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Optical response of artificial opals oriented along the Γ*X* direction

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The optical response of artificial opals in the surroundings of the [100] crystallographic direction has been measured by means of microreflectance and transmittance spectroscopies. The results indicate that for such sample orientation, the optical properties are determined entirely by low dispersion bands responsible for out-of-plane diffraction. This is corroborated by phase-sensitive spectroscopy which shows strong anomalies in the measured phase above the onset of diffraction. Such anomalies translate into group velocity values of $\pm c/20$, evidencing slow and superluminal light propagation. These findings could be relevant in developing future routes for enhanced light-matter interaction. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2746936]

Artificial opals[1] have become quite popular materials over the past decade due to the fact that they allow us to test predictions regarding light propagation in photonic crystals[2] in the optical regime, where most applications for such systems are envisaged. These phenomena may include the existence of forbidden spectral bands,[3] modification of radiation dynamics,[4] or the possibility of obtaining regions of ultraslow[5–8] or superluminal light propagation.[7,8] What makes artificial opals so attractive is the fact that, contrary to other three-dimensional photonic crystals, they can be fabricated in a simple and inexpensive way. These samples grow naturally in a face centered cubic (fcc) lattice with the {111} planes parallel to their free surface, and therefore optical characterization is usually performed along the Γ*L* direction and its surroundings in reciprocal space.

When growing samples by some particular methods, such as the vertical deposition one,[9] regions with their surface parallel to {200} planes may also appear, although such behavior is currently not understood and cannot be fully controlled.[10–12] On the other hand, if one wishes to obtain samples along other crystallographic directions in a controlled manner, this can be done by employing patterned substrates to direct the growth,[13–15] cleaving already grown samples,[16] carving bulk thick opals,[17] or, as shown recently, by means of spin coating techniques.[18] However, despite such large efforts to produce artificial opals along crystallographic directions different from the [111], up to now optical characterization has only appeared in the form of normal incidence reflectance on samples exhibiting {100},[10,13,14,16–18] {110}, and {210} (Ref. 16) planes parallel to their surface.

In this letter we present a detailed optical study of artificial opals having the {100} planes parallel to their free surface. Such study is performed by means of polarization- and angle-resolved microreflection and transmission spectroscopies, as well as phase-sensitive spectroscopy. The results compare favorably with theoretical photonic bands and allow us to explore the dispersion relation of this type of materials along regions of the reciprocal space not probed before. From such comparison we see that the optical response of samples oriented like this is entirely due to energy bands presenting low dispersion which are known to originate from diffraction by planes not parallel to the surface.

Samples employed in the present work were grown by means of the vertical deposition method from aqueous suspensions of polystyrene colloids having a diameter of 505 nm (3% polydispersity). Further details on the sample growth may be found elsewhere.[19] In this type of samples, regions with dimensions of the order of hundreds of microns and presenting a [100] orientation can be found.[12] The regions under study had seven layers of spheres parallel to the sample surface. Reflectance and transmittance measurements were collected in a wide spectral range (0.4–3 μm) with a microreflectometer coupled to a Fourier-transform spectrometer (Bruker IF-66s). Spectra were collected from regions 50 μm large for several angles of incidence in the range of 0°–70°. Phase measurements were performed on identical regions employing an experimental setup based on a modified Mach-Zehnder interferometer coupled to a scanning Michelson interferometer.[8,20] Photonic bands were calculated employing a numerical code based on the plane wave expansion method.[21]

Figure 1 shows reflection and transmission spectra collected at normal incidence together with the calculated bands along the Γ*X* direction in reciprocal space. For reduced frequencies under $a/\lambda = 0.9$ the sample behaves as a transparent thin film, showing high transmission and a small reflection consisting of Fabry-Pérot oscillations. This behavior corresponds to a spectral regime where the dispersion relation is that of a homogeneous transparent medium, i.e., a straight line folded at the edge of the Brillouin zone due to the periodicity present in the sample. Opposite to the {111} crystallographic direction no forbidden interval appears at the folding point where the Bragg diffraction condition is met as a consequence of which mode coupling takes place and energy bands split. This is due to the fact that for a fcc lattice the (100) Fourier component of the dielectric constant, which determines the mode coupling strength, becomes zero for this particular filling fraction and refractive index contrast[22] and is a direct consequence of the structure factor taking a null value for this lattice in this direction.[23]

For reduced frequencies above 0.9 a very low reflectance with nearly no structure is observed. On the other hand, in

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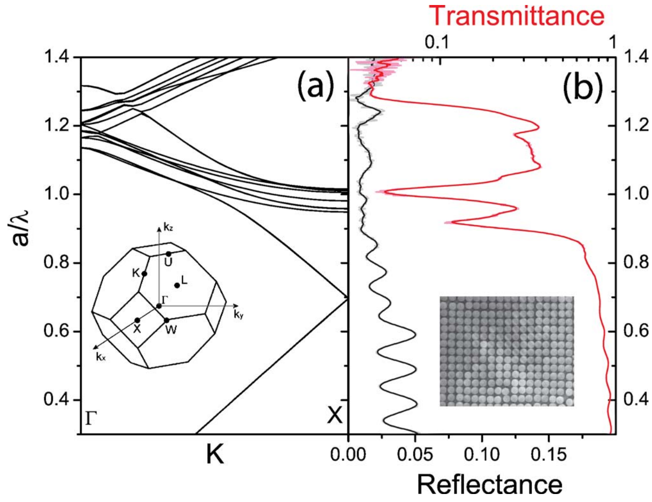

FIG. 1. (Color online) (a) Photonic bands along the Γ*X* segment in reciprocal space. Inset shows the Brillouin zone for a fcc lattice, with the highest symmetry points labeled. (b) Reflectance (black line) and transmittance (red line) spectra measured for an angle of incidence of 3° for a sample seven layers thick oriented with the {100} planes parallel to the sample surface. Inset shows a scanning electron microscopic image of a sample with that orientation.

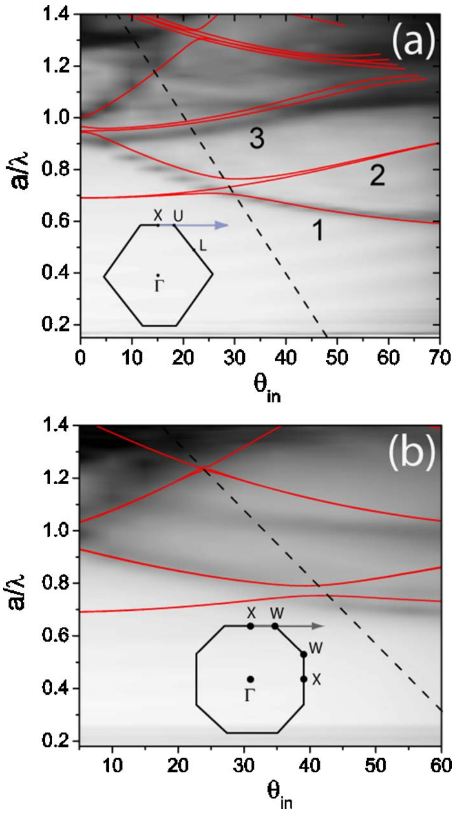

FIG. 2. (Color online) (a) Transmission spectra represented as a contour plot (logarithmic scale) for *s*-polarized light as a function of angle of incidence for a seven layers thick sample having the {100} planes parallel to the sample surface. Calculated dispersion for antisymmetric modes appears as red lines. Inset shows the transverse section of the fcc Brillouin zone across the plane containing the incident and transmitted beams for this sample orientation. The gray arrow indicates the parallel component of the incident wave vector. (b) Same results for the other sample orientation. Black dashed lines indicate the *U* and *W* points in reciprocal space in Figs. 2(a) and 2(b), respectively.

this range the transmission presents sharp dips. For the same frequencies, the calculated dispersion relation shown in Fig. 1(a) presents a number of low dispersion bands which can be associated with out-of-plane diffraction.[24–26] Hence light is not transmitted nor reflected, and it is out-of-plane diffraction which determines the optical response. Therefore, if one wants to retrieve information on the band structure of these systems, transmission measurements are best suited.

The above mentioned optical behavior makes these regions difficult to distinguish from the ubiquitous {111} ones in an optical experiment. In the low energy region they just appear as a transparent medium, and for high energies, the optical response is similar to that of the [111] direction.[27,24] Therefore if broad probe beams are employed in the optical characterization, no evidence of their presence will be found for this type of samples, where such regions coexist with those oriented along the {111} direction.

Next, we study the angle resolved evolution of the optical response for *s*-polarized light along high symmetry directions of the Brillouin zone. In order to orient the samples, diffraction patterns obtained at normal incidence were employed. Figure 2 shows the angular evolution of transmittance spectra for *s*-polarized light, along the corresponding reciprocal lattice paths. Experimental results are shown as contour plots in order to better appreciate the evolution of the optical response. For each sample orientation, photonic bands were calculated along a straight line in reciprocal space, as indicated in the drawings of Figs. 2(a) and 2(b). A symmetry characterization of the eigenvalues was performed in order to identify symmetric and antisymmetric ones, which were subsequently compared with results for *p*- and *s*-polarized light we found that the dispersion relation and the optical response were similar for both polarizations.

In Fig. 2(a) we show a contour plot containing transmission spectra for *s*-polarized light with a wave vector having its normal component along the (100) direction (Γ*X* segment in the Brillouin zone) and a parallel one along the (110) direction (*XU* segment) which increases with the angle of incidence (see inset). We can see that certain features appear in the optical spectra which closely match the dispersion of the photonic bands. In particular, in the spectral region below $a/\lambda = 1.2$ pronounced dips in transmission (dark streaks in the contour plot) following the dispersion of two sets of bands (marked 1 and 3 in the plot). These bands arise from out-of-plane diffraction and, as such, cause light to propagate into directions other than the straight line where the detector is placed in a transmittance measurement. On the other hand, the set of bands (marked 2) does not have any correspondence in the contour plot. This is to be expected, since such set of photonic bands corresponds to the frustrated band splitting at reciprocal lattice points lying on the square facet of the Brillouin zone.

For higher frequencies, experimental features match less accurately the dispersion of the photonic bands. A possible reason for this discrepancy is the finite size of the samples. We should also consider the fact that when measuring transmission, the optical response of the sample is dictated by the dispersion of photonic bands along a certain vector in reciprocal space joining a point at the surface Brillouin zone and its center at the Γ point. Here, instead, we are only considering for comparison the bands at the surface of the Brillouin zone, as is usually done in this type of experiments. Therefore one expects some differences between the dispersion of the bands and the optical response, which in our case takes place for high frequencies.

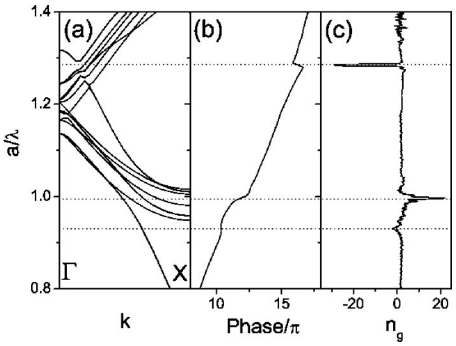

FIG. 3. (a) Photonic bands along the Γ*X* direction in reciprocal space. (b) Absolute phase delay and (c) group index measured at normal incidence for a seven layers thick sample. Dotted horizontal lines indicate the spectral position of the main transmission dips in Fig. 1.

A similar behavior is seen in Fig. 2(b), corresponding to a different sample orientation. Now the parallel component of the incident wave vector is contained in the *XW* segment of the Brillouin zone. Besides the above mentioned small discrepancies, we emphasize the good correspondence between energy bands, calculated assuming an infinite sample, and optical spectra from opals having just seven layers. This is in agreement with previous experimental evidence for (111) oriented samples, which showed that the optical response of the high energy region usually reaches the bulk behavior predicted by the bands for a smaller number of layers[24] as compared to the low energy region.[19]

The fact that the optical response is dictated by diffraction bands makes these sample regions highly interesting from the point of view of light propagation. Diffraction bands are characterized by a complex behavior which includes low dispersion and abrupt changes in slope. This implies a nontrivial scenario for light propagation where the group velocity, defined as the derivative of the band, may take values far from those observed in uniform media.[2] In order to investigate the optical response from this point of view the optical phase of the transmitted light beam, which carries information on the dispersion relation,[7,8] was measured at normal incidence. Figure 3(b) shows that strong anomalies in the measured phase are observed for the same frequencies where diffraction bands occur ($a/\lambda > 0.9$). To understand how does this influences the way light propagates inside the sample, we have extracted the group index $n_g = c/v_g$, where the group velocity is given by $v_g = dk/d\omega$. We can see [Fig. 3(c)] that slope changes in the phase correspond to strong dips in transmission spectra, as one would expect from Kramers-Krönig relations. These are accompanied by peaks in the group index presenting both large positive ($n_g = 20$) and negative values ($n_g = -30$), which correspond to slow and fast (superluminal) propagations, respectively. Notice that the latter, although counterintuitive, have been shown not to violate causality as no signal traverses the sample faster than the speed of light in vacuum.[28]

These results evidence that nonconventional light propagation can be obtained in opal based photonic crystals. While negative group velocities are interesting from a fundamental point of view, slow values may find applications in optical memories, dispersion compensators, or devices where enhanced light-matter interaction is pursued. An example of the latter is the recent observation of laser action in opal base systems at wavelengths where diffraction bands appear.[29]

In summary we have presented an in-depth optical study of artificial opals oriented with their surface parallel to {100} planes. Angle- and polarization-resolved transmission measurements have been presented which find a good correspondence with theoretical dispersion relations and which show that the optical response in this type of samples is entirely determined by diffraction processes involving families of planes other than the ones parallel to the sample surface. The effect of diffraction bands on light propagation has been explored by means of phase measurements evidencing spectral regions of slow and superluminal light propagation which warrants further study to obtain enhanced light-matter interaction.

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