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# Realization of multi-paired photonic crystals by the multiple-exposure nanosphere lithography process

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Arrays of two-dimensional multi-paired photonic crystals (PCs) have been fabricated by a multiple-exposure nanosphere lithography (MENSL) method utilizing a self-assembled nanosphere as a lens mask and an expanded He–Cd laser. The nanospheres were self-assembled on a photoresist. The masked PR was then multi-exposed with changing rotation angle ($\theta$) and tilt angle ($\gamma$). Scanning electron microscopy reveals that MENSL is a useful tool for fabricating multi-paired PCs with various lattice structures.


*Keywords:* MENSL; Photonic crystals; Multi-paired photonic crystals; Nanosphere lithography

Photonic crystals (PCs) are periodic dielectric material arrays characterized by photonic bandgaps (PBGs) or electromagnetic stop bands, which have a functional similarity to the electronic energy gap of semiconductor crystals. Thus, PCs can prohibit the propagation of a certain frequency range of light within PBGs [1,2]. In the same manner that semiconductors play a pivotal role in the electronics industry, PCs are expected to be employed in many applications in opto-electronics and optical communications [3,4].

PCs with one- (1-D), two- (2-D) and three-dimensional (3-D) periodic dielectric lattices can be designed to possess a proper PBG. Naturally, the ideal 3-D PCs exhibit a perfect PBG, in which photon propagation is completely prohibited for arbitrary polarization in any direction (solid angle $\Omega = 4\pi$). Although 1-D and 2-D PCs do not possess a perfect PBG, they can be realized with simple planar deposition techniques or conventional lithographies. In particular, 1-D and 2-D PCs are very receptive to incorporation of the defect state in the PBG [5,6], required for applications of the PC to various functional devices such as selective transmission filters [7], optical waveguides [8], microcavities [9], etc. In addition, it has been reported that a well-designed 1-D PC can totally reflect arbitrary polarized photons at all incident angles with the omnidirectional (omni-) PBG of $\Omega = 2\pi$ [10]. Similarly, 2-D PCs could be considered to have an omni-PBG in the crystal plane perpendicular to the PC columns.

A number of researchers have investigated optimal conditions for achieving wide-range omni-PBGs using various 1-D PC structures based on periodic or aperiodic sequences [11–14]. In a previous paper, we proposed that the omni-PBG of 1-D PC can be enlarged by including multiple-periodic structures, and demonstrated theoretically and experimentally that the normalized frequency range for the omni-PBG in double-periodic PC can be enhanced approximately 2-fold compared with a single PC [14]. Moreover, the PBG characteristics of the 1-D PC are determined by the design rule of the period and filling factor [5]. In similar manner, the PBG characteristics in the 2-D PC strongly depend on its primitive lattice structure. Many researches of PBG engineering have focused on the PC array with a square or triangle-type lattice that is the representative primitive cell structure capable of being realized with relative ease [15]. The lattice points in the conventional 2-D PCs consist of cylindrical columns with equal sizes and shapes, which could be considered as atomic sites in an elemental semiconductor. If a 2-D PC with complex lattices, instead of a simple triangle lattice, could be designed and realized, this might prove to be a fruitful avenue of research in the field of PBG engineering and towards subsequent applications. The aim of the present work is the realization of multi-paired PC arrays with complex lattices using the multiple-exposure nanosphere lithography (MENSL) method. Although the proposed multi-paired

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PCs are based on the triangle lattice, its one lattice point consists of twin, triple or quadruple points. In addition, it is possible to fabricate the PCs with two interpenetrating triangle sublattices as in a binary compound semiconductor.

2-D PCs can be fabricated using several methods, including focused-ion beam lithography, nano-imprint lithography and e-beam lithography. However, many disadvantages have been reported with the above methods, including damage to the substrate, long processing times (low yield) and high cost. In a previous paper, the authors reported that dot- and anti-dot-type 2-D PC arrays of a large area can be fabricated by a double holographic lithography (DHL) and in particular, their primitive lattice structures can be controlled from a triangle to square via a change in the rotation angle of the double beams [16]. In addition, the multiple-exposure holographic lithography (MEHL) modification of DHL method was employed to fabricate 2-D photonic quasi-crystals (PQCs) [17], which have an excellent rotational symmetry of their morphological characteristics. MEHL appears to be a highly suitable technique to realize 2-D P(Q)Cs without any damage to the substrate, with high throughput over a large area, and at a low cost. However, it is difficult to fabricate the multi-paired PCs proposed in this work. The MENSL method, which is a modification of nanosphere lithography (NSL), utilizes a self-assembled nanosphere as a lens mask and an expanded laser as multiple-exposure source.

A nanosphere was fabricated with a styrene monomer and methacrylic acid (MAA) by emulsion (or dispersion) polymerization. The *p*-type Si (1 0 0) wafer was used as a substrate after cleaning with acetone, trichloroethylene, isopropyl alcohol and deionized water. Then, a diluted polymer resist (PR) DMI-150 with a propylene glycol monomethyl ether acetate (PGMEA) was spin-coated onto the prepared substrate and prebaked at 80 °C for 60 s [18]. The measured thickness of the PR layer was 0.4–1.2 μm. The fabricated nanosphere was self-assembled onto the PR-coated Si substrate by a stepped spin-coating method.

The nanosphere was approximately 1.0 μm in size under all conditions, and the shape was a perfect sphere at 10% MAA. The nanospheres are generally polymerized in the hydrophobic portion of the inner surfactant micelle by an emulsion (or dispersion) polymerization process; accordingly, the outer surfaces of the nanospheres revealed a hydrophilic property. Thus, the PR substrate was simply treated by dipping the PR samples into a developer solution for a hydrophilic surface, and the nanospheres are well ordered on the substrates. A scanning electron microscopy (SEM) image of the self-assembled nanosphere coated onto the PR substrate is shown in Figure 1a, and a clear monolayer of nanospheres with a triangle lattice can be seen to have formed.

The patterning was optically exposed onto the sample position shown in Figure 1a using an expanded laser exposure set-up. A single laser beam (He–Cd laser, 442 nm, $h\nu = 2.80$ eV) was utilized as a light source for the exposure system. The beam was routed by two dielectric mirrors and expanded to a diameter of ~100 mm using a beam expander consisting of an objective lens (numerical aperture of 0.65), a pinhole (diameter of

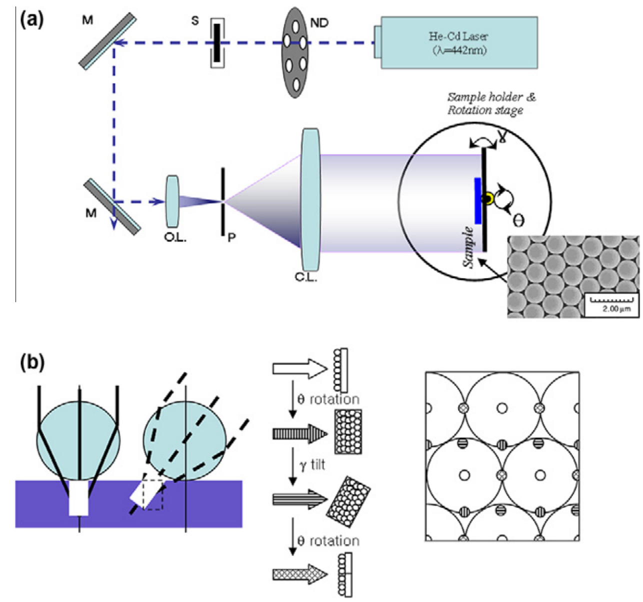


**Figure 1.** (a) Schematic diagram of the rotation and tilt multiple-exposure system using the He–Cd laser: M, mirror; S, shutter; ND, neutral density filter; OL, objective lens; P, pinhole; CL, collimating lens; Sample, SEM images of the self-assembled nanosphere. The intensities of the He–Cd laser measured immediately in front of the sample were 6.5–15.0 mW $cm^{-2}$. (b) The multiple-exposure mechanism in the self-assembled nanosphere sample: solid line, exposing ray before the $\theta$ rotation; dashed line, after the $\theta$ rotation. The expected patterns for MENSL.

10 μm) and collimating lens, as detailed in Figure 1a. The He–Cd laser, 442 nm and 120 mW, was expanded to 70 mm of the beam diameter. The beam intensity (irradiance) immediately in front of the sample, measured using an optical power meter, ranged from 6.5 to 15.0 mW $cm^{-2}$. The sample holder and rotation stage enabled simple adjustment of the rotation and tilt angles ($\theta$, $\gamma$), allowing the incident angle to be easily changed and precisely controlled. Figure 1b shows the mechanism of the multi-exposure and photosensitization for the MENSL. The first exposure shown in Figure 1b was conducted to form a 2-D PC array (periodic dot) in the PR. The PR sample was rotated to the $\theta$ angle and the second exposure was performed. The nanosphere was formed by a microcircular lens. Figure 1b shows a multiple-exposure with the sample rotation and tilt. The multi-exposed sample was then developed using tetramethyl ammonium hydroxide (TMAH 2.38%) at room temperature. All images of the developed pattern were observed by SEM.

Figure 2a–c show the SEM images of 2-D PC pattern arrays fabricated in the PR via the process of the single-exposure NSL and development, where the exposure times for Figure 2a–c were 5, 10 and 15 s, respectively. The 2-D PCs achieved by NSL with different filling factors can be fabricated by controlling the exposure time: increasing the exposure time leads to an expansion in the size of the patterned dot. In this case, the exposure time does not affect the depth of the patterned dot; it was simply affected by focusing the depth of the nanosphere. Accordingly, the PR thickness of the NSL and MENSL needed to be controlled. Figure 2d shows a SEM image of a patterned side view onto a thicker PR (diluted PR