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[DOI: 10.1117/1.OE.53.6.069804]

Publisher's note: The introduction of this paper [*Opt. Eng.* **52**(9), 095103 (Sep. 2013)] has been revised by the authors to include two additional references and explanatory text. This erratum includes the revised introduction and the updated references. The full published article was updated online to include this new material on 26 June 2014.

1 Introduction (revised)

Photonic crystals (PhCs) are artificial crystals specially made for applications¹ in visible and near-infrared spectral regions. Different approaches have been used to fabricate PhCs, e.g., electron-beam lithography,² selective oxidation,³ holography,⁴ crystal building up,⁵ self-organization,⁶ etc. Among these approaches, holographic lithography (HL)⁷ attracts intense attention since multibeam interference can form almost all basic Bravais lattices⁸ and the periodic lattice patterns can be fabricated into photosensitive materials quickly with low cost. In addition, HL is an appealing optical technique to create nanopatterns over wafer-scale dimensions in a high throughput way without electron beams, ion beams, prefabricated masks, prepatterned wafers, or atomic layer deposition. In the application regime of surface enhanced Raman spectroscopy (SERS) substrate, the periodic nanopatterns could be filled with Au or silver films or particles.⁹ Because of the ability to support surface plasmonic polarization or localized surface plasmonic resonance, the nanopatterns can be used as a Raman substrate. Meanwhile, the ordering degree of nanopatterns can affect the amplitude of enhanced coupling electric field when adding an electric field on the array surface, thereby increasing the intensity of Raman scattering signals when the lattice surface is adhered by the molecules to be detected.¹⁰ The photonic array with an improved ordering degree and the smaller period obtained in our work could produce an ideal candidate as a Raman substrate.

Here, we report a compact HL system to fabricate large-scale photonic phased array with four beams interference. The four-beam interference geometry can not only effectively fabricate the 3-D photonic crystals structure reported by Divliansky et al.,¹¹ but also be a method to determine the translational symmetry and lattice constant as well as the contrast of the interference pattern by modulating the wave vector distribution, real amplitudes, polarizations, and relative phase difference of four beams, respectively.¹ In this paper, the tuned pattern properties (especially the relative phase modulation of the four-beam interference) and

controlled thickness of the photoresist were combined to obtain a 2-D large-scale photonic array for an ideal Raman substrate. The geometry of four beams interference is based on the geometry proposed by Campbell et al.,¹² with one beam in the center and three outer beams symmetrically around the central one, however, the four beams are obtained using a specially designed holographic optical element (HOE) in our system. MATLAB numerical simulations have shown that the phase modulation between four beams could redistribute the lattice array. Experimental results have verified that the phase modulation and the nonlinear photoresist response have combined actions on the change trend of photonic array in the photoresist. Because of the improved ordering degree and enhanced coupling field, the large-scale photonic phased array with a smaller period could be an ideal candidate as a Raman substrate.

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