Dynamic picosecond holography produced by means of photochemical hole burning

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A train of picosecond laser pulses is recorded and read by burning out a spectral hologram photochemically and then probing its temporal response by weak picosecond pulses.

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1. In holography the spatial-angular characteristics of fields are recorded on standard photographic film. In this paper we demonstrate that if the medium recording the interferogram is a highly selective photochromic medium, then it is also possible to record and reproduce spectral-temporal characteristics of the radiation.

Such media include low-temperature matrices with impurity molecules, which are suitable for photochemical hole burning (with typical width 10^{-3} cm^{-1} with monochromatic irradiation) in their inhomogeneously broadened absorption spectrum.[1–3]

If a train of picosecond pulses is focused on such a medium, a grating of narrow transmission bands appears in its spectrum with a spacing $\Delta\nu = 1/\tau$, where τ is the distance between the pulses in the train,[4] i.e., a spectral hologram is recorded in the form of a Fourier transform of the temporal form of the train. When a single picosecond pulse is transmitted through such a hologram functioning as a spectral filter, the spectrum of this ("reading") pulse is transformed, leading to the appearance of repeated pulses with spacing τ in the response.

2. A polystyrene matrix with a porphyrazine additive at 1.8 K (inhomogeneous broadening ≈ 500 cm^{-1}) was used in the experiment as the photochromic medium. The holograms were burned out by irradiating the specimen with a train of picosecond pulses (peak intensity 10^{-1} W/cm^2, pulse duration 2–3 ps, pulse spacing in the train 80 ps, and repetition frequency of the trains 82 MHz), obtained by multiplying pulses from a synchronously pumped picosecond rhodamine-6G dye laser.

Figure 1 shows the spectral hologram.[1)]

The temporal response of the hologram was investigated with the help of an electro-optical converter (EOC) with synchronous scanning.[5] The intensity of the light transmitted through the specimen, when single laser pulses weakened by a factor of 10^5 were incident on it, was unfolded in time on the screen of this converter.

It is evident in Fig. 2 that an "echo" in the form of two additional pulses of directed radiation, delayed by 80 and 160 ps, respectively, appear in the temporal response of the specimen, after the spectral hologram is burned out with sufficient contrast, i.e., the recorded picosecond signal is reproduced.

3. To clarify the relation between the phenomenon being examined and previously

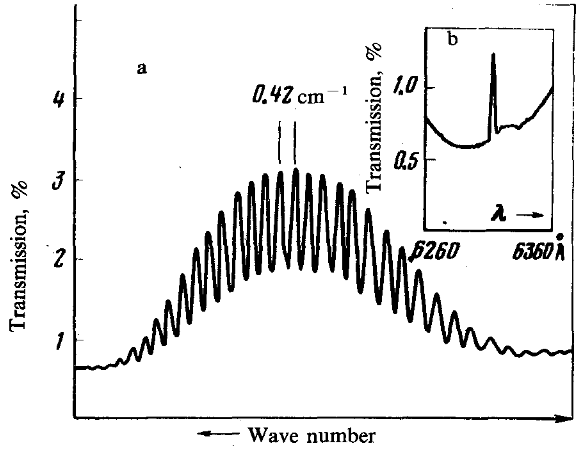

FIG. 1. Transmission spectrum of specimen after exposure to a train of picosecond pulses with total energy density 20 mJ/cm^2. a) Fine structure of hole, arising via absorption in no-phonon transitions of impurity molecules, measured by scanning of laser line with half-width 0.075 cm^{-1}; the distance between the peaks is equal to the inverse magnitude of the interval between the pulses in the train; b) the general form of the region of the hole measured with a resolution of 1 Å.

known phenomena, as well as to clarify the possibilities of using it in the holography of signals of arbitrary form, we point out the following facts. First, the spectral fine structure of the hole can be viewed just as the result of interference: the coherent excitation of molecules created by the first (reference) pulse interacts with the subsequent ("objective" or "code") part of the radiation. Second, the additional pulses in the time response of the hologram physically represent spontaneous coherent emission of an ensemble of coherently excited dipoles with a specially prepared inhomogeneous distribution of transition frequencies. It follows from here that a) the duration of the recorded and reproduced signals is limited by the phase relaxation times $T_2 \approx 10^{-9}$ s of the molecules and b) the phenomenon can be interpreted as a new, intrinsic to highly selective photochromic media, modification of the (stimulated) light echo[2)] or photochemically accumulated stimulated light echo (PASLE).

The differences between the phenomena stem from differences in the mechanisms of depletion and characteristic regeneration times of the ground state of the absorbers (for the stimulated echo, the actual lifetime T_1 of the excited state of molecules; for PASLE, the lifetime of the photoproducts $T_\Phi \gtrsim 10$ hours and possibly years). We see a number of advantageous properties of PASLE: 1) it is easy to accumulate the interference pattern in the medium up to high contrast, even in the case of weak pulses, by repeating their recording many times (by a factor of 10^{10} in the given experiment) over the course of a long time; 2) the recording and reading processes of the picosecond events are separable: the formation of PASLE reduces to linear filtering of the reading pulse, whose power, time of incidence, and even repetition do not have a significant effect on the relative intensity of the response pulses.

4. To estimate the prospects for using the phenomenon discovered in creating a fast-acting holographic memory,[6] devices for inverting the wave front, and so on, it is

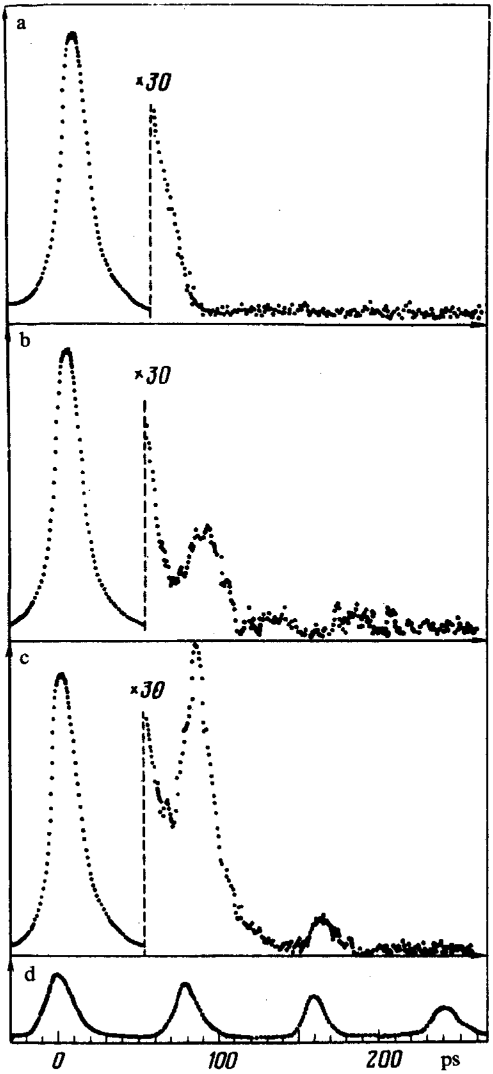

FIG. 2. Temporal response of specimen to picosecond pulse: a) before burning; b) after exposure 10 mJ/cm^2; c) after exposure 20 mJ/cm^2. The curve *d*, shown for comparison, is an image of part of the train of burning pulses. The visible half-width of the pulses (20 ps) is due to the resolution of the EOC.

important to emphasize that PASLE pulses with relative intensity of the order of several percent (see Fig. 2) with an unprecedented small, for experiments involving the photon echo, energy of reading pulses 10^{-12} J/cm^2 = 10^2 photons/cm^2 have already been obtained in the first experimental results.

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[1)]The spatial aspect of the problem is not examined in this work: The rays are collinear with the field distribution over the cross section which is standared for a laser.

[2)]The possibilities of the light echo in ultrafast holography of spatial-temporal events have been pointed out in papers by outstanding specialists in these fields (see, for example, Refs. 6–8).

[1]A. A. Gorokhovskiĭ, R. K. Kaarli, and L. A. Rebane, Pis'ma Zh. Eksp. Teor. Fiz. **20**, 474 (1974) [JETP Lett. 20, 216 (1974)].

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

[3]L. A. Rebane, A. A. Gorokhovskij, and J. V. Kikas, Appl. Phys. B **129**, 235 (1982).

[4]A. K. Rebane, R. K. Kaarli, and P. M. Saari, Opt. Spektrosk. 55 (1983) [in press].

[5]A. Freĭberg, A. Raĭdaru, A. Aniyalg, and K. Timpmann, Izv. Akad. Nauk Est. SSR, Ser. Fiz. -Matem. **29**, 187 (1980).

[6]Yu. N. Denisyuk, "Holography and its prospects," in: Problemy opticheskoĭ golografii (Problems in Optical Holography), Ed. by Yu. N. Denisyuk, Nauka, Lenigrad, 1981, pp. 7–27.

[7]V. A. Zuĭkov, V. V. Samartsev, and R. G. Usmanov, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 293 (1980) [JETP Lett. **32**, 293 (1980)].

[8]S. O. Elyutin, S. M. Zakharov, and É. A. Manykin, Opt. Spektrosk. **52**, 577 (1982) [Opt. Spectrosc. (USSR) **52**, 345 (1982)].

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