

A simple a priori determination of optical transmission gaps in photonic crystals of weak symmetry

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Abstract

Pair correlation and radial distribution functions of a disordered SiO₂ colloidal photonic crystal (PC) were generated directly from binary color scale rendering of a focussed ion beam milled cross-section that was imaged by common electron microscopy techniques. The bandwidth and mid-gap frequency of the optical band gap are calculated to within 6% and 30%, respectively, of their measured values by considering a simple variation of the Ioffe–Regel conditions and symmetries derived from the experimentally determined correlation functions. This work provides a simple a priori approach to a first-order prediction of the structure/optical property relationships in PCs directly from standard microstructural imaging techniques.

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1. Introduction

Spatial periodicities in an optical material's dielectric function promote coherent interference by multiple scattering when the wavelengths of incident electromagnetic waves are comparable in size to the periodicity scale. Allowed and forbidden directions for light of certain energies to propagate may result and can provide added control over the properties of the light interacting with such a

'photonic crystal' (PC). These structured dielectrics are enabling a myriad of useful optical effects ranging from enhanced or inhibited light generation [1], omni-directional reflection [2], and ultra-low threshold lasing [3], to name just a few. While a fully defined photonic band structure is only manifested by an array of scatterers that expresses a perfect 3D point symmetry, structural analogies to amorphous semiconductors suggest that disordered systems also can exhibit photon localization phenomena. We have demonstrated previously that optical band gap effects are preserved, though with lessened magnitude, in the limit that the dielectric periodicity goes to zero [4] as has been

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theoretically predicted [5,6]. Thus, these disordered ‘photonic glass’ systems still exhibit some PC effects and can play an enabling role in future applications.

This work introduces a simple combination of microscopy and image processing to determine directly the transmission properties of PCs. Image analysis of focus ion beam (FIB) sectioned samples is used to generate a pair correlation function (PCF), and corresponding radial distribution function (RDF), that is representative of the PC’s symmetry. From a simple application of the Ioffe–Regel (I–R) conditions, we can understand the transmission spectra of the solid in a straightforward manner. This novel approach is conceptually simple, yet powerful, and permits a priori the quantification of a PC’s optical properties directly using common microscopic imaging techniques. Although this paper only discusses random assemblages of compositionally and geometrically equivalent scatterers, the approach is not limited in microstructural form and can be extended to periodic or functionally graded structures as well as ones with conceivably any distribution in particle sizes, morphologies, compositions, etc.

2. Experimental procedures

The reader is referred to our previous work for specifics regarding the synthesis of the SiO_2 particles and that of the PCs as well as the electron microscopy [4]. The disordered PCs were sectioned using FIB milling (Materials Analytical Services,

Inc., Raleigh, NC) after being imbedded in epoxy and polished to a $1\ \mu\text{m}$ finish using standard metallographic polishing techniques. FIB milling was performed using 30 keV Ga^+ ions in an FEI 200 TEM FIB (FEI Inc., Hillsboro, OR). Image analysis of the electron micrographs was performed using Photoshop 5.0 (Adobe Systems Inc., San Jose, CA) equipped with additional image processing and analysis add-ins (Reindeer Games Inc., Asheville, NC). Five, $5\ \mu\text{m} \times 5\ \mu\text{m}$ sections were selected for analysis from the original micrographs. The particles were highlighted using a thresholding filter that converted the images to binary color images. Noise in the images was reduced via hole filling, erosion and dilation filters. Feature analysis was used to generate a table of the particle center coordinates. The five particles closest to the center of each image were chosen and the radial distances between those particles and the centers of all other particles in the image were determined. The bulk packing density of the particle compact was estimated from the cross-sectional area of the particles divided by the area of the images. The RDF curve then was calculated using the methods outlined by Allen and Thomas [7]. Calculations were performed for a shell size of 60 nm.

3. Results and discussion

Fig. 1(left) shows an electron micrograph of a fracture cross-section from a disordered PC. The particles are randomly ordered and monosized

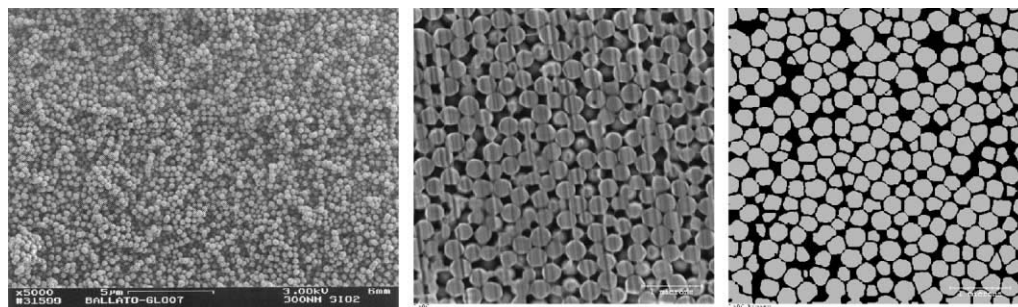


Fig. 1. FE-SEM micrograph of a colloidal solid containing a random arrangement of monosized SiO_2 spherical particles (left). FE-SEM micrograph of FIB-milled disordered colloidal crystal (center). Binary color scale image of FIB-milled disordered colloidal crystal (right).

with diameters of approximately 300 nm. As has been discussed previously, the short-range particulate order in these types of structures may partially satisfy the conditions for a photonic band gap although the optical cutoffs (i.e., the optical Brillouin zone boundaries) will not be well defined [4]. This is validated by the observation of an angle dependent weak opalescence from this sample and a broad band gap with a 3 dB bandwidth of ~600 nm that was centered about ~550 nm (not shown, see Ref [3]). Electronic image analysis directly from the Fig. 1(left) micrograph is not useful because it does not provide a well-defined flat 2D surface. Thus, the ‘surface’ will have particles above and below a given plane, leading to a miscounting of the number of particles over a given distance from a central particle. This error in particle number will manifest itself as an error in the RDF and, thereafter, in the calculated optical transmission spectrum. A flat surface is produced with particulate placements well maintained using FIB milling, as compared to microtoming or traditional polishing, as is shown in Fig. 1(center). Particles below the surface plane are visible in some instances. This should not lead to an overly large error in particle counting provided an adequate contrast is selected when this micrograph is converted to a binary color scale for image analysis, as is shown in Fig. 1(right).

Fig. 2 shows the PCF, $g(R)$, as a function of the distance, R , from a central particle normalized

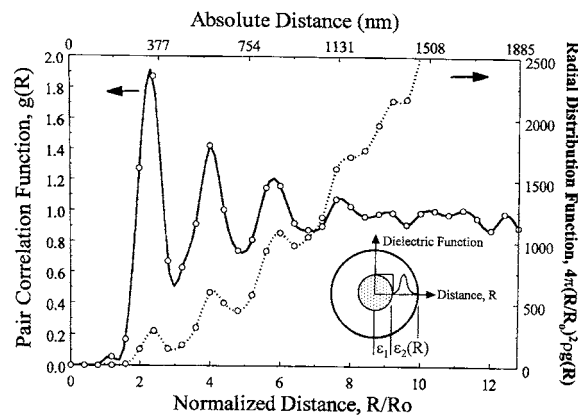


Fig. 2. PCF, $g(r)$, as a function of normalized distance from a central particle, R/R_0 , generated directly from the binary colored microscopy image in Fig. 1(right).

to the particle’s diameter, R_0 (a constant in this monosized system) computed directly from the Fig. 1(right) data. The first peak, corresponding to the first-nearest-neighbor normalized distance, occurs at about $2R/R_0$. There is a small but finite correlation at distances $< 2R/R_0$ which arises from particles milled off center. Interestingly, for these monosized systems, this information can be used to extract the 3D microstructure from the 2D cross-section [8]. As expected, the placement of higher order nearest neighbors is observed at nearly integral multiples of $2R/R_0$. Past approximately $8R/R_0$ (i.e., fourth-nearest neighbor) the PCF limits to unity which corresponds to the distance where the effective density of the compact approaches its bulk value as is characteristic of the atomic RDF from noncrystalline solids [9].

Numerous studies have considered approaches to quantifying the level of order or disorder in PCs. Most rely on a generic order parameter that is quantified using a photon localization length being itself defined in terms of a logarithmic average of the system’s optical transmission over the various configurations [6,10]. We suggest here that the PCF (or the related RDF), being a physical observable and now a direct measurand, is a well-suited metric on which to define certain aspects of order and disorder. The RDF provides information regarding the underlying symmetries, though it stops short of generating the full space group needed to compute the complete photonic band structure. Intuitively, it is clear that some information about the optical response (i.e., the transmission) of an array is contained within the RDF given that we know the dielectric properties of the scatterers.

In John’s reinterpretation of the standard I–R condition for photon localization in weakly disordered systems, the photon is assumed to be nearly free. Degenerate perturbation theory is used to describe the Bragg scattering at the zone edges and the effects of geometric resonances are treated exactly before any disorder averaging [11]. In the case of the PCs the RDF provides only an indication of geometric order radially away from a single scatterer and thus averages the effects of disorder directly. In fact, the degree of disorder can be quite high in the array with only minimal

variation in the appearance of the RDF. Therefore a direct application of John's approach does not seem possible with only the information provided in the RDF. In order to gain some insight however, we can examine the localization criterion in the case where only spherically continuous symmetries of the RDF are known. To do this we apply a simple 1D model intended to capture only the essential features of the scattering. Further, to focus on the effects of long-range geometric scattering, we treat only the case where the Mie scattering resonances are far from the photonic band gap.

If we consider an incident wave scattering from an individual sphere within the PC, that wave will see a spherically symmetric dielectric function, on average, into which it must scatter. This dielectric susceptibility will vary as a function of radial distance from the scatterer as shown in the Fig. 2 inset. We define ε_1 as the dielectric constant of the central scatterer ($= 1.18$).¹ The dielectric susceptibility then repeats as a function of radius (radial coordinate, R) with a shape over one repeat unit that can be taken directly from the PCF: $\varepsilon_1 \sim g(R)$. The index difference between the scattering shells at $2R/R_0$ is then 0 and at $1R/R_0$ is $\varepsilon_0 - \varepsilon_1$, where ε_0 is taken to be the dielectric constant of air and ε_1 is the maximum value of the dielectric constant in the silica medium. From a simple Fourier transform of the PCF the distance between scattering shells is roughly 377 nm (not shown). Now in this pseudo-1D context, we apply the conditions suggested by John. The wave vector at which the gap in transmission is to occur is $\mathbf{K}_0 = \mathbf{G} / (1 + \varepsilon_0/\varepsilon_1)^{1/2}$. Taking $\pi/377$ (nm^{-1}) to be \mathbf{G} , this gives a mid-gap wavelength of ~ 565 nm. The breadth of the transmission gap is then approximately $\Delta\omega = \mathbf{G} \times c \{ (2\varepsilon_1)^{1/2} / (\varepsilon_0 + \varepsilon_1) - (5/12)^{1/2} \times (\varepsilon_0 + \varepsilon_1/5)^{1/2} \}$ giving a gap bandwidth of ~ 400 nm. Comparing these estimates to the measured values of the gap [4], yields an error in the mid-gap wavelength and bandwidth of 6% and 30%, respectively—a reasonable agreement for so simple a

model. Aside from the first order approximation to the I–R conditions, the major errors in the calculated band gap property values are believed to arise from the estimation of the dielectric constant of the partially porous SiO_2 as well as the shell size used to generate the PCF, which affects its spectral profile and the resultant Fourier transform. Preliminary results indicate an equally satisfactory extension of this approach to ordered systems where the RDF and resulting transmission characteristics are generated along specific crystallographic directions. This leads to the possibility for the direct experimental computation of the angle dependent photonic band gap properties.

4. Conclusions

This work has provided a simple analysis of disordered PCs by sectioning monosized SiO_2 assemblies by FIB milling and using image analysis of electron micrographs. The image analysis was used to generate directly PCFs that, when applied to simplified I–R conditions, provide an a priori quantification of the PC's mid-gap wavelength and bandwidth without necessarily also having to perform angle- and energy-resolved optical spectroscopy. Further, it suggests that the PCF is a good metric of the disorder limit necessary to achieve photon localization in these crystals.

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¹ Defined as: $\varepsilon_1 = (n_1)^{1/2} \cong (1.4)^{1/2} = 1.18$ where $n_1 \cong 1.4$ derives from an estimate of the porosity of the solution-derived SiO_2 particles.

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