# High-resolution and large-area nanoparticle arrays for model systems in catalysis using EUV lithography techniques

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Catalysts are of vital importance in biological processes and large-scale production of fuel and chemicals. Chemically synthesized model systems for heterogeneous catalysis and electrocatalysis do not feature well-defined size and lack lateral order of the active sites which are important to study surface structure - activity relationships, reaction mechanisms, and reaction kinetics. Surface nanopatterning using top-down methods has, therefore, attracted substantial interest to develop reproducible model systems for catalysis. Replication of realistic catalysts requires sub-10 nm particles with uniformity in diameter, height and shape over a very large area. Conventional lithography techniques are either low-throughput, e.g. electron beam lithography (EBL), or do not enable the required resolution, e.g. laser interference lithography. Extreme ultraviolet interference lithography (EUV-IL) has the throughput and simplicity advantages of interference lithography as well as high resolution due to its short wavelength of 13.5 nm. Since we aim for large area and uniform patterns, we adopted achromatic Talbot lithography (or achromatic spatial frequency multiplication (ASFM)), which has a unique self-healing property, i.e. it is robust to individual defects on the mask. Here, we demonstrate EUV lithography techniques to obtain nanoparticle arrays for catalysis using a novel high-quality transmission mask fabrication strategy with one-step EBL and electroplating Ni.

A fabrication process for masks, performed on thin 100 nm $Si_3N_4$ membranes with high transmission to EUV, using EBL with a 2-D array of holes in Ni over an area of 500×500 $\mu m^2$ has been developed based on ASFM method (Fig. 1). Ni is chosen for its high absorbance to EUV and low stress of electroplated thin films, a requirement because of the fragile nature of membranes. The whole process involves one EBL and Ni electroplating step, significantly simplifying fabrication and increasing the yield of these highly efficient EUV masks. Several masks are fabricated with this approach enabling periodic dot patterns down to 100 nm pitch and minimum feature size as low as 10 nm (Fig. 2). We compare performance of ASFM method with multiple-beam interference lithography by fabricating 4-beam masks with Ni gratings that give hole or dot arrays (Fig. 3).

We have established an effective lithographic approach for fabricating well-defined model systems for catalysis. High-resolution Pt nanoparticle arrays with uniformity over large areas are achieved. Further studies aim for feature sizes well below 10 nm over much larger areas in order to study catalytic systems, in particular Pt nanoparticles and their size effects.

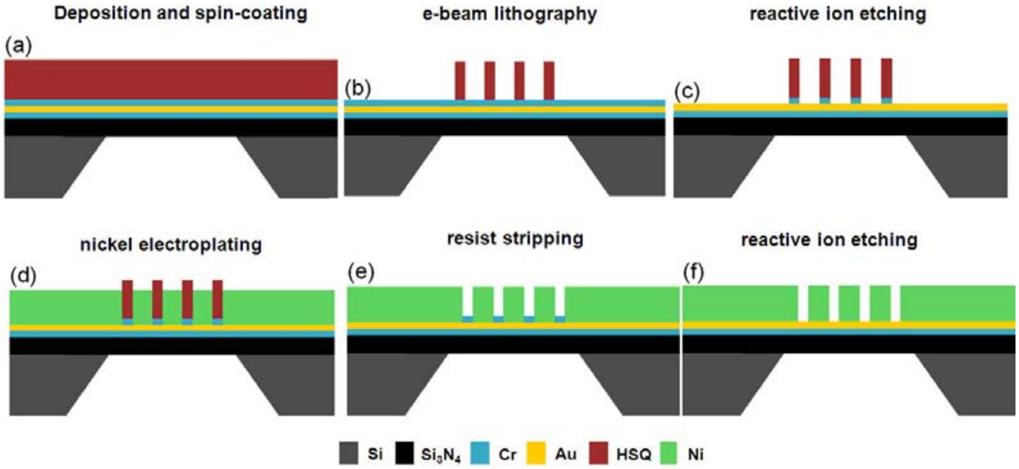

*Figure 1: Schematic of the EUV transmission mask fabrication process flow. (a) $Si_3N_4$ membrane with thermally evaporated Cr/Au/Cr multilayer and spin-coated hydrogen silsesquioxane (HSQ) resist; (b) EBL and development; (c) chlorine reactive ion etching of Cr layer; (d) Ni electroplating; (e) resist removal with hydrofluoric acid; (f) chlorine reactive ion etching of Cr layer*

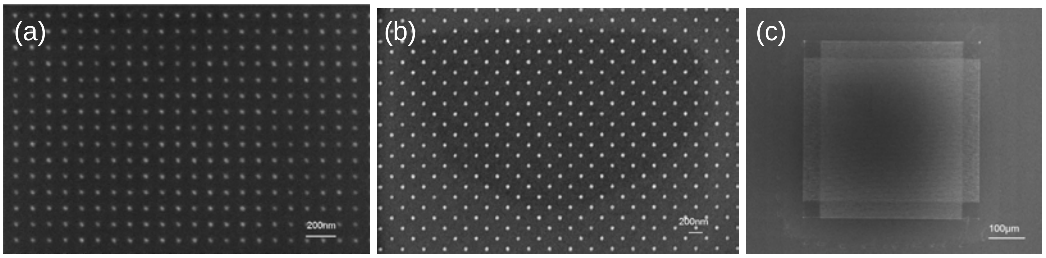

*Figure 2: SEM images of HSQ on Si from EUV exposure using high-resolution larger-area transmission mask based on ASFM method, single EBL step and nickel electroplating (a) Dot-array with feature size 20 nm; period 105 nm on wafer using mask with period 150 nm; (b) Dot-array with period 212 nm using mask with period 300 nm; (c) Exposure using the 500×500 $\mu m^2$ mask gives dot-arrays with uniformity over a large-area.*

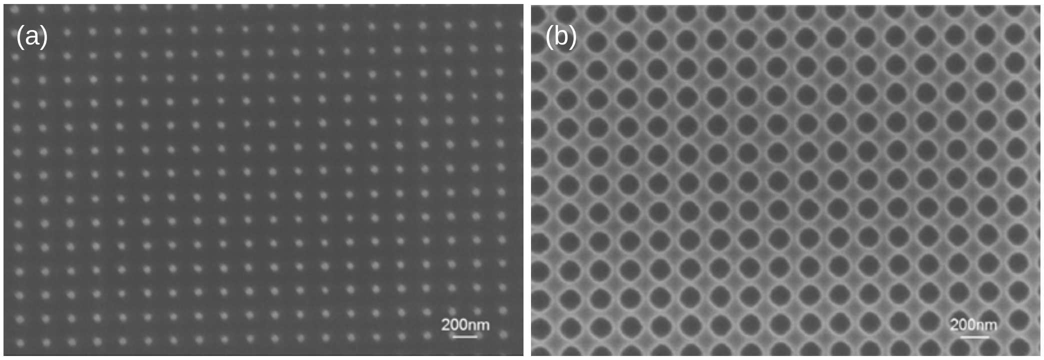

*Figure 3: SEM images of HSQ in Si from exposure using large-area 4-beam mask with 1-step EBL and grating in electroplated Ni (a) Uniform 30 nm dot array with period 200 nm at low dose, and (b) Uniform 175 nm hole-array with period 200 nm at high dose over 400×400 $\mu m^2$ area.*