# Ultrafast Time-and-Space-Domain Holography by Spectral Hole Burning in Dye-Doped Polymers

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*Abstract.* Certain photochromic materials exhibit at liquid-helium temperature a special property of very high selectivity in frequency dimension. This phenomenon, commonly known as spectral hole burning (SHB), makes it possible to extend conventional spatial-domain optical data storage into the dimensions of frequency and time. We have applied SHB for ultrafast recording of holograms and coherent optical processing on the timescale of $10^{-12}$–$10^{-13}$ s. To achieve ultrafast performance in the time domain, we use special organic dye-doped polymer materials, which provide SHB recording in a broad optical band width of 5–10 THz. In this paper, we discuss recording and playback of holograms of pico- and femtosecond time-and-space-domain signals using dye-doped SHB polymers at liquid-helium temperature. We discuss unusual properties of SHB holograms such as causality-related asymmetry of diffraction and inversion of the time coordinate, ultrafast frequency-domain processing, and associative recall of events.

## 1. Introduction

Currently, there is a large interest in new physical and chemical principles, which would allow to store and process data with much higher speed and with much higher capacity than it is currently possible with existing technologies. In this respect, certain photochromic materials, which exhibit unique property of very high selectivity in frequency dimension, are of special interest. This phenomenon occurs typically at liquid-helium temperatures and is commonly known as spectral hole burning (SHB). Spectral hole burning can be used to increase by several orders of magnitude the capacity and speed of data storage and processing, basically because it allows to extend conventional optical storage techniques into the new dimensions of frequency and time.

We focus our investigations on prospective applications of SHB for ultrafast holography and coherent optical process-

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ing on the timescale of $10^{-12}$–$10^{-13}$ s. The advantage of using the holography consists in the way how massive amount of data can be addressed and processed in parallel. Furthermore, holographic method provides a natural way to combine and manipulate different optical degrees of freedom – the three spatial degrees of freedom, the frequency coordinate, and connected to it *via Fourier* transformation the time coordinate. All these storage dimensions are simultaneously available in SHB medium. Time-and-space-domain holography serves as a multidimensional generalization of conventional holography to utilize the four or five optical storage dimensions.

To achieve the performance on the ultrafast timescale, we apply special SHB materials, which provide up to 10 THz spectral band width. In this paper, we discuss recording and playback of holograms of pico- and femtosecond time-and-space-domain signals using polymer films activated with porphine-type dye molecules at liquid-helium temperature. We also discuss unusual properties of SHB holograms such as causality-related asymmetry of diffraction and inversion of the time coordinate, ultrafast frequency-domain processing, and associative recall of events.

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His current research interests are optics and spectroscopy of spectrally selective materials, ultrafast nonlinear optics and holography for applications in future high-speed optical communications and data storage. For his work on time-and-space-domain holography *Aleksander Rebane* has been awarded International Commission for Optics Prize in 1993.

## 2. Spectral Hole Burning

SHB is based on the fact that the absorption spectrum of chromophores in solids at low temperature, *e.g.*, dye molecules doped into polymers, have broad inhomogeneous absorption bands, which are composed by superposition of a large number of narrow homogeneous resonance lines – purely electronic zero-phonon lines (ZPL) [1]. By choosing chromophores, which are photochemically active at liquid-helium temperature, it is possible to selectively bleach out these dye molecules whose ZPLs are exactly at the wavelength of the illuminating light, while the rest of the inhomogeneous absorption band remains practically unchanged. In the case of monochromatic illumination, this creates a narrow dip or hole in the inhomogeneous absorption profile, corresponding to a 'negative' image of the ZPL [2].

The minimum width of spectral holes is approximately the double homogeneous ZPL line width, $\Gamma_{hom}$, and can be as narrow as $\Gamma_{hom} = 1$–$10^{-2}$ GHz for organic dye

molecules at temperatures below 4–10 K and even considerably narrower for rare-earth ions in crystals. Because the inhomogeneously broadened band is formed by superposition of a great number of ZPLs, its width, $\Gamma_{inh}$, can be as much as $10^3$–$10^6$ times larger than the width $\Gamma_{hom}$. SHB in inhomogeneously broadened bands with large ratio between $\Gamma_{inh}$ and $\Gamma_{hom}$ provides for high density optical storage, where the frequency dimension accommodates thousands or even hundreds of thousands of data bits in each tightly focussed laser spot [3]. It is most remarkable that SHB allows changing the absorption coefficient and correlated to it by *Kramers-Kronig* dispersion relations index of refraction of the media with high precision in the frequency dimension. Illumination reduces the number of resonantly absorbing molecules at a particular frequency in proportion to the amount of the absorbed energy, which in turn alters the complex index of refraction of the media. In some cases, the absorption can be reduced by several orders of magnitude, which is useful for constructing wide-aperture narrowband spectral filters [4].

The persistence or the lifetime of the spectral holes depends largely on the relaxation of the molecule's excited states as well as on the thermally activated conformations of the molecule and its closest surrounding. Organic dye molecules such as free-base porphyrines possess an intrinsic phototautomerization mechanism with very slow relaxation at liquid-helium temperatures. This permits recording of permanent holograms, which persist for days or longer. A variety of different SHB processes have been investigated in organic dye molecules, rare-earth ions, and color centers, both in ordered hosts like single crystals as well as in disordered hosts like glasses and polymers. Certain SHB mechanisms have short nanosecond relaxation times, which is useful for transient storage and processing (for a review on SHB mechanisms see [5] and references therein).

One way of addressing data is to do it with a narrow-band laser, which is tuned over the inhomogeneous band from one frequency to another. Alternatively, all frequencies may be addressed in parallel by using a pulsed laser with a broad spectrum, which covers the whole inhomogeneous band at once. Based on these two approaches, frequency-domain holography [6] and time-domain holography [7][8] were proposed. In particular, the serial frequency-domain approach has been used to demonstrate recording and playback of holographic movies, containing several thousands of holographic image frames,
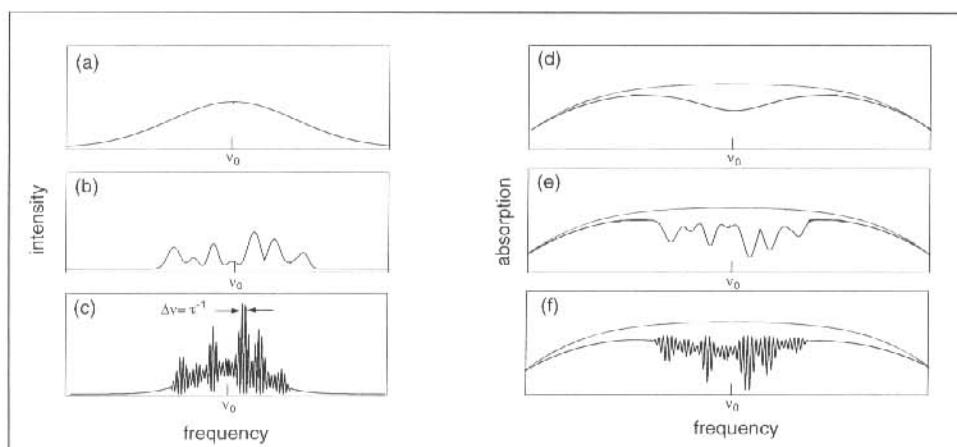


Fig. 1. a) *Intensity spectrum of short, temporally smooth reference pulse with center carrier frequency $\nu_0$.* b) *Intensity spectrum of a temporally (and spectrally) modulated object pulse with the same center carrier frequency.* c) *Combined intensity spectrum of the reference and object pulses showing an interference in the frequency dimension. The period of the fringes is equal to inverse value of the delay t between the pulses.* d) *Hole burned into inhomogeneously broadened absorption band by the reference pulse. The dashed line corresponds to the absorption before illumination.* e) *Hole burned by the application of the object pulse. The shape of the hole corresponds to the intensity spectrum of the object. No or very limited phase information is stored.* f) *Combined burning with object and reference pulses. The hole profile records the frequency-domain interference pattern which contains information about the amplitude and phase of the object.*

each frame recorded in a narrow frequency interval of a single projector-slide-size sample [9]. In combination with *Stark* effect [10], 'molecular computing' has been demonstrated by making use of interference between holographic images recorded at different frequencies [11].

In our time-domain experiments, the holographic images are played back not as separate frames, but rather as an entire time-and-space-domain scene. Instead of spending time on shifting the laser frequency to access different image frames, the whole hologram is accessed at once and is played back in one flash. This is possible because the hologram contains information about both, the amplitudes as well as the relative phase of the spectral components of the signal. Time-and-space-domain holograms recall, in addition to the spatial structure of the object also the delay, the duration and the complete temporal shape of the object.

In organic SHB materials, the maximum duration $\Delta t$ of the recorded time-and-space-domain scene or event is limited to about a few nanoseconds basically by the finite value of the homogeneous ZPL line width, $\Delta t < (\Gamma_{hom})^{-1}$. On the other hand, the inverse value of the inhomogeneous band width defines the shortest duration event that can be recorded. We utilize the fact that in some dye-doped polymers the inhomogeneous width is as large as 5–10 THz, which permits recording and playback with femtoseconds resolution in time domain.

A review on physical background and theory of time-and-space-domain holo-

graphy was presented recently in [12]. Here, we give a brief description of the basic concepts and provide experimental illustrations on time-and-space-domain storage with short laser pulses.

## 3. Time-and-Space-Domain Holography with Femtosecond Pulses

The idea of time-domain holographic storage is based on extending the notations of interference and diffraction to describe frequency- and time-domain amplitudes [13]. *Fig. 1, a* and *b* shows, correspondingly, the frequency-domain amplitude of a brief reference pulse and of a structured object pulse. Both pulses have the same central carrier frequency $\nu_0$. The combined intensity (*Fig. 1, c*) of the two pulses contains a pattern of interference fringes with the period equal to the inverse value of the time delay between the pulses, $\Delta \nu = \tau^{-1}$. The holographic information about the object pulse is contained in the frequency-domain interference pattern.

The role of the SHB material is to record the intensity pattern in the frequency dimension. *Fig. 1, d* and *e* shows how the inhomogeneous absorption profile is modified by illumination with the object and reference pulse taken separately. *Fig. 1, f* shows the combined effect of the two pulses: the absorption profile memorizes the frequency-domain interference between the object and reference. Note that the two pulses need not to overlap in time. It is needed, however, that the time delay is not larger than the inverse value
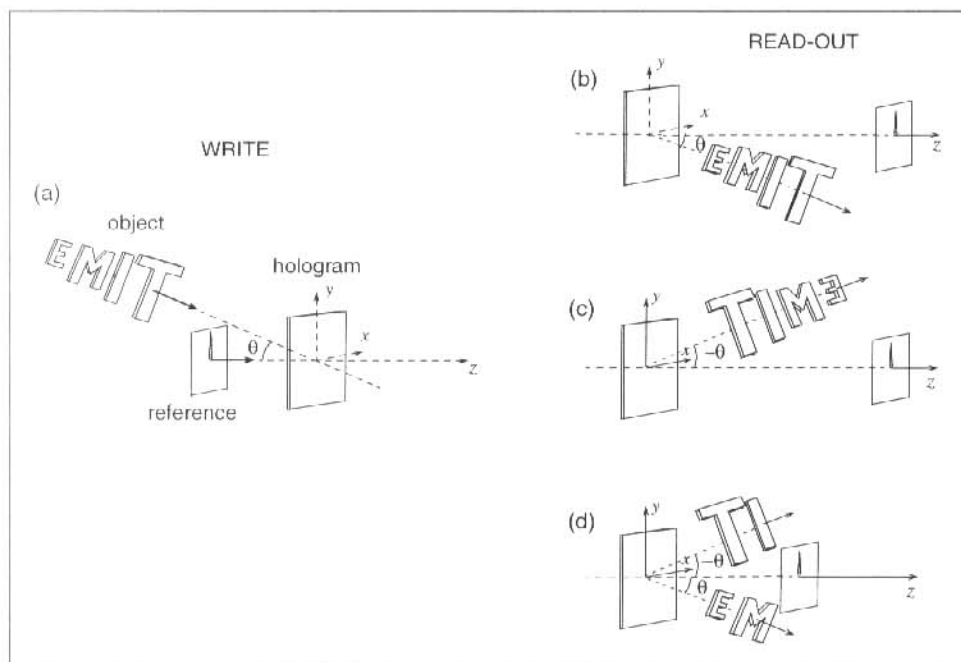
Fig. 2. *Scheme of recording (a) and playback (b–d) of time-and-space-domain holograms* (see text for explanations)
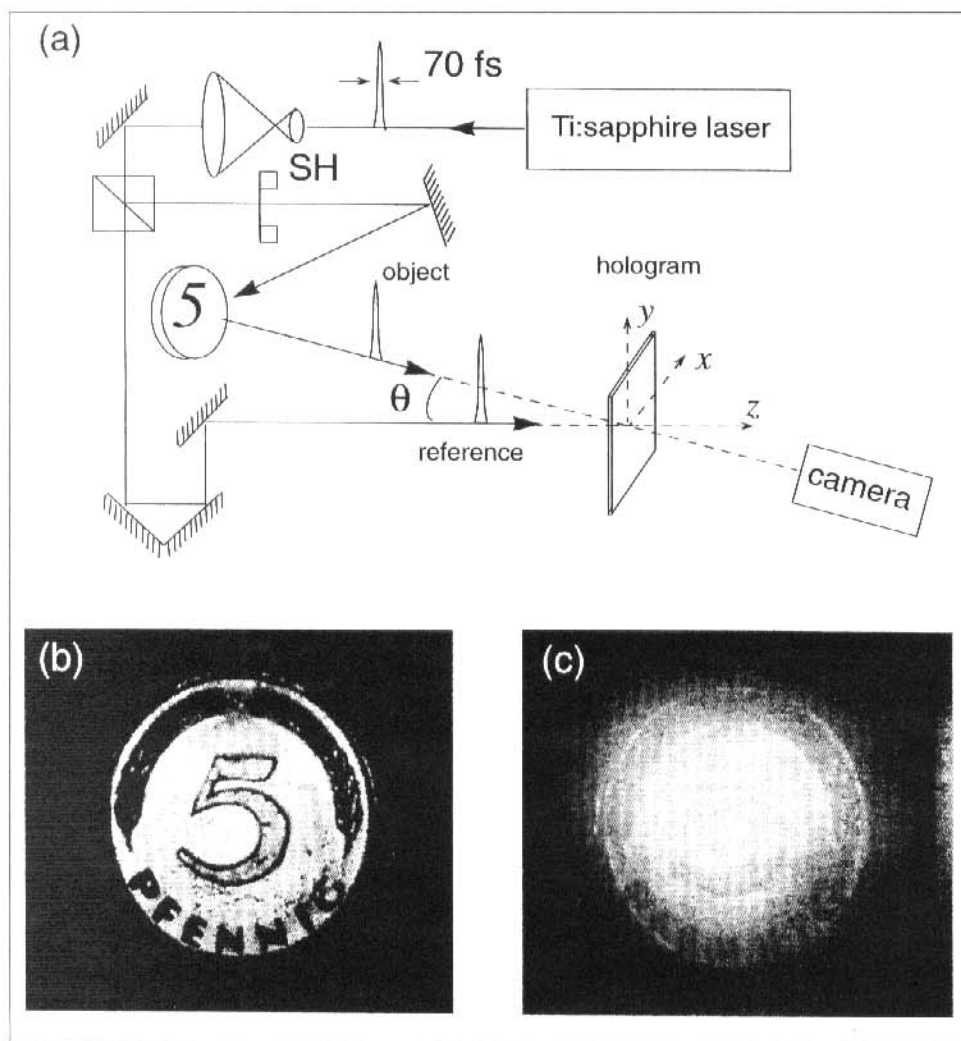


Fig. 3. a) *Experimental arrangement to write SHB holograms with 70-fs-duration pulses.* The reference beam propagates along the z-axis, whereas the object beam is scattered from the coin. Shutter (SH) is used to block the object beam during the readout. b) *The original image of the coin illuminated with the Ti:sapphire-laser beam.* c) *Image reconstructed from the hologram.*

of the homogeneous ZPL line width, $\tau < (\Gamma_{hom})^{-1}$.

The hologram is interrogated by illuminating the medium with a replica of the reference pulse. Due to scattering (diffraction) by the spectral pattern, the transmitted interrogating pulse is accompanied by a time-delayed coherent optical response, which is a replica of the object pulse. This property is closely related to photon echo [14–17], a well-known phenomenon in coherent optical spectroscopy.

Next, we assume that the object pulse has not only a distinct temporal structure, but also a certain structure of the wave front. *Fig. 2, a* shows a plate of SHB material illuminated by the object pulse and by a plane-wave reference pulse, applied at a different angle. In this geometry, each spatial location of the SHB material may be recording a different interference structure in the frequency dimension. The resulting complicated frequency-and-space-domain interference pattern contains all the necessary information about the object. Here, the object is depicted to consist of four pulses, each one arriving at a different time. *Fig. 2, b–d* shows the interrogation of the hologram with a replica of the reference pulse, depending on the relative temporal ordering of the object and the reference during the recording. When the reference pulse precedes the object pulse, then the hologram plays back a replica of the object including its temporal structure. However, if the temporal ordering of the writing pulses is reversed, *i.e.*, if the object pulse is applied before the reference pulse, then the hologram plays back the conjugated wave front of time-inverted replica of the object (*Fig. 2, c*). In the intermediate case (*Fig. 2, d*) the reference arrives simultaneously with the object pulse and the recalled signal is then split into two parts propagating in different directions: the earlier half of the object is recalled as a conjugated time-inverted replica, whereas the later half is reproduced as a non-inverted replica. This occurs because causality principle serves as a 'time arrow', which distinguishes between the signals that arrive before and after the reference pulse. Recently, we have used this effect to study 'time-edge' holograms, where the direction of diffraction changes abruptly as a function of the delay [18], and have shown that such holograms can be used to measure the homogeneous spectrum of the chromophore molecules in a very short interval of time. In addition, causality-related effects can be used to view objects hidden behind opaque screen [19].

*Fig. 3* shows an experiment where we record an image of a coin illuminated with 70-fs-duration pulses from a mode-locked Ti:sapphire laser. For this experiment, we use 200-μm-thick polyvinylbutyral film activated with special phthalonaphthalocyanine dye molecules absorbing at the 750–780-nm-wavelength of Ti:sapphire laser. The solubility of these molecules in polymer was chemically enhanced by introducing the 2,4-dimethyl-3-pentoxy substituents [20]. The sample was contained at temperature $T = 2K$ in an optical cryostat. It is interesting to note that the coherence length of the illuminating light (*ca.* 20 μm) was several orders of magnitude shorter than the optical beam-path difference between the writing pulses (several cm), which excludes any conventional interference between the beams. *Fig. 3* shows that holograms can be recording even if the light shows no interference in conventional sense – all the interference occurs in the spectral dimension of the SHB medium.

*Fig. 4* shows an experiment, which illustrates the effect of inversion of the time shape of the object pulse. *Fig. 4, a* shows the writing object pulse consisting of a train of picosecond pulses, followed by single reference pulse. The object-pulse train starts at time $t = 0$ and extends over one nanosecond, whereas the delay of the reference pulse is 400 ps. *Fig. 4, b* shows the signal obtained from the hologram in the conjugated diffraction direction. The hologram reproduces a time-inverted replica of that part of the object, which preceded the reference during writing.

## 4. Associative Recall of Time-and-Space-Domain Images

Time-and-space-domain holograms can be recorded also in an associative manner, without using a separate reference pulse. In this case, the recollection of the information is performed by illumination with a selected fragment of the original object [21]. *Fig. 5, a* shows a beam of picosecond laser that passes through a stack of optical delays of 0, 34, 68, and 102 ps. Subsequently, the beam scatters from ground glass plate in the forward direction, reaching the SHB hologram plate inside a cryostat. The writing procedure was accomplished simply by illumination with the object beam, without using any special reference. The readout was performed by illuminating the hologram with a part of the attenuated object beam. *Fig. 5, b* shows the original spatial image of the object observed with a photographic cam-
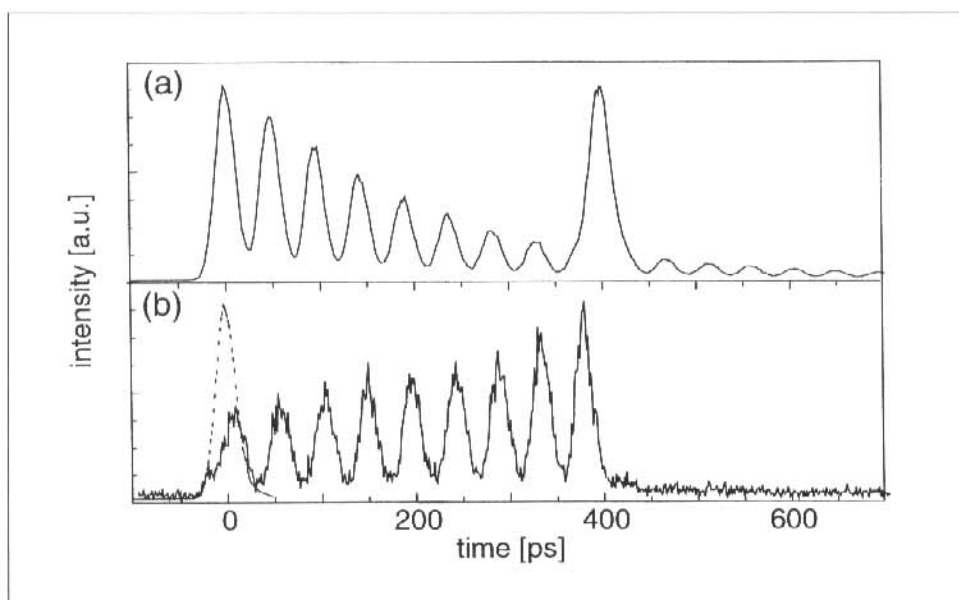


Fig. 4. *Inversion of time-domain signal by SHB hologram. a*) The writing object pulse train (starting at time zero) followed by reference pulse at delay 400 ps; the SHB sample is a 100-μm-thick chlorin-doped PVB film at $T = 2K$. The laser source is a synchronously pumped dye laser with pulse duration of 2–3 ps and spectral width of 5 cm$^{-1}$. The pulse train is created by a *Fabry-Perot* etalon and detection is made with a 30-ps-resolution streak camera. *b*) Hologram signal observed in conjugated diffraction direction. Dashed line shows the applied readout pulse.
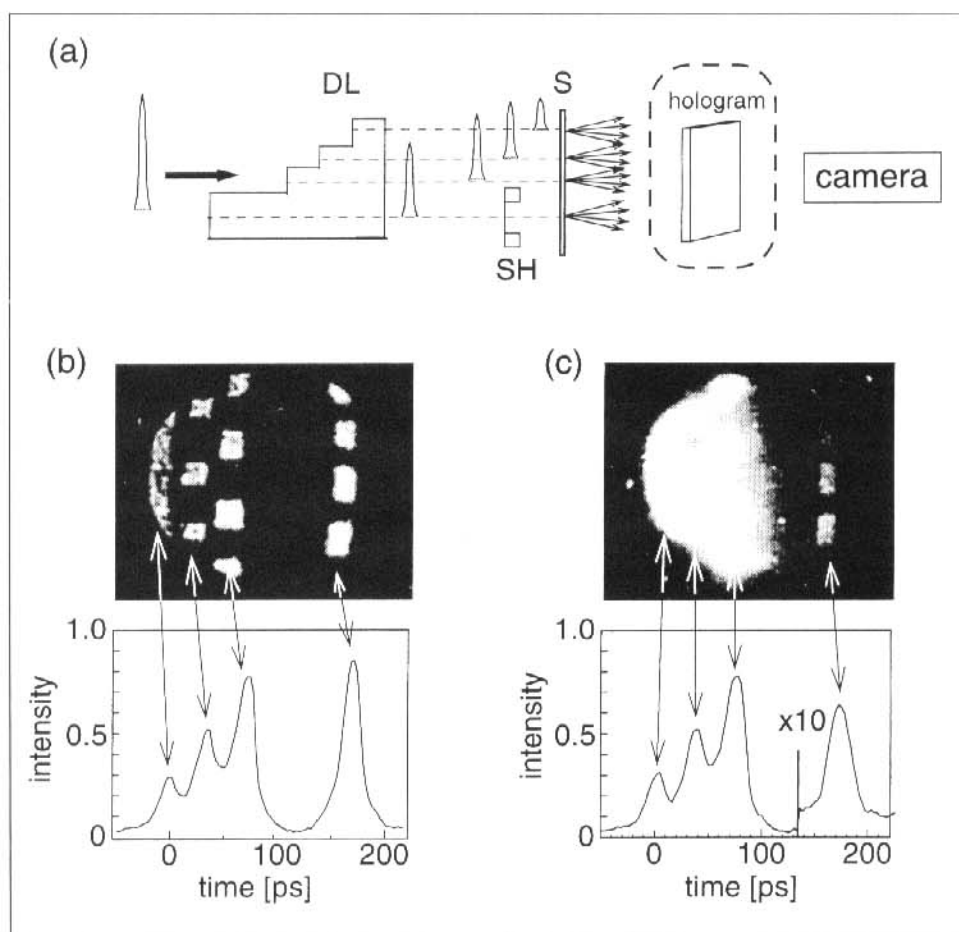


Fig. 5. *Associative recall of time-and-space-domain holograms. a*) Experimental arrangement; DL – optical delay; S – scattering screen. *b*) The original spatial image and the corresponding time-resolved structure of the object. Arrows indicate the correspondence between the spatial image parts and time-domain pulses. *c*) Associatively reconstructed signal when the 'key' fragment consisting of three first pulses (brightly illuminated part on the left side of the image). The signal reconstructed by the hologram appears on the right side of the image.

era and the corresponding temporal intensity profile measured with a picosecond time-resolution streak camera. The arrows indicate which part of the object in the time domain corresponded to which part of the spatial image. *Fig. 5, c* shows the result of associative readout with an interrogating 'key', which consisted of the three first pulses. The hologram recalls by association the image and the temporal shape of the missing fragment. However, if we used a 'key' that was the last fragment, then the hologram did not play back any of the missing parts of the object. This demonstrates again that the causality-related 'time arrow' depends on the relative time ordering of the illuminating light signals, which is prohibiting associative recall of those parts of the scene, which are in time later than the 'key' fragment [22]. This property, pointed out earlier by *Gabor* [23], resembles the function of human memory.

A digital version of associative SHB memory was studied in [24]. In this case, the holograms served as programmable multidimensional (spatial, frequency, and time dimension) interconnecting optical elements between arrays of digitally coded input and output signals. SHB materials allow to increase the dimensionality of optical interconnections and were used to implement error-corrective autoassociative memory in time and in frequency dimension.

## 5. Spectral Programming of Ultrashort Time-Domain Signals

In this experiment, we show that it is possible to produce arbitrary time-domain pulse shapes by using SHB holograms synthesized in the frequency dimension [25]. We recorded a set of SHB gratings at different frequencies with variable, predefined relative amplitudes and phases by using a narrow-band tunable laser. We then applied a short subpicosecond pulse to generate a photon echo signal and measured its intensity profile in time domain. As the tunable narrow-band laser we utilized a dye laser with the line width of 0.5 cm$^{-1}$. The SHB material was chlorin (dihydroporphyrin) in PVB (= polyvinylbutyral) at $T = 2$K. The hole-burning proper-

ties of chlorin are discussed in [26]. The maximum diffraction efficiency of the recorded gratings was on the order of 1%. The readout was carried out by 200-fs-duration white-light continuum in the wavelength range of 630–635 nm generated with the help of an amplified femtosecond Ti:sapphire-laser system. The time profile of the hologram signal was measured by cross-correlation with Ti:sapphire-laser pulses.

*Fig. 6, c–e* shows different pulse trains, which were produced by this method. For each pulse train, a separate set of frequency-domain gratings was recorded according to a given calculated amplitude and phase algorithm. The measured duration of the individual pulses in the train was *ca.* 350 fs, which gives the time resolution of the synthesized signals. The maximum time span of the pulse trains was *ca.* 0.5 ns and was limited by the finite homogeneous ZPL line width. The potential practical application of ultrafast synthesized pulse trains is in transmission and routing of data in optical fibers links and in ultrafast optical processors.

In all experiments described above, the recorded holograms are gradually erased by illumination with the readout light. In certain applications such as permanent memories and pulse-train generators, it is required that the recorded holograms are resistant with respect to irreversible erasure during the readout. To resolve this problem, it is of great interest to use two-color photon-gated SHB materials [27]. In these materials, permanent holes are produced only if an additional illuminating beam, typically of a different color or wavelength, is added. Readout with only one color will not cause irreversible erasure. We have shown recently that photon-gated hole-burning allows practically nondestructive readout of time-and-space-domain holograms [28]. It also allows to read out holograms with a much higher intensity than during the writing, without actually destroying the recorded information.

## 6. THz Bit-Rate Optical Processing by Two-Pulse Photon Echo

In this experiment, we applied low-temperature polymer to demonstrate multiplication of two 8-bit binary vectors in less than 10 ps by using two-pulse photon echo [29]. We used a two-channel diffraction-grating pulse shaper to code binary input data into the intensity spectrum of 100-fs-duration excitation pulses derived from a regenerative-amplifier system. The
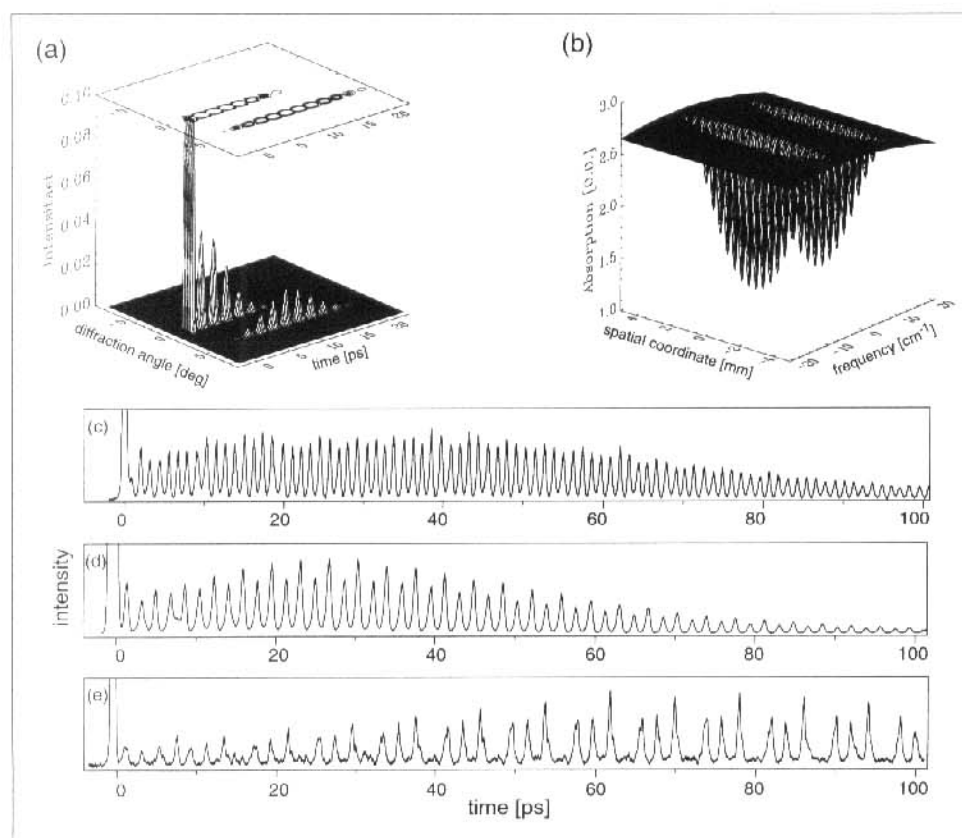


Fig. 6. *Spectrally programmed time-domain SHB holograms.* a) Quasi-three-dimensional representation of one possible time-domain response function. The axis perpendicular to the time coordinate represents the diffraction angle. b) Calculated spatial spectral structure that has to be written into the absorption spectrum of the SHB plate in order to produce the desired response function in time domain. c–e) Three different time-response functions implemented in the experiment. The synthesized holograms are producing different series of subpicosecond pulses with predefined amplitudes and phases.
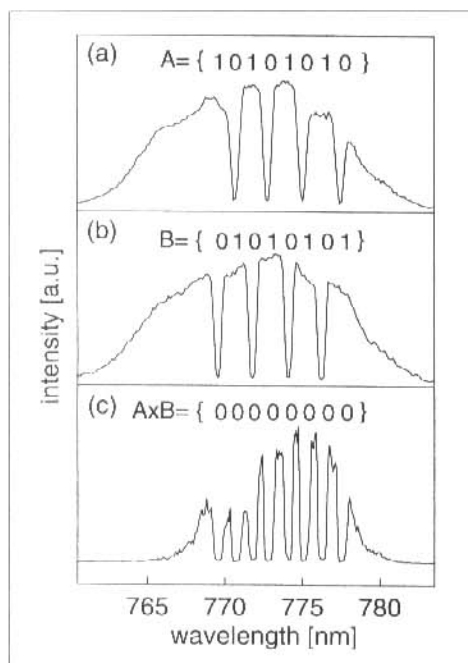
Fig. 7. *Ultrafast bit-to-bit multiplication by two-pulse photon echo. a)* Intensity spectrum of the first excitation pulse. *b)* Intensity spectrum of the second excitation pulse. *c)* Intensity spectrum of the photon echo signal. The duration of the excitation and echo pulses is *ca.* 10 ps.

spectral coded pulses illuminated a sample contained in a low-temperature cryostat. A few-picoseconds time delay was set between the pulses to avoid temporal overlap. The result of the multiplication is obtained by measuring the intensity spectrum of the echo. *Fig. 7, a* and *b* shows the intensity spectrum of the excitation pulses, presenting two binary vectors A = {1, 0, 1, 0, 1, 0, 1, 0} and B = {0, 1, 0, 1, 0, 1, 0, 1}. By choosing appropriate power of the excitation pulses, we observed that the intensity spectrum of the echo signal corresponds to bit-to-bit multiplication of binary vectors A × B = {0, 0, 0, 0, 0, 0, 0, 0} (*Fig. 7, c*). Due to parallel access of the data in frequency dimension, this method sets a record of the speed of optical processing, allowing multiplication of several terabits per second.

## 7. Summary

Our experiments have shown that spectral hole-burning holography in time and frequency domain holds great promises for future generation of ultrafast storage and processing devices. The special properties of SHB materials open up principally new ways to record and manipulate broad-band optical signals. The important fact is that the frequency selectivity is an intrinsic property of the material, and is therefore not restricted to any specific

beam propagation direction or spatial location. Because of this property, we can speak about true multidimensional optical recording, where the spatial coordinates are fully independent from frequency and/or time coordinate.

Currently available organic SHB materials provide at liquid-helium temperature up to tens of thousands of frequency channels within inhomogeneous band width of 5–10 THz. This matches well the band width required for operation on femtosecond timescale. We have used dye-doped polymers immersed in liquid helium to demonstrate time-and-space-domain storage and processing of signals as short as few tens of femtoseconds in the near-infrared range of wavelengths. Shifting the operation wavelength further towards longer wavelengths would allow to implement devices for fiber communications. All materials used in our experiments so far lose their frequency-selective properties at higher temperatures. For several practical applications, it will be highly desirable that the SHB materials would preserve the large ratio between the inhomogeneous and the homogeneous line width also at higher temperatures, up to room temperature. Photon-gated spectral hole-burning materials, which resist erasure of the information during the readout are also required. Optimization of the SHB material properties along with the investigation of the nonlinear coherent optical effects on ultrafast timescale will be the focus of further work in this area.

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