Nondestructive Read-out of Two-color Photon-gated Spectral Holeburning Holograms

DANIEL REISS, ALEXANDER REBANE, URS P.WILD
Physical Chemistry Laboratory, Swiss Federal Institute of Technology,
ETH Zentrum, CH - 8092 Zürich, Switzerland

We recorded picosecond photochemically accumulated stimulated photon echo (PASPE) holograms by using two-color photon-gated persistent spectral hole-burning at liquid-helium temperature. Images stored using this technique have more than three orders of magnitude higher read-out stability compared with the one-color process. We used Zn-tetra-phenyl-tetrabenzoporphyrin in a polystyrene film with 20% hexachloroethan (C_2Cl_6) as recording material. The long lifetime of the triplet state allows the observation of a transient image prior to the fixation of the image. By restricting the green gating beam to a part of the transient hologram image on the polymer film, the photoreaction takes place only in the area illuminated with both colors, and this part of the image is stored permanently.

INTRODUCTION

Spectral hole burning (SHB) relies 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. Persistent SHB shows a potential to be an extremely high density frequency domain optical storage technique [1].

Typical SHB materials 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. SHB then occurs with a quantum yield (i.e. number of photo-transformed chromophores per one ab-

sorbed photon) which is independent from the intensity of the illumination. Because of 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 optical storage applications this characteristic is a serious drawback because each read-out of the data inevitably causes an erasure of a part of the recorded information.

The first examples of photon gating utilized the mechanism of two-step photoionisation of the absorbing molecule or ion and subsequent trapping of the ejected electron in the nearby host matrix [2]. A mechanistic investigation of photoinduced donor-acceptor electron transfer (DA-ET) was reported by Carter et al. [3] and it was concluded that electron transfer occurs from the highly excited triplet molecule to the halocarbon acceptor. The dependence on the variation of different organic halocarbon additives is shown in [4]. In a number of papers, photon-gated hole-burning in different organic and inorganic materials has been studied [1]. All these experiments used transmission or absorption to detect the holes. Holographic techniques have important advantages for use in data storage, but they require a much higher optical quality of the samples[5].

In this paper we report the nondestructive read-out of holographic images recorded by locally controlled two-color photon-gated spectral holeburning.

Experimental

The optical density of the sample at the maximum of the S_1 - S_0 absorption of ZnTPTBP was 1.3 at room temperature. During the whole experiment the sample was kept in superfluid helium below 2K.

The light source used was a 76 MHz-repetition-rate picosecond dye laser synchronously pumped by a Coherent Antares 76s frequency-doubled Nd:YAG laser. The dye laser delivered pulses with a duration of 4-5 ps, a spectral width of 0.56 nm and an average power of 300 mW. A standard holographic setup was used: the dye laser output was attenuated, divided and afterwards expanded with two telescopes into two plane wave beams. The reference beam was applied at

normal incidence with respect to the SHB film. The object beam (with transmission mask) was applied in at an angle $\theta=8^0$ with respect to the reference. The di-
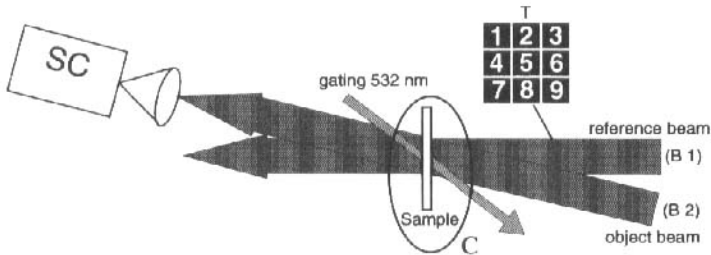

FIGURE 1   Schematic setup for local controlled two color photon gated image storage: The parallel object beam (B 2) was let through a transmission mask (T) before hitting the sample in the cryostat (C) with the 100 ps delayed reference beam (B 1). To fix the transient holographic grating, the green beam has to illuminate the sample for some seconds simultaneously with B 1 and B 2. During read-out only B 1 hits the sample and the stored image is detected by the camera (SC).

ameter of the spot illuminated with both writing beams at the sample was 8 mm, the reference beam ($60\,\mu W\,/cm^2$) was delayed by 100 ps with respect to the object beam ($140\,\mu W\,/cm^2$). 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 ZnTPTBP in the range λ_1=630 - 645 nm. Because of the long lifetime of the triplet state, a transient holographic grating forms, which was observed after closing the object beam by use of the high resolution camera (Sony SSC-M370CE). By repeating this cycle of building a transient grating and looking at the deflected hologram image at rates of at 2 to 5 Hz we could optimize the optical setup in real time (see image (A) in figure 2).

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 directed to the SHB film from the opposite side with respect to the two hologram writing beams. The diameter of the spot illuminated on the SHB sample by the gating beam was 2 mm and it covered the area of one

number of the transmission mask. The image storage experiments were done with an intensity of the gating beam between 150 to 600 mW/cm^2.

Results and Discussion

The simultaneous illumination of the transient hologram with the green gating beam for approximately 3 s on the "1" fixes the image (see image (B) in figure 2). By moving the gating beam over the sample the whole image of the transmission mask was stored (image (C) in figure 2).
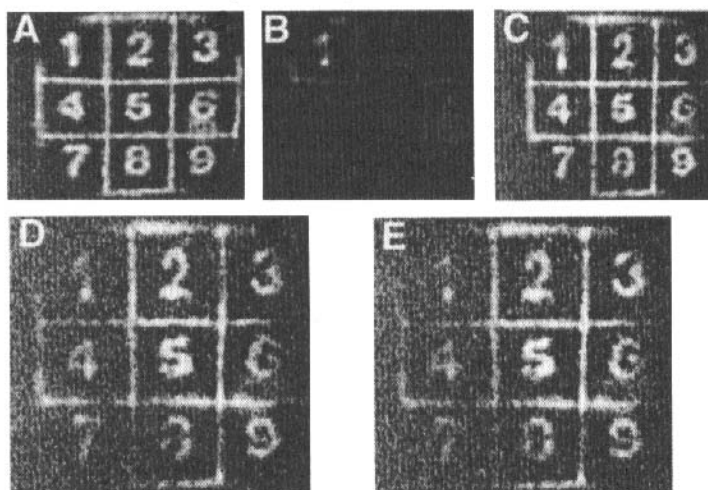

FIGURE 2   Local controlled image storage: Because of the long lifetime of the triplet state, a transient holographic grating is formed. By closing the object beam, a stored image may be observed (A). To permanently store an image, the transient grating has to be illuminated with an additional green gating beam for some seconds. This procedure was performed with only the "1" illuminated by the green beam (B). Then all the numbers were illuminated (C).
Nondestructive read-out: The numbers 1, 4 and 7 were read with 6000 (D) and 12000 (E) times the burning energy. The signal intensity of this area is decreased, but the numbers are still recognizable.

During the following read-out only the reference beam illuminated the area

of the numbers on the left side (1,4,7) for 7200 s with 10 times the burning intensity (600 μW /cm^2), the other numbers were shielded. Every 5 minutes the reference beam was attenuated to the writing power and the signal intensities of the whole image was analyzed (see figure 2 images (D) and (E) after 3600s and 7200s continuous reading respectively). The degeneration of the image quality may be clearly seen. All numbers are still recognizable however, even after read-out with about 10000 times the writing energy. This is a dramatic improvement on the results obtained using one-color holeburning[6]. For better signal visibility the pictures were treated with histogram equalization.

To quantify the difference between one color and two color holeburning, the numbers in the transition mask were replaced by circles. A one-color hologram was stored for 6 s. The read-out signal was fitted with a single exponential (see figure 3, left) which gave a decay time. τ was in the order of 2 times the writ-
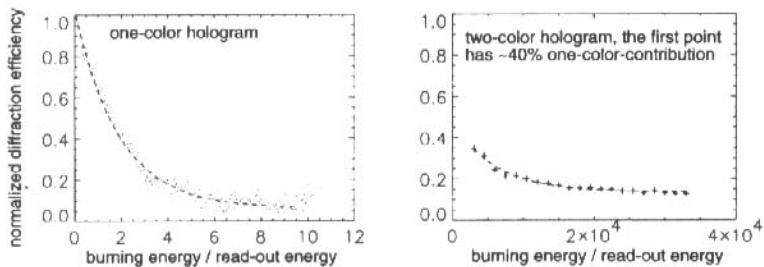

FIGURE 3 Read-out stability of one- and two-color images. Holograms stored with one color undergo a fast bleaching ($\tau \sim 2$ of the writing energy, left), while the two-color photon gated hologram decays much more slowly ($\tau \sim 6000$, right). Note the different scales of the figures and that the first point of the two-color hologram contains ~40% signal intensity of the one-color hologram.

ing energy. A two-color photon-gated image was stored under the same conditions (6 s simultaneous two color illumination). The hologram was read out with 10 times the writing intensity in the reference beam. The decay curve of this read-out gives us a time constant of about 6000 times the writing energy (see fig-

ure 3, right; fitted without the first point, discussion see below).

The read-out stability of the two-color photon gated holograms consists of at least of two different processes. We observed in the first seconds a fast decrease of the signal which can be described by a one-color photoreaction (~ 40% contribution).

Conclusions

We have demonstrated nondestructive read-out of time and space domain holograms stored by area selective two-color persistent spectral holeburning. The holographic technique allows a background free detection of the images. From the comparison of the holographic signal, and the corresponding transmission signal, the advantage of this technique may be estimated. Investigation of the photoreaction would help optimize the choice of dye, electron acceptor and host materials.

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