

Interferometric technique for the measurement of photonic band structure in colloidal crystals

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Under suitable conditions polystyrene microsphere colloids form photonic crystals capable of diffracting visible light, analogous to x-ray diffraction from atomic crystal planes. The lattice spacings of these crystals can be tailored to satisfy the Bragg condition along a certain direction for a particular desired wavelength. A modified Mach–Zehnder interferometer has been developed for accurately measuring relative phase shifts of light propagating in photonic crystals to determine the dispersion resulting from photonic band structure near the band edges.

Photonic band-gap materials forbid propagation of photons within a certain frequency range irrespective of their direction of propagation, permitting the possibility of studying new physics within the gap. For example, spontaneous emission and radiative electron–hole recombination will be strongly inhibited if the frequency of the photon to be emitted lies in the forbidden gap. The concept of a three-dimensional photonic band structure was introduced by Yablonovitch as an extension of a familiar one-dimensional optical device: a Fabry–Perot resonator constructed from dielectric mirrors.¹ Of great interest to those interested in photon localization, John² suggested that the addition of some disorder to this otherwise ordered structure would require a reinterpretation of the Ioffe–Regel criterion for Anderson localization of photons and that localized states would exist in the vicinity of the gap. Structures exhibiting full photonic band gaps in the microwave,³ millimeter,⁴ and submillimeter⁵ regimes have already been fabricated, but scaling these structures down to the optical regime has remained a challenge.

The photonic crystal considered here is a three-dimensional dielectric structure with a periodic modulation of its dielectric constant with spatial periods of the order of the wavelength of light. Owing to this periodicity, light incident along a particular direction will be Bragg diffracted by the crystal planes for a certain range of frequencies, forming stop bands. When the stop bands are wide enough and overlap for both polarization states along all crystal directions, the material possesses a complete photonic band gap. To build such a structure there are several important parameters that need to be controlled: lattice type, filling fraction, and dielectric contrast between the low and high dielectric regions. One way of fabricating a photonic crystal suitable for Bragg diffraction of visible light is by growing colloidal crystals formed by submicrometer polystyrene microspheres suspended in water.⁶ Although such a structure is not expected to exhibit a *complete* photonic band gap, it does permit study of photonic band structure effects, as reported here. Under suitable conditions such crystals may yield a pseudogap, with a consequent depression of the

density of propagating photonic states, of interest in photon localization studies.

A photonic crystal was grown for this work from a colloidal suspension of polystyrene microspheres having a diameter of $0.110\ \mu\text{m}$ and monodisperse to within 4.3%.⁷ For preparation of the crystal, the stock solution was diluted to the desired volume fraction and prepared by tumbling with ion exchange resin before being transferred to an adjustable thin cell.⁸ The microspheres have a permanent net negative surface charge counterbalanced by free ions in the solution. Once the free ions are removed with ion-exchange resin, the microspheres interact with a short-range repulsive Coulomb force in addition to a long-range attractive van der Waals force.⁹ As a result of this interaction the colloid undergoes a phase transition from a disordered state to a face-centered-cubic lattice (for this volume fraction and microsphere diameter) with the densest planes (111) parallel to the walls of the containers. Ion-exchange resin enclosed in nylon mesh was also placed in the cell to inhibit the destruction of the crystal by additional free ions introduced over time. After crystals formed in the cell, their quality was confirmed by examination of the Kossel line patterns,^{10,11} which were also used to orient the single crystals.

The transmission spectrum of the crystal along the [111] direction shown in Fig. 1 displays a stop band (Bragg notch) more than 2 orders of magnitude deep. The transmission minimum is centered near 599 nm with a bandwidth (photonic stop band) of 7 nm at 1% transmission. Assuming 1.34 for the average index of refraction of the crystal, we calculate the distance between (111) planes from Bragg's law to be $0.224\ \mu\text{m}$. This corresponds to a lattice spacing of $0.387\ \mu\text{m}$ and a volume fraction of 4.8%. The decrease in transmission near 400 nm arises from increased single-particle scattering cross section at lower wavelengths (mainly because of residual disorder in the sample such as microsphere diameter variation and dislocations), increasing absorption, and possibly the second-order Bragg minimum at about 300 nm.

The dispersion relation of the crystal along the [111] direction was obtained with a modified Mach–Zehnder interferometer as shown in Fig. 2. This version of the

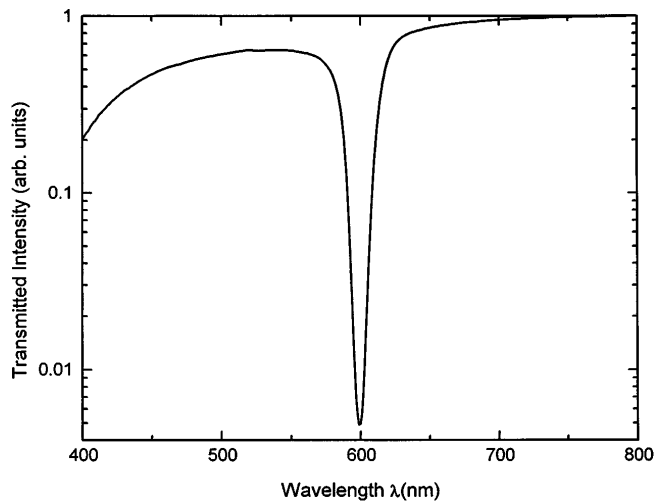


Fig. 1. Transmission spectrum along the [111] direction. The photonic crystal is a colloidal crystal of polystyrene microspheres, with 0.110- μm diameter and 4.8% volume fraction.

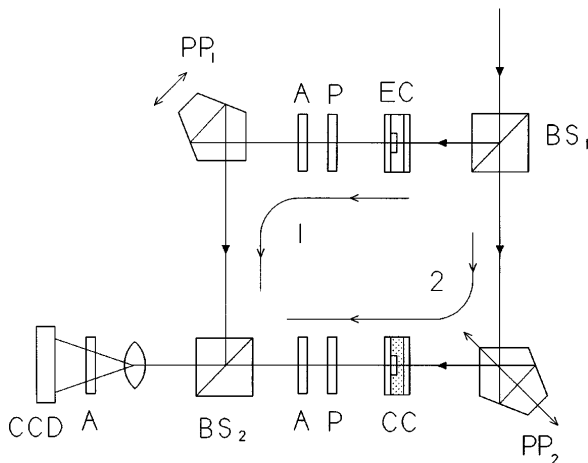


Fig. 2. Modified Mach-Zehnder interferometer. The fringe pattern is positioned in front of the CCD camera by adjustment of pentaprism PP_1 . The optical paths are equalized by adjustment of pentaprism PP_2 along a line perpendicular to PP_1 . The relative intensity is optimized for maximum fringe visibility around the band edge by rotation of the polarizer in the first arm. EC, empty sample cell; CC, crystal sample cell; BS's, beam-splitter cubes; A's, analyzers; P's, polarizers.

Mach-Zehnder interferometer¹² was chosen because of its orthogonality of adjustments. The optical path lengths and dispersions were equalized in each arm as accurately as possible with the sample absent. The intensity in the first arm was adjusted relative to the second arm to increase the overall fringe visibility as the wavelength was tuned to the band edges. The fringe patterns were captured with a CCD camera for each step in wavelength, as shown in Fig. 3. A nonlinear least-squares fit of the following equation to each fringe pattern was used to extract the desired phase information:

$$I(x) = I_0 \exp\left[-\left(\frac{x - \mu}{w}\right)^2\right] [1 + \mathcal{V} \cos(2\pi fx + \Delta\phi)]. \quad (1)$$

Here I_0 is the intensity and μ and w are the center position and the width, respectively, of the Gaussian envelope. The visibility, spatial frequency, and phase of the fringe pattern are \mathcal{V} , f , and $\Delta\phi$, respectively.

The absolute phase difference between the two interferometer arms can be considered in two parts, $\Delta\Phi(\lambda) = \phi_0(\lambda_0) + \Delta\phi(\lambda)$, where ϕ_0 is the initial phase difference arising from the insertion of the crystal at the initial wavelength λ_0 and $\Delta\phi$ is the incremental phase difference [from Eq. (1)] between the phase of the fringe pattern at a particular wavelength λ and λ_0 . The relative phase shift across the tuning curve of the Rhodamine 6G dye laser (570–640 nm) is shown in Fig. 4, where the frequency of the light (independent variable) is plotted on the vertical axis so that the figure resembles a typical dispersion relation.

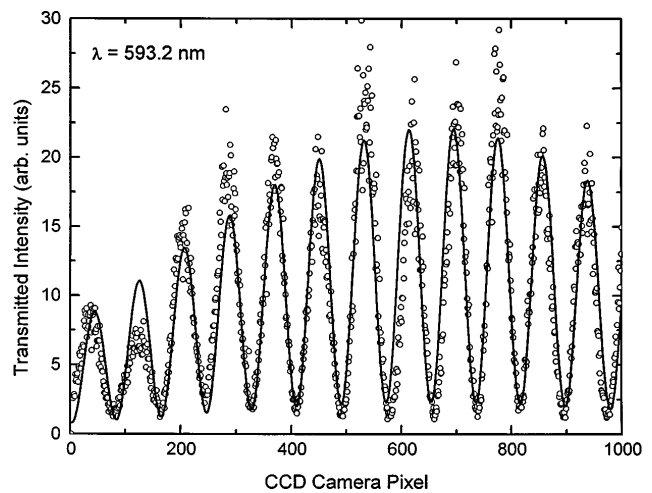


Fig. 3. Typical interferometer fringe pattern with optimized fringe visibility. The solid curve is the resulting nonlinear least-squares fit of Eq. (1).

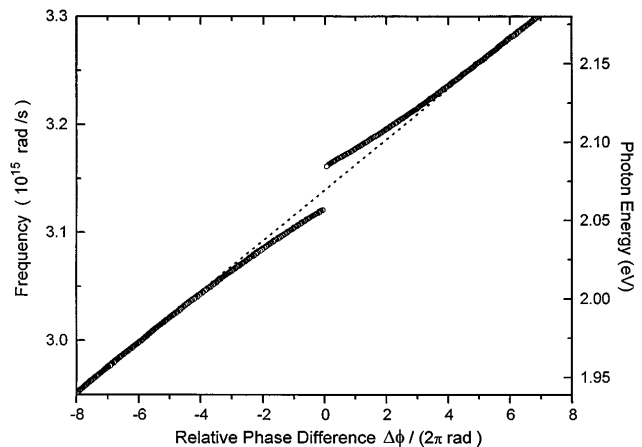


Fig. 4. Photon dispersion obtained from the wavelength dependence of relative phase of the fringe patterns. The broken region corresponds to the [111] stop band; the low transmitted intensity prohibits formation of a measurable fringe pattern. The dotted line represents the free photon dispersion in the crystal away from the gap along with the residual imbalance in the optical elements of the interferometer.

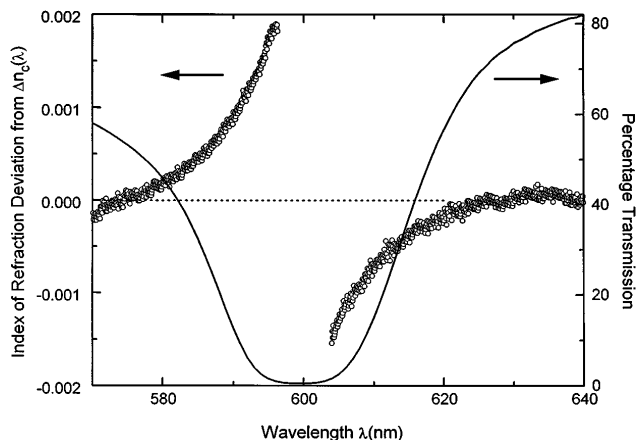


Fig. 5. Deviation of the index of refraction near the stop band. The free photon dispersion has been linearly approximated and subtracted from the residual index of refraction obtained from Fig. 4. Note that the scanned region lies well inside the stop band, where the solid curve is the percentage transmission from Fig. 1.

The absolute phase difference between paths 1 and 2 is also given by

$$\Delta\Phi(\lambda) = 2\pi[n_c(\lambda) - 1]L/\lambda, \quad (2)$$

where n_c is the index of refraction and L is the thickness of the crystal, determined to be $240 \mu\text{m}$ for this sample. Thus the incremental change in the index of refraction of the crystal, $\Delta n_c(\lambda)$, can be related to the incremental phase difference by

$$\Delta n_c(\lambda) = n_c(\lambda) - n_c(\lambda_0) = \frac{\Delta\phi(\lambda)}{2\pi} \frac{\lambda}{L}. \quad (3)$$

One can extract the deviation from the free photon case around the band edges by obtaining a residual from $\Delta n_c(\lambda)$ by linear approximation and subtraction. The resulting deviation $\Delta n \sim \pm 0.002$ at the band edges is clearly evident in Fig. 5 and is expected to grow as the center of the stop band is approached. Note that the scanned region lies well inside the stop band, shown as percentage transmission in Fig. 5 (taken from Fig. 1). The dramatic decrease of transmitted intensity makes measurement of fringe patterns difficult as the stop band is entered; hence a gap appears in the relative phase and refractive index reported.

The ability to tune the stop band to the wavelength of interest is an essential feature of using colloidal crystals for photonic band structure measurements, since the lattice parameter is controlled by the volume fraction rather than by the ball size. The width of the stop band is dictated by the ball size and the relative dielectric strength of the microsphere. Because of the limited tuning curve of the dye laser used, a relatively small microsphere diameter was selected to generate a narrow stop band. Much wider gaps can be generated with balls closer to a Mie resonance.

In summary, the feasibility of measuring photonic band structure in colloidal crystals in the optical regime has been demonstrated by use of a modified Mach-Zehnder interferometer. To our knowledge, this is the first reported measurement of optical photonic band structure in a three-dimensional photonic crystal. The self-organized crystals formed from polystyrene microspheres provide a novel system for examining photonic band structure. By expansion of the effective range of the interferometer, complete mapping of photonic band structure and its associated anomalous refractive index will be possible.

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