

MICROCAVITÉS ET CRISTAUX PHOTONIQUES *MICROCAVITIES AND PHOTONIC CRYSTALS*

Three-dimensional photonic crystals as a cage for light

A. Femius Koenderink, Patrick M. Johnson, Juan F. Galisteo López, Willem L. Vos
Van der Waals Zeeman Instituut, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

Received and accepted 23 November 2001

Note presented by Guy Laval.

Abstract

We review recent developments in three-dimensional photonic crystals. State of the art fabrication methods, such as layer-by-layer micromachining, self-assembly and various etching, lithographic and holographic techniques are discussed. We present an overview of optical studies of photonic band gap formation, such as reflectivity, transmission and time-resolved pulse propagation experiments. The fundamental issues associated with disorder and absorption are also considered. Progress towards the ultimate goal of full spontaneous emission control is reviewed. Finally, remaining open questions are summarized. To cite this article: A.F. Koenderink et al., C. R. Physique 3 (2002) 67–77. © 2002 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

photonic band gaps / microphotonics / microfabrication / X-ray diffraction / spontaneous emission / quantum optics / diffraction / scattering

Les cristaux photoniques tridimensionnels : une cage pour la lumière

Résumé

Nous examinons les développements récents dans le domaine des cristaux photoniques tridimensionnels. Nous présentons et discutons des méthodes de fabrication parmi les plus avancées, telles que le micro-usinage couche par couche, l'auto-assemblage et les diverses techniques de gravures, lithographies, holographies. Nous donnons une vue globale des études optiques visant à mettre en évidence la formation de gaps photoniques, telles que la réflectivité, la transmission et les études de propagation d'impulsion résolues en temps. Les questions de fond soulevées par la présence du désordre et de l'absorption sont aussi considérées à leur tour. Nous passons en revue les progrès faits vers le but ultime, à savoir le contrôle total de l'émission spontanée. Finalement, les questions restant ouvertes sont résumées. Pour citer cet article : A.F. Koenderink et al., C. R. Physique 3 (2002) 67–77. © 2002 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

bande interdite photonique / micro-photonique / micro-fabrication / diffraction aux rayons X / émission spontanée / optique quantique / diffraction / diffusion

1. Introduction

There is a fast growing interest in photonic crystals: two- or three-dimensional (3D) periodic composite materials with typical length scales a matching the wavelength of light λ [1,2]. The essential optical

E-mail address: fkoender@science.uva.nl (A.F. Koenderink).

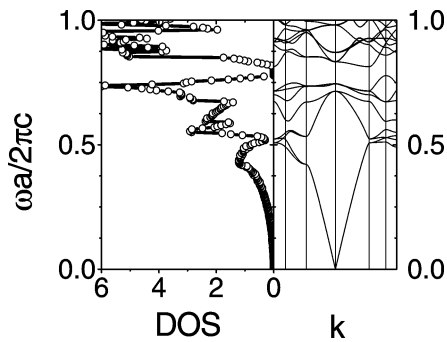


Figure 1. Photonic density of states (DOS) of an fcc inverse opal of air in dielectric with $\epsilon = 11.9$ as a function of normalized frequency $\omega a / (2\pi c)$. With its strong modulations, the DOS differs widely from the quadratic dependency in a homogeneous system. The photonic band gap is the range of zero DOS near $\omega a / (2\pi c) = 0.8$. The right panel shows the complex dispersion relations for wave vectors between the high-symmetry points X, U, L, Γ , X, W, and K that are indicated by the vertical lines.

properties of photonic materials are determined by the spatially varying refractive index, analogous to the periodic potential for an electron in a crystal. The interaction between light and a photonic structure can be gauged with a photonic ‘strength’ parameter Ψ , defined as the polarizability α per volume v of each scattering unit. If Ψ is large, e.g. near atomic resonances or due to large refractive index variations, multiple scattering dominates the linear and non-linear optical properties [3].

The main ‘raison d’être’ for 3D photonic crystals is to provide a ‘photonic band gap’. At frequencies inside the band gap, the density of states (DOS) is zero in these structures because Bragg diffraction prevents propagation of light in any direction (Fig. 1). Therefore, photonic crystals should play an important role in quantum optics and cavity QED, as illustrated by the original paper of Yablonovitch [4]. Probably the most eagerly awaited phenomenon is complete inhibition of spontaneous emission: excited atoms inside a crystal with their transition frequencies tuned to the band gap cannot emit photons, since vacuum fluctuations are suppressed by the band gap [4–6]. In the presence of controlled disorder, Anderson localization of light occurs [7]. In this case a photon is caged at a defect, since it is unable to traverse the bulk of the photonic band gap crystal. Such a defect serves as a cavity with a high quality factor [8]. In addition, spectacular modifications of quantum optical dynamics have been predicted such as transparency, fractional localization, or (multi-) photon–atom bound states [9–11]. In this dossier, we review the current status of experiments on 3D photonic crystals, starting with the fabrication aspects.

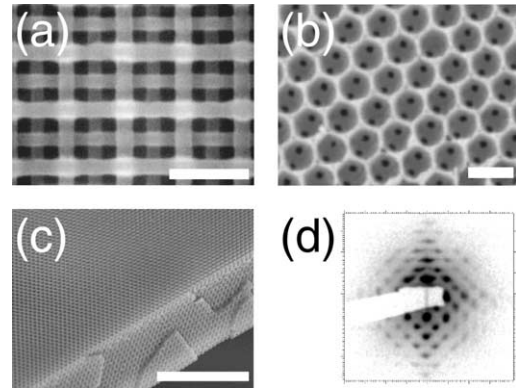
2. Synthesis of 3D photonic crystals

The synthesis of photonic band gap crystals continues to be a rich problem after over a decade of work. This may come as a surprise since the first photonic band gap material was created a few years after the first theoretical paper on photonic crystals. However, this photonic crystal functioned in the microwave range [12]. A main goal of the field is the fabrication of photonic band gap materials at optical frequencies, which will allow both fundamental studies and industrial applications to go forward. The numerous fabrication methods adapted to achieving this goal can roughly be categorized into three groups: layer-by-layer fabrication; self-assembly using mostly colloidal particles; and etching by lithographic and holographic techniques. In some cases, the power and flexibility of these methods may be increased by casting the high index photonic crystal from a low index template. While photonic band gaps have recently been claimed at near IR wavelengths [13–16], the challenges of disorder and finite size effects, as well as the inherent difficulty of proving the existence of a photonic band gap leave the field of fabrication open for new ideas.

2.1. Layer-by-layer micromachining

The layer-by-layer fabrication approach has allowed fabrication of photonic crystals for near-IR frequencies from high-index semiconductors. A diamond symmetry may be achieved, which should result in band gaps in the frequency range of first order Bragg diffraction. Lin and collaborators created a

Figure 2. (a) Scanning Electron Micrograph (SEM) of a layer-by-layer micromachined woodpile structure from reference [14], courtesy of Susumu Noda. (b) SEM of a TiO_2 inverse opal, from reference [27]. (c) SEM of a 3D photonic crystal template in photoresist produced holographically, from reference [31], courtesy of Andrew Turberfield. (d) Typical SAXS pattern from an inverse opal, from reference [27]. Scalebars represent (a) 1 μm , (b) 0.5 μm and (c) 10 μm .



micromachined so-called ‘woodpile’ arrangement [13]. In this technique, the periodic array is built up by depositing lines of polycrystalline silicon into micron sized SiO_2 trenches in successive layers. Once the structure is built up, the SiO_2 is removed with HF leaving a silicon–air photonic crystal. Noda et al. have produced photonic crystals in GaAs using a similar layering concept (Fig. 2a) [14]. These photonic crystals are created by carefully aligning, stacking, and fusing prefabricated GaAs wafers.

These techniques are powerful because they use known semiconductor fabrication techniques and produce high- Ψ photonic crystals. Also, there is considerable freedom to choose the structure. This freedom may allow certain waveguide geometries to be introduced into the photonic crystal for applications purposes [17]. One current issue is the rather limited thickness that can be achieved, and it may be a challenge to fabricate optical circuits within each layer. Other approaches to layer-by-layer fabrication may overcome some of these restrictions. For example, using autocloning bias-sputtering, Kawakami and coworkers have deposited 2D periodic patterns, one layer at a time, to build up a 3D structure [18]. When combined with an e-beam lithography etch step, a wide photonic band gap is expected.

2.2. Self-assembly

Self-assembly of colloidal crystals results in truly 3D periodic arrays, easily reaching hundreds of microns of thickness, thus solving the thickness issue mentioned above. While the existence of such colloidal crystals or opals has been known for a long time [19], the use of such structures as photonic crystals per se is limited by a low photonic strength Ψ . A major step forward has been the recent use of self-assembled structures as templates for high index materials [20–23]. See for reviews references [24–26]. In our group, highly ordered ‘inverse opals’ are created via precipitation of TiO_2 from a tetra-propoxy-titane precursor infiltrated into a latex opal, and subsequent removal of the template by calcination (Fig. 2b) [27]. Inverse opals with higher index materials such as silicon have been fabricated by chemical vapor deposition onto silica opals [15,16]. Such innovations in 3D templating can be applied to a wide variety of fabrication schemes. While inverse opals offer an elegant solution to the problem of size, they are accompanied by their own challenges. Widely used colloids with a hard-sphere like interaction potential have a tendency to form random stacks of hexagonal planes, a structure with intrinsic disorder along the c -axis. Charged colloids as used in our lab, yield well-ordered crystals with the fcc arrangement [27], a structure that develops a band gap in the range of second order Bragg diffraction (Fig. 1). Higher order gaps are relatively narrow and sensitive to disorder, however, hence such photonic band gaps are expected to be fragile [28]. To date, fabrication of a diamond lattice, and fabricating intentional defects from colloids remain open issues. On the other hand, innovative surface deposition methods are being developed that yield crystals with well-defined thickness, morphology, orientation, or shells [16,29,30].

2.3. Etching, lithography, holography

Various etching and other lithographic techniques may be used to create a periodic structure by removing material from a solid block. These techniques have the potential to combine the size of the colloidal crystal with the controlled order of layer-by-layer assembly. An elegant approach, recently described in reference [31], uses a four-laser holographic method to create a 3D periodic interference pattern in a block of photoresist. The high intensity regions in the interference pattern render the photoresist soluble, allowing the 3D periodic template to be formed (Fig. 2c). TiO₂ inverse structures can then be formed from the template. The holographic technique has the advantage of speed since the entire pattern in the photoresist is created in nanoseconds. Also, a variety of crystal structures can be created by varying the orientation and polarization of the four laser beams. It remains to be tested to what degree ideal defect-free structures can be experimentally realized, how absorption of the laser beams affects uniformity in thickness, and how intentional defects and waveguides could be incorporated in the structure. Direct techniques for carving diamond structure and other 3D structures from a solid block of material using X-ray lithography [32] and focused-ion-beam etching [33] have also been recently reported. Modulated photo-electrochemical etching in silicon has yielded an interesting simple hexagonal structure [34].

2.4. Structural characterization

Since the structure of photonic crystals is defined on the scale of hundreds of nanometers, scanning electron microscopy (SEM) is ideal for characterizing surfaces and cross sections of samples (Fig. 2, a–c). Understanding the 3D degree of order, however, is essential for the interpretation of optical experiments, but requires innovative techniques. Recently, we have initiated small angle x-ray diffraction experiments to identify the crystal structure of colloidal crystals, opals, and inverse opals unambiguously as fcc and to determine the content of the unit cell (Fig. 2d) [27,35]. In addition, essential parameters that gauge disorder were quantified: the size polydispersity of colloids [36] and for the first time of macropores in inverse opals [27]. The variations were within the theoretical limits for retaining a photonic band gap in the presence of disorder [28]. Moreover, particle displacements in colloidal crystals were found to decrease with increasing density, and were smaller than theoretically predicted [37]. Thus, small angle X-ray diffraction is a powerful tool to study the structure of 3D photonic crystals, and the method is complementary to microscopy since it yields parameters that are otherwise difficult to access.

3. Optical studies of gap formation

The periodic dielectric structure of a photonic crystal gives rise to Bragg diffraction, which is associated with the appearance of frequency gaps in the dispersion relation for certain propagation directions (Fig. 1). The frequency width of the gaps increases with increasing interaction Ψ between light and the crystal. This allows stop gaps associated with different sets of lattice planes to merge into a full photonic band gap for sufficient dielectric contrast. Since stop gaps are the precursors to a photonic band gap, the study of their optical properties is essential. The center frequency of the first order stop gaps, often used to determine the lattice spacing and average refractive index, is well understood [38–40]. The intrinsic width of the stop gaps has been demonstrated to gauge the photonic interaction strength for colloidal crystals [41,42], or depend on the thickness of hollow-shell crystals [43]. In addition, stop gaps strongly affect the propagation of ultrashort optical pulses. An essential aspect in understanding optical experiments, including spontaneous emission from inside photonic crystals, is the scattering of light by defects and irregularities. Here we will review several common techniques to study gaps, and the first efforts to quantify the scattering by disorder.

3.1. Reflectivity

Gaps in the band structures of thick, strongly interacting photonic crystals are conveniently probed by angle-resolved broad band reflectivity. In the first such study, on TiO₂ inverse opals, broad stop

gaps were observed and their angular behavior was found to agree well with plane wave band structure calculations [44]. It was concluded that for frequencies at the top of the fcc L-gap, the propagation of light was limited to $\leq 45\%$ of all available directions. Similar broad reflectivity peaks have been observed by other groups in inverse opals [45,46], and may be monitored in between the fabrication steps of inverted opals [47].

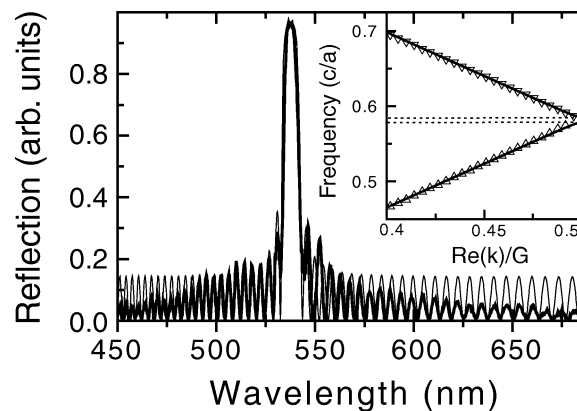
Experiments on inverse opals at frequencies beyond the first stop gap yielded evidence of an intriguing splitting of Bragg peaks, and a concomitant avoided crossing of stop gaps over a wide range of angles and frequencies [48]. Theoretical analysis revealed that the phenomena were caused by interactions and band repulsions between multiple Bloch modes in the crystals that yield coupled modes. Flat bands were observed over a considerable wave vector range, and were identified as the precursor to band gap formation, that is, when bands are dispersionless throughout the whole Brillouin zone. Avoided crossing of Bragg peaks have also been observed in opals by Romanov et al. [49].

At frequencies in the range of the second order Bragg diffraction where the band gap is expected for fcc crystals, peculiar triple reflection peaks have been observed in inverse opals by several groups [15,50]. Angle-resolved experiments revealed that the reflectivity features are dispersionless throughout a range that corresponds to the whole Brillouin zone. Such dispersionless behavior could be interpreted as evidence for a photonic band gap, but a theoretical analysis revealed that this is rather the signal of strong coupling of many Bloch modes, that even occurs in absence of a band gap. Moreover, the triple peak structure was identified with high-order stop gaps [50].

A diffraction peak is theoretically expected to have a flat top with 100% reflectivity, that abruptly falls off at the edges [51]. The width of the top is equal to the width of the stop gap, and the full width at half maximum (FWHM) is only slightly larger. In presence of extinction, diffraction peaks become rounded and asymmetric with maxima less than 100%. The FWHM, however, remains the same, therefore it is a reliable measure of the widths of stop gaps for crystals with extinction [43,44,48]. In most measurements, many crystal domains of a sample are illuminated by coarse white light beams. Reflectivity spectra usually differ from the expected peak shape due to extinction by imperfections such as mosaic spread, grain boundaries, surface roughness, or polydispersity of the unit cell components, which scatter light diffusively. In cases where reflectivity was calibrated [44,48], the maxima of the peaks were considerably less than 100%. Care should be taken to avoid broadening of the diffraction peaks, which may be caused by strain, or finite-sized crystal domains [51].

Recently, square-top peaks with near-unity reflectivity have been established at normal incidence, by using optical microscopy to focus on single-crystal opals [52]. In this way, the photonic width of the peak is best estimated, since structural effects are most effectively removed. For thin samples with parallel front- and back planes, the main reflectivity peak is accompanied by numerous fringes, as shown in Fig. 3. Interestingly, the frequencies of these Fabry–Pérot-like resonances map the dispersion relation, since they

Figure 3. Single-domain reflectivity measurements on opals reveal reflectivities close to unity and Fabry–Pérot fringes resulting from the parallel front and back faces. Figure from reference [52], courtesy of David Norris.



occur at equidistant k -points [52,53]. Single crystal angle-resolved experiments have been done on inverse opals in reference [54], yielding reflectivity up to 95% and a small mosaic spread of $\leq 3^\circ$. The angle-dependent widths were found to agree with earlier coarse-beam experiments, hence the inhomogeneous broadening in the crystals is small.

3.2. Transmission

Transmission measurements have been used successfully to probe stop gaps in weakly photonic crystals [41,55–60] and in strongly interacting thin crystals [13,14,61]. Stop gaps are apparent as decreased transmission over a range of frequencies. In experiments on impressive GaAs layer by layer structures, Noda et al. observed deep stop gaps with up to 40 dB attenuation at normal incidence, and 7 dB over the full range of the expected band gap [14].

An interesting quantity that may be probed in transmission is the Bragg attenuation length for frequencies within a stop gap. This penetration depth of the incident beam can be determined from the transmission as a function of the number of crystal layers, and is predicted to be less than 10 crystal layers for frequencies at the gap center of strongly interacting crystals. Surprisingly, several studies of weakly photonic samples have revealed attenuation lengths in excess of the theoretical value by a factor 2 to 5, although the stop band edges agreed well with calculations [58,60]. While the mechanism is not well understood quantitatively, it is suspected that the Bragg attenuation length depends sensitively on the amount of diffuse scattering, in contrast to the stop gap edges.

Transmission experiments are complementary to reflectivity: in a transmission measurement, the beam passes through the whole sample, including possibly different crystallites which may be differently oriented [41]. In a reflectivity experiments only the first crystal planes near the surface are probed. Differently oriented crystallites do not add to the reflectivity, but do cause significant inhomogeneous broadening of stop gaps in transmission. Furthermore, outside stop gaps, transmission versus frequency usually decreases similar to Rayleigh's law due to scattering by defects [58,62,63]. Consequently, transmission measurements seem well-suited for the study of samples thinner than a mean free path ℓ , described below.

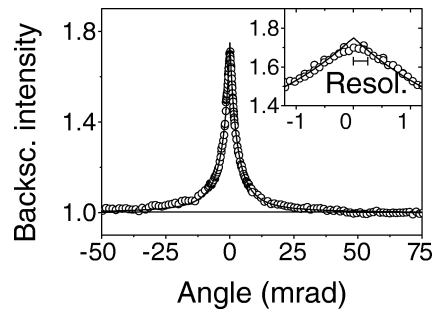
3.3. Time-resolved measurements

Time-resolved experiments are essential to investigate the dynamics of electromagnetic fields in photonic crystals. At the edges of photonic stop bands, the dispersion curves become highly dispersive. As a result, a considerable slowing down and reduction of the group velocity has been observed in opals and colloidal crystals, using ultrashort pulses [64,65]. In a phase-sensitive ultrashort-pulse interferometric experiment, the group velocity dispersion has also been measured. It was observed that this dispersion diverges near stop band edges, with branches of both normal and anomalous dispersion, whereas the dispersion vanished inside the stop band [42,65]. In analogy with electrons, the group velocity dispersion may be interpreted as an 'effective photon mass', that also diverges near gaps [66]. An interesting feature of the group velocity dispersion is that it allows the mapping of photonic gaps *without* any theoretical modeling, in contrast to all other methods we are aware of. Consequently, this technique is expected to be powerful for the identification of photonic band gap, certainly in the case of structures with complex optical behavior [15,16,50] such as the ubiquitous fcc structure.

3.4. Disorder and absorption

Apart from the fundamental interest in the study of Anderson localization [67] of light in disordered photonic band gap materials [7], scattering by defects is relevant for the understanding of light propagation in all real photonic crystals. The length scale which characterizes the degree of disorder is the transport mean free path ℓ , which is the distance over which light propagates before its propagation direction is randomized by scattering. The relevance of ℓ has already been pointed out in the discussion of transmission

Figure 4. The diffuse intensity shows an enhancement due to interference in the backscatter direction, known as the enhanced back scattering cone. For an opal of polystyrene spheres in air with radius $r = 180$ nm the transport mean free path, found from the FWHM of the cone, is $\ell = 15 \pm 2$ μm . The inset shows a closeup of the top of the cone, which is cusped for infinite nonabsorbing samples. The rounding relative to the cusped fit, here limited by the angular resolution, provides an upper bound to the sample thickness and absorption length. From reference [68].



experiments, and it also plays an important role in emission experiments. Regarding applications, the disorder in photonic crystals must be controlled to the extent that ℓ remains larger than both the Bragg attenuation length L_B and the sample thickness L , i.e., $\ell > L > L_B$.

A convenient technique, originally developed in our lab, to study the transport of light in multiple scattering media is enhanced backscattering. Fig. 4 shows that it can be used [68,69] to determine ℓ in photonic crystals and probe the degree of disorder optically. Mean free paths of the order of $\ell = 10$ μm are found, about five times longer than L_B , hence the formation of Bragg diffraction and stop gaps is hardly affected by disorder. We have found that ℓ is limited by polydispersity and mean-squared displacements of $\sim 2\%$ [68], typical for state of the art structures. This result suggests that considerable improvements in crystal fabrication are required to achieve mean free paths long enough to permit extended optical chips in 3D photonic crystals.

Optical absorption is detrimental for photonic band gap effects, which rely on multiple scattering and interference. Because very long light paths are sampled near the top of enhanced backscatter cones (see Fig. 4), i.e., many times the sample thickness, this method is also well suited to investigate absorption. From the sharpness of the cusp at 0° in Fig. 4, we determine an upper bound to the absorption length of at least 0.5 cm, that is $> 10^4 \times \lambda$, or $10^2 \times L$.

4. Spontaneous emission

Since 3D photonic crystals fundamentally modify the DOS, it is natural to study their influence on spontaneous emission. To this end, internal light sources such as excited atoms, quantum dots, fluorescent molecules or thermal radiation sources are placed inside photonic crystals. In essence, such a light source will experience two effects: (i) a change of the local density of states at its spatial position [70] that results in a change of the radiative lifetime in a dynamic experiment or a change of total emitted power in a cw experiment, (ii) an angular redistribution of intensity due to stop gaps for propagation in certain directions.

The first effect has yet to be clearly observed. Several time resolved emission experiments in search of lifetime changes in colloidal photonic crystals showed no modified emission rate due to insufficient dielectric contrast of the crystals [71–74]. The study of a modified radiative rate necessarily involves a suitable reference system, which provides the same chemical surroundings for the light source as well as a quadratic photonic density of states. It has recently been shown that even in the absence of a band gap, strongly interacting photonic crystals dramatically affect the total emitted power with large spatial variations of the local density of states [75].

The second effect is commonly observed: an attenuation band appears when a stop gap overlaps the emission spectrum [57,76–82]. These stop gaps may be extracted by comparing emission spectra for different crystal orientations. As an example, emission spectra of a laser dye in a TiO_2 opal are shown in Fig. 5A for various angles relative to the (111) reciprocal lattice vector. The attenuation band shifts with angle from 14800 cm^{-1} to higher frequencies, in qualitative agreement with Bragg's law. For 60° the attenuation band has shifted out of the emission spectrum. We extract the stop gap from the spectra

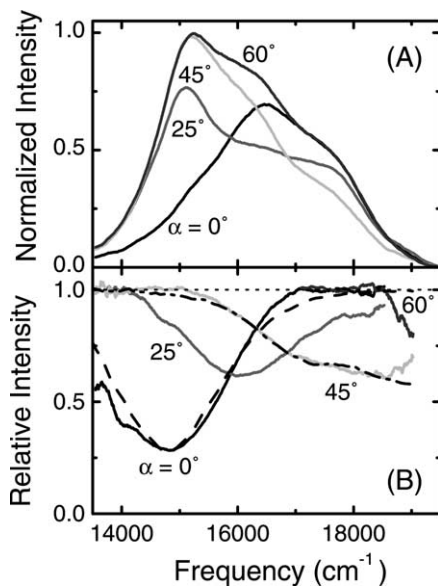


Figure 5. Emission from the laser dye Nile Blue in titania inverse opals is strongly redirected, as demonstrated by the spectra in (A) as a function of angle α relative to the (111) reciprocal lattice vector. An attenuation band sweeps to higher frequencies with angle, according to the band structure. Dashed curves in (B) correspond to inverted reflectivity spectra to illustrate the correspondence of attenuation bands with measured stop gaps in reflectivity. From reference [82].

by normalizing the emission to this 60° spectrum, as shown in Fig. 5B. The attenuation bands appear at the same frequencies and show similar behavior as a function of angle as reflectivity peaks [82]. For large angles α relative to the (111) vector, there appears to be a transition to a double stop band. The stop band edges exhibit an avoided crossing as a function of α , which is the result of the (200) Bloch modes mixing with the (000) and (111) modes, similar to what was discussed above for reflectivity measurements [48].

It is striking that the attenuation of spectra in stop band regions (see Fig. 5B) is far less than the attenuation in typical transmission experiments [80,82]. Scattering of fluorescence by disorder plays a crucial role in determining the measured luminescent spectra, hence the crystal does not simply act as a filter. Light from internal sources can propagate in all but the stop gap directions, and percolates through the whole crystal by multiple scattering. Light may exit the crystal along a stop gap direction if it is scattered toward the detector at a distance within L_B from the crystal surface. Consequently, the degree of attenuation along the direction of a stop gap is limited to about $(1 - L_B/\ell)$ or 80%. In contrast, an external beam is usually appreciably attenuated in transmission, since both diffuse scattering and Bragg diffraction along the full sample thickness remove intensity from the incident beam.

5. Open issues

Theoretically, a full band gap, that is, zero DOS in some frequency window, is only defined for infinitely extended, perfect structures. An important question is: which experimental signatures allow (reasonable) proof of a photonic band gap in a carefully crafted structure? Several teams have observed overlapping stop gaps over a wide range of propagation directions [13,14,61]. Omnidirectional stop gaps, although a consequence of a photonic band gap, do not necessarily imply the absence of all propagating modes, since modes may exist that do not couple to the outside world [83]. It has been proposed that a photonic band gap can be identified from cw emission spectra of internal sources [80], spontaneous emission dynamics of such sources, or a measurement of group velocity dispersion for many directions [42].

It is of prime importance to limit the structural disorder such as size randomness, positional disorder and roughness of the photonic crystal building blocks. Recent calculations have addressed the effect of size randomness and positional disorder on the photonic DOS [28], but we are unaware of any experimental investigations to date. The gap in the density of states which ideally occurs for fcc inverse opals is destroyed

by positional disorder of $\sim 2\%$ of the lattice parameter, even for very high index contrasts. In contrast, the first-order pseudogap is very robust. Therefore, the band gap in diamond structures are quite robust.

Many of the exciting phenomena predicted to occur in a photonic band gap material rely on the existence of suitable luminescent sources with a high quantum efficiency. The requirement of a high refractive index severely constrains the choice of material for a photonic band gap structure. In turn, this limits the number of alternative light sources that are compatible with the photonic crystal backbone. For instance, in the case of luminescent dyes, it is known that sensitization can occur, which must be prevented to allow a high efficiency [84]. For each of the different materials and fabrication mechanisms, the design, position control and pumping strategy remain unexplored territory to date.

We would like to point out that experiments designed to probe photonic crystals at frequencies near the photonic band gap will be difficult to interpret due to the complex dispersion of photonic crystals, especially at higher order where the gap is expected for fcc structures. Even the incoupling, dispersion and propagation direction of pump and control beams in optical experiments will be nontrivial to control. As most reflectivity and transmission experiments to date have been limited to low frequencies, the experimental study of beam propagation in photonic crystals at high frequencies is urgently called for. A reward of such studies could be the design of 3D beam-steering devices, such as superprisms.

In future experiments, near field optical microscopy techniques may provide novel insight in the mode distributions and local density of states near the interface of photonic crystals. An exciting new experimental challenge lies in the tuning and switching of photonic crystals. Impressive first experiments have already been performed using liquid crystals [85–87], while new strategies have been proposed for ultrafast switching of photonic band gaps [88,89]. If a photonic band gap is achieved, the next challenge is to engineer line and point defects, which allow efficient waveguiding and high- Q cavities.

Acknowledgements. We thank A. Lagendijk and all members of the group Waves in Complex Media for continuous encouragements and support. This work is part of the research program of the “Stichting voor Fundamenteel Onderzoek der Materie (FOM)”, which is supported by the “Nederlandse Organisatie voor Wetenschappelijk Onderzoek” (NWO).

References

- [1] J.D. Joannopoulos, R.D. Meade, J.N. Winn, Photonic Crystals; Molding the Flow of Light, Princeton University Press, Princeton, NJ, 1995.
- [2] C.M. Soukoulis (Ed.), Photonic Crystals and Light Localization in the 21st Century, Kluwer, Dordrecht, 2001.
- [3] A. Lagendijk, B.A. van Tiggelen, Phys. Rep. 270 (1996) 143–216.
- [4] E. Yablonovitch, Phys. Rev. Lett. 58 (1987) 2059–2062.
- [5] Y. Yang, S.Y. Zhu, Phys. Rev. A 62 (2000) 013805.
- [6] Z.Y. Li, L.L. Lin, Z.Q. Zhang, Phys. Rev. Lett. 84 (2000) 4341–4344.
- [7] S. John, Phys. Rev. Lett. 58 (1987) 2486–2489.
- [8] E. Yablonovitch, T.J. Gmitter, R.D. Meade, A.M. Rappe, K.D. Brommer, J.D. Joannopoulos, Phys. Rev. Lett. 67 (1991) 3380–3383.
- [9] E. Paspalakis, N.J. Kylstra, P.L. Knight, Phys. Rev. A 60 (1999) R33–R36.
- [10] K. Busch, N. Vats, S. John, B.C. Sanders, Phys. Rev. E 58 (1998) 4168–4175.
- [11] G.M. Nikolopoulos, P. Lambropoulos, Phys. Rev. A 61 (2000) 053812.
- [12] E. Yablonovitch, T.J. Gmitter, K.M. Leung, Phys. Rev. Lett. 67 (1991) 2295–2298.
- [13] S.Y. Lin, J.G. Fleming, D.L. Hetherington, B.K. Smith, R. Biswas, K.M. Ho, M.M. Sigalas, W. Zubrzycki, S.R. Kurtz, J. Bur, Nature 394 (1998) 251–253.
- [14] S. Noda, K. Tomoda, N. Yamamoto, A. Chutinan, Science 289 (2000) 604–606.
- [15] A. Blanco, E. Chomski, S. Grachtak, M. Ibsate, S. John, S.W. Leonard, C. López, F. Meseguer, H. Míguez, J.P. Mondia, G.A. Ozin, O. Toader, H.M. van Driel, Nature 405 (2000) 437–440.
- [16] Yu.A. Vlasov, X.Z. Bo, J.C. Sturm, D.J. Norris, Nature 414 (2001) 289–293.
- [17] A. Chutinan, S. Noda, Jpn. J. Appl. Phys. 39 (2000) 2353–2356.
- [18] M. Notomi, T. Tamamura, T. Kawashima, S. Kawakami, Appl. Phys. Lett. 77 (2000) 4256–4258.
- [19] J.V. Sanders, Nature 204 (1964) 1151–1153.
- [20] A. Imhof, D.J. Pine, Nature 389 (1997) 948–951.
- [21] B.T. Holland, C.F. Blanford, A. Stein, Science 281 (1998) 538–540.

- [22] J.E.G.J. Wijnhoven, W.L. Vos, *Science* 281 (1998) 802–804.
- [23] A.A. Zakhidov, R.H. Baughman, Z. Iqbal, C. Cui, I. Khayrullin, S.O. Dantas, J. Marti, V.G. Ralchenko, *Science* 282 (1998) 897–901.
- [24] O.D. Velev, E. Kaler, *Adv. Mater.* 12 (2000) 531–534.
- [25] Y. Xia, B. Gates, Y. Yin, Y. Lu, *Adv. Mater.* 12 (2000) 693–713.
- [26] D.J. Norris, Yu.A. Vlasov, *Adv. Mater.* 13 (2001) 371–376.
- [27] J.E.G.J. Wijnhoven, L. Bechger, W.L. Vos, *Chem. Mater.* 13 (2001) 4486–4499.
- [28] Z.Y. Li, Z.Q. Zhang, *Phys. Rev. B* 63 (2001) 125106; *Phys. Rev. B* 62 (2000) 1516–1519.
- [29] A. van Blaaderen, R. Ruel, P. Wiltzius, *Nature* 385 (1997) 321–324.
- [30] P. Jiang, J.F. Bertone, K.S. Hwang, V.L. Colvin, *Chem. Mater.* 11 (1999) 2132–2140.
- [31] M. Campbell, D.N. Sharp, M.T. Harrison, R.G. Denning, A.J. Turberfield, *Nature* 404 (2000) 53–56.
- [32] C. Cuisin, A. Chelnokov, J.-M. Lourtioz, D. Decanini, Y. Chen, *Appl. Phys. Lett.* 77 (2000) 770–772.
- [33] A. Chelnokov, K. Wang, S. Rowson, P. Garoche, J.-M. Lourtioz, *Appl. Phys. Lett.* 77 (2000) 2943–2945.
- [34] J. Schilling, F. Muller, S. Matthias, R.B. Wehrspohn, U. Gösele, K. Busch, *Appl. Phys. Lett.* 78 (2001) 1180–1182.
- [35] W.L. Vos, M. Megens, C.M. van Kats, P. Bösecke, *Langmuir* 13 (1997) 6004–6008.
- [36] M. Megens, C.M. van Kats, P. Bösecke, W.L. Vos, *Langmuir* 13 (1997) 6120–6129.
- [37] M. Megens, W.L. Vos, *Phys. Rev. Lett.* 86 (2001) 4855–4858.
- [38] W.L. Vos, R. Sprik, A. van Blaaderen, A. Imhof, A. Lagendijk, G.H. Wegdam, *Phys. Rev. B* 53 (1996) 16231–16235.
- [39] H. Míguez, A. Blanco, F. Meseguer, C. López, H.M. Yates, M.E. Pemble, V. Fornés, A. Mifsud, *Phys. Rev. B* 59 (1999) 1563–1566.
- [40] V.N. Astratov, A.M. Adawi, M.S. Skolnick, V.K. Tikhorimov, V. Lyubin, D.G. Lidzey, M. Ariu, A.L. Reynolds, *Appl. Phys. Lett.* 78 (2001) 4094–4096.
- [41] W.L. Vos, M. Megens, C.M. van Kats, P. Bösecke, *J. Phys.: Condens. Matter* 8 (1996) 9503.
- [42] W.L. Vos, H.M. van Driel, M. Megens, A.F. Koenderink, A. Imhof, in: C.M. Soukoulis (Ed.), *Photonic Crystals and Light Localization in the 21st Century*, Kluwer, Dordrecht, 2001, pp. 191–208.
- [43] R. Rengarajan, P. Jiang, V. Colvin, D. Mittleman, *Appl. Phys. Lett.* 77 (2000) 3517–3519.
- [44] M.S. Thijssen, R. Sprik, J.E.G.J. Wijnhoven, M. Megens, T. Narayanan, A. Lagendijk, W.L. Vos, *Phys. Rev. Lett.* 83 (1999) 2730–2733.
- [45] A. Richel, N.P. Johnson, D.W. McComb, *Appl. Phys. Lett.* 76 (2000) 1816–1818; Addenda: *Appl. Phys. Lett.* 77 (2000) 1062–1063.
- [46] G. Subramania, R. Biswas, K. Constant, M.M. Sigalas, K.M. Ho, *Phys. Rev. B* 63 (2001) 235111.
- [47] A. Blanco, H. Míguez, F. Meseguer, C. López, *Appl. Phys. Lett.* 78 (2001) 3181–3183.
- [48] H.M. van Driel, W.L. Vos, *Phys. Rev. B* 62 (2000) 9872–9875.
- [49] S.G. Romanov, T. Maka, C.M. Sotomayor Torres, M. Müller, R. Zentel, D. Cassagne, J. Manzanares-Martinez, C. Jouanin, *Phys. Rev. E* 63 (2001) 056603.
- [50] W.L. Vos, H.M. van Driel, *Phys. Lett. A* 272 (2000) 101–106.
- [51] R.W. James, *The Optical Principles of the Diffraction of X-rays*, Bell, London, 1962.
- [52] Yu.A. Vlasov, M. Deutsch, D.J. Norris, *Appl. Phys. Lett.* 76 (2000) 1627–1629.
- [53] M. Born, E. Wolf, *Principles of Optics*, Pergamon, Oxford, 1954.
- [54] J.F. Galisteo Lopez, W.L. Vos, *Phys. Rev. B*, submitted.
- [55] Í.Í. Tarhan, G.H. Watson, *Phys. Rev. Lett.* 76 (1996) 315–318.
- [56] R.D. Pradhan, Í.Í. Tarhan, G.H. Watson, *Phys. Rev. B* 54 (1996) 13721–13726.
- [57] V.N. Bogomolov, S.V. Gaponenko, I.N. Germanenko, A.M. Kapitonov, E.P. Petrov, N.V. Gaponenko, A.V. Prokofiev, A.N. Ponyavina, N.I. Silvanovich, S.M. Smolovich, *Phys. Rev. E* 55 (1997) 7619–7625.
- [58] Yu.A. Vlasov, V.N. Astratov, O.Z. Karimov, A.A. Kaplyanskii, V.N. Bogomolov, A.V. Prokofiev, *Phys. Rev. B* 55 (1997) R13357–13360.
- [59] V.N. Astratov, Yu.A. Vlasov, O.Z. Karimov, A.A. Kaplyanskii, Yu.G. Musikhin, N.A. Bert, V.N. Bogomolov, A.V. Prokofiev, *Phys. Lett. A* 222 (1996) 349–353.
- [60] J.F. Bertone, P. Jiang, K.S. Hwang, D.M. Mittleman, V.L. Colvin, *Phys. Rev. Lett.* 83 (1999) 300–303.
- [61] J.G. Fleming, S.Y. Lin, *Opt. Lett.* 24 (1999) 49–51.
- [62] Yu.A. Vlasov, V.N. Astratov, A.V. Baryshev, A.A. Kaplyanskii, O.Z. Karimov, M.F. Limonov, *Phys. Rev. E* 61 (2000) 5784–5793.
- [63] Yu.A. Vlasov, M.A. Kaliteevski, V.V. Nikolaev, *Phys. Rev. B* 60 (1999) 1555–1562.
- [64] Yu.A. Vlasov, S. Petit, G. Klein, B. Hönerlage, Ch. Hirlimann, *Phys. Rev. E* 60 (1999) 1030–1035.
- [65] A. Imhof, W.L. Vos, R. Sprik, A. Lagendijk, *Phys. Rev. Lett.* 83 (1999) 2942–2945.
- [66] N.W. Ashcroft, N.D. Mermin, *Solid State Physics*, Holt, Rinehart and Winston, New York, 1976.
- [67] P.W. Anderson, *Phys. Rev.* 109 (1958) 1492–1505.

- [68] A.F. Koenderink, M. Megens, G. van Soest, W.L. Vos, A. Lagendijk, Phys. Lett. A 268 (2000) 104–111.
- [69] J. Huang, N. Eradat, M.E. Raikh, Z.V. Vardeny, A.A. Zakhidov, R.H. Baughman, Phys. Rev. Lett. 86 (2001) 4815–4818.
- [70] R. Sprik, B.A. van Tiggelen, A. Lagendijk, Europhys. Lett. 35 (1996) 265–270.
- [71] J. Martorell, N.M. Lawandy, Phys. Rev. Lett. 65 (1990) 1877–1880.
- [72] E.P. Petrov, V.N. Bogomolov, I.I. Kalosha, S.V. Gaponenko, Phys. Rev. Lett. 81 (1998) 77–80.
- [73] M. Megens, J.E.G.J. Wijnhoven, A. Lagendijk, W.L. Vos, Phys. Rev. A 59 (1999) 4727–4731.
- [74] M. Megens, H.P. Schriemer, A. Lagendijk, W.L. Vos, Phys. Rev. Lett. 83 (1999) 5401;
E.P. Petrov, V.N. Bogomolov, I.I. Kalosha, S.V. Gaponenko, Phys. Rev. Lett. 83 (1999) 5402.
- [75] A.F. Koenderink, L. Bechger, H.P. Schriemer, A. Lagendijk, W.L. Vos, Phys. Rev. Lett., submitted.
- [76] S.G. Romanov, A.V. Fokin, V.I. Alperovich, N.P. Johnson, R.M. De La Rue, Phys. Status Solidi A 164 (1997) 169–172.
- [77] T. Yamasaki, T. Tsutsui, Appl. Phys. Lett. 72 (1998) 1957–1959.
- [78] K. Yoshino, S.B. Lee, S. Tatsuhara, Y. Kawagishi, M. Ozaki, Appl. Phys. Lett. 73 (1998) 3506–3508.
- [79] A. Blanco, C. Lopez, R. Mayoral, H. Miguez, F. Meseguer, A. Mifsud, J. Herrero, Appl. Phys. Lett. 73 (1998) 1781–1783.
- [80] M. Megens, J.E.G.J. Wijnhoven, A. Lagendijk, W.L. Vos, J. Opt. Soc. Am. B 16 (1999) 1403–1408.
- [81] S. Lin, J.G. Fleming, E. Chow, J. Bur, K.K. Choi, A. Goldberg, Phys. Rev. B 62 (2000) 2243–2246.
- [82] H.P. Schriemer, H.M. van Driel, A.F. Koenderink, W.L. Vos, Phys. Rev. A 63 (2001) R011801.
- [83] Y. Fink, J.N. Winn, S. Fan, C. Chen, J. Michel, J.D. Joannopoulos, E.L. Thomas, Science 282 (1998) 1679–1682.
- [84] L. Bechger, A.F. Koenderink, W.L. Vos, Langmuir, in press.
- [85] K. Yoshino, Y. Shimoda, Y. Kawagishi, K. Nakayama, M. Ozaki, Appl. Phys. Lett. 75 (1999) 932–934.
- [86] S.W. Leonard, J.P. Mondia, H.M. van Driel, O. Toader, S. John, K. Busch, A. Birner, U. Gösele, V. Lehmann, Phys. Rev. B 61 (1999) R2389–R2392.
- [87] D. Kang, J.E. MacLennan, N.A. Clark, A.A. Zakhidov, R.H. Baughman, Phys. Rev. Lett. 86 (2001) 4052–4055.
- [88] S.W. Leonard, H.M. van Driel, A. Birner, U. Gösele, in preparation.
- [89] P.M. Johnson, A.F. Koenderink, W.L. Vos, Phys. Rev. Lett., submitted.