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Light-controlled band shift in synthetic opals filled with an optically nonlinear dye solution*

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***Abstract*. The light-induced nonlinear response of SiO_2 synthetic opals filled with a dye solution in ethanol is experimentally studied. Excitation of samples by radiation with a power density of ~ 10 W cm^{-2} causes changes in their transmission and reflection due to the light-induced dynamic shift of the stop band of a photonic crystal. Synthetic opals studied in the paper can be used in optical gates and light-controlled switches.**

Keywords: *opal-like nanomatrices, photonic crystals, optical nonlinearity, optical switches.*

Advances in the manufacturing technology of opal-like SiO_2 nanomatrices (ONM) allow the fabrication of structures consisting of closely packed periodically repeated amorphous SiO_2 spheres of diameter 100 – 800 nm.

A considerable volume of voids between spheres (from 24 % in normal opals to 76 % in inverse opals) allows the filling of ONMs with other substances, which can, for example, change efficiently their optical parameters. In particular, the introduction of inorganic and organic liquids into nanosize voids in opals can reduce light scattering, superluminescence and other processes at SiO_2 sphere – void interfaces by several orders of magnitude, thereby increasing optical transmission. In addition, of great interest is the possibility of fabricating new opal-like materials with strong nonlinear-optical properties. The presence of the forbidden photonic band, or the stop band, is important for possible applications of photonic crystals and ONMs in optical data processing and communication systems [1, 2]. The combination of a drastic change in the transmission/reflection of ONM samples in the region of the stop band with the optical nonlinearity of a material filling them can lead to the development of promising optical switches with improved energy and temporal parameters (see, for example, [3]).

In this paper, we present experimental angular and temperature dependences of the transmission of SiO_2 ONM plates filled with the methyl red dye solution in ethanol and study light-induced changes of this transmission caused by low-intensity laser radiation.

We studied 0.5 – 1.5-mm-thick opal-like samples of area 0.5 – 2 cm^2 based on SiO_2 nanospheres of diameter 200 – 300 nm. The samples were manufactured at the Tekhnomash Central Research Technological Institute (Russia). Excitation and probing were performed with the cw and modulated radiation from an argon laser at 488 and 514 nm and radiation from a 632-nm He – Ne laser. These conditions determined the choice of the spectral position of the stop band and the spectral region of the maximum nonlinearity of the filling medium. We also took into account intense diffusion scattering in comparatively thick opal-like samples, which complicated the measurement of transmission spectra of opals and simulation of optical switches with the use of transmitted radiation. Scattering is related to the difference in the refractive indices of SiO_2 spheres ($n = 1.37$) forming the nanomatrix lattice and of air-filled pores ($n = 1$). For this reason, most of the measurements were performed by immersing matrices into ethanol with $n = 1.361$.

First of all, we measured the spectral parameters of samples to estimate their quality and to determine the initial conditions for studying the temperature and light-induced changes in them. The transmission spectra were recorded with PV 1251B spectrophotometer (Solar, Belarus).

Figure 1 illustrates the angular dependence of the spectral position of the stop band for an opal-like sample, which was selected for the study upon excitation at 488 nm and probing by a 632-nm laser beam. The general tendency of the stop band to exhibit the blue shift with deviation from the normal incidence of light agrees with the data in the literature [4] and correlates with the general physical picture of the formation of spectral characteristics of a volume Bragg reflector. Variations in transmission and the stop-band depth observed in Fig. 1 can be caused by the propagation of the beam through different volume regions of the inhomogeneous synthetic opal of thickness 1.5 mm with changing the angle of incidence; as a result, different sets of monodomains are involved in the formation of the spectral characteristic of the sample.

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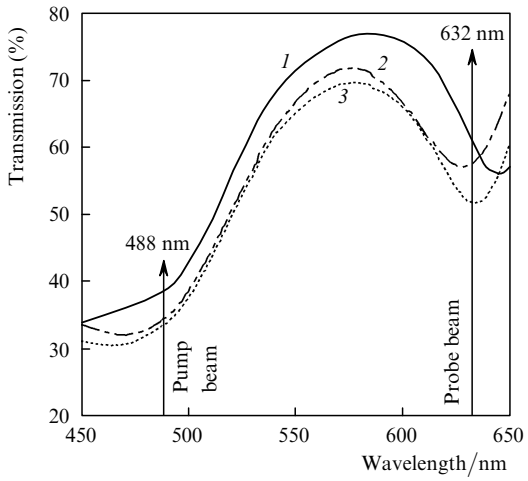

Figure 1. Transmission spectra of a SiO_2 synthetic opal immersed in ethanol recorded for the angles of incidence equal to 0 (*1*), +9° (*2*), and −7° (*3*).

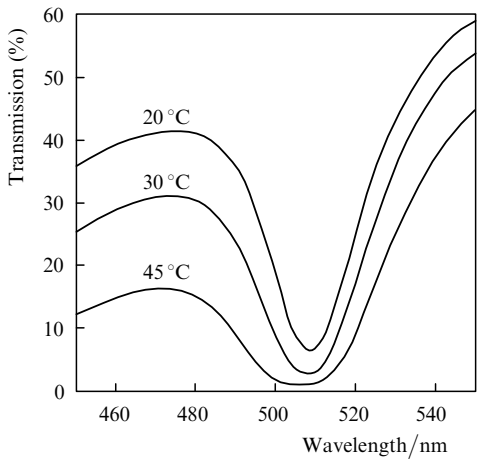

Figure 2. Temperature dependences of the transmission spectra of SiO_2 synthetic opals immersed in ethanol. The stop bands of samples are located near 510 nm.

The ONM samples consist of SiO_2 nanoglobules and have a weak intrinsic nonlinearity. To enhance the nonlinear properties of ONM samples, they were filled with a dye solution in ethanol. The dye type was selected according to the excitation wavelength. In particular, the methyl red dye was used for the blue–green spectral region. The dye concentration should not be too high to avoid strong absorption of radiation. At the same time, the absorption of laser radiation by the dye should provide the local heating of the solution and the corresponding temperature change in the refractive index of solution in the matrix. The value and sign of the induced refraction are determined by the temperature dependence of the refractive index of ethanol with the coefficient $dn/dT = -4 \times 10^{-4}$ K^{-1}.

The nonlinear response of the ONM sample was simulated by changing its temperature upon heating the sample by a thermal source and its cooling. The temperature dependences of transmission studied at the excitation wavelengths were used to estimate the influence of nonlinearity on the spectral and amplitude characteristics of samples filled with ethanol. Figure 2 presents the results of measurements performed for an ONM sample with the stop band located at 0.5 μm. One can see that the stop band broadens with temperature and the transmission minimum shifts to the blue in accordance with the temperature dependence of the refractive index of the medium filling the ONM. The spectral shift of the transmission band is one of the most important conditions for obtaining optical switching and limitation.

The nonlinear optical properties of immersed ONMs were studied by recording transfer characteristics in reflected and transmitted light upon excitation of samples by rectangular 488-nm laser pulses with the power density from several W cm^{-2} to tens of kW cm^{-2}. According to the spectral properties of the sample under study (Fig. 1), the probe radiation from a 632-nm He–Ne laser was used. The probe beam was not modulated, had a power of ~ 1 mW (i.e. considerably lower than the excitation beam power) and did not produce any substantial changes in the transmission of samples. Figure 3 shows the optical scheme for nonlinear optical measurements.

A sample was mounted on an angular positioning rotation stage providing the tuning of the stop band

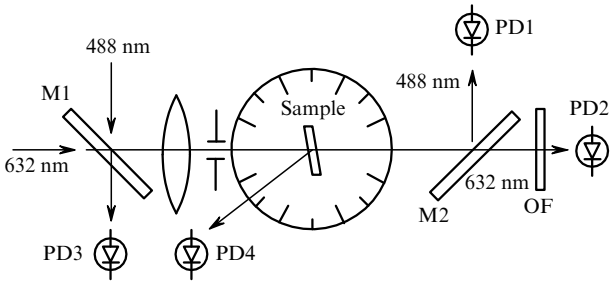

Figure 3. Scheme of the experiment on measuring changes in the sample transmission induced by radiation from an argon laser and probed by radiation from a He–Ne laser. A SiO_2 synthetic opal sample is immersed in the methyl red dye solution in ethanol; PD1–PD4 are photodiodes; M1, M2 are dichroic mirrors with the maximum reflectance at 490 nm; OF is a 590-nm cut-off optical filter.

with respect to the probe radiation wavelength by tilting an ONM. An objective with the focal distance from 60 to 400 mm provided the required excitation power density. Radiation transmitted through the sample and reflected probe-beam radiation were detected with photodiodes PD2 and PD3, respectively. An optical filter suppressing exciting radiation was mounted in front of PD3, if necessary. Photodetector PD4 was used to control the spectral shape and power of exciting radiation, and PD1 was used to record the kinetics of this radiation transmitted through a sample.

Samples with pores filled preliminarily were mounted in a closed cell on a rotation stage and immersed into a solution. Excitation was performed in the sample region projecting over the solution level. This prevented rapid evaporation of ethanol from ONM pores and, in addition, excluded the nonlinear interaction with the solution, which would be located between the cell wall and sample surface in the case of completely immersed sample.

Figure 4a shows the change in transmission of the ONM sample at 632 nm, whose transmission spectra are presented in Fig. 1. The intensity fronts of the probe beam correspond to the fronts of a rectangular excitation pulse. The initial conditions correspond to spectrum (*1*) in Fig. 1, when the probe-beam wavelength is located at the short-wavelength wing of the stop band. The excitation power density at

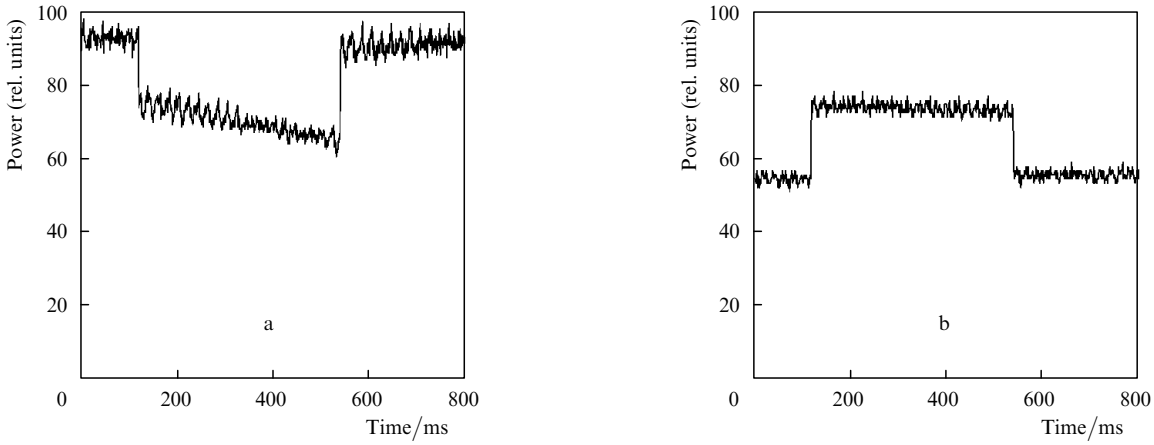

Figure 4. Changes in the transmission of a synthetic opal sample at 632 nm (probe beam of a He – Ne laser) excited by an argon laser at 488 nm (light-induced switching); the probe-beam wavelength corresponds to the short-wavelength (a) and long-wavelength (b) wings of the stop band.

488 nm was ~ 35 W cm^{-2}. A lens was not used in the scheme in Fig. 3 and the aperture diameter was 0.5 mm. The noise of the signals presented in Fig. 4 is caused by the electric interference in input circuits of detectors.

One can see that, along with optically controlled modulation achieving 20 %, comparatively slow changes in transmission become already noticeable for the given excitation power density. These changes reduce the intensity of the probe and transmitted exciting beams. They can be caused by a number of physical processes such as an induced thermal lens in a sample, intense evaporation of the solution from the excited region and even its local boiling. The slope of the curve presented in Fig. 4a is probably caused by the same mechanisms as the general decrease of transmission in Fig. 2. It is important in our case to determine the total effect of such processes and to exclude, where possible, their distorting influence on optical switching in such systems.

Figure 4b shows the change in the power of obliquely incident probe radiation. In this case, the probe-beam wavelength falls on the long-wavelength wing of the stop band [the initial spectrum approximately corresponds to curve (*3*) in Fig. 1]. The amplitudes of signals in Figs 4a, b are normalised identically and are related to different wings of the transmission contour. The excitation power density in the case in Fig. 4b was ~ 20 W cm^{-2} and the modulation depth was ~ 30 %. One can see that the sample has a higher switching contrast at the lower initial transmission. For the given power density, the long-term decrease in transmission becomes virtually inessential. The estimate of the spectral shift of the stop band resulting in the observed change in transmission is ~ 10 nm and approaches to the value sufficient for the realisation of devices for controlling optical signals. At the same time, for practical applications of ONMs in such devices it is desirable to use samples having stop bands with steeper wings.

As the power density of the excitation beam is further increased, slow changes in the ONM transmission considerably exceed the switching-pulse amplitude (Fig. 5), which is most likely caused by overheating of the solution in the volume exposed to the laser beam over its boiling temperature.

The kinetics was recorded under the same conditions as before, but using a focusing lens in the optical scheme. The diameter of a region excited by the focused laser beam was 20 μm and the power density achieved a few kW cm^{-2}.

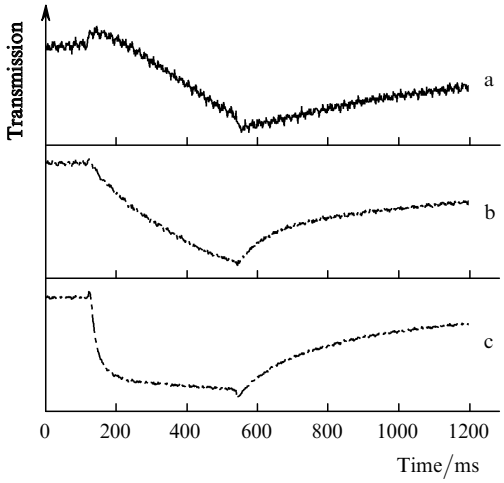

Figure 5. Changes in the transmission of a synthetic opal sample exposed for a long time to radiation with intensities 75 (a), 150 (b), and 500 W cm^{-2} (c).

Initially, the probe-beam wavelength was located in the spectral region approaching the stop-band bottom from the short-wavelength side. One can see that, beginning from the power density of the order of a hundred of W cm^{-2}, the attempts to increase the switching contrast by simply increasing the excitation power lead to a drastic increase in the influence of comparatively slow variations in the ONM transmission. As a result, such changes in transmission can exceed the switching amplitude by many times, thereby reducing the switching contrast to zero.

To optimise the conditions for manifestation of the nonlinear response in ONM systems, we measured the diagrams of angular reflection spectra for the ONM growth surfaces (normal to the [111] axis of a face-centred cubic lattice) both from the growth surface side and the side facing a substrate in the technological process of sample fabrication. The spectra were recorded for wavelengths of an argon and a He – Ne lasers at 476.5, 488, 514.5, and 632.8 nm. Measurements were also performed after additional polishing of these surfaces.

The measurements were used to determine the wavelength dependences of the optimal reflection angle associated with the stop band at which the maximum contrast is achieved for the intensity peak in the angular

distribution diagrams for reflected and scattered radiation. In particular, the contrast measured for a radiation wavelength of 488 nm and the angle of incidence 30° was 15. The angular width of the peak did not exceed 10°. The total radiation power in the angular distribution peak achieved 35 % of the incident power.

We observed the correlation between the optimal reflection angle and the angular width of the peak for radiation of a shorter wavelength with respect to the band gap. For the long-wavelength radiation at 632.8 nm, the type of reflection changes considerably. The angular profile of the reflection peak corresponds quite accurately to that of the incident beam, and for large angles of incidence (exceeding 35°), an additional diffraction peak, blurred due to scattering, appears in the vicinity of half the angle of incidence.

The quality criteria for ONM samples providing a strong nonlinear response take into account the sharpness of the reflection peak in the radiation directional pattern and the contrast of this peak against the background (Rayleigh) scattering. The optimisation of samples includes the ordering of their structure, the reduction of the size dispersion of nanospheres forming the matrix, and careful matching of the lattice period of a photonic crystal, the refractive indices of the matrix and a medium filling pores in samples with the excitation wavelength.

Thus, we have observed the light-induced dynamic shift of the stop band in the SiO_2 opal-like nanomatrix filled with the methyl red dye solution in ethanol upon excitation by low-intensity pulses from an argon laser. The two mechanisms of the induced change in the ONM transmission and shift of the stop band with substantially different temporal characteristics have been pointed out. The switching of the signal light beam in the ONM has been performed by using control radiation pulses with power densities ~ 10 W cm^{-2}.

Our study suggests that ONMs filled with nonlinear media can be promising for the development and improvement of the basic devices for optical data processing systems, including optical modulators, limiters, bistable elements, and multiplexers/demultiplexers.

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References

1. Joannopoulos J.D., Villeneuve P.R., Fan S. *Nature*, **386**, 143 (1997).
2. Fleming J.G., Lin S.-Y. *Opt. Lett.*, **24**, 49 (1999).
3. Mazurenko D.A., Akimov A.V., Pevtsov A.B., Kurdyukov D.A., Golubev V.G., Dijkhuis J.I. *Physica E*, **17**, 410 (2003).
4. Comoretto D., Cavallo D., Dellepiane G., Grassi R., Marabelli F., Andreani L., Brabec C., Andreev A., Zakhidov A., in *Mater. Res. Soc. Symp. Proc.* (2002) Vol. 708, p. BB10.19.1.