Nanofabrication with Laser Holographic Lithography for Nanophotonic Structures

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Nanoscale periodic structures are gaining strong interests and finding more applications with their unique nanophotonic properties arising from photonic bandgap, plasmonic, and subwavelength structures. Integrating nanofabrication technologies from three dimensional laser holographic lithography, atomic layer deposition, nano machining, and nano desposition provides unique solutions by defining volumetric or surface nano structures, backfilling them with the active or passive materials of interest, and selectively creating functional aperiodic regions. We have demonstrated the fabrication of 3D photonic crystal templates in SU-8 on glass and Si substrates using laser holographic lithography. The voids in the photonic structures are filled with tungsten metal using atomic layer deposition. We have also investigated the feasibility of applying the laser lithograhically defined 2D structures to produce periodic nanoscale structures on a Si wafer using positive and negative photoresists. We have created functional defects in the template of periodic structures by focused ion beam milling to remove individual pillars or by direct write ion or e-beam deposition with gas injection system to fill holes or voids in the photonic structures. These device structures can lead to many photonic applications including lighting, display, sensor, and optical information processing.

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1. Introduction

Nanoscale periodic structures are gaining strong interests and finding more applications with their unique properties arising from photonic bandgap, plasmonic, and subwavelength structures. Structures formed in the wavelength scales of interest can enable disruptive light modulation solutions in display and photonics where these device scales were not accessible before with limited fabrication capability. Conventional approach is based on the advanced very large scale integration (VLSI) fabrication with deep ultraviolet (UV) lithography that requires many steps for an extended time. Upcoming competitive approaches include nanoimprint process, focused ion beam or e-beam machining, self-assembly of nanoscale beads, direct laser writing with near field scanning for two-photon absorption process [1-5]. They are still more expensive or time consuming or lacking precision to align three dimensional structures in the visible wavelength scale of interests. Three dimensional laser holographic lithography is enabling technology for the rapid prototyping of two or three dimensional nanoscale photonic structures [6]. Integrating Hewlett-Packard Company's nanofabrication technologies from three dimensional holographic lithography, atomic layer deposition (ALD), nano machining, and nano desposition provides unique solutions by defining volumetric or surface nano structures, backfilling them with the active or passive materials of interest, and selectively creating functional aperiodic regions. These device structures can lead to many photonic applications including lighting, display, sensor, and optical information processing.

2. Laser Holographic Lithography

Flexible and rapid prototyping of nanoscale volumetric structures are achieved by integrating laser holographic lithography and atomic layer deposition. Laser holographic lithography provides 1) flexibility, the ability to adjust lattice spacing, lattice structure, and lattice size by controlling the angle of incidence, the exposure fluence, and developing time, 2) resolution, the ability to provide down to ~10 nm resolution and subwavelength feature size, 3) rapid prototyping, the ability to fabricate nanoscale volumetric periodic structures with a single exposure of ~10 ns, and 4) cost advantage, the ability to achieve all the above at significantly lower cost than competitive technologies.

Principles of laser holographic lithography is using the optical interference of highly coherent laser beam to introduce three dimensional intensity profile, thus exposing SU8 photoresist in periodic patterns. Final periodic lattice structure and the optical set up of incoming beams have reciprocal relationship. A holographic intensity grating produced by laser interference has a discrete Fourier spectrum where the intensity distribution is given by the equation (1) [7].

$$I(r) \propto \sum_{n,m=1}^{4} a_{nm} e^{i\bar{G}_{nm} \bullet \bar{r}} \qquad (1)$$

Where G_{nm} represents the reciprocal lattice vector ($G_{nm} = k_n - k_m$), k_i is the wave vector, and r is the position vector. While the wave vectors (wavelength and angle) of the Fourier components determine the translational symmetry of the structures, the magnitude (laser beam intensity) of the Fourier components determine the basis of lattice point in each unit cell (fill-ratio). 2 beam, 3 beam, and 4 beam interference can lead to submicron structures in 1D, 2D, and 3D respectively.

In our laser holography set up, we use single pulse from injection-seeded, frequency tripled Q-switched Nd:YAG laser (wavelength of 355 nm) exposes photoresist with interference patterns. Short duration of laser pulse (8 ns) allows the interference to be unperturbed by photo induced changes in the precursor and the mechanical fluctuation of the optics over time yielding well defined volumetric photonic crystal structure. The detailed information on the laser holography set up can be found elsewhere [6-8].

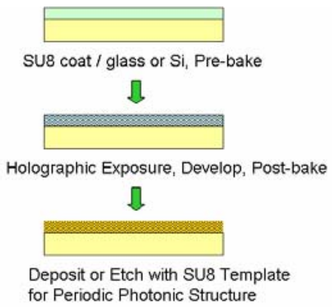


Fig. 1 Process Steps for Laser Nanofabrication.

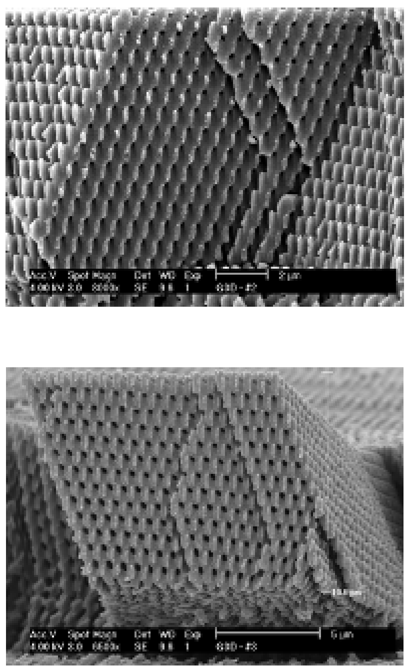


Fig. 2 Cross-sectional SEM of three-dimensional photonic crystal structures.

Figure 1 shows the brief summary of process steps for laser nanofabrication processes. Depending on the type of photonics template to fabricate, negative photoresist of SU8 or positive photoresist of Shipley i123 is exposed with holographic process. SU8 has high functionality with very dense crosslinks, providing high contrast suitable for the high aspect ratio micro or nano mechanical structures [6]. SU8 process involves the cleaning and rinse of glass and Si wafers in organic solvents, pre-bake to remove all the solvent in the SU8, illumination with various dosage to optimize the process, post exposure bake for cationic photopolymerization of the epoxy at a higher temperature than the glass transition temperature, development in ethyl-lactate, followed by rinse and dry process. Flood exposure under homogeneous UV and bake step is applied when necessary to prevent the flowing of SU8 structure.

Figure 2 shows the cross-sectional scanning electron micrograph (SEM) pictures of a three dimensional nano-scale periodic structure fabricated from our process. In ref. [6], the calculated constant-intensity surfaces in four-beam interference patterns show face centered cubic (f.c.c.) pattern with lattice constant of 922 nm (pitch of 652 nm) when the three outer beams are placed with an angle of 38.9^o symmetrically with the central beam. Our result indicates that the photonic crystals of SU8 are formed according to this periodic intensity patterns. The SEM cross-sectional images of cleaved sample clearly indicate the preferential cleavage planes of {111} and direction of <110> for f.c.c. lattice where the plane defines an energy-privileged plane for the easy slip and the direction defines an small distance between partial dislocations forming the wave type transverse slip.

Figure 3 shows the reflectances from SU8 with and without photonic crystal structures. The reflectance is suppressed in the subwavelength regime where wavelengths are smaller than a pitch of the structure. These surface structures form a surface-relief grating in which the grating period is small compared to the wavelength of incident light so that no diffractive orders other than the reflected or transmitted zeroth orders are allowed to propagate. The light sees a homogeneous region where an effective index of refraction continuously varies from the incident medium to the substrate. Due to the fact that SU8 is relatively transparent in the range of wavelength measured, the reflectance curve shows interference fringes. As the refractive index contrast (~1.6 for SU8) is lower than the required condition for photonic bandgap, we have not seen meaningful photonic bandgap effect from the photonic crystals based on SU8 alone.

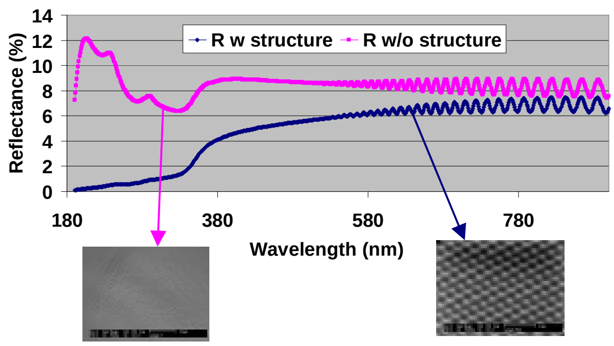


Fig. 3 Reflectances of SU8 surface with and without photonic crystal structure.

3. Atomic Layer Deposition

Atomic layer deposition (ALD) is a chemical vapor thin film deposition method based on sequential, self-limiting surface reactions that provides thin film deposition with atomic layer control [9-13]. Atomic layer deposition provides unique capability for controlled layer-by-layer deposition of films in high-aspect ratio and non-line of sight structures at temperatures below 200 ºC to fill up the pores introduced with holographic lithography without affecting the structural integrity of photopolymer forming photonic crystal template. The hard baked SU8 has a degradation temperature of over 350 ºC allowing the deposition of various materials using atomic layer deposition process. This whole process allows the fabrication of three-dimensional photonic crystals with required optical properties such as higher index contrast or surface plasmon resonance to produce active or passive photonic devices.

In ALD, chemical precursors are pulsed one at a time separated by inert gas purges. The pulse-purge sequence is repeated to grow thin films in a layer-by-layer fashion. Because of the self-limiting nature of ALD, highly conformal films with excellent uniformity, step coverage, and thickness control can be deposited not only on flat surfaces, but also inside trenches and narrow openings. These properties make ALD a superior technique for coating and backfilling the interconnected network of openings in three-dimensional photonic crystals. In our demonstration, we backfill the 3D photonic structures with the atomic layer deposition of tungsten at 150 ºC. For the growth of tungsten with ALD process, WF_6 and B_2H_6 or Si_2H_6 can be used as precursors. Deposition rate is dependent on substrate temperature and ranges from 2 to 5 Angstroms/cycle.

In our experiment, we deposited 40 nm thick tungsten without filling up the voids completely but once the voids are filled completely at lower temperature where SU8 is stable, original polymeric structure can potentially be ashed out at higher temperature and by using plasma ashing or chemical process. SEM analysis of cross-sectional images indicates that thin tungsten layers are formed inside the structure as well. Figure 4 shows the comparison of reflectances from a tungsten thin film and the photonic crystal backfilled with tungsten. In this Figure, reflectance spectra no longer show the interference fringes seen from the structure with SU8 alone. It still shows the characteristic subwavelength response from the nanoscale photonic structure.

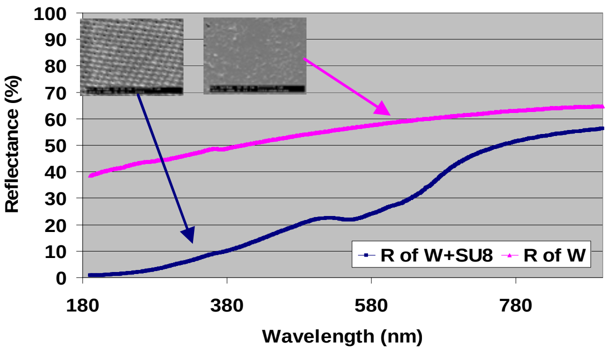
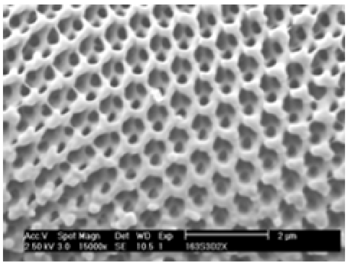


Fig. 4 Comparison of reflectances with and without tungsten deposited on SU8 photonic crystal.

4. Performance of Photonic Crystals

We have demonstrated the photonic bandgap with holographically fabricated SU8 photonic crystal templates and the backfilling of voids with metal on glass and Si substrates for enhanced index contrast. In order to produce three-dimensional interference, a single beam from highly coherent laser is split into four beams and re-converged at the photoresist. The reference beam in the center is circularly polarized while the three side beams are linearly polarized. Fluences of 40 to 100 mJ/cm^2 provide open structures with 30 seconds of developing time in ethyl lactate.

Figure 5 shows the top view of 3D photonic crystal with a reflectance spectrum indicating sharp photonic band edge at ~1 um with full bandgap from ~1.3 um. The photonic bandgap effect is characterized by Fourier Transform Infrared Spectrometer (FTIR). While SU8 based structures also show similar optical response with the weak contrast of dielectric constants, this particular sample has W deposited via ALD to fill up the pores in the optimized photonic crystal sample introduced by holographic lithography.



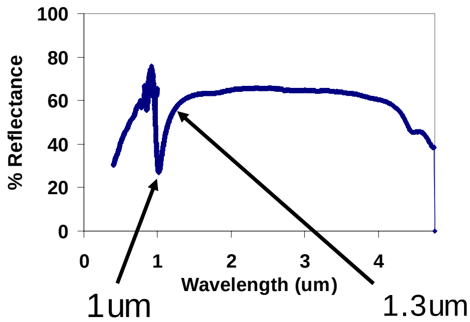
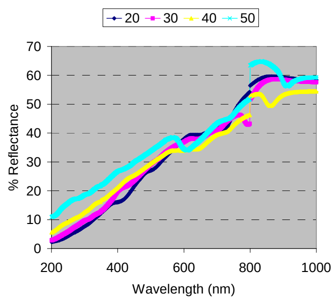
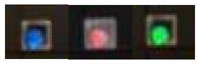


Fig. 5 Top view of 3D photonic crystal in SU8 (above) and a reflectance spectrum showing a photonic bandgap (below).

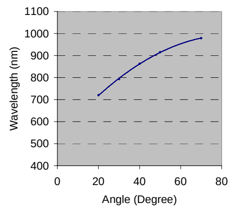
Figure 6 shows unique optical response from a 15 um thick sample, strongly reflecting different colors at different angles. In order to characterize the angular dependence of reflectance, Variable Angular Spectrum Reflectance Accessory for Cary 6000i from Varian is used for the measurement angles varying from 20 degree to 50 degrees. Higher angles broaden the size of beam too large to calibrate properly. Angular measurement shows characteristic resonance modes from reflectance that shifts with angle. The shift in reflectance intensity at 800 nm is due to the change of detector in Cary 6000i and is not relevant to the properties of the crystal. At higher angle, more than one mode appears and this is due to the diffraction by coupling between incident light and the surface modes as discussed in ref. [6] in detail.



(a) Angular reflectance



(b) Angle dependant reflection



(c) Shift of resonance mode

Fig. 6 Angular dependence of reflectance from the tungsten filled SU8 photonic crystal and the shift of resonance modes with angle.

In 4 beam interference, circularly polarized beam at the center repeats and extends the 2 dimensional intensity modulations to 3 dimensional intensity profiles by individually interacting with each side beam. By blocking the center beam, we can produce 2 dimensional triangular lattice photonic crystals. Figure 7 shows the 2 dimensional and 3 dimensional structures fabricated on the SU8 layer of same thickness. While the image on the left indicates the two dimensional structure with a pitch of 360 nm from 3 side beam interference, the image on the right shows the result from full 4 beam interference. With 1 um thick layer, we have about 2 units of 3D structure formed when center beam is applied. This demonstrates the flexibility of laser lithography techniques for rapidly fabricating multi-dimensional structures.

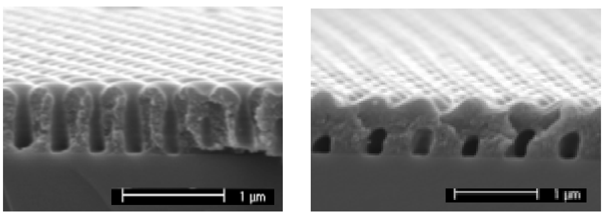


Fig. 7 2D structure (left, pitch of 360 nm) and 3D structure (right, pitch of 600 nm).

5. Laser Lithography and Nanoscale Editing

We have also investigated the feasibility of applying the laser lithograhy to a positive photoresist layer of Shipley i-123 in addition to the negative SU8 photoresist to create 2D structures with a pitch of 360 nm by blocking a center beam. By changing the chemical structure where the high intensity exposure with interference occurs to be more soluble during development, the positive photoresist allows the formation of pillar based structure having a symmetry of three fold axis. Using these 2D structures as etch masks, we have produced periodic nanoscale structures on a Si wafer.

We created functional defects in the template of periodic structures by Focused Ion Beam (FIB) milling to remove individual pillars or features from the photonic structures. We also createrd functional defects by direct write FIB or e-beam Gas Injection System (GIS) deposition to fill holes or voids in the photonic structures. Gas dispense needle is placed 50-100 um from substrate to release organo or organo-metallic carrier gas for deposition or reactive carrier gas for etchant. Beam cracks the gas adsorbed onto the surface enabling either, deposition or etching of metals or dielectrics allowing maskless patterning process.

The defects in the structure can be used to control or guide the light within the photonic crystal structure [14]. Crystal edits of features as small as 10-15 nm are possible. In the examples shown, we have used the ion beam at 30kV and 1pA. A dwell of 20 s is used for milling the SU8, while a dwell of 200 ns is used to deposit the W to fill the structure selectively in the region of interest. Other GIS gases are available for direct write deposition of Pt, Pd, Au, C, TEOS, etc as well as for selective or enhanced etching of C, dielectrics, or metals. This demonstrates the capability to fabricate Application Specific Integrated Photonic Circuits formed of photonic crystal waveguides or photonic crystal resonant cavities for a specific wavelength.

Figures 8 and 9 show the original template made on positive and negative photoresist, etched pattern on Si with generic Si semiconductor etching process using laser lithographically defined templates, and nano machining and nano deposition to define defects respectively. Positive photoresist has narrower range of optimized fluence so the area of uniform surface structure is smaller when exposed with a beam of Gaussian profile. Compared to cross-connected holed structure with negative photoresist, positive photoresists with individual pillars do not result in uniform structure in Si etching process requiring further study as shown in Figure 8.

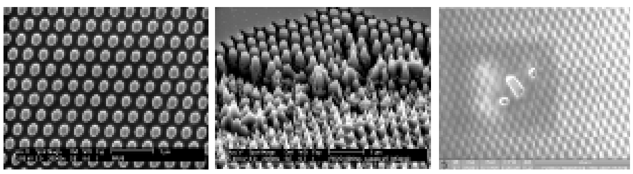


Fig. 8. SEM images of 2D structures in positive photoresist (left), nano pillars and spires etched on Si (center), and nano FIB machining to create functional defects (right).

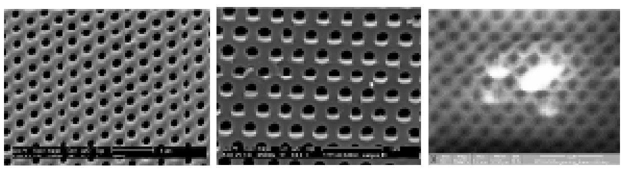


Fig. 9. SEM images of 2D structures in negative photoresist (left), nano holes etched on Si (center), and nano FIB GIS depositied W to selectively create functional defects (right).

6. Summary

Nanophotonics is continuously adding the ability to produce and detect light with nanoscale structures of various materials. It also provides the ability to guide and modulate optical signals with Si based photonic crystals that allows the monolithic integration of the photonic devices on Si. Nanoscale periodic photonic structures with customized defects are crucial to enable these applications and laser holographic lithography combined with other nanofabrication capability provides low cost, high throughput, precision, and flexible fabrication processes. For this purpose, we have demonstrated laser based nanofabrication capability in this paper by integrating nanofabrication technologies from three dimensional laser holographic lithography, atomic layer deposition, nano machining, and nano desposition processes using positive and negative photoresists to create periodic nanoscale structures of opposite polarity and use them as templates for further downstream processing.

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