Fabrication of Silicon Nanostructures with Spatially Gradient Periodicity by Stretched Nanotransfer Printing

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Silicon metasurface can provide a CMOS compatible platform for optical wavefronts modulation with high transmittance efficiency. It inspires many emerging applications, for example, holographic encoding and refractometric sensing, to name a few.¹⁻² Compared to their plasmonic counterparts, silicon metasurfaces composed by silicon nanoresonators are not compromised by the strong absorption loss in metallic nanostructures, which makes them particularly suitable for many optical applications including polarization beam splitting, perfect broadband reflectors, and spontaneous emission control.³ However, Mie-type resonances of silicon nanostructures strongly depend on the nanostructure sizes, orientations and periodicity. The resonance and spectral properties of individual silicon nanostructures therefore need to be finely tuned by theoretically and experimentally optimizing using a lot of structures with different geometric properties using conventional nanolithographic methods like e-beam lithography or focused ion beam lithography is costly and time-consuming.

In this research, we have developed a novel nanopatterning technique to obtain silicon nanostructures with spatially gradient periodicity from a nanoimprint mold with homogenous periodic feature. The fabrication process is schematically illustrated in Figure 1. A 500-nm pitch gold nanodisk array is first fabricated by nanoimprint lithography and electrodeposition or e-beam evaporation, then stripped by an adhesive poly(vinyl alcohol) (PVA) film. By tailoring the PVA film to a designed trapezoid shape and mechanically stretch it, a distribution of strain is generated and preserved within the PVA film which also induce a gradient in the periodicity of the gold nanodisk array. After transfer the stretched PVA film to a quartz substrate deposited with a thin layer of silicon, the PVA can be dissolved in water, leaving the gold nanodisks with spatially varying periodicity transferred as a mask for the following reactive ion etching (RIE) process. Finally, the gold nanomask is wet etched to complete the fabrication of silicon nanodisks with spatially gradient periodicity on the quartz substrate. Figure 2 shows the gold nanodisk mask with gradient periodicity transferred to a quartz substrate deposited with 50-nm thick silicon through the proposed method. The PVA film embedded with 500-nm-pitch gold nanodisk array was tailored to a trapezoid shape and then stretched to 200 % of its original length. As expected, periodicity of the transferred gold nanodisks varies with the spatial location from 537 nm to 870 nm.

1 Yang, Y., Kravchenko, I., Briggs, D. and Valentine, J. *Nat. Comm.* **2014**, *5*, 5753. 2 Reineke, B., Sain, B., Zhao, R., Carletti, L., Liu, B., Huang, L., De Angeis C. and Zentgraf, T. *Nano Lett.*, **2019**, 19(9), 6585-6591. 3 Moitra, P., Slovick, B. A., Li, W., Kravchencko, I. I., Briggs, D. P., Krishnamurthy, S. and Valentine, J. *ACS Photonics*, **2015**, 2(6), 692-698.

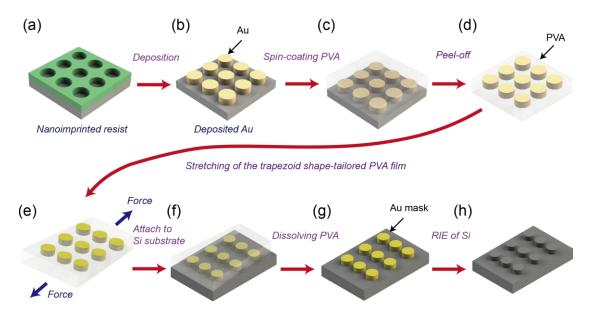


Figure 1. Schematic illustration of the fabrication process.

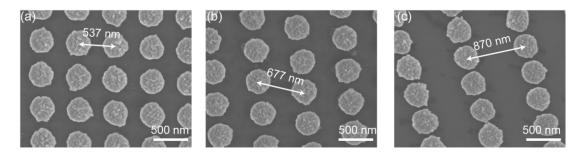


Figure 2. SEM micrographs of the transferred gold nanodisks with varying periodicity of (a) 537 nm, (b) 677 nm and (d) 870 nm on different locations of the sustrate, using a trapezoid PVA transferring film embedded with 500-nm-pitch gold nanodisk array and stretched by 100%.