Experimental evidence of polarization dependence in the optical response of opal-based photonic crystals

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We report angle resolved reflectivity measurements showing the polarization dependence of the pseudogap in artificial opals. This kind of photonic crystals consist of silica spheres ordered in a face-centered-cubic lattice. The analyzed gap originates from the (111) family of planes. It is shown that the width of the Bragg peak observed in the reflectance spectra follows the bands determining the pseudogap, which are selectively excited according to the polarization (*s* or *p*) of the light impinging on the opal. Moreover, it is found that the coupling of light with the photonic bands occurs according to their predicted symmetry, which was assigned by arguments based on group theory. [DOI: 10.1063/1.1582379]

Photonic crystals,[1,2] composite materials for which the dielectric constant undergoes a spatial periodic variation with a period comparable to the wavelength of light, are currently attracting much attention for their possible applications in optoelectronics,[3,4] and the physics associated with the interaction between these structures and electromagnetic radiation.[2,5] In these structures, propagation of light is forbidden along certain directions because of diffraction effects in a range of frequencies, known as a stop band. For a sufficiently large contrast in the dielectric function, stop bands could overlap for all directions leaving a range of forbidden frequencies inside the crystal: A full photonic band gap (PBG).

While some of the systems predicted to feature a full PBG have already been realized,[6–8] the intricate ways in which electromagnetic radiation interacts with these structures is yet to be fully understood. Thus, effective photonic band gaps produced by uncoupled modes[9] (i.e., bands with eigenmodes that cannot be excited by the incident light because of symmetry reasons) have been reported of some two-dimensional photonic crystals.[10,11] Moreover, some authors have predicted a similar phenomenon also taking place in three-dimensional materials.[12–16]

Recently, the classification of eigenmodes according to their symmetry properties has been reported for the case of artificial opals,[17] which consist of silica spheres ordered in a close-packed face-centered-cubic (fcc) lattice. This work shows that the symmetry of the photonic bands provides valuable information to predict the result of polarized light interacting with opals in optical experiments.

The purpose of this letter is to present experimental evidence on how light with different polarization can only couple to certain bands in the lowest-energy range of the band structure of artificial opals. In order to achieve our goal, we analyze the four bands determining the pseudogap in the vicinity of the L point of the Brillouin zone. This can be thought of as the pseudogap associated with diffraction by (111) set of crystalline planes. Angle and polarization resolved transmission measurements were carried out by Tarhan and Watson for dilute colloidal crystals,[18] but their results gave rise to controversial interpretations.[19]

The four lowest bands in the vicinity of the L point for an opal structure are identical irrespective of the direction along the surface of the Brillouin zone for an internal angular range of 0–34° and slightly departing from a common behavior only for higher angles. That is within a circle inscribed in the hexagonal face of the Brillouin zone the photonic bands are isotropic. So, for the sake of simplicity, we can take the ΓLU triangle as representative and assume incident light with its wavevector $\mathbf{k}$ contained in that plane, the tip of $\mathbf{k}$ lying on the LU segment (see Fig. 1). This will permit us to classify the bands by their behavior under mirror reflection with respect to that plane, which coincides with the diffraction plane. Both symmetric (second and third) and antisymmetric (first and fourth) bands are to be found bound to the first Bragg peak.[17]

Let us now consider the symmetry properties of the incident field for any wavevector $\mathbf{k}$ contained in the ΓLU plane. The electric field $\mathbf{E}$ can always be written as a linear combination of two base vectors contained in a plane perpendicular to $\mathbf{k}$. In particular, it is convenient to choose such vectors as parallel $(\mathbf{e}_p)$ and perpendicular $(\mathbf{e}_s)$ to the diffraction plane (see Fig. 1). Hence, $\mathbf{E}=\alpha\mathbf{E}_s+\beta\mathbf{E}_p$ with $\mathbf{E}_s=E_0\mathbf{e}_s$; $\mathbf{E}_p=E_0\mathbf{e}_p$; $\mathbf{e}_s\cdot\mathbf{e}_p=\mathbf{e}_k\cdot\mathbf{e}_p=\mathbf{e}_k\cdot\mathbf{e}_s=0$, and $\alpha^2+\beta^2=1$, where $\mathbf{e}_k$ is a unit vector in the direction of propagation $\mathbf{k}$. When light is linearly polarized perpendicular to the diffraction plane ($\alpha=1$ and $\beta=0$), the electric field $\mathbf{E}$ is equal to $\mathbf{E}_s$. On the other hand, $\mathbf{E}=\mathbf{E}_p$ for the case of parallel polarization ($\alpha=0$ and $\beta=1$). Then, in the p-polarized configuration, the $\mathbf{E}$ field is contained in the diffraction plane, and the mirror reflection with respect to that plane leaves the field vector unchanged. However, for s polarization, the $\mathbf{E}$ field is perpendicular to the mirror symmetry plane so that the symmetry operation changes $\mathbf{E}$ into $-\mathbf{E}$. Details on how

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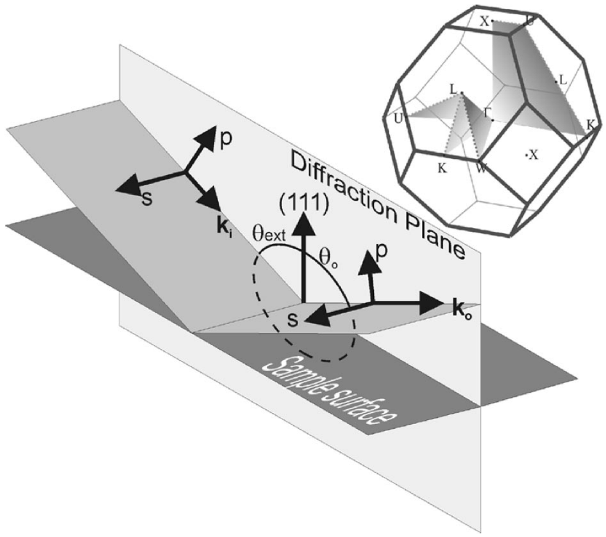

FIG. 1. Geometry of the experiment. The diffraction plane is represented in light gray. The sample surface plane is represented in dark gray. The inset shows the first Brillouin zone for a fcc structure. High symmetry points are indicated.

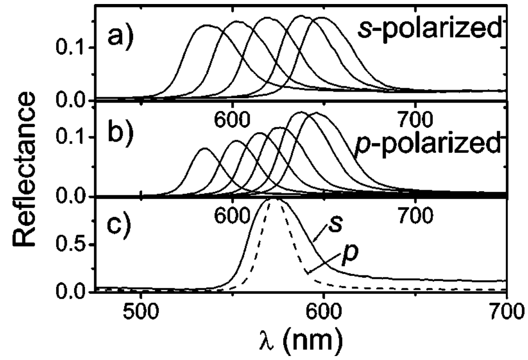

FIG. 2. (a) Reflectivity spectra as a function of external angle of incidence for *s*-polarized light for $\theta_{ext}=6°$, 14°, 23°, 29°, and 34°. (b) Reflectivity spectra as a function of external angle of incidence for *p*-polarized light for $\theta_{ext}=8°$, 14°, 21°, 25°, 30°, and 35°. (c) Normalized reflectivity spectra are shown in solid (dashed) line for *s*- (*p*-) polarized light, for an external angle of incidence of 39°.

this can be done may be found in Ref. 17. Thus, the symmetric (A') eigenstates along the *LU* direction can only be excited by symmetric *p*-polarized incident fields, whereas the *s*-polarized fields can only couple with the antisymmetric (A'') states. So, a polarization sensitivity of the Bragg peak width for oblique incidence is expected, according to band structure calculations.

When studying the optical properties of photonic crystals, reflectivity and transmission are customary techniques. In transmission information is gathered on all possible diffraction processes since these are revealed as reductions in transmitted intensity. By the nature of the experiment, the full thickness of the sample is probed, which ineluctably involves diffuse scattering, loss of signal, and other unwanted effects.[18] On the other hand, reflectance experiments offer information on selected diffraction planes determined by the geometry of the scattering; which makes it suitable for our purposes.

The samples employed in the experiments are artificial opals consisting of silica spheres 297 nm in diameter according to scanning electron microscopy (SEM) characterization. The samples were grown by natural sedimentation, and consist of an ordered fcc array, with the surface parallel to the (111) crystallographic plane. A detailed description of the synthesis can be found elsewhere.[20] A typical sample presents a surface of 9 mm^2 and a thickness of 0.5 mm. Reflectivity measurements were carried out using a collimated beam from a tungsten lamp 1 mm^2 in cross section. The reflected beam was diffracted by a grating and collected by a photomultiplier tube. Angle resolved measurements were performed with linearly polarized light with its electric field perpendicular (*s* polarization) or parallel (*p* polarization) to the diffraction plane, as shown in Fig. 1(b). Reflectivity spectra for *s*- and *p*-polarized light as a function of angle are presented in Fig. 2. In both cases, the shape of the spectra deviate from the flat top peaks with 100% reflectivity predicted by theory for perfect infinite crystals.[21] Extinction caused by diffuse scattering taking place at defects in the bulk and surface of the crystal is known to affect the intensity and shape of the peak rounding the edges but not affecting the full width at half maximum (FWHM).[22,23] In the presence of a mosaic spread, besides an additional decrease in reflected intensity due to nonspecular reflections, inhomogeneous broadening could take place. Another source of peak broadening could be the finite size of crystallites in the sample.[24] These issues will be discussed next, as a comparison with calculated bands is carried out. When comparing the spectra from Figs. 2(a) and 2(b), a difference in the behavior of the absolute reflectivity as a function of the angle of incidence is observed. While the spectra for *s*-polarized light show a nearly constant reflectivity with changes below 1.5% over the entire angular range, the spectra associated to *p*-polarized light present a stronger decrease of nearly 6% in the same range. This difference will be addressed next. Two spectra for *s*- and *p*-polarized light, for an external angle of incidence θ_{ext} of 39°, are presented in Fig. 2(c). Both spectra have been normalized in intensity for comparison. Here, we can already appreciate the difference in width between both peaks, indicating a strong polarization sensitivity.

In order to compare our experimental results with numerical calculations,[17] several issues have been taken into account. Because of the geometry of the sample in our experiment, we are mapping the bands for **k** in the vicinity of the hexagonal face of the Brillouin zone [see Fig. 1(a)]. As the angle of incidence increases, the wavevector **k** approaches the boundaries of the hexagon centered at *L* in the direction of one of the high symmetry points located at the corners (*W*) and centers (*U*, *K*) of the sides of the hexagon. This will be determined by the orientation of the sample surface relative to the diffraction plane. For each possible orientation, different results are expected.

Determining the orientation of the samples is not trivial since they are composed by a mosaic of monodomains typically 20–50 μm in size.[20] While all domains show a preferential out-of-plane orientation with the surface having a (111) orientation, they present a random in-plane orientation. The size of the probe beam being larger than a typical domain will cause averaging over many domains. To avoid this uncertainty, reflectivity spectra were collected from normal incidence to an internal angle of 32° so that the four lowest

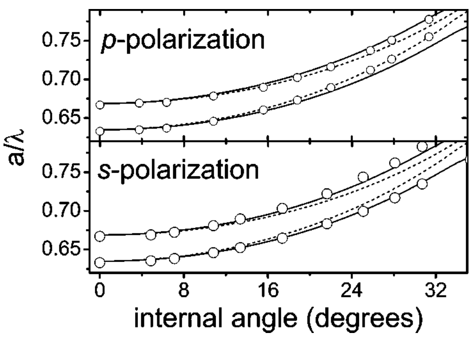

FIG. 3. Calculated photonic bands (lines) and measured band edges from reflectance spectra (circles). Continuous (dashed) lines represent bands antisymmetric (symmetric) with respect to mirror symmetry. Experimental data correspond to light plane polarized parallel (p) and perpendicular (s) to the diffraction plane.

lying energy bands are identical irrespective of the direction of tilting. The correspondence between internal and external angles can be obtained from Snell's law using an effective refractive index n_{eff} which is calculated by fitting the experimental angle dependence to Bragg's law: $\lambda_{\max}=2d_{111}\sqrt{n_{\mathrm{eff}}^2-\sin^2\theta_{\mathrm{ext}}}$, where d_{111} is the interplanar distance for the (111) family of planes, $\lambda_{\max}$ is the peak center and θ_{ext} is the angle formed by the normal to the crystal surface and the incident beam. This expression has proven valid for artificial opals[25] at angles below the avoided crossing region near the U, K, and W points in the first Brillouin zone. The values obtained from this fit were: $n_{\mathrm{eff}}=1.336$ and $d_{111}=244$ nm. If we estimate the diameter of the spheres from this value of d_{111}, we obtain 299 nm in agreement with the value obtained from SEM.

We have assumed the widely used criterion of associating the FWHM of reflectance peaks with the edges of a stop band to compare experimental reflectivity peaks with calculated bands. In the absence of finite size effects or mosaic spread, this approach would be correct as explained herein. Following this criterion, Fig. 3 shows the experimental results for p- and s-polarized light, respectively (symbols), plotted on top of the calculated bands (lines). A fair agreement is observed in both cases. The fact that the FWHM of the peak for small angles of incidence matches with the predicted width implies that finite size effects are negligible and inhomogeneous broadening due to a mosaic spread, if present, will be small. As a matter of fact, the broadening observed for large angles of incidence could be due to a mosaic spread, since its effect on the peak width would become more pronounced as the θ_{ext} increases.[26] According to the previous analysis, the outer bands (1 and 4) couple to s-polarized light, while the inner bands (2 and 3), defining a narrower stop gap, couple to p-polarized light. This explains the difference in intensity for s- and p-polarized light reflectivity as a function of angle shown in Fig. 2. In the case of p-polarized light, the stop band defined by the symmetric bands to which this polarization couples dramatically narrows as we move away from normal incidence, therefore, the penetration length of the incident light into the crystal increases[27] and the effect of extinction by bulk defects will increase, while for s-polarized light the width of the stop band suffers a much smaller narrowing and therefore the change in intensity is smaller. Let us remark that in an experiment using unpolarized light, the outer bands would be probed while the inner ones would remain hidden since the broader peaks contain the narrower.

In this letter, we have measured angle and polarization resolved reflectivity from artificial opals, a system that, while not having a PBG, proves to be an interesting playground for studying the complex interaction of light with photonic crystals. We have experimentally demonstrated how electromagnetic radiation with different polarization will only couple to certain energy bands depending on their symmetry character, in accordance with predictions based on group theory. From these results, it is evident that the symmetry character of the photonic bands must be taken into account in order to properly interpret optical measurements.

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