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Photonic Bandgap Structures of Core-Shell Simple Cubic Crystals from Holographic Lithography

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Comments

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Photonic Bandgap Structures of Core-Shell Simple Cubic Crystals from Holographic Lithography

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We report the investigation of photonic bandgap properties of a core-shell simple cubic structure (air core with a dielectric shell) using a two-parameter level-set approach. The proposed structure can be obtained by partially backfilling high refractive index materials into a polymeric template fabricated by multi-beam interference lithography. We find that the shell formation in the inverted simple cubic structure increases the complete photonic bandgap width by 10 – 20 % in comparison to that of a completely filled structure. The bandgap between the 5th and 6th bands begins to appear at a refractive index contrast of 2.7. This study suggests the importance to investigate the core-shell formation in three-dimensional photonic crystals through backfilling, which may offer an additional control over their photonic bandgap properties.

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A three-dimensional (3D) photonic crystal (PC) that possesses a complete photonic band gap (PBG) is highly desirable to confine, manipulate and guide photons for a broad range of applications, including low-threshold lasers,[1] light-emitting devices,[2] optical biosensors,[3] and microphotonic devices.[4] To fabricate 3D photonic crystals, many methods have been used, such as self-assembly of colloidal particles and block copolymers,[5,6] layer-by-layer photolithography,[7] direct-write assembly,[8] and two-photon lithography.[9] Among them, interference (or holographic) lithography[10] is a very promising candidate, which enables rapid production of defect-free 3D crystals over a large area with considerable control over both lattice size and symmetry.

In holographic lithography, the focused laser beams interfere to generate periodic patterns in photosensitive polymers, which, however, typically have low refractive indices. For example, the refractive index of SU-8 photoresist is ~1.6. Therefore, back-filling with a higher index inorganic material is needed, followed by removal of the polymer template to fabricate PC with a complete PBG. Chemical vapor deposition and liquid phase deposition have been used to infiltrate high index materials, such as titania ($n = 2.5 – 3.0$),[11] silicon ($n = 3.5$),[12] and germanium ($n = 4.0$)[13] into sacrificial polymeric templates. It should be noted that the deposition reaction usually occurs between the liquid or vapor precursors and the corresponding functional groups on the template surface. A shell structure is formed at first, which grows continuously normal to the initial surface to fill the interstitial voids (e.g. Fig 1(b) and (c)). Previously it was found that in a partially filled inverted face-centered cubic opal, the shell formation could enhance the PBG, and the bandwidth of a directional L-stopgap could be tuned with the core-to-shell ratio.[14,15]

In this letter, we use a two-parameter level-set approach to investigate the PBG properties of core-shell formation in a simple cubic P (Pm3m) structure. Of the various PC structures, diamond D and gyroid G have received much attention due to their large gap width and robustness.[16-19] However, it is not straightforward to produce these structures by multi-beam interference lithography since each term in the level-set equation is dependent on all 3D (x, y, and z) coordinates. The accessible lattice size is limited by the choice of beam parameters and the wavelength of the laser. In comparison, the simple cubic P (Pm3m) structure can be size-scalable via triple exposures of a two-beam interference pattern, where the angle between the beams of the individual gratings can be varied.[20] This primitive structure shows a relatively wide and full PBG with a maximum gap to mid-gap ratio of 13% between 5th and 6th bands for a dielectric contrast of 13:1 and a volume fraction of 0.26.[21] Moreover, the pseudo gaps along the (100) direction (X-gap) in the simple cubic structure are very wide and appeared over a large range of filling ratios.[22] The position of the pseudo gap between the 2nd and 3rd bands is more sensitive to the lattice constant than that of the L-gap in fcc structure, which makes the simple cubic structure attractive for tunable photonic crystals and optical sensors. In this letter we investigate the photonic bandgap properties of the core-shell P structure.

The formation of the P structure has been analyzed via the level-set approach.[23] In this approach, the surface of a porous dielectric structure is represented by the solution to scalar-valued functions $F(x, y, z)$ of three independent variables, and the volume fraction can be controlled by varying the parameter, t. Therefore, the structure is defined as

$$F(x, y, z) > t \text{ for dielectric, and } F(x, y, z) < t \text{ for air} \qquad (1)$$

Using a similar approach, we propose two level-set surfaces of interference patterns to simulate triply periodic structures with a shell-like morphology given by

$$t_1 < F(x, y, z) < t_2 \text{ for dielectric, and}$$

$$t_1 > F(x, y, z) \text{ and } t_2 < F(x, y, z) \text{ for air} \qquad (2)$$

This structure is divided into the inner-core (air) and the outer-shell (dielectric) surfaces defined by t_2 and t_1, respectively. The resulting two level surfaces share the same normal vectors such that they are in parallel to each other. Because the shell morphology can be formed from either liquid phase deposition or CVD process, the use of two t parameters in the equation offers close approximation to the real deposited surfaces. For example, Fig. 1 shows a schematic diagram of a simple cubic structure formed from interference lithography, and the evolution of its replicas from air-core-shell to completely-filled structures. Since the sum of a level surface and the surface translated by half a lattice period gives a constant maximum value in the level-set equation, i.e. $F(-x, -y, -z) = -F(x, y, z)$, the volume fraction is symmetrically related to the exposure intensity. Therefore, the desired volume fraction of the primitive photonic crystals with high refractive index (Figure 1c) can be obtained from templates (Figure 1a) with controlled exposure intensity.

In the bandgap calculation, we use the level-set surface of $F(x, y, z) = \sin(x) + \sin(y) + \sin(z)$ for Eq. 2 and two-parameters, t_1 and t_2, for the outer and inner surfaces of dielectric materials ($n = n_d$), respectively.[20] The band structures of the PC are calculated using the MIT photonic bands (MPB) software package.[24] We first calculate the complete PBG with only a single parameter t_1 (i.e., completely filled structure) ranging from 0.6 to 0.9. A bandgap appears between the 5th and 6th bands. The maximum PBG is found at $t_1 = 0.83 - 0.85$ with $n_d = 3.50$, and the corresponding volume fraction is 0.25 – 0.26, which agrees well with the results from literature.[20] Then, we optimize t_2 to maximize the complete bandgap width while keeping the t_1 fixed (i.e., core-shell structure). For t_1 ranging from 0.65 to 0.90, the maximum PBG is obtained at $t_2 = 2.5 – 2.7$, which is smaller than $t_2 = 3.0$ required for a completely filled simple cubic structure (see Fig 2(a)). Therefore, the formation of shell in a simple cubic structure increases the complete PBG by 10 – 20 % within the range of aforementioned t_1. By analogy, it has been reported that bandgap of 2D square lattice can be increased by the introduction of additional square lattice of smaller unit atoms, which effectively reduces the crystal symmetry.[25] Fig 2(b) shows the calculated photonic band structure of a core-shell P structure with the maximum PBG. It is found that the introduction of an air core lifts the 6th band especially at wavevector M (the bottom of the 6th band), resulting in the increase of PBG. The inset shows the optimized structure. Compared to the volume fraction of 0.26 for completely filled structures, the optimized core-shell P structure has a slightly lower volume fraction of 0.25. However, such photonic bandgap enhancement is not observed in core-shell diamond and gyroid structures.

Figure 3 shows the gap/mid-gap ratios of the P structure shown in Fig 2(b) as a function of the refractive index. The bandgap is calculated from the P surface with the largest PBG, that is $t_1 = 0.84$ for complete filling, and $t_1 = 0.84$ and $t_2 = 2.70$ for a core-shell structure. Previously, it has been reported that the minimum refractive index required to open a complete PBG is approximately 2.0 for D, 2.2 for G and 2.8 for P structures.[21,22] In the core-shell P structure, the minimum index contrast required to open a complete PBG is found to be less than 2.7. Thus, in practice a simple cubic photonic crystal with a complete PBG at optical wavelengths can be achieved by the deposition of anatase titania into photoresist templates.[11]

In conclusion, we have investigated the photonic bandgap properties of a core-shell simple cubic structure using a two-parameter level-set approach. The 3D structure is defined by the inner-core (air) and the outer-shell (dielectric) surfaces. The proposed structure can be fabricated by backfilling a sacrificial polymer template created by multi-beam interference lithography. The photonic bandgap width is found increased in the core-shell primitive structure in comparison to that of the completely filled one. A complete bandgap between the 5th and 6th bands begins to appear at a refractive index contrast of 2.7. This suggests that the core-shell formation may offer additional controls over photonic properties. The two-parameter level-set approach presented in this letter may provide a useful guidance for the fabrication of 3D photonic structures through backfilling.

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Figure Captions:

Figure 1 (color online). Templating method to produce PC of high refractive index materials. (a) Template, (b) air core/dielectric shell, (c) air core, and (d) completely filled structure.

Figure 2.(color online) (a) Complete photonic bandgap of the simple cubic lattice with core-shell and completely filled structures, respectively. The t_2 is optimized with respect to t_1 to maximize the bandgap width. (b) Photonic band structure of the simple cubic lattice with t_1=0.85 and t_2 = 2.7 or 25% filled, and n_d =3.50, showing complete PBG between 5th and 6th bands.

Figure 3. Gap/mid-gap ratio as a function of refractive index contrast in the simple cubic structure shown in Fig. 2(b).

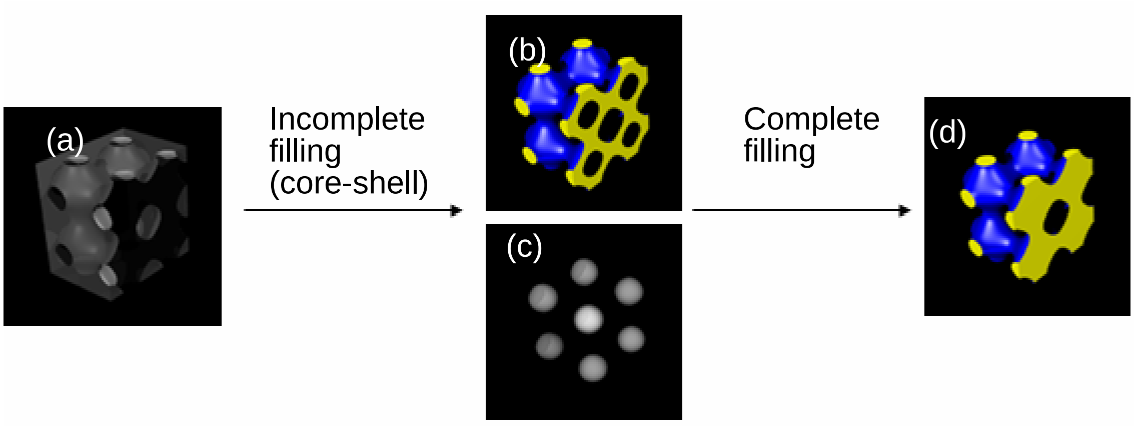

Figure 1 (color online)

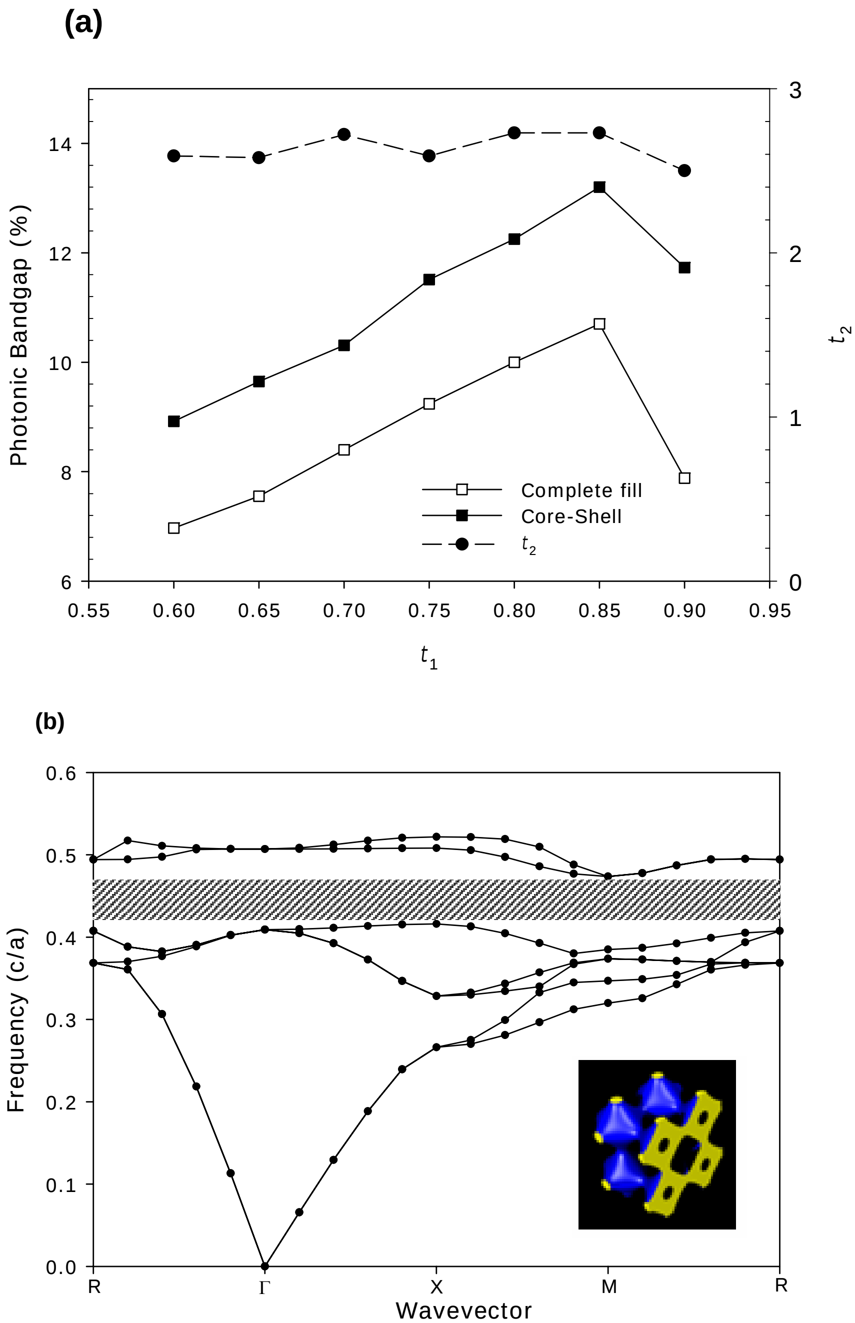

Figure 2 (color online)

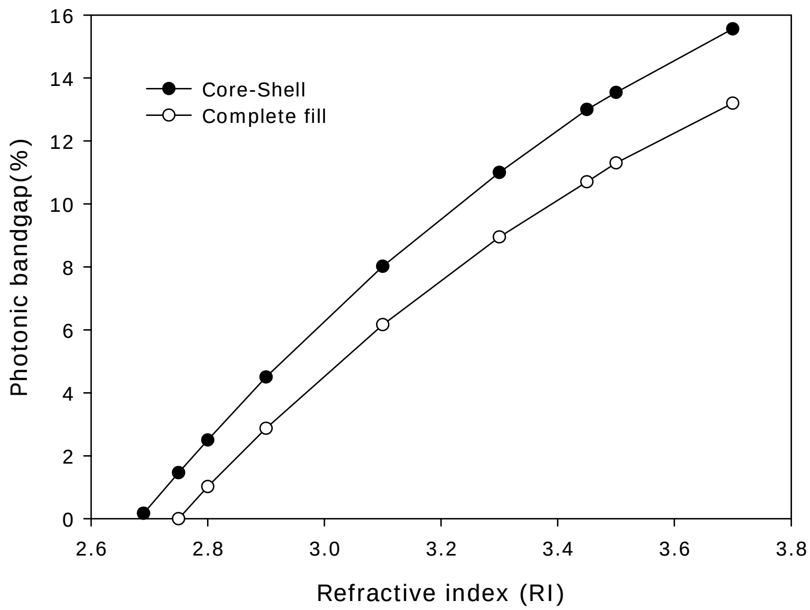
16
14
12
10
8
6
4
2
0
Photonic bandgap(%)
2.6
2.8
3.0
3.2
3.4
3.6
3.8
Refractive index (RI)
Core-Shell
Complete fill

Figure 3