decrease of the relative intensity of the echo pulse according to the law $\exp(-4t/T_2)$ [4]. Thus the relaxation time T_2 of the impurity molecule can be found from the dependence of the relative intensity of the echo signal on the delay t.

2. Parallel Detection of Photochemical Holes

The employment of photochemical burning of persistent holes in the impurity absorption band of low-temperature solutions allows the density of binary information stored in optical memories to be increased at the expense of a spectral coordinate by several orders[18,19]. In principle the number of the holes that can be formed in the inhomogeneously-broadened impurity absorption band by photochemical burning is limited by the ratio of the widths of the ingomogeneous absorption band and the homogeneous nophonon line. For a purely electronic line this ratio may be as much as 10^4 - 10^5, that leads to a limit information packing of 10^{12}-10^{13}bit/cm^2. Unfortunately, a successive recording of holes by monochromatic excitation is a time-consuming procedure. The process of hole burning and detection can be essentially accelerated by using the PASPE phenomenon. This allows one to perform the parallel recording as well as reading of photochemical holes. Thus, by means of PASPE it has been shown[20] that it is possible to burn 1600 holes in the absorption band of object II whereas such amount of holes does not engender any distortion in their form or contrast. In conslusion, this approach allows a parallel information recording and reading over all frequency components of the memorizing medium and the use of ultrahighspeed light pulse modulation for information coding.

3. Synthesis of Light Pulses with a Given Arbitrary Form and Wave Front

By using CW tunable single-frequency lasers it is possible, in principle, to synthesize and fabricate such spectral amplitude-phase holograms which on transmitting a reading pulse allow one to obrain pulses of any form and wavefront. In other words, we can form light pulses of any spatial and temporal properties. For that it is necessary to create the spectral grating-hologram in each volume element (of λ-size magnitude) of the selectively photosensitive medium by means of the laser so that on restoration the scattering from the whole area of the hologram should result in the formation of a pulsed light field with the desired spatial and

APPENDIX

Experimental Methods and Samples

As a spectrally selective photosensitive medium solid polymerized solution of H_2-tetra-tret-butyl-porplyrazine (sample I) and octa-ethylporphin (sample II) in styrol, cooled down to liquid helium temperature, were used. The inhomogeneous width of the operating absorption bands were respectively 500 cm^{-1} and 200 cm^{-1}, the homogeneous width of purely electronic lines, $\sim$0.1 cm^{-1} and $\sim$0.05 cm^{-1}. The samples were prepared in the form of 3-10 mm-thick platelets with optical density D = 1-3.

Photochemical holograms were burned and probed by picosecond CW dye (R6G) laser, which was synchronously pumped by an argon laser with active mode-locking (Spetra Physics, models 375 and 171). Pulse repetition rate (duration 2-3 ps, spectral width 5 cm^{-1}) was 82 MHz. Spectral measurements were performed by the same dye laser working in the CW mode and having a complementary scanning etalon in the resonator (oscillating linewidth 0.075 cm^{-1}). Transmission spectra were recorded by a double-channel photon counting system.

The hologram time response was studied by a synchroscan streak camera system[20] with time resolution of about 20 ps or by up-conversion of the echo signal in a $LiIO_3$ crystal in a noncollinear scheme by using a gating picosecond pulse with variable delay. The dependence of up-conversion signal on the delay was recorded by a phase-sensitive detector.

The average intensity of laser emission of burning (recording) and probing (image restoring) was 0.1 mW/cm^2 and 0.1 W/cm^2, respectively.

To form the reference and object pulses delayed with respect to one another Fabri-Perot and Michelson-type interferometers were used.

temporal characteristics.

4. Wave Front Conjugation

As was shown above, if on recording in the selectively photochromic medium an object pulse is sent first then a time-reversed replica of the object pulse is restored. When including to the treatment 3 space dimensions, on restored by a pulse of a direction reverse to the reference one ($-\vec{K}_R$), a simultaneous conjugation of the wave front occurs.

In conslusion, the restored pulse is a backward-propagating phase conjugated and time reversed exact copy of the signal pulse.

5. CONCLUSION

In the case under consideration the holographic process is generalized to a case of photosensitive materials which are able to memorize not only the spatial distribution of the field intensity but also its spectral compositon. The proposed and experimentally realized method of the holographic recording in the familiar media with photochemically active impurity absorption centers allows an object scene of 10^{-8} to 10^{-13} s duration to be restored in its full time dependence with an efficiency sufficient for practical applications. In essence the space-time holography solves the problem of real recording of moving space domain pictures, if to bear in ming that the cinematographic method of imagewise shooting does not allow the image changes to be restored in the direct sense of the word.

It is also shown that photochemically accumulated photon echo and the space-time holography based on this phenomenon have a number of promising applications.

The authors are indebted to Prof. K. Rebane for fruitful and stimulating discussions.

Two geometries were used: (a) collinear direction of all beams to investigate only spectral-time dependences; (b) the signal and reference beams separated to an angle $\Theta \leqslant 1°$ to study both spatial and temporal dependences. For a spatial formation of the signal pulse one shoulder of the Michelson interferometer was supplied with 2m-focal-length mirror. Behind the cryostat, where the samples were introduced, on a special screen it gave an image of a point adjacent to the spot from the passed-through plane reference wave. The angle between the reference and object wave was chosen by detuning of the interferometer.

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