PHOTOCHEMICAL TIME-DOMAIN HOLOGRAPHY OF WEAK PICOSECOND PULSES

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We show that weak picosecond optical pulse propagation through an absorbing medium with a photochemically burned-in persistent spectral hologram of a picosecond pulse train makes the sample emit coherently a replica of the pulse train applied in the burning-in cycle.

1. Introduction

Photochemical hole-burning (PHB) [1,2] has proved to be a very useful method in low-temperature spectroscopy providing effective ways to get rid of inhomogeneous broadening [3,4]. This method has also opened new possibilities for technical applications, for example in developing optical memory devices with high storage density [5,6].

The starting-point of stationary PHB techniques, commonly used both in research and applications, lies in burning and detecting of photochemical holes by means of CW radiation. The homogeneous widths of purely electronic lines (PEL) in impurity centres at low temperatures are very small (10^{-3}–10^{-4} cm^{-1} and less [7]) and the usual aim in PHB experiments is to have the excitation linewidths as narrow as possible. Therefore the characteristic times of excitation are usually much longer than the phase relaxation time T_2 of the impurity centre.

On the other hand, PHB with light pulses short compared to the relaxation time T_2 and with a spectrum much broader than the homogeneous PEL-width, can also be utilized for research and application purposes. As shown experimentally in a recent work of three of us [8], under conditions where inhomogeneous broadening is much larger than the width of the excitation spectrum, the PHB by means of a sequence of pairs of coherent relatively low-intensity picosecond pulses shifted in time by τ ($\tau \ll T_2$) results in a persistent photochemical hole with a complex spectral shape. The envelope of the hole reflects the spectrum of an individual pulse, while the hologram-like substructure appears due to the interference of the second pulse with the excitation produced in the matter by the first pulse.

Now let us ask: what is the effect of a δ-like light pulse propagating through a medium with such a persistent hologram-like transparency spectrum?

To answer this question in the case of strong excitation, one must solve a complex problem of coupled Maxwell—Bloch equations, which describe such nonlinear processes as free induction decay, stimulated echo, etc..

In the limit of weak excitation the answer can be derived from the general rules of linear filtration by postulating that the output pulse shape is determined by the Fourier transform of the transparency spectrum.

So it may be supposed that if a spectral hologram, produced by a sequence composed from a number of pulses, is "scruned" with a weak picosecond pulse, the free-decay signal of the excitation should also have a form of several retarded spontaneous bursts of coherent radiation, as in the case of stimulated photon echo.

In order to prove this simple consideration we arranged an experiment, in which we burned a persistent photochemical spectral hologram and subsequently probed its pulse response to weak picosecond pulses.

2. Experimental

The scheme of the set-up is presented in fig. 1. The photochemical holograms were burned and probed by means of a picosecond rhodamine 6G dye laser synchronously pumped by an actively mode-locked Spectra-Physics model 171 argon-ion laser. The picosecond dye laser provided 82 MHz repetition rate pulses with 2–3 ps duration and 5.5 cm^{-1} spectral FWHM. For spectral measurements we used a 0.075 cm^{-1} FWHM and a 12 cm^{-1} tuning range laser line delivered by the same dye laser readjusted to CW regime and by scanning fine tuning etalons inserted in its cavity. A broader spectral region of the sample transparency was measured with a glow lamp and a MDR-4 monochromator. The spectra were recorded with a conventional photon-counting system.

The temporal analysis of the radiation was performed by focusing the beam on the entrance slit of a synchroscan streak camera system [9,10], which provided temporal resolution better than 20 ps (FWHM).

The sample, prepared by polymerization of a porphyrazine [11] styrol solution, was embedded in a He cryostat at 1.8 K. The mean intensity delivered on the sample during burning and probing cycles was 0.1 mW/cm^2 and 0.1 μW/cm^2, respectively. For generating a coherent sequence of burning pulses a 12 mm basis Fabry–Perot etalon was inserted in the optical path of the burning beam. During the experiment four equal 5.0 mJ/cm^2 dose exposures were made. After each exposure the Fabry–Perot etalon was removed and the beam was attenuated with neutral filters 10^3 times to avoid the destruction of spectral holograms during streak camera measurements.

3. Results and discussion

In fig. 2 the transparency spectrum of a photochemical hologram produced by the picosecond pulse train generated in the Fabry–Perot etalon from the initial picosecond laser pulse is presented. The envelope of the spectrum refers to the spectrum of a single pulse entering the Fabry–Perot etalon, while the observable sharp features appear to be due to interference between the pulses merging from the etalon.

The high contrast of the spectral hologram arises from the accumulation of the PHB effects, produced by nearly 10^{10} identical pulse trains delivered during the exposure at 82 MHz repetition rate on the sample.

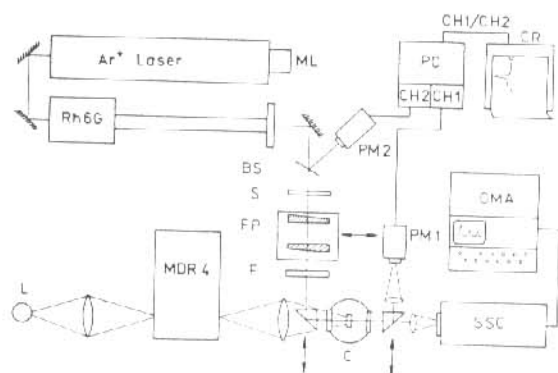The temporal response of the hologram, as it ap-

Fig. 1. Scheme of the experimental arrangement. ML – mode locker, PM – photomultiplier, BS – beam splitter, S – shutter, FP – Fabry–Perot etalon, F – filter, L – glow lamp, C – He-cryostat, SSC – synchroscan streak camera, OMA – optical multichannel analyser, PC – photon counter, CR – chart recorder.
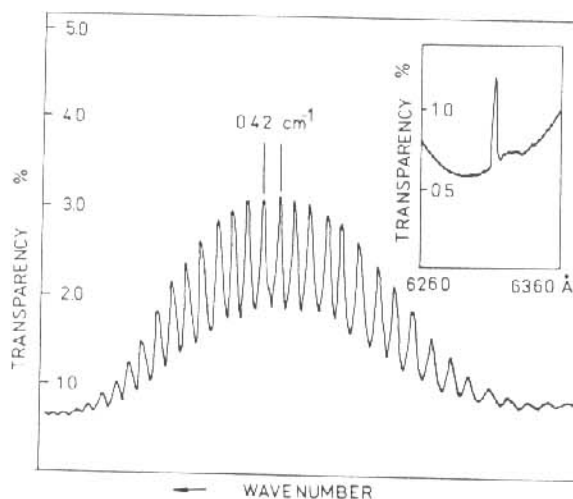

Fig. 2. Transparency spectrum of the sample after the fourth exposure. Insert – burned-in photochemical hole and its pseudo-photon-sideband; main frame – fine structure of the hole, measured by scanning of the dye laser line with FWHM = 0.075 cm^{-1}.
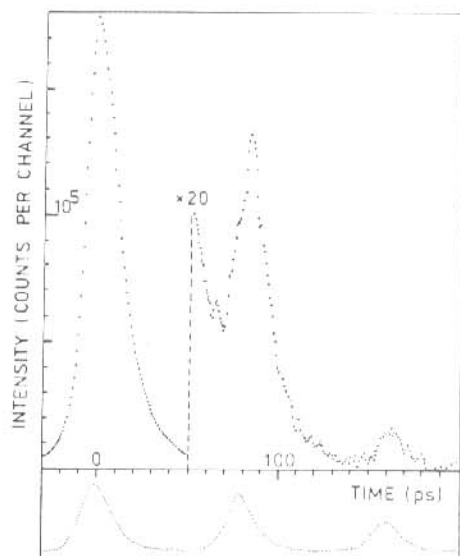
Fig. 3. The streak image of the picosecond laser beam passed through the spectral hologram after the last exposure. Upper left is the passed-through laser pulse, while the two pulses on the right of it are the "echo" pulses produced by free decay of excitation. For actual time reference the streak image of the first three pulses from the coherent burning sequence is also presented in the lower part of the figure.

pears on the streak camera image, is presented in fig. 3. It is clearly seen that the passed-through laser pulse is followed by two additional retarded bursts of radiation. The delays of these "echo" pulses are 80 ps and 160 ps, which correspond exactly to the delay produced in the 12 mm Fabry—Perot etalon.

From our viewpoint these experimental results prove the simple consideration that even in the limit of weak excitation any spectral modulation of transparency gives also rise to the modulation of the temporal response function.

The fact that the observed pulse response of the hologram does not correspond exactly to that of the Fabry—Perot etalon can be qualitatively explained by the effect of saturation broadening of photochemical holes, which leads to the deformation of the PHB spectrum [12] *. On the other hand it should also be taken into account that to carry out a complete spectral holography of the pulse train, a still shorter δ-like reference pulse with a broad uniform spectrum must be available.

It is interesting to point out that we noticed no changes in the hologram during the experiment and so the lifetime of the hologram, determined by that of the photoproduct, could be several hours or even days [3,13].

In order to discuss the applications of the photochemical holography it will be useful to compare it to some aspects of the recently published theory of time-domain optical data storage [14], where the possibility of PHB with short optical pulses was also mentioned. But as this theory requires nearly 10^5 times more intense excitation pulses than those applied in our experiment, it does not correspond to the weak excitation case of the general problem raised in section 1. This circumstance brings it close to the recently reported effect of correlation of the shapes of single nanosecond and picosecond pulses in stimulated photon echo experiments [15,16], rather than to the phenomenon discussed in this paper.

Nevertheless, as far as the conditions * $\rho \gg \Delta$ and $T_2 \gg \tau$ are satisfied, all the suggested applications of the time-domain optical data storage hold also for the photochemical holography of weak pulses. Even more the application of subpicosecond pulses, which is possible due to the large inhomogeneous broadening (≈ 500 cm^{-1}), will result in $10^3 - 10^2$ bits per nanosecond data read-out speed, which is three orders of magnitude more than in the mentioned theory. The high contrast of spectral holograms, created in our case by the accumulation of the PHB effect, results in a high-contrast data read-out free-decay signal, which in the far-from-ideal conditions of our experiment was 4%, i.e. an order of magnitude higher than that offered by the most optimistic theoretical predictions in ref. [14].

In conclusion, it should be said that besides optical data storage the photochemical holography of weak pulses may be applied for many other purposes, for example, for phase conjugation of laser pulses, for monitoring picosecond laser operation, and also for direct time-domain investigation of ultrafast phase relaxation processes.

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* A full quantitative theory of the photochemical time-domain holography will be published elsewhere.

* ρ is the width of the inhomogeneous broadening, Δ is the maximum spectral width of data pulses, and τ is the duration of the data writing pulse train.

Acknowledgement

References

[1] A.A. Gorokhovskij, R.K. Kaarli and L.A. Rebane, JETP Lett. 20 (1974) 474.

[2] B.M. Kharlamov, R.I. Personov and L.A. Bykovskaya, Optics Comm. 12 (1974) 191.

[3] L.A. Rebane, A.A. Gorokhovskij and J.V. Kikas, Appl. Phys. B29 (1982) 235.

[4] R.K. Kaarli, A.K. Rebane and P.M. Saari, Eesti NSV Tead. Akad. Toimet. Füüuus. Matem., to be published.

[5] G. Castro, D. Haarer, R. Macfarlane and R. Trommsdorff, U.S. Patent 4,103,346 (1978).

[6] K.K. Rebane, Laser Study of Inhomogeneous Spectra of Molecules in Solids, Proceedings of Conference "Lasers-82", New Orleans, USA, Dec. 1982.

[7] K.K. Rebane, Impurity spectra of solids (Plenum Press, New York, 1970).

[8] A.K. Rebane, R.K. Kaarli and P.M. Saari, Opt. Spectry., to be published.

[9] A. Freiberg, A. Raidaru, A. Anijalg, K. Timpmann, P. Kukk and P. Saari, Eesti NSV Tead. Akad. Toimet. Füüs. Matem. 29 (1980) 187.

[10] A. Anijalg, A. Freiberg, R. Kaarli, P. Kukk, P. Saari and K. Timpmann, Proceedings of the Second International Symposium UPS-80, Oct. 30—Nov. 5, 1980, Reinhardsbrunn, GDR.

[11] V.N. Kopranenkov, L.S. Gonchareva and E.A. Lukyanets, Zh. Obschey Khim. 47 (1977) 2143.

[12] A.A. Gorokhovskij and J.V. Kikas, Zh. Prikl. Spektrosk. 28 (1978) 832.

[13] A.A. Gorokhovskij, Opt. Spectry. 40 (1976) 477.

[14] T.W. Mossberg, Optics Lett. 7 (1982) 77.

[15] G.O. Eljutin, S.M. Zakharov and E.A. Manykin, Opt. Spectry. 52 (1982) 577.

[16] V.A. Zuikov, V.V. Samartsev and R.G. Usmanov, Pis'ma Zh. Eskp. Teor. Fiz. 32 (1980) 293.