The Synthesis and Calculation of Heterostructural Photonic Crystals with Large Bandgap

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Abstract. Heterostructural photonic crystals (HPC) have attracted considerable interest due to its large photonic bandgap. Here we report a facile method to calculate and fabricate the heterostructural photonic crystals by using optical transfer-matrix method and polystyrene (PS) spheres with different particle sizes. Results suggest that the numerical simulation match well with the experimental spectra. In addition, compared with the single-structural photonic crystals (SPC), the HPCs have larger photonic bandgap, which is equal to a total of the photonic bandgaps of each SPC. Therefore, the HPCs with large bandgap can be well designed and then synthesized.

1. Introduction

The photonic crystal, proposed by Yablonovitch [1] and John [2] in 1987, is a dielectric structure with the periodical refractive index. Electromagnetic waves [3-5] of specific frequency in a photonic crystal can be absorbed completely. Therefore, the photonic crystal is also known as the photonic bandgap materials. With excellent performances such as high optical transmission and extremely fast response ability, the photonic crystals have attracted significant attention in recent years.

To obtain larger photonic bandgap, considerable work has been done and it has been found that photonic crystal with different structures or patterns may enlarge the photonic bandgap efficiently. Early research shows that heterostructures can effectively enlarge the frequency range of multiple-channeled filters [6] using numerical simulation method. Recently, theoretical calculation [7] demonstrates that heterostructural photonic crystals with large bandgap are composed of a series of photonic bandgaps of different structural photonic crystals.

At present, the researches on HPCs mainly base on the theoretical deduction, however, relatively lack in the combination of theory with experimental results. In this paper, the preparation of HPCs with PS spheres were introduced first. Then the calculation of HPC was established and the numerical simulated results of the photonic bandgap were obtained using optical transfer-matrix method [8,9]. Results illustrate that the calculated location and breadth of the bandgap are in accordance with that of the optical spectra obtained from experimental fabrication. Thus, HPC can be designed well according to the calculation and has great potential in photonic crystal fiber [10,11] and filter[12].
2. Experimental Preparation

2.1. Preparation of PS microspheres
PS spheres are prepared by non-soap emulsion polymerization of experiments in three groups. The details are as follows: 1) to add 100ml deionized water to four-neck flask and then ventilate nitrogen for disposing-oxygen; 2) to add styrene monomer with its dosage respectively in 8 ml, 9 ml and 10 ml successively in three groups of experiments when the system was slowly heated to 60°C; 3) when the system was heated to 70°C, adding 0.2g aqueous solution containing potassium persulfate as the initiator and 1.2g α-methacrylic acid as the surface modifier, through 24h reaction under the protection of nitrogen, the polystyrene (PS) microspheres dispersion liquid was obtained; and 4) after repeated centrifuge washing and ultrasonic cleaning, the PS microspheres samples were obtained and labeled as PS-A, PS-B and PS-C, kept in the refrigerator.

2.2. Preparation of heterostructural photonic crystals
HPCs were synthesized by the multiple vertical deposition method. First, the single-structural photonic crystals, PC-A, PC-B, PC-C, were deposited respectively using the PS microspheres samples PS-A, PS-B and PS-C. Secondly, the PS microspheres were placed in the beaker of the ethanol solution (volume fraction as 30%), then hydrophilic treated glass slides were vertically immersed into the solution. The vertical deposition was carried on at 66°C as well as a humidity at 90%. Finally, the as deposited photonic crystal samples were heated at 85°C for 5 hours. Afterwards the HPCs were prepared by adopting the multiple vertical deposition method in different order. Table 1 shows the samples obtained in the corresponding deposition sequence.

| Table 1 The heterostructure composition of photonic crystal samples |
|---------------------------------|-----------------|
| Sample Number | Composition & Order |
| PC-AB | PC-A→PC-B |
| PC-BA | PC-B→PC-A |
| PC-AC | PC-A→PC-C |
| PC-CA | PC-C→PC-A |

3. Results and discussions

3.1. Transfer-matrix method

![Figure 1. SEM images of surface of the heterostructural photonic crystal samples: (a) Sample PC-A; (b) Sample PC-B and (c) Sample PC-C.](image-url)
Figure 2. SEM images of section of the heterostructural photonic crystal samples: (a) PC-BA and (b) PC-CA.

Figure 1. shows that PS microspheres in the photonic crystal were packed densely and arranged in order. The particle size of PC-A is about 155nm, the particle size of PC-B is about 190nm and the particle size of PC-C is about 200nm. Figure 2. demonstrates the interface between different single-structure photonic crystals marked in white lines. On both sides of the interface, PS microspheres have smaller particle size above the line.

3.2. Transfer-matrix method

Figure 3. Model diagram of transfer-matrix theory.

Considering the wave propagation in the materials composed of alternating arrangement with different relative dielectric constant $\varepsilon$ (material refractive index $N$) and different thickness $d$ of the dielectric layer as shown in Figure 3., supposing incident wave is plane wave, then the tangential value of the electric and magnetic fields below the interface of the first layer is $E_1, H_1$. By inference, the tangential value of the electric and magnetic fields below the interface of the $N$ layer is $E_N, H_N$. When the light transmits the single-layer medium, the relationship of light wave fields both on the dielectric incident surface and on the dielectric exit surface can be shown by the matrix:

\[
\begin{bmatrix}
E_1 \\
H_1
\end{bmatrix} =
\begin{bmatrix}
\cos \delta_i & -\frac{i}{\eta_i} \sin \delta_i \\
-i\eta_i \sin \delta_i & \cos \delta_i
\end{bmatrix}
\begin{bmatrix}
E_2 \\
H_2
\end{bmatrix}
\]

Matrix $M_j$ is a characteristic matrix of the single-layer medium:
$$M_j = \begin{bmatrix} \cos \delta_j & -i \frac{\sin \delta_j}{\eta_j} \\ -i\eta_j \sin \delta_j & \cos \delta_j \end{bmatrix}$$

(2)

$$\delta_j = \frac{2\pi}{\lambda} N_j d_j \cos \theta; \eta_j = \frac{N_j}{\cos^2 \theta}; N_j$$ is the refractive index of the single-layer medium; $$d_j$$ is the thickness of the single-layer medium. For a periodical structure, the following formula is obtained by continuous application of the above transmission relationship:

$$\begin{bmatrix} E_1 \\ H_1 \end{bmatrix} = M_1 M_2 \ldots M_N \begin{bmatrix} E_{N+1} \\ H_{N+1} \end{bmatrix} = \begin{bmatrix} A & B \\ C & D \end{bmatrix} \begin{bmatrix} E_{N+1} \\ H_{N+1} \end{bmatrix}$$

(3)

According to the formula above, the transmittance coefficient $$T$$ can be solved when the wave propagation occurs in the photonic crystal as follows:

$$T = \left( \frac{2\eta_0}{A\eta_0 + B\eta_0\eta_{N+1} + C + D\eta_{N+1}} \right)^2$$

(4)

3.3. Properties of single-structural photonic crystals

Figure 4. Model of the single-structural photonic crystal: (a) Composition diagram and (b) Physical model diagram.

SEM images indicate that the deposited photonic crystals are formed periodically with two different refractive indexes materials, polystyrene and air, as shown in Figure 4a. Suppose the normal angle between the incident light and the photonic crystal interface is 0 degree, i.e. $$\cos \theta = 1$$, then the photonic crystal along the light incident direction can be regarded as the periodical alternating arrangement of PS layer with the thickness $$d_1$$, refractive index $$N_1$$ and air dielectric layer with the thickness $$d_2$$, refractive index $$N_2$$, as shown in Figure 4b.

Suppose $$M_1$$ represents the matrix of the PS and $$M_2$$ represents the matrix of the air, then the matrix of the whole photonic crystal is $$M = [M_1 M_2]$$, of which $$\delta_1 = \frac{2\pi}{\lambda} N_1 d_1$$, $$\delta_2 = \frac{2\pi}{\lambda} N_2 d_2$$, $$\eta_1 = N_1$$, $$\eta_2 = N_2$$ ($$N_1 = 1.59$$ representing the PS refractive index, $$N_2 = 1$$ representing the air refractive index, $$d_1 = d_2$$). Using the particle sizes of samples PC-A, PC-B and PC-C as parameters for calculation ($$d_1 = 155nm$$ for PC-A; $$d_2 = 190nm$$ for PC-B; $$d_2 = 200nm$$ for PC-C), the numerical simulation (calculated by MATLAB 7) and experiment results are shown in Figure 5.
Figure 5. Spectra comparison of single-structural photonic crystals between numerical simulation and experiment results: (a) PC-A; (b) PC-B and (c) PC-C.

Figure 5. shows the sample PC-A with bandgap central wavelength of 402nm and bandwidth of 38nm in calculation and with bandgap central wavelength of 396.5nm and bandwidth of 26nm in experiment; the sample PC-B with bandgap center wavelength of 493nm and bandwidth of 46nm in calculation and with bandgap center wavelength of 493.5nm and bandwidth of 35nm in experiment; and the sample PC-C with bandgap center wavelength of 520nm and bandwidth of 50nm in calculation and with bandgap center wavelength of 526nm and bandwidth of 41nm in experiment. Still with the minimal deviation, the photonic bandgap locations of the three samples in both numerical simulation and experiment matched well. The deviation may result from the following factors: the approximation of isotropic, non-dispersive and non-magnetic medium, the instability of environmental condition in the preparation of photonic crystals, the intrinsic absorption of polystyrene material in the optical examination. Consequently, results imply that, as PS particle size increases, the SPCs show red shifted and larger photonic bandgap.

3.4. Properties of heterostructural photonic crystals
In experiment, the heterostructures can be obtained by adjusting PS spheres’ particle size. For instance, first depositing the photonic crystal A using PS particle in the same size, then preparing the photonic crystal B of PS particle in another size, and so on, as shown in Figure 6a. Suppose the normal angle between the incident light and the photonic crystal interface is 0 degree as shown in Figure 6b.

Figure 6. Model diagram of the heterostructural photonic crystal: (a) Composition diagram and (b) Physical model diagram.

Suppose $M_1$ represents a characteristic matrix of the PS dielectric layer and $M_2$ represents a characteristic matrix of the air dielectric layer in the photonic crystal A, then the characteristic matrix of the photonic crystal PC-A can be expressed as $M_A = [M_1M_2]^n$, in the same way,
\[ M_B = \left[ M_3 M_4 \right]^{n_b}, \quad M_C = \left[ M_5 M_6 \right]^{n_c}, \ldots \]

Thus, the characteristic matrix of heterostructural photonic crystals can be expressed as:

\[ M = M_A M_B M_C \ldots = \left[ M_1 M_2 \right]^{n_1} \left[ M_3 M_4 \right]^{n_3} \left[ M_5 M_6 \right]^{n_6} \ldots \]

Using the photonic crystal samples PC-AB, PC-BA, PC-AC and PC-CA, the calculation of heterostructures is carried out: \( d = 155nm \) of \( A \), \( d = 190nm \) of \( B \) and \( d = 200nm \) of \( C \). The numerical simulation and experimental results are shown in Figure 7.

![Spectra comparison of double heterostructural photonic crystals between numerical simulation and experiment results: (a) PC-AB; (b) PC-BA; (c) PC-AC and (d) PC-CA.](image)

Figure 7 shows that two peaks with the bandwidth of 38nm and 46nm located at 402nm and 493nm in the numerical simulation of sample PC-AB and sample PC-BA while in experiment two peaks with the bandwidth of 22nm and 30nm appeared at 397nm and 492.5nm, also, two peaks with the bandwidth of 38nm and 50nm appeared at 402nm and 520nm in the numerical simulation of sample PC-AC and sample PC-CA while in experiment two absorption peaks with the bandwidth of 24nm and 41.5nm appeared at 397.5nm and 524nm. Still with the minimal deviation, the photonic bandgap locations of the samples in both numerical simulation and experiment matched well. The two sets of data in both numerical simulation and experiment show that the bandgap of HPCs is equal to a total of the bandgap of each SPC, for instance, the two bandgap positions in sample PC-AB are in accordance with that in sample PC-A and PC-B respectively. In another word, the superposition fits the commutative law, unrelated to the superposition order.

4. Conclusion

HPC samples are successfully synthesized with PS spheres. The photonic bandgap of HPCs were calculated using the optical transfer-matrix method. It is found that the numerical simulation results match well with the experimental spectra. The conclusions are shown as follows: 1) as PS particle size increases, SPC show red shifted and larger photonic bandgap; 2) photonic crystals using heterostructures can enlarge the photonic bandgap obviously; 3) the bandgap of HPC is equal to a total of the bandgaps of each SPC, unrelated to the superposition order. Therefore, based on this work, the
required photonic crystal with special performance such as heterostructures, large photonic bandgap and filtering properties can be well designed.

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