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### ▶ To cite this version:

Vincent Reboud, Timothy Kehoe, Javier Romero-Vivas, Nikolaos Kehagias, Marc Zelsmann, et al.. Polymer photonic band-gaps fabricated by nanoimprint lithography. Photonics and Nanostructures - Fundamentals and Applications, 2012, 10 (4), pp.632-635. 10.1016/j.photonics.2012.06.001 . hal-00808929

## HAL Id: hal-00808929 https://hal.science/hal-00808929

Submitted on 5 Jan 2024

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### Polymer photonic band-gaps fabricated by nanoimprint lithography

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We report on the fabrication and characterization of photonic band-gaps structures by nanoimprint lithography in a dye-doped polymer. Photonic band calculations show that photonic crystal slabs composed of a triangular array of polymer pillars could exhibit photonic band-gaps for the magnetic-like modes. The resulting structures show that the nanoimprint lithography process is well-suited to fabricate in a single-step process, these challenging photonic structures opening perspectives to realize integrated photonic band-gap circuits.

Keywords: Photonic crystal; Photonic band gap; Nanoimpression

Research in applications of photonic crystals (PhCs) has been focused on demonstrating new functionalities of two-dimensional structures to generate and/or to control light, such as filters [1], couplers [2], waveguides [3] and lasers [4] at telecommunication wavelengths using semiconductor technology. Components are usually fabricated using direct electron beam lithography and dry-etching processes to transfer the pattern into a higher dielectric constant substrate. Nanoimprinted polymers can be used as an etch mask to be transferred into inorganic substrates or can be

employed directly as functional components leading to the realization of organic PhCs. So far a polymer waveguide-type wavelength filter [5,6], a micro-ring resonator [7], 1D and 2D band-edge lasers [8,9] and photonic crystals for light extraction applications [10,11] have been recently fabricated by NIL. Recently two-dimensional polymer photonic crystals displaying photonic band gap (PBG) were fabricated using an anodic porous alumina mould [12]. The incorporation of structural point defects into photonic band-gap slabs can lead to various functional nanophotonic devices [13]. Here we report a strategy to produce in a single step dye-doped polymer photonic band-gap structures exhibiting a PBG in the visible wavelength range for laser applications. Two-dimensional polymer photonic crystal based on structural defects have been designed by the plane wave expansion method (PWE) and

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successfully imprinted in a dye-doped polymer. Our results demonstrate the potential of nanoimprint lithography for the one step fabrication of low-cost, compact two-dimensional active polymer photonic band-gap structures.

A UV-crosslinkable polymer, namely mr-NIL 6000 from *Microresist Technology*, has been chosen for its mechanical properties [14] and its relatively high refractive index. The refractive index of a 400 nm thick mr-NIL 6000 layer on magnesium fluoride substrate has been measured by ellipsometry at a wavelength of 600 nm to be 1.60 (Fig. 1a). For comparison, the well-known poly(methyl methacrylate) has a refractive index of approximately 1.49 for visible wavelengths. Nanoimprinted polymer PBG structures were fabricated with pillars in the polymer as simulation shows a larger bandgap than holes in the polymer [15].

Fig. 1b presents an example of nanoimprinted dyedoped polymer pillars on a magnesium fluoride substrate (pillars pitch: 320 nm, pillars diameter: 185 nm). The width of the first band-gap as a function of the ratio of the pillar radius and the PhC lattice constant was calculated by PWE for TM polarization (Fig. 1c). A band gap width of 10.5% was obtained for a pillar radius of 0.29a where *a* is the lattice constant of the PhCs. For comparison a maximum band-gap width of 7.3% in TE polarization will be created with holes in the polymer (radius of 0.38a). An optical cavity can be then formed by removing rods to confine modes in the photonic band-gap. Fig. 1d shows the photonic band structure for TM-like modes of a triangular array of mr-NIL 6000 polymer pillars calculated by the two-dimensional plane wave expansion method.

The PhCs exhibit a complete PBG for TM-like modes between the reduced frequencies 0.468 and 0.524 for a hole radius of r = 0.29a. The low refractive index of the polymer is responsible for this narrow PBG.

For a single-step imprint of dye-doped polymeric photonic band structures, Si stamps were patterned in a silicon wafer by electron-beam lithography and etched to a depth of 175 nm by inductively coupled plasma etching. A self-assembled anti-adhesive monolayer of



Fig. 1. (a) Refractive index of 400 nm mr-NIL 6000 film measured by ellipsometry, (b) SEM micrographs of (a) tilted view of nanoimprinted PBG structure in a dye-doped polymer (mr-NIL 6000 and Rhodamine B), (c) width of the first bandgap as a function of the normalized lattice unit calculated by PWE, (d) photonic band diagram of triangular polymer pillars array calculated by PWE in reduced frequency for TM polarization. The hole radius is r = 0.29a, a is the lattice constant,  $\lambda$  is the wavelength, and c is the speed of light in vacuum. Red bands correspond to the PBG for the TM polarization.



Fig. 2. Emission spectra for mr-NIL 6000 doped with Rhodamine B taken before (solid line) and after the nanoimprint process (dot line). The spectra are normalized to the maximum intensity of the spin-coated dye-doped polymer film.

tridecafluor-1,1,2,2-tetrahydrooctyl trichlorosilane was deposited on the stamp by vapour deposition. For the dye doping of the polymer, Rhodamine B (from Sigma Aldrich) was been directly dissolved with a concentration of  $5 \times 10^{-3}$  mol/L, in mr-NIL 6000.

One further advantage of mr-NIL 6000 is its relatively low glass transition temperature of about 40–45 °C, which is lower than that of other printable polymers, e.g. PMMA. This reduces the possible thermal degradation of Rhodamine B during the nanoimprinting process. In fact, the photoluminescence (PL) spectrum of the spin-coated dye-doped polymer on the magnesium fluoride substrate was compared to that of the same substrate imprinted with a flat stamp and a decrease intensity of less than 10% was measured after patterning at 60 bar for 5 min at 90 °C (Fig. 2). The emission spectrum of the dye is centred at 602 nm.

From the PWE calculations (Fig. 1d), lattice constant of 300 nm and 340 nm was chosen to obtain a TM-like mode PBG.

Fig. 3a shows the scanning electron micrograph of the silicon stamp with a deliberate structural defect. The lattice constant of the PhC is 300 nm in this case and the diameter of the holes is 175 nm (r = 0.29a). The silicon stamp has then been imprinted in a 400 nm thick dyedoped polymer film spin coated on a magnesium fluoride substrate. Fig. 3b shows the excellent pattern transfer in the optically active polymer. Undoped imprinted samples were cleaved to allow measurement of the reflection spectrum of the polymer pillar array in the  $\Gamma$ -M direction (Fig. 3b). A magnification 100× optical microscope objective was employed using



Fig. 3. (a) SEM micrographs of a silicon stamp containing twodimensional triangular array of holes (pitch: 300 nm, hole diameter: 175 nm). (b) SEM micrographs of nanoimprinted photonic crystals in dye-doped polymer.

polarized white light from a tungsten halogen lamp to shine the light of the cleaved facet of the undoped imprinted photonic crystal sample. The beam was shined on a defectless photonic crystal without a cavity. The reflected light was collected through the same microscope objective and separated by a beam splitter to be sent to an optical spectrometer.

Fig. 4a shows the reflection spectra of polymer photonic crystals without cavities with a pitch of 300 nm and 340 nm having the same filling factor. For the transverse magnetic (TM) polarization a spectral stop band is observed for the two PhCs. For the transverse magnetic (TM) polarization a spectral stop band is observed for the two PhCs. The position and the width of the stop band are in relatively good agreement with the one calculated by plane wave expansion method. The position of the stop band shifts to a higher wavelength when the pitch of the pillar array increases,



Fig. 4. Reflectance spectra of undoped polymer pillar arrays with a pitch of 300 nm and 340 nm along the  $\Gamma$ -M direction of the photonic crystals.

as expected. After showing the presence of photonic band-gap, the dye-doped polymer was optically pumped with 514.5 nm light incident to normal to the surface with a power of 50  $\mu$ W. The 20  $\mu$ m diameter spot was focused on the defect of the PhCs. The reflected light was collected thought the same microscope objective perpendicularly to the surface. The PL spectra were measured through a polarizer by an optical spectrum analyser. No cavities effect of the PhCs was observed suggesting a weak coupling regime between the emitters and the cavity. To observe a possible lasing effect, experiments should be performed with a pulsed laser to reach higher power density pumping with a low repetition rate to avoid photo-bleaching.

In conclusion, an original NIL process has been developed to realize, in a single imprint step, active polymer PBG structures on magnesium fluoride substrates. Designs have been proposed to obtain polymer photonic band-gaps operating in the visible range. NIL conserves the high tolerances needed for such PBG devices and has the potential of cost-efficient production for compact PBG circuits. Optical characterization showed the presence of photonic stop bands which depend on the periodicity of the 2D nanoimprinted photonic crystals.

#### Acknowledgements

We gratefully acknowledge M. Brøkner Christiansen and A. Kristensen for the dye-doped polymer. The support of the EC-funded project NaPaNIL is gratefully acknowledged. The content of this work is the sole responsibility of the authors.

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