Optical properties of opal-based photonic crystals

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In this work the results of modeling the optical properties of a particular class of artificial metamaterials, namely opal-based photonic crystals, are presented. An approximate model that allows to calculate the photonic band dispersion and the optical spectra of these systems is developed. Subsequently, numerical results in a large spectral range are discussed and compared to experimental measurements, proving the validity of the method.

1 Introduction

In recent years a great deal of attention has been devoted towards photonic crystals (PhCs) [1–4]. Essentially, these artificial materials are the optical analogous of ordinary crystals, since their periodicity in refractive index determines a structure of allowed and forbidden bands for the frequency dispersion of light with respect to the wavevector inside the structure. When light propagation is inhibited for any polarization and any direction inside the crystal a complete photonic bandgap is achieved.

In particular, three dimensional (3D) PhCs can be an answer to the many demands that arise in the field of photonics, since they may allow to achieve a complete control on light propagation due to strong modifications of the 3D density of states. Among this class of materials an important role is played by artificial opals, as they can be produced with easy and low cost processes based on self-assembly from colloidal solutions, such as natural sedimentation or vertical deposition [5]. Many interesting issues can be studied in these structures: to name only a few, we can mention superprism effects [6], superluminality [7–9], diffraction phenomena [10–14] and the control of spontaneous emission [15–17]. Bare opals consist of dielectric spheres arranged in a close-packed face centered cubic (fcc) lattice oriented along the [111] crystallographic direction and they may be infiltrated with high-index materials to obtain the so-called inverse opals. The latter, provided that the index contrast is high enough, present a complete photonic bandgap as it has been demonstrated both theoretically [18] and experimentally [19].

The advancements in self-assembly fabrication, which lead to high-quality crystals, give the possibility to probe the optical properties in the region around and above the second-order stop band, otherwise hidden by disorder and imperfections, where diffraction effects due to higher-order crystalline planes come into play. The aim of the present work is to introduce a reliable theoretical model that allows to calculate optical spectra of opal-based PhCs to be compared to experimental measurements.

The work is organized as follows. In Sec. 2 we give a description of the opal structure, and we compare the photonic band dispersion of the actual opaline structure with that obtained by replacing each sphere with a set of cylinders. In Sec. 3 we present results for optical spectra in both low and high energy range. Section 4 contains a brief summary of the main results.

2 Theoretical model

A direct opal is a crystal made of dielectric spheres in air, which are arranged in an fcc lattice. Usually (111) planes are parallel to the substrate on which the sample is grown, hence the opal structure can be viewed as a stacking of layers in a close-packed arrangement: each layer perpendicular to the [111]...
Fig. 1: (a) Sketch of a close-packed fcc lattice as viewed from [111] direction (hexagonal face in dashed black line). (b) Cross-sectional view of an fcc-lattice oriented along [111] direction; the region where two consecutive planes are interpenetrating is defined by two dashed-dotted lines. The quantities $R_{\text{cap}}$ and $H_{\text{cap}}$ are also defined.

direction is a triangular lattice with periodicity equal to sphere diameter $d = a/\sqrt{2}$, where $a$ is the fcc lattice constant, and it is shifted with respect to the previous one by a quantity equal to $d/\sqrt{3}$ following a sequence ABC, ABC, ... as shown in Fig. 1a. A vertical cross section of the opal structure in the $yz$ plane is shown in Fig. 1b. It can be seen that two consecutive layers are interpenetrating, i.e. the distance between the two is smaller than the sphere diameter. In particular, the distance $d_{111}$ between two planes of spheres along the [111] direction is given by

$$d_{111} = \frac{\sqrt{2}}{3} d = \frac{a}{\sqrt{3}}.$$  

(1)

The height $H_{\text{cap}}$ of the sphere caps and their radius $R_{\text{cap}}$, also shown in Fig. 1b, at the contact point are the following:

$$H_{\text{cap}} = \left(1 - \sqrt{\frac{2}{3}} \right) d, \quad R_{\text{cap}} = \frac{d}{2\sqrt{3}}.$$  

(2)

These sphere caps are arranged in a graphite lattice, i.e. a triangular lattice with a basis of two atoms per unit cell.

The Fourier modal scattering matrix approach [20] gives us the possibility to calculate optical properties of a system constituted by one or more dielectric layers, which may present an in-plane periodicity, but have to be homogeneous along the stacking direction. This method is very convenient as it allows to distinguish zero-order Bragg reflection and transmission from higher-order processes, corresponding to diffraction in directions other than those of the transmitted and reflected beams.

The actual opal structure can be implemented in the code, provided a staircase approximation is introduced, i.e. by subdividing each sphere in a set of cylindrical slices in $xy$ plane in order to fulfill the condition of having a stacking of homogeneous layers along vertical direction. By means of plane wave expansion method [21] one can calculate the photonic band dispersion of an infinite crystal whether the lattice basis is made by spheres or by cylinders. Hence, a comparison between the two band structures can be used to optimize cylinder parameters or to test the validity of the approach, since it is proved that the optical properties of PhCs are strongly dependent on band dispersion.

We obtained very good results by subdividing the spheres in five cylindrical slices [22]. Figure 2 presents results for band structure obtained with this approach; in the same figure the relevant dimensions (cylinder radii and heights) are defined. In particular, cylinders 1 and 5 are equal, as well as cylinders
Fig. 2: Five-cylinder approximation. In thick grey line the photonic band dispersion for the sphere layout is presented; in thin red line the dispersion obtained substituting each sphere with three cylinders. Cylinders parameters are also defined.

2 and 4, to preserve symmetry with respect to the horizontal plane. The total height of the five cylinders is equal to the sphere diameter $d$. The outermost cylinders (no. 1 and 5) have the same height $H_{cap}$ as the overlapping layers and the same radius $R_{cap}$ as the sphere caps at the contact point. Those cylinders are arranged in a graphite lattice shifted by $R_{cap} = d/(2\sqrt{3})$ with respect to the previous layer, while the three inner cylinders (2 to 4) are arranged according to the usual triangular lattice in their respective planes. The total dielectric filling fraction is fixed to that of the spheres.

Once all the previous conditions are satisfied, the best choice for the cylinder parameters is as follows:

$$
H_1 = H_5 = 0.1835d, \quad R_1 = R_5 = 0.2886d,
$$
$$
H_2 = H_4 = 0.1721d, \quad R_2 = R_4 = 0.4309d,
$$
$$
H_3 = 0.2888d, \quad R_3 = 0.5000d.
$$

The radius of cylinder no. 3 turns out to be the same as the radius of the sphere. The condition that the total height is equal to sphere diameter is essential in order to give a good representation of light coupling at the interfaces for an opal of finite size. In this case the interface between air and opal (or substrate and opal) has a corrugated profile due to sphere caps, that is reproduced, at least roughly, by the outermost cylinders.

In Fig. 2 the band dispersion for the fcc structure with both spheres and cylinders is shown. The agreement of the photonic bands is amazingly good, even in the high-energy region (where the folding of free-photon bands corresponds to the occurrence of diffraction) and also along directions different from ΓL, i.e. for non-normal incidence. The overall error between the two band dispersions does not exceed 2%: this means that the approximation error lies within the polydispersity of the spheres in real samples and, consequently, within the possible experimental error.

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3 Numerical results

By means of the scattering matrix method and exploiting the approximate approach previously described, we analyse the optical properties of opal based PhCs. Hereafter we will present a brief overview of two of the main result that have been extensively explained in Ref. [23].

3.1 Optical spectra in the low energy region

We investigate the optical behaviour of opals for light impinging at normal incidence on sample surface and having a frequency corresponding to the region of the first pseudogap in the L point of fcc Brillouin zone and its surroundings. Numerical calculations are compared to experimental measurements, thoroughly presented in Ref. [9].

In Fig. 3a transmittance spectra of samples having an increasing number of layers (from bottom to top) are shown. A region of low transmission is clearly noticeable in the frequency region of the first stop band. The dip becomes more pronounced for increasing sample thickness: Bragg diffraction, responsible of this forbidden energy band, is more efficient if the number of periods along the vertical direction is greater. This is an evidence of an evolution towards the infinite crystal behaviour: for a bulk PhC light would be totally reflected for a frequency corresponding to a forbidden band, where only imaginary values for the light wavevector are allowed inside the crystals.

From the phase delay, also obtained from the scattering matrix method, we retrieved the group index \(n_g = c/v_g\), where \(c\) is the speed of light in vacuum and \(v_g\) the group velocity for light propagating through the crystal. The results are shown in Fig. 3b, for the same sample thicknesses as the transmittance spectra. When the samples are thick enough a region of superluminal propagation is present in the frequency window of the Bragg peak: in this energy range the light beam undergoes a strong extinction inside the crystal, hence the group velocity is no more equivalent to energy velocity and the causality principle is not violated. Moreover slow light behaviour occurs at the two edges of the stop band. For an ideal infinite
Fig. 4: In left and right panels supercell calculations performed for samples having 4 and 5 layers are shown. In middle panel calculated phase delay for samples of the same thickness.

crystal the light velocity would vanishes at these frequencies, since the bands become flat at the edges of a forbidden region. In this example we observe a narrowing of the two peaks for increasing number of layers, evidencing once again the evolution towards the optical properties of a bulk PhCs.

3.2 Diffraction effects at normal incidence

When the wavelength of the light impinging on the medium is nearly the same as the lattice constant, diffraction effects may occur. The optical properties of thin opal films in this energy region are strongly affected by their finite size and can be very different from those of a bulk crystal. By using a supercell approach, i.e. by calculating the photonic band structure of a lattice whose primitive cell is the finite system, embedded in air or in a dielectric medium, that we want to study, we can identify those features which are typical of a structure having a finite number of layers.

Results for these calculations are presented in left and right panel of Fig. 4, together with a close view of the phase delay for 4 and 5 layer samples in the middle panel. An asymmetric supercell has been taken into account, as the opal is surrounded by 4 equivalent layers of air on one side and the same thickness of glass on the other side. Phase anomalies, leading to superluminal values of group velocity, are associated with regions wherein only diffraction, indicated by the flat bands in the frequency dispersion, is present: this phenomenon is clearly ascribed to the 3D periodicity. Besides the $2\pi$ flip between the two phase curves, other phase jumps appear whenever an anticrossing or a stop band occurs in photonic band structure.

When there is no permitted mode in photonic band dispersion for the transmitted beam, such as in the second pseudo gap or in some anticrossing with a low dispersion band, only diffraction states are allowed, resulting in a strong extinction for light traveling through the crystal. In these regions we observe abrupt jumps both in theoretical and experimental phase delay, measured for samples having an increasing number of layers. The group index, derived from phase data, denotes a transition from slow to superluminal and even negative group velocity depending on sample thickness. The latter does not represents a violation of causality principle since we are in a region of low, but non-zero, transmission.
4 Conclusions

The scattering-matrix method is seen to be a powerful approach for calculating the optical response of opal photonic crystals, provided the spheres in the fcc structure are approximated with cylindrical layers. It is important to take into account the caps of the spheres in the overlapping layers in order to obtain a good representation of the photonic bands in the region where diffraction effects become relevant. The optical properties of these systems in the low energy region are fairly well reproduced by our calculations, evidencing a region of superluminal behaviour inside the first stop band and the presence of slow light effect at the two band edges. In the high energy region, where out-of-plane diffraction occurs, a number of anomalous features in the phase delay are present, related to flat bands in the photonic band dispersion of a structure presenting a finite number of layers.

The theoretical approach and an extensive investigation of the optical properties of those systems, for finite incidence angles and in direction different from [111] are fully investigated in the PhD thesis work related to this publication [23].

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References