Fabrication of hetero-binary and honeycomb photonic crystals by one-step holographic lithography

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(Received 26 August 2005; accepted 3 February 2006; published online 2 March 2006)

We report the simulation and fabrication of two-dimensional hetero-binary and honeycomb photonic crystals by one-step holographic lithography. These structures are realized by introducing one or three auxiliary beams into the three basic beams forming a regular hexagonal structure. The size contrast between the center rod and its six neighbors in a hetero-binary structure can be tuned by adjusting the intensity contrast between the auxiliary beam and basic beams. The idea of creating heterogeneity into the interference pattern by auxiliary beams could be easily extended to three-dimensional case by adding more beams in the vertical direction. © 2006 American Institute of Physics. [DOI: 10.1063/1.2181270]

Since its first introduction by Yablonovitch¹ and John,² photonic crystal (PhC) has attracted steadily growing interest for the past decades due to the fascinating properties and intriguing application for the miniaturization and integration of optical devices. Photonic crystals also exhibit a variety of new physical phenomena, including the suppression or enhancement of spontaneous emission, low-threshold lasing, and quantum information processing. Fabricating controllable complex micro/nanostructures has been a hot topic in the experimental studies on photonic crystals. Various techniques, such as e-beam lithography, self-assembly and interference lithography have been applied to fabricate PhCs including one-dimensional, two-dimensional (2D) and three-dimensional (3D) structures.³⁻⁷

Holographic lithography was first proposed and implemented by Berger, Gauthier-Lafaye, and Costard⁸ to fabricate 2D hexagonal patterns in a photosensitive polymer, which subsequently served as an etching mask for transfer to a high-index silicon substrate. This technique was extended by Campbell *et al.*⁶ and Shoji and Kawata⁹ by introducing up to two additional laser beams to create 3D structures. As being able to produce defect-free, nanometer-scale structures over large area uniform PhCs in a single step fabrication, this technique is a very economical and powerful tool and might hold the key to volume production of photonic structures.^{6,10} Much effort has been devoted to develop this method to fabricate 2D and 3D micro/nanostructures.¹¹ However, most of the work on this topic has been concentrated on fabricating 14 simple Bravais lattices except that by Wang *et al.* on 2D quasicrystals.^{11(b)}

Compared to the regular hexagonal PhC structure, honeycomb lattice of dielectric rods in air exhibits a very large absolute gap that extends over a wide range of filling

fractions.¹² Symmetry breaking can lead to an even larger absolute band gap when another rod is placed in the center of the honeycomb cell,¹³ which is called hetero-binary PhC structure in this letter. The hetero-binary structure could also provide a profound modulation in the propagation of photons such as defect engineering¹⁴ and filters.¹⁵

Generally, three laser beams form a 2D interference pattern. In this letter, by introducing one or three auxiliary beams separately, we create 2D hetero-binary and honeycomb PhC microstructures by holographic lithography. To avoid the alignment complexity and inaccuracies due to differences in the optical path length and angles among the interfering beams as well as vibrational instabilities in the optical setup, we applied single refracting prism holographic lithography technique, which was described in our previous work.^{7,16}

The projection of the specially designed prism for this letter, which is a critical component in the optical setup, is schematically shown in Fig. 1. The top and bottom surfaces are hexagonal and surrounded by six symmetrically separated beams.

FIG. 1. Projection of the specially designed prism along the optical axis. The top (smaller) and bottom (larger) surfaces are hexagonal, being surrounded by six symmetrical side-refracting surfaces, B_1 - B_6 . Several types of structures can be obtained by choosing different groups of beams. Details about beam combinations for this work are summarized in Table I.

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TABLE I. Selection of laser beams for various structures.

Name of structures	Selected beams
Hexagonal structure	$B_1+B_2+B_3$
Hetero-binary structure	$B_1+B_2+B_3+B_4$
Honeycomb structure	$B_1+B_2+B_3+B_4+B_5+B_6$

rated side-refracting surfaces. Several types of structures can be obtained by choosing different combinations of the beams. Table I summarizes those used in the present study. Intensity of the independent beams can be controlled by polymer polarizers sitting in the front of the refracting prism mounted on the homemade mask.

The samples were fabricated in a standard holographic lithography procedure, as described previously.^{16(a)} A Nd:YVO4 continuous wave laser at 532 nm was used as the irradiation light source to expose the negative photoresist SU-8 (from Shell) film with 2 wt. % Irgacure 261 (from Ciba. Co.) as cationic photoinitiator. After being exposed to the interference pattern, the polymerization process was completed by a postbake. Those unpolymerized regions were washed away first by propyleneglycolmethylether acetate and then cleaned with acetone, leaving behind a copy of the interference pattern permanently embedded in the polymer film. Fabricated results were observed by scanning electron microscope (SEM) (JEOL 6300F).

The intensity profile of the holography pattern from interference of N beams is given by

$$I(\mathbf{r}) \propto \left| \sum_{n=1}^N \mathbf{E}_n e^{i(\mathbf{k}_n \cdot \mathbf{r} - \omega t)} \right|^2 = \sum_{n,m=1}^N a_{nm} e^{i(\mathbf{G}_{nm} \cdot \mathbf{r})}.$$

The differences of the wave vectors for the incident plane waves, $\mathbf{G}_{nm} = \mathbf{k}_n - \mathbf{k}_m$, can be regarded as the reciprocal lattice vectors. The pattern displays maxima at locations satisfying Bragg diffraction condition: $\mathbf{G}_{nm} \cdot \mathbf{r} = 2q\pi$ (q is an integer).

$N=3$ is sufficient to create a 2D lattice. For example, $\mathbf{k}_1 = (2\pi/\lambda)(1, 0, 0)$, $\mathbf{k}_2 = (2\pi/\lambda)(\cos(2\pi/3), \sin(2\pi/3), 0)$, and $\mathbf{k}_3 = (2\pi/\lambda)(\cos(4\pi/3), \sin(4\pi/3), 0)$ form a regular hexagonal lattice with $\{\mathbf{G}_1 = [(2\pi/\lambda)(3/2, \sqrt{3}/2, 0)], \mathbf{G}_2 = [(2\pi/\lambda)(\sqrt{3}, 0, 0)]\}$ as the reciprocal basis vectors. The beam layout is illustrated in the inset between Figs. 2(a) and 2(b). This beam configuration can be realized by choosing three of the refracted beams (B_1, B_2, B_3) from the prism in Fig. 1, which are the basic beams for all the beam arrangements in this letter. The calculated intensity distribution and SEM image for the fabricated structure are demonstrated in Figs. 2(a) and 2(b), respectively.

By introducing an auxiliary beam, $\mathbf{k}_4 = (2\pi/\lambda) \times (\cos(1\pi/3), \sin(1\pi/3), 0)$ into the above basic beam arrangement, as shown in the inset between Figs. 2(c) and 2(d), the size of the center rod in the hexagonal lattice is able to be modulated. From Fig. 2(c), it can be seen that there is a size contrast, i.e., the size difference between the rod in the center of the hexagonal structure and its six closest neighbors, which is called hetero-binary structure here. This structure has been realized by picking up four refracted beams (B_1, B_2, B_3, B_4) in Fig. 1. The fabricated result is illustrated in Fig. 2(d), where the rod in the center is smaller than its six closest neighbors, being consistent with the simulation result. In Figs. 2(b) and 2(d), some remaining resist bridges between

FIG. 2. Calculated intensity distributions on the left hand side and corresponding SEM images for the surface morphology on the right hand side. The insets in the middle show the beam configurations. (a) (b) Regular hexagonal structure from three symmetrically separated beams ($B_1+B_2+B_3$) at the same intensity. (c) (d) Hetero-binary structures by introducing an auxiliary beam B_4 into $B_1+B_2+B_3$. The intensity contrast between the auxiliary beam and basic beams is $I_{B4}/I_{Ba}=52\%$. The size contrast between the central rod and its six closest neighbors is clearly observed. (e) (f) Hetero-binary structures from different intensity contrast ($I_{B4}/I_{Ba}=26\%$). It can be seen that the size contrast between the center rod and side rods is reduced.

neighbor rods are visible, which were explained as from a kind of chemical self-assembling process due to surface tension occurring at the sample drying stage.^{11(d)}

The size contrast between the central rod and its six neighbors can be tuned by the intensity contrast between the auxiliary beam and three basic beams. As exhibited in Fig. 2(c), when the intensity contrast $I_{B4}/I_{Ba}=52\%$ (in this letter, three basic beams, denoted as I_{Ba} , are intensity equivalent during the fabrication, i.e., $I_{B1}=I_{B2}=I_{B3}$), the size contrast is very obvious. By decreasing the intensity contrast to 26%, the size contrast is reduced, i.e., the size of the center rod is closer to that of its neighbors, as shown in Fig. 2(e). The fabricated result in Fig. 2(f) matches with that from the simulation.

Auxiliary beams can be chosen more flexibly. By adding three auxiliary beams, $\mathbf{k}_4 = (2\pi/\lambda)(\cos(1\pi/3), \sin(1\pi/3), 0)$, $\mathbf{k}_5 = (2\pi/\lambda)(\cos(3\pi/3), \sin(3\pi/3), 0)$, $\mathbf{k}_6 = (2\pi/\lambda)(\cos(5\pi/3), \sin(5\pi/3), 0)$ with same intensity, the honeycomb structure can be created. Six beams, $B_1+B_2+B_3+B_4+B_5+B_6$, were used to yield this structure. Beam configuration is shown in the inset of Fig. 3. The honeycomb structures are clearly exhibited in Fig. 3, both from simulation and fabrication.

All the above beam configurations are based on the fact that three basic beams have equal intensity, thus the rod shape is nearly round. In fact, the shape of the rod is tunable

FIG. 3. Calculated intensity distribution for the honeycomb structure in (a) and SEM image in (b). Honeycomb structure is realized by introducing three symmetrically separated auxiliary beams between three basic beams. All the six beams have the same intensity.

in the same lattice structure by adjusting the intensity ratio of these three basic beams. Also by modifying the phase and polarization of the independent beams, different heterobinary or hetero-ternary structures are obtainable. Furthermore, by controlling the exposure intensity, very different configurations such as holes and rods can be switched (these are not shown here). Therefore, by introducing auxiliary beams, holographic lithography could become a very flexible and useful technique to obtain heterostructures or compound structures other than 14 Bravais structures. This method is also possible to be extended to fabricate 3D heterostructures by adding more beams in the vertical direction,¹⁷ which is very difficult by other techniques such as e-beam lithography or self-assembly.

To summarize, by introducing one or three auxiliary beams separately, we have obtained hetero-binary and honeycomb PhC structures with a specially designed prism having six side-refracting surfaces. The size contrast between the central rod and its six closest neighbors can be tuned by adjusting the intensity contrast between the auxiliary beam and basic beams. More hetero-microstructures could be realized by designing the beam configuration, phase, polarization and intensity ratio between different beams properly.

The authors gratefully acknowledge Professor Wing Yim Tam and Professor Che Ting Chan for useful discussions. This work is partially supported by the Research Grant Council (RGC) of Hong Kong by the Grant No. N_HKUST610/02. The author G.P.W. acknowledges the financial support from the Program for New Century Excellent Talents in University (04-0678) and the National Natural Science Foundation of China (Grant No. 60278018).

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