Technical Physics Letters, 2025, Vol. 51, No. 4

08

Features of calculation and investigation of optical characteristics of a gradient photonic crystal

© E.N. Egorova[1,2], E.V. Panfilova[2], I.O. Azarnin[2], M.S. Kuzikova[2]

[1] National Research Center „Kurchatov Institute“, Moscow, Russia
[2] Bauman Moscow State Technical University, Moscow, Russia
E-mail: egorova_EN@nrcki.ru

Received October 22, 2024
Revised December 10, 2024
Accepted December 15, 2024

A method for synthesizing gradient photonic colloidal crystals is shown. A method for calculating the effective refractive index and the position of the photonic band gap of gradient structures is proposed. Successful verification of the computational model was carried out on the obtained samples.

Keywords: gradient photonic crystal, photonic band gap.

DOI: 10.61011/TPL.2025.04.61004.20165

Colloidal photonic crystal structures hold much promise for use in devices for selective interaction with incident radiation in the visible wavelength range. For example, a narrow-band filter based on a photonic crystal [1] is a reflective Bragg grating formed by a periodic ordered closely packed structure of colloidal silicon dioxide microspheres. The phenomenon of self-organization underlies the formation of such structures. Technological conditions of fabrication of a colloidal photonic crystal are set in accordance with the desired optical properties of the structure and the corresponding morphology.

The optical characteristics of photonic crystals (reflection coefficient R [%], position of the peak of the photonic band gap (PBG) λ [nm], and its width Δ [nm]) are determined by the material of the photonic crystal and its structure. Colloidal solutions of silicon dioxide (SiO_2), polystyrene monodisperse latex, and polymethyl methacrylate are the systems that are traditionally used to produce self-organized optical structures. Their dispersed phase is monodisperse spheres with diameters ranging from several tens of nanometers to several micrometers, which are arranged in closely packed monolayers and 2D and 3D objects [2,3]. The research in this direction has spurred interest in combined gradient structures [4]. A gradient photonic crystal is a solid-state ordered structure formed from particles with a layer-by-layer change in diameter. This structuring affects the geometric and dielectric response functions. Gradient photonic crystals have a specific optical property: since each layer is characterized by a gradually varying particle diameter, a combination of layers allows one to obtain a set of band gaps in the light reflection spectrum. In other words, a gradient grating forms a chirped photonic crystal [5,6].

It is characterized by optical isotropy: the reflection curve is preserved as the angle of incidence of light on the crystal surface changes. This feature expands the scope of application of a gradient structure, since it functions as a three-dimensional reflector and may be used in devices requiring a wide viewing and detection angle.

The research into gradient photonic structures is focused on their properties, advancement of techniques for their formation, and possible usage scenarios [7–10]. In order to produce a direct, composite, or inverse gradient photonic crystal with specified output characteristics, one needs to use mathematical models of the effects that arise in the interaction of radiation with a gradient multilayer structure. Existing applications for modeling of optical elements, such as the open-source OghmaNano tool, are used widely in the calculation of solar cells, optical filters, and organic light-emitting diodes. Unfortunately, these applications cannot make an allowance for the gradient nature of a photonic crystal. Therefore, in the present study, we performed proprietary modeling to determine the effective refraction index of a gradient photonic structure, which specifies its key optical characteristics: the reflection coefficient and wavelength λ of the maximum reflection coefficient in the photonic band gap.

Wavelength λ was determined in accordance with the Wulff–Bragg condition

$$N\lambda = 2\sqrt{\frac{2}{3}}d\sin\theta n_{eff}, \tag{1}$$

where N is the diffraction order ($N = 1$ for synthetic opal), d is the crystal sphere diameter, θ is the Bragg diffraction angle, and n_{eff} is the effective refraction index of the material.

A photonic crystal consisting of a monolayer of spheres of the same diameter with refraction index n_{sp} and air in the intersphere space with refraction index n_{air} has an effective refraction index given by the following expression [11]:

$$n_{eff} = \sqrt{n_{sp}^2 f_{sp} + n_{air}^2 f_{air}}, \tag{2}$$

where f_{sp} is the factor of filling the crystal with spheres and f_{air} is the factor of filling the intersphere space of the crystal with air.

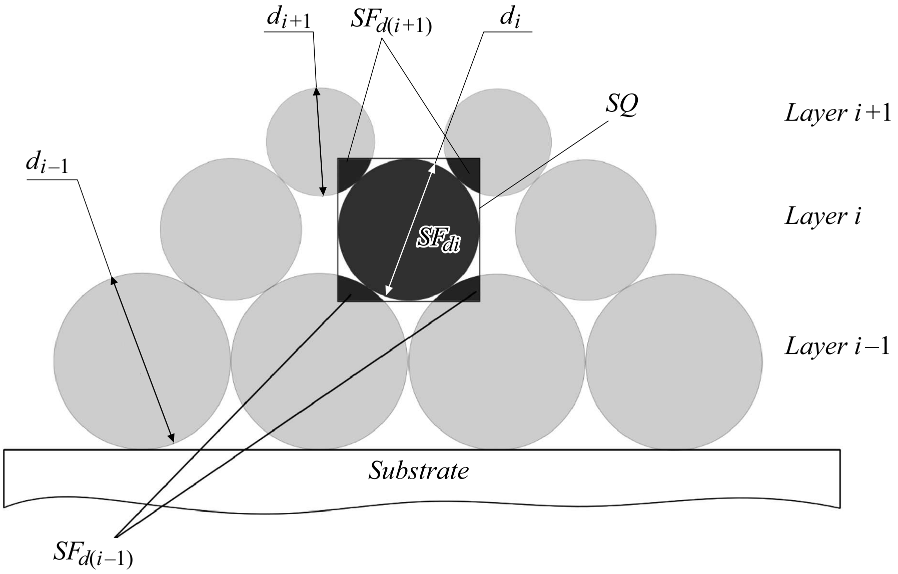

Figure 1. Geometry of the calculation layer.

Comparison of the model and the experiment

Layer formula	Peak number in Fig. 4, *b*	λ_{des}, nm	λ_{exp}, nm	Relative error δ, %
0−400−300	*1*	863	850	1
400−300−200	*2*	660	670	1
300−200−0	*3*	474	484	2

A nanocomposite photonic crystal is a structure modified by the introduction of particles of additional materials with the aim of adjusting the output optical parameters or enhancing the functionality of the structure. The introduced functional fragments with intrinsic refraction index n_{int} fill a fraction of space occupied by air; therefore, factor f_{int} of filling with the introduced material is also used. The effective refraction index may then be calculated as follows:

$$n_{eff} = \sqrt{n_{sp}^2 f_{sp} + n_{air}^2 f_{air} + n_{int}^2 f_{int}}. \tag{3}$$

The factor of filling the volume of a photonic crystal with a certain component is defined as the ratio of the volume occupied by this component to the volume of the structure as a whole, so that $f_{sp} + f_{int} + f_{air} = 1$. To solve the problem layer by layer, we introduce the concept of a unit area of a cleaved photonic crystal face: a square with side d and its center coinciding with the center of a sphere. This assumption limits the domain of calculations and allows one to simplify the actual calculation structure and subsequent mathematical description (including the description of derivative structures).

The calculation model for determining the filling factors of a gradient crystal is proposed in Fig. 1, where unit area SQ of a cleaved photonic crystal face is highlighted in black. This region includes the sphere of calculation layer i and fragments of the adjacent layers. The geometry of the system specifies the expression for calculating the factor of filling the crystal with the sphere material

$$f_{sp} = \frac{SF_{di} + 2SF_{d(i+1)} + 2SF_{d(i-1)}}{SQ}, \tag{4}$$

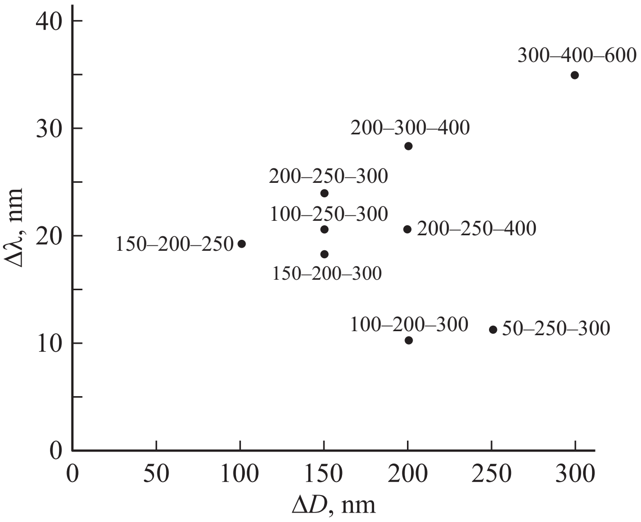

Figure 2. Diagram of the dependence of PBG wavelength shift $\Delta\lambda$ on the difference in layer diameters.

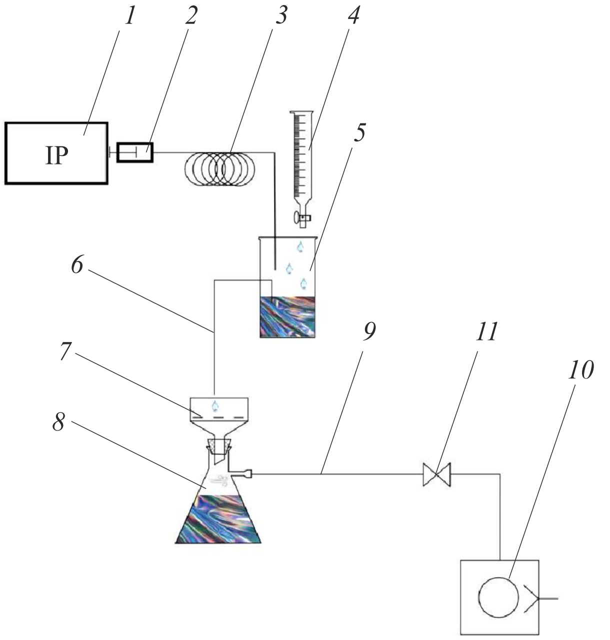

Figure 3. Schematic diagram of the setup for fabrication of gradient structures. *1* — Infusion pump, *2* — syringe, *3* — tube with a special inset, *4* — burette, *5* — chemical beaker, *6* and *9* — connecting hoses, *7* — Buchner funnel, *8* — Bunsen flask, *10* — vacuum pump, and *11* — check valve. The original version of the circuit was missing elements *9–11*.

where SF_{di} is the cross-sectional area of the sphere of the calculation layer, $SF_{d(i-1)}$ is the cross-sectional area of fragments of the lower layer falling within the calculation domain, and $SF_{d(i+1)}$ is the cross-sectional area of fragments of the upper layer falling within the calculation domain.

The problem of determining the wavelength corresponding to the maximum reflection coefficient was solved in accordance with the Wulff–Bragg condition under the following assumptions:

– the particle size increases „from the bottom up": ideally round nanospheres with the largest diameter form a closely packed layer on a glass-ceramic substrate;

– the center of nanospheres of subsequent layers lies on the projection of the point of contact of the lower spheres, and the layer formed by them is not packed closely in the plane parallel to the surface of the substrate;

– the gradient structure is formed from particles with their size varying from 50 to 500 nm in 50 nm steps.

The ΔD parameter, which is a sum of the absolute values of the difference between $d_i - d_{i+1}$ and $d_i - d_{i-1}$, was introduced in order to estimate the influence of particle sizes in layers $i+1$ and $i-1$. Parameter $\Delta\lambda$ characterizes the shift of wavelength λ within a unit area of a cleaved gradient photonic crystal face relative to the same parameter in a monolayer of particles with diameter d_i.

The values of $SF_{d(i-1)}$ and $SF_{d(i+1)}$ are determined by double integration of the equation of a part of a circle with variable integration limits for the plane parallel to the substrate plane. The solution to the problem is to calculate the value of f_{sp} for subsequent analysis of the modeled structure with the use of known formulae (1) and (3).

A diagram (Fig. 2) illustrating the dependence of shift $\Delta\lambda$ on the difference in layer diameters was plotted in order to visualize the set of calculation results. For example, a point with coordinates (150; 23) characterizes the shift of the photonic band gap peak for the calculation layer from 557 nm for a monolayer to 580 nm in the gradient structure with formula „200–250–300" (i.e., a photonic crystal formed by monolayers of particles with diameters of 200, 250, and 300 nm stacked in accordance with the simulated sample diagram; Fig. 2).

It was found in calculations that shift $\Delta\lambda$ is no smaller than 15 nm when $i+1$ and $i-1$ layers are formed with a significant (more than 50 nm) difference in particle diameters falling within the range of 50–500 nm.

Samples with polystyrene particles with a diameter of 400, 300, and 200 nm were prepared by layer-by-layer evaporation of a solution on a heated glass-ceramic substrate and deposition onto the surface of a filter for experimental testing of the mathematical model of a gradient photonic crystal. The obtained samples were imaged with a Crossbeam 550 (Carl Zeiss Microscopy, Germany) scanning electron microscope (SEM) and examined with an EPSILON (IZOVAK, Belarus) optical spectrophotometer.

In layer-by-layer evaporation, a certain amount of colloidal solution was poured onto the substrate, and the solvent was evaporated at elevated temperatures. Since polystyrene is extremely unstable at high temperatures, the processing temperature did not exceed 50 °C. To prevent premature coagulation during sedimentation, a solution with a concentration of 0.5 % was used. Peeling of the already deposited layers at the edges due to contact with the liquid phase was suppressed by reducing the volume of each subsequent portion by 40 % relative to the preceding one.

In the process of filtering, the colloidal suspension was deposited fractionally onto a hydrophilic polytetrafluoroethylene filter with pores 200 nm in size through which the dispersion medium was removed. The diagram of the setup used for this purpose is shown in Fig. 3. The resulting gradient photonic crystal film formed on the filter was transferred to the substrate. The structure and reflection spectra of the produced gradient photonic crystals were similar for samples obtained by different methods. A typical structure is shown in Fig. 4, *a*; the SEM image of the cleaved face of a sample with a total thickness of 4522 nm reveals layers with different particle diameters. However, these layers are not continuous, which is why particles from one layer fill the discontinuities of other layers and layer intermixing occurs. This is also evidenced by the results of measurements of layer thicknesses in SEM images. The analysis of reflection spectra from the surface parallel to the substrate surface (Fig. 4, *b*) reveals the presence of three PBGs, suggesting that a structure containing layers of particles of different diameters was formed. The interference of radiation scattered by them yields a pattern that agrees with

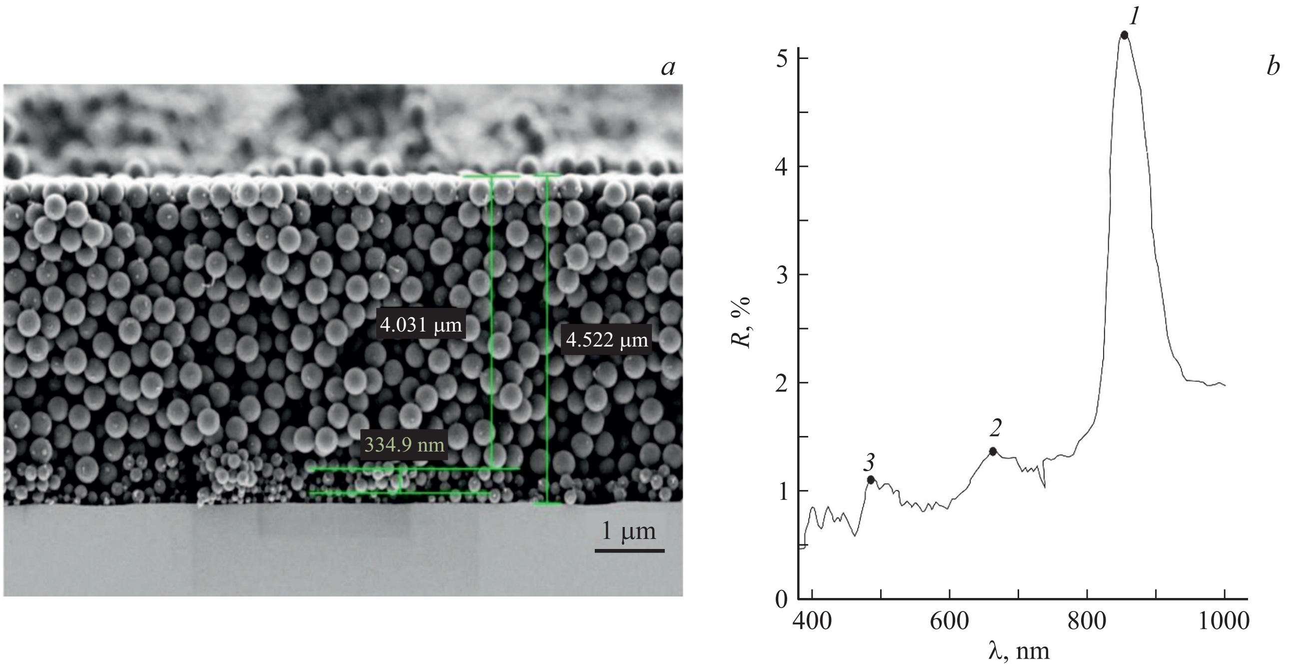

Figure 4. Experimental results: *a* — SEM image of the cleaved face of a sample with a gradient structure on the substrate side; *b* — reflection spectrum of the sample separated from the substrate recorded from the gradient structure side. The sample was produced by layer-by-layer evaporation.

theoretical models. The difference between the calculated wavelengths corresponding to the maximum coefficient (λ_{des}) and the values obtained after spectrophotometry of the experimental sample (λ_{exp}) (see the table) may be attributed to the significant thickness of the upper layer, the above-mentioned intermixing of layers, and the uncertainty of the polystyrene particle size. Polystyrene particles may get deformed due to internal elastic stresses in the course of sample formation, since a heated substrate was used. To prevent the formation of these defects in future studies, we propose to use exclusively the filtration method in combination with a pumping device in order to produce a pressure drop in the Bunsen flask for more efficient removal of the dispersion medium.

The influence of contacting structure layers on the effective refraction index of a photonic crystal formed by particles with a continuous gradient of diameters is taken into account in the proposed model. The comparison of calculated values with experimental data revealed an average discrepancy no greater than 1.7 %, which verifies the applicability of the developed approach. The obtained results may be used in the design of structures with extended PBGs, lasers, coherent light sources, and optical signal analysis devices and in the development of techniques for their production with the aim of expanding the range of Russian electronic components.

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] R.R. Kumar, H.K. Tsang, Opt. Lett., **46** (1), 134 (2020). DOI: 10.1364/OL.409784

[2] M. Pan, X.B. Li, C. Xiong, X. Chen, L. Wang, X. Chen, Y. Li, Part. Part. Syst. Charact., **37** (4), 1900495 (2020). DOI: 10.1002/ppsc.201900495

[3] Y. Häntsch, G. Shang, B. Lei, B. Winhard, A. Petrov, M. Eich, K.P. Furlan, ACS Appl. Mater. Interfaces, **13** (17), 20511 (2021). DOI: 10.1021/acsami.1c01392

[4] A.E. Schedl, I. Howell, J.J. Watkins, H.W. Schmidt, Macromol. Rapid Commun., **41** (8), 2000069 (2020). DOI: 10.1002/marc.202000069

[5] J.P. Vigneron, V. Lousse, Opt. Quantum. Electron., **39**, 377 (2007). DOI: 10.1007/s11082-007-9098-0

[6] I.S. Amiri, A.N.Z. Rashed, K. Sarker, B.K. Paul, K. Ahmed, J. Opt. Commun., **44** (3), 333 (2023). DOI: 10.1515/joc-2019-0146

[7] B. Zhou, Z. Qi, D. Yan, Angew. Chem. Int. Ed., **61** (39), e202208735 (2022). DOI: 10.1002/anie.202208735

[8] M. Schöttle, T. Lauster, L.J. Roemling, N. Vogel, M. Retsch, Adv. Mater., **35** (7), 2208745 (2023). DOI: 10.1002/adma.202208745

[9] A.H. Gevorgyan, N.A. Vanyushkin, M.S. Rafayelyan, Eur. Phys. J. Plus., **137** (6), 719 (2022). DOI: 10.1140/epjp/s13360-022-02944-9

[10] A.H. Gevorgyan, N.A. Vanyushkin, I.M. Efimov, Opt. Mater., **148**, 114839 (2024). DOI: 10.1016/j.optmat.2024.114839

[11] V.S. Gorelik, Quantum Electron., **37** (5), 409 (2007). DOI: 10.1070/QE2007v037n05ABEH013478.

Translated by D.Safin