Photonic Crystals: Numerical Predictions of Manufacturable Dielectric Composite Architectures

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Abstract—**Photonic properties depend on both dielectric contrast in a microscopic composite and the arrangement of the microstructural components. No theory exists for direct prediction of photonic properties, and so progress relies on numerical methods combined with insight into manufacturable composite architectures. We present a discussion of effective photonic crystal production techniques and several numerical methods to predict dispersion relations of hypothetical but fabricable structures.**

Index Terms—**Photonic Crystals, Dielectrics, Microstructural Properties, Computational Materials Science**

I. Introduction

PHOTONIC properties that derive from microstructural composites of materials with dielectric contrast may be useful in nanotechnological applications. Unfortunately, there is no known simple way to predict photonic properties for a given periodic microstructure with composite material properties besides numerical calculation. A search for technologically feasible structures with optimal properties must be guided by the the fabrication techniques that must be applied to produce the dielectric composites. Thus, processing methods and computational materials science become tools for materials exploration.

Examples of photonic band gaps are now well known. Computed examples of the reflectivity of the two polarization modes as a function of inbound plane waves as a function of composite size are illustrated in the appendix.

II. Background

A. Microstructures of Dielectric Composites

Photonic crystals are periodic arrangements of dielectric

materials which can be one- (1D), two- (2D), or three-dimensional (3D). Incoming light with a wavelength λ and at an angle θ scatters off the various interfaces in the crystal. For 1D and 2D photonic crystals, two distinct orientations for the electric and magnetic fields exist, the TE and TM modes. For a given photonic crystal geometry, light with certain wavelengths and incident angles scatters off the interfaces in such a way that no light is transmitted and total reflection occurs. This range of wavelengths and incident angles is called the photonic band gap.

The property of a photonic device depends on both its macroscopic geometry and its dielectric composite architecture. We investigate methods of simultaneously controlling both composite architecture and geometry through electrochemical techniques. The unique characteristics of electrochemistry could allow novel photonic crystal devices to be produced. Specifically, electrochemical techniques are scaleable, non-directional, simple, and inexpensive. The merits of this technique, not yet explored in this context, are its use to create non-planar 1D, pseudo-3D, and particulate photonic crystal geometries. Two- and three-dimensional finite element modeling are employed to computationally explore the optical properties of these novel devices. The model is verified with experimental studies, allowing it to then be used for optimization and development of other devices with unique geometries.

Photonic crystals have been widely studied for a variety of applications, including low-loss waveguides, omnidirectional (highly reflective at all incident angles) mirrors, and band pass filters. Because photonic crystals can have 100% reflectivity at even very large incident angles, they can be used to eliminate losses due to bends in waveguides as well as to allow signals to travel very long distances with little attenuation. This property also allows photonic crystals to function as highly reflective mirrors to light at all angles of incidence, which have been utilized in devices such as very efficient laser cavities. Furthermore, if a defect cavity (a dielectric section that is a different size than the other dielectric units in the crystal) is inserted into a photonic crystal, the resulting device will be

a narrow optical filter, allowing only a single wavelength (the defect wavelength) within the band gap of the crystal to be transmitted.

Because the position of the band gap can be tuned to any wavelength, these devices can be manufactured to function at any wavelength, provided that the appropriate dielectric materials and geometries of the dielectric regions can be obtained. Although these devices could be achieved with 2D and 3D photonic crystals, many have chosen to focus on manufacturing such devices from 1D photonic crystals due to easier processing techniques. It has been demonstrated that 1D photonic crystals can have a complete omnidirectional band gap provided that a few constraints on the relative indices of refraction of the materials are satisfied.

This project investigates a means to calculate the photonic properties of manufacturable photonic crystal devices with precisely controlled dielectric architectures and explores methods of a scalable process using electrochemical techniques. There are several advantages to using electrochemistry instead of other traditional techniques. Although there are few constraints on the materials that can be used to create useful photonic crystal devices, there are still several material systems and structures that can be produced electrochemically.

In order to manufacture a photonic crystal device that has a band gap targeted to a specific wavelength, precise control over the size and uniformity of the dielectric regions is needed. For 1D photonic crystals, traditional techniques, such as spin coating or evaporation, deposits uniformly over small areas and on limited surface topologies. More uniform coatings on larger flat areas can be produced using evaporation with a special chamber that rotates and swings the substrate around the boat, but this is complicated and not easily modified for large-scale manufacturing.. However, large-scale and mass production of photonic crystals is achievable with electrochemical techniques. In fact, electrochemical techniques are already used for manufacturing of large-scale metallic coatings. Additionally, the electrochemical production of 2D photonic crystals through anodization involves a balance of forces, which naturally constrains the sizes of the dielectric regions to be very monodisperse and well-ordered over a macroscopic length scale.

There are many possible material systems that can be used to create a photonic crystal. However, there are a few requirements on the materials that can be used in most photonic crystal devices.

B. Dielectric Behavior

Any system of two or more optically different materials can be used to produce a photonic crystal device---however the properties are generally enhanced with large dielectric contrast and with judicious choice of microstructure. Metallic and semimetal systems are typically more absorbing near the visible wavelengths. Nevertheless, these materials can be used to make photonic devices even if they are absorbing. These devices may utilize the absorption, minimize the absorption, or work in a regime where the absorption is negligible. Most photonic crystal devices require materials that behave as dielectrics (i.e. negligible absorption) in the wavelength regime of their intended use.

C. High Index Contrast

In order to have an omnidirectional band gap, there are a few requirements on the relative sizes of the refractive indices of the materials that make up the photonic crystal. The specific constraints differ for different micrstructural configurations. However, in general, a high index contrast is needed between the two materials and the average index of the materials must be significantly higher than the surrounding medium. Several numerical techniques can be used to determine if the index contrast and index values for a particular photonic crystal structure are sufficient for an omnidirectional band gap. These techniques include the transfer matrix technique (for 1D finite crystals) and the frequency domain technique (for infinite crystals), which are computational methods that can be used to determine the band diagram for a given structure.

D. Electrochemically Feasible

In addition to the above considerations, if electrochemistry is to be utilized to create the devices, then the photonic crystal materials and structures must be able to be produced electrochemically. Not all material systems are electrochemically stable, and not all structures can be produced using electrochemistry. Still, there is a significant range of material systems that can be used and structures that can be produced.

III. 3D PHOTONIC BAND GAP DIELECTRIC NETWORK STRUCTURES[1]

Several new three-dimensional dielectric network structures possessing large photonic band gaps are presented. Two of these photonic crystals show gaps as large as 26% of the midgap frequency for a refractive index contrast 3.6:1. Four of the proposed structures consist of 3-connected networks and a fifth structure has mixed 3- and 4-connected nodes. The realization of these structures using block polymer self assembly, interference lithography and layer by layer lithography is discussed

Photonic crystals possessing large photonic band gaps have been based on a small number of structures: the single primitive, the single gyroid, and especially the single diamond structures. It is noteworthy that these structures are all dielectric networks with 6-, 3-, or 4-connected nets respectively. Many of the leading band gap structures (spheres on a diamond lattice, Yablonovite, rod-connected diamond, the woodpile structure, the MIT structure, ABC type hexagonal structure, and the bicontinuous diamond D level set) are in fact, all based on the 4-connected diamond structure. In this thesis, dielectric structures possessing

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[1] Material in this section was submitted to Applied Physics Letters by M.Maldovan, W.C. Carter, and E.L. Thomas.

large photonic band gaps were found by considering the systematic replacement of the 4-connected points in the diamond structure by pairs of 3-connected points. Our approach utilizes A. Wells' comprehensive work on the 3-D nets, as summarized in his monograph "Three-dimensional Nets and Polyhedra." Building upon Wells' work, five structures with complete 3-D photonic band gaps were found. Four of these consist of 3-connected networks [structures denoted by Wells as "(10,3)-a, (10,3)-b, (10,3)-d and (10,3)-f"] with the fifth structure comprised of a mixture of 3- and 4-connected points [denoted "(8,3/4)-b"]. The photonic properties (band diagrams) of the structures were calculated using the standard plane-wave method. The real space unit cells employed in calculations were modeled by connecting neighboring Wyckoff sites in the networks with cylindrical rods of radius r. A sphere of the same radius was placed at the joints in order to smooth the connections. The radius of the cylindrical rods was varied to control the volume fraction of the structure. The refractive index of the dielectric material was fixed to $n = 3.6$.
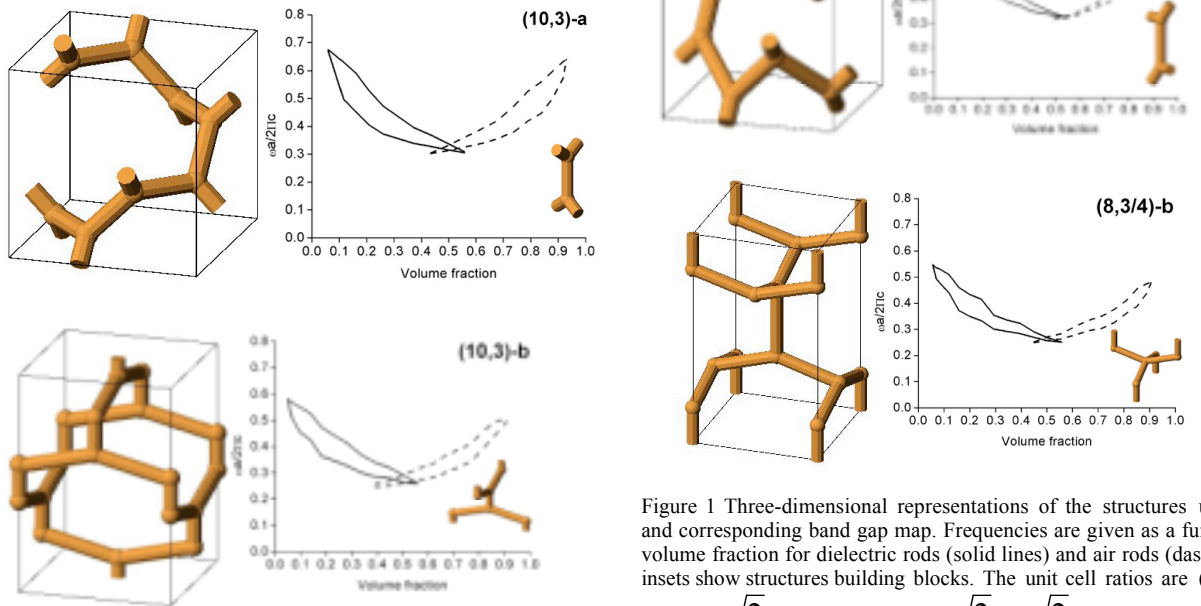

Figure 1 Three-dimensional representations of the structures unit cells and corresponding band gap map. Frequencies are given as a function of volume fraction for dielectric rods (solid lines) and air rods (dash). Small insets show structures building blocks. The unit cell ratios are (a) a:a:a, (b) a:a: $\sqrt{2}$ a, (c) a:a:a, (d) $\sqrt{2}$ a: $\sqrt{2}$ a:a, (e) a:a: $\sqrt{3}$ a. Maximum direct-inverse gaps values are (a) 26.6%–22.6%, (b) 26.2%–19.6%, (c) 16.2%–7.0%, (d) 8.0% (e) 22.0%–15.0%.

IV. PHOTONIC PROPERTIES OF BICONTINUOUS SIMPLE CUBIC P, BODY-CENTERED CUBIC G, AND FACE-CENTERED CUBIC D STRUCTURES[2]

Band structures of three-dimensionally periodic bi- and tri-continuous cubic structures have been calculated using the plane-wave method for solving Maxwell's equations. In particular, the single primitive (SP), single diamond (SD), single gyroid (SG), double primitive (DP), double gyroid (DG) and double diamond (DD) level surface families as examples of such structures found in self-organizing systems are considered. Design guidelines are provided for

creating 3D photonic crystals with a complete photonic band gap from block copolymer systems and for other self-organizing systems.

Self-assembly is emerging as a viable technology for the formation of large-scale photonic materials. The pattern forming capabilities of self organizing systems have been explored for a wide variety of applications requiring nanoscale structures. More recently photonic crystals have been formed from self-organizing colloids and block copolymer systems (BCPs). BCPs promise simple, non-lithographic routes to complex morphologies that have interesting and important photonic properties.

Block copolymers belong to a class of self-organizing materials including amphiphiles and surfactants which form a wide range of microstructures. These microstructures can be one-, two-, or three-dimensionally periodic and may be discrete or continuous. These phases are separated by a sharp interface known as the intermaterial dividing surface (IMDS). Among the most interesting microdomains structures are the single primitive (SP), single diamond (SD), single gyroid (SG), double primitive (DP), double diamond (DD), and double gyroid (DG) that form two or three independent but interconnected domains (one or two networks and a matrix) of cubic symmetry.

Because the networks and matrices are both three-dimensionally periodic and continuous, if one type of domain is extracted via etching, for example, and the other component was a rigid material, the structure would be free standing. The 3D continuity also provides access throughout the bulk of the material for reagents, etchants, and fillers, which affords their use as material templates. Controlled dielectric contrast can be achieved in such photonic materials by etching and/or back filling. These properties make these structures desirable for large three-dimensional photonic devices. In order to use these materials in photonic applications, it must be understood how to design the system for maximum performance. This is most effectively accomplished by mapping the photonic band structures of these morphologies over a range of materials parameters and structural characteristics. From these photonic properties or photonic "gap" maps, the necessary information can be extracted for constructing a materials system that will form, via self-assembly, a 3D photonic crystal with a full band gap for a desired frequency range. This will bring self-assembled photonic devices one step closer to realization.

V. A NEW LAYER-BY-LAYER STRUCTURE WITH A LARGE PHOTONIC BAND GAP[3]

A new layer-by-layer periodic dielectric structure with a large photonic band gap is presented. It consists of a layer-by-layer approximation of the triply periodic bicontinuous level set diamond D. The new structure retains the ease of fabrications of the standard woodpile while increasing the maximum quality factor of the gap by 28%. The refractive

index contrast was fixed at 3.6:1. Photonic band gap properties of this structure were calculated using the plane-wave method and its band gap optimized.

Three-dimensional periodic dielectric structures possessing a complete photonic band gap offer many applications as optical devices. For example, these types of materials are candidates for inhibiting spontaneous emission in direct-gap semiconductors, enhancing the performance of semiconductor lasers, providing new means for wave guiding, and supplying a platform for optical microcircuits. Recently, the capability of the woodpile structures as components in all-optical microchips has been theoretically demonstrated. Although the new structure presented in this proposal can be used for all the applications mentioned above, it is more suitable for optical microcircuits not only because it increases the space available for air wave-guiding, but also presents an enhanced photonic gap.

One important parameter determining the efficiency of the gap is the quality factor (QF), defined as the gap width divided by the midgap frequency. Those structures exhibiting wider gaps at low frequencies will be able to perform more efficiently (high QF) than those with narrow gaps at high frequencies (low QF). However, the benefits of gap robustness must be weighed against the ease of their fabrication.

A layer-by-layer approximation of the bicontinuous diamond D will be investigated as part of this thesis. The diamond D is a triply periodic bicontinuous structure with the symmetry of the diamond *Fd3m* space group. This structure was proposed as a candidate for a realizable photonic crystal Fabrication of this structure has been proposed through a 3-D interference lithography approach. Furthermore, its large and robust band gap makes it a suitable template for a layer-by-layer approximation.
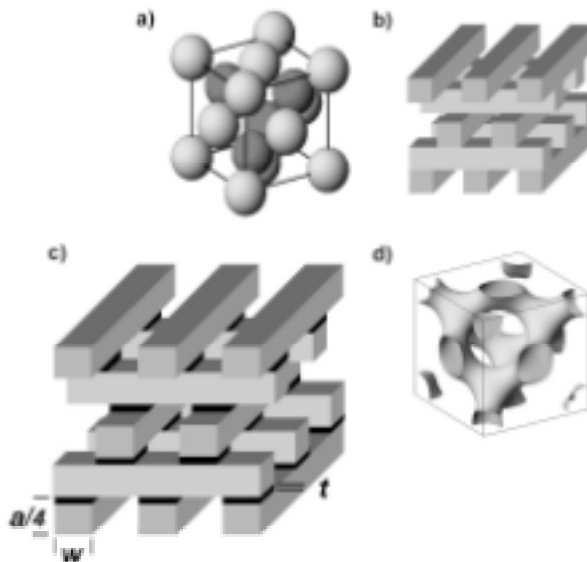
Figure 2 Schematic three-dimensional representation for (a) spheres on a diamond lattice, (b) standard woodpile, (c) diamond woodpile, and (d) bicontinuous diamond D level set.

The proposed approximation to the level set D surface structure is shown in Figure 2c. A comparison of the new structure with the standard woodpile structure is instructive. Such a comparison shows that the proposed structure is similar to the standard woodpile and can be produced by simply adding a square prism of dielectric material (with optimized thickness) in the area where the rods touch. This small modification results in an increase in the quality factor of the gap from 18% to 23% and is thus a major design innovation.

VI. MODIFIED DOUBLE GYROID STRUCTURE WITH A COMPLETE PHOTONIC BAND GAP

A. Hypothesis

Recently, a self-assembling block copolymer system was employed to form the double gyroid morphology at an appropriate length scale to interact with visible light. This process can serve as the basis to fabricate a three-dimensional photonic band gap at visible wavelengths. The relative composition of the copolymer determines the periodic morphology, whereas the molecular weight determines the length scale of the structure. The double gyroid structure consists of two intertwined single gyroid networks.

Numerical calculations of the photonic properties of the double gyroid structure show that is does not possess a complete photonic gap, however a pseudo-gap is present.

Preliminary numerical calculations on the double gyroid suggest that some modifications on the structure open the pseudo-gap. These modifications are basically deformation along a particular direction and the removing of the matrix. With these modifications, it is expected that the double gyroid structure, which more closely resembles a diamond geometry, will have a complete photonic band gap.

B. Summary and Conclusions

Photonic band gap structures are difficult to make. Processing and its relation to structure and robust band gaps are important.

Materials with high dielectric constant are needed to produce robust gaps.

The plane-wave method can be used to model photonic composites. Up to date, the diamond structure presents the best gap (~30%).

Bicontinuous structures single P, single G, and single D, which can be fabricated through an interference lithography technique, show complete photonic band gaps for a large range of volume fractions.

A level set method based on crystallography provides a systematic way of studying the symmetry dependence on gaps and exploring for photonic band gap structures.

Topology and connectivity can be explored by the replacement of the 4-connected points in the diamond by 3-connected points.

A layer-by-layer approximation of the bicontinuous diamond results in a diamond woodpile structure with a 23% increase in gap over the standard woodpile.

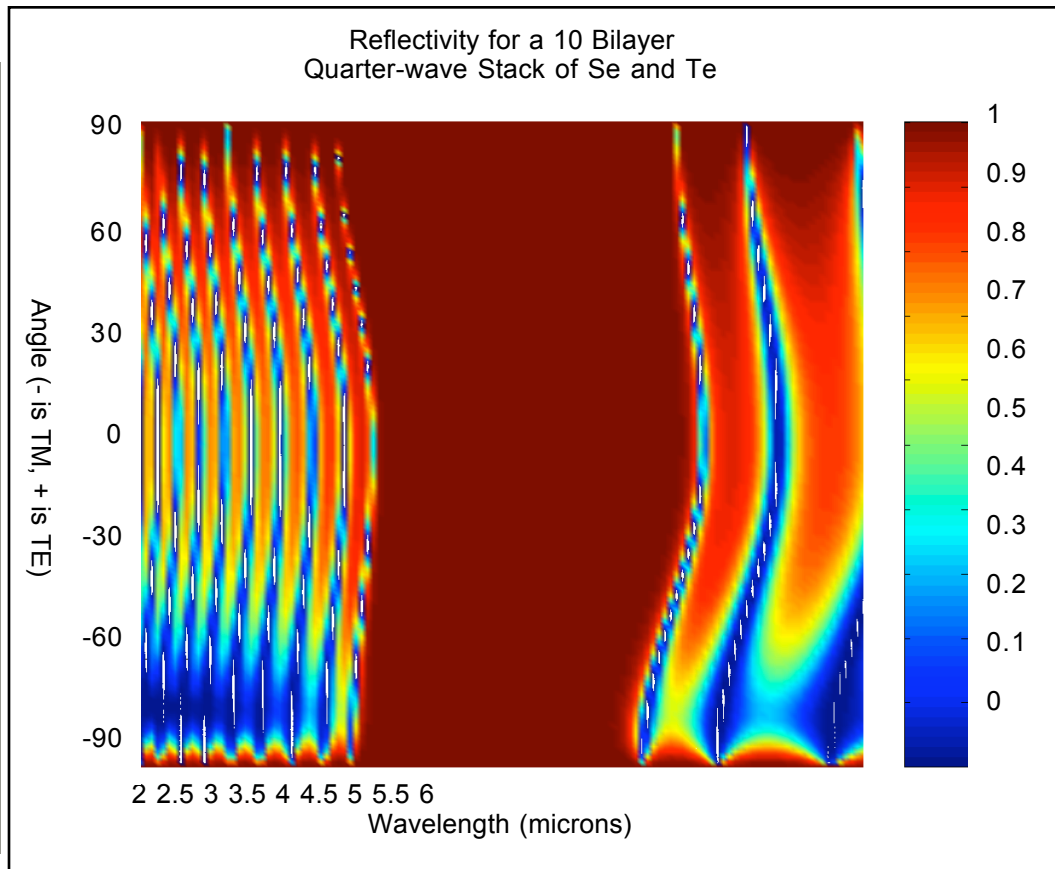Modified double gyroid structure is expected to present a complete photonic band gap.

Figure 3: A band diagram for a ten bilayer quarter-wave stack of selenium and tellurium tuned to 4μm. The omnidirectional band gap centered at 4μm is clearly apparent.
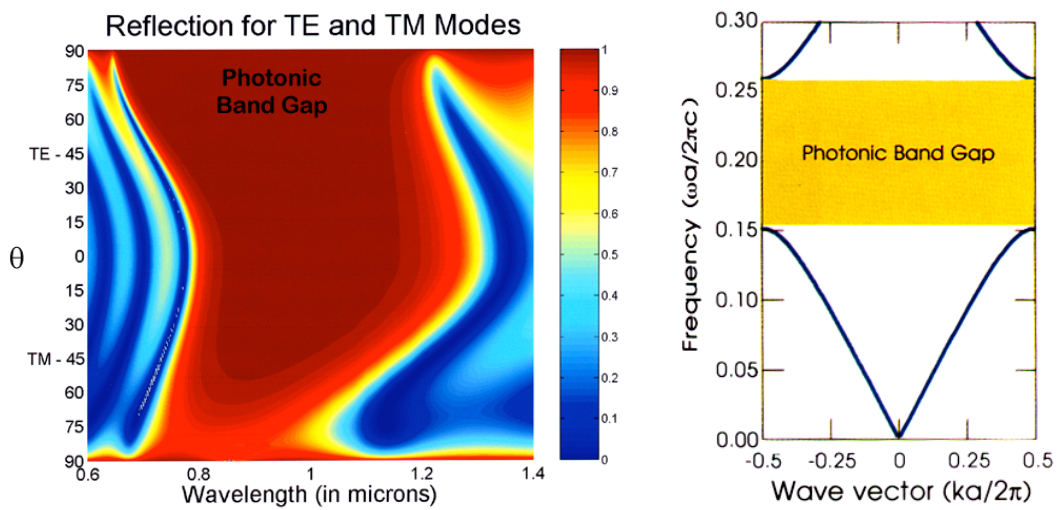

Figure 4: Left: A plot of reflectivity versus Incident angle and wavelength reveals a region of extremely high reflectivity. This region is indicative of the photonic band gap of the crystal. The above plot was produced using the transfer matrix technique. Right: An alternative plot can be obtained by converting the incident wavelength to a frequency and the incident angle to a component of the wave vector k.