Holography of space–time events

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An investigation is made of the process of holographic recording and a reconstruction of spatial and temporal characteristics of an optical field. It is shown theoretically and experimentally that this can be achieved by recording spectral amplitude–phase holograms in spectrally selective light-sensitive materials capable of storing harmonic components of the reference and object waves. A report is given of experiments on low-temperature solid solutions of large organic molecules in which stable narrow dips in the impurity absorption spectrum can be "burnt out" by photochemical reactions. The causality principle operating via the phase part of a hologram results in selection of one of the images: either a virtual image with a direct time dependence or a real image with a reversed time dependence. An experimental check of the properties of space–time holograms yielded a fairly high (for practical applications) diffraction efficiency of $\sim 50\%$.

1. INTRODUCTION

In conventional holography it is the spatial distribution of the field scattered by an object which is recorded in a light-sensitive material. In the hologram retrieval stage the image of the object is reconstructed provided that during exposure a certain steady-state distribution of the field intensity has existed in the interior of a hologram. This requirement follows from the fact that a light-sensitive medium can record only the total optical radiation dose in which the information on the time dependences is lost (averaged out).

A series of investigations[1–5] has shown that holography of time dependences of optical fields is possible in spectrally selective photochromic materials, i.e., in media capable of recording the spectral composition of the absorbed light. Experiments carried out on frozen matrices containing photochemically active impurity molecules, which have been used to record picosecond light signals and to reconstruct them with a high ($\sim 50\%$) efficiency, have demonstrated that such materials are promising for practical applications.

We shall present a theory of space–time holography as a method for recording an object scene and then reconstructing the image while retaining its time dependence, and we shall report the results of an experimental check of these theoretical conclusions.

When use is made of low-temperature impurity systems with an inhomogeneous distribution of transition frequencies that can be used to burn out photochemically a dip in the absorption spectrum,[6–8] the maximum duration of scenes which can be recorded in this way can reach hundreds of nanoseconds and the time resolution can be tens of femtoseconds. This follows from the basic idea of the method[2]: a mutual spectrum of the object and reference pulses at a given point is recorded in each element of the light-sensitive medium; subsequently, each element acts as a spectral filter in respect of the reconstructing pulse, which is transformed into a replica of the object pulse. The mutual spectrum or "spectral hologram" is recorded by destruction of some of the absorbing centers within the inhomogeneous width ($\sim 10^2$ cm^{-1}) of the absorption and of a frozen matrix with an accuracy to within the homogeneous or zero-phonon width ($\sim 10^{-4}$ cm^{-1}) of the absorption line of the centers. The reciprocals of these widths determine the time limits just given.

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Recovery of the object pulse from the reconstructing pulse can be interpreted in the same way as the formation of the signal representing the decay of free polarization in an element of a light-sensitive medium with a special distribution of the dipole frequencies established in the recording stage. In this time treatment it is clear that the phase relaxation time T_2 of a resonance transition of molecular dipoles (which is the reciprocal of the line width) limits the permitted duration of the reconstructing pulse. On the other hand, the phase memory of the medium makes it possible to separate the object and reference pulses in time during the recording state without losing the interference between them, which—as demonstrated below—together with the causality principle determines the essential features of the holograms under discussion. It also follows from the time treatment that the processes of hologram recording and reconstruction in such media are based on a physical phenomenon similar to the stimulated photon echo in conventional resonant media. Therefore, the concept of photochemically accumulated stimulated optical echo (PASOE) was introduced in Ref. 2.

The potentialities of the optical echo in dynamic holography[1)] have been analyzed fundamentally in several treatments (see, for example, Refs. 9–11) and, in particular, it has been shown that there are correlations between the time profiles of the stimulated echo and pump pulses.[12] A theoretical analysis[13] published during the planning of our first experiments demonstrated how the space–time behavior of a laser pulse can be recorded in a spectrally inhomogeneous resonant medium and reconstructed as a stimulated echo signal.

However, in practice the use of the optical echo in holography is limited by the circumstance that a hologram of this kind has a very short lifetime (limited to the lifetime of the excited state of molecules), requires high-power pump

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pulses (1 MW in the picosecond range), and has a low diffraction efficiency. In contrast, the holograms based on the PASOE concept are real in the sense that they can be subjected to repeated reconstruction after hours and possibly after years (depending on the stability of the products of photochemical transformations of the absorption centers); moreover, a high contrast is readily achieved using relatively weak pulses by repeating the recording cycles (for details see Refs. 2–5).

It is interesting to note that, in principle, such space–time holography is attainable also in conventional photographic materials. A method has been proposed theoretically[14,15] in which the spectral composition of an object pulse is recorded using a special reference wave characterized by a carrier frequency which varies with depth in a hologram.

In the next section we shall give a theory of space–time holography. In describing the holographic process we shall assume the use of the PASOE concept. This allows us to consider only the approximation of weak fields, so that we can ignore details of the physical effect underlying the process and use formulas which demonstrate clearly the holographic aspect of the problem. This aspect is of general nature and, therefore, the main conclusions of the theory apply also to the case of a conventional stimulated echo. In the third section we shall give data on a light-sensitive material and experimental apparatus. In the fourth section we shall analyze the experimental results on the basis of the theory in Sec. 2.

2. THEORETICAL DESCRIPTION OF THE HOLOGRAPHIC PROCESS

A theoretical description of the interaction of optical fields with a spectrally selective photochromic medium is in linearity of such a medium (its permittivity varies under the influence of light), the hologram being formed reacts on the fields which are being recorded and this may be particularly important in the case of volume holograms. Moreover, a rigorous calculation of a reconstructed wave requires a solution of the Maxwell equations for a field in a spatially and spectrally inhomogeneous medium, which is unattainable in the general case. Therefore, we shall consider the approximation of a low contrast in the recording stage and we shall treat the propagation of a field across a hologram in the Kirchhoff approximation, as is usually done in holography and coherent optics.

We shall consider a highly selective photochromic element in the form of a plate of thickness d which is located in the $z = 0$ plane and has the transverse dimensions $2x_{\max} \times 2y_{\max}$. We shall assume that an optical pulse is traveling from an object in the direction of the plate (Fig. 1) and the field intensity in this pulse can be described by a scalar signal $S(\mathbf{r}, t)$ in the plane of the plate is

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

The complex amplitude s depends weakly (compared with the exponential function) on its arguments and its time dependence may reflect both the amplitude and frequency (relative to the average carrier frequency ω_0) modulation of the object (source) and its motion. We shall assume that the small bandwidth condition is satisfied; $\Delta\omega_s \ll \omega_0$, where $\Delta\omega_s$ is the width of the signal spectrum. Representation of the signal in the form of Eq. (1) implies that the nature of the waves is practically unaffected as a result of traversal of a distance of the order of d. We shall consider a specific time scale such that at the moment $t = 0$ the front of the pulse reaches the plate and at the moment t_s it leaves the plate.

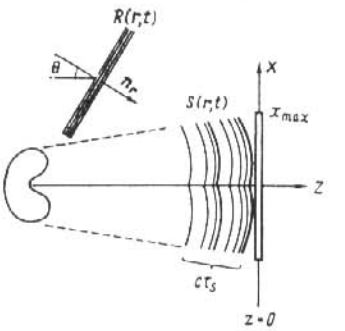

FIG. 1. Recording of a space–time hologram. A signal pulse $S(\mathbf{r}, t)$ emitted by an object scene in the form of a train of waves is shown at the moment $t = 0$; for $t \leqslant 0$ or $t \geqslant t_s$ the value of S in the $z = 0$ plane is practically zero; a reference pulse $R(\mathbf{r}, t)$ is shown after a delay $t_R > t_s$.

We shall select a reference signal $R(\mathbf{r}, t)$ in the form of a plane-wave pulse incident on the plate at an angle θ (Fig. 1):

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

where $\mathbf{n}_R = (-\sin\theta, 0, \cos\theta)$ is a unit vector in the direction of propagation of the wave [for the sake of simplicity, we shall assume that $\mathbf{n}_R = (-\theta, 0, 1)$; t_R is the moment at which the peak of a pulse of amplitude R_0 crosses the origin of the coordinate system. The assumption of δ-like behavior reduces to the requirement that the width of the spectrum of the reference wave $\Delta\omega_R$ is greater than the width of the object spectrum and the spectrum itself is constant in the region of the object spectrum.

We shall assume that the photochromic medium has the permittivity

$$\varepsilon(\mathbf{r}, \omega) = \varepsilon_0 + (\sigma c/2\pi\omega)\int[g(\mathbf{r}, \Omega)/(\Omega - \omega + (i/T_2))]d\Omega, \quad (3)$$

where the second term is due to the fact that the medium contains absorbing centers with an inhomogeneous distribution of the transition frequencies near ω_0 and it corresponds to an absorption band formed from zero-phonon lines of width T_2^{-1} of impurity molecules with an integral absorption cross section σ; the product $g(\mathbf{r}, \Omega)d\Omega$ gives the number of centers per unit volume (before or after recording of the hologram), which have the zero-phonon line frequency at the interval between Ω and $\Omega + d\Omega$ (we shall ignore the presence of phonon wings of the lines); ε_0 is the part of the permittivity due to other internal vibronic transitions and due to the medium (for the sake of simplicity, we shall assume that $\varepsilon_0 = 1$). We shall also postulate that the characteristic width $\Delta\omega_g$ of the distribution g is greater than the widths $\Delta\omega_s$ and $\Delta\omega_R$, and that the phase relaxation time T_2 of the resonance transition is much greater than the total duration of the "object + reference pulse" event. Then, Eq. (3) can be replaced with

$$\varepsilon(\mathbf{r}, \omega) = 1 - (\sigma c/2\omega)[ig(\mathbf{r}, \omega) - \hat{H}\{g(\mathbf{r}, \omega)\}], \quad (3')$$

where $\hat{H}\{g(\mathbf{r}, \omega)\}$ is the Hilbert transform of the function $g(\mathbf{r}, \omega)$. the expression $\varepsilon(\mathbf{r}, \omega)$ given by Eq. (3') shows that

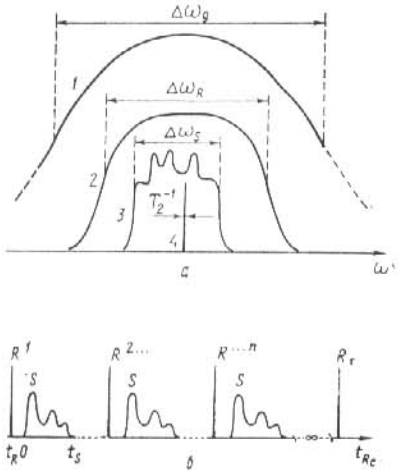

FIG. 2. a) Relationship between the widths of the spectra of an inhomogeneous absorption band (1), a reference pulse (2), an object pulse (3), and a homogeneous zero-phonon line of an absorption center (4). b) Time sequence of application of pulses to a hologram. Recording is carried out by one or n pairs of pulses R and S, and the intervals between the pairs are greater than the phase relaxation time $T_2 \sim 10$ nsec; after an arbitrary time interval from the time of recording (which may be of the order of hours and maybe months) a reconstructing pulse is applied.

spectral modulation of the absorption in a medium is always accompanied (because of the causality principle) by modulation of the refractive index; in our case an amplitude spectral hologram is accompanied by a phase hologram.

The reported relationships governing the frequency–time characteristics of the model are illustrated in Fig. 2a. In the case of low-temperature matrices containing organic dye impurities these relationships apply in the case of subnanosecond signal pulses.

Further analysis of the holographic process reduces[16] to a calculation of the total field (I) acting during the recording stage, of changes in the permittivity of the medium as a result of exposure (II), and of the field at the exit from a hologram in the reconstruction stage (III). We shall begin by considering differences from the conventional holography of stationary spatial scenes. Firstly, during the recording stage it is not necessary (and we shall show later that it is even undesirable) that the object and reference fields should act simultaenously on a hologram. Secondly, the requirement of coherence of the object and reference fields has to be satisfied only when repeated exposures are used in the recording stage, i.e., if the object and reference pulses act on a hologram repeatedly separated by time intervals exceeding T_2 (Fig. 2b). Thirdly, we shall expand the fields into monochromatic components since the photosensitivity of the medium is spectrally selective.

Calculations (I) and (II) yield the following expression for the density of the centers after recording, which has to be substituted in Eq. (3′):

$$g(\mathbf{r}, \omega) = g_0 |1 - \sigma n \eta I(x, y, \omega) \exp(-g_0 \sigma z)|, \qquad (4)$$

where n is the number of pairs of the object and reference pulses used in the exposure stage; η is the probability of a photochemical transformation of an optically excited center; g_0 is the density of the centers before burning out a dip in the absorption spectrum of the hologram (it is assumed that the plate is opticlly thick, i.e., that $\sigma g_0 d \gg 1$);

$$I(x, y, \omega) = R_0^2 + |\bar{s}(x, y, \omega - \omega_0)|^2 + R_0 \bar{s}(x, y, \omega - \omega_0) \exp|-i\omega(x\theta - t_R)| + R_0 \bar{s}^*(x, y, \omega - \omega_0) \exp i\omega(x\theta - t_R) \qquad (5)$$

is the spectral intensity of the light performing the burning-out operation; a bar above a symbol denotes a Fourier transform (for the sake of simplicity, we shall assume that $c = 1$).

The expresison (4) is written down in the linear approximation, i.e., it is assumed that the changes in g during the recording stage are fairly small: $\sigma n \eta \ll 1$. The exponential functions in Eq. (5) show that the changes in $\varepsilon(\mathbf{r}, \omega)$ in the medium during the recording stage result in the formation of a spatial amplitude–phase grating at each freqeuncy, although in the integral sense (i.e., for white light) there may be no such gratings. Envelopes of the spatial–spectral gratings carry full information on the object field. Therefore, if we use Eqs. (4) and (5) to change from Eq. (3′) to a complex transmission coefficient of a hologram plate, we find that such a plate can reconstruct the time Fourier components of the object field when illuminated with a plane monochromatic reconstructing wave (for example, by a wave with $\mathbf{n} = \mathbf{n}_R$ and with a frequency ω_0):

$$\bar{E}^{out}_{\omega_0} = e^{-\frac{1}{2}\sigma g_0 d}\left[1 - \frac{1}{2}\varkappa(1 + i\hat{H})\{R_0^2 + |\bar{s}(x, y, \Omega)|^2\}\right] \times \exp i\omega_0(t + \theta x - z)| + e^{-\frac{1}{2}\sigma g_0 d}\left[\frac{1}{2}\varkappa R_0(1 - i\hat{H})\{\bar{s}(x, y, \Omega) \exp - i\Omega(\theta x - t_R)\}\right] \times \exp i\omega_0(t + t_R - z) + e^{-\frac{1}{2}\sigma g_0 d}\left[\frac{1}{2}\varkappa R_0(1 + i\hat{H})\{\bar{s}^*(x, y, \omega) \exp i\Omega(\theta x - t_R)\}\right] \times \exp i\omega_0(t - t_R - 2\theta x - z), \quad \Omega \equiv \omega - \omega_0 = 0; \ \varkappa = \sigma n \eta; \ z = +0. \qquad (6)$$

We can therefore see that in the case of a conventional plane hologram, three waves appear at the output and these correspond to the terms in Eq. (6). The first term describes a distorted reconstructing wave traveling in the direction $\mathbf{n}_R$; the second travels in the Z direction and gives a virtual image of the object scene which in this case appears as if observed through a narrow-band filter tuned to the frequency ω_0; the third term describes a wave in the direction $(-2\theta, 0, 1)$ and gives a real image of the scene.

If a set of plane monochromatic waves with frequencies distributed uniformly over a band $I(x, y, \Omega)$ is incident on the plate from the direction $\mathbf{n}_R$, then each wave is transformed at the output in accordance with Eq. (6) and the resultant wave is found by integration over all the frequencies Ω. Consequently, when a reconstructing pulse has characteristics similar to the refrence pulse which crosses the origin of the coordinate system at a time $t = 0$, the field $E^{out}(x, y, t)$ emerging in the XY plane is found by integration of Eq. (6) using a phase factor $\exp[i\Omega(t + \theta x)]$, i.e., applying the inverse Fourier transformation. The final result is that reconstruction of the object field by a pulse is described by the expression

$$E^{out}(x, y, t) = e^{-\frac{1}{2}\alpha_0 d}\left\{\left[\left(1 + \frac{1}{2}\varkappa R_0^2\right)\delta(t - \theta x) + \varkappa Y(t + \theta x) \times \int s(x, y, \tau) s^*(x, y, t + \theta x - \tau)\, d\tau\right] \times \exp i\omega_0 (t + \theta x) + R_0 \varkappa Y(t + \theta x) \times s(x, y, t + t_R) \exp i\omega_0 (t + t_R) + R_0 \varkappa Y(t + \theta x) s^*(x, y, -t + t_R - 2\theta x) \times \exp i\omega_0 (t - t_R + 2\theta x)\right\}. \quad (7)$$

We an see that the field emerging as a pulse response of a hologram obeys the causality principle: unit Heaviside steps Y, which (for $z \geqslant 0$ and exact recording) have the argument $ct = x \sin\theta - z\cos\theta$, ensure the absence of the field at any point beyond the hologram up to the moment when this point is crossed by the reconstructing pulse or by the plane of the front of this pulse (at the points where the pulse itself does not arrive because of its limited aperture). The expression (7) describes three waves which become separated at some distance (Fig. 3). The first term with the amplitude factor in the brackets describes the transmitted reconstructing pulse with a "tail" governed by the signal autocorrelation function.

If $t_R \leqslant -|\theta x_{max}|$, where x_{max} is the coordinate of the hologram edge (in the recording stage a reference pulse reaches the plate before an object pulse), we find that the function $Y(t + \theta x)$ in the second term may be dropped. Consequently, the second term in Eq. (7) then describes a fully reconstructed signal field [compare with Eq. (1)] emerging from the hologram after a delay t_R relative to the reconstructing pulse. An observer along the Z axis receives this field as a virtual image of the recorded scene. It is important to stress that a complete reconstruction of the object scene is achieved together with its time dependence, including a possible frequency modulation of the radiation emitted by the sources. If the condition $t_R \leqslant -|\theta x_{max}|$ is not satisfied, then an object pulse forms because of the function Y. For example, if the object signal consists of two pulses, the second of which reaches the plate after the reference pulse, then the second pulse is absent from the field reconstructed along the Z direction.

Finally, we can readily demonstrate[16] that the third term in Eq. (7) describes the formation of a real image of the object scene in the direction $(-\sin\theta, 0, \cos 2\theta)$ with the reversed time dependence (Fig. 3). If $t_R \geqslant |\theta x_{max}| + t_s$, i.e., when a reference pulse is applied after an object pulse crosses the plate, reconstruction is possible without truncation of the front of the reconstructing pulse. Conversely, when the condition for untruncated reconstruction of a virtual image $t_R \leqslant -|\theta x_{max}|$ is satisfied, the term in question vanishes, i.e., there is no real image.

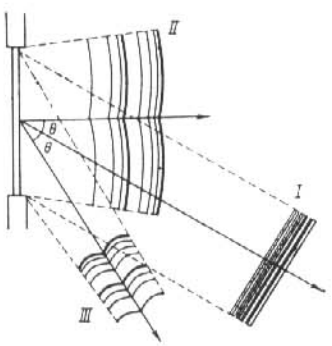

FIG. 3. Pulses emerging from a hologram in the reconstruction stage; the Roman numbers correspond to the terms in Eq. (7).

It therefore follows that for one direction of a reconstructing pulse, we can obtain either a real image or a virtual reconstructed image (history of an event) depending on the order in which the reference and object pulses are applied during the recording stage.

It remains to consider the effects of an increase in the thickness of the hologram under consideration. An analysis[16] shows that in this case we can expect a complete reconstruction of the object scene including its time dependence only for two directions of the reconstructing pulse. Moreover, the formation of images is subject not only to the restrictions imposed by the causality principle, but also to certain additional limitations: if a reconstructing pulse is directed along the direction of the reference pulse, only a virtual image is obtained; a real image (with a reversed time dependence) can be reconstucted if we direct a reconstrucitng pulse antiparallel to the reference pulse and in this case we do not obtain a virtual image. We may conclude by noting that an increase in the hologram thickness results in limitations due to wave phase matching known to apply to volume holograms (see,for example, Ref. 17).

The effects of a nonlinearity of the medium are more serious because in the case of a hologram of considerable thickness the above assumption of a low contrast (a small change in the distribution of the centers) is not satisifed throughout the hologram. It follows from general considerations that time replicas of the reconstructed signal pulses may appear. A quantitative analysis of these effects is possible in a nonlinear variant of the above theory.

3. EXPERIMENTAL METHOD

Our spectrally selective light-sensitive media were solid polymerized solutions of H_2-tetra-tert-butylporphyrazine and octaethyl porphin in styrene, cooled to liquid helium temperatures. The inhomogeneous width of the active absorption bands was 500 and 200 cm^{-1}, respectively, and the homogeneous width of purely electronic lines was ~ 0.1 and ~ 0.05 cm^{-1}, respectively. Samples were plates 3–10 mm thick and their optical density was $D = 1$–3.

In these systems a narrow stable dip was formed in the inhomogeneously broadened impurity absorption spectrum because of photoinduced changes caused by resonant monochromatic excitation and the width of this dip was governed by the homogeneous zero-phonon line width (dip burning effect[6,7]). Photochemical holograms were burnt and probed by a picosecond laser utilizing the rhodamine 6G dye, which was pumped synchronously by an argon laser with active mode locking (Spectra Physics, models 375 and 171). The pulse repetition frequency (duration 2–3 psec, width of the spectrum, 5 cm^{-1}) was 82 MHz. Spectral measurements and

holography experiments on a stationary field were carried out using the same rhodamine dye laser but employed in the cw regime with an additional scanning etalon in the resonator (emission line width 0.075 cm^{-1}) or with a two-plate Lyot filter (1–1.5 cm^{-1}).[18] The transmission spectra were recorded employing a two-chanel photon counting system.

The time response of the holograms was investigated employing a system with image conversion and synchronous scanning,[18] characterized by a time resolution of ~20 psec or by frequency up-conversion of sections of an echo signal in an $LiIO_3$ crystal using a noncollinear system with a picosecond gate pulse characterized by a variable delay. The dependence of the up-conversion signal on the delay was determined with a phase-sensitive detector. The average intensities of the laser radiation in the burning procedure (recording) and in the probing (image reconstruction) stage were 0.1 MW/cm^2 and 0.1 $\mu W/cm^2$, respectively.

Reference and object pulses delayed relative to one another were generated employing Fabry–Perot and Michelson interferometers.

Two geometries were used in these experiments: 1) with collinear directions of all the rays for investigating only the spectral–time dependences; 2) with the object and reference rays separated by an angle of $\theta \leqslant 1°$ in studies of spatial dependences. The spatial formation of the object beam was ensured by placing a mirror with a focal length 2 m in one arm of the Michelson interferometer; beyond a cryostat in which a sample was placed, it was found that this arrangement produced on a distant screen an image of a point together with an unfocused spot from the plane reference wave. The angle between the reference and object waves was selected by misalignment of the interferometer.

4. DISCUSSION OF RESULTS

In this section we shall consider and interpret, on the basis of the above theory, the results of experiments described in Refs. 1–3 and 5, and also of the experiments carried out in order to test the most intriguing predictions made theoretically.

It follows from Eq. (5), which describes $\varepsilon(\mathbf{r}, \omega)$ of the medium after recording, that an interference structure in the transmission spectrum of a hologram becomes stronger on increase in the interval t_R between the pulses compared with the duration of the object pulse (they should be compared with the condition for simultaneous action of light beams under usual interference conditions). Therefore, the burning of a spectral grating was detected in the following experiment.[1,2] A sample was subjected to trains of pulses of ~3 psec duration separated by intervals of 80 psec. In view of the collinear geometry and the similarity of the pulses in a train, there was no need to select the reference pulse and $I(\omega)$ was given by the spectrum of the train identical with the spectrum of a single pulse but with a factor oscillating at a frequency $\Delta\omega = 2\pi/t_R$. The results of a determination of the transmission spectrum of the hologram sample confirmed this conclusion (Fig. 4).

In accordance with the basic idea behind time holography, the response of such a spectral hologram to a reconstructing pulse should restore the train just described. Measurements carried out with direct time resolution of the light transmitted by the hologram indicated that this indeed took place[2–5] (Fig. 5). This experiment differed from the system described in Sec. 2 because of the absence of an explicit reference pulse, the spectrum of which was wider than that of the rest of the signal. Therefore, this experiment was essentially realization in time of a generalized holographic method of reconstructing the whole object from a fragment.

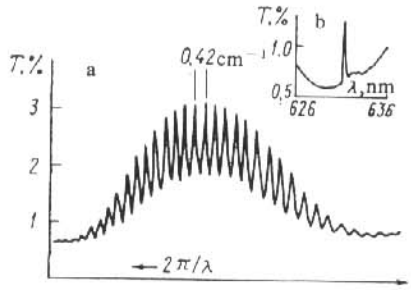

FIG. 4. Transmission spectrum of H_2-tetra-tert-bytylporphyrazine in polystyrene after exposure to trains of picosecond pulses with a total energy density 20 mJ/cm^2. the inset shows the general appearance of a dip measured with a resolution of 0.1 nm.

The ability of a hologram to reverse the time dependence of a given event in accordance with the last term in Eq. (7) was checked in an experiment[4] carried out employing an explicit reference pulse. A photochromic medium was subjected first to an object pulse of asymmetric shape and then to a delayed short reference pulse. In the reconstruction stage (Fig. 6) the response pulse which appeared after the transmitted reconstructing pulse did indeed have a reversed time profile.

The most interesting theoretical prediction was the ability of a hologram to distinguish the "future" from the "past" in the literal sense: a virtual image of the object events occurring after the application of a reference pulse is emitted in one direction, whereas a real image of the object events before the reference pulse is emitted in another direction. In other words, depending on the order of application of the

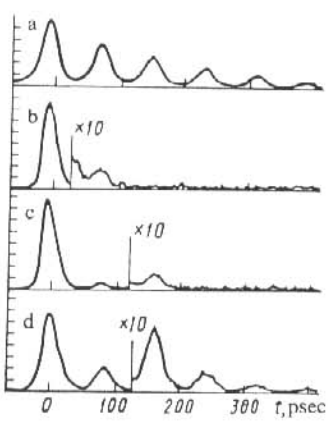

FIG. 5. Dependence of the response of octaethyl porphin in styrene on the exposure during photochemical burning by trains of picosecond pulses at intervals of 80 psec (a), compared with the responses after exposure to 0.5 (b), 1.5 (c), and 2.5 mJ/cm^2 (d).

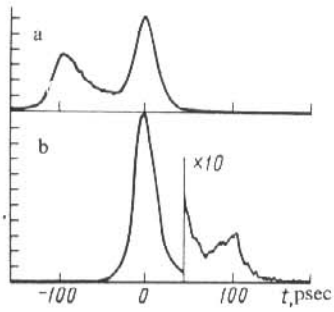

FIG. 6. Asymmetric signal and delayed reference pulses (a), and the reconstructing and reconstructed pulses characterized by a reversed time dependence (b).

object and reference pulses during the reconstruction stage, one of the images is not obtained and this is true of thin and thick holograms. This prediction was confirmed as follows. A hologram was subjected to a picosecond reference pulse with a plane wavefront and to an object picosecond pulse inclined at a small angle to the reference pulse and characterized by a converging wavefront, so that it became focused behind the hologram. If in the course of the hologram recording stage the first to reach the medium was the reference pulse, then reconstruction by a pulse in the direction $\mathbf{n}_R$ produced a converging wave in the direction $\mathbf{n}_s$ and this wave was focused at the point of location of the signal wave focus, i.e., the recorded event was reconstructed. The scattered pulse was delayed relative to the reconstructing pulse by the same amount as the delay during the recording stage.

The identity of the reconstructing and object pulses during the reconstruction stage was checked by the application also of the object pulse. Then, interference between the delayed object and hologram-scattered pulses was observed clearly beyond the hologram and it was manifested by quenching and enhancement of the focused spot as the phase of the signal pulse varied within one wavelength. Such interference confirmed the direct time measurements of the delay of the scattered pulse and also the reconstruction of the phase structure of the latter. If the first to reach the object was the object pulse, then on reconstruction in the direction $\mathbf{n}_s$ there was no image in the direction $\mathbf{n}_R$, in agreement with Eq. (7). However, in the direction $2\mathbf{n}_z - \mathbf{n}_s$ allowed by the causality principle [in Eq. (7) this corresponds to the direction $(-2\theta, 0, 1)$], we did not observe an image because this was forbidden by the angular phase-matching condition applicable to volume holograms. It should be pointed out that in this case a plane scattered wave appeared in the direction $\mathbf{n}_R$ when reconstruction was performed by a converging (initially signal) wave behind the hologram. This experiment essentially involved signal recognition.

The diffraction efficiency of the holograms was found to be very high and in some cases it reached $\sim 50\%$ of the intensity of the transmitted undiffracted wave. Switching of the direction of propagation of the wave scattered in the hologram or forbiddenness of reconstruction of an image were due to the causal behavior of the polarization of an element of the medium. In the spectral pattern this corresponded to one-sided diffraction due to the always-valid dispersion relationships between the absorption coefficient and the refractive index of the medium [see Eq. (3′)]. Clearly,the conclusions that follow from the causality principle do not just apply to the holographic process described in Sec. 2. For example, one-sided diffraction should also occur in the case of dynamic gratings in resonant media, for which a theory is developed in Ref. 10 but without an allowance for the spectral modulation of the refractive index. Moreover, a grating burnt in a spectrally selective medium by cw laser radiation should also be one-sided. This was confirmed experimentally by recording holograms with a cw dye laser characterized by an emission spectrum $\sim 1.5\ \text{cm}^{-1}$ wide. The reference and object beams were applied to a sample separated by a path difference greater than the coherence length of the laser radiation, so that there was no interference pattern in the region of intersection between the beams. When reconstruction took place, the familiar pattern was observed in the $\mathbf{n}_R$ direction: when the reference wave arrived after delay, then the wave converging in the $\mathbf{n}_s$ direction was reconstructed, whereas in the case when the object wave arrived first, there was no image in the $\mathbf{n}_s$ direction. This follows mathematically from the properties of the operator $1 + i\hat{H}$ in Eq. (6), which can assume the values 0 to 2 provided the function on which it acts can be represented as a product of a rapidly oscillating exponential function and a slower factor. This slower factor can be the power spectrum of cw laser radiation, because it depends on the frequency more slowly than an exponential function of the shift if the absolute delay of one beam relative to the other exceeds the correlation time of laser radiation.

It should also be pointed out that the appearance of the factor 2 in Eq. (7) in front of the term describing diffraction in the allowed direction (due to the phase part of the hologram) is an indication of an increase in the diffraction efficiency by a factor of 4 compared with pure amplitude holograms characterized by the same transmission and the same modulation depth.

These predictions were confirmed experimentally and high diffraction efficiencies were observed.

5. CONCLUSIONS

The theory of the conventional holographic process is generalized to the case of photosensitive materials which are capable of recording not only the spatial distibution of the field intensity, but also its spectral composition. The proposed and experimentally realized method of holographic recording in familiar media with photochemically active impurity absorption centers makes it possible to reconstruct the image of an object with an efficiency sufficient for practical applications and this can be done over a wide range of time intervals from 10^{-8}–10^{-13} sec. Essentially, space–time holography solves the problem of the true reconstruction of the events if we bear in mind that the line method of recording individual frames does not provide a direct reconstruction of motion in the image of a scene.

Moroever, the results show that the properties of such holograms that follow from the causality principle are of general nature and apply also to holography of stationary

fields in spectrally selective media, to dynamic holography in resonant media, and to other processes of the interaction between light and matter in which spectral modulation of the susceptibilities takes place.

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[1)]To avoid possible terminological confusion, we must point out that dynamic holography is defined as that taking place in media in which the formation of a hologram and the transformation of waves in the hologram occur in real time.[11] In the limiting case a dynamic hologram follows changes in the spatial distribution of the instantaneous intensity of light in an interference pattern; by definition, a time hologram records (and retains for an interval considerably greater than the duration of the event in question) the time dependence of such a distribution.

[1]A. K. Rebane, R. K. Kaarli, and P. M. Saari, Opt. Spektrosk. **55**, 405 (1983) [Opt. Spectrosc. (USSR) **55**, 238 (1983)].
[2]A. K. Rebane, R. K. Kaarli, and P. M. Saari, Pis'ma Zh. Eksp. Teor. Fiz. **38**, 320 (1983) [JETP Lett. **38**, 383 (1983)].
[3]A. Rebane, R. Kaarli, P. Saari, A. Aniyalg (Anijalg), and K. Timpmann, Opt. Commun. **47**, 173 (1983).
[4]A. Rebane and R. Kaarli, Chem. Phys. Lett. **101**, 317 (1983).
[5]A. K. Rebane and R. K. Kaarli, Izv. Akad. Nauk SSSR Ser. Fiz. **48**, 545 (1984).
[6]A. A. Gorokhovskiĭ, R. K. Kaarli, and L. A. Rebane, Pis'ma Zh. Eksp. Teor. Fiz. **20**, 474 (1974) [JETP Lett. **20**, 216 (1974)].
[7]B. M. Kharlamov, R. I. Personov, and L. A. Bykovskaya, Opt. Commun. **12**, 191 (1974).
[8]K. K. Rebane, Elementary Theory of the Vibrational Structure of Impurity Centers in Crystals [in Russian], Nauka, Moscow (1968).
[9]E. I. Shtyrkov and V. V. Samartsev, in: Electromagnetic Superradiance [in Russian], Kazan Branch of the Academy of Sciences of the USSR (1975).
[10]E. I. Shtyrkov and V. V. Samartsev, Phys. Status Solidi **45**, 647 (1978).
[11]Yu. N. Denisyuk, in: Problems in Optical Holography (ed. by Yu. N. Denisyuk) [in Russian], Nauka, Leningrad (1982), p. 7.
[12]V. A. Zuĭkov, V. V. Samartsev, and R. G. Usmanov, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 293 (1980) [JETP Lett. **32**, 270 (1980)].
[13]T. W. Mossberg, Opt. Lett. **7**, 77 (1982).
[14]V. A. Zubov and T. I. Kuznetsova, Kvantovaya Elektron. (Moscow) No. 1(13), 135 (1973) [Sov. J. Quantum Electron. **3**, 85(1973)].
[15]T. I. Kuznetsova, Tr. Fiz. Inst. Akad. Nauk SSSR **84**, 62 (1975) [see Chap. IV, p. 136].
[16]P. M. Saari and A. K. Rebane, Izv. Akad. Nauk Est. SSR Fiz. Mat. **33**, 322 (1984).
[17]L. M. Soroko, Holography and Coherent Optics, Plenum Press, New York (1980).
[18]A. O. Aniyalg, P. M. Saari, T. B. Tamm, K. É. Timpmann, and A. M. Freĭberg, Kvantovaya Elektron. (Moscow) **9**, 2449 (1982) [Sov. J. Quantum Electron. **12**, 1597 (1982)].

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Gasdynamic lasers utilizing carbon gasification

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A theoretical investigation was made of the influence of the processes of carbon gasification by combustion products and oxidants on the chemical composition of the active medium and the energy characteristics of a gasdynamic CO_2 laser. Conditions were found under which the stored energy of the active medium was greater than 100 J/g.

1. As is well known,[1] the output power of gasdynamic CO_2 lasers is proportional to the specific energy E stored in the active medium, which is determined by the expression

$$E = (h\nu/N)\sum \gamma_i [n_i(T_i) - n_3(T_3')]/\sum \gamma_j M_j, \qquad (1)$$

where $h\nu$ is the photon energy of the laser radiation; N is the total particle concentration in the gas stream; the summation in the numerator is performed only over the vibrational modes of the energy-carrying components of the mixture (N_2, CO, asymmetric vibrations of CO_2), and that in the denominator over all the components of the mixture; γ_j and M_j are the molar fraction and mass of the particles of the jth component; $n_i(T_i) = [\exp(\theta_i/T_i) - 1]^{-1}$ is the average number of vibrational quanta with a characteristic temperature θ_i per molecule of the ith type with a vibrational temperature T_i; the subscript 3 denotes the asymmetric CO_2 mode; $T_3' = T_1\theta_{00^01}/\theta_{10^00}$ (Ref. 2) is the vibrational temperature for which the gain is zero; T_1 is the vibrational temperature of the symmetric CO_2 vibrations.

It can be seen from Eq. (1) that the larger the value of E the higher T_i and the lower T_1, so that in order to maximize the specific stored energy one must raise the gas temperature T_0 in the forechamber and lower the relaxational losses of the energy-carrying molecules which result from collisions with the quenching components. Among such components are, for example, water vapor, atomic impurities, and in part CO_2 and H_2O molecules. At the same time CO_2 is an emitting component and the presence of a certain amount of water vapor in the mixture is necessary in order to clear the lower active levels, so that the requirements for maximizing E as applied to the combustion products of the widely used hydrocarbon fuels are contradictory. In fact, high values of

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