Supplementary Information

Arrays of Highly Complex Noble Metal Nanostructures using Nanoimprint Lithography in Combination with Liquid-Phase Epitaxy

Eredzhep Menumerov,^a Spencer D. Golze,^a Robert A. Hughes^a and Svetlana Neretina^{*a,b}

^aCollege of Engineering, University of Notre Dame, Notre Dame, Indiana, 46556, USA ^bDepartment of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, Indiana, 46556, United States **E-mail: sneretina@nd.edu*

1. Pneumatic Nanoimprinting Press

Figure S1 shows images of the pneumatic press that was built to facilitate the nanoimprinting process. The function of the press is to apply a pressure of 40 bar to a silicon nanoimprint lithography (NIL) stamp while maintaining the temperature at 140 °C. During this process the stamp-resist-substrate combination rests on the bottom platen as the upper platen is lowered onto its surface. Both platens were precision ground and machined to maximize parallelism. The upper platen moves on four guide rods where bronze bushings provide stability and accommodate a slight angular tilt that allows for the self-alignment of the top platen. The top platen is driven by a double-acting pneumatic cylinder with a 1.5 in. diameter bore where two hydraulic dampers prevent the top platen from arriving to the stamp at excessive speeds. The sample is heated using five stainless steel 90 W cartridge heaters that were inserted into holes drilled into the top and bottom platens. The temperature of each platen was monitored and controlled independently using type J thermocouples and CN7833 temperature controllers (Omega) employing an on/off algorithm. A thermal resistor consisting of a 0.25 in. thick Macor® machinable ceramic stacked on a notched stainless steel plate was inserted between the pinned piston connection and upper platen to slow the rate of thermal conduction so as to prevent the pistons seals from exceeding their temperature tolerance. The device operates in air.



Fig. S1. Images of the pneumatic press that was designed to imprint a moldable polymer resist with a nanopatterned silicon stamp at elevated temperatures and pressures.

2. Seeds Produced as Large-Area Periodic Arrays

The figures below show SEM images and analysis aimed at characterizing the seed arrays that show both large area capabilities as well as some of the limitations of the process.



Fig. S2. SEM image of Au nanostructures formed using nanoimprint lithography in combination with templated dewetting.



Fig. S3. SEM images showing both the large-area capability of the nanofabrication process as well as the missing structures that can develop as the NIL stamp becomes damaged.



Fig. S4. Histograms showing the variations in (a) Au seed diameter and (b) the center-to-center distance between the arrayed seeds.



Fig. S5. SEM image of a periodic array of Au nanostructures where multiple dot defects form at some of the array positions because the deposited Au pedestal thickness was too thin. In this scenario, natural instabilities in the film cause it to divide into multiple islands where each forms a nanostructure.

3. Core-Void-Nanoframes



Fig. S6. (a) Top- and (b) tilted-view SEM images of periodic arrays of the Au@void@Au core-void-nanoframes.