Methods of characterization of synthetic opal films

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Abstract. We developed methods for determination of thickness, number of layers and filling fraction of silica particles for synthetic opals. We show that the filling fraction is considerably less than for ideal close-packed structure, which is important for practical and theoretical applications.

1. Introduction
Photonic crystals (PC) are materials with a spatial periodicity in their dielectric function (or, equivalently, a periodical index of refraction) [1]. PC can have a photonic bandgap, i.e. a frequency window in which propagation of light through the crystal is difficult [2]. The inhibition of light propagation makes it possible to control spontaneous emission [3] that has many important applications [4]. Recently it has been shown [5] that the modification of the interaction of an electron with its own radiation field in the PC medium in fact is equivalent to that the electron mass changes its value and this opens up new vistas for their practical applications. This effect, for example, can in turn give rise to the significant modification of the strong interaction of emitters in PCs with a resonant laser field, predicted in [6]. At present there are numerous approaches to the creation of PC by using the lithography [7], interferential holography [8] and self-assembly of colloidal particles [9]. We use self-assembly of colloidal silica particles (SiO₂). It forms synthetic opal films consisting of close-packed spherical particles of silica. The photonic crystal samples based on (SiO₂) were prepared by vertical deposition onto a glass substrate by capillary action [9]. The process is quite simple in terms of hardware design and have no fundamental restrictions both on the sample size and the number of photonic crystals produced in one cycle of synthesis. So our purpose is to investigate methods for quick and easy determination of parameters such as thickness, number of layers and filling fraction of silica particles. Such methods and parameters required at multiple syntheses for different experiments with PC. Effective refractive index, thickness of layer and filling fraction of silica particles can be determined by spectrum position of band gap. Thickness of PC and number of layers can be determined by different methods depending on the number of layers.

2. Spectroscopic methods of parameters determination
Effective refractive index, thickness of layer and filling fraction of silica particles can be determined by transmission or reflection spectra measured at different incidence angles with the help of Bragg’s law. Taking account of Snell’s law, it is written as:

$$\lambda = 2d_{(11)} \left( n_{ef}^2 \sin^2 \theta \right)^{1/2},$$

(1)
where $\lambda$ is the wavelength corresponding to the bandgap, $d_{(111)}$ is the period of the structure (the distance between the (111) planes, so-called thickness of layer), $n_{\text{eff}}$ is the effective refractive index, $\theta$ is the angle between the normal to the sample surface and the direction of the incident light.

Figure 1 shows the transmission and reflection spectra of PC samples based on silica. The dependence of band gap spectral position ($\lambda$) on the incidence light angle ($\theta$) is linear in terms of $\lambda^2 - \sin^2 \theta$ (figure 2).

![Figure 1](image1.png)

**Figure 1.** Transmission (a) and reflection (b) spectra of the PC samples.

![Figure 2](image2.png)

**Figure 2.** The spectral positions of the band gaps for different angles of incidence of light on the sample surface: a) for the transmission spectrum; b) for the reflection spectrum.

Using a linear least-squares approximation, we have calculated the effective refractive index as well as the period of the structure. The effective refractive index of the PC is

\[
n_{\text{eff}} = \left( n^2 \cdot f + n_{\text{air}}^2 \cdot (1 - f) \right)^{1/2},
\]

where $n$ is the refractive index of the SiO$_2$ particles ($n = 1.46$), $n_{\text{air}}$ is the refractive index of air ($n_{\text{air}} = 1$), $f$ is the filling fraction of silica particles. According to close-packing of equal spheres the $f$ parameter is equal to 0.7405 and $n_{\text{eff}}$, in turn, is 1.36. Using the equation 2, the filling fraction of silica particles can be calculated as follows:

\[
f = \frac{n_{\text{eff}}^2 - n_{\text{air}}^2}{n^2 - n_{\text{air}}^2}.
\]
The calculation results for several different samples showed that the filling fraction lies in the range from 0.59 to 0.66, while the average value of $d_{(111)}$ and $n_{eff}$ was 280 nm and 1.30, respectively. Filling fraction obtained for the PC samples is significantly less than for the ideal structure with close-packing of identical spheres. Since the defects of lattice affect a packing density, conventional PC samples have a larger volume of cavities.

3. Thickness and number of layers for thick films

Thickness of the thick films (number of layers greater than about 20) can be determined by microscopy of cross-section or measurement the mass of the samples. Microscopy of PCs cross-section is the obvious way for thickness determination using the image from optical microscope which is shown on figure 3 (a).

![Figure 3](image)

**Figure 3.** a) Snapshot of PCs cross-section by using optical microscope; b) scheme of PCs cross-section.

Thickness of PC can be determined via mass of samples:

$$H = \frac{m}{\rho \cdot S \cdot f},$$

where $H$ is the thickness of PC, $m$ is the mass of the sample, $\rho$ is the density of silica particles, $S$ is the square of PC surface. Number of layers ($N_L$) can be determined via thickness of PC and thickness of the layer:

$$N_L = \frac{H}{d_{(111)}}.$$

The calculation results for one of the samples were as follows: $m = 17$ mg, $N_L = 26$ and $H = 17.3$ µm. The proposed method for determining the thickness cannot be fine applied to a relatively thin film, because their mass is so small that it becomes comparable with the measurement error.

4. Thickness and number of layers for thin films

For the thin PC films number of layers can be determined by oscillations on the transmission spectrum (figure 4). The interference of light reflected by the top and bottom surfaces causes the well-known Fabry-Perot interferences [10].
Combining the Bragg’s law and interference conditions we obtain the following expression:

\[ N_L = \frac{p \cdot \lambda_0 \cdot \lambda_p}{\lambda_{Br} \left( \lambda_0 - \lambda_p \right)}, \]  

(6)

where \( \lambda_0, \lambda_p \) is wavelengths corresponding to different fringes, \( p \) is a positive integer numbering consecutive minima between the fringes, \( \lambda_{Br} \) – the Bragg diffraction wavelength (bandgap). Thickness of PC can be obtained via number of layers and thickness of layer: \( H = N_L \cdot d_{(111)} \), where thickness of the layer \( d_{(111)} \) is calculated by the method described above.

Thus, the thickness of the PC film for the sample in figure 4 (a) amounted to 3.6 µm, number of layers – 12. For the sample in figure 4 (b), these parameters were 4.4 µm and 15, respectively.

In the case of a film of thickness more than 8 µm, intensity oscillations cannot be clearly observed in the visible light. Thus, determination of number of layers for thick films by Fabry-Perot oscillations is not possible.

5. Conclusion

In summary, we have developed methods for quick and easy determination of synthetic opal’s parameters such as thickness and number of layers for thick-film PCs. We have shown that filling fraction of silica particles obtained for conventional PC are considerably smaller than for ideal PC structure due to the presence of packing defects.

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