Anomalous group velocity at the high energy range of a 3D photonic nanostructure

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Abstract: We report on a study of electromagnetic waves propagation in thin periodically ordered photonic nanostructures in the spectral range where the light wavelength is on the order of the lattice parameter. The vector KKR method we use allows us to determine the group index from finite photonic structures including extinction providing confirmation of recently emerged results. We show that for certain frequencies the group velocity of opal slabs can either be superluminal or approach zero depending on the crystal thickness and the unavoidable presence of losses. In some cases, group velocity can be negative. Such behavior can be clearly attributed to the finite character of the three-dimensional structure and reproduces previously reported experimental observations. Calculations show that contrary to the predictions of extraordinary group velocity reductions for infinite periodic structures, the group velocity of real opals may exhibit strong fluctuations at the high energy range. Hence, a direct identification between the calculated anomalous group velocities, for an actual opal film, and the predicted propagating low dispersion modes for an ideal infinite ordered structure seems difficult to establish.

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References and links

1. K. Sakoda, "Optical properties of photonic crystals," Springer-Verlag, Berlin (2005).
2. A. A. Fedyanin, O. Aktsipetrov, D. A. Kurdyukov, V. G. Golubev, and M. Inoue, "Nonlinear diffraction and second-harmonic generation enhancement in silicon-opal photonic crystals," Appl. Phys. Lett. **87**(15), 151111 (2005).
3. M. Scharrer, A. Yamilov, X. Wu, H. Cao, and R. P. H. Chang, "Ultraviolet lasing in high-order bands of three-dimensional ZnO photonic crystals," Appl. Phys. Lett. **88**(20), 201103 (2006).
4. H. Noh, M. Scharrer, M. A. Anderson, R. P. H. Chang, and H. Cao, "Photoluminescence modification by a high-order photonic band with abnormal dispersion in ZnO inverse opal," Phys. Rev. B **77**(11), 115136 (2008).
5. M. Botey, M. Maymó, A. Molinos-Gómez, L. A. Dorado, R. A. Depine, G. Lozano, A. Mihi, H. Míguez, and J. Martorell, "Second Harmonic Generation at the High Energy Range in a Nonlinear Opal Film," Opt. Express **17**, 12210 (2009), http://www.opticsinfobase.org/oe/abstract.cfm?URI=oe-17-15-12210.
6. J. F. Galisteo-López, M. Galli, A. Balestreri, L. C. Andreani, and C. López, "Optical response of artificial opals oriented along the ΓX direction," Appl. Phys. Lett. **90**(23), 231112 (2007).
7. J. F. Galisteo-López, M. Galli, A. Balestreri, M. Patrini, L. C. Andreani, and C. López, "Slow to superluminal light waves in thin 3D photonic crystals," Opt. Express **15**(23), 15342–15350 (2007), http://www.opticsinfobase.org/oe/abstract.cfm?URI=oe-15-23-15342.
8. L. A. Dorado, R. A. Depine, G. Lozano, and H. Míguez, "Interplay between crystal-size and disorder in the high-energy optical response of photonic crystals slabs," Phys. Rev. B **76**(24), 245103 (2007).
9. R. J. P. Engelen, D. Mori, T. Baba, and L. Kuipers, "Two regimes of slow-light losses revealed by adiabatic reduction of group velocity," Phys. Rev. Lett. **101**(10), 103901 (2008).

10. M. Botey, L. A. Dorado, R. A. Depine, G. Lozano, H. Miguez, and J. Martorell, "Anomalous group velocity in a 3D photonic nanostructure," in *Conference on Lasers and Electro-Optics Europe/European Quantum Electronics Conference,* Technical Digest (CD), (Optical Society of America, 2009), paper CK3.2Tue.
11. J. G. Pedersen, S. Xiao, and N. A. Mortensen, "Limits of slow light in photonic crystals," Phys. Rev. B **78**(15), 153101 (2008).
12. N. Stefanou, V. Karathanos, and A. Modinos, "Scattering of electromagnetic waves by periodic structures," J. Phys. Condens. Matter **4**(36), 7389–7400 (1992).
13. N. Stefanou, V. Yannopapas, and A. Modinos, "Heterostructures of photonic crystals: Frequency bands and transmission coefficients," Comput. Phys. Commun. **113**(1), 49–77 (1998).
14. K. Ohtaka, "Scattering theory of low-energy photon diffraction," J. Phys. C: Solid State **13**(4), 667–680 (1980).
15. A. Modinos, "Scattering of electromagnetic waves by a plane of spheres-formalism," Physica A **141**(2-3), 575–588 (1987).
16. L. A. Dorado, R. A. Depine, G. Lozano, and H. Míguez, "Physical origin of the high energy optical response of three dimensional photonic crystals," Opt. Express **15**(26), 17754–17760 (2007), http://www.opticsinfobase.org/oe/abstract.cfm?URI=oe-15-26-17754.
17. F. García-Santamaría, and P. V. Braun, "Are artificial opals non-close-packed fcc structures?" Appl. Phys. Lett. **90**(24), 241905 (2007).
18. L. A. Dorado, R. A. Depine, D. Schinca, G. Lozano, H. Míguez, "Experimental and theoretical analysis of the intensity of beams diffracted by three-dimensional photonic crystals," Phys. Rev. B **78**(7), 075102 (2008).
19. G. S. Lozano, L. A. Dorado, R. A. Depine, and H. Míguez, "Towards a full understanding of the growth dynamics and optical response of self-assembled photonic colloidal crystal films," J. Mater. Chem. **19**(2), 185–190 (2008).
20. M. Botey, M. Maymó, and J. Martorell, "Band-structure determination for finite 3-D photonic crystals," Appl. Phys. B **81**(2-3), 277–281 (2005).
21. X. Ma, J. Q. Lu, R. S. Brock, K. M. Jacobs, P. Yang, and X.-H. Hu, "Determination of complex refractive index of polystyrene microspheres from 370 to 1610 nm," Phys. Med. Biol. **48**(24), 4165–4172 (2003).
22. G. Lozano, L. A. Dorado, D. Schinca, R. A. Depine, and H. Míguez, "Optical analysis of the fine crystalline structure of artificial opal films," Langmuir **25**(22), 12860–12864 (2009).
23. L. A. Dorado, and R. A. Depine, "Modeling of disorder effects and optical extinction in three-dimensional photonic crystals," Phys. Rev. B **79**(4), 045124 (2009).

1. Introduction

In recent years, a growing interest arose in systems that can either provide a reduction of the speed of electromagnetic waves or superluminal light propagation since they could play a key role in the field of optical technology. Such anomalous light propagation has been observed in a variety of settings and these studies have captured significant attention both within and outside the optics community. Among them are two and three-dimensional (3D) periodically nanostructured materials. In such systems, perfectly periodic band structure calculations predict propagating modes with an almost vanishing slope, at the spectral range where the wavelength of light is on the order, or smaller, than the lattice constant. Such modes are associated with strong group velocity reductions [1]. In 3D ordered systems, interesting applications that take advantage of such phenomenon have been proposed to enhance various optical processes [1–5]. Besides, experiments have confirmed the existence of not only slow light, but also superluminal properties in 3D ordered thin nanostructures [6,7]. Although, using periodic boundary conditions only a reduction of the group velocity was predicted. Several experimental and theoretical works have already been devoted to explain the optical new features appearing at this energy range [7,8,5].

In the present paper, we consider the band structure of a perfect infinite face-centered-cubic (fcc) opal made of polystyrene spheres (dielectric constant ε_{sph}=2.5) in air (ε_{air}=1). In the ΓL crystallographic direction, and for $1.05 \leq a/\lambda \leq 1.35$ in reduced frequency units, the geometry of the structure gives rise to such high order extremely flat bands (see Fig. 1). In that energy region of almost vanishing dispersion, the group velocity of ideal infinite opals is expected to reach almost zero for a broad spectrum of wave vectors within the structure. Actual 3D systems exhibit a slowing down of two or three orders of magnitude [9–11,1]. On the other hand, also transitions from slow-light to superluminal regimes with either positive or even negative group velocities have been observed in this same spectral region in high quality opals with known thickness domains [7,5].
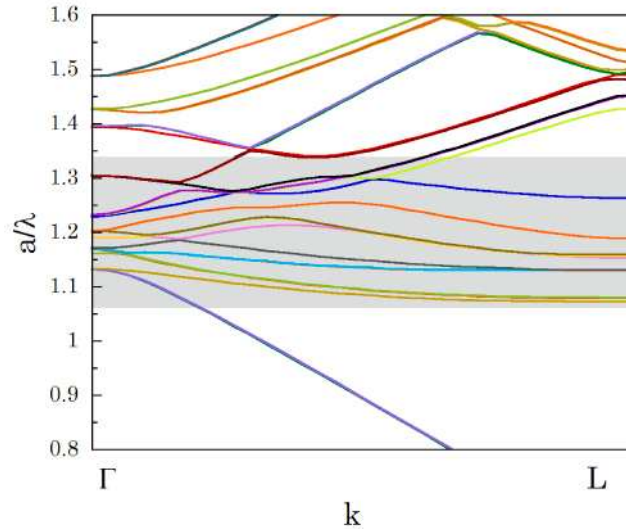
Fig. 1. High-energy band structure in the ΓL direction of an ideal close-packed fcc opal made of polystyrene spheres (ε_s=2.5) in air (ε=1). Energy is expressed in reduced frequency units, lattice parameter, a, over wavelength in vacuum, λ. The shaded region corresponds to the considered high energy range of the spectrum, $1.05 \leq a/\lambda \leq 1.35$.

Recently, calculations based on the code reported by Stefanou et al. [12,13] developed from the vector Korringa-Kohn-Rostoker (KKR) method [14,15] have provided very accurate predictions of the transmission and reflection spectra of opal films [8]. This approach is adequate to treat ordered assembles of spherical scatters and requires reduced computational effort. Besides, this method has also allowed finding an explanation of the physical origin of the optical response in terms of the electromagnetic resonances occurring in the finite sphere ensemble and even reproduces fine details such as the spectral and angular dependence of the intensity of the diffracted modes [16–19,8]. Furthermore, this same approach has already accounted for the experimental observation of a nonlinear light-matter interaction enhancement by slowing light at a particular high energy frequency range [5].

In this work we perform a comprehensive analysis on the influence of finite-size effects on the group velocity and group index of thin artificial opals, in a wide high energy spectral range. Group velocity represents an important aspect to determine light propagation in real periodic structures. In particular, we reproduce the experimental results presented in ref [7]. which were partially described using the scattering matrix method. To this purpose we adopt the same code used in refs [8,10]. which allows considering a finite-width crystal slab and the presence of a substrate. Moreover, the effect of diffuse scattering due to the unavoidable structural imperfections is also accounted by the introduction of an imaginary part in the dielectric constant of the spheres [8,20]. We are able to fully explain the experimentally observed finite-size effects on group velocity and evidence that it may also be strongly affected by the presence of losses.

2. Group velocity determination at the high energy range

We focus our study on light propagation in the ΓL direction of a 3D ordered close-packed face-centered-cubic (fcc) lattice made of polystyrene spheres in air. To provide a rigorous analysis on light transmission from a thin sample, taking into account all interfaces between the crystal and the glass support, we used a vector KKR method with extinction. Since the KKR method refers propagation to a given 2D crystallographic plane, the fields at either side of the crystal are expressed using a plane wave expansion of the reciprocal lattice vectors of the plane. The relative amplitudes for the reflected and transmitted fields are denoted by R_{ml} and T_{ml}, respectively, where the integers (ml) refer to the reciprocal lattice vectors of the 2D

plane. Thus, T_{00} represents the forwardly transmitted and R_{00} the specularly reflected electric fields. Figure 2 displays the lower reflected and transmitted modes for a 10 [111]-layers glass-supported structure. For convergence reasons, the values of the code parameter, LMAX, that controls the cut-off value of the angular momentum in the spherical-wave expansion of the electric fields is taken as LMAX = 9. For calculations shown in this paper, the code parameter, RMAX, of the plane wave expansion of the electric field, is taken as RMAX = 26; meaning that we use the first 41 reciprocal-lattice vectors [13,5]. From the condition of conservation of the tangential component of the wave vectors in a 2D close packed triangular geometry:

$$\frac{a}{\lambda} \geq \frac{\sqrt{2}}{n}\sqrt{l^2 + \frac{(2m+l)^2}{3}} \tag{1}$$

one finds that diffraction in air starts at $a/\lambda \sim 1.63$ — in glass at $a/\lambda \sim 1.07$ — for the first six diffracted modes ($ml=1\text{-}1,\text{-}11,10,\text{-}10,01,0\text{-}1$) and at $a/\lambda \sim 2.83$ —$a/\lambda \sim 1.85$ in glass— for the next six ($ml=-21,\text{-}12,2\text{-}1,1\text{-}2,11,\text{-}1\text{-}1$). Hence, at normal incidence on the [111] planes, and for $a/\lambda \leq 1.63$, the only reflected non-vanishing mode is the specular one, see Fig. 2.a.
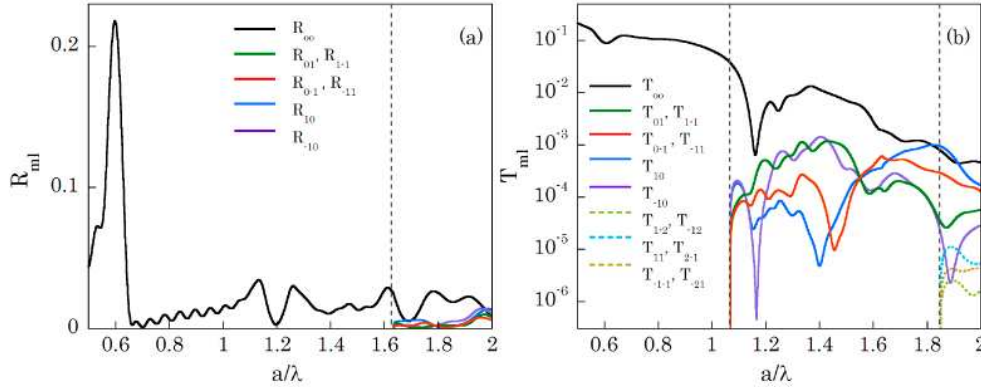

Fig. 2. a) Reflected modes for a glass supported opal made of 10 [111] layers, taking ε_{sph}=2.5+0.1i for the dielectric spheres and ε_g=2.34 for the glass substrate. b) Transmitted modes for the same structure. The vertical dotted lines corresponds to the diffraction cut-off for the six lower order diffracted modes and the next six respectively. All curves are plotted as a function of frequency in reduced, a/λ, units.

As we can appreciate in Fig. 2b, even for a glass supported opal, and the spectral range we are interested in, $a/\lambda \leq 1.4$, the forwardly transmitted field dominates transmittance since it carries more power than all the diffracted modes [8,10]. Despite the high complexity of the scattering phenomena that take place inside ordered nanostructures, the group velocity can be calculated from the forwardly transmitted and specularly reflected fields.

We first calculate the phase delay, θ, introduced by the structure in these fields for a beam propagating in the ΓL direction of the fcc lattice as a function of the frequency, ω. By means of the usual definition:

$$n_g = \frac{c}{v_g} = \frac{c}{L}\frac{\partial \theta}{\partial \omega} \tag{2}$$

—where L is the crystal thickness and c the speed of light in vacuum— we easily obtain the corresponding group index, to which the group velocity v_g is inversely proportional. Figure 3 displays the frequency dependence of both, the phase delay introduced to the forwardly transmitted beam from a 10 [111]-layers-thick crystal, and the group index, for different index contrasts between the nanospheres and the surrounding medium. We observe that for

very low dielectric contrast the phase dependence is linear and the group index constant. Increasing the contrast results in a more complex phase function with either positive or negative slopes, which translates into an anomalous behavior of the group velocity, see Fig. 3b. Positive slope of the phase profile tends to produce slow light, a negative slope fast light. Group index may attain exceptionally high values, greater than the values of refractive indices of the materials used, as well as superluminal negative values, becoming zero for certain frequencies.
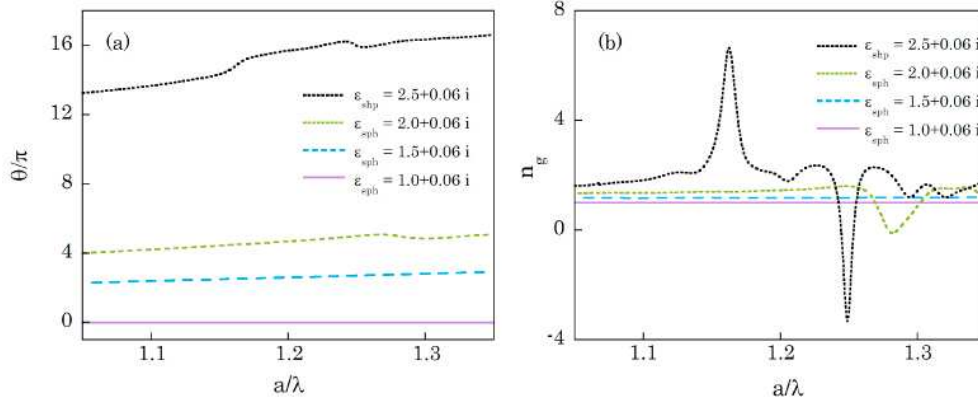
Fig. 3. a) Phase delay, in units of π, introduced to a forwardly transmitted beam propagating along the [111] direction of a 10 [111]-layers opal made of spheres, for increasing values of the dielectric contrast. The imaginary part of the sphere-dielectric function is kept as constant. b) Corresponding group indexes. All curves are plotted as a function of frequency in reduced, a/λ, units.

3. The effect of crystal thickness on the group velocity

For the considered geometry of an fcc lattice made of dielectric spheres in air, an anomalous phase behavior takes place for light propagating in the ΓL direction at the high energy range. In the investigated range $1.05 \leq a/\lambda \leq 1.35$ we find three particularly interesting frequency regions, namely $a/\lambda \sim 1.15$, $a/\lambda \sim 1.25$ and $a/\lambda \sim 1.30$, see Fig. 4. For a given opal, at these three spectral positions, the phase delay introduced to the transmitted electric field does not show a linear dependence on the frequency but exhibits strong slope fluctuations. Since these spectral positions slightly depend on the actual crystal properties we will refer to them as 'A', 'B', and 'C', respectively. Besides, the phase slope depends on the sample thickness, indicating a strong influence of the finite-size effects on the crystal properties.
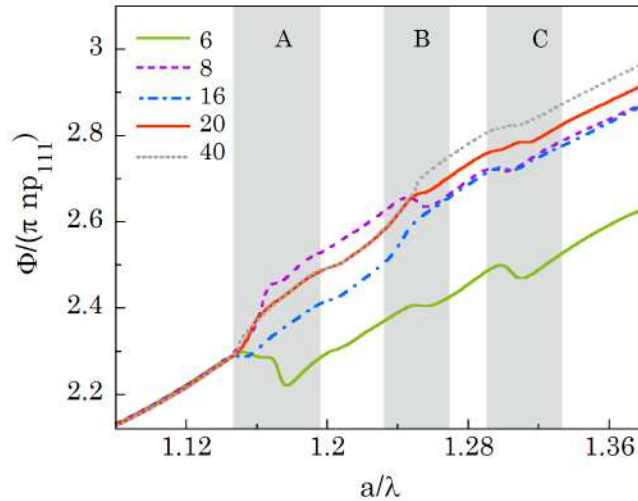
Fig. 4. Phase delay divided by the number of [111] planes, in units of π, introduced to a forwardly transmitted field propagating in the ΓL of the fcc glass supported (ε_g=2.34) structure. All phases are plotted as a function of frequency in reduced, a/λ, units. Nanospheres dielectric function ε_{sph}=2.5+0.04i. The number of [111] planes is indicated in the inset of the figure.

Such phase fluctuations give rise to an anomalous group velocity; the corresponding group index being plotted in Fig. 5. Note that, for a small number of planes, in both spectral positions, A and B, group index exhibits either negative (fast-light) or positive (slow-light) high values depending on the sample thickness. If we increase the number of [111] planes, the opal group index tends to present high positive values. Therefore, for a given thickness, adding only one layer introduces a flip of the phase slope. Such phase jump translates into a change of the group index sign, from negative group index to a high positive group index value. Taking for instance ε_{sph} = 2.5+0.04i, the slope flip at A takes place between 7 and 8 [111] layers, and at B between 12 and 13, see Fig. 5. It is important to note that even for good quality samples, where the effect of the losses is weak, a small imaginary part of the dielectric function has to be considered to describe actual crystals. In Fig. 4 such imaginary part is taken in accordance to the best fits of self-assembled samples [8]. Contrary to what could be expected, increasing the opal thickness does not cause the disappearance of the oscillations and even a second and third change in the group velocity sign takes place at A. In the case here considered, while we observe slow light for a 15-[111]-layers-thick sample, group velocity is superluminal for 16 [111] planes. The third flip takes place between 18 and 19 layers recovering a slow-light regime. Finally, for already thick systems, further increasing the number of [111] planes tends to smooth the fluctuations in this high energy spectral range, either at A or B. Nevertheless, rather than giving rise to the expected broad slow light regime, the average group velocity reduction is small, except for very narrow resonances. In the case of the frequency range denoted by C, group index is negative for opal films and the negative superluminal regime persists even for a 40-[111]-layers-thick structure.
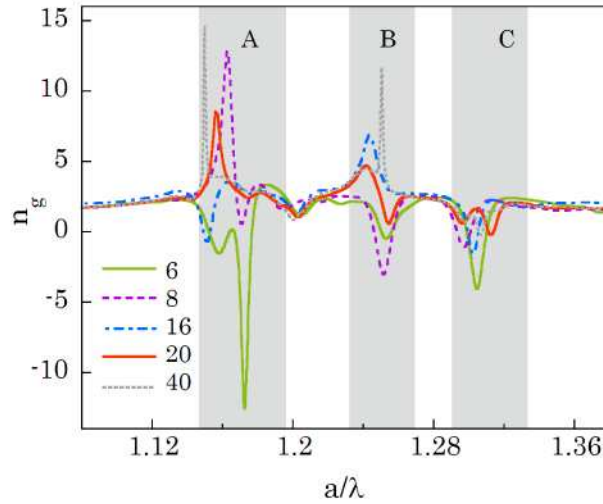
Fig. 5. Group index for different opal thicknesses; all plotted as a function of frequency in reduced units. Nanospheres dielectric function ε_{sph}=2.5+0.04i. The number of [111] planes is indicated in the inset of the figure.

The above predicted anomalous behavior of the phase delay was experimentally measured by Galisteo-López et al. at the spectral range A for a glass supported opal slab made of polystyrene spheres in air [7]. In that case, it was observed that the transition between negative superluminal light propagation to a low positive group velocity took place when the thickness was increased by a single [111] layer, from 7 to 8. The difference in the phase delay introduced to the forwardly transmitted field between both samples, across the jump, was measured to be of 2π. Here we use the vector KKR method with extinction to calculate the phase difference occurring in this particular case in order to contrast our theoretical results with the experimental ones. The sphere's dielectric function is taken in accordance to polystyrene spheres dispersion [21] and a slight lattice relaxation [22]. Taking ε_g = 2.34 for the glass substrate and no other adjusting parameter, the calculations give precisely a monotonous 2π phase shift at the exact frequency where the flip of the group index sign takes place, in excellent agreement with the results reported in ref [7].

We also calculate the group index for different opal thicknesses; keeping all other parameters constant and just changing the number of [111] layers. Our numerical results provide the group index dependence on the number of [111] layers displayed in Fig. 6, which also reproduce with great accuracy the reported experimental results on group index of ref [7]. For a 4-[111]-layers-thick opal the group index is negative and it decreases while increasing the sample thickness. Our predictions confirm theoretically that for an 8-[111]-layers thick opal, group index inverts its sign, changing from superluminal to slow-light propagation. Group index reaches a high positive value which decreases for a 9-[111]-layers thick opal.
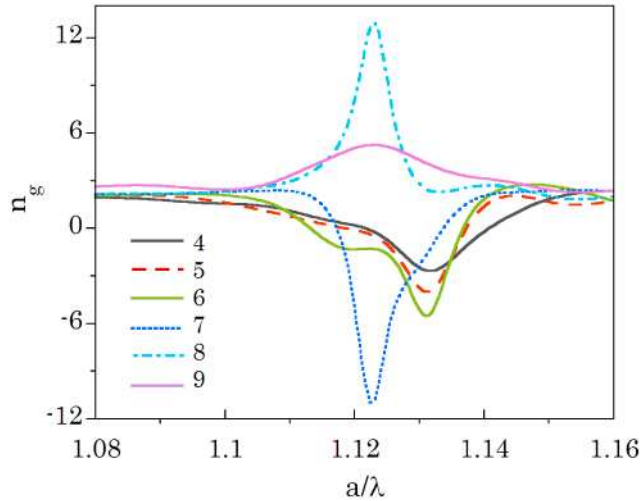
Fig. 6. Calculated group index for different opal thickness as a function of reduced frequency in the A region. The graphic inset corresponds to the number of [111] layers. The dielectric constant of the spheres and the glass support are ε_{shp}=2.6+0.0575i and ε_g=2.34, respectively [21].

4. The effect of extinction on the group velocity: an approach to the analysis of the role of disorder

Since disorder is unavoidable in any practical realization of periodic nanostructures we have also investigated the effect of losses caused by lattice imperfections or dislocations. We have seen that the group velocity does not only depend on the crystal geometry and dielectric contrast, but also the presence of imperfections play a crucial role. For simplicity reasons we have modeled extinction only by the introduction of an imaginary part to the sphere's dielectric function which accounts for the diffuse light scattering produced by imperfections in the crystalline structure [23]. In fact increasing or decreasing such imaginary part of the dielectric function causes group velocity to change. Precisely for the case considered in Fig. 6, a 10% reduction induces a sign flip of the phase between 6 and 7 crystal layers instead of taking place between 7 and 8 layers. This fact suggests that the lack of coincidence between the scattering matrix method prediction and the observations in ref [7], could lie precisely in the role of losses.

For a given opal thickness, the group index transitions from negative to positive values depend on extinction, following the description given above. This holds for any high quality artificial opal film not only at the spectral region A but also at B. As mentioned, the values normally required for the imaginary part of the dielectric constant to fit the reflection spectra of real samples range from 0.04i to 0.12i [8]. For 0.14i we still find superluminal light propagation. For higher extinctions, the group index fluctuations smooth out in general. However, we do not observe a broad slow-light regime but only a narrow resonance at A. Moreover, it is important to note that for a sufficiently high absorption the superluminal character may persist even for thick samples made of over 200 [111] planes. On the other hand, at frequency region C, we observe that, while for small losses group velocity is always negative, for higher values it approaches zero, and finally all effects disappear and we do not find any sign of anomalous group velocity.

5. Conclusions

To conclude, we have theoretically obtained the group index of thin real opals from the phase delay introduced to the forwardly transmitted field, by using a vector KKR method with extinction. At the high energy range of a 3D opal-like structure, we have determined the

presence of three frequency ranges with strong fluctuations of the group velocity. Not only is found the expected slow-light regime but also a much richer optical behavior. Group velocity becomes either superluminical, positive or negative or approaches zero, depending on the crystal length. The above calculated group index explains very accurately previously reported experimental results for thin opal samples [7]. We have also determined that group velocity has not only strong finite-size effect dependence, but it also depends on extinction. For small absorptions, phase jumps and superluminal propagation persist even for a large number of planes, indicating that the finite character of the structure plays a crucial role even for relatively thick samples. Hence, it seems difficult to establish a direct identification between the above calculated anomalous group velocities for a real opal film with extinction and the predicted propagating low dispersion modes of the band structure, for an ideal infinitely periodic nanostructure. Moreover, lattice disorder, losses and finite size effects may represent a limiting factor to the predicted attainable minimum group velocities at that energy range. Increasing the crystal thickness smoothes the group index profiles but does not give rise to a broad slow-light regime. Therefore, we have showed that at the high energy range of a finite periodic structure, real aspects as imperfections and finite-size effects can no longer be ignored to predict light propagation [8,9]. Although great effort has always been made to avoid structural imperfections in largely ordered nanostructures we find that precisely disorder and finite-size effects may give rise to interesting optical properties. Such new features appearing at the high energy range of 3D periodic structures could be advantageous for some applications; they open new ways for the control of light propagation and could have a significant impact on the field of photonic technology.

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