# Three-Dimensional Nanolithography Using Colloidal Nanospheres with Oblique and Multiple Exposures

***Xu A. Zhang**, Zhiyuan Xu, Bin Dai, and Chih-Hao Chang***

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

Colloidal assembly is a simple, low-cost, and high-throughput nanofabrication approach. In these techniques, monodispersed nanospheres and their assemblies are utilized as physical masks or templates for additive and subtractive nanopatterning processes [1-3]. In recent work, we demonstrated that the Talbot effect from periodic nanosphere assemblies under light illumination can be exploited to fabricate complex 3D periodic nanostructures, extending colloidal assembly to the photolithography regime [4]. Light scattering from single nanospheres can also create interesting hollow-shell "nano-volcanoes" using a single normal-incident exposure, adding additional flexibilities to colloidal lithography [5]. In this work, we explore the fabrication of more complex, unconventional 3D geometries using light scattering from single particles under oblique light incidence and multiple exposures.

The proposed fabrication approach and associated scanning electron microscope (SEM) images of the fabricated 3D nanostructures are shown in Figure 1. We explored the sequential single, double, triple and quadruple exposures at oblique incidence on single polystyrene nanospheres assembled on positive photoresist surfaces, as shown in Figure 1(a)-(d). The sequential exposure beams were arranged symmetrically with user-defined incident angle $\theta$. Side and top views of the fabricated 3D nanostructure are shown below the diagram of each exposure configuration. For single exposure in Figure (e) and (i), as a result of the combined effects of light scattering and refraction at the air-resist interface, the hollow-shell structure has mirror symmetry and is tilted toward the incident direction, adding directionality to the structure. For double exposures in Figure 1(f) and (j), the structure is a doublet of the single-exposure case, resulting in the same symmetry as the double beam arrangement. The fabrication results for triple and quadruple exposures are similar, but with more structure complexity and symmetry. Note that the multiple exposures were applied sequentially rather than simultaneously, resulting in the summation of the multiple exposure intensity. More complex exposure is possible using simultaneous multi-beam exposures, which can result in the coherent interference pattern.

In this work, we investigate the three-dimensional nanolithography using particle scattering from multiple oblique light exposures. Preliminary results have demonstrated the feasibility of this technique and its capability of fabricating more complex 3D nanostructures with high symmetry and directionality. The light scattering patterns under these configurations will be modeled and studied, and the design of the structure geometries by varying the particle size, incident angle, and polarization will be studied. Possible applications of these nanostructures in micro/nanofluidics and light trapping will be explored.

*xzhang27@ncsu.edu

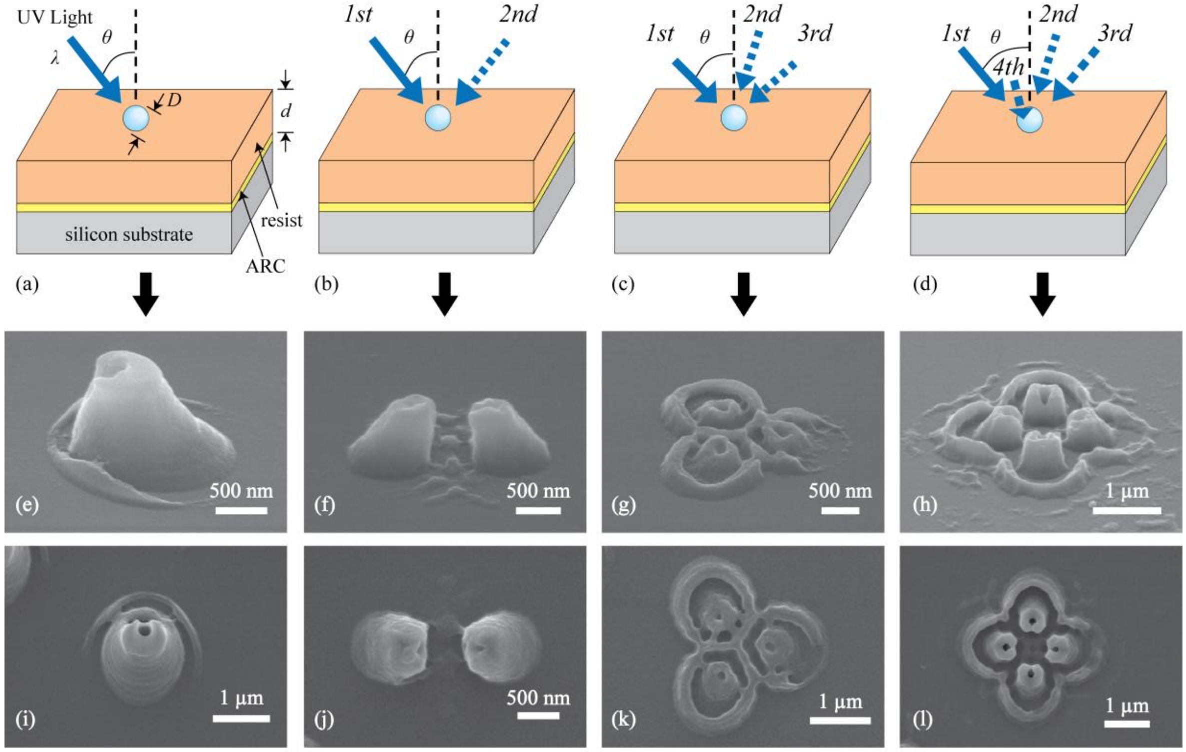


**Figure 1.** Diagrams of the nanolithography setups using a single nanosphere for (a) single, (b) double, (c) triple and (d) quadruple exposures arranged symmetrically around the surface normal at incident angles of $\theta$. SEM images of (e)-(h) side and (i)-(l) top views of the 3D nanostructures fabricated, respectively. The fabrication parameters are: (e, i) $\lambda$=325 nm, $D$=390 nm, expsoure dose=110 mJ/cm$^2$ and $\theta$=45˚; (f, j) $\lambda$=325 nm, $D$=390 nm, expsoure dose=125 mJ/cm$^2$ and $\theta$=45˚; (g, k) $\lambda$=325 nm, $D$=505 nm, expsoure dose=120 mJ/cm$^2$ and $\theta$=45˚; (h, l) $\lambda$=325 nm, $D$=505 nm, expsoure dose=115 mJ/cm$^2$ and $\theta$=45˚.

**References:**

[1] J. C. Hulteen and R. P. Van Duyne, *J. Vac. Sci. Technol. A* 1995, Vol. 13, No. 3 1553-1558.
[2] Moon, J. H. *et. al., Chem. Commun.* 2005, 4107-4109.
[3] Velev, O. D. *et. al*., *Nature* 1999, 401, 548–548.
[4] C.-H. Chang *et. al., Nano Lett.* 2011, 11, 2533-2537.
[5] X. A. Zhang, *et. al.*, *ACS Nano*, 2013, 7(7), 6212-6218.