Bicontinuous cubic photonic crystals via level set and 3-D interference lithography

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We present a combined analysis on the creation and fabrication of three-dimensional bicontinuous photonic crystals with large complete gaps based on the modulation of the dielectric material along principal directions and its relation to the interference lithography technique.

1. INTRODUCTION

Interesting phenomena arise from the interaction of electron waves and periodic semiconductor materials. These electronic materials permit the control of electron flows and set the foundations for the technological revolution on computers. A similar system, in which the analogue effects of the periodic atomic potential are created by a periodic dielectric structure, can allow researchers to control the propagation of electromagnetic waves[1]. These artificial "photonic crystals" are three-dimensional composite structures made of high and low dielectric materials that extend periodically in space. An appropriate dielectric architecture, combined with a correct selection of materials, can create a photonic crystal possessing a large and complete three-dimensional photonic band gap[2]. The existence of this photonic band gap creates a medium in which electromagnetic waves, with frequencies within the gap, are forbidden to propagate irrespectively of their propagation direction. Although two-dimensional photonic crystals can also be fabricated, the creation of three-dimensional photonic crystals with large gaps is essential to fully take advantage of the technological applications of these novel materials. They can be used for the inhibition of spontaneous emission, the improvement of semiconductor lasers, wave-guiding, low-loss mirrors, etc[1,3].

The existence of these frequency gaps is an interesting phenomena arising from the basic interaction of waves and periodic structures. It is intrinsically due to the space/temporal cyclic character of the wave and the spatial periodicity of the underlying structure. This interaction between the wave and the structure produces multiple wave scattering with strong interference. The opening of a large complete gap is obtained when the wavelength of the propagating wave is of the order of the spatial periodicity of the structure. This is an important aspect determining the size of the structure unit cell with respect to the frequency range of operation of the photonic

Nanofabrication: Technologies, Devices, and Applications, edited by Warren Y-C. Lai,
Stanley Pau, O. Daniel López, Proc. of SPIE Vol. 5592 (SPIE, Bellingham, WA, 2005)
0277-786X/05/$15 · doi: 10.1117/12.580234

crystal. Since the electromagnetic wave equation is scalable, a particular structure possessing a complete gap for a specific frequency range, will also present a complete gap for higher/lower frequencies if the lattice constant of the structure is increased/reduced. This allows researchers to test structures with predicted photonic properties at large scales where the fabrication process is much simpler. However, in order to control the flow of light, it is necessary to create photonic crystals to operate at optical frequencies, and this set a formidable challenge, to fabricate three-dimensional periodic dielectric structures at sub-micron length scales.

2. THE THEORETICAL PROBLEM

The basic theoretical problem for the photonic crystal community is the following: under which conditions (symmetry, geometry, material properties) does a large and complete gap develop ?.

Yablonovitch[1] proposed the first 3-D complete band gap structure by the generalization of the photonic properties of 1-D systems. However, neither the first proposed 3-D checkerboard lattice nor its experimental version (air spheres in the fcc lattice) possess a complete gap in the fundamental lower bands[4-6]. Later, Ho *et al.*[6] introduced the diamond structure by breaking symmetries in the original fcc structure to lift the degeneracy of the bands that were disallowing the creation of the gap. This diamond structure was the first theoretical photonic crystal exhibiting a complete photonic band gap. Moreover, the diamond gap is so large that strong structural modifications on the diamond structure do not make the gap close. In this regard, researchers can transform the diamond structure into diamond-like structures that are "easy-to-craft"[6-19]. We have recently shown how twenty outstanding 3-D photonic crystals are obtained by the modification of the original diamond structure into fabricatable designs[20]. In particular, the versatility of the diamond structure allows researchers to create diamond-like structures that can be fabricated by the use of the well-developed lithography techniques. A layer-by-layer design of the diamond structure[10] or "woodpile structure" is one of the favorite architectures for the fabrication community to build at several different length scales[21-27]. However, the need for the physical realization of photonic crystals in order to test predicted theoretical properties, and the discovering of some additional intriguing uses as photonic devices, have motivated no research on the original question: *how does a complete gap develop* ?

3. A THEORETICAL SOLUTION

We propose a systematic three-dimensional generalization of one-dimensional photonic crystals[20]. It is known that a one-dimensional multilayer system presents photonic band gaps at least for normal incidence. We represent such a system by the trigonometric function $f(x) = \sin(2\pi x/a)$, where a is the lattice constant of the multilayer system. A parameter t is included in order to control the relative volume fraction of the different materials in the composite. The region for which $f(x) + t > 0$ is filled with high-dielectric material, whereas the region for which $f(x) + t < 0$ is filled with low-dielectric material. When combined with the lattice constant a, the parameter t determines the absolute thickness for each layer. The resultant dielectric material distribution corresponds to a one-dimensional periodic step function with high and low dielectric materials.

The generalization of this one-dimensional description of photonic crystals to three-dimensions is straightforward. An evident 3-D structure can be obtained by the choice of three sinusoidal functions along the *x*, *y*, and *z* axes[20]. For example, the photonic crystal created by the function $f(x)= \sin(2\pi x/a) + \sin(2\pi y/a) + \sin(2\pi z/a) + t = 0$ possesses a maximum 13% complete photonic band gap for a dielectric volume fraction $f=0.26$ [28]. Yablonovitch's original three-dimensional generalization of the one-dimensional gap system was to suggest a 3-D checkerboard structure comprised of cubes of high and low dielectric material. Unfortunately this structure does not have a complete gap[1]. However, by simply modulating the dielectric material along the three normal directions in real space, a complete photonic band gap is created. This simple cubic photonic crystal described by $f(x) + t = 0$ (Fig. 1) is a member of the single P surface family[28,29].

The most general case corresponds to the creation of complete photonic band gap structures by the modulation of dielectric material along several different directions in real space[20]. However, since the creation of the gap arises from the interaction between wave and structure, in order to obtain a complete forbidden frequency range it is reasonable that the modulations along different directions have the same length. This assures that gaps created along different directions overlap. Mathematically speaking, the dielectric modulations should be, in principle, be described by set of wavevectors with same moduli. Some structures with complete gaps that we obtained by the use of this approach are presented in the results section.

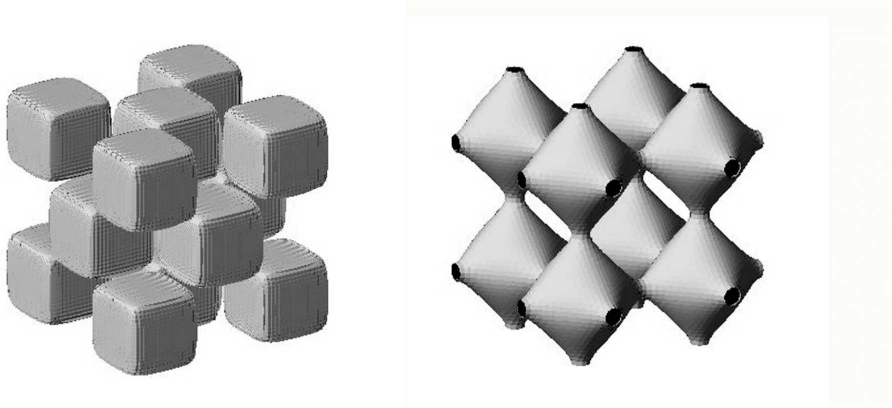

Figure 1. Left: The first proposed photonic crystal (a 3-D checkerboard structure) failed to provide a complete gap. Right: The single P structure. By simply modulating the dielectric material along the three normal directions in real space, a complete photonic band gap is created.

4. THE EXPERIMENTAL PROBLEM

As mentioned previously, the characteristic length of the periodic dielectric structure determines the range of frequencies for which electromagnetic propagation is forbidden. In order to create structures that can be readily used by the telecommunication industry, or to manage visible light, sub-micron length scales are needed to obtain a gap at appropriate frequencies. The lattice constant of the structure is thus an important design parameter. In addition, once a particular architecture possessing a complete gap is identified, its fabrication is not straightforward due to the required small length scales. Moreover, the spatial periodicity of the dielectric structure and the dielectric contrast between the materials are both important characteristics. As a result, the choice of materials, size and specific design of the structure are the three critical parameters to fabricate a photonic crystal for optical wavelengths.

From inspection of the known large complete band gap photonic crystals, it is apparent that photonic crystals with large gaps are bicontinuous. That is, the high and low dielectric regions, which consist of two distinct regions, or phases, are completely connected within themselves and self supporting. However, while bicontinuity is a necessary condition it does not guarantee the existence of a large complete gap. In fact, some bicontinuous structures can present no gap at all. As a result of this necessary condition, the fabrication process must allow the creation of bicontinuous structures. From theoretical analysis, in general, the larger the dielectric contrast between the materials the larger the width of the gap. Thus, fabrication techniques must allow the processing of materials with high dielectric constants and, for optimal conditions, leave one of the connected subspaces as air.

Some of the techniques that demonstrated potential access to three-dimensional bicontinuous structures at small lengths scales are, for example, layer-by-layer lithography[22], and interference lithography[30]. Through the processing of high dielectric constant materials, layer-by-layer lithography has the advantage of producing structures with desired size, architecture, and dielectric contrast at the same time. However, the processing is slow and costly. On the other hand, relatively inexpensive techniques such as interference lithography can produce structures with correct length scales and architectures provided that an appropriate identification of the beam parameters have been made to obtain the desired architecture[31,32]. This is important since not all periodic dielectric structures possess complete gaps even if the architecture looks similar to the diamond structure. In their first stage, structures obtained by interference lithography do not have the dielectric contrast needed to open the gap, but, in a second stage process, the structures can be infiltrated with high dielectric materials and then the polymer network phase degraded and replaced by air such that the necessary dielectric contrast can be reached. In this case, the original polymer network structure to be made is the complement to the final high dielectric netwrok structure.

5. AN EXPERIMENTAL SOLUTION

We propose a combination of the proposed theoretical solution and the interference lithography technique to obtain fabricatable structures with large complete gaps. The modulations along principal directions scheme can be used to create large complete photonic bandgap structures. Due to the analytical representation, this structures are described by level set equations. In fact, these trigonometric representations are Fourier series of low order terms that are compatible with the interference pattern of laser beams. As a result, from the number and type of terms present in the series, it is possible to calculate the beam parameters needed to create the original structure. This three-step process is described in Figure 2. Note that the bicontinuous P-surface is particularly easy for infiltration[33] since the complementary structure (the air dielectric network) is also a single P surface.

1 Identification of a level set photonic crystal with a large complete gap by modulating the dielectric material along principal directions. For example, the single P structure $f(x,y,z) = \sin(2\pi x/a) + \sin(2\pi y/a) + \sin(2\pi z/a) + t = 0$ shows a maximum 13% gap at dielectric volume fraction f=0.26.

2 Calculation of the beam parameters (wave-vectors, polarizations) that create the desired bicontinuous structure by the interference of the beams.

3 Fabrication of the structure by interference lithography where the 3-D interference pattern is transferred into a template followed by infiltration of a high dielectric material.

Figure 2. A three-step scheme to create a photonic crystal with a large complete gap

6. RESULTS

By the use of the dielectric modulations along principal directions scheme, we found some interesting and novel photonic crystals. For example, in the 2-D case, we created a photonic crystal having p4mg symmetry by the use of the function $f(x,y) = \cos[2\pi(2x+y)] - \cos[2\pi(x+2y)] - \cos[2\pi(2x-y)] + \cos[2\pi(x-2y)]$ (Fig. 3). This photonic crystal presents a maximum complete gap of about 6% at $f \sim 0.50$. The quality of the gap is comparable to the gap of the photonic crystal created by only two modulations ($f(x,y) = \cos[2\pi x] + \cos[2\pi y]$), which has p4mm symmetry, but the gap of the p4mg photonic crystal extends for a larger range of volume fractions. However, the use of three principal modulations ($f(x,y) = \cos[2\pi(x-y/\sqrt{3})] + \cos[2\pi(x+y/\sqrt{3})] + \cos[2\pi(2y/\sqrt{3})]$) yields the photonic crystal with the largest gap (18%).

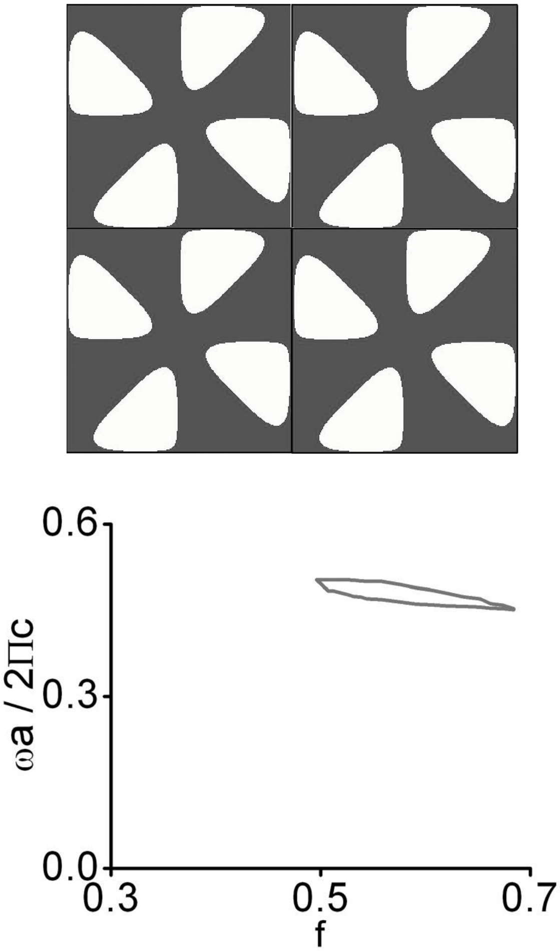

Figure 3 A photonic crystal with p4mg symmetry created by symmetric modulations along principal directions. Grey: high dielectric material, white: air.

In the 3-D case, a structure belonging to the 216 space group can also be obtained by the use of symmetric modulations along principal directions. As it can be seen in Figure 4, this 216 photonic crystal is made of a diamond-like dielectric network. However, the distribution of dielectric material around (0,0,0) is different from the one around (a/4,a/4,a/4). This modification breaks some symmetries within the unit cell and transforms the structure from space group 227 diamond to 216 "unbalanced diamond". More importantly, the structure retains the 2-3 diamond gap and also presents and additional 5-6 gap. This structure, having two simultaneous gaps, can be used for multifrequency amplifiers.

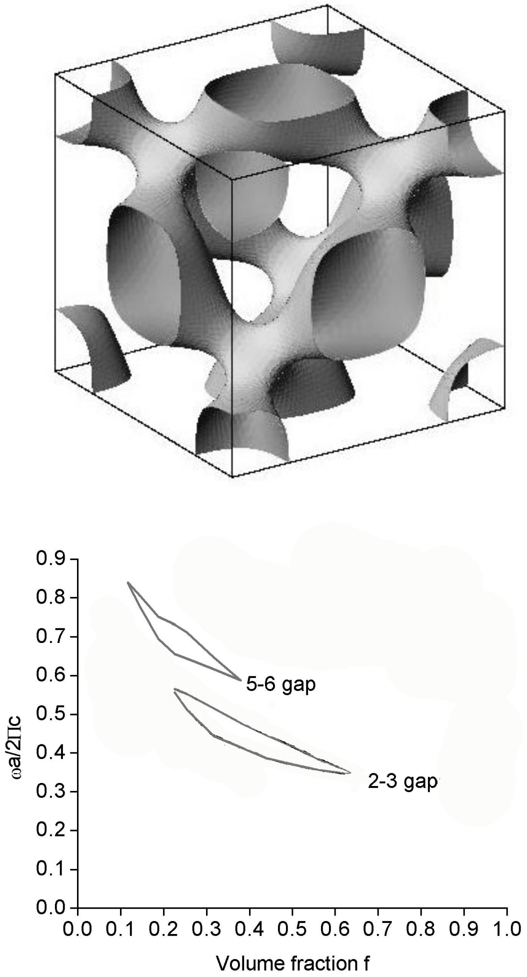

Figure 4 A photonic crystal belonging to the 216 space group created by symmetric modulations along principal directions. This 216 unbalanced diamond presents two simultaneous gaps. The original 2-3 diamond gap and an additional 5-6 gap.

As a proof of demonstration of the abiltiy to create a bicontinuous photonic crystal through interference lithography, the P structure is transferred into a modified SU-8 photoresist platform.[34] An important practical aspect that needs to be considered is the refraction of the beams and associated change in polarization due to the high refractive index *n*=1.62 of the SU-8. In order to avoid this distortion it is useful to employ prisms with an index equal to that of the photoresist with faces perpendicular to the incoming beams.[35]

In the particular case of the bicontinuous P structure since the three terms of the level set equation are mutually orthogonal, registration between respective gratings is not a problem. In order to achieve a size scalable simple cubic structure, while retaining the symmetry elements, three exposures can be used employing pairs of beams. While exposing the three gratings it is important that the beams of different gratings do not interfere with one another. In order to avoid this, the exposures can either be separated in time i.e. a multiple exposure route or, the polarizations between any two sets of gratings must be perpendicular. An important consideration in the multiple exposure technique is the requirement that the photoresist platform employed must be additive in nature. Figure 5 shows SEM micrographs of the P structure along the [100] direction with a diffraction pattern along the [111] direction. The diffraction pattern shows that the structure formed is cubic.

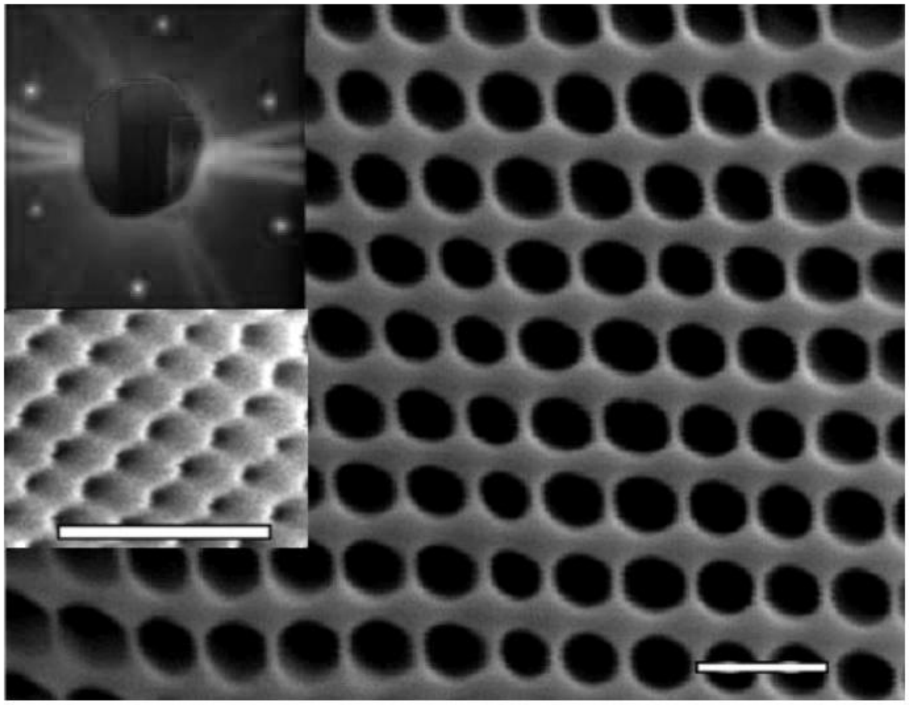

Figure 5 SEM micrograph of the (100) surface for a bicontinuous P surface structure having a lattice constant a=1.1 μm. The inset shows a SEM image of a P surface structure having a periodicity of 0.5 microns demonstrating size scalability. The inset diffraction pattern is from another P surface structure showing the (111) orientation. The scale bars shown are 2 microns.

CONCLUSIONS

A comprehensive study on photonic crystals via level set equations and 3-D interference lithography has been presented. The modulations along principal directions scheme can be used to obtain photonic crystals with large complete gaps. Once an appropriate photonic crystal, and corresponding level set equation, has been identified, the beams parameters to create the structure through interference lithography must be found. The desired photonic crystal is then created by the 3-D pattern generated by the interference of the beams and appropriate infiltration of the structure by a high dielectric material[33].

ACKNOWLEDGMENTS

The authors acknowledge the financial sources: the U.S. Air Force Defense University Research Initiative on Nanotechnology in conjunction with the University of Buffalo and the Institute for Soldier Nanotechnologies of the U.S. Army Research Office. Note: The research was partially sponsored by the U.S. Army Research Office through the Institute for Soldier Nanotechnologies at MIT, and the content of the information does not necessarily reflect the position or the policy of the Government, and no official endorsement should be inferred.

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