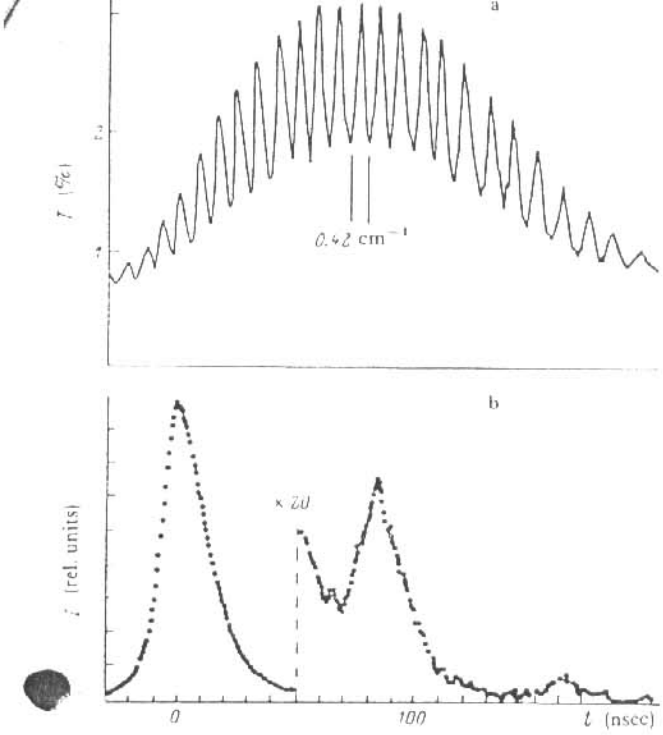


FIG. 1. Lattice of holes in transmission spectrum of a sample of polystyrene activated with $H_2$-tetra(tert-butyl)-porphyrazine (a) and time response of sample to pulsed excitation (b).

where

$$T(\omega) = T_0 + T_{\text{Л}} \sin\left(\frac{2\pi\omega}{\Delta} - \varphi\right). \quad (1)$$

The relative depth of the modulation is

$$M = \frac{T_{\text{Л}}}{T_0}, \quad (2)$$

while the contrast of the spectral lattice, i.e., the ratio of minimum to maximum transmission, is

$$G = \frac{1 + M}{1 - M} = \frac{T_0 + T_{\text{Л}}}{T_0 - T_{\text{Л}}}. \quad (3)$$

The time response of such a spectral lattice to pulsed excitation consists of two pulses separated by the time interval $\tau^{-1}$. Let $Q_0$ and $Q_L$ be the energy of the first and second pulses of the response, respectively, while $q = Q_L / (Q_0 + Q_L)$ is the relative energy of the second pulse. One can show that

$$M = \frac{2\sqrt{Q_0 Q_{\text{Л}}}}{Q_0 + Q_{\text{Л}}} = 2\sqrt{q(1-q)} \quad (4)$$

and

$$G = \frac{\left(\sqrt{Q_0} + \sqrt{Q_{\text{Л}}}\right)^2}{\left(\sqrt{Q_0} - \sqrt{Q_{\text{Л}}}\right)^2} = \frac{\left(\sqrt{1-q} + \sqrt{q}\right)^2}{\left(\sqrt{1-q} - \sqrt{q}\right)^2}. \quad (5)$$

To obtain a spectral lattice of holes of large extent in the present work, a combined method of parallel sequential burning with frequency scanning of the emission of a picosecond dye laser (rhodamine 6G) was employed (5-$cm^{-1}$, spectral halfwidth of pulse 3-psec duration) and transmission of laser pulses through a Fabry-Perot interferometer with a 30-mm spacing.

A block of 9-mm thickness of a solid solution of octaethylporphyn in polystyrene with an admixture concentration of $2.5 \times 10^{-4}$ M was used as a sample. The optical density of the sample at the maximum of the 0–0 band of the admixture $S_1 \leftarrow S_0$ absorption ($\lambda = 618$ mn) was 1.5; the temperature of the measurements was 1.8 K.

A lattice of 1600 holes was burned in the spectral region from 613 to 623 nm with 266-$cm^{-1}$ extent. The spectral distance between neighboring holes was 0.167 $cm^{-1}$, and the density of the irradiation dose integrated over the spectrum was 50 J $cm^{-2}$. The contrast of the lattice was estimated from the relative intensity of the retarding response, which, depending on its place in the spectrum, comprised from 1 to 10%. From Eqs. (4) and (5) then $0.2 \leqslant M \leqslant 0.6$ and $1.5 \leqslant G \leqslant 4$. No substantial dependence of these indices on the spectral extent of the lattice of holes was detected; i.e., on the number of burned out molecules. This shows that under the conditions of the experiment inhomogeneous broadening induced by static action of the photoproduct did not appear, which is extremely essential for practical application of the photochemical hole-burning method.[4]

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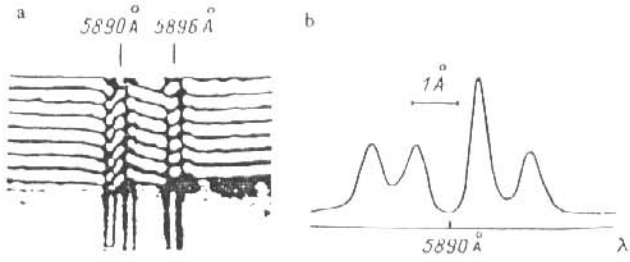


FIG. 2. (a) Interferogram and absorption spectrum of sodium resonance doublet; (b) densitometer tracing of absorption spectrum close to the line ($\lambda = 5890$ Å).

trum as a consequence of the negative absorption of the barium atoms (amplification without population inversion) on resonance interaction. We note that amplification without population inversion was observed earlier in the region.[7] The behavior of the refractive index also quantitatively agrees with this model.

To study the behavior of a multiplet in a light wave field the yellow sodium doublet was chosen. In Fig. 2(a) an interferogram and absorption spectrum close to the sodium resonance doublet are given. Splitting of the two absorption lines is evident. The line corresponding to the $3^2S_{1/2}$–$3^2P_{3/2}$ transition is split into four components and that corresponding to the $3^2S_{1/2}$–$3^2P_{1/2}$ transition into two components. In Fig. 2(b) a densitometer tracing of the spectrum close to the absorption line corresponding to the $3^2S_{1/2}$–$3^2P_{3/2}$ transition is given.

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## Limiting number of photochemical holes in an inhomogeneously broadened spectrum

Ya. V. Kikas, R. K. Kaarli, and A. K. Rebane
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The limit in principle to the number of holes formed in the inhomogeneously broadened band of an extrinsic absorption by photochemical burning[1,2] is the ratio of the widths of the inhomogeneous absorption band and the homogeneous phononless line. For a purely electronic absorption band this value can reach $10^4$–$10^5$ for allowed transitions[3] and possibly several orders of magnitude greater for forbidden ones. In the papers known to us on burning many holes in one inhomogeneously broadened spectral band,[4] their number is of the order of $10^2$ or less, which is considerably smaller than the theoretical limit. Besides the experimental difficulties, however, several physical causes may hinder attainment of the latter (inhomogeneous broadening induced by formation of a photoproduct). By this fact experiments on burning a large number of holes are of interest for their clarification. In the present paper a substantially closer approach to the theoretical limit is announced—the burning in an inhomogeneously broadened spectrum of more than $10^3$ holes.

Usually burning and recording holes in an inhomogeneous spectrum are accomplished by frequency scanning of a narrowband laser (sequential burning). The possibility of burning a large number of holes (spectral lattice) by a series of mutually coherent light pulses (parallel burning) was shown recently.[5,6] Excitation by a series of equidistant mutually coherent light pulses equivalent to periodic modulation of the spectrum of a single exciting pulse also leads, as a result of the photochemical burning process, to formation of a periodic structure (lattice) in the transmission spectrum of the sample. In Fig. 1(a) such a structure in the transmission spectrum of a sample of polystyrene with a $H_2$-tetra (tert-butyl)-porphyrazine admixture at 1.8 K is represented. The burning was conducted by a series of picosecond pulses obtained upon passage of a pulse from a synchronously pumped dye laser through a Fabry-Perot interferometer with a spacing of 12 mm (80 psec). The width of the laser line when recording the spectral lattice was 0.1 $cm^{-1}$.

As experimentally demonstrated in Ref. 6, the presence of such a spectral lattice leads to the arising of a multipulse response of the system to pulsed excitation. In Fig. 1(b) the response is shown from a sample, the transmission spectrum of which is shown in Fig. 1(a). The duration of the pulses represented in Fig. 1 (25 psec) is determined by the apparatus function of the synchronously scanned electron-optical converter.

The contrast of the spectral lattice can be determined from the time response parameters. Let us examine the case of sinusoidal modulation of the transmission spectrum

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