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SYNTHESIS AND PROPERTIES OF SiO_2 PHOTONIC CRYSTALS MODIFIED BY DNA

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Background: Photonic crystals are structures characterized by periodic modulations of the refractive index with a period commensurate with the wavelength. This periodicity is associated with the existence of a complete band gap in the spectrum of the electromagnetic states of the crystal. The stop zone is called the band gap for the highlighted direction in the crystal. Globular photonic crystals are called three-dimensional photonic crystals, which consist of the same diameter globules. The pores between the globules in the opal allow one to change the refractive index and optical contrast of the material. The task of controlling the stop-zone frequency limits of a globular photonic crystal without changing its physical structure is of practical interest. The easiest way to control the stop-zone parameters is to fill the pores of the photonic crystals with materials with different refractive indices, for example, DNA. Control of the optical parameters of a globular photonic crystal can be used for the creation of optical detectors, sensors, test systems, a quantum biocomputer as well as analyzing and studying a conformational state of DNA.
Objectives: the creation of SiO_2 globular photonic crystals modified by DNA and studying of the influence of DNA on their optical properties.
Materials and Methods: Ethyl alcohol, distilled water, ammonium hydroxide, tetraethoxysilane and DNA were used to synthesize SiO_2 photonic crystals. Aqueous DNA solution was used to infiltrate the photonic crystals. We used a visible range spectroscopy for optical experiments and a finite-difference time-domain (FDTD) method for numerical calculations**.**
Results: SiO_2 globular photonic crystals modified by DNA were synthesized with 195 nm globules. The reflection spectra of the obtained photonic crystals were measured. A red-shift of the stop-zone maximum after the infiltration of photonic crystals with DNA molecules was found. The electric field distribution was calculated for the photonic crystal with 200 nm globules**.**
Conclusions: FDTD calculations in the linear mode show that the presence of point defects in the structure of the photonic crystal influences the amplification of the local electric field in the interglobular space of the photonic crystal, which houses the DNA molecule at infiltration. The DNA infiltration into the pores of a photonic crystal changes the effective refractive index of the system by 5.99%. Synthesis SiO2 photonic crystals with DNA leads to the formation of a more ordered structure at the macro levels. Thus, DNA serves as a template-like structure for photonic crystals to be assembled on. In this case, the effective refractive index of the system increases by 6.01%.
KEY WORDS: SiO_2 globular photonic crystal; stop-zone; a molecule of DNA.

СИНТЕЗ І ВЛАСТИВОСТІ SiO_2 ФОТОННИХ КРИСТАЛІВ, МОДИФІКОВАНИХ ДНК

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Актуальність. Фотонними кристалами називають структури, яким властиві періодичні модуляції коефіцієнта заломлення з періодом, співмірним з довжиною світлової хвилі. З наявністю цієї періодичності пов'язане існування повної забороненої зони в спектрі власних електромагнітних станів кристалу. Стоп-зоною називають заборонену зону для виділеного напрямку в кристалі. Особливий інтерес становлять тривимірні фотонні кристали, які складаються з глобул однакового діаметру. Такі структури називають глобулярними фотонними кристалами. Наявність в фотонному кристалі пор між глобулами дозволяє змінювати показник оптичного контрасту матеріалу. Задача керування положенням забороненої зони глобулярного фотонного кристалу без зміни його фізичної структури становить практичний інтерес. Найпростішим способом керування параметрами

забороненої зони є заповнення пор фотонних кристалів матеріалами з різними показниками заломлення, наприклад, ДНК. Керування оптичними параметрами глобулярного фотонного кристалу може бути використано для створення оптичних детекторів, сенсорів, тест-систем, квантового біокомп'ютера, а також аналізу та вивчення конформаційного стану ДНК.
Мета роботи. Метою роботи є створення SiO_2 глобулярних фотонних кристалів, модифікованих ДНК, та вивчення впливу ДНК на їх оптичні властивості.
Матеріали і методи. Для синтезу SiO_2 фотонних кристалів використовувався спирт етиловий, дистильована вода, гідроксид амонію, тетраетоксисилан, ДНК. Було застосовано оптичну спектроскопію видимого діапазону для експериментальних досліджень та математичний пакет — метод моделювання скінченних різниць в часовій області (FDTD) — для чисельних розрахунків.
Результати. Було синтезовано SiO_2 глобулярні фотонні кристали, модифіковані ДНК, з діаметром глобул 195 нм. Виміряно спектри відбивання одержаних фотонних кристалів. Показано зсув в червону область максимуму стоп-зони фотонних кристалів при наявності в них молекул ДНК. Проведено розрахунки розподілу електричного поля в структурі фотонного кристалу з діаметрами глобул 200 нм.
Висновки. Розрахунки методом FDTD в лінійному режимі показують, що наявність точкових дефектів в структурі фотонного кристалу впливають на підсилення локального електричного поля в міжглобулярному просторі фотонного кристалу, в якому розміщена молекула ДНК при інфільтрації. Інфільтрація ДНК в SiO2 фотонний кристал викликає зміни ефективного показника заломлення системи на 5,99%. Синтез SiO2 фотонного кристалу з ДНК призводить до утворення більш впорядкованої структури фотонного кристалу на макрорівні. Отже, ДНК є своєрідним темплейтом при створенні фотонного кристалу. При цьому ефективний показник заломлення системи збільшується на 6,01%.
КЛЮЧОВІ СЛОВА: SiO_2 глобулярний фотонний кристал; стоп-зона; молекула ДНК.

СИНТЕЗ И СВОЙСТВА SiO_2 ФОТОННЫХ КРИСТАЛЛОВ, МОДИФИЦИРОВАННЫХ ДНК

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Актуальность. Фотонными кристаллами принято называть структуры, которые обладают периодическими модуляциями коэффициента преломления с периодом, сопоставимым с длиной волны. С этой периодичностью связано существование полной запрещённой зоны в спектре собственных электромагнитных состояний кристалла. Стоп-зоной называют запрещённую зону для определённого направления в кристалле. Особый интерес представляют трёхмерные фотонные кристаллы, состоящие из глобул одинакового диаметра. Такие структуры принято называть глобулярными фотонными кристаллами. Наличие в опале пор между глобулами позволяет изменять показатель оптического контраста материала. Задача управления положением запрещённой зоны глобулярного фотонного кристалла без изменения его физической структуры имеет практический интерес. Самым простым способом управления параметрами запрещённой зоны является заполнение пор фотонных кристаллов материалами с разными показателями преломления, например, ДНК. Управление оптическими параметрами глобулярного фотонного кристалла мажет быть использовано для создания оптических детекторов, сенсоров, тест-систем, квантового биокомпьютера, а также анализа и изучения конформационного состояния ДНК.
Цель работы. Целью работы является создание SiO_2 глобулярных фотонных кристаллов, модифицированных ДНК, и изучение влияния ДНК на их оптические свойства.
Материалы и методы. Для синтеза SiO_2 фотонных кристаллов использовались этиловый спирт, дистиллированная вода, гидроксид аммония, тетраэтоксисилан, ДНК. Использовалась спектроскопия в видимом диапазоне для оптических экспериментов и математический пакет — метод моделирования конечных разностей во временной области (FDTD) — для расчетов.
Результаты. Были синтезированы SiO_2 фотонные кристаллы, модифицированные ДНК, с диаметром глобул 195 нм. Измерены спектры отражения полученных фотонных кристаллов. Показано смещение в красную область максимума стоп-зоны при инфильтрации фотонных кристаллов молекулами ДНК. Проведены расчёты распределения электрического поля в структуре фотонного кристалла с диаметрами глобул 200 нм.
Выводы. Расчеты методом FDTD в линейном режиме показывают, что присутствие в структуре фотонного кристалла точечных дефектов влияет на усиление локального электрического поля в межглобулярном пространстве фотонного кристалла, в котором располагается молекула ДНК при инфильтрации. Инфильтрация ДНК в SiO2 фотонный кристалл приводит к изменению эффективного показателя преломления системы на 5,99 %. Синтез SiO2 фотонного кристалла с

ДНК приводит к образованию более упорядоченной структуры фотонного кристалла на макроуровне. Таким образом, ДНК может быть своеобразным темплейтом при создании фотонного кристалла. При этом эффективный показатель преломления системы увеличивается на 6,01 %.
КЛЮЧЕВЫЕ СЛОВА: SiO_2 глобулярный фотонный кристалл; стоп-зона; молекула ДНК.

Photonic crystals (PC) are space-periodic structures whose permittivity varies with a period commensurate with the wavelength in the visible and near-infrared ranges. The PC has either a full three-dimensional band gap associated with the periodicity of the structure [1] or a stop-zone (a band gap in a given direction in the crystal) [2]. The interaction of the electromagnetic field with such structures is determined by the interference effects that accompany the diffraction of visible light in three-dimensional periodic lattice, which can be compared with a strong modification of the energy spectrum of the electromagnetic modes [3, 4]. Among the most promising materials for PC formation are synthetic opals formed by monodisperse spherical SiO_2 particles, which form densely packed layers parallel to growth surfaces [2, 5], colloidal structures composed of TiO_2 spherical microparticles [6, 7], and inverted opals [8, 9, 10]. Technological features of the growth of synthetic photonic structures in different laboratories lead to the samples with different properties and different degree of ordering of the crystal lattice. Therefore, studying the effect of the real crystalline structure of the PC on the stop-zone parameters is still an urgent task [11].

Up to 26% of the total volume of a PC makes pores formed between globules [12]. By infiltrating the PC with different substances, e.g. DNA, it is possible to model a new conformation of DNA, namely, stretching DNA [12] and to change drastically the effective refractive index of the system with DNA, and therefore to control its optical parameters [13]. Studies of the active centers infiltrated into a photonic crystal are of fundamental interest and allow us to investigate the unique optical properties of PC composites. Activated by rare earth ions, SiO_2 spheres can be used as active elements in nanosensors, microlasers [14, 15], and luminescent markers [16]. Particles of highly dispersed SiO_2 have an effective ability to adsorb amino acid and DNA molecules onto their surface [17]. Increased interest in such systems is associated with the ability to effectively control the propagation of light inside the PC [2]. Controlling of the optical parameters of a globular PC can be used for creation of optical isolators, modulators, polarization converters, detectors, sensors, optical computers, or test systems.

In this work we study the influence of DNA on the forming of SiO_2 photonic crystal and its stop-zone parameters and do numerical experiments of light propagation through the PC structure.

MATERIALS AND METHODS

In this work, SiO_2 photonic crystal and DNA-SiO_2 photonic crystal were synthesized.

SiO_2 photonic crystal

In the first stage of the process, the synthesis of a monodisperse suspension of SiO_2 spherical particles was performed using a modified Stöber method [18] by hydrolysis of tetraethoxysilane (TEOS) in alcohol-aqueous medium with ammonium hydroxide as a catalyst. The size of the particles depends on the concentration of the components in the reaction mixture. Specifically, a mixture of C_2H_5OH (96%) — 25 ml, TEOS-1 ml, H_2O-13.25 ml, NH_4OH (25%) — 0.89 ml was used. The resulting aqueous suspension of these particles was placed in a cell for a long period (up to 1 month). Under the action of gravity, SiO_2 particles were deposited at the bottom and self-assembled into a three-dimensional periodic structure. In the second step, the precipitate obtained was dried and annealed in a muffle furnace for 12 hours: first at 150°C, and then the temperature was elevated to 750°C and the samples were kept at this temperature for 4 hours. The obtained samples have a porous structure with a continuous void network formed between SiO_2 spheres.

SiO_2 photonic crystal infiltered with DNA

Deoxyribonucleic acid (DNA) sodium salt from salmon testes (Sigma-Aldrich CAS Number 438545-06-3) was used to infiltrate the SiO_2 photonic crystal. 1 mg of DNA was added to 0.5 ml of distilled water. The solution was maintained at 5°C for 24 hours. During this time, it became gel-like. The PC specimens were twice covered with the gel solution with a periodicity of 3 h until complete drying.

DNA-SiO_2 photonic crystal

Another experiment was carried out in such a way that 1 mg of DNA was added to 13.25 ml of distilled water at the first stage of the SiO_2 synthesis-with the same proportions of the remaining components as above. So, we used a mixture of C_2H_5OH (96%) — 25 ml, TEOS-1 ml, DNA+H_2O-13.25 ml, NH_4OH (25%) — 0.89 ml. The resulting aqueous suspension with DNA was placed in a cell for a 1 month for getting three-dimensional periodic structure. However, the precipitate obtained was not dried in an oven but was maintained for 10 days at room temperature. As result we obtained the DNA-SiO_2 photonic crystals.

The photo of the surface of the DNA-SiO_2 photonic crystal is shown in Fig. 1. The photo was obtained with DM 2500M Leica microscope and has a 50x magnification. A clear structure of the synthesized crystals can be observed.

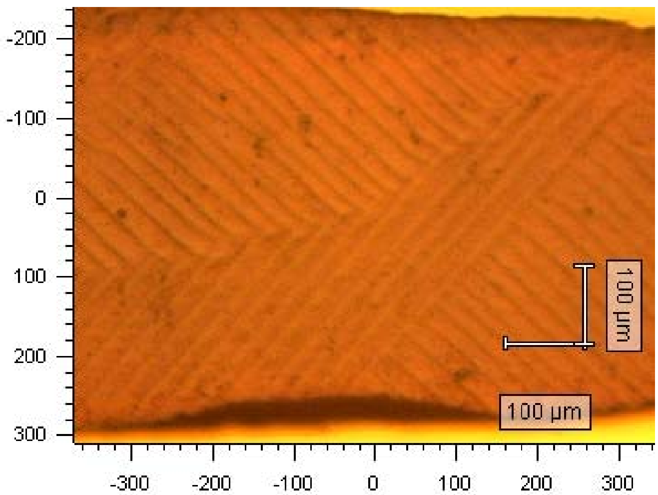

Fig. 1. Microscopic image of DNA-SiO_2 photonic crystal.

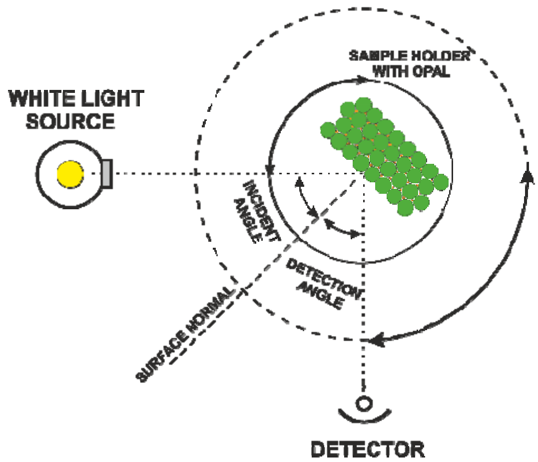

Fig. 2. Optical scheme for the Bragg scattering measuring.

Spectrometer Ocean Optics USB 2000 (USA) was used. The scheme of the optical experiment is presented in Fig. 2. Broadband white light from a halogen lamp was incident onto the PC surface. It was possible to change a sample holder and a detector angular position to get an angular dependence of the PC reflection spectra.

To simulate light wave propagation through PC structure we used Lumerical high-performance photonic simulation software FDTD [19].

RESULTS AND DISCUSSION

Optical experiments

We experimentally investigated the optical properties of the SiO_2 photonic crystal as well as the SiO_2 photonic crystal infiltrated with DNA, and DNA-SiO_2 photonic.

Fig.3. shows the reflection spectra measured at different angles of incidence light for: (a) SiO_2 photonic crystal synthesized according standard technology; (b) SiO_2 photonic crystal infiltrated with DNA; (c) DNA-SiO_2 photonic crystal without annealing.

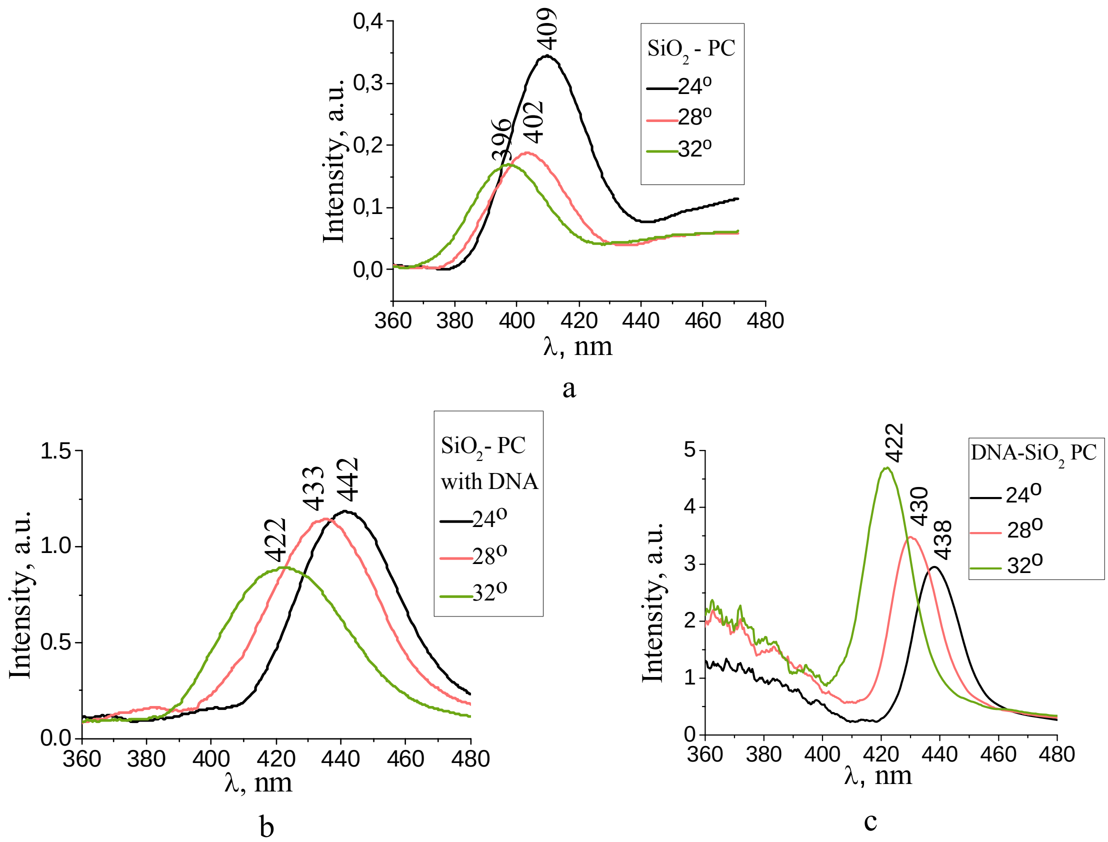

Fig. 3. Reflection spectra of (a) SiO_2 photonic crystal, (b) SiO_2 photonic crystal infiltrated with DNA and (c) DNA-SiO_2 photonic crystals measured at the light incidence angles 24°, 28°, and 32°.

The position of the stop-zone is determined by the angle of incidence light. For pure SiO_2 photonic crystal (Fig. 3a), the stop-zone with a maximum at λ = 409, 402 and 396 nm is observed at the light incident angles of 24°, 28°, and 32°, respectively. By the formula

$$\lambda_{max} = 2D\sqrt{2/3} \cdot \sqrt{n_{eff}^2(\lambda) - sin^2(\theta)} \quad (1)$$

we can estimate the size of the PC globules. There, λ_{max} is stop-zone maximum; D — diameter of PC globules; $n_{eff}(\lambda)$ — effective refractive index, calculated as

$$n_{eff}(\lambda) = \sqrt{\sum_{i=1}^{N} n_i^2(\lambda) \cdot f_i} \quad (2)$$

and depending on the refractive index $n_i(\lambda)$ of i-components comprising the system, and their volume fraction f_i; θ — light incident angle. For a pure photonic crystal, we have a two-component system consisting of SiO_2 globules and the air. In publications [20, 21] it was shown that the pores make up 26% of the total PC volume. Therefore, we take f_1 = 0.74 and f_2 = 0.26. More information about the calculated parameters n_{eff} (λ) and D, measured values of λ_{max} at different angles, the refractive indexes taken from work [22], and their volume fraction are presented in Table 1, block "SiO_2-PC".

Table 1. Experimental values and calculated parameters of the studied PCs.

Light incidence angle Θ	Stop-zone maximum λ_{max}, nm	n_{SiO_2}	f_{SiO_2}	n_{air}	f_{air}	n_{H_2O}	f_{H_2O}	n_{DNA}	f_{DNA}	n_{eff}	D, nm
SiO_2-PC											
24°	409	1.4692	0.74	1.0003	0.26					1.3629	192.55
28°	402	1.4699	0.74	1.0003	0.26					1.3635	192.32
32°	396	1.4706	0.74	1.0003	0.26					1.3640	192.94
SiO_2-PC infiltrated with DNA											
24°	442	1.4662	0.74	1.0003	0.02	1.3373	0.18	1.6031	0.06	1.4446	195.26
28°	433	1.4669	0.74	1.0003	0.02	1.3377	0.18	1.6052	0.06	1.4454	193.97
32°	422	1.4679	0.74	1.0003	0.02	1.,3381	0.18	1.6079	0.06	1.4446	192.29
DNA-SiO_2 PC											
24°	438	1.4665	0.74	1.0003	0.02	1.3375	0.18	1.6031	0.06	1.4449	193.46
28°	430	1.4672	0.74	1.0003	0.02	1.3378	0.18	1.6052	0.06	1.4456	192.59
32°	422	1.4679	0.74	1.0003	0.02	1.3381	0.18	1.6079	0.06	1.4464	192.03

DNA infiltration into the pores of the PC (Fig. 3b) leads to a red-shift of the stop-zone maximum by 33, 31, and 26 nm at the light incidence angles of 24°, 28°, and 32°, respectively. The studied system should be considered as a four-component one consisting of SiO_2 glubules, which occupy 74% of the total volume, as well as air, water and DNA, accounting for 26% of the total volume. In the process of DNA infiltration, the air is displaced from the pores by the DNA aqueous solution. However, within a margin of error, not more than 2% of the air can remain in the pores. DNA and water occupy the remaining 24% of interglobular volume. The calculated data are presented in Table 1, "SiO_2-PC infiltrated with DNA" block. The effective refractive index of the system "SiO_2-PC infiltrated with DNA" increases by 5.99%.

The addition DNA into the suspension of spherical SiO_2 particles showed that the stop-zone maximum (Fig. 3c) is red-shifted by 29 nm, 28 nm, and 26 nm at the light incidence angles of 24°, 28°, and 32°, respectively. We observe a peculiarity in the reflection spectra: as the angle of incidence increases, the intensity of the reflected light increases. This is due to the reduction of scattering losses in the presence of aqueous DNA solution in the pores, and due to the PC was not annealed, since the PC annealing at high temperatures would result in DNA burning. The addition of DNA into the PC suspension leads to the formation of a more ordered structure, as evidenced by the narrowing of the half-width of the Bragg peak in the reflection spectrum by 15 nm. The effective refractive index of the system "DNA-SiO_2 PC" increases by 6.01%. So, DNA influence on the forming of structure of photonic crystal. DNA serves as template-like structure for photonic crystals to be assembled on. The calculated data are presented in Table 1, block "DNA-SiO_2 PC".

Previously, it was shown in work [12] that the introduction of water into the pores of the PC leads to a slight shift of the stop-zone maximum (by 5 nm). Infiltration of DNA aqueous solution into the pores of the PC as well as DNA introduction into the SiO_2 suspension at the stage of synthesis, leads to more significant optical changes.

Numerical experiments

FDTD is a numerical technique used for modeling computational electrodynamics. It is a really powerfull tool for modeling optical and electrical effects in PC. If you want to find field distribution inside the PC, the FDTD method is one of the most advanced. You take a mesh and replace the continuum space by a discrete set of nodes. Then solve the system of Maxwell's equations for the discrete mesh using boundary conditions.

Synthetic opal was built of SiO_2 spheres packed in a face-centered cubic (FCC) lattice. The calculated refractive index was 1.46, the SiO_2 sphere diameter was 200 nm, which is comparable with the experimental data. There were 10 layers in Z direction. The plane waves with linear polarization and different wavelengths were incident on the surface in [111] direction.

We first calculated the opal stop-zone wavelength using the Bragg law. After this we calculated the transmission spectrum of a broadband source using FDTD method and compare the results. In both cases the stop-zone wavelength was 440 nm. According to the obtained experimental results, for the further simulation we choose the wavelength 440 nm — as the value that matches the center of the opal stop-zone.

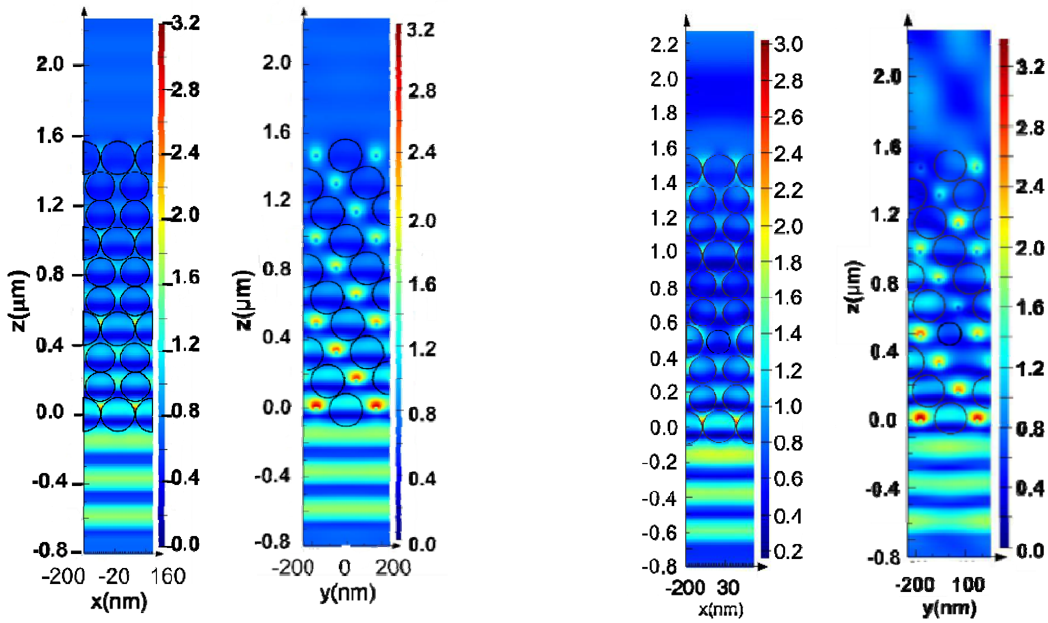

Fig. 4 Electromagnetic field distribution in XZ and YZ opal planes calculated for the wavelength 440 nm propagation in: (a) perfect periodic structure, (b) structure with defects

Near-field electromagnetic field distribution in XZ and YZ opal planes for the wavelength 440 nm was calculated at the following approximations. The electromagnetic pulse propagates in the z-direction, spheres diameter — 200 nm with the refractive index n =1.45 are surrounded by air; light is linearly polarized in the y-direction. Figure 4a presents the simulation results. The right picture (Fig. 4b) corresponds to opal with defects. The left picture (Fig. 4a) corresponds to opal with perfect periodic structure.

The results were taken from three orthogonal plane monitors. The black circles mark the opal silica globules. The color scale shows the intensity of the electric field at different space points. The electric field at some regions of the PC is about 3 times larger than the incoming field. The filed energy tends to concentrate in the regions with low permittivity, in our case in the air pores between silica globules.

As a next step, opal periodic structure with two point defects was considered. One sphere was completely removed and the diameter of another adjacent sphere was reduced by one third. The field distribution was calculated for 440 nm wavelength (Fig. 4b). The field pattern was the same. All graphs are depicted in two colour scales — maximum field value for the current plane, and maximum value obtained for all measurements. In the case of structural defects that may be present in any real synthetic opal, it can lead to even greater redistribution of the field intensity (~10 times, not for data presented here) [23] and concentration of light energy in the interglobular space. Though the defects are not good for the opal stop-zone, they may cause higher inhomogeneity of the electromagnetic field distribution in the PC structure.

CONCLUSION

The photonic crystals made of SiO_2 and DNA were synthesized by the Stöber method and characterized. It has been shown that DNA can be used as a template-like structure for photonic crystals synthesis leading to a more orderly structure on a macroscale, namely hundreds of microns and more.

Computer simulation of light propagation through the PC structure demonstrates the localization of the electromagnetic field in the regions with less refractive index, i.e. in the pores between the globules of synthetic opal. Thus, for certain wavelength in certain points we can expect an enhance a local field inside the photonic crystal up to 3 times in perfect crystal and order more in defect crystal using it for further application.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest

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