PICOSECOND PULSE SHAPING BY PHOTOCHEMICAL TIME-DOMAIN HOLOGRAPHY

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Received 21 July 1983

Very high relative intensity (≥30%) coherent optical free-decay signals can be stimulated by weak picosecond excitation of persistent spectral holograms photochemically burned in inhomogeneously broadened impurity absorption bands. The feasibility of phase relaxation-time measurement as well as time-reversal of picosecond optical signals is demonstrated.

1. Introduction

Photochemical hole burning (PHB) [1,2] is widely used as a spectroscopic method for the investigation of the homogeneous spectral characteristics of impurity molecules in the presence of large inhomogeneous broadening [3], but it has also found several technical applications, such as in the development of optical memory devices with high storage density [4,5].

We have recently shown [6,7] that it is possible to perform time-domain holography of picosecond events by PHB, i.e. to fix, store and reproduce temporal shapes of coherent picosecond optical signals.

To explain the main principles of time-domain holography, it must be pointed out that the traditional PHB methods consist of two distinct stages: first, spectral "holes" are burned by monochromatic excitation of inhomogeneously broadened absorption bands of photochemically unstable impurity molecules, and secondly, these "holes" are detected by fine scanning of the excitation over the modified inhomogeneous spectrum.

Now let us suppose that, instead of monochromatic excitation, PHB is carried out by coherent sequences of picosecond pulses with the spectrum width Δ much larger than the homogeneous widths of impurity absorption lines (the homogeneous widths of purely electronic lines are 10^{-3} cm^{-1} and less at low temperatures [8]) but still less than the width of the inhomogeneous broadening ρ. One can see [9] that if $\rho \gg \Delta$ and $T_2 \gg \tau$ (T_2 is the impurity-molecule phase relaxation time and τ is the overall duration of the applied pulse sequence) the shape of the resulting photochemical hole appearing in the sample absorption spectrum will correspond to the fine Fourier spectrum of the pulse sequence.

If a sample with such a burned-in spectral hologram is consequently probed by a weak single picosecond pulse, the temporal shape of the transmitted pulse will, in agreement with general rules of linear filtration, "play back" the sequence of pulses fixed in the sample transparency spectrum.

To perform time-domain holography of picosecond optical signals, a coherent reference pulse should be added, in the same way as is done in ordinary space-domain holography.

The aim of the present paper is to demonstrate some developments of photochemical time-domain holography, particularly the controlled shaping of picosecond pulses.

2. Experimental

In our experiments * we utilized a synchronously pumped rhodamine-6G picosecond dye laser, which generated at a rate of 82 MHz 2–3 ps duration pulses (5.5 cm^{-1} spectral fwhm), and a synchroscan streak camera system [10,11] which provided 20 ps (fwhm) temporal resolution.

* For further details of the experimental apparatus see ref. [7].

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For photochemical burning of spectral holograms, the laser beam was directed through either a Fabry–Pérot or a Michelson-type interferometer, which transformed the input single picosecond pulses into suitable coherent pulse sequences. The interferometers' output beams were collinearly directed on a sample positioned in a 1.8 K helium cryostat with pass-through windows.

The transmitted laser beam along with the signal stimulated from spectral holograms was focused on the entrance slit of the streak camera, and to detect the temporal shapes of these signals the interferometers were replaced by a pack of neutral filters attenuating the incident picosecond laser beam to minimize destructive hole burning during measurement cycles.

The sample was prepared by polymerization of an octaethylporphin 2.5×10^{-4} M styrol solution.

3. Results and discussion

First, to demonstrate the pulse-shaping effect of spectral holograms, the picosecond laser beam was passed through a 12 mm basis Fabry–Pérot etalon, which produced an exponentially decreasing sequence of picosecond pulses with a fixed 80 ps delay between them (see fig. 1). An optimum exposure spectral hologram burned by such pulse sequences and probed by single laser pulses is seen to irradiate several retarded "echo" pulses which reproduce exactly the applied PHB pulse sequence. Note that the nearly 30% relative intensity of the first "echo" pulse (fig. 1d) is obtained

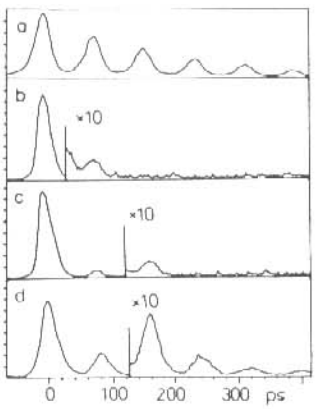

Fig. 1. Streak-camera images of the applied PHB sequence (a) and of the resulting "echo" signals after 0.5 mJ/cm^2 (b), 1.5 mJ/cm^2 (c) and 2.5 mJ/cm^2 (d) PHB exposures.

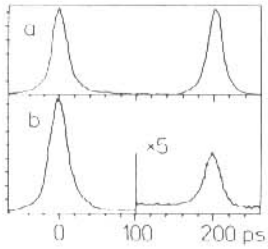

Fig. 2. Streak-camera images of twin burning pulses (a) and of the probing pulse passed through along with the retarded "echo" (b).

without any attenuation of the probing pulse passed through.

Secondly, to show that by burning and probing of spectral holograms it is also possible to find the phase relaxation time T_2, sequences of twin collinear burning pulses (peak intensity 10^{-1} W/cm^2) with a variable delay τ were produced by splitting of the laser beam in a Michelson-type interferometer. If we consider that the intensity of the "echo" signal falls off as $\exp(-4\tau/T_2)$ [12], T_2 can be calculated from the dependence of the "echo" pulse intensity on the delay τ *. We estimated T_2 to be about 300 ps in our experiment (fig. 2).

Lastly, to show the possibility of time-reversal of picosecond optical signals, we inserted an additional 1 mm basis etalon in the shorter arm of our Michelson-type interferometer. This resulted in a rather complicated pulse sequence consisting of a rapidly decreasing pulse train and of a retarded single pulse (fig. 3a) serving as a reference for the non-symmetric signal pulse train. It was expected that every individual pulse from the signal pulse train combined with the reference pulse would give rise to an "echo" with a delay and intensity corresponding to the position of the pulse in the signal train, which, in turn, would result in a time-reversed replica of the whole signal. Results shown in fig. 3b confirm that we were indeed able to detect the time-reversed signal, but the mechanical instability of the interferometer did not allow us to obtain a better signal-to-noise ratio.

To discuss these experimental results, it should be noted that the simple considerations presented in sec-

* In the intervals between PHB with different τ, the initial inhomogeneous bandshape should be restored (for example, by illumination of the sample with white light).

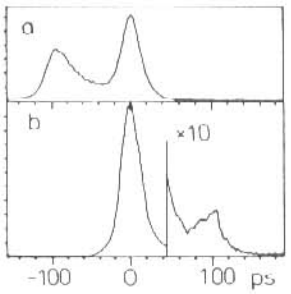

Fig. 3. Streak images of the burning sequence consisting of signal and reference pulses (a) and of the time-reversed "echo" of the signal pulse (b).

tion 1, based on the linear filtration theorem, provide no implications for the origin of the processes responsible for the formation and read-out of spectral hologram.

As we have already pointed out in our previous work [7,9], the formation of spectral holograms can be more properly treated as an interference of the trailing pulses of PHB sequences with the coherent excitation produced in the matter by preceding parts of the same picosecond sequence.

The origin of the echoes lies in turn in the coherent spontaneous emission of a coherently excited molecular ensemble with a specially prepared inhomogeneous distribution of transition frequencies.

If we also consider that the duration of both stored and reproduced picosecond pulse sequences is limited by the phase relaxation time T_2 of impurity molecules, then the time-domain holography along with the observed pulse-shaping effects may be regarded as a new modification of optical free-decay transients, closely related to the phenomenon of three-pulse stimulated photon echoes [13,14]. While the stimulated photon echo can occur only within the relaxation time of induced frequency-space population gratings, in our experiments the lifetime of free-decay "echoes" is determined by the lifetime of PHB photoproducts (usually estimated to be several days or even months [15]).

The extremely long population relaxation time allows the frequency space gratings or holograms to be accumulated to a very high contrast simply by repeating the PHB procedure many times ($\approx 10^{10}$ in our case) with low-intensity picosecond pulses.

Acknowledgement

The authors are indebted to K. Rebane and P. Saari for useful discussion of the paper and to A. Freiberg, A. Anijalg and K. Timpmann for cooperation in the streak camera performance. We also wish to thank J. Kikas for providing the samples and for discussing the PHB problems.

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