

# Arbitrary-lattice photonic crystals created by multiphoton microfabrication

Hong-Bo Sun

*Satellite Venture Business Laboratory, The University of Tokushima, 2-1 Minamijyosanjima, Tokushima 770-8506, Japan*

Ying Xu, Saulius Juodkazis, Kai Sun, Mitsuru Watanabe, Shigeki Matsuo, and Hiroaki Misawa

*Department of Ecosystem Engineering, The University of Tokushima, 2-1 Minamijyosanjima, Tokushima 770-8506, Japan*

Junji Nishii

*Optical Materials Division, Osaka National Research Institute, 1-8-31 Midorigaoka, Iketa, Osaka 563-8577, Japan*

Received October 16, 2000

We used voxels of an intensely modified refractive index generated by multiphoton absorption at the focus of femtosecond laser pulses in Ge-doped silica as photonic atoms to build photonic lattices. The voxels were spatially organized in the same way as atoms arrayed in actual crystals, and a Bragg-like diffraction from the photonic atoms was evidenced by a photonic bandgap (PBG) effect. Postfabrication annealing was found to be essential for reducing random scattering and therefore enhancing PBG. This technique has an intrinsic capability of individually addressing single atoms. Therefore the introduction of defect structures was much facilitated, making the technique quite appealing for photonic research and applications. © 2001 Optical Society of America

OCIS codes: 160.4670, 160.4760, 160.6030, 190.4180, 220.4000, 230.3990.

Microfabrication of femtosecond lasers through the nonlinear processes of photon–matter interactions has recently attracted much research effort. It has been successfully applied to fields such as prototyping by photopolymerization of resins<sup>1,2</sup> and three-dimensional (3D) optical memory in transparent media.<sup>3–6</sup> These applications rely on highly localized light excitation and modification of material properties induced by two-photon or multiphoton absorption. For optical memory, a reversible (photorefraction<sup>7,8</sup>) or a permanent (bond-breaking<sup>3–6,9,10</sup>) change of refractive index is induced at the focal point, where voxels of subwavelength or near-wavelength size are recorded. Writing voxels plane by plane in, e.g., silica, yielded a recording density as high as 0.8 Tbits/cm<sup>3</sup>.<sup>6</sup> Readout based on the differences in refractive indices of voxels and matrices has been demonstrated.<sup>3,7,8</sup>

If the deposited energy of the laser and the focusing conditions are properly chosen, the voxels will take the form of a well-defined shapes similar to that of a sphere. After processing of the internal morphology by, e.g., annealing, etching, and (or) change of composition by dye filling, the voxel with a radial refractive-index distribution may possess complete optical functions, such as light confinement. Therefore in this Letter we term the voxels “photonic atoms.” An isolated photonic atom may function as a microcavity, and this is particularly interesting when such atoms are regularly organized. This possibility is attractive when generic crystal structures are concerned, for which the properties of particular atoms and their periodic array determine all aspects of the properties of the materials. If the photonic atoms were arranged in the same way as atoms in crystals, the variation in refractive index would produce periodic modulation of light as Coulombic potential does for electrons. Such structures are actually photonic crystals (PhC’s),<sup>11</sup>

with lattices directly scaled up from atom point lattices of crystals that exist in nature. Although photonic devices such as fibers<sup>12</sup> and two-dimensional PhC’s made by piling gratings<sup>13</sup> have been reported, the structures that we propose here are more attractive and are better suited for use in for engineering novel photonic materials and in controlling their properties.

In this Letter we report what is to our knowledge the first experimental realization of this idea. Notably, photonic bandgap (PBG) effects that arise from Bragg-like photonic atom diffraction were clearly observed. Postfabrication annealing that was utilized to depress random scattering was found to be essential for the formation of PBG’s.

Laser pulses of 150-fs duration and 400-nm wavelength (second harmonic of a mode-locked Ti:sapphire laser) at repetition rates adjustable from 1 to 1000 Hz were focused into silica (10GeO<sub>2</sub>:90SiO<sub>2</sub> mol. %) by an objective lens (100×; N.A., 1.35). The energy of laser fabrication was typically three times that of the laser-induced breakdown threshold, ~0.55 μJ/pulse. A small portion of the average beam power (~1 W) was split by a beam splitter and detected by a photodiode, generating pulsed signals to trigger the piezoelectric stage movement. With stage translation synchronized to the laser pulse output, the designed 3D dot patterns were faithfully replicated to matter structures. Figure 1 shows one plane of a simple cubic (sc) lattice. Nearly spherical voxels with outer diameters of ~1.0 μm are recorded by a single shot per voxel. The voxel, as revealed by atomic-force microscopy, is hollow, with a core diameter of approximately 200–300 nm. This submicrometer-sized feature confined at the focal point is smaller than the lateral diffraction limit, 360 nm, of the fabrication laser as estimated by the Rayleigh criterion:  $\omega_0 = 1.22\lambda/N.A.$ , where  $\lambda$  is the laser wavelength.

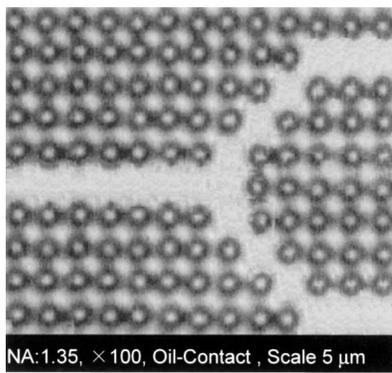


Fig. 1. (100) Plane of an *sc* photonic lattice in Ge-doped silica. The empty Y-shaped area consists of missing photonic atoms and functions as a microintegrated optical waveguide.

For the electronic bandgap of silica (8.9 eV) and the photon energy (3.1 eV), the electron excitation is a three-photon absorption process. Multiphoton absorption does not play an important role in that process unless the laser field strength becomes comparable with the atomic field strength (typically  $10^5$ – $10^8$  V/m). In the current experimental scheme, a highly transient photon flux that arises from intense compression of the pulses in the time domain ( $\sim 150$  fs FWHM) and tight focusing in the spatial domain ( $\sim 360$ -nm waist diameter of a Gaussian beam) gives a field of the order of  $10^8$  V/m. Consequently, material was excited to the free-carrier plasma state primarily by multiphoton absorption tunneling, and, partially, by impact ionization during the pulse duration. The highly excited and tightly localized plasma expands in an explosive way before it can transfer the energy to a lattice, generating a hollow core surrounded by a denser phase. Indirect spectroscopic evidence for such a physical process has been presented elsewhere.<sup>9,10</sup>

Because the absorption at a chosen fundamental wavelength was negligible, the laser light could penetrate the surface and was focused at arbitrary sites inside the materials. Photonic atoms can be configured to various lattice points, scaled up from 7 systems, 32 classes, and 230 space groups of generic crystals. The relevant wavelength also shifts from an x-ray waveband to optical and infrared regions. None of currently available techniques that are used for the production of PhC's has such a capability. As an example, we present results from a face centered cubic (*fcc*) lattice, one of the most widely investigated structures. Figure 2(a) shows a point lattice of monatomic *fcc* crystals. Dots can be written in a random order. Nevertheless, for convenience in programming and *in situ* observation during fabrication, low-index plane packing is preferred. Both (100) and (111) planes can be used as primitive piling planes, repeated at every second [for (100)] and third [for (111)] layer. Figure 2(b) is a photograph of one layer (111) plane in a 3D array that is produced by (111) packing. PhC's occupied an area of  $40 \mu\text{m} \times 40 \mu\text{m}$ , and the thickness of the array was  $20 \mu\text{m}$ , which corresponds to seven lattice periods.

After the spots were organized into a photonic lattice as a replica of atomic point lattices, new optical-photonic phenomena were expected; the principal phenomenon is the formation of PBG's.<sup>11,14,15</sup> The PBG effects of the PhC as shown in Fig. 2(b) were investigated (samples need annealing, as discussed below) by Fourier-transform infrared spectroscopy. A typical transmission spectrum is shown in Fig. 3 (filled squares). The minimum transmittance occurred at  $3490 \text{ cm}^{-1}$ . Changing the lattice constant caused a synchronous variation of the wavelength of the transmission valley, as predicted by Bragg law, showing that the narrow valley is indeed a result of interatom interference.

We calculated a transmission spectrum to reproduce the wavelength of the experimentally observed transmission dip. For this purpose we used the transfer matrix technique of Bell *et al.*<sup>14</sup> The difference in refractive index  $\Delta n$  and hole diameter ( $2r$ ) was in fitting parameters. As a result,  $\Delta n \sim 0.45$  and  $2r \sim 250$  nm resulted in good agreement between the measured and the calculated dips. The refractive-index difference of 0.45 is much larger than those achieved by photorefractive techniques, which typically are of the order of  $10^{-4}$  to  $10^{-3}$  ( $\Delta n/n$ ).

It should be emphasized that commonly no wavelength-dependent transmission dip is observable in as-fabricated samples (upward-pointing triangles in Fig. 3). It was observed previously that the local

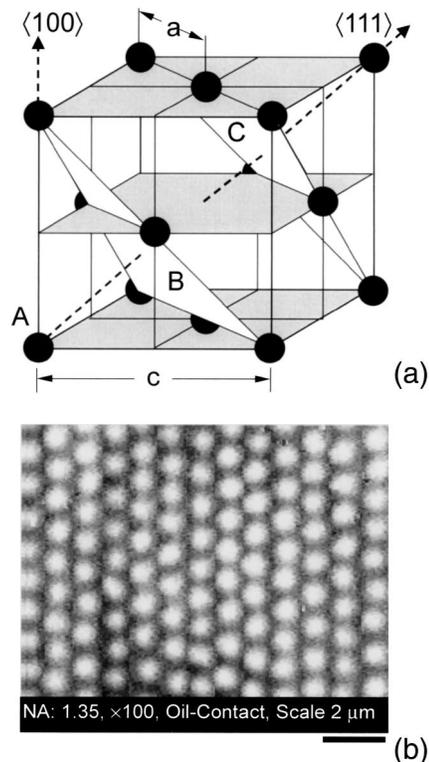


Fig. 2. PhC of the *fcc* lattice fabricated in Ge-doped silica ( $10\text{GeO}_2:90\text{SiO}_2$  mol.%). (a) Schematic of the spatial voxel array, where *a* and *c* denote voxel spacing in (111) planes and the lattice constant, respectively. We created the structure by alternating (111) planes in the order ABC(ABC)... (b) Plane (111) in a fabricated 3D photonic lattice.

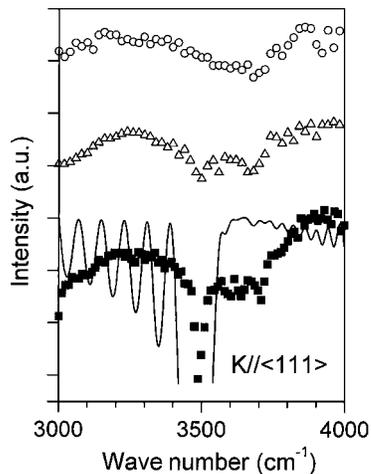


Fig. 3. Measured and simulated transmission spectra (one division on the intensity axis denotes a 10% variation of transmission). Squares, upward-pointing triangles, and circles denote spectra from annealed PhC's, as-fabricated PhC's, and annealed randomly distributed dots, respectively (the latter two curves were offset upward for clarity). The solid curve is from a simulation.

explosion at focal points leads to intense structural modifications and the formation of material defects<sup>9,10</sup>; thus-induced metafeatures (e.g., size approximately tens of nanometers) can result in strong light scattering, which may obscure any potential PBG effects. It has been observed that point defects about the voxels were reduced by annealing and diminished at 700 °C.<sup>9,10</sup> The disappearance of defects should be accompanied with a variation of internal morphology of voids, changed to a smoother state. In this research, samples that contained PhC's were annealed at an even higher temperature, 900 °C, for 30 min. An attenuation of frequency gaps did appear after annealing (squares in Fig. 3). The same thermal treatment was performed on samples consisting of randomly distributed dots; no similar band appeared (circles in Fig. 3), which shows that the band is not caused by absorption.

The PBG behavior of the photonic lattices implied an ability to guide light<sup>15</sup> if a proper defect structure of photonic atoms, for example, the lattice that was shown in Fig. 1, where the missing spots form a Y-shaped waveguide, is introduced. If a complete bandgap existed in the background lattice, the light within the frequency range of the PBG would be forced to propagate along the prescribed path because the modes are prevented from escaping into the crystal. Here we emphasize that we achieved this waveguide only by programming (skipping points in the lattice design or shuttering the exposure of specific points) because of the intrinsic capability of the direct laser writing technique used here to individually address single atoms. This technique clearly contrast with some conventional techniques such as self-organization of dielectric spheres (despite the utilization of templates and recent endeavors to develop reverse opal structures<sup>16</sup>). The ease of defect induction makes the technique even more promising for designing 3D opti-

cal circuits, especially in transparent electronic materials, that involve more-complicated photonic-optoelectronic functions.

In future, two problems need to be solved: (i) low filling ratio, that is, a volume percentage of holes that is smaller than that necessary for opening a full bandgap. This problem may be solved by wet etching, as used by Blanco *et al.*<sup>17</sup> As we recently found that the spot region is chemically active, that region can be selectively etched off by HF acid, for example. (ii) Low dielectric ratio. A utilization of dielectrics with high refractive indices, for example, use of amorphous TiO<sub>2</sub> ( $n = 2.4\text{--}2.6$ ; transparent at  $\alpha = 1\text{ cm}^{-1}$  in  $0.43\text{--}5.3\text{ }\mu\text{m}$ ) and GaP ( $n = \sim 3.1$ ; transparent at  $\alpha = 10\text{ cm}^{-1}$  in  $0.52\text{--}7.2\text{ }\mu\text{m}$ ), will solve this problem.

In conclusion, by utilizing annealed voxels induced by femtosecond lasers in transparent media as photonic atoms, we produced photonic lattices as a replica of atomic point lattices of crystals on scaled-up dimensions. The crystalline order of the photonic atoms was confirmed by pronounced PBG effects.

This research was supported in part by a grant-in-aid for scientific research (A)(2) 09355008 from the Ministry of Education, Science, Sports and Culture, the Marubun Research Promotion Foundation, and the Satellite Venture Business Laboratory of the University of Tokushima. H. Misawa's e-mail address is misawa@eco.tokushima-u.ac.jp.

## References

1. S. Maruo, O. Nakamura, and S. Kawata, *Opt. Lett.* **22**, 132 (1997).
2. H.-B. Sun, S. Matsuo, and H. Misawa, *Appl. Phys. Lett.* **74**, 786 (1999).
3. J. H. Strickler and W. W. Webb, *Opt. Lett.* **16**, 1780 (1991).
4. E. N. Glezer and E. Mazur, *Opt. Lett.* **21**, 2023 (1996).
5. E. N. Glezer and E. Mazur, *Appl. Phys. Lett.* **71**, 882 (1997).
6. M. Watanabe, S. Juodkakis, H.-B. Sun, S. Matsuo, and H. Misawa, *Appl. Phys. Lett.* **77**, 13 (2000).
7. D. Day, M. Gu, and A. Smallridge, *Opt. Lett.* **24**, 948 (1999).
8. H. Ueki, Y. Kawata, and S. Kawata, *Appl. Opt.* **35**, 2456 (1996).
9. H.-B. Sun, S. Juodkakis, M. Watanabe, S. Matsuo, H. Misawa, and J. Nishii, *J. Phys. Chem. B* **104**, 3450 (2000).
10. M. Watanabe, S. Juodkakis, H.-B. Sun, S. Matsuo, and H. Misawa, *Phys. Rev. B* **60**, 9959 (1999).
11. E. Yablonovitch, *Phys. Rev. Lett.* **58**, 2059 (1987).
12. K. Miura, J. Qiu, H. Inouye, T. Mitsuyu, and K. Hirao, *Appl. Phys. Lett.* **71**, 3329 (1997).
13. H.-B. Sun, Y. Xu, S. Matsuo, and H. Misawa, *Opt. Rev.* **6**, 396 (1999).
14. P. M. Bell, J. B. Pendry, L. Martin-Moreno, and A. J. Ward, *Comp. Phys. Commun.* **85**, 306 (1995).
15. J. D. Joannopoulos, P. R. Villeneuve, and S. Fan, *Nature* **386**, 143 (1997).
16. K. Busch and S. John, *Phys. Rev. Lett.* **83**, 967 (1999).
17. A. Blanco, E. Chomski, S. Grabtchak, M. Ibsate, S. John, S. W. Leonard, C. Lopez, F. Meseguer, H. Miguez, J. P. Mondia, G. A. Ozin, O. Tader, and H. M. van Driel, *Nature* **405**, 437 (2000).