K. K. Rebane. *Space-time holography of ultrafast events through spectral hole burning.* The photoburning of stable holes in the spectra of impurities in solid matrices has proved an effective tool for high-resolution spectroscopy. Research is being carried out on atoms, ions, and small and large molecules (including chlorophyll and its analogs) in single-crystal, polycrystalline, glass, and polymer matrices and also on the structure of these matrices and the processes which occur in them at ultralow temperatures, including the properties of two-level systems and possible manifestations of a fractal structure of glasses and polymer films (see the original papers,[1,2] the reviews,[3–6] etc.[7,8]). Spectral hole burning, however, has applications beyond spectroscopy. It is also a highly selective tool for acting on the spectral characteristics of a substance: on the absorption spectrum and on the spectrum of the refractive index, which is related to the absorption. In addition to the new possibilities for studying substances, there is a new opportunity to develop some devices of practical importance—extremely narrow-band optical filters and spectral memories for computers[8,9]—and to develop a holography of ultrafast space-time events in the pico- and nanosecond ranges.[7,10,12] This new field of holography is based on the possibility of making use of spectral hole burning to fabricate stable spectral gratings in a substance and also on the phenomenon of a photochemically accumulated stimulated photon echo.[12,13] The optical fields required here are not intense; the contrast of the spatial-spectral gratings in a substance depends on the illumination dose, rather than on the light intensity. The recording persists for a time determined by the lifetime of the products of the burning or the

scale time for diffusion in the matrix; at liquid-helium temperatures, the recording persists for many hours or even a matter of years. For example, Breinl *et al.*[14] have reported that the decay half-life of a hole burned in the spectrum of a molecular impurity system (quinizarin in a deuterated organic glass) ranges up to 20 000 yr at liquid-helium temperatures, but studies of the lifetime of the holes and of their temperature dependence are still in their infancy.

Here we would like to report experiments carried out by Saari's group at the Institute of Physics, Academy of Sciences of the Estonian SSR, on space-time holography, in complete accordance with a theory developed earlier.[10] This is a linear theory: Effects related to the photoecho arise in a medium which acts as a linear filter on a probing pulse (i.e., the theory uses the approximation of a linear electric susceptibility).

The holograms are written near the peak of the absorption band of octaethylporphyrin molecules in a polystyrene matrix at a wavelength of 620 nm. The sample photosensitive medium is placed in an optical helium cryostat with windows allowing the transmission of a light beam through the cryostat. The sample is cooled to 1.8 K. The holograms are recorded and read out with the help of a quasi-cw picosecond laser using the dye rhodamine 6 G, synchronously pumped by an argon ion laser. The length of the pulses from the picosecond laser is 3 ps; the spectral width of the pulse is 6 cm^{-1}; and the pulse repetition frequency at an average laser output power of 100 mW is 82 MHz.

The following procedures are used to record and read out the holograms. The beam from the picosecond laser is expanded to a transverse dimension of 5 cm in a telescope and then split into reference and object beams. Both of these beams are directed by a system of prisms and mirrors to the sample in the cryostat, where the two beams intersect at an angle of 6°. The pulses in the object beam are delayed by a time interval τ with respect to the corresponding pulses in the reference beam. This delay can be adjusted over the interval $\tau = \pm 300$ ps with respect to the front of the object pulse by moving one of the reflecting prisms in the reference channel. We recall that the interference which is required for holographic recording arises only if the delay between the pulses does not exceed the phase relaxation time of the excited electronic state of the impurity molecule,[12,13] which is 200–300 ps.

At the entrance to the cryostat the reference beam has a plane wavefront and a time-average intensity of 0.5 $\mu W/cm^2$. The wavefront of the signal beam is formed by reflection from the object (a coin) or by passage through a transparency (an image of an arrow) and has a complex space-time structure. The average intensity of the object beam is two or three times lower than that of the reference beam.

The reproduced signal reaches its maximum contrast at an exposure of several tens of seconds. Over this time, the hole-burning effect of many (about 10^{10}) identical sequences of reference and object pulses, arriving at a frequency of 82 MHz, accumulates in the medium. For reproduction of the recording in the hologram, the object beam is blocked with a screen, and either the reference beam, attenuated by neutral filters, or the same beam, directed opposite to the beam of reference pulses during the recording, is used as the probing beam. The images reproduced from the hologram are photographed 20 cm from the exit windows of the cryostat on screens positioned for the direct and backward readout beams, respectively, on the rear and front sides of the

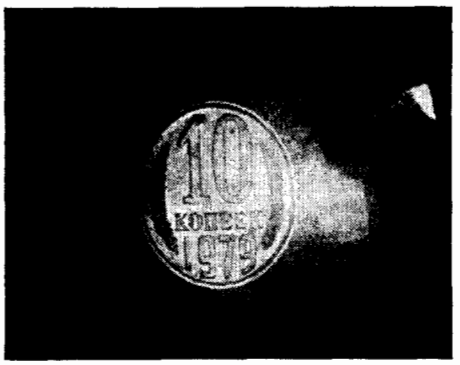

FIG. 1. The holograms were recorded by means of a photchemical burning of spectral holes at a wavelength of 620 nm in the inhomogeneous band of a purely electronic transition (the inhomogeneous width is 200 cm^{-1}) of octaethylporphyrin molecules in a polystyrene matrix at 1.8 K (the homogeneous width of the purely electronic line is no more than 0.05 cm^{-1}). The picosecond reference pulse leads the signal scattered from the coin by 10 ps during the recording. The same delay of the signal is observed when the hologram is read out with a picosecond probing pulse. The exposure in the recording of the hologram is 1 $mJ \cdot cm^{-2}$. This exposure is achieved by accumulating the hole-burning effect of 10^{10} identical pairs of reference and object pulses, arriving at the repetition frequency of 82 MHz of the picosecond laser.

FIG. 2. Correction of a picosecond signal through phase conjugation with a space-time hologram. The picosecond signal, distorted by passage through an irregular glass plate, is recorded under conditions similar to those used to record the image of the coin, except that the order of the application of the pulses is reversed: The reference pulse reaches the recording medium after the object pulse. A photograph of the distorted recorded signal is shown at the left. The hologram is read out for the purpose of correction in the direction opposite to the direction of the reference pulse during the recording. The delayed conjugate signal scattered from the hologram is inverted in both space and time. After it passes through the same distorting medium (the plate) in the opposite direction, the signal reacquires its original, undistorted shape: an image of an arrow (at the right).

cryostat in terms of the direction of the recording beams.

Figure 1 is a photograph of a holographic space-time image of a coin recorded through the photochemical burning of spectral holes. The time evolution of the event—the passage over a coin of the band illuminated by a picosecond pulse—is lost in this photograph because of the time averaging of the delay in the photography. However, the existence of this time evolution can be and was established by the authors on the basis of the appearance of an interference between the image reconstructed from the hologram and the direct image of the coin illuminated by the picosecond pulses.

In contrast with the holography of stationary waves, in the holography of time-varying events there arises the problem of a correct interpretation of the principle of causality in the theoretical description. In the theory of Ref. 10 this problem is dealt with by incorporating the Kramers-Kronig dispersion relations; this approach is equivalent to using a Hilbert transformation (in place of a Fourier transformation) in the transition from a frequency distribution to a time dependence. Experimentally, of course, there is automatic adherence to the principle of causality. This property of holograms can be exploited for phase conjugation and thus a corresponding correction of signals. Figure 2 shows an example of a reconstruction, distorted by an irregular glass plate, of a (space-time) image of an arrow.

[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]V. M. Kharlamov, R. I. Personov, and L. A. Bykovskaya, Opt. Commun. **12**, 191 (1974).

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

[4]J. Friedrich and D. Haarer, Angew. Chem. (Intern. Ed.) **23**, 113 (1984).

[5]H. P. H. Thijssen, R. van der Berg, and S. Völker, in: Photoreaktive Festkörper: Konferenzbuchband der VW-symposiums. Mettelberg, 16–21 September 1984, Wahl-Verlag, Karlsruhe, 1984, p. 763

[6]R. I. Personov, in: Spectroscopy and Excitation Dynamics of Condensed Molecular Systems (ed. V. M. Agranovich and R. M. Hochstrasser), North-Holland, Amsterdam, 1983, p. 55.

[7]K. K. Rebane, J. Lumin. **31-32**, 744 (1984).

[8]E. W. Moerner, J. Mol. Electron. (in press).

[9]A. Szabo, U. S. Patent No. 3.896.420 (July 22, 1975); G. Castro, D. Haarer, R. MacFarlane, and H. P. Thommsdorff, U. S. Patent No. 4.101.976 (July 18, 1978).

[10]P. M. Saari and A. K. Rebane Izv. Akad. Nauk. ESSR, Fiz.-Mat. **33**, 320 (1984).

[11]A. Rebane, P. Kaarli, and P. Saari, Iz. Akad. Nauk. ESSR, Fiz.-Mat. **34**, 444 (1985).

[12]A. K. Rebane, R. K. Kaarli, and P. M. Saari, Pis'ma Zh. Eksp. Teor. Fiz. **38**, 320 (1983) [JETP Lett. **38**, 383 (1983)]; A. Rebane, R. Kaarli, P. Saari, K. Timpmann, and A. Anijalg, Opt. Commun. **47**, 173 (1983); P. M. Saari, P. K. Kaarli, and A. K. Rebane, Kvantovaya Elektron. (Moscow) **12**, 672 (1985) [Sov. J. Quantum Electron. **15**, 443 (1985)].

[13]K. K. Rebane, Usp. Fiz. Nauk **143**, 487 (1984) [Sov. Phys. Usp. **27**, 541 1984)].

[14]W. Breinl, J. Friedrich, and D. Haarer, Chem. Phys. Lett. **106**, 487 (1984).