Photonic crystals with an arbitrary number of photonic band gaps made of porous quartz with a gradual change in the refractive index

© S.E. Svyakhovskiy, N.I. Pyshkov

Moscow State University, Moscow, Russia e-mail: sse@shg.ru Received April 24, 2023 Revised June 02, 2023 Accepted June 02, 2023

One-dimensional photonic crystals with an arbitrary number and spectral position of photonic band gaps in the optical spectral range are experimentally demonstrated. The absence of mutual influence of the photonic band gaps was shown. No features corresponding to higher harmonics or combination frequencies were found in the spectral response of the multifrequency photonic crystals created.

Keywords: photonic crystals, photonic band gap, quasicrystals.

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1. Introduction

Photonic crystals (PCs) are optical media that have their refraction index spatially periodically modulated in one or more dimensions [1]. The periodicity of PCs results in a photonic band gap (PBG) in their optical spectrum, i.e. a frequency region where light propagation is impossible [2]. Thanks to the PBG, photonic crystals are used to effectively control light, to concentrate and amplify the field of light wave, and to enhance linear and nonlinear optical effects.

Many tasks require simultaneous amplification of the optical response at several frequencies, for example, generation of the second [3] and the third [4] optical harmonics, cascade generation of harmonics [5]. In such applied areas as laser technology [6], production of photosensitive matrices [7], spectral filtration [8], protection from powerful laser radiation, PCs are required simultaneously with several PBGs. Moreover, nearly any spectral positions of these PBGs can be on demand depending on the problem being solved.

Several different techniques have been proposed for creating PCs with multiple PBGs. Combining several photonic crystal lattices into one [9,10] according to various logical rules makes it possible to obtain multiple PBGs, however, parasitic phenomena are present in the spectrum along with them: multiple frequencies that take place in any photonic lattice with its profile different from the ideal harmonic function [11], combination frequencies that arise from the summation of two spatial lattices [12]. Another approach is to create quasicrystalline [13] and aperiodic deterministic structures [14,15], which have found successful application in problems of photoluminescence signal amplification [16], optical filtration [17], and many others.

Previously, in [10] the authors experimentally demonstrated quasiperiodic two-frequency photonic crystals based on porous silicon oxide. Two-level refraction index modulation was used, which caused the presence of artifacts in the form of multiple and combination frequencies in the reflectance spectrum, which were successfully reduced thanks to the correct analytical selection of the refraction index modulation function. However, the two-level modulation method turned out to be unsuitable for implementing three or more PBGs in one crystal.

In this study, we demonstrate quasiperiodic multifrequency photonic crystals with the spectral response having three or more independent PBGs. The crystals were fabricated using electrochemical etching of silicon with continuous modulation of the etching current, resulting in continuous spatial modulation of the refraction index. As a result, the spatial profile of the refraction index approximates a harmonic function or the sum of a number of such functions with an accuracy sufficient for the fabrication of high-quality photonic structures.

2. Methods

The reflectance spectra of photonic crystals were theoretically calculated using the propagation matrix method, described, for example, in [18]. The continuous change in refraction index was approximated by a step function, thus the photonic crystal structure was approximated by a onedimensional multilayer sequence. It is known [19] that this method is correct. In the calculation, the thickness of a single layer was chosen to be 20 nm. The modeling step along the spectrum was 1 nm, unless otherwise indicated.

Experimental samples of photonic crystals were prepared using the method of electrochemical etching of silicon; the procedure for electrochemical etching is described in detail in [20]. In this study, the following parameters are used: continuous modulation of the etching current density, minimum and maximum current densities of $j_{min} = 40 \text{ mA/cm}^2$ and $j_{max} = 180 \text{ mA/cm}^2$, the concentration of fluoric acid in electrolyte solution of 28%. Photonic crystals were formed due to periodic spatial modulation of the refraction index.

During the electrochemical etching, pores are formed inside the silicon. In the (001) crystallographic oriented

silicon used in this study, the pore growth occurs vertically downward. The pore diameter depends monotonically on the etching current density, therefore it can be controlled during the etching process creating a spatial modulation of the pore thickness and, consequently, the etching current density.

The pore size in the fabricated porous silicon in the considered range of current densities ranged from 20 to 50 nm, which was measured using a scanning electron microscope [21]. This pore size is much smaller than the wavelength of radiation in the visible range, which allows considering the porous medium as optically homogeneous.

After fabrication, the photonic crystal (PC) samples were subjected to thermal oxidation, as a result of which silicon was oxidized to silicon oxide. The porous structure of the samples was preserved. Due to the fact that the refraction index of a continuous layer of silicon oxide is less than the refraction index of silicon, the refraction index of the porous substance decreases during oxidation.

The refraction index values of the final porous silicon oxide were measured from the interference spectra of homogeneous layers with a thickness of $10\,\mu\text{m}$ without modulation of the refraction index. For current densities of j_{\min} , j_{\max} the refraction indices were 1.22 and 1.14, the middle of this range $n_0 = 1.18$ was taken as the average value of the refraction index against which modulation was applied.

The reflectance spectra of photonic structures were measured using the technique described in [20] and [10]. Briefly: all spectra were measured at normal light incidence on the sample; the light source was a halogen lamp with a spectrum of 400-1200 nm, and the detector was a OceanInsight QEPro spectrometer.

3. Spectrum of a sinusoidal lattice

Let us imagine a transparent medium with an average refraction index $n_0 = 1.18$ and optical thickness $n_0L = 50 \,\mu$ m. Let the coordinate axis x be laid out in the depth of this medium and measured in units of the optical path. Let us introduce in this medium a spatial modulation of the refraction index according to a harmonic law of the following form:

$$n(x) = n_0 + \delta n \sin\left(\frac{4\pi}{\lambda_0}\right),$$
 (1)

where δn is modulation amplitude, λ_0 is some wavelength. The modulus of the reciprocal lattice vector of such a structure is $k = 4\pi/\lambda_0$. It is known [22] that when a light wave with a wave vector whose modulus is half that of k, namely $2\pi/\lambda_0$, is incident normally on a structure, this wave will experience diffraction reflection in the opposite direction. Therefore, at a wavelength of λ_0 there will be a PBG in the reflectance spectrum.

To test the theoretical and experimental methods used, a PC with n modulation according to (1) and with parameters

1.0 1.21 0.8 1.1 Reflectance 0.6 n 0.5 1.0 1.5 2.0 $x, \mu m$ 0.4 Theory 0.2 Exp 200 300 400 500 600 700 800 900 Wavelength, nm

Figure 1. Measured reflectance spectrum of PCs with sinusoidal spatial modulation of the refraction index in comparison with the calculated one. The inset shows the spatial profile of the refraction index.

 $\lambda_0 = 850 \text{ nm}, \ \delta n = 0.02 \text{ was fabricated, measured, and its}$ reflection spectrum was calculated (Fig. 1).

The sample obtained in the experiment has a band gap center at a wavelength of 853.5 nm. The theoretical spectrum was also built up for this wavelength. The inset shows the sinusoidal refraction index profile approximated by the step function from which the reflection spectrum of the multilayer structure was calculated.

With the exception of a shift of 3.5 nm relative to the planned one, the fabricated PC fully corresponds to expectations, as can be seen from the comparison of the experimental and theoretical spectra. The shape of the PBG in terms of width and amplitude corresponds to the calculated one, the period of lateral oscillations coincides with the theoretical one.

It should be noted that both the theoretical and experimental spectra are completely free of high-order parasitic zones. The asymmetry of the sinusoid would manifest itself in the presence of a second-order zone, which, even if present in a real sample, has an amplitude less than the experimental noise.

There are no high-order zones in the theoretical spectrum, which is indicative of the fact that the approximation of the sinusoid by a step function does not affect the calculation results.

4. Combination frequencies and higher harmonics

Let us consider a quasiperiodic two-frequency refraction index lattice of the following form:

$$n(x) = n_0 + \delta n \sum_{j=1}^N \sin\left(\frac{4\pi}{\lambda_j}\right), \qquad (2)$$





Figure 2. Calculated reflectance spectrum of a quasiperiodic dual-frequency PC with given wavelengths of $\lambda_1 = 700$ nm, $\lambda_2 = 990$ nm. Inset shows zoomed in ranges of 409–411 nm and 2375–2405 nm.

where N = 2, and check whether there are higher harmonics and combination frequencies in its spectrum. As part of this study, two-frequency lattices were numerically modeled in the range of crystal thicknesses *L* from 100 to 500 μ m; the lattice frequencies varied within the optical range: λ_1 remained constant and was equal to 700 nm, λ_2 varied from 710 to 1400 nm, and the spectral ranges corresponding to combination frequencies and higher harmonics were modeled with increased spectral resolution of 0.01 nm.

The manifestation of combination frequencies in the spectrum is expected at wavelengths of

$$\frac{1}{\lambda_{\pm}} = \frac{1}{\lambda_1} \pm \frac{1}{\lambda_2}.$$
 (3)

According to the modeling results, combination frequencies and higher harmonics were almost absent in the spectrum. These effects are too subtle and can only be observed in high-Q crystals, which means large optical thickness of the crystal and strict adherence to periodicity. They could be observed in the calculated spectrum only with a significant increase in the optical thickness and modulation of the refraction index (Fig. 2).

The modeling parameters were $\lambda_1 = 700$ nm, $\lambda_2 = 990$ nm, $\delta n = 0.05L = 500 \,\mu$ m. It should be noted that the last two parameters are the technologically permissible limits for the sample fabrication technology under consideration.

The expected spectral positions of the combination frequencies, according to (3), are at wavelengths of $\lambda_+ = 410$ nm, $\lambda_- = 2390$ nm. The insets in Fig. 2 show the corresponding fragments of the spectrum.

So, with the maximum technologically permissible refraction index contrast and crystal thickness, it is found that the sum frequency has the form of a peak with a width of 0.1 nm, and the difference frequency has the form of a dip with a depth of 0.015. In addition, two maxima were discovered at wavelengths of 350 nm and 495 nm, which correspond to the second harmonics of the fundamental frequencies. Their amplitude is less than the amplitude of the maximum at λ_+ . It was decided that detecting such subtle effects in experiments is very difficult and does not make much sense, since the effects are parasitic.

Also, it should be noted that the found wavelengths at which the combination peaks have the maximum amplitude are in the following approximate relation to each other $\lambda_2/\lambda_1 \approx \sqrt{2}$. This frequency ratio is considered dissonant in the music theory due to the strong combinational harmonics.

5. Implementation of structures with arbitrary band gaps

To implement PCs with multiple band gaps, a series of three-frequency PCs with modulation of form (2)) at N = 3 were modeled and experimentally created. Fixed values of $\lambda_1 = 475$ nm, $\lambda_3 = 950$ nm were chosen, the λ_2 took 7 equidistantly chosen values from the interval of $[\lambda_1, \lambda_3]$.

The theoretical reflectance spectra of a series of 7 samples are shown in Fig. 3; the spectrum of each sample exhibits 3 band gaps without any additional spectral features. Fig. 4 shows, as an example, the structure of samples 2 and 6, namely the refraction index profile calculated according to (2). The profile has the form of a quasiperiodic multi-frequency harmonic function approximated by a step function.

This series of 7 samples was implemented experimentally. Fig. 5 shows the corresponding reflectance spectra.

The experimental implementation of a series of 7 samples meets theoretical expectations: there are three band gaps in the spectrum, the outermost of them are in the same spectral ranges of 470-480 nm and 940-960 nm, respectively, the middle band gap shifts from sample to sample, the position of the middle band gap is approximately



Figure 3. Calculated reflectance spectra of a series of PCs with three PBGs. The graphs are shifted by unity relative to each other.

equidistant. The spectra have no other spectral features; there are minor lateral oscillations.

Based on the results of the above, an experimental sample was created with five band gaps (Fig. 6), the outermost of them are located at wavelengths of $\lambda_1 = 475$ nm, $\lambda_5 = 950$ nm, the middle ones are located equidistantly in this range.

First, the refraction index sequence (2) was specified with N = 5. This sequence, in the form of the same table of refraction indices and layer thicknesses, was used both for theoretical calculations and for numerical control of the electrochemical etching setup program. Fig. 6 shows comparison of the theoretical and experimental spectra. It follows from the graph that spectral positions of the five experimental maxima differ from the calculated ones by 2.3 nm, 2.9 nm, 2.9 nm, 1.9 nm, 2.4 nm, respectively. We



Figure 4. Section of the spatial profile of the refraction index of a PC with three PBGs in the range of $50-54 \,\mu\text{m}$, samples N° 2 and N° 6.



Figure 5. Measured reflectance spectra of PCs with three PBGs. The graphs are shifted by unity relative to each other.



Figure 6. Experimentally measured reflectance spectrum of the photonic crystal with five band gaps in comparison with the theoretically calculated one.

consider this agreement between theoretical and experimental results to be very good.

In this case, we could choose an arbitrary arrangement of PBG central wavelengths, so it should be noted that an equidistant arrangement of the band gaps in wavelengths is more difficult to implement and more demanding on the quality of the sample than, for example, an equidistant arrangement in frequencies, because in the latter case multiple and combination frequencies may be masked by other band gaps.

6. Conclusion

This study has demonstrated a method for producing multiband PCs with an arbitrary number of band gaps. The method features the complete absence of interference between band gaps; with reasonable experimental parameters, no multiple or combination frequencies are detected in the reflectance spectrum. High-quality PC samples with 3 and 5 band gaps in arbitrary spectral regions were experimentally obtained.

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Conflict of interest

The authors declare that they have no conflict of interest.

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