
Reflection Spectra of and Infiltrated Synthetic Opal Photonic Ba(NO₃)₂ Crystals

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Abstract

Angular dependences of Bragg reflection spectra for the bare and infiltrated synthetic opal photonic crystals of Ba(NO₃)₂ have been studied in the wavelength range of 420–630 nm for the geometry of specular reflection and the incidence angles 5°, 20°, 30° and 45°. Theoretical relation between the diffracted light wavelengths at the maximum reflectance and the angle of incidence has been used and compared with the experimental one. The distance between (111) planes of opal and the diameter of silica globules have been determined. Angular distribution of diffuse reflection has been measured in the vicinity of direction of specular reflection.

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Introduction

Photonic crystals represent a class of new optical materials with unique properties [1–3]. Synthetic opal photonic crystal is an artificial, three-dimensionally periodic dielectric structure with a period close to the wavelength of light. The interest to different optical phenomena occurring in the photonic crystals is caused by a presence of forbidden gaps in their photonic band structure. Photonic forbidden gap emerges as a result of light scattering on a three-dimensional lattice formed by modulation of dielectric constant. The most interesting phenomenon is a complete forbidden photonic gap, a range of wavelengths where light cannot propagate through photonic crystal in any direction, thus enabling a variety of applications, in particular those associated with photonics and optoelectronics [2]. A notable advance in fabrication of synthetic opal photonic crystals and a wide range of possible modifications of

their optical properties achieved by infiltrating them with various substances have stimulated extensive experimental studies of those materials [4–16]. At the same time, experimental researches of optical properties for the pure synthetic opal photonic crystals have been mainly carried out with the methods of light reflection and transmission. As a consequence, the obtained experimental material has rather limited character. Moreover, the studies for synthetic opal photonic crystals with dielectric filling have not been performed earlier.

The purpose of the present work is to study angular dependences of the reflection spectra for both the bare and infiltrated synthetic opal photonic Ba(NO₃)₂ crystals.

Samples and experimental setup

We studied samples of synthetic opal made of a-SiO₂ globules. The dimensions of samples were approximately 1.0x0.5x0.5cm³. The

reflection spectra for the case of reflection from the plane of growth (111) were measured for both the bare opal sample and the sample with interstitials filled with $\text{Ba}(\text{NO}_3)_2$. The samples were illuminated with incandescent lamp or light emitting diodes. Light from the source was collimated with a collimator of goniometer GS-5 and an iris diaphragm. The divergence of the incident collimated beam (its diameter being equal to 3 mm) was not more than $1'$. The reflected light, which included the component related to diffuse reflection, was collected with a lens positioned at two focal distances from the sample and a fibre-optical cord with the cross section diameter 3.5 mm positioned at two focal distances from the lens. The resulting viewing angle of radiation was less than 10° . The other end of the fibre-optical cord was placed in the plane of entrance split of a double grating monochromator of spectrometer DFS-12. Such the position of optical elements enabled us to direct all the reflected light into the cord and so measure not only a general shape of the reflection spectra but conduct quantitative measurements. The reflection spectra were measured in the geometry of specular reflection for the incidence angles of 5° , 20° , 30° and 45° . The data obtained for the incidence angle of 5°

contained only qualitative information which was used for further analysis of the spectra measured for the oblique incidence reflection. The angular distribution of the diffuse reflection was derived with the aid of a slit diaphragm in a close vicinity of the direction of specular reflection at the wavelength that corresponded to the maximum specular reflectance.

Results and interpretation

The measured Bragg reflection spectra of the bare synthetic opal photonic crystals and those impregnated with $\text{Ba}(\text{NO}_3)_2$ are shown respectively in Fig. 1 and 2. In case of sample 1 (bare synthetic opal), the maximum reflectances for the incidence angles 5° , 20° , 30° and 45° are detected at the wavelengths of 520, 506, 482 and 445 nm, respectively. The maximum reflectances for sample 2 (synthetic opal with interstitials filled with $\text{Ba}(\text{NO}_3)_2$) and the same incidence angles are detected at the wavelengths of 606, 590, 564 and 510 nm, respectively. The magnitude of the maximum reflectance for the incidence angle 20° reaches 25% for the both samples. The reflectance decreases with increasing incidence angle: it is equal to 24% for 30° in case of both samples, while for 45° we have 18% for the sample 1 and 20% for the sample 2.

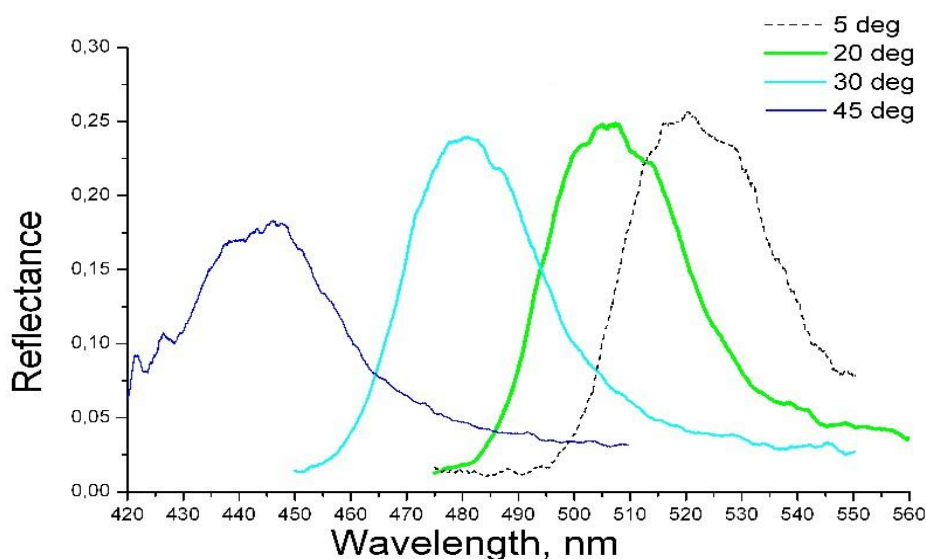


Fig. 1. Angular dependence of the Bragg reflection spectrum for the bare synthetic opal photonic crystals.

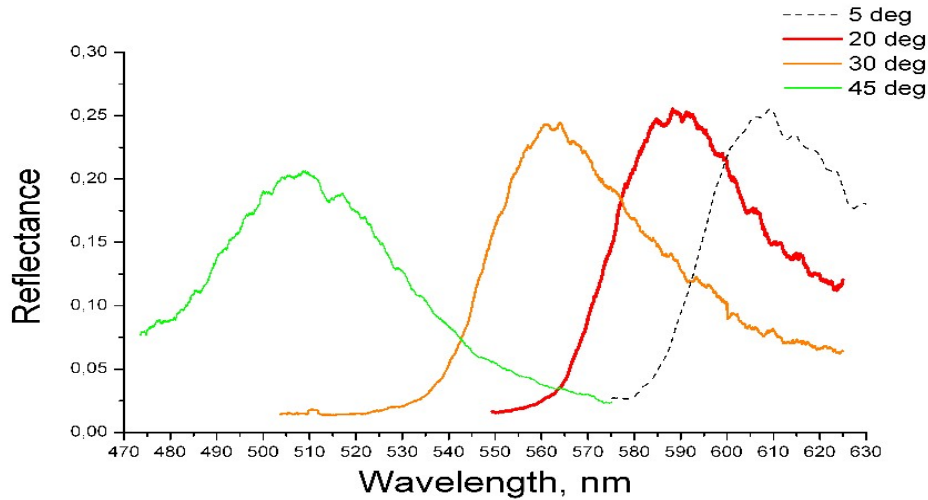


Fig. 2. Angular dependence of the Bragg reflection spectrum for the synthetic opal photonic crystals infiltrated with $\text{Ba}(\text{NO}_3)_2$.

We have analysed the obtained spectra in the approximation of Bragg diffraction on one-dimensional periodic structure, using a well-known expression for the diffracted light wavelength:

$$\lambda(\theta) = 2d\sqrt{n_{\text{eff}}^2 - \sin^2 \theta} ,$$

where d is the distance between (111) planes of opal ($d = (2/3)^{1/2} a$, with a being the diameter of silica globules), θ the angle of incidence, $n_{\text{eff}}^2 = x n_{\text{sph}}^2 + (1-x) n_{\text{int}}^2$ denotes the effective refractive index of the opal (with x being the packing fraction of the spheres, n_{sph} and n_{int} the refraction

indices of materials of the spheres and interstitials, respectively). We have thus calculated the theoretical dependence of the wavelength of diffracted light on the incidence angle.

The following parameter values have been used for the sample 1: $n_{\text{sph}} = 1.47$, $n_{\text{int}} = 1$, $x = 0.74$, $n_{\text{eff}} = 1.363$ and $a = 234$ nm (notice that the a value has been calculated from the nearly normal reflection data). The wavelengths 504, 484 and 445 nm have been obtained respectively for the incidence angles of 20° , 30° and 45° . For the sample 2, the corresponding parameters are

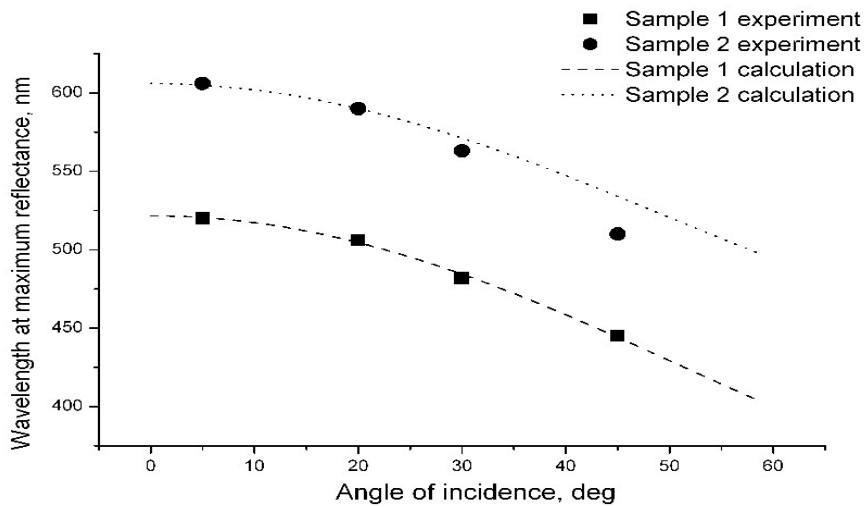


Fig. 3. Theoretical and experimental dependences of the maximum reflectance wavelengths for the Bragg diffracted light on the angle of incidence.

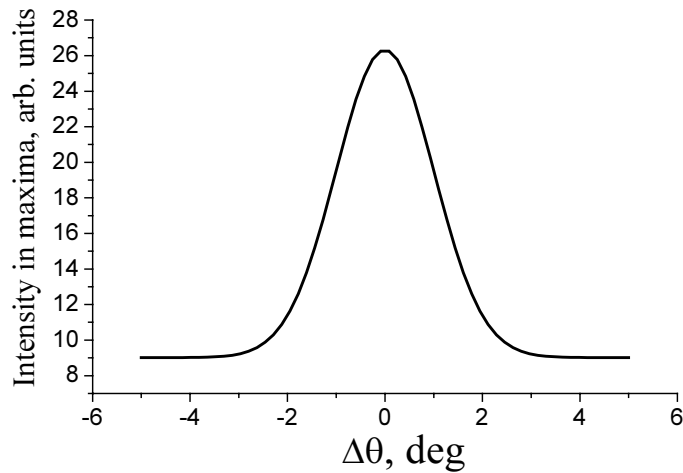


Fig. 4. Angular distribution of diffusely reflected light. The sample is illuminated with the light characterized with the wavelength $\lambda_{\max} = 513$ nm, using light emitting diodes.

as follows: $n_{sph} = 1.47$, $n_{int} = 1.57$, $x = 0.74$, $n_{eff} = 1.497$ and $a = 248$ nm. Then we get the wavelengths 591, 572 and 535 nm respectively for the angles of incidence mentioned above. Hence, the experiment and calculation give very close results for the sample 1, while the data for the sample 2 somewhat differ (see also Fig. 3).

The latter discrepancy seems to be due to some features of filling of the interstitials with substance. The halfwidth of the peak in the reflectance spectra is generally defined by the stopgap width. For the sample 1, the peak halfwidths are 30, 30 and 36 nm respectively for the angles of incidence 20° , 30° and 45° . The corresponding values for the sample 2 are 47, 47 and 50 nm. Additional broadening of the peaks is due to light scattering on the defects of opal structure. Misalignments of densely packed growth planes (111) in the domains from which the bulk opal consists [15], interfaces of the domains and defects in the domains like stacking faults and point defects might be mentioned among those defects. In order to obtain quantitative data about disordered orientation of the planes (111) in the blocks, the angular distribution of the diffuse reflection has been measured at $\lambda_{\max} = 513$ nm for the angle of incidence $\theta = 13.5^\circ$. Diffusely reflected light has been studied near the direction of specular reflection in the angular region $\theta' = \theta \pm \Delta\theta$ (Fig. 4).

The obtained data have shown that $\Delta\theta_{\max} = 3.2^\circ$. This corresponds to the angle of 1.6° between the normal to densely packed hexagonal layers (111) in different areas on the sample surface and its growth axis. The result agrees well with the data reported in the work [4]. The intensity of scattering increases with increasing angle of incidence, thus yielding additional spectral broadening for the large angles. One more mechanism of spectral broadening for the peak in the reflection spectra, available in the case of impregnated opal, is possibly due to defects in filling of the interstitials.

Conclusion

Thus, in the present work we have evidenced experimentally that the spectra of Bragg reflection from the non-polished growth planes (111) contain an asymmetric peak, whose parameters depend upon the angle of incidence. The magnitude of the maximum reflectance for the incidence angle 20° reaches 25% for both samples studied by us. The reflectance decreases with increasing angle: for 30° it is equal to 24% for both samples, while for 45° it is 18% for the bare synthetic opal photonic crystals and 20% for those impregnated with $\text{Ba}(\text{NO}_3)_2$. We have also determined the distance d between (111) planes of opal and the diameter a of silica globules. They are $d = 191$ nm, $a = 234$ nm and

$d = 203$ nm, $a = 249$ nm respectively for the bare synthetic opal photonic crystals and the crystals impregnated with $\text{Ba}(\text{NO}_3)_2$. It is established that the additional broadening of the Bragg reflection peak is caused by scattering of light on the defects of opal structure. We have shown on the basis of the obtained data that the angle between the normal to densely packed hexagonal layers (111) in different areas of the sample surface and the growth axis is $\leq 1.6^\circ$. Estimation of the photonic stopband gap has given the value $\Delta_g = 0.15\text{eV}$.

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