

Picosecond time- and space-domain holography by photochemical hole burning

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We demonstrate persistent storage, recall, and conjugation of picosecond light signals from various model objects, including a coin, by making use of coherent optical responses in photochemically active media. A simple linear theory of holographic storage and playback of both the spatial and the temporal behavior of the signal field is shown to describe well the experimental results obtained by utilizing octaethylporphyrin-doped polystyrene at 1.8 K as a spectrally selective recording material.

1. INTRODUCTION

Photochemical hole burning (PHB),^{1,2} in addition to its well-known applications in high-resolution laser spectroscopy of impurities in solid matrices,^{3,4} has also been considered a promising method for optical data storage,⁵⁻⁸ since PHB allows the data bits to be attached not only to different spatial locations of an optical memory but also to different optical frequencies within an inhomogeneously broadened impurity absorption band.

Traditionally, in both spectroscopic and data-storage applications, PHB is performed by burning spectral holes through tunable monochromatic laser excitation in inhomogeneously broadened absorption bands of impurity molecules. Consequently, these persistent holes are detected by fine scanning of the excitation over the modified inhomogeneous spectra either to perform the readout of spectral data bits or to obtain homogeneous spectral characteristics of impurity molecules.

In Ref. 9, a radically different approach, designated time-domain frequency-selective data storage, is proposed in which all the data bits assigned to a given spatial location are written in parallel by a temporally modulated laser pulse. We have demonstrated experimentally that time-domain PHB can be carried out by sequences of coherent picosecond pulses, with the spectral width 2 orders of magnitude larger than the homogeneous hole width (0.05 cm^{-1}).¹⁰ As the duration of the applied sequences of burning pulses is less than the impurity-molecule phase relaxation time T_2 , the complex shape of the resulting hole corresponds to the Fourier spectrum of the pulse sequence, i.e., the width of the hole envelope is reciprocal to the pulse duration, whereas the fine spectral structure (containing up to 10^2 peaks) of the hole originating from the interference of the burning pulses is determined by the number and the intervals of the pulses in the sequence.¹¹

We have also shown^{11,12} that passing a single arbitrarily weak probe picosecond pulse through a sample with such a hole in its inhomogeneous absorption spectrum will result in delayed echo pulses that completely reproduce the burning-pulse sequence and have very high (up to 50%) intensity compared with the transmitted probe pulse. These delayed pulses were interpreted as the temporal response of the

sample acting as a linear filter with a burned-in persistent spectral transparency grating.

On the other hand, in terms of time-domain coherent phenomena, the physical nature of these coherent optical responses lies in free-induction decay under weak excitation. For that reason, we have termed the observed phenomenon, photochemically accumulated stimulated photon echo (PASPE), in that it is most closely related to the accumulated three-pulse stimulated photon echo.^{13,14} However, PASPE displays several special features. First, although the stimulated photon echo can occur only within the relaxation time limits of light-induced transient frequency-domain population gratings, the lifetime of PASPE is determined by the very long lifetime (at least many hours, maybe even years) of PHB photoproducts, and so the time available to recall the frozen-in echo is practically infinite. This allows the spectral gratings in PASPE to be accumulated to a very high contrast, which in turn results in very high relative-intensity echo signals.

Second, because of the accumulation effect, PASPE experiments can be performed well under modest linear excitation conditions, and, in the first approximation, a simple theory based on a linear dielectric susceptibility approach can be established.¹⁵

Third, since only weak readout pulses are required, the replica of the signal, once it is stored, can be recalled from the sample many times before distortions may appear.

These special features make PASPE useful in determining impurity excited-state phase relaxation times.^{16,17}

But, from our viewpoint, one of the most interesting applications of PASPE lies in the persistent holographic recording of both temporal and spatial characteristics of ultrashort light pulses.^{15,18} Let us suppose that the PHB sequence comprises an object pulse with arbitrary temporal and spatial amplitude and phase distribution and a plane-wave reference pulse, which is short enough to be considered a δ pulse as compared with the signal. In Ref. 15, where the theory of time- and space-domain holography is developed by extending the formalism of ordinary holography into the fourth dimension, we have demonstrated that, if the phase relaxation time T_2 of the PHB media is long enough compared with the duration of the signal, exhaustive information about the signal can be stored by means of spatial and spec-

tral distribution of the PHB bleaching effect and later reproduced by PASPE.

Again, bearing in mind the special features of PASPE, it should be stressed that PASPE holograms, unlike the echo holograms in ordinary resonant media,^{9,19,20} are able to play back object scenes with their temporal dependence at arbitrary moments after storage.

The purpose of the present paper is to report the experimental results on recording temporal and spatial characteristics of ultrashort light signals by using PHB and on playing back their holographic images by using PASPE as well as to demonstrate accordance of the results with theoretical predictions. In Section 2 we shall give a brief account of the linear theory of time- and space-domain holography in spectrally highly selective media. Then, after listing main experimental details, we shall discuss the features of direct and phase-conjugated holographic images of light signals formed from picosecond pulses by model transparencies. Particularly, we shall demonstrate the possibility of complete elimination of wave-front distortions by using the conjugated image (earlier accomplished in nonlinear media; see Ref. 21 and references therein). Finally, we consider a PASPE hologram of a coin illuminated by a picosecond pulse that demonstrates the far-reaching prospects of this type of holography for practical applications.

2. THEORY

Following Ref. 15 we outline here the derivation of formulas governing the holographic process.

Consider a plate with dimensions $(2x_{\max}, 2y_{\max}, d)$ containing photochemically active dye molecules having zero-phonon absorption lines (ZPL's) of homogeneous width T_2^{-1} around frequency ω_0 . Let the plate be illuminated from the $z < 0$ side, as shown in Fig. 1, by an object pulse

$$S(\mathbf{r}, t) = s(x, y, t - z/c) \exp[i\omega_0(t - z/c)], \quad (1)$$

with a spectral width $\Delta\omega_s$ and a duration $t_s \ll T_2$. If the front edge of the object pulse reaches the plate at moment t

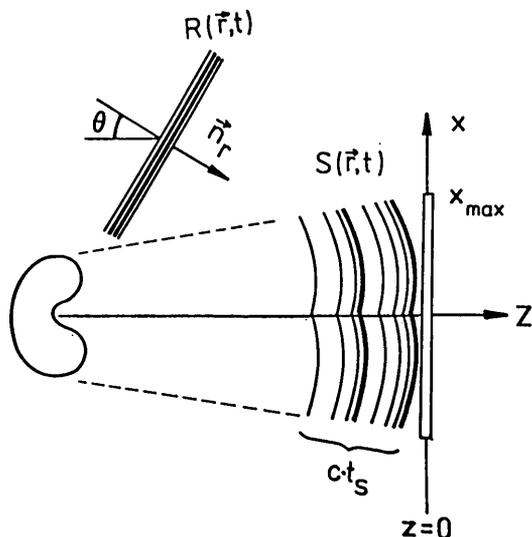


Fig. 1. Diagram of the proposed holographic-recording procedure. Object pulse $S(\mathbf{r}, t)$ reaches the hologram plate at the moment $t = 0$; reference pulse $R(\mathbf{r}, t)$ is shown to be delayed, i.e., $t_R > t_s$.

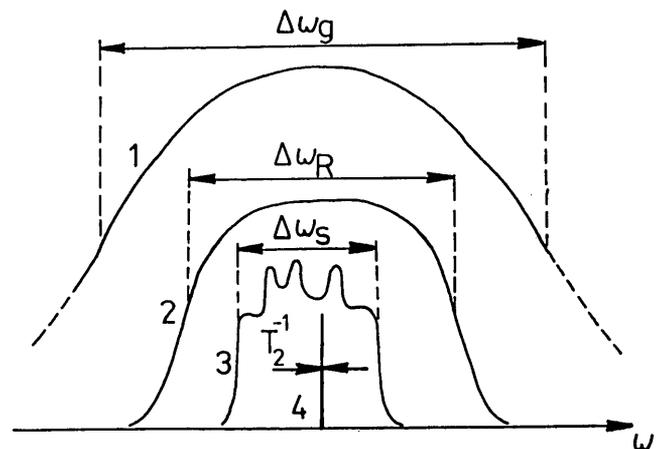


Fig. 2. Relations among spectral widths of 1, inhomogeneous absorption band; 2, reference pulse; 3, object pulse; and 4, homogeneous ZPL linewidth.

$= 0$, the trailing edge of the pulse will leave the plate at $t = t_s$.

Let us further suppose that, with the delay t_R relative to the front edge of the object pulse, the plate is also illuminated by a short plane-wave reference pulse, which is passed through the plate at a small angle θ with respect to the Z axis (all the approximations made and inequality relations used have been discussed in Refs. 15 and 18; see also Fig. 2). If the reference pulse has a duration small enough compared with that of the object pulse, it can be considered a δ pulse and can be written as

$$R(\mathbf{r}, t) = R_0 \delta(t - \hat{n}_R \cdot \mathbf{r}/c - t_R) \exp[i\omega_0(t - \hat{n}_R \cdot \mathbf{r}/c - t_R)], \quad (2)$$

where $\hat{n}_R = (-\sin \theta, 0, \cos \theta) \sim (-\theta, 0, 1)$ is a unit vector in the direction of the reference pulse.

The intensity spectrum of the sequence of object and reference pulses acting jointly on the medium is given by

$$I(x, y, \omega) = R_0^2 + |\bar{s}(x, y, \omega - \omega_0)|^2 + R_0 \bar{s}(x, y, \omega - \omega_0) \exp[-i\omega(x\theta/c - t_R)] + R_0 \bar{s}^*(x, y, \omega - \omega_0) \exp[i\omega(x\theta/c - t_R)], \quad (3)$$

where \bar{s} stands for the Fourier image.

Optical properties of the medium of the plate are described by means of its dielectric permittivity

$$\epsilon(\mathbf{r}, \omega) = \epsilon_0 + (\sigma c / 2\pi\omega) \int d\omega' g(\mathbf{r}, \omega') / (\omega' - \omega + iT_2^{-1}), \quad (4)$$

where σ is the integral absorption cross section of the ZPL, T_2^{-1} is the homogeneous width of the ZPL, and $g(\mathbf{r}, \omega)$ is the inhomogeneous distribution function, altered in the course of being recorded, that depends on spatial coordinates within the recording medium and is defined so that $g(\mathbf{r}, \omega)d\omega$ is the number of impurity centers in the unit volume of the medium that have their ZPL frequencies in the interval $(\omega; \omega + d\omega)$. The constant nonresonant part of the permittivity will be further taken as $\epsilon_0 = 1$.

Assuming that T_2 is much longer than the overall duration of the pair of object and reference pulses, the permittivity [Eq. (4)] can be written in the form

$$\epsilon(\mathbf{r}, \omega) = 1 - (\sigma c/2\omega)\{ig(\mathbf{r}, \omega) - \hat{H}[g(\mathbf{r}, \omega)]\}, \quad (5)$$

where the symbol \hat{H} means the Hilbert transformation

$$\hat{H}[f(\omega)] \equiv \pi^{-1} \int d\omega' f(\omega')/(\omega' - \omega). \quad (6)$$

If the intensity of object and the reference pulses is moderate so that nonlinear-saturation and power-broadening effects can be overlooked, the PHB bleaching effect resulting from the resonant excitation can be considered proportional to intensity [Eq. (3)]. If we further assume that the plate has a considerable resonant optical density and that the spectrally selective bleaching does not change it significantly, the inhomogeneous distribution function may be expressed as

$$g(\mathbf{r}, \omega) = g_0[1 - m\sigma\eta I(x, y, \omega)\exp[-g_0\sigma\eta]], \quad (7)$$

where g_0 is the density of centers before the PHB, η is a coefficient of PHB efficiency, and m is an integer counting the number of applied sequences of reference and object pulses, i.e., taking into account the accumulation process during the storage.

Using Eqs. (5) and (7), we can write the complex linear transmittance function of the plate as follows:

$$K(x, y, \omega) = \exp[-d(i\omega/c - g_0\sigma/2)] \\ \times \{1 + (\kappa/2)(1 + i\hat{H})[I(x, y, \omega)]\}, \quad (8)$$

where $\kappa = m\sigma\eta$ and the exponential factor describes the attenuation and phase shift of the output signal after crossing the plate of thickness d . One can see that the second term in Eq. (8) contains, as a hologram, exhaustive information about the object pulse. Considering the plate now as a spectral and spatial filter, one can calculate the response of the plate for an arbitrary read-out field. In this case we use a read-out pulse with properties identical to those of the

reference pulse; the resulting optical signal in the output of the hologram is

$$F^{\text{out}}(\mathbf{r}, t) = f_0(\mathbf{r}, t)\exp[i\omega_0(t + x\theta/c)] \\ + f_s(\mathbf{r}, t)\exp[i\omega_0(t + t_R)] \\ + f_s^*(\mathbf{r}, t)\exp[i\omega_0(t - t_R + 2\theta x/c)], \quad (9)$$

where

$$f_0(\mathbf{r}, t) = [1/(R_0\kappa) + R_0/2]\delta(t + \theta x/c) + R_0^{-1}Y(t + \theta x/c) \\ \times \int s(x, y, \tau)s^*(x, y, \tau - t - \theta x/c)d\tau, \\ f_s(\mathbf{r}, t) = Y(t + \theta x/c)s(x, y, t + t_R), \\ f_s^*(\mathbf{r}, t) = Y(t + \theta x/c)s^*(x, y, t_R - t - 2\theta x/c). \quad (10)$$

In writing Eq. (9) we have dropped the constant factor $R_0^2 \kappa \exp[-\sigma g_0 d/2]$ common for all three terms and assumed that $z = +0$; in Eqs. (10), $Y(\tau)$ is the Heaviside unit step function.

Equations (9) and (10) show that, as in the case of ordinary two-dimensional holograms, in the output of the spectral hologram there will appear three different pulsed-light waves (see Fig. 3). The first term, with amplitude f_0 , propagates in the direction \hat{n}_R and describes a transmitted readout pulse with a distorted train determined by the autocorrelation function of the signal. The second term, with amplitude f_s , propagates along the Z axis and represents the playback of the stored object signal as a virtual image of the event recorded. The last term shows the possibility of recalling the conjugated image of the object with reversed time behavior.

In Eqs. (10), the causality function $Y(\tau)$ guarantees that no output signal appears at any point behind the hologram before the readout pulse has arrived. It also cuts off a part of the reproduced object pulse in dependence on the delay of the reference pulse during the process of writing the hologram: If $t_R \leq -|\theta x_{\text{max}}|$ is valid, i.e., the reference pulse came before the object pulse, the latter is completely reproduced but the conjugated object pulse is entirely cut off; if, on the contrary, $t_R \geq |\theta x_{\text{max}}| + t_s$, the nonconjugated object pulse is absent; finally, in an intermediate case of overlapping object and reference pulses $-|\theta x_{\text{max}}| \leq t_R \leq |\theta x_{\text{max}}| + t_s$, both conjugated and nonconjugated object pulses are partially reproduced.

It can also be shown that, if the condition of two-dimensional hologram $\theta^2 d \leq 2\pi c/\omega_0$ is violated so that the synchronism of writing and readout pulses must be considered, the conjugated object pulse can be reproduced only if a counter-propagating probe pulse with the direction $-\hat{n}_R$ is applied.

3. EXPERIMENT

In our experiments we utilized a Rhodamine 6G laser synchronously pumped by an actively mode-locked Ar-ion laser (Spectra-Physics Model 171). The picosecond dye laser provided 82-MHz repetition-rate pulses with 2–3-psec duration ($5\text{--}6\text{-cm}^{-1}$ spectral FWHM) at 100-mW average output power.

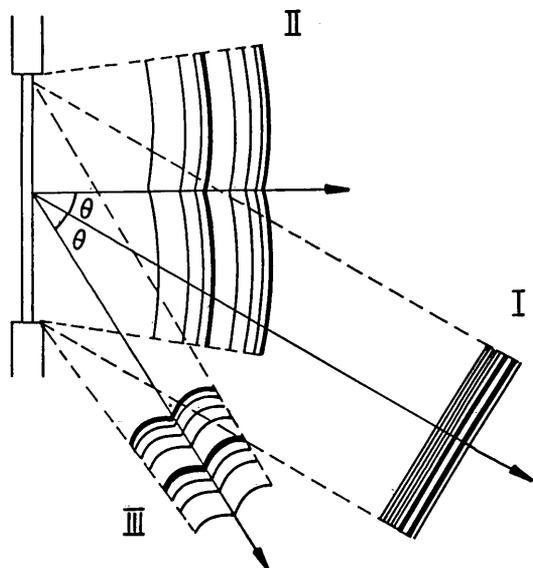


Fig. 3. Diagram showing three different kinds of readout pulses: I, passed-through probe pulse; II, reproduced object pulse; and III, conjugated object pulse. In the case of a three-dimensional hologram, the conjugated object occurs only if the probe pulse is applied in a direction opposite the direction of the reference pulse.

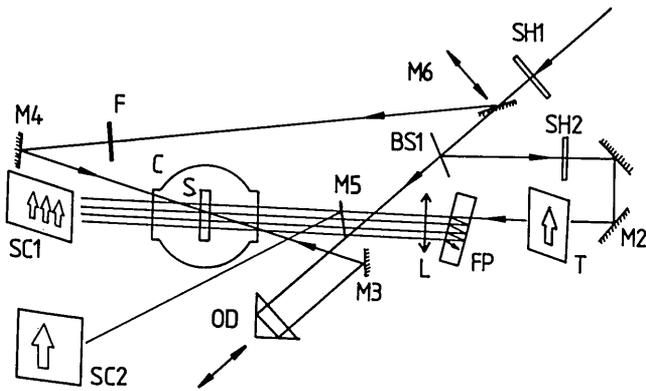


Fig. 4. Schematic diagram of experimental setup for recording spatially modulated picosecond signals. Beam splitter BS1 divides the expanded input picosecond laser beam between reference and object channels; mirrors M1 and M2 direct the object beam through the transparency T, the Fabry-Perot étalon FP, and the lens L with focal length 2 m; optical delay OD is used to place the reference pulse in the 50-psec intervals between object pulses; movable mirrors M5 and M6 have been inserted to obtain conjugated images by passing the probe pulse in the opposite direction; echo signals are visualized on screens SC1 and SC2. C, cryostat; S, sample; F, neutral-density filter; SH1 and SH2, shutters.

Picosecond time analysis was performed by means of a synchroscan streak camera coupled to an optical multichannel analyzer. This system provided 20-psec temporal resolution.²²

To study the spatial properties of time- and space-domain holography, a plane wave-front reference beam was applied at an angle of 6° with respect to the beam of the object pulses (see Fig. 4). After PHB exposures, probe pulses were passed through the hologram either in the direction coinciding with the reference beam or in the opposite direction. The spatial and temporal structure of the picosecond signal was formed by passing the expanded laser beam through a transparency and an étalon. The étalon was tilted by 45° with respect to the beam axis so that the passed-through beam consisted of spatially separated reflections with the contour of the transparency, whereas the 50-psec delay between the reflections was determined by the double pass-through time of the étalon. The wave fronts of the object arrows to be recorded on the hologram were curved by a 2-m focusing lens. The spatial structure of picosecond signals was photographed as the pulses were scattered from sheets of scale paper used as screens to block the output object and conjugated object beams.

As a spectrally selective medium for recording time and space holograms by PHB, we used polystyrene doped with octaethylporphyrin at concentrations of 10^{-4} – $10^{-3}M$. The inhomogeneously broadened 0–0 impurity absorption band was 200 cm^{-1} FWHM and had a maximum at 617 nm. Homogeneous ZPL widths were less than 0.05 cm^{-1} at 1.8 K. Samples were prepared in blocks with a thickness of 0.3–1.0 cm and an optical density of 1–3 and were contained in a liquid-He cryostat with pass-through optical windows.

To record high-contrast spectral holograms, PHB exposures over 0.1 mJ/cm^2 were needed. Depending on the average intensity of the incident light, the PHB effects of 10^9 – 10^{11} identical sequences of writing pulses were accumulated.

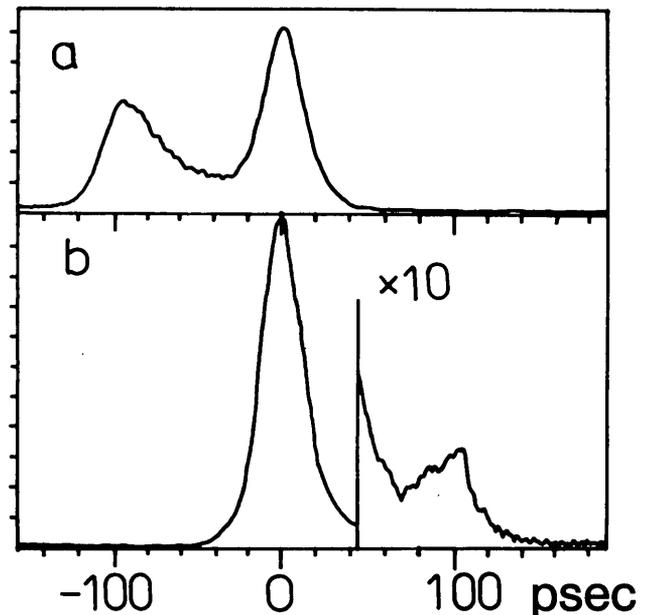


Fig. 5. a, Streak-camera images of the applied PHB sequence comprising an unsymmetrical object pulse and a reference pulse; b, the passed-through probe pulse followed by the time-reversed PASPE signal. The object pulse was formed by a thin étalon from the laser pulse and has a duration of 50 psec. The apparent 20-psec FWHM of both reference and probe pulses is determined by streak-camera time resolution.

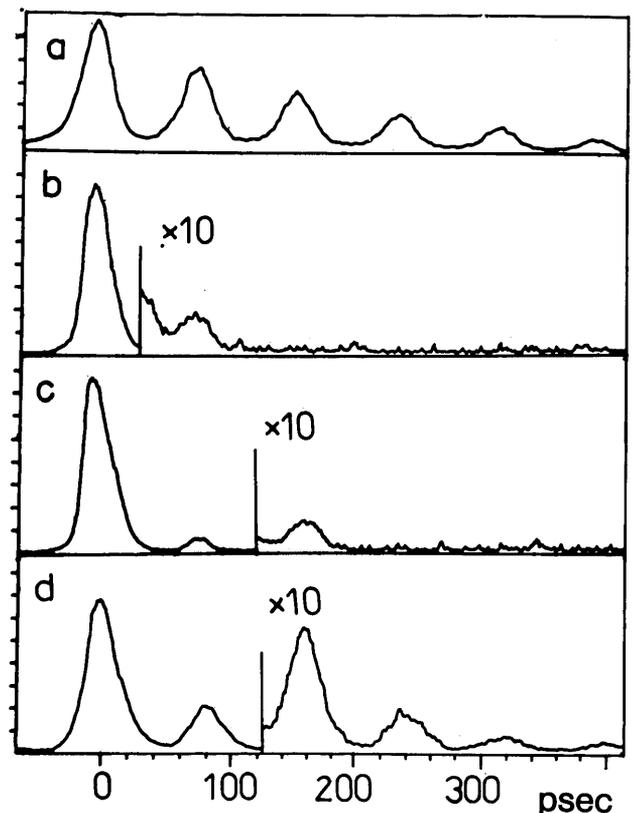


Fig. 6. Streak-camera images of a, the applied PHB sequence and of the resulting PASPE signals after b, 0.5-mJ/cm^2 ; c, 1.5-mJ/cm^2 ; and d, 2.5-mJ/cm^2 PHB exposures. Average intensity of the PHB beam was 0.1 mW/cm^2 .

4. RESULTS AND DISCUSSION

In order to gain some insight into the temporal responses of the PASPE holograms obtained, let us first consider streak-camera experiments in which all beams were collinear and spatially unmodulated.^{12,16} In this geometry ($\theta = 0$; see Section 2) all output pulses propagate collinearly and can be collected onto the entrance slit of the camera in order to measure the entire response of the hologram directly.

Figure 5 represents the result in the case when PHB was performed with the reference pulse delayed with respect to the object pulse by 100 psec. Since $t_R = 100$ psec $>$ $t_S = 50$ psec here, in full accordance with the theory, the time-reversed replica of the object appears in the response after the 100-psec delay (Fig. 5b).

Figure 6 represents the result in the case when PHB was

performed with an exponentially decaying pulse sequence (Fig. 6a). After an optimum exposure (Fig. 6d), as many as four echo pulses delayed by 80, 160, 240, and 320 psec were recorded in the response. As far as the applied pulse sequence can arbitrarily be divided into signal and reference parts in this experiment, in terms of holography the result is nothing else but the restoration of the object by its time-domain fragment.

Let us consider now experiments carried out in accordance with the full scheme of time- and space-domain holography, i.e., experiments with both spatially and temporally modulated picosecond signals and variable delayed reference pulses.

Figure 7(a) gives an idea of the model signal used. It should be recalled that the arrows were 1-mm-thick wave packets flying one after another. To discuss the responses of holo-

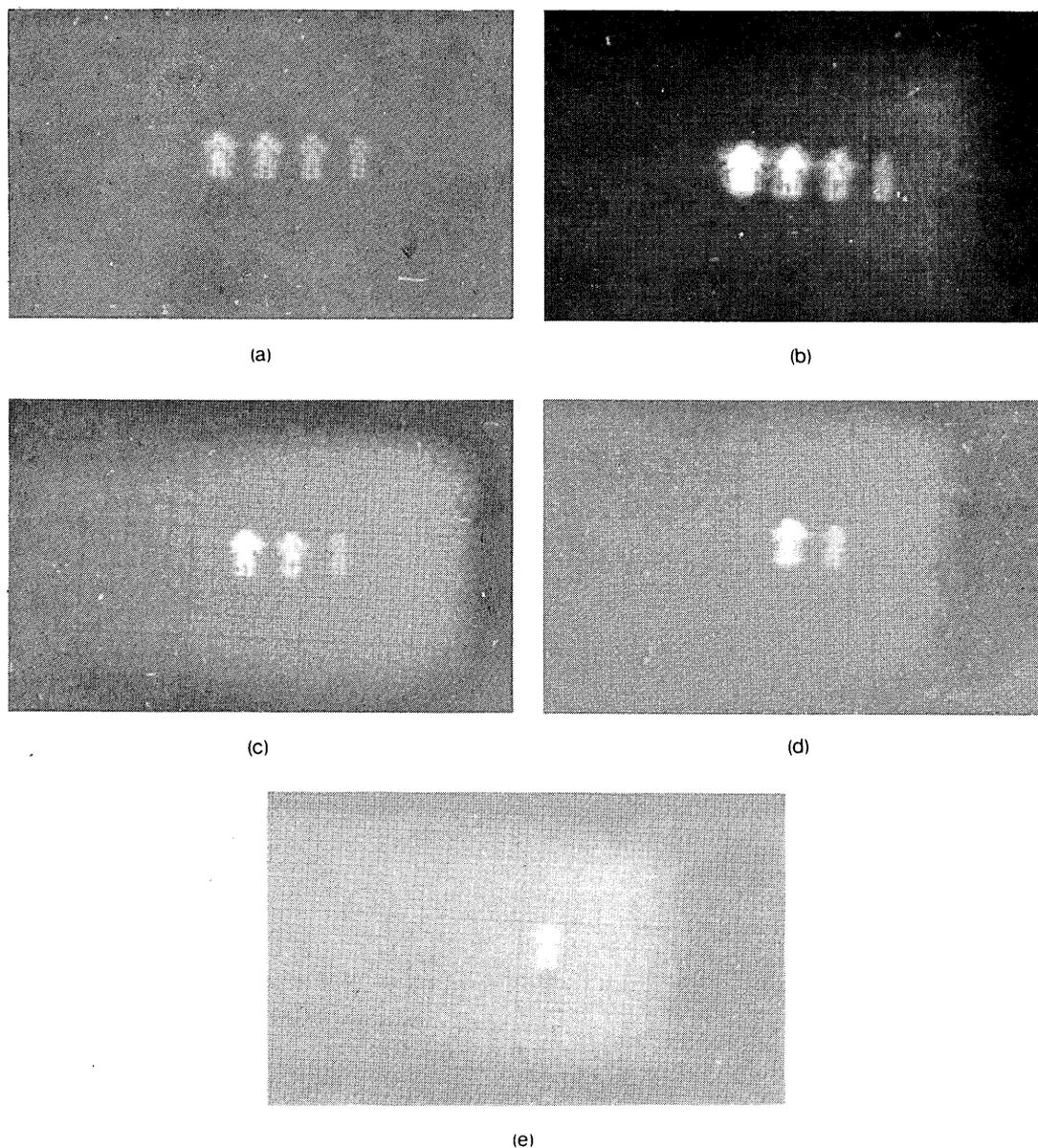
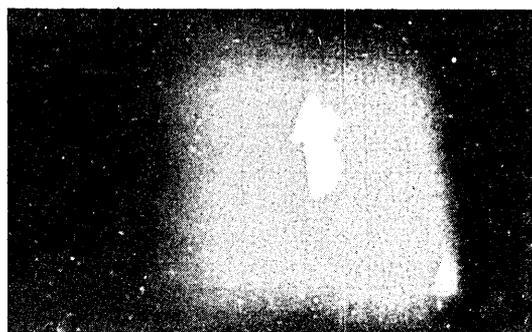
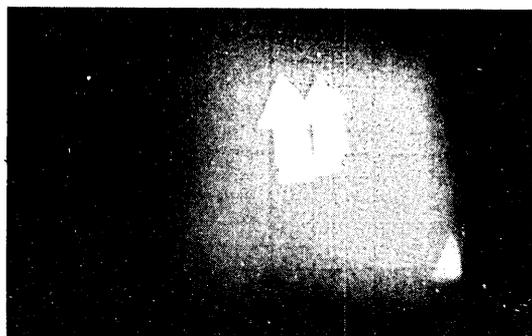


Fig. 7. Photograph images of (a) the pulses used for the PHB and (b) those of echo signals reproduced from the hologram after PHB exposures with the reference pulse applied before the first object pulse, (c) between the first and the second object pulses, (d) between the second and the third object pulses, and (e) between the third and the fourth object pulses.



(a)



(b)

Fig. 8. Image of conjugated echo signals photographed from the screen SC2 with the probe pulse passed through the hologram in the opposite direction. For the PHB the reference pulse has been set between (a) the first and the second object pulses and (b) the second and the third object pulses.

grams obtained at various settings of the reference-pulse delay [Figs. 7(b) and 7(c)], let us also keep in mind that, according to Section 2, the nonconjugated image of the retarded part and the conjugated image of the advanced part of the signal pulse should be reproduced in the case of the forward-propagating probe beam. Figure 7(b) indicates that all temporally separated spatial components of the object were completely restored in the case when the reference pulse had been applied first. Further, in Figs. 7(c)–7(e) one can see that the signal components preceding the reference pulse in the recording procedure are absent in the restored image, showing the temporal selectivity of the hologram. All observed images correspond only to the nonconjugated signal wave. The reason lies in the fact that the holograms were recorded in a thick sample ($\theta^2 d \sim 2\pi c/\omega_0$; see Section 2).

In accordance with the theory, conjugated images appeared when a counterpropagating picosecond probe beam was applied (Fig. 8). In that case, on the contrary, an increase of the reference-pulse delay resulted in the successive appearance of the preceding components of the object that were played back in a reversed temporal sequence. Note that the conjugated images are magnified and that the diffraction patterns present in Fig. 7 are absent here. These effects were also expected from the theory, as the conjugated images are reversed not only temporally but also spatially.

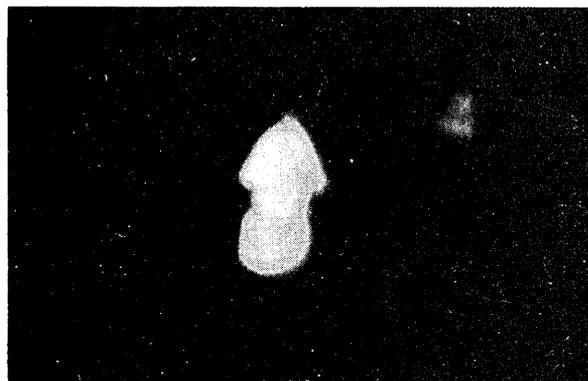
To demonstrate the conjugation effect more explicitly, the experimental setup was modified by replacing the lens and the étalon by a distorter, which scattered the input object-beam contour over an angle of 4° (see Fig. 9). The distorted

object image was recorded on the hologram simultaneously with the delayed reference pulse. Afterward, with the help of a counterpropagating probe beam, a conjugated picosecond PASPE signal was produced. PASPE pulses traveled through the distorter in the opposite direction and reconstructed the initial object image of transparency [Fig. 9(b)]. It should be noted that the temporal distortions of picosecond object pulses were also compensated for.

As the last step of our experiments, we recorded a PASPE hologram of a coin illuminated by a 2-psec-duration pulse. The pulse scattered from the coin modeled an arbitrary spatially and temporally modulated object pulse that passed



(a)



(b)

Fig. 9. (a) Distorted object image photographed at the input of the cryostat, (b) reproduced object image obtained after conjugated echo pulses are passed through the distorter in the opposite direction.



Fig. 10. Image of a coin photographed from the hologram.

through the recording medium at an angle of 10° and some 20 psec later than the plane wave-front reference pulse. The recorded hologram was later illuminated, as in previous experiments, by a beam of 2-psec-duration forward-propagation probe pulses. The reproduced image of the coin was photographed from behind the cryostat (Fig. 10). Although no temporal analysis of the image was performed, on the basis of the theoretical and experimental data presented the reconstruction of the time structure of the object wave can also be stated. It could be partially proved by illuminating the hologram simultaneously by the probe and the object waves. A constructive interference between the object waves scattered from the coin and those reproduced from the hologram was observable, i.e., the reproduced object wave had the same spatial- and temporal-phase structure as the wave scattered from the object.

5. CONCLUSIONS

In this paper, the concept of holographic storage and reproduction of optical signals was generalized for the case of a spectrally highly selective recording medium, which not only can fix the spatial intensity distribution of the incident light but is also able to memorize its intensity spectrum. The proposed method of time- and space-domain holographic recording utilizing well-known spectrally selective media—solid matrices doped with impurity molecules liable to cause PHB—provides the storage and reproduction of time-dependent optical signals with a duration of 10^{-8} – 10^{-13} sec. This presents unique possibilities for ultrahigh-speed data storage and optical signal processing.

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(see overleaf)

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P. Saari was born in Tallinn, Estonian SSR, in 1945. He graduated from the Tartu State University, Tartu, USSR, in 1968. He received the Ph.D. and the Doctor of Sciences degrees from the Institute of Physics, Estonian SSR Academy of Sciences, in 1972 and 1980, respectively. His thesis research introduced hot luminescence into the spectroscopy of resonant secondary light emission of solids. He is now the director of the Institute of Physics, Estonian Academy of Sciences. His research interests include vibronic relaxation processes, ultrafast and nonlinear spectroscopy and, particularly, their application in coherent optics.

include vibronic relaxation processes, ultrafast and nonlinear spectroscopy and, particularly, their application in coherent optics.

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research associate. His research interests include ultrafast and hole-burning spectroscopy, coherent phenomena in photochemically active media, and their application in time-domain holography.

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