

# Polarization anisotropy in light propagation inside opal-based photonic crystals

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**Abstract:** Photonic crystals is expected to be the backbone of future optical integrated circuits. To realize this goal, light propagation and interaction with matters must be understood and controlled. In this work, we investigate the propagation of light inside opal-based photonic crystals along certain paths at the edge of its Brillouin Zone. Opal films made of polystyrene particles were prepared using self-assembly approach, the capillary deposition method. The structures and the optical properties of the resulting opals were characterized using scanning electron microscopy and polarization-resolved spectroscopy, respectively. The opal films have a face-centered cubic structure consisting of two domains showing preferential orientations. Domains in the form of ABC and ACB-type fcc crystals are oriented along the growth direction of the opal films. Light with frequencies near optical band gap shows a strong anisotropy. Light propagation inside opals depends on the polarization of the incident light. The intensity and the width of the extinction peaks for p-polarized incident light differ significantly from those of s-polarized light. The anisotropy disappears at frequencies above the optical band gap. The anisotropic light propagation is related to the strong anisotropy in equifrequency surface of band structure around the band gap. The shift of the extinction peaks and the variation of intensity of the extinction peaks will be discussed using the combination of kinematic and simplified dynamical diffraction theory.

**Keywords:** opals; photonic crystals; polarization-resolved spectroscopy; anisotropy; band structure

## 1. Introduction

Photonic crystals are structures with a periodic modulation in their dielectric constants. The modulation can be in one-, two-, or three-dimension. Photonic crystals allows light concentration in an ultra-small volume and almost lossless light propagation even through a sharp-bend path due to the existence of optical band gap. The optical band gap exists for a certain range of frequency because of destructive interference of light reflected at each boundary of materials having different dielectric

constants. The band gap occurs when the periodicity of dielectric constant modulation is comparable to the wavelength of light (Joannopoulos, *et al.*, 2008). Controlling light propagation and its interactions with matters at nanoscale would open the possibility for the fabrication of many high-performance optical devices. Photonic crystals are believed to serve as the backbone of future integrated optical devices.

One- and two-dimensional photonic crystals can be fabricated effectively using well-established lithographic techniques. These techniques are very precise and accurate, but unfortunately do not work well for the fabrication of 3D photonic crystals. They are expensive, slow, and suffers from alignment problem between consecutive layers. Therefore, an alternative approach based on natural tendency of colloidal particles to arrange themselves becomes the most-widely used fabrication techniques (Marlow, *et al.*, 2009). Three dimensional structures are needed for the realization of omnidirectional (full) optical band gap. The resulting face-centered cubic crystals made of colloidal particles are known as artificial opals or simply opals. Opals are starting materials for further modification such as inversion with high-refractive-index material and chemical or biological functionalization to be used for a specific application.

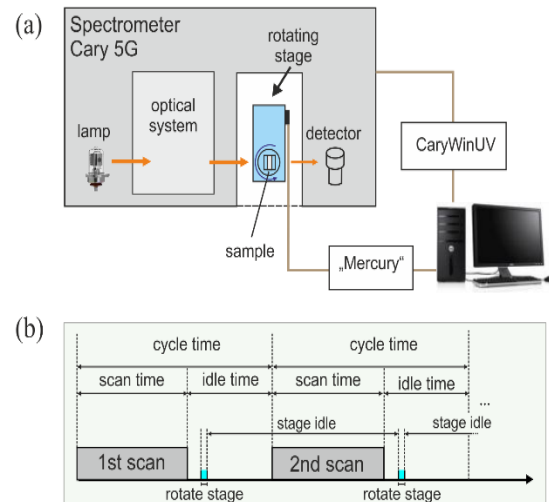
There are many available opal deposition methods (Marlow, *et al.*, 2009). Each deposition method has different degree of controllability that results in opals with different degree of crystallinity. Among other methods, the capillary deposition method (CDM) is known to results in high-quality opals with controlled film thickness (Muldarisnur and Marlow, 2011). In the CDM, the flow of suspension inside a confining cell leads to the formation of highly-ordered particles in the form of large and oriented domains.

In this work, we investigate propagation of light inside CDM-made opals using *Polarization Angular-Resolved Spectroscopy* (Muldarisnur and Marlow, 2016). The measurements were focused on light with frequency comparable to the optical band gap (stop band) of the opal. The shift and intensity variation of extinction peaks with incident angle for two different polarizations will be compared and discussed using diffraction theory and calculated band structures.

## 2. Experiments

Thin opal films were fabricated using the CDM method. In this method, opals are grown inside a planar cell made of sandwiched two glass slides separated by two thin polymer spacers of known thickness. The spacers determine the thickness of the resulting opals. Besides, micrometer-size space between the glass slides due to the presence of spacers causes capillary force when the cell is filled with colloidal suspension. The force leads to the spreading of the suspension inside the cell during deposition process. The evaporation of the solvent increases the concentration of colloidal particles at the open edges of the cell. Disorder-to-order transition to form ordered arrangement of colloidal particles occurs when a critical filling fraction of particles is reached. Deposition process ends when opals fill the entire capillary cell and the solvent evaporates completely.

An aquabidest-diluted suspension of 200 nm polystyrene particles was located inside an ultrasonic bath for 10 minutes before deposition to ensure its homogeneity. In the CDM, deposition is started when capillary cell is connected to suspension container and finished within three days, depending on the intended thickness. Opal films were deposited in a well-regulated room. Relative humidity and temperature were kept at  $35 \pm 1.5\%$  and  $23 \pm 1$  °C, respectively.



**Figure 1.** (a) Measurement set-up, (b) rotation and measurement synchronization (Muldarisnur, *et al.*, 2011).

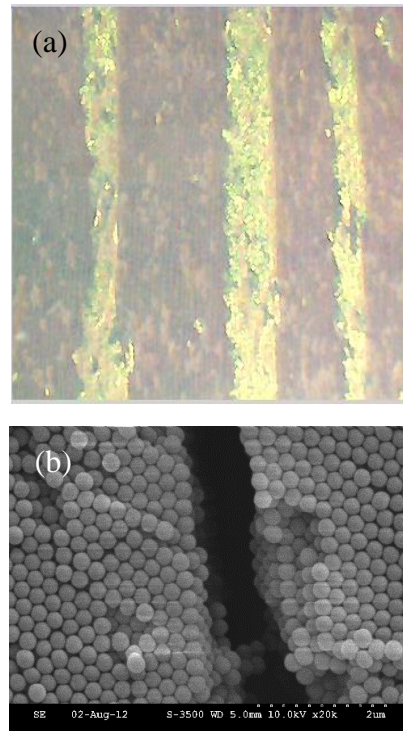
A Cary 5G UV/Vis-NIR spectrometer was used to perform optical characterization. The measurements were carried out at a fixed beam position while sample is rotated. The variation of incident angle was managed by using a C-862 Mercury rotating stage allows precise incident angle definition down to a few milidegree. The polarization angular-resolved spectroscopy was done by synchronizing of sample rotation and transmission measurements. Sample was rotated within the idle-time of the transmission measurement (see Figure 1). A polarizer was placed in the front of opal sample to obtain polarized incident light.

CDM-made opals have an fcc structure with two orientation preferences. The (111) plane is always oriented parallel to the glass substrate while the  $[1\bar{1}0]$  of the crystals is oriented along the opal growth direction. Knowing crystal orientation allows easy determination of probed crystal planes when sample is rotated during measurement. Opal samples were rotated to probe crystalline planes along two scanning paths lie at the edge of irreducible Brillouin zone of fcc lattice. The first sample orientation (Case V, vertical) corresponds to scanning along the KWLWK path, while the second one (Case H, Horizontal) correspond to probing along XULKL path. Details can be found in our previous work (Muldarisnur, *et al.*, 2011).

Band structure of opal-based photonic crystals was calculated by using a free software the *MIT Photonic Bands* or MPB (Johnson and Joannopoulos, 2001). MPB uses plane wave expansion method to solve Maxwell's equations for periodic structure. For calculation, the refractive index of polystyrene was assumed to be constant that is 1,59. Weak frequency dependence of polystyrene refractive index was ignored.

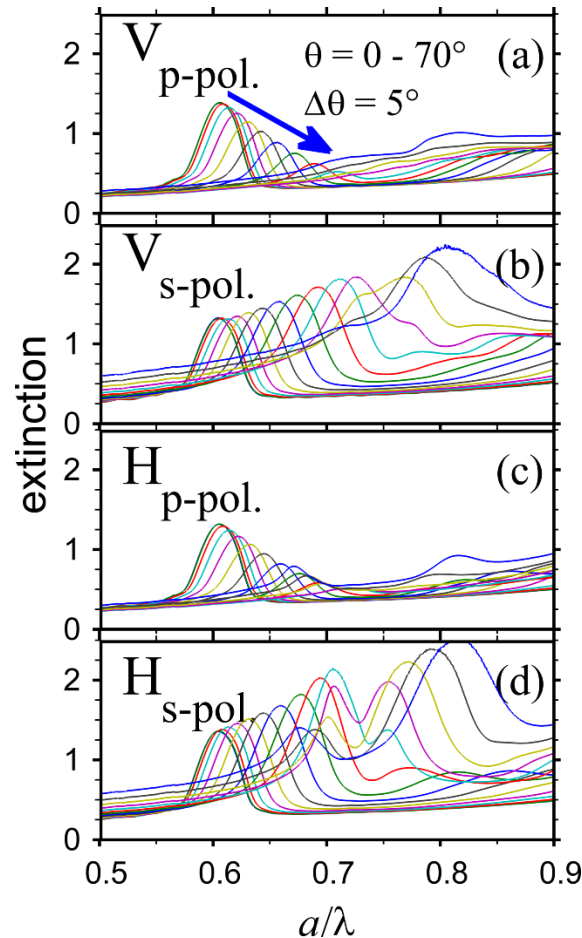
### 3. Result and Discussion

Microscope images of opal films are shown in Figure 2. Stereo optical Microscopes shows crystalline domains. Striking different color indicates two types of domains corresponding to differently-packed crystals. Different colors are observed when opal sample is illuminated from one side only. The sample was illuminated using flexible fiber-optic arm of the stereomicroscope at incident angle of  $45^\circ$ . When the sample was illuminated from the other side at the same angle, the colors of the stripes are interchanged. The size of the stripes varies from  $100 - 300 \mu\text{m}$ .



**Figure 2.** Microscope images of opal film, (a) stereomicroscope image, and (b) electron microscope image.

Electron microscope image shows nice particle ordering (Figure 2b). Hexagonal pattern of particle arrangement is the (111) plane of fcc crystal. SEM characterization also reveals that stripes are oriented along  $[1\bar{1}0]$  direction. Long-range particle ordering observed in SEM image is manifested in sharp extinction peak. The extinction indicates that light at the frequencies lie within the band gap cannot propagate inside opals, instead the light will be reflected at the surface of opal.



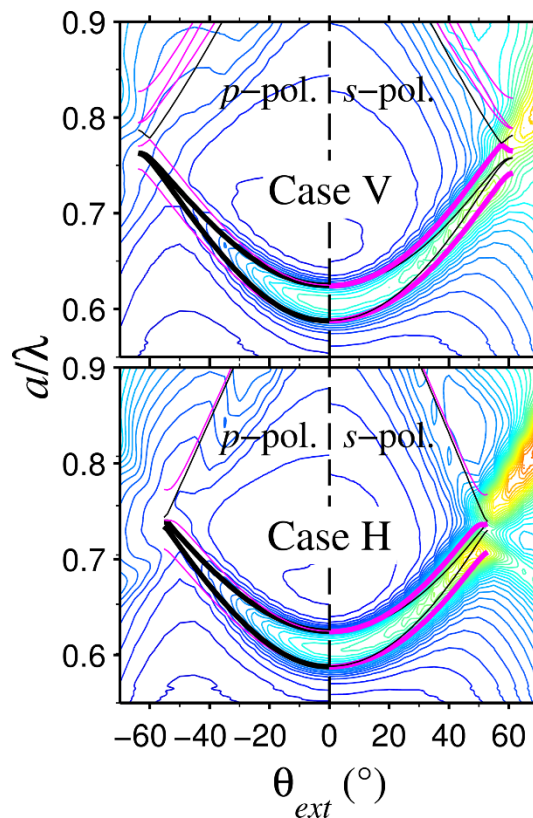
**Figure 3.** extinction ( $-\log T$ ) spectra for, (a) Case V with p-polarized light, (b) Case V with s-polarized light, (c) case H with p-polarized light, and (d) Case H with s-polarized light. Note:  $T$ ,  $a$ , and  $\lambda$  are transmittance, fcc lattice constant, and wavelength of light, respectively.

Extinction spectra for incident angle varies from  $0^\circ$  to  $70^\circ$  and angle step  $5^\circ$  are shown in Figure 3. Spectra were taken for two sample orientations, vertical V and Horizontal H, and for two polarization orientations, perpendicular (s-polarized light) and parallel (p-polarized light). Incident light orientation is defined with respect to opal surface. The shift of the extinction peak with angle satisfies modified Bragg's law. The extinction peak result from diffraction by (111) plane along  $TL$  direction. At incident angle above  $55^\circ$ , light will be simultaneously diffracted by other crystal planes. As a consequence, two or sometime three peaks appear.

There are significant difference between intensity of the extinction peak when rotated sample is illuminated with differently-polarized incident light. The intensity of p-polarized light decreases significantly to nearly zero at incident angle of  $55-60^\circ$  before re-increases. On contrary, the intensity of s-polarized light continues to increase when sample is rotated. Besides, the full width at a half of maximum of the of the stop band decreases significantly for p-polarized light while nearly constant for s-polarized light. The variation of the intensity and the width of the extinction peaks is related to the width of opal stop band. The p-polarized light excites optical modes at the second and

third bands in opal band structure, whereas s-polarized light excites the first and the fourth bands (López-Tejeira, *et al.*, 2002). Larger stop band means a higher light attenuation, or equivalently larger extinction. The calculated band structure of opal films is shown in Figure 4. The bands correspond to s-polarized light are displayed in magenta while the bands correspond to p-polarized light are shown as black curves. The agreement between band width and the width of the extinction peaks is remarkable. A slight increase of the extinction peak for s-polarized light fits nicely with the band structure. However, it must be kept in mind that accurate descriptions of the relation between the width of the stop band and the width and the intensity of the extinction spectra needs a much deeper theoretical consideration that is beyond the scope of this work.

Another interesting feature is the presence of the double peaks at a certain large angle,  $55^\circ$  for Case H and  $62^\circ$  for Case V. The multiple peaks appear due to simultaneous diffraction by crystal planes when light propagation approach edge of Brillouin zone. The avoided crossing of the peaks is observed only for s-polarized light, on contrary the extinction peak disappears at these angles. The avoided crossing for Case H is much stronger than that of Case V. The reason is simultaneous probing of differently-packed opal domains for Case H.



**Figure 4.** Contour plot of the spectra overlapped with calculated opal band structure. The extinction for p-polarized light is plotted with negative incident angle.

At high frequencies, above the band gap, multiple light diffractions causes polarization mixing. Therefore, no significant anisotropy is observed above the stop band. Strong anisotropy at frequencies that corresponds to optical band gap results from anisotropy of equifrequency surface in the band structure.

#### 4. Conclusions

The width and intensity of extinction peak of light propagating inside opal-based photonic crystals show a strong dependency on the polarization of incoming light. The width and the intensity of extinction peak for p-polarized light decrease with incident angle, while they increase for s-polarized light. The decreases in the width and the intensity agrees very well with the width of the stop band upon sample rotation. At a certain large angle, an avoided crossing of two extinction peaks is observed. The avoided crossing is observed for s-polarized incident light. The avoided crossing is much stronger for Case H than case V. The existence of a region where extinction peaks varies significantly at the avoided crossing angle open the possibility for the opal to be used as optical switch where light transmission can be controlled in on and off states by slight variation in incident angle.

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