

# Slow to superluminal light waves in thin 3D photonic crystals

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**Abstract:** Phase measurements on self-assembled three-dimensional photonic crystals show that the group velocity of light can flip from small positive (slow) to negative (superluminal) values in samples of a few  $\mu\text{m}$  size. This phenomenon takes place in a narrow spectral range around the second-order stop band and follows from coupling to weakly dispersive photonic bands associated with multiple Bragg diffraction. The observations are well accounted for by theoretical calculations of the phase delay and of photonic states in the finite-sized systems.

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OCIS codes: (050.1940) Diffraction and gratings : Diffraction, (260.2030) Dispersion.

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## 1. Introduction

In the last century, a number of beliefs regarding the propagation of light through a material have been re-examined after the theoretical anticipation and subsequent experimental observation of group velocities which lie either below or well above the causal limit of  $c$ , the speed of light in vacuum [1-13]. Such extremes in the propagation of light are commonly termed *slow-light* and *superluminal* behaviors, respectively. A striking example of superluminal behavior is that of propagation with a *negative group velocity*, where the peak of a pulse propagates across a medium with a negative time delay, appearing as if the peak exits the medium before even entering it [2,3,5,6]. The occurrence of superluminal behavior is often due to the presence of anomalous dispersion and it is acknowledged that such phenomena do not represent a violation of causality. Indeed, although the peak of the pulse travels with a speed above  $c$ , no signal travels faster than light in vacuum [1,7,8]. A number of media have been shown to present such behavior in the visible and infra-red range of the electromagnetic spectrum, including bulk absorptive media [2,3], reflective dielectric multilayers [4], transparent media presenting gain [5], diffraction gratings with sub wavelength apertures [6], metamaterials [9] or serial loop structures operating on the MHz [10], to name a few. At the opposite end, group velocities several orders of magnitude below  $c$  have been obtained. A number of approaches have been used to achieve the so-called *slow-light* regime such as employing media presenting electromagnetically induced transparency [11], Bose-Einstein condensates [12] or periodically nanostructured waveguides [13]. In this work we present experimental evidence, by means of phase measurements, of the existence of

negative group velocities in a three-dimensional (3D) photonic crystal [14] of micrometer thickness, and the possibility to perform a transition from such behavior to a slow-light regime. The crossover from slow to superluminal propagation is controlled by the physical size of the samples and follows from simultaneous diffraction by several families of crystal planes.

The system under study is a 3D photonic crystal (PhC) [14] based on an artificial opal [15]. These nanostructured systems can be easily produced by self-assembly growth. While results for the group velocity have been presented in similar systems [16-19], these previous works were concerned with the spectral region where a pseudogap or forbidden frequency interval appears in the dispersion relation as a consequence of Bragg diffraction by (111) planes, that are parallel to the sample surface and perpendicular to the wave vector. Hence the optical response is similar to that of a one-dimensional (1D) dielectric stack. In the present work we deal with the spectral interval located at higher energies, where Bragg diffraction is dominated by non-(111) crystallographic planes [20-24] and the system displays a truly 3D behavior. Phase measurements are performed on samples with different sphere size and varying number of crystal layers. A spectral region of anomalous dispersion with an abrupt jump in the phase takes place around the frequency where the onset of diffraction occurs. Such jump becomes more pronounced and eventually changes its sign with the number of layers, a behavior which denotes a transition from negative (superluminal) to positive group velocity, as evaluated from the derivative of the measured phase. This unexpected behavior is reproduced by Scattering-Matrix simulations of the phase delay, while the interpretation in terms of multiple diffraction is supported by supercell calculations of photonic eigenmodes in the finite-size system.

## 2. Samples and experimental set-up

For the present study artificial opals made of polystyrene spheres (3% polydispersity in diameter) and grown on glass substrates were employed. These samples grow forming an FCC lattice with the (111) planes parallel to the sample surface. They have an extremely high optical quality and can be grown with control over the thickness [25]. We present results on samples of two kinds, named A and B, with sphere diameter 705 and 505 nm, respectively. The corresponding FCC lattice constants are  $a = \sqrt{2}d = 997$  nm (sample A) and 714 nm (sample B). The phase in transmission at normal incidence is retrieved by means of white-light interferometry employing a commercial scanning Michelson interferometer (Bruker, IFS-66/S) coupled to a modified Mach-Zehnder interferometer [26]. With such a set-up one may measure with a high degree of accuracy both the intensity and the phase of the transmitted beam. In order to obtain the *absolute* phase delay introduced by the sample, one must subtract the contribution of the substrate and determine the number of layers from the condition that the phase tends to zero at vanishing frequency. By this method it was possible in the past to obtain both the real and imaginary parts of the effective refractive index in the low energy spectral region around the first L gap [19].

## 3. Theory

The absolute phase delay across the sample has been calculated employing a Scattering Matrix Method approach in which the spheres forming the opal are divided in cylindrical slices [27,28]. Using a set of five cylinders allows a precise modeling of the opal structure even in the high-energy region. This approach was previously shown to be in very good agreement with experiments performed up to the diffraction cutoff [19]. The parameters employed in the calculation are those mentioned in Refs 19 and 28, taking into account a slight relaxation of the FCC lattice. In order to describe the dispersion relation of a finite-size PhC we have performed band structure calculations with the plane wave expansion method [29] using a supercell oriented along the [111] crystallographic direction.

## 4. Results and discussion

### 4.1 Optical phase and transmission

Figure 1(a) presents transmission measurements for sample A. Figures 1(b) and 1(c) show measured absolute phase for samples B and A, respectively. Figure 1(d) shows calculations of the phase, taking into account the material dispersion [19], for the sample with 705 nm spheres. All results are represented in terms of reduced frequency  $a/\lambda$ , where  $\lambda$  is the wavelength of light in vacuum. Results for the phase are given for samples of increasing thickness, from 4 layers (bottom curve) to 9 layers (top curve).

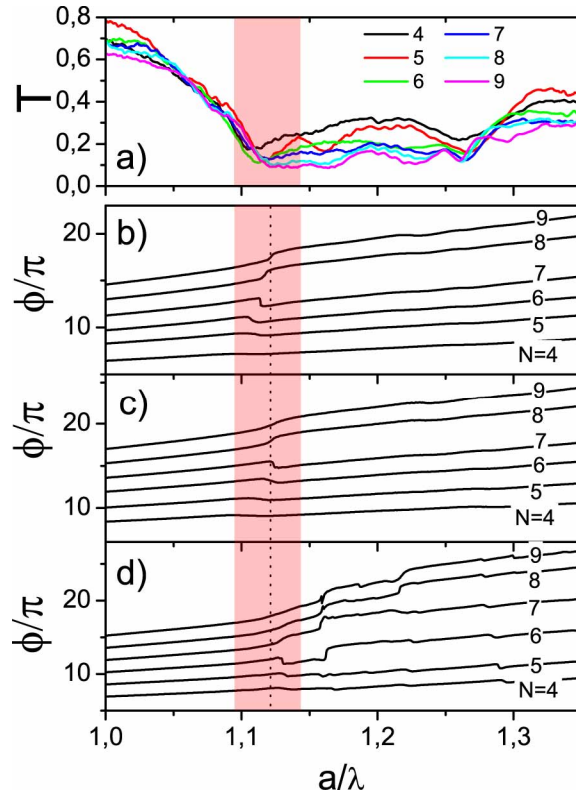


Fig. 1. (a) Transmission for opals made from 705 nm spheres having an increasing thickness. (b,c) Absolute phase in transmission for opals of increasing thickness made from 505 and 705 nm spheres, respectively. (d) Calculated phase for the 705 nm spheres sample.

In the experimental phase a linear behavior is observed which is truncated at  $a/\lambda \sim 1.12$  by a jump. Such jump becomes better defined with growing number of layers and eventually, when going from 7 to 8 layers, it flips its sign. For thicker samples the jump becomes less defined again. For higher frequencies, the measured phase shows little or no structure. The phase jump takes place at the same reduced frequency for both sphere diameters, hence it scales with the lattice parameter, pointing to an intrinsic property of the interaction of the incident light with the 3D periodic lattice.

The theoretical calculations in Fig. 1(d) reproduce the jump in the phase at a similar reduced frequency (variations of 1% are found, below the uncertainty associated to the polydispersity of the spheres). For frequencies above the phase jump a well defined structure is found with other jumps which can also change their sign, as is the case of those at  $a/\lambda \sim 1.16$  and 1.22. Further, in the theoretical calculations the sign flip takes place as the number of (111) planes goes from 6 to 7 instead of 7 to 8, as in the experimental case. These two

discrepancies are probably associated with the simplified model of the sample structure. For the frequency range under consideration the probe wavelength is smaller than the lattice parameter, and hence sensitive to any deviations between the real and the modeled, possible necking of the spheres, etc. Also the small disorder present in the samples has a more intense effect for smaller wavelengths [30]. Nevertheless, theory reproduces the main feature present in the absolute phase at  $a/\lambda \sim 1.12$ , within the spectral region where the onset of out-of-plane diffraction is expected for these samples [23,24]. Figure 2 shows the difference between the phase in transmission before and after the sign flip (6-7 layers for the theory and 7-8 for the experiment). Notice that the change in phase across the jump is  $2\pi$  (see arrow in the Fig. 2), as precisely reproduced by the theory.

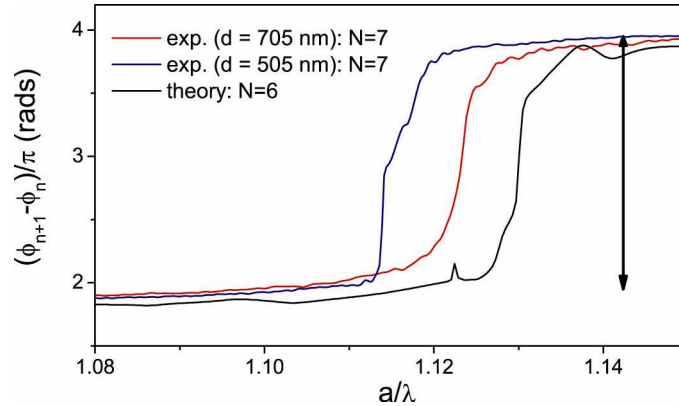


Fig. 2. Difference between the absolute phase delay for two sample thicknesses, before and after the phase flip has taken place, from 6 to 7 layers in the calculation (black curve) and 7 to 8 in the experiment (blue and red curves).

#### 4.2 Group velocity

The implications of such behavior in the way light propagates through the system are better seen if one calculates the group index, commonly defined as:

$$n_g = \frac{c}{v_g} = \frac{c}{D} \frac{d\phi}{d\omega} \quad (1)$$

where  $v_g$  is the group velocity and  $D$  the sample thickness. Results for sample A and the corresponding theoretical calculation are represented in Fig. 3(a) and 3(b), respectively. The effect of the phase jump at  $a/\lambda \sim 1.12$  is now strongly enhanced. For a thin opal (4 layers), a region of negative group velocity is present. As the number of layers increases, this region becomes spectrally thinner and more pronounced. Eventually, when going from 7 to 8 layers in the experiment (6 to 7 in our model) the peak in the group index changes its sign, evidencing small but positive group velocities. As the sample thickness keeps increasing, the peak becomes less intense, in accordance with the phase jump becoming less sharply defined. Therefore, just by changing the sample thickness by one layer, we may switch between two distinct light propagation scenarios where superluminal and slow-light regimes are observed in a given frequency window.

It is important to notice that the determination of the phase and of the group velocity is fully valid even in the presence of extinction. Indeed, even in previous situations where anomalous dispersion is associated with extinction [2-4,6,9,31], the group velocity has nevertheless proved to maintain its definition of the derivative of the phase with respect to the frequency. In our case, the negative group velocity is also associated with a region of strong extinction, but the transmission through the sample is high enough (absolute transmittance ca. 10%, see Fig. 1(a)) to allow a very precise determination of the phase.

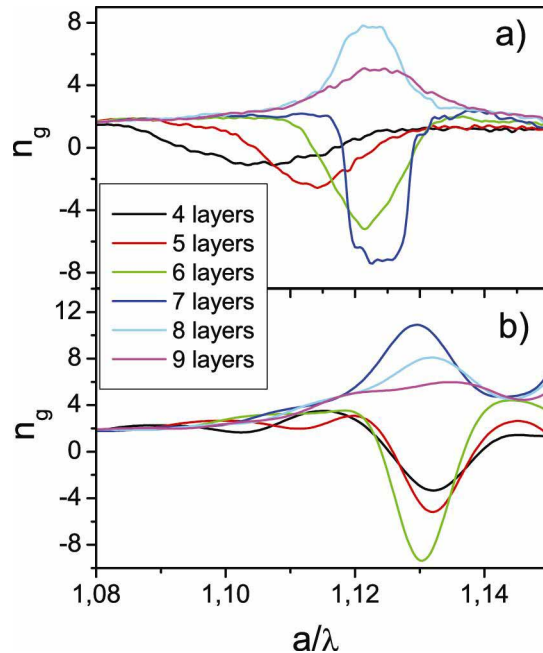


Fig. 3. Group index as a function of reduced frequency for samples made of 705 nm spheres, with increasing number of layers: (a) experimental, (b) calculated results.

We now consider the implications of the above results in terms of real transit times. In particular, we consider the sample with 705 nm spheres and 7 layers, for which a spectral region 8 nm wide is found where  $v_g$  is negative an almost constant (see Fig. 3(a)). The shortest Gaussian pulse with such spectral width would have a 290 fs duration. The time needed for a reference pulse to traverse a distance in vacuum equal to the sample thickness (4  $\mu\text{m}$ ) is 13.5 fs. On the other hand, the group delay when traversing the sample is  $-107$  fs, which would result in a pulse advance.

Bearing in mind the fact that transmission is 10% in this spectral range, the transmitted beam would remain within a reference pulse which propagated in vacuum. It remains to be further studied whether such advance could be enhanced. The dispersion relation of artificial opals, and hence its optical response, may be modified by appropriately modifying the topology of the unit cell [32]. Future studies could focus on trying to optimize the pulse advance; however lower transmission is also expected under such conditions, which would keep the advanced pulse within the reference one.

#### 4.3 Comparison with theory

In order to gain further understanding on the physical origin of this phenomenon we have compared the evolution of the calculated phase for increasing number of layers with the dispersion relation of the infinite system (see Fig. 4). To calculate the dispersion relation we must assume a constant refractive index for polystyrene ( $n=1.59$ ) and an ideal FCC structure, hence we show the calculated phase for the same parameters [30]. In Fig. 4 the grayed areas highlight the spectral regions where normal dispersion of collinear propagation in effective medium is disrupted either by zone-edge gap formation or in-zone anticrossings by coupling with non-collinear propagating bands of appropriate symmetry [22]. It can be noticed from Fig. 4 that in the present case the phase jumps are close to the lower edge of the second-order stop band (indicated by the wider grey bar in the figure). It is evident that strong anomalies in the phase, which translate into superluminal values of the group velocity, take place when only diffraction bands, which send light into directions other than the incident one, are available. In this spectral range light propagation inside the crystal is far from simple, and

takes place in the form of multiple diffracted beams which interact mutually. Such interaction is probably the origin of the complex phase gathered by the traveling beam, which is measured in the experiment. This effect is then caused by a 3D periodicity and its physical mechanism differs dramatically from that described in previous work on light propagation because of the existence of a complicated multiple diffraction process. Further, it is the finite size of the system that plays a crucial role in dictating the transition from positive to negative group velocities.

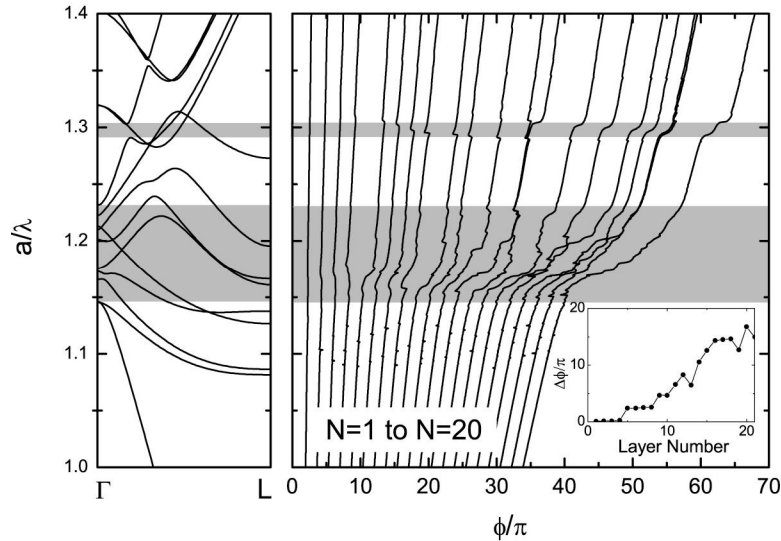


Fig. 4. Band structure along the  $\Gamma L$  direction for an artificial opal made from spheres of refractive index 1.59 (left panel). Calculated phase for a sample having an increasing number of layers, from 1 to 20, left to right (right panel). Grey boxes indicate the spectral regions where only diffraction bands are found in the dispersion relation. Inset shows the phase difference between the two edges of the second order pseudogap (taken from  $a/\lambda=1.14$  to  $1.23$ ).

This mechanism differs from that reported in the micro-wave region by Solli et al [31], who observed similar phase anomalies arising from a birefringent behavior producing mutual interaction between two forward-propagating beams with orthogonal polarizations. In the present case the situation is more complex, as the interacting beams are a consequence of Bragg diffraction inside the sample, and therefore a simple model cannot properly describe such behavior. The crucial role played by finite-size effect is highlighted by the inset to Fig. 4, which shows the calculated jump in phase across the second-order stop band as a function of the number of layers. This curve has an overall increase, albeit with plateaus that seem to occur at particular values of  $\Delta\phi$ .

In order to properly describe the optical response of the samples, one should consider the dispersion relation of the finite system, which may be obtained by means of band structure calculations employing a supercell. For the present case we have considered a hexagonal supercell consisting of blocks of  $N$  (111) crystal planes separated by the equivalent of four (111) planes, two of air and two of glass, in order to calculate the dispersion relation of a  $N$ -plane crystal embedded between air and glass. Considering a larger air thickness did not affect the spectral position of the dispersionless bands present in the dispersion relation. Only a slight change in the slope of the normal dispersion bands arises, because the average dielectric constant of the system is reduced as the air thickness increases. Notice that the calculation takes into account the actual crystal structure and interfaces, as well as possible presence of surface states. The resulting photonic dispersion, together with the calculated phase, for two samples having  $N = 4$  and  $N = 5$  (111) planes are shown in Fig. 5. The band structure shows a number of flat dispersionless levels which correspond to localized states (that do not

propagate across the sample) coupled to a set of bands (highlighted in the figure) with a normal dispersion corresponding to the forward propagating modes in the infinite structure. A number of small phase anomalies are present around  $a/\lambda \sim 1.11$  which correspond to regions where normal dispersion bands undergo anticrossings with dispersionless bands, although their small spectral width probably prevents them from being observed in the experiment. At those frequencies where the phase jump changes its sign, only diffraction bands are present even for the finite-system dispersion relation. This demonstrates the presence of an intimate relation between the sign flip of the group velocity and multiple diffraction processes in the finite-sized system under consideration. For a complete description of the physical phenomenon, one should extract the exact coupling efficiencies to each band as well as their Fourier components, something which is out of the scope of the present work.

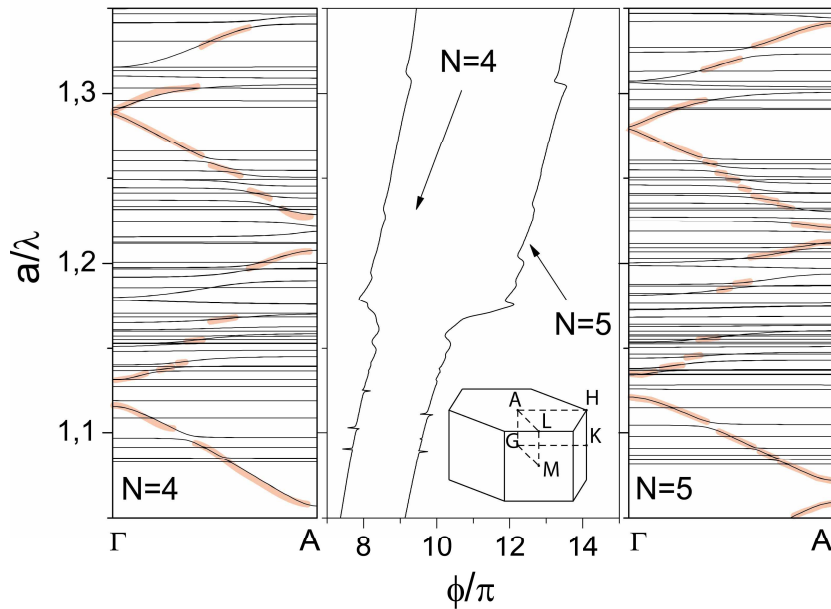


Fig. 5. Left and right panels show dispersion relations for opals having 4 and 5 (111) layers, obtained with a supercell method (the hexagonal Brillouin zone with symmetry points is shown in the inset). Bands reminiscent of normal dispersion are highlighted with red semi-transparent lines. Central panel shows calculated phase for the same structures.

## 5. Conclusion

In summary, we have studied a nanostructured material with a new scenario for light propagation in which the group velocity displays an extremely rich behavior. In particular we have presented experimental results, supported by theoretical calculations, which show a transition between slow and negative (superluminal) group velocity in 3D PhCs based on artificial opals operating in the visible-infrared part of the electromagnetic spectrum. Such behavior is associated with the existence of strong anomalies in the phase delay, which take place in spectral regions where strong out-of-plane diffraction occurs and light propagates inside the sample as a set of interfering diffracted beams. These phenomena are crucially dependent on the 3D nature of the investigated PhCs. Further, the existence of a frequency window with a transition between positive and negative values of the group velocity is a finite-size effect which manifests itself when the sample thickness is varied. The present findings open up exciting questions regarding the interaction of electromagnetic radiation with matter in nano-structured materials in the high-energy spectral region. Recently, it has been



shown that for similar samples in this spectral range strong angular redistribution of spontaneous emission of internal light sources [34] as well as lasing action [35] can be observed. Such results further confirm the interest of phenomena regarding the strong modification of light propagation achievable in a small-sized nanostructured material.

### **Acknowledgements**

This work has been partially financed by the EU network of excellence FP6-511616-NoE PHOREMOST, the ESF COST action P11, the Spanish MEC through MAT-2003-01237 and MAT2006-09062 projects, the Italian MiUR through Cofin program and the CARIPLO Foundation. J. F. Galisteo-López was sponsored by the Postdoctoral Program of the Spanish Ministry of Science and Education.