Continuous Patterning of Three-Dimensional Periodic Nanostructures using Roll-to-Roll System

I-Te Chen,* Xu A. Zhang, Joong-Hee Min, and Chih-Hao Chang

Department of Mechanical and Aerospace Engineering, North Carolina State University Raleigh, NC 27695, USA

Periodic three-dimensional (3D) nanostructures show many unique properties that cannot be observed in macroscale materials, and can have applications in photonics, acoustics, and mechanics [1-3]. One effective method to fabricate 3D nanostructures is phase-shift lithography using lithographically defined [1-2] or colloidal particles as near-field optical masks [4-5]. In this approach, under normal illumination the photoresist below the masks will be exposed by complex 3D intensity distribution as described by the Talbot effect. However, fabricating 3D structures in large area by this method is still arduous since it requires high quality phase masks or colloidal assemblies, and previous work is restricted to laboratory demonstration scale. The goal of continuously printing 3D nanostructures over large areas in a scalable fashion remains challenging.

In this work, we introduce a hybrid assembly and lithography system by employing continuous automated Langmuir-Blodgett assembly for colloidal particles coating and near-field lithography for 3D patterning. The proposed fabrication approach, illustrated in Figure 1, allows for continuous printing of periodic 3D nanostructures on a variety of substrates. Before loaded on the system, a 4-inch silicon wafer was coated with antireflection coating and positive photoresist (Sumitomo PFi-88). Monodispersed silica nanoparticles are first injected on the surface of the fluid, forming a monolayer of hexagonal close-packed assembly on the surface of a photoresist layer through continuous Langmuir-Blodgett assembly in the "Colloidal Assembly Module." Upon drying, the whole stack is then exposed by normal illumination from a UV laser, as shown in Figure 1(b) and (c), in the "Lithography Module." The relative ratio of the particle to incent wavelength can then determine the patterned 3D geometry. For a 1000-nm-thick Sumitomo PFi-88 positive photoresist, 500 nm diameter nanospheres are used under 325 nm HeCd laser exposure with the dose of 110 mJ/cm². Negative SU-8 resist can also be used, which requires an exposed SU-8 buffer layer to increase adhesion between ARC and SU-8. Necessary post exposure baking might be included based on the selected photoresist. Subsequent cleaning and development process remove the nanospheres and produce the periodic 3D nanostructures in the photoresist.

Scanning electron micrographs (SEM) image and photograph of initial results are shown in Figure 2(a) and (b). Under proper coating rate, which is 1.5 mm/sec in this experiment, a 2D monolayer of nanospheres can be arranged in a hexagonal close-packed geometry on top of a thick photoresist layer. The nanosphere diameters we used in this experiment is 500 nm. The reflection spectra of the fabricated structures will be characterized to demonstrate applications in photonics. We will present detailed analysis on the quality of the patterned 3D structure, pattern area, and limitations for a wide range of particle materials and sizes. The proposed process is an efficient method which may make the continuous, roll-to-roll manufacture of 3D nanostructures available. We will keep integrating other modules, like the lithography system and nanospheres recycling system, to the current system to make the whole process more convenient and inexpensive.

^{*} ichen5@ncsu.edu



Figure 1. Fabrication process for continuous 3D nanostructure manufacture (a) Assemble a monolayer of nanoshperes on the photoresist, (b) dry the coating, (c) UV exposure.



Figure 2. Fabrication result (a) a monolayer of 500 nm diameter nanoshperes covers whole 4-inch silicon wafer region, (b) SEM image of a monolayer of 500 nm diameter nanoshperes on the surface photoresist, (c) SEM image of the hexagonal close-packed assembly pattern of positive photoresist, (d) cross-sectional SEM image of the 3D patterned positive photoresist.

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