Small-angle x-ray scattering from photonic colloidal crystals

L. Lottermoser, C. Song, A.I. Goldman, G. Subramania, M.M. Sigalas, R. Biswas and K.-M. Ho
Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames IA 50011

L. Lurio
Physics Department, Massachusetts Institute of Technology, Cambridge MA 02139

Introduction

Over the last decade photonic crystals have become the subject of intense interest spurred by potential applications including optical switches, micro-fabricated lasers and light-emitting diodes. The essential requirement for a band-gap at optical frequencies is the fabrication of a three-dimensional periodic dielectric structure (face-centered cubic) with a lattice parameter on the order of optical wavelengths. A very attractive method for forming these structures involves polystyrene microspheres which, in a titania suspension, self-assemble into periodic structures[1,2]. After processing, these microspheres are "burned" out leaving a periodic structure of "spherical" holes in a titania matrix.

The characterization of these crystals is commonly done by scanning electron microscopy, revealing information about the surface structure. However, it is very desirable to probe the bulk structure as well. In particular, calculations of the optical properties of these crystals (the energy of the gap) are in disagreement with experiments. This has been attributed to a compression of the lattice along the direction perpendicular to the plane of the film that results from processing. Small-angle x-ray scattering is an ideal non-destructive probe for studying both the in-plane and out-of-plane structures of these films.

Methods and Materials

The photonic crystal was formed on a glass substrate at the Ames Laboratory using a non-toxic mixture of polystyrene microspheres (radius of 200 +/- 20 nm) and titania[3]. Holes, 1mm in size, were etched through the glass substrate for the x-ray measurements. The SAXS measurements were performed on the IMM-CAT small-angle scattering beam line at the Advanced Photon Source using an incident wavelength of 1.51Å and a beam size of 50μm X 50μm. Scattering images were taken using a CCD-camera located 5.6 m from the sample. Data sets were taken both in the transmission geometry to obtain in-plane structural data, and in a grazing-incidence reflection geometry to obtain out-of-plane structural information.

Results

In-plane structure - Data taken in the transmission geometry reveals information about the in-plane structure of the films. CCD images showed regions of highly ordered single grains, several hundred μm in size, with their surface normal parallel to the <111> direction of the cubic lattice. However, other regions were observed to contain twinned crystals or crystals with random orientation. The CCD images were azimuthally averaged to obtain a one-dimensional, in-plane, radial scattering pattern. All of the diffraction lines in this pattern could be indexed to the FCC structure with a lattice constant of 568 nm, which is expected for spheres of this size (e.g. a nearest neighbor distance of 400 nm). No evidence of stacking faults in the structure was found for these "good" single grain regions.

Out-of-plane structure - Perhaps the most interesting result of this study was revealed in the scattering from the film in the grazing-incidence reflection geometry. Here the diffraction pattern reveals information about structure along the surface normal direction that can not be easily obtained by SEM measurements. Three relatively broad diffraction peaks were observed at positions that indicate a significant (30%) compression of the lattice along the <111> direction. This also implies that the voids themselves are not spherical, but compressed along the direction parallel to the film surface normal.

Discussion

It has long been suspected that the photonic crystals fabricated by this method are compressed along the direction parallel to the surface normal. This most likely results from a cold-isostatic pressing of the sample during processing and anisotropic shrinkage of the cavities during heat treatment of the film. The compression of about 30% determined in this measurement, also helps to reconcile the discrepancy between the calculated and observed optical band-gap energy. Further work is planned to measure the form factor of these cavities to further refine the calculated optical properties.

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