

# Photonic Crystal Formed by the Imaginary Part of the Refractive Index

By Juntao Li, Bing Liang, Yikun Liu, Peiqing Zhang, Jianying Zhou,\*  
Sergey O. Klimonsky, Alexander S. Slesarev, Yuri D. Tretyakov,  
Liam O'Faolain, and Thomas F. Krauss

Photonic crystals (PhCs)<sup>[1]</sup> are widely studied photonic structures that provide unprecedented control over the propagation of light. The large majority of PhCs are formed by a periodic modulation of the refractive index, i.e., a modulation of the real part of the dielectric constant. Additional functionality can be created by including absorbing features into the structure, thus creating PhCs out of materials with complex dielectric index. This is particularly interesting when the photonic resonance created by the refractive index contrast overlaps with the absorption feature, hence creating a “resonantly absorbing PhC”. An example of such a resonantly absorbing structure is that of a semiconductor saturable absorber mirror (SESAM)<sup>[2]</sup> used in mode-locked lasers where quantum wells are incorporated into a multilayer Bragg stack. Similar structures referred to as Resonantly Absorbing Bragg Reflectors (RABR)<sup>[3]</sup> have been used to demonstrate optical switching,<sup>[4]</sup> optical storage,<sup>[5]</sup> and nonlinear optical conversion.<sup>[6]</sup>

The examples above are based on Bragg mirrors or 1D photonic crystals. An extension to 2D and 3D structures has been proposed and demonstrated by backfilling the voids of a conventional photonic crystal with resonantly absorbing materials such as quantum dots<sup>[7]</sup> and metal.<sup>[8]</sup>

Most of these structures are based on refractive index modulation with absorption providing additional features. In contrast, the structure we propose and demonstrate here, an extension of the 1D case approaches of Prineas et al.<sup>[4]</sup> and Kozhekin et al.,<sup>[5]</sup> is formed exclusively by the absorbing feature, hence it is a true “imaginary refractive index” structure. A refractive index contrast naturally exists near the absorption feature, as required by the Kramers–Kronig relationship, but away from this feature, the refractive index contrast is practically zero.

In order to demonstrate this effect experimentally, we created a template using holographic lithography<sup>[9]</sup> with a diffractive

optical element (DOE) (Fig. 1), which generates a 2D photonic lattice of SU-8 polymer disks (Fig. 2a). The disks are doped with a high concentration of the organic dye Rhodamine B (RhB) that has an absorption peak around 564 nm, so the absorption of the lattice is strongly dependent on wavelength. Subsequent filling of the voids with the same SU-8 polymer, but without dye doping, gives rise to an imaginary index photonic lattice (Fig. 2b). The diffraction pattern of the structure before back-filling is given by the dielectric modulation that exists for all wavelengths as in a conventional photonic lattice and is much like a rainbow (Fig. 3a). By contrast, only yellow/green light diffraction around 564 nm can be observed once the structure has been back-filled (Fig. 3b–d). The wavelength dependent diffraction clearly shows that the structure only acts as a PhC in the vicinity of the absorption window. Out of this window, the structure behaves as a uniform polymer layer. This new type of photonic crystal offers intriguing properties for further study in the field of saturable light absorption and emission control. The PhC may also be applied for optical switching and optical logic operation.

The imaginary index structure can be treated as a 2D grating with wavelength-dependent phase and intensity modulation. According to diffraction theory,<sup>[10]</sup> the intensity distribution  $I$  created by the grating is given by the Fourier transform of the transmission function of the grating  $t$ :

$$I(f_x, f_y, \lambda) \propto |FT(t(x, y, \lambda))|^2 \quad (1)$$

Where  $x$  and  $y$  are the coordinates of the grating,  $f_x$  and  $f_y$  are the Fourier transforms corresponding to  $x$  and  $y$ , which are related to the diffraction angle at the wavelength  $\lambda$ .

We determine the structural parameters from the SEM image shown in Figure 2a and calculate the corresponding transmission function  $t$ :

$$t(x, y, \lambda) = \begin{cases} \exp(i2\pi dn_b/\lambda) & (\text{air or SU8 without dye}) \\ \sqrt{T(\lambda)} \exp(i2\pi dn_r/\lambda) & (\text{SU8 with dye}) \end{cases} \quad (2)$$

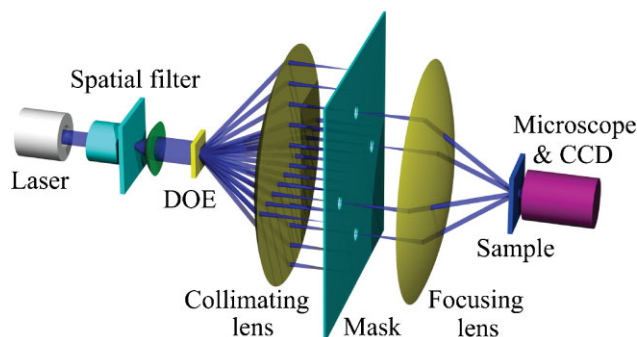
In Equation 2,  $n_b$  is equal to 1 for a normal template and  $n_0 = 1.62$  for the resonantly absorbing PhC, which corresponds to the refractive index of air and undoped SU8, respectively;  $d = 2.6 \mu\text{m}$  is the thickness of the sample; the transmission  $T$  of the RhB doped SU8, which is modified by the absorption of RhB, was measured by spectrometer (Fig. 4a). Based on Figure 2a, the lattice constant and the normalized rod size of the sample are set to be  $a = 2.86 \mu\text{m}$  and  $r/a = 0.22$ , respectively. The good

[\*] Prof. J. Y. Zhou, Dr. J. T. Li, B. Liang, Y. K. Liu, P. Q. Zhang  
State Key Laboratory of Optoelectronic Materials and Technologies  
Sun Yat-sen University  
Guangzhou, 510275 (China)  
E-mail: stszjy@mail.sysu.edu.cn

Prof. Yu. D. Tretyakov, Prof. S. O. Klimonsky, A. S. Slesarev  
Department of Materials Science  
M. V. Lomonosov Moscow State University  
Lenin Hills, Moscow, 119991 (Russia)

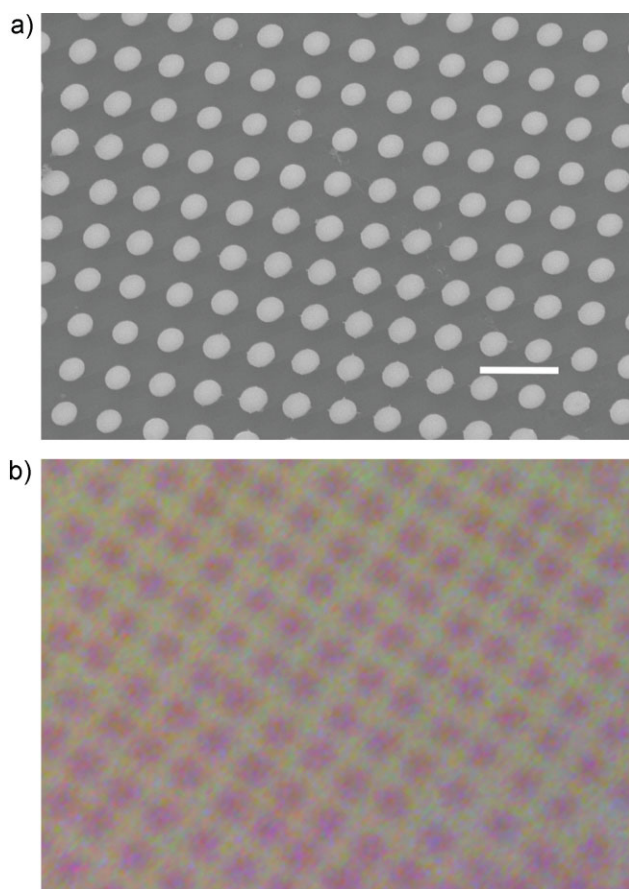
Prof. T. F. Krauss, Dr. L. O'Faolain  
School of Physics and Astronomy  
University of St Andrews  
St Andrews, Fife, KY169SS (UK)

DOI: 10.1002/adma.200903938

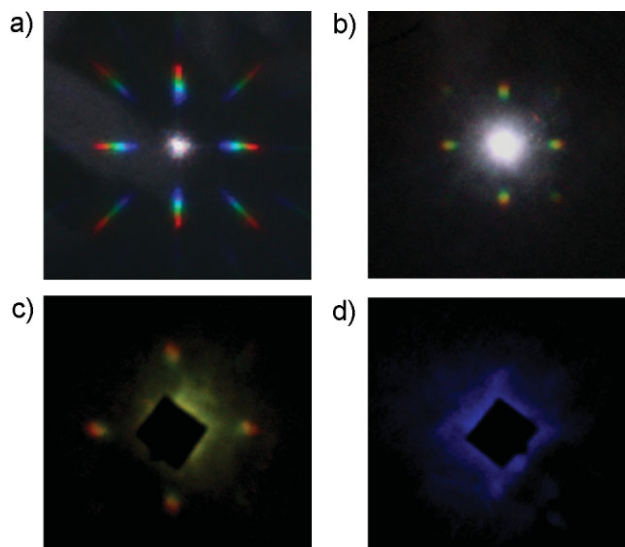


**Figure 1.** Experimental setup for the creation of the 2D PhC sample. The sample was exposed by 4-beam interference lithography, using a DOE, a mask and suitable lenses.

agreement between the measured and the calculated diffraction efficiency shown in Figure 4b is strong evidence that there is no refractive index contrast for the wavelength away from the absorption region. Hence  $n_r = n_o + \Delta n$ , with  $\Delta n$  being the refractive index change near the absorption center of the dye (Fig. 4a), which is calculated using the Kramers–Kronig relationship based on the absorption of the RhB.<sup>[11]</sup> Figure 4b



**Figure 2.** a) SEM image of the 2D square lattice PhC created by holographic lithography. Scale bars: 5  $\mu\text{m}$ . b) Microscopic image of the same structure after back-filling to create the imaginary index PhC. The fact that the lattice appears as red spots indicates the wavelength-dependence of the periodic corrugation.



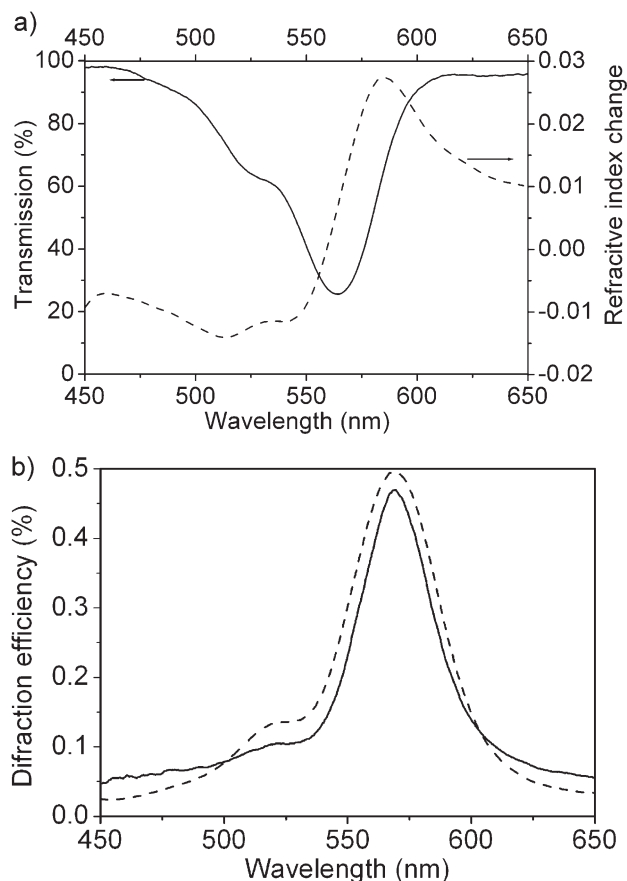
**Figure 3.** Diffraction pattern taken with a Xenon lamp showing: a) the lattice before and b) after backfilling with SU-8. It is clear that diffraction in a, occurs for all wavelengths, while in b, the wavelength range is rather limited, centering on green/yellow. The wavelength-dependence of the diffraction of the structure after back-filling is further highlighted in c and d using Xenon lamp with color filters. Diffraction of green light is shown in c and the absence of diffraction for blue light is shown in d. It is clear from all these observations that mainly the green light, near the absorption peak of the RhB, produces a diffraction pattern.

shows the numerical simulation of the diffraction efficiency of the structure by substituting Equation 2 into Equation 1. The corresponding experimental curve was also measured and is shown in the same figure, which shows a striking agreement with the numerical simulation, especially when considering experimental imperfections.

As a final check to confirm that our structure operates on the imaginary part of the refractive index rather than on absorption-related thermally-induced refractive index change, we measured the response time of the diffraction-pattern with a picosecond laser of 25 ps pulse length at 564 nm wavelength. A photomultiplier and oscilloscope were used to measure the zero order and the first-order diffraction signals with a time resolution of 10 ns. No time delay was observed between the two signals, so we can indeed confirm that the effect is not thermal but caused by an instantaneous refractive effect.

There are many applications for an imaginary index photonic crystal. For example, by exploiting saturable absorption or the ultrafast ac Stark effect<sup>[3]</sup> to shift the absorption peak of the material, an efficient all-optical switch can be realised. The structure also has the potential to generate a large family of solitons, i.e., distributed solitons<sup>[12]</sup> and slow light gap solitons.<sup>[5]</sup> Furthermore, the fact that the structure allows a convenient measurement of the complex refractive index and its wavelength dependence is a useful tool for the characterisation of novel optical materials, such as polymers loaded with colloidal quantum dots.

In conclusion, we have proposed a novel class of photonic crystals that are based solely on the imaginary part of the refractive index. The structures have been characterized by the wavelength dependent diffraction near the absorption center of the



**Figure 4.** a) Transmission (solid line) of SU8 with RhB and the corresponding refractive index change (dashed line) near the absorption center of the RhB, which is calculated via the Kramers–Kronig relationship based on the absorption of the RhB. b) Numerical stimulation (dashed line) and experimental result (solid line) of the diffraction efficiency of the imaginary index PhC.

incorporated dye, which is found to be in good agreement with the simulation. We believe that this structure can also be extended to 3D and applied in many areas of Photonics, such as optical switching, coherent and incoherent PhC structure manipulation, refractive index measurement as well as soliton generation.

## Experimental

**Experimental Setup:** The experimental setup for the fabrication of samples is shown in Figure 1. The output beam from a continuous-wave Ar<sup>+</sup> laser with 488 nm wavelength was spatially filtered to improve the quality of the beam. The beam was then collimated and passed through a DOE, which produced a dot cross pattern with 21 beams of nearly equal intensity. The multiple beamlets were collimated and passed through a mask in order to select 4 symmetry beamlets to produce the 2D square lattice PhC. The 4 parallel beams generate an intensity pattern for holographic lithography, which is recorded with photosensitive SU8 polymeric material. Two achromatic doublets lenses with 50.8 mm diameter and 75 mm focal length were used as the collimating and focusing lenses, which can produce a 2D square lattice PhC with a lattice constant of 2.86 μm. A microscope and a CCD detector were used to monitor the intensity distribution, thereby examining the formation of a

hologram. This experimental setup has already been applied to fabricate large area and high quality PhCs [13].

The transmission of the SU8 doped with RhB was measured with a Xenon lamp and a fiber optic spectrometer (Ocean Optic USB2000 +). The Xenon lamp was passed through a pinhole, then collimated and focused by lenses with 63 mm focal length. The diameter and aperture angle of the focal spot were 80 μm and 1°, respectively. For the diffraction measurement, the same Xenon lamp was used. Two filters with 60 nm bandwidth were placed behind the lamp to observe the diffraction of green and blue light in Figure 3c and d, respectively. The central wavelength of the filters is 570 and 435 nm. The different order diffraction signals for the light fields at different frequencies were collected by an aspheric condenser lens with a large numerical aperture.

**DOE Preparation:** The DOE was fabricated on a glass substrate coated with ZEP520A electron beam resist with thickness of 400 nm thickness. The sample was coated with a conductive polymer, Mitsubishi RAYON AquaSAVE, to dissipate charge during the exposure. The pattern, which was designed using the annealing algorithm (SA) method [14] to generate the dot cross diffraction pattern, was exposed using a hybrid ZEISS GEMINI 1530/RAITH ELPHY electron beam writer and developed using Xylene. The DOE used a pixel size of 180 nm, while the size of the sample is 600 μm × 600 μm. The fabrication was carried out in the framework of the ePIXnet Nanostructuring Platform for Photonic Integration [15].

**Sample Preparation:** The photoresist used to record the periodic structure was coated at 7000 rpm on glass substrates. It contained the resin Epon-SU8 (from Shell) dissolved in γ-butyrolactone (from Aldrich) with 2 wt. % Benzenecyclopentadienyliron (II) hexafluorophosphate (from Sigma-Aldrich) acting as cationic photoinitiator and 1.5 wt% RhB dye. Soft-baking of the SU8 and the evaporation of solvent were both conducted at 95°. The 2.6 μm thick film was exposed with the holographic light pattern described above. To fabricate the PhC template by a 4-beam interference holographic lithography, the intensity of each beam was 0.3 mW and the exposure time was 50 s. For reference, a uniform thin film was also prepared by exposure for 3 min with a single beam. After exposure, another post bake at 95 °C was performed to complete the cross-linking of the photoresist. The unlinked regions were developed in propyleneglycol-methylether acetate and then rinsed with acetone. After drying at room temperature, the sample was finished.

Back-filling the sample with SU8 without dye was performed in a vacuum environment and the polymer drop-cast onto on the template; the interstitial voids are then simply filled by the effect of air pressure. The sample was then spun at 2000 rpm to remove the extra SU8, followed by solvent evaporation and a flood-exposure in order to ensure that the SU8 with and without dye-doping experienced the same process.

## Acknowledgements

J.T.L. and B.L. contributed equally to this work. This work is supported by the National Key Basic Research Special Foundation (G2010CB923204), Chinese National Natural Science Foundation (10934011, 10774193), the Russian Foundation for Basic Research (07-03-92113 and 08-03-00938), the Program for Fundamental Research of Russian Academy of Sciences and the Federal Target Science and Engineering Program (02.740.11.0135). T.F.K. and L.O'F. is supported by the UK Silicon Photonics EPSRC grant.

Received: November 18, 2009

Published online:

- [1] a) E. Yablonoitch, *Phys. Rev. Lett.* **1987**, *58*, 2059. b) S. John, *Phys. Rev. Lett.* **1987**, *58*, 2486.
- [2] U. Keller, D. A. B. Miller, G. D. Boyd, T. H. Chiu, J. F. Ferguson, M. T. Asom, *Opt. Lett.* **1992**, *17*, 505.
- [3] A. Kozhokin, G. Kurizki, *Phys. Rev. Lett.* **1998**, *81*, 3647.

- [4] J. P. Prineas, J. Y. Zhou, J. Kuhl, H. M. Gibbs, G. Khitrova, S. W. Koch, A. Knorr, *Appl. Phys. Lett.* **2002**, *81*, 4332.
- [5] a) J. Y. Zhou, H. G. Shao, J. Zhao, K. S. Wong, *Opt. Lett.* **2005**, *30*, 1560.  
b) I. V. Mel'nikov, J. S. Aitchison, *Appl. Phys. Lett.* **2005**, *87*, 201111.  
c) R. Khomeriki, J. Leon, *Phys. Rev. Lett.* **2007**, *99*, 183601.
- [6] J. T. Li, J. Y. Zhou, *Opt. Express* **2006**, *14*, 2811.
- [7] a) A. Kaso, S. John, *Phys. Rev. E* **2006**, *74*, 046611. b) P. D. García, Á. Blanco, A. Shavel, N. Gaponik, A. Eychmüller, B. Rodríguez-González, L. M. Liz-Marzán, C. López, *Adv. Mater.* **2006**, *18*, 2768.
- [8] W. Y. Zhang, X. Y. Lei, Z. L. Wang, D. G. Zheng, W. Y. Tam, C. T. Chan, P. Sheng, *Phys. Rev. Lett.* **2000**, *84*, 2853.
- [9] a) M. Campbell, D. N. Sharp, M. T. Harrison, R. G. Denning, A. J. Turberfield, *Nature* **2000**, *404*, 53. b) L. Wu, Y. Zhong, C. T. Chen, K. S. Wong, G. P. Wang, *Appl. Phys. Lett.* **2005**, *86*, 241102. c) Y. V. Miklyaev, D. C. Meisel, A. Blanco, G. von Freymann, K. Busch, W. Koch, C. Enkrich, M. Deubel, M. Wegener, *Appl. Phys. Lett.* **2003**, *82*, 1284.
- [10] M. C. Huley, in: *Diffraction Gratings*, Academic Press Inc, London, UK **1982**.
- [11] M. Born, E. Wolf, in: *Principle of Optics*, Cambridge University Press, UK **1999**.
- [12] Y. Y. Li, W. Pang, Y. Z. Chen, Z. Q. Yu, J. Y. Zhou, H. R. Zhang, *Phys. Rev. A* **2009**, *80*, 043824.
- [13] T. Kondo, S. Juodkazis, V. Mizeikis, S. Matsuo, H. Misawa, *New J. Phys.* **2006**, *8*, 250.
- [14] G. S. Mageras, R. Mohan, *Med. Phys.* **1993**, *20*, 639.
- [15] See <http://www.nanophotonics.eu>, (last accessed April 2010).