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Non-destructive readout of picosecond time-and-space-domain holograms recorded by photon-gated spectral hole burning

Alexander Rebane, Daniel Reiss, Indrek Renge, Urs P. Wild

Physical Chemistry Laboratory, Swiss Federal Institute of Technology, ETH-Zentrum, CH-8092 Zürich, Switzerland

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Abstract

Two-color photon-gated persistent spectral hole burning in zink tetrabenzoporphine with a solid halocarbon additive 1,1-bis(4-chlorophenyl)-2,2,2-trichloroethane in polystyrene film at liquid-helium temperature has been used for recording of picosecond photochemically accumulated stimulated photon echo (PASPE) holograms. In the presence of the gating illumination at 532 nm the hole-burning yield increased by a factor of up to 100. With the gating illumination switched off we were able to read out the recorded time-and-space-domain holograms for a long time and with high intensity practically without destroying the contrast of the recorded information.

1. Introduction

Spectral hole burning (SHB) [1,2] is based on the existence of very narrow and intense purely electronic zero-phonon lines (ZPL) which constitute the inhomogeneously broadened absorption band of a chromophore in a solid matrix at low temperatures. Potential practical applications of SHB in optical storage devices [3–5] make use of the fact that by illumination one can change the optical properties of the media such as absorption coefficient and correlated to it via Kramers–Kronig dispersion relations index of refraction in the frequency dimension, where the precision in the frequency is essentially given by the homogeneous width of the ZPLs.

New versions of holography using frequency-domain writing and readout with a narrow-band laser [6,7] and time-domain writing and readout with spectrally broad short pulses [8–11] have been developed offering new possibilities for high-capacity and high-speed optical storage. Typical SHB materials used in these experiments are either rare-earth ions in crystals or dye-doped polymers. The hole burning occurs following one-step excitation of the chromophores from the ground state to an excited electronic state with a quantum yield (i.e. number of photo-transformed chromophores per one absorbed photon) practically independent from the intensity of the illumination. Due to this linear character of the hole-burning process, any resonant illumination will bleach the SHB medium in proportion to the dose of the absorbed optical energy. In the prospective optical storage applications this means a serious drawback because each readout of the data inevitably causes an erasure of a part of the recorded information.

With normal SHB substances such as dye-doped polymer films, where the hole burning occurs follow-

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ing one-photon excitation to the first singlet excited electronic state, it was recently demonstrated that the erasure of frequency-domain holograms can be reduced by using a special hologram readout technique [12]. It has been shown that the erasure rate of the information is slower if during the readout the frequency of the illuminating narrow-band laser is restricted to certain discrete frequencies where the absorption of the material is minimal and the index of refraction contribution to the diffraction efficiency of the hologram is maximal. However, this method cannot be applied directly to time-and-space-domain holograms because in this case the readout is generally performed with a broad continuous spectrum rather than with a set of discrete frequencies.

The 'photon-gated' SHB materials promise a more universal solution to the problem of non-destructive readout of holograms. Two- or multiple-step photoprocesses which require the absorption of additional (gating) photons at a different wavelength from an intermediate excited electronic state of the molecule [13] satisfy these conditions. When the gating beam is turned off, then the photons absorbed in the first step alone cannot cause permanent bleaching of the media and the erasure of the information can be avoided. A two-quantum photoprocess in the course of resonant excitation of zero-phonon transitions was observed for tetracene molecules embedded in a glassy solvent [14]. Two-color hole burning was accomplished for a derivative of zinc tetrabenzoporphine (ZnTBP) in poly(methyl methacrylate) (PMMA) matrix [15]. The hole-burning efficiency increased at least by an order of magnitude when the selective radiation of the He–Ne laser was combined with pulsed UV excitation of a N_2 laser [15]. A detailed study of the same system with liquid halocarbon additives (CH_2Cl_2, CH_2Br_2, $CHCl_3$) has demonstrated that the triplet state is involved in the phototransformation process [16]. It was concluded that probably electron transfer occurs from the highly excited triplet molecule to the halocarbon acceptor. In a number of following papers photon-gated hole burning in different organic and inorganic materials has been studied [17]. However, all these experiments used simple transmission- or absorption-type of detection of the holes rather than holography. Holographic techniques have important advantages in the data storage but they are setting also much higher requirements on the optical quality of the samples and on the efficiency of the gated hole burning process.

In this Letter we report a new organic photon-gated SHB material which consists of polystyrene matrix doped with zink tetrabenzoporphine (ZnTBP) molecules and with a solid halocarbon 1,1-bis(4-chlorophenyl)-2,2,2-trichloroethane (DDT) as the activator of the gated SHB process. This material can be prepared as a thin film of good optical quality, it has at liquid-helium temperature a high gated hole-burning efficiency and does not deteriorate when stored at room temperature for a long time. This allowed us, for the first time, to record gated coherent optical transients such as photochemically accumulated stimulated photon echo and perform essentially non-destructive readout of picosecond time-and-space-domain holograms.

2. Experimental

To prepare the sample, the solutions of polystyrene (Aldrich, average molecular weight ≈ 280000) and DDT both in dichloromethane, and ZnTBP in pyridine were mixed and then cast and let to dry over several days in a Petri dish which was covered with a glass plate to ensure slow evaporation. The resulting raw film was kept under vacuum at 60°C for 24 h to remove the residual solvent. The dry film was 100 μm thick and contained 20% (0.56 M) of DDT and 2.5×10^{-4} M of ZnTBP. The film was removed from the glass and a 1 cm^2 piece was cut out and affixed to the cryostat sample holder. The optical density of the sample at the maximum of the S_1–S_0 absorption of ZnTBP was 0.6. During the whole experiment the sample was kept in superfluid helium at 2 K.

A scheme of the experimental arrangement is shown in Fig. 1. As the light source we used a 76 MHz repetition rate picosecond dye laser synchronously pumped by a Coherent Antares 76s frequency-doubled Nd:YAG laser. The dye laser delivered 0.6 nm spectral width and 3–4 ps duration pulses with an average power of 300 mW. In a standard holographic setup the dye laser output was expanded with a telescope and divided into two plane wave beams. The reference beam was applied

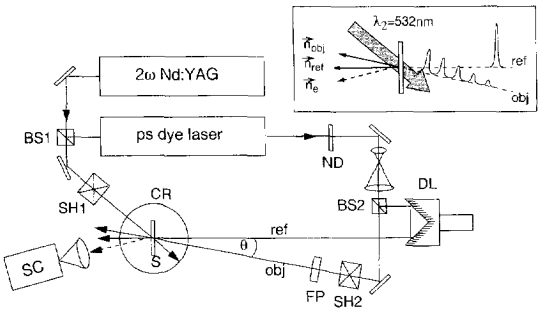

Fig. 1. Scheme of the experiment. BS1, BS2: beam splitters; SH1, SH2: shutters; DL: optical delay line; ND: neutral density filter; CR: cryostat; S: sample; SC: streak camera; FP: Fabry–Perot etalon. Insert shows detailed view of the arrangement of the illuminating beams. The gating beam is indicated as bold arrow.

in the direction $\boldsymbol{n}_{\mathrm{ref}}$ at normal incidence angle with respect to the SHB film. The object beam was applied in the direction $\boldsymbol{n}_{\mathrm{obj}}$ at an angle $\theta = 60°$ with respect to the reference. The diameter of the spot illuminated by both writing beams at the sample was 6 mm. The average power of the beams was 60 μW. The reference beam was delayed by 130 ps with respect to the object beam. The wavelength λ_1 of the picosecond dye laser was tuned into resonance with the inhomogeneously broadened absorption band of the S_1–S_0 transition of ZnTBP in the range λ_1 = 627–637 nm.

In order to simulate a train of picosecond data pulses we used a plane-mirror Fabry–Perot etalon that was inserted into the object beam.

To detect picosecond signals we used a Hamamatsu 00S-1 streak camera synchronized with the mode-locker of the Nd:YAG laser. The time resolution of the detection (defined as full width at half maximum of the pulses) was about 35 ps. In the experiment we used lenses to focus the light diffracted from the hologram on the entrance slit of the streak camera.

Part of the frequency-doubled output of the mode-locked Nd:YAG pump laser at the wavelength λ_2 = 532 nm was used to provide the gating illumination. The gating beam was expanded and was directed to the SHB film from the opposite side with respect to the two hologram writing beams. The diameter of the spot illuminated by the gating beam was 7 mm and it completely covered the area that was illuminated by the two red-color beams. The average intensity of the gating beam was 180 mW/cm^2.

3. Results and discussion

To write the holograms we opened both the object and the reference beams and simultaneously illuminated the sample with the gating beam. For the readout the hologram was illuminated only with the reference beam, while the object beam and the gating beam were blocked. The hologram diffraction signal was monitored in the direction $\boldsymbol{n}_{\mathrm{echo}} = 2\boldsymbol{n}_{\mathrm{ref}} - \boldsymbol{n}_{\mathrm{obj}}$. Note that in this diffraction direction the phase-matching condition for the stimulated photon echo is fulfilled only approximately. However, because of the small thickness of the SHB film and because of the small value of the angle θ this did not affect the detection of the echo signal.

Fig. 2 (solid line) shows the low-temperature absorption spectra of the sample before the writing exposure. The maximum of the S_1–S_0 in homogeneous band is at 630 nm and it partially overlaps with the other band at 626 nm which belongs to a higher excited electronic state of the ZnTBP chromophore and which displays no sharp zero-phonon lines.

In addition to the persistent SHB our sample exhibited also transient hole burning due to shelving

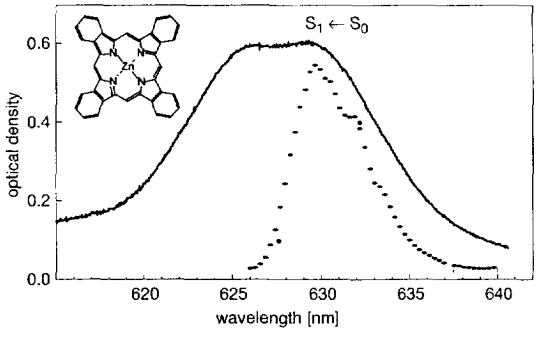

Fig. 2. Transmission spectrum of ZnTBP–polystyrene sample at liquid-helium temperature measured before the writing exposures. The S_1–S_0 transition inhomogeneously broadened absorption band of ZnTBP with the maximum at 630 nm partially overlaps with the transition to a higher excited electronic state with the maximum at 626 nm. Dots show the measured relative intensity of triplet-accumulated stimulated photon echo measured at different laser wavelengths.

of the population in the metastable triplet state of ZnTBP having a lifetime of 40 ms [18]. By this mechanism, simultaneously with the writing of the permanent hologram we also created a transient accumulated stimulated photon echo [19–21] that was detected as diffraction of the writing reference beam. The lifetime of the transient hologram was about 50 ms. Fig. 2 (dots) shows how the intensity of the transient hologram signal depended on the excitation wavelength. The transient signal was observed in a wavelength range of about $\Gamma_{inh} \approx 6$ nm and corresponds approximately to the frequency distribution of the purely electronic zero-phonon lines of the S_1–S_0 transition, i.e. the inhomogeneous distribution function.

With the gating illumination switched on we needed a rather small exposure dose of only about 1 mJ/cm^2 (corresponding to about 3–5 s illumination time) to write a well detectable permanent hologram. In comparison, without the gating beam the minimum writing exposure to obtain a detectable hologram was about 60 mJ/cm^2. Fig. 3 shows time-domain diffraction signals from two permanent SHB holograms which were written, correspondingly, with (a) and without (b) the gating light illumination. From this result we can estimate that the increase in the hole burning efficiency due to the illumination at 532 nm is at least by a factor of 100. The actual gating ratio may be even higher because the hologram signal without the gating remains close to the level of the scattering background noise.

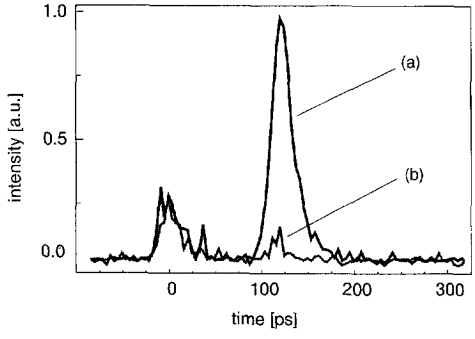

Fig. 3. Permanent SHB time-and-space-domain hologram signal measured with picosecond streak camera. The delay between the reference pulse and the object pulse is 130 ps. The pulse detected at zero delay is the scattered readout pulse. (a) Hologram after 30 s writing exposure when the sample was simultaneously illuminated with 180 mW/cm^2 average intensity gating beam; (b) after a similar hologram writing exposure but with the gating beam blocked.

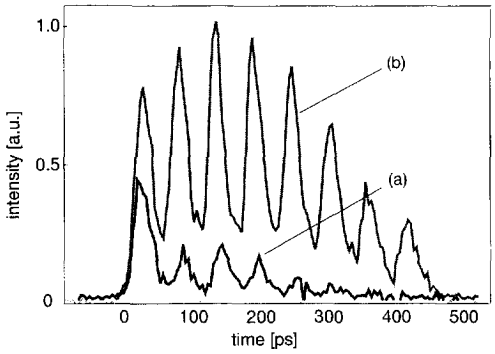

Fig. 4. Comparison of permanent (a) and transient (b) SHB hologram signals obtained when the object was a train of picosecond pulses with 60 ps interval. The gated SHB hologram writing exposure time was 30 s with total illumination intensity in the reference and object beam of 1 mW/cm^2. The intensity of the gating light was 100 mW/cm^2. Triplet-accumulated hologram signal was detected with the same power of the writing beams and with the gating beam blocked.

In fact, the SHB efficiency without the gating illumination was low enough to allow us to read out the time-and-space-domain hologram for about 40 min with a reading beam intensity of 0.85 mW/cm^2. The total reading fluence required for a 50% hologram reduction was about 2 J/cm^2.

In our experiment we also compared the diffraction efficiency of an optimally recorded permanent gated hologram with that from a transient triplet-accumulated hologram. Fig. 4 shows a signal diffracted from a permanent SHB hologram (a) and a transient triplet-accumulated hologram (b) under comparable experimental conditions. Pulse trains occur in this experiment because in the writing object beam we had placed a Fabry–Perot etalon to simulate a train of picosecond data pulses. We observed that the highest relative diffraction efficiency of transient holograms was about 1%, while the gated permanent holograms had a relative efficiency of about 0.1%. We can explain the low efficiency of the gated holograms if we assume that in our material only a fraction of the ZnTBP molecules in resonance with the laser illumination can be permanently burned. The chromophores that cannot be permanently bleached will also not contribute to the formation of the diffraction grating, while in the transient holo-

gram storage there is no such limitation and all resonant zero-phonon lines can contribute to the diffraction. It is possible that in our SHB material the concentration of the activator molecules is not high enough to have on the average an efficient electron transfer from all excited chromophore molecules to an acceptor. By assuming that the diffraction efficiency is proportional to the contrast of the recorded spectral-and-space-domain grating, we can estimate that the fraction of the ZnTBP chromophores that can be permanently burned away is about 10–20%. In fact, we observed that a considerable fraction of the chromophores were not bleached away even after an extended illumination of the sample simultaneously with λ_1 and λ_2. Our material may actually have a broad distribution of different permanent burning efficiencies depending on the relative position of the activator molecules and the chromophore molecules in the matrix.

The absorption of the ZnTBP chromophore at the gating illumination wavelength corresponds to an optical density of the sample < 0.1. Although the absorption at λ_2 was small, we observed that by increasing the gating illumination intensity there was a shortening of the optical dephasing time of the ZnTBP chromophores zero-phonon transition. The dephasing time T_2 was estimated by varying the delay between the object and the reference and by monitoring the intensity of the transient triplet-accumulated echo signal. Without λ_2 illumination or with the λ_2 intensity under 10 mW/cm^2, we obtained $T_2 \approx 500 \pm 50$ ps, while at higher gating light intensity the dephasing was faster. A manifestation of this effect can be seen in Fig. 4 where the signal pulse train corresponding to the triplet-accumulated hologram is almost by a factor two longer than the pulse train reproduced from the gated permanent hologram. We note that the shorter dephasing time can be caused by a temperature increase due to the absorption of λ_2 in the presence of λ_1.

4. Conclusions

By using an organic SHB material with both improved optical quality and higher hole-burning efficiency we have demonstrated the storage of photon-gated coherent transients. By using this technique we have produced picosecond time-and-space-domain holograms that can be read out without destruction of the original recorded information.

On the other hand, our results suggest that in the current SHB material the relative diffraction efficiency of the photon-gated holograms is restrained to about 0.1%. We can explain this behavior if we assume that the majority of the ZnTBP molecules does not have in the polymer matrix enough closely located activator molecules to facilitate permanent bleaching. Since only 10–20% of the chromophores can actually undergo permanent hole burning, the contrast of the hologram gratings remains limited and further work is needed to improve the photon-gated SHB system.

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