Fabrication of three-dimensional void photonic crystals using ultrafast-laser driven micro-explosion in a solid polymer material

Guangyong Zhou, Michael James Ventura, Michael Ross Vanner, and Min Gu* Centre for Micro-Photonics and Centre for Ultrahigh-Bandwidth Devices for Optical Systems, Faculty of Engineering and Industrial Sciences, Swinburne University of Technology, PO Box 218, Hawthorn, Victoria 3122, Australia

ABSTRACT

Micron-sized void dots have been generated in a solidified resin by using ultrafast-laser driven micro-explosion method. Side view confocal images of the void dots show that the void dots are almost spherical. The diameter of the void dots can be controlled by adjusting the laser power and exposure time. Three-dimensional (3D) structures, stacked in the [100] lattice direction, of diamond, FCC and BCC lattices have been fabricated, respectively. Multi-order stop gaps are observed for all three different types of structures. The suppression rate of the first order gap can be up to 70% for diamond and FCC structures. The angle dependence of the bandgap properties of a diamond structure reveals that the observed first order gap shifts to the longer wavelength whereas the second gap shifts to the shorter wavelength as the angle of incidence increases. Such a sensitive angular dependence of the bandgap structure may find applications in photonic crystal superprisms.

Keywords: Micro-explosion, polymer, three-dimensional photonic crystals, diamond lattice, face-centered-cubic lattice, body-centered-cubic lattice, photonic bandgap properties, microfabrication

1. INTRODUCTION

In recent years the fabrication of photonic crystals has attracted extensive interest as such artificial periodic structures may control the behavior of light in such a way that semiconductors control the behavior of electrons.^{1,2} The unique properties of photonic crystals may result in novel devices which could find broad applications in integrated optics and all-optics communication networks. By tightly focusing ultrafast laser light into transparent solid materials, micro-explosion occurs at the focal point, where the material is ejected from the center forming a void cavity surrounded by a region of compressed material.³⁻⁵ This method was firstly proposed to be used in three-dimensional read-only optical data storage.^{4,5} Smooth void dots and void channels have been generated in glass^{6,7} and polymer materials⁸⁻¹¹ and stacked into two-dimensional (2D) and three-dimensional (3D) photonic crystals. This technique is a one-step approach, which does not require chemical post processing, and results in photonic crystals with a high degree of perfection. Although the refractive index of polymer and glass is not high enough to open a complete photonic bandgap, partial bandgaps (stop gaps) can be achieved in the near infrared wavelength range and could be used for superprisms where a complete bandgap is not prerequisite.^{12,13} The lower power threshold to generate void dots in polymer materials compared with glass materials.

Void channel based woodpile structures in a solidified optical adhesive NOA 63 (Norland Product Inc. USA) can easily open a partial bandgap (stop gap) and higher order gaps in the stack direction.^{9,14} However, it is not easy to achieve different types of lattice structures and the gaps are not sensitive to the angle of incidence. It is more flexible to arrange void dots to fabricate arbitrary lattices.^{7,8,11} In this paper, we report the generation of spherical void dots in a solidified NOA 63 resin and fabrication of photonic crystals with diamond, face-centered-cubic (FCC) and body-centered-cubic (BCC) lattices. The photonic bandgap properties of the diamond structures are studied in detail.

Nanophotonics, Nanostructure, and Nanometrology, edited by Xing Zhu, Stephen Y. Chou, Yasuhiko Arakawa, Proceedings of SPIE Vol. 5635 (SPIE, Bellingham, WA, 2005) 0277-786X/05/\$15 · doi: 10.1117/12.569944

^{*} Email: mgu@swin.edu.au; fax: 61-3-92145435; phone: 61-3-92148776

2. EXPERIMENTAL SETUP

Figure 1 shows the experimental setup used to generate void dots and photonic crystals. A 740 nm laser light beam came from a Tsunami femtosecond oscillator (Spectra-Physics, USA). The laser light beam was expanded by an inverse telescope system and tightly focused by a 1.45 numerical aperture (N.A.), $60\times$ oil immersion objective. A shutter was used to control the laser light power. The fabrication process was *in situ* monitored with a CCD camera. The sample was affixed to a 3D piezoelectric scanning system (PI, Germany) which was controlled by a computer. Two different types of samples were used in this work. In order to study the side view of the void dots, a cuboid bulk sample was used. For all other experiments, thin films with a thickness of approximately 100 μ m were used. The void dots were characterised by use of a confocal microscope (Fluoview, Olympus, Japan). A Fourier transform infrared (FTIR) spectrometer fitted with a 32×, 0.65-NA reflective objective (Thermo Nicolet, Madison, WI, USA) was used to measure the transmission spectra of the fabricated photonic crystals.



Figure 1: Sketch of a femtosecond laser microfabrication system.

3. RESULTS AND DISCUSSIONS

3.1 Void generation and characterisation

When the laser power is above the threshold, micro-explosion occurs at the focal point and a void cavity is generated therein. Figure 2(a) shows the confocal reflection (left side) and transmission images (right side) of an individual void dot generated with a power of 40 mW and an exposure time of 10 ms. The strong reflection single in the confocal reflection image indicates high refractive index contrast between the fabricated and unfabricated regions, which verifies the generation of a void cavity.⁵ From Fig. 2(a) one can see that the transverse diameter of the void is approximately 1.8 μ m. To further confirm the generation of the void cavity, x-z scanning confocal images were measured, as shown in Fig. 2(b). Two strong reflection signals were observed in the confocal reflection image (left side), which correspond to the top and bottom surfaces of the void cavity. The right side of Fig. 2(b) shows a typical Becke line phenomenon. As shown in the transmission image in Fig. 2(b), as the void dot move away from the objective (from bottom to top), the bright halo moves into the pure resin, which indicates that the refractive index of the fabricated region is lower than the unfabricated region.

The size of the void dots dependents on the laser power and the exposure time. Increasing the laser power/exposure time, the diameter of the void dots increases correspondly. As shown in Fig. 3, at the power of 40 mW, the diameter of the void dots increases from $\sim 1 \ \mu m$ to $\sim 1.8 \ \mu m$ as the exposure time increases from 5 ms to 30 ms. An approximately linear relationship has been observed. At the power of 60 mW, the diameter of the void dots is larger than that at the power of 40 mW with the same exposure time. But the dependence of the void diameter on the exposure time is not linear. When the exposure time is longer than 20 ms, the increase of the diameter becomes slower and the diameter of the void dots tends to saturate. Therefore, the maximum diameter of void dots can be achieved is approximately 2.2 μm with a 1.45-NA objective at a wavelength of 740 nm.





Figure 2: x-y scanning and x-z scanning images of individual void dot. Left side: confocal reflection images; right side: confocal transmission images.

Figure 3: Diameter of void dots as a function of exposure time and laser power.

In order to view the fabricated void dots in the fabrication (longitudinal) direction, a cuboid bulk sample was prepared and 22 rows of void dots were fabricated at different depths with dot spacing and row spacing of 6 μ m and 3 μ m, respectively, as illustrated in Fig. 4(a). After fabrication, the sample was rotated by 90 degrees and viewed by use of a confocal microscope. Fig. 4(b) shows the reflection (left) and transmission confocal images (right) of the void dots. Smooth and uniform void dots can be seen from the zoom-in transmission confocal images in Figs. 4(c) and (d). Due to the slight effect of spherical aberration caused by the refractive index mismatch between the polymer (1.56) and immersion oil (1.52),¹⁵ the peak power at the focal point becomes lower at a deeper depth, which results in smaller voids, as shown in Figs. 4(c) and (d). As a result, the diameter of the dot decreases from approximately 1.8 μ m near the top surface down to approximately 1.2 μ m at a depth of 60 μ m. It should be pointed out that the void dots are almost spherical which is different from the elliptical cross section of the fabricated rods in the two-photon polymerisation method.¹⁶ The possible reason is that, in the case of micro-explosion, high pressure gas exists in a void and a spherical shape is a stable state. Due to the flexibility of the polymer, voids may be reshaped to the stable spherical shape right after the formation of the void dots.



Figure 4. Side view of fabricated void dots at different depths. (a) Sketch of the void dots arrangement within a bulk polymer sample; (b) confocal reflection (left side) and transmission (right side) images of void dots at different depths; (c) zoom-in image of void dots at smaller depths; (d) zoon-in image of void dots at larger depths

3.2 Photonic bandgap properties of 3D photonic crystals with a diamond lattice

By stacking void dots layer-by-layer, we fabricated 3D structures with three different lattice arrangements, i.e. a diamond lattice, a face-centered-cubic (FCC) lattice and a body-centered-cubic (BCC) lattice. The unit cells of these

lattices are shown schematically Fig. 5, where the [100] direction has been indicated. In this section, we report in detail the photonic bandgap properties of diamond structures.



Figure 5. Sketches of diamond (left), face-centered-cubic (center) and body-centered-cubic (right) lattices.

The best way to check the uniformity of the void dots lattices is to measure the transmission spectra of the fabricated structures. A Nicolet Nexus Fourier transform infrared (FTIR) spectrometer was used to provide infrared light with the wavelength from 1 μ m to 5 μ m and a 32×, NA 0.65 reflective objective was used to focus the light beam on the structure. This objective provides an incident hollow light cone with an outer angle of 40° and an inner angle of 15° (which corresponds to 25° and 10° in the sample). A tiny off-centered aperture was attached to the objective to reduce the angle of incidence range to approximately 5 degrees (inside the sample),¹¹ which corresponds to approximately 3% of total illumination energy. Illumination at different angles of incidence was achieved by adjusting the position of the aperture and tilting the sample. Figure 6 shows the transmission spectra of 32-layer diamond structures in the [100] direction with the lattice constant of 4.12 μ m, 4.44 μ m and 4.72 μ m. One can see strong suppression peaks for all three lattice constants. The strong suppression rate of the observed first and second order gaps, approximately 65% and 75% respectively, indicates that the fabricated void dots are well correlated each other. One can also see that the gaps shift to longer wavelengths as the lattice constant. As expected, a linear relationship is observed.



Figure 6: Baseline-corrected transmis-sion spectra of 3D void diamond photonic crystals stacked in the [100] direction with a lattice constant of 4.12 μ m, 4.44 μ m and 4.72 μ m. The insert shows the mid-gap wavelength of the observed three gaps as a function of the lattice constant.

The photonic bandgap properties of 3D diamond lattice photonic crystals depend on the lattice direction. Therefore the angle dependence of the bandgap properties of a 3D diamond structure stacked in the [100] direction with a lattice constant of 4.44 μ m was investigated. Figure 7 shows the baseline-corrected transmission spectra of different angles of incidence. The observed first order gap shifts to a longer wavelength where as the observed second order gap shifts to shorter wavelength when the angle of incidence increases. The mid-gap wavelength as a function of the angle of incidence of the first and second order gaps is shown in the inset of Fig. 7. It is expected that the observed first and second gaps would overlap in the [100] direction. The dependence of the suppression rate of the observed gaps on the layer number was studied. Figure 8 shows the baseline-corrected transmission spectra of the photonic crystals consisting of different layers of void spheres with a lattice constant of 4.28 μ m. The transmittance of the three observed gaps as a function of the layer number is shown in the inset. Increasing the layer number from 12 to 32, the suppression rate increases from 46% to 75% for the observed second order gap.



Figure 7. Baseline-corrected transmission spectra of 3D diamond photonic crystals stacked in the[100] direction ($a = 4.44 \mu$ m) at different directions of light incidence. The dependence of the mid-gap wavelength on the angle of incidence is given in the inset.

Figure 8. Baseline-corrected transmission spectra of 3D diamond void photonic crystals stacked in the [100] direction ($a = 4.28 \mu$ m) for different numbers of layers. The mid-gap wavelength of the observed first, second and third order gaps as a function of the layer number is plotted in the inset. The labels L12, ...L32 denote the layer number of 12, ... 32, respectively.

3.3 Photonic bandgap properties of 3D photonic crystals with FCC and BCC lattice

Structures with FCC and BCC lattices have also been fabricated and partial photonic bandgaps have also been observed. Figure 9 shows the transmission spectra of 28-layer FCC structures with the lattice constant of $3.4 \,\mu\text{m}$, $4.0 \,\mu\text{m}$ and $4.96 \,\mu\text{m}$. Two orders of stop gaps were observed, which are marked in the figure. The suppression rate of transmission of the observed first order gap is as large as 74%. The second order stop gap located at exactly a half of the wavelength of the first order gap is also observed, with a suppression rate of approximately 20%. Both the first and second order stop gaps shift to the longer wavelengths with an increase in the lattice constant. Figure 10 shows the transmission spectra of a 28-layer BCC structure stacked in the [100] direction with a lattice constant of $4 \,\mu\text{m}$. Three order stop gaps can be observed. Because the equivalent void dot density is much lower (2 dots per unit cell) compared with that of the FCC (4 dots per unit cell) and the diamond (8 dots per unit cell), the suppression rate (approximately 30% for the first gap) of the stop gap is much lower for BCC structures.



Figure 9. Transmission spectra of 3D FCC void photonic crystals stacked in the [100] direction with different lattice constants of $3.4 \,\mu\text{m}$, $4 \,\mu\text{m}$ and $4.96 \,\mu\text{m}$ [(a)-(c)].



Figure 10. Transmission spectra of a BCC structure with a lattice constant of 4 μ m stacked in the [100] direction. The observed three stop gaps are marked.

4. CONCLUSIONS

In conclusion, spherical void dots have been fabricated in solidified NOA 63 resin by use of ultrafast laser driven micro-explosion method. The size of the void dots can be controlled by adjusting the laser power and the exposure time. 3D structures with three different lattices, i.e. diamond, FCC and BCC lattices, have been fabricated in the [100] direction. Multi-order stop gaps have been observed for all three types of structures. The suppression rate of the first gap can be up to 70% for diamond and FCC structures. The results indicate that the micro-explosion method can be used to fabricate 3D void-based photonic crystals with arbitrary lattice. The angle dependence of the bandgap properties of a diamond structure reveals that the observed first order gap shifts to the longer wavelength whereas the second gap shifts to the shorter wavelength as the angle of incidence increases. Such a sensitive angular dependence of the bandgap structure may prove advantages in angle-depedent photonic devices such as superprisms.

ACKNOWLEDGEMENT

This work was produced with the assistance of the Australian Research Council under the ARC Centres of Excellence Program. CUDOS (the Centre for Ultrahigh bandwidth Devices for Optical Systems) is an ARC Centre of Excellence

REFERENCES

1. E. Yablonovitch, "Inhibited Spontaneous Emission in Solid-State Physics and Electronics," Phys. Rev. Lett. 58, 2059-2062 (1987).

- 2. S. John, "Strong localization of photons in certain disordered dielectric superlattices," Phys. Rev. Lett. 58, 2486-2489 (1987).
- 3. E. N. Glezer and E. Mazur, "Ultrafast-laser driven micro-explosions in transparent materials," Appl. Phys. Lett. **71**, 882-884 (1997).
- K. Yamasaki, S. Juodkazis, M. Watanabe, H. –B. Sun, S. Matsuo, and H. Misawa, "Recording by microexplosion and two-photon reading of three-dimensional optical memory in polymethylmethacrylate films," Appl. Phys. Lett. 76, 1000-1002 (2000).
- 5. D. Day and M. Gu, "Formation of voids in a doped polymethylmethacrylate polymer," Appl. Phys. Lett. **80**, 2404-2406 (2002).
- 6. H.-B. Sun, Y. Xu, S. Matsuo, and H. Misawa, "Microfabrication and characteristics of two-dimensional photonic crystal structures in vitreous silica," Opt. Rev. 6, 396-398 (1999).
- 7. H.-B. Sun, Y. Xu, S. Juodkazis, K. Sun, M. Watanabe, S. Matsuo, H. Misawa, and J. Nishii, "Arbitrary-lattice photonic crystals created by multiphoton microfabrication," Opt. Lett. 26, 325-327 (2001).
- H.-B. Sun, Y. Xu, K. Sun, S. Juodkazis, M. Watanabe, S. Matsuo, H. Misawa, and J. Nishii, "Inlayed 'atom'-like three-dimensional photonic crystal structures created with femtosecond laser microfabrication," Proc. Mater. Res. Soc. 605, 85-90 (2000).
- 9. M. J. Ventura, M. Straub, and M. Gu, "Void channel microstructures in resin solids as an efficient way to infrared photonic crystals," Appl. Phys. Lett. **82**, 1649-1651 (2003).
- 10. G. Zhou, M. J. Ventura, M. Straub, and M. Gu, "In-plane and out-of-plane band-gap properties of a twodimensional triangular polymer-based void channel photonic crystal," Appl. Phys. Lett. 84, 4415-4417 (2004).
- 11. G. Zhou, M. J. Ventura, M. R. Vanner, and M. Gu, "Use of ultrafast-laser-driven microexplosion for fabricating three-dimensional void-based diamond-lattice photonic crystals in a solid polymer material," Opt. Lett. **29**, 2240-2242 (2004).
- 12. S. Lin, V. M. Hietala, L. Wang, and E. D. Jones, "Highly dispersive photonic band-gap prism," Opt. Lett. 21, 1771-1773 (1996).
- 13. T. Baba and M. Nakamura, "Photonic crystal light deflection devices using the superprism effect," IEEE J. Quantum Electron. 38, 909-914 (2002).
- 14. M. Straub, M. J. Ventura and M. Gu, "Multiple Higher-Order Stop Gaps in Infrared Polymer Photonic Crystals," Phys. Rev. Lett. Vol. **91**, 043901 (2003).
- 15. D. Day and M. Gu, "Effects of Refractive-Index Mismatch on Three- Dimensional Optical Data-Storage Density in a Two-Photon Bleaching Polymer," Appl. Opt. **37**, 6299-6304 (1998).
- 16. M. Straub and M. Gu, "Near-infrared photonic crystals with higher-order bandgaps generated by two-photon photopolymerization," Opt. Lett. Vol. 27, 1824-1826 (2002).