II-G. Nanoimprint

Master mold
Deform resist
Etching
Fast

Fig. 1. Schematic of nanoimprint lithography process: (a) imprinting using a mold to create a thickness contrast in a resist, and (b) pattern transfer using anisotropic etching to remove residue resist in the compressed areas.
Nanoimprint-2

Nanoimprint-3 Stamper

Nanoimprint-4
Laser Assisted Direct Imprint

Figure 1 Schematic of laser-assisted direct imprint (LADI) of nanostructures in silicon. a, A quartz mould is brought into contact with the silicon substrate. A force presses the mould against the substrate. b, A single XeCl (308 nm wavelength) excimer laser pulse (20 nm pulse width) melts a thin surface layer of Si. c, The molten silicon is embossed while the silicon is in the liquid phase. d, The silicon rapidly solidifies. e, The mould and silicon substrate are separated, leaving a negative profile of the mould patterned in the silicon. f, The reflectivity of a HeNe laser beam from the silicon surface versus the time, when the silicon surface is irradiated by a single XeCl (308 nm) laser pulse with 1.6 mJ cm² fluence and 20 ns pulse duration. Molten Si, becoming a metal, gives a higher reflectivity. The measured reflectivity shows the silicon in liquid state for about 220 ns.

Nanoimprint-5
Laser Assisted Direct Imprint

Figure 3 SEM image of the cross-section of samples patterned using LADI. a, A quartz mould. b, Imprinted patterns in silicon. The imprinted silicon grating is 140 nm wide, 110 nm deep and has a 300 nm period, an inverse of the mould. We note that the 10 nm wide and 15 nm tall silicon lines at each top corner of the silicon grating are the inverted replicas of the small notches on the mould (the notches were caused by the reactive ion etching trenching during mould fabrication). This indicates the sub-10-nm resolution of the LADI process. (These images are representative only. In fact, the Si structure in the image was probably not imprinted by the structure shown in the mould image.)

II-H. Shadow Mask

Alumina Mask

Mask Application

Deposition

Liftoff

Thickness ~ hole size

Liu, et. al. APL 81, 4434, (2002)

FIG. 1. Scanning electron micrograph of the 67 nm diameter nanodot sample. Inset is a histogram showing the distribution of nanodot sizes.

FIG. 2. Typical SEM images of a porous alumina mask (anodized in 0.3M oxalic acid at 40 V) with magnifications of (a) 35,000 and (b) 90,000; structural characterization of the porous mask shown in (b). (c) Diameter distribution (average of 63 nm and standard deviation of 6 nm). (d) Periodicity distribution (average of 101 nm and standard deviation of 12 nm). (e) Number of nearest-neighbor pores (average of 5.97, standard deviation of 0.18). (f) Angle between the directions to nearest neighbor pores (average of 59.9°, standard deviation of 10.6°). (g) Pore shape is analyzed by fitting the pores as ellipses. The average length ratio (minor axis/major axis) of the ellipses is 0.85, with standard deviation of 0.11. (h) FFT image of porous structure. The pore periodicity (~98 nm) calculated from the FFT spot distance agrees well with the result shown in (d). Both the structural characterization and FFT analyses are consistent with a hexagonal ordering of nanopores.

Li, et. al. JAP 100, 074318, (2006).
Shadow Mask-3


Fig. 2. SEM images of an array of multiple dots of Au at two different magnifications.

Fig. 3. Images of the array of bimetallic dots of Au and Ag: a) SEM image, b) SAM image. In (b) the signals from Au, Ag, and Si are shown in green, red, and blue, respectively.
II-I. Self-Assembly: Nucleation-1

Ni dots on Au (111)

Other systems:
- Co on Au (111)
- Fe & Co on Cu (11)
- Co on N₂ adsorbed Cu (100) & Cu (110)
- Fe-Ag on Mo (110)


FIG. 2. Correlation of Ni island nucleation with Au reconstruction. (a) Typical reconstructed section of large terrace on clean Au(111). Light zigzagging bands are ~0.2-Å-high ridges where atoms are near bridge sites. “Elbows” in ridges lie on two nearly horizontal domain boundaries. The lower boundary contains “pinched” elbows; the upper one, “bulged” elbows. $V_t = -2.0$ V. (b) Completed nucleation and polygonal shape of Ni islands at 0.14 ML. $V_t = -0.61$ V, deposition rate 0.1 ML/min. Of thousands of islands observed in dozens of STM images that reveal both islands and reconstruction ridges, ~99% form at these elbow sites. In (b) and (c) a nonlinear gray scale is used to make ridges visible. (c) Nucleation of Ni islands at elbows at $\theta = 0.01$. Three islands are seen on each of two domain boundaries of the herringbone pattern, running diagonally from upper left to lower right. $V_t = -1.11$ V, deposition rate 0.05 ML/min. (d) Sketch of herringbone pattern and nucleation sites. Two pairs of ridges are shown as dark bands. Arrows on the upper pair show directions of Burgers vectors, which alternate in type-x ridge and remain constant in type-y ridge. On the lower pair, small circles mark island sites [cf. (c)], located symmetrically about the central axis of the type-x ridge (fine line).
Self-Assembly: Nucleation-2

Co dots on Au (111)


FIG. 1. Reconstructed Au(111) surface (730×730 Å²) with rotational domains along the (112) direction forming an ordered zigzag pattern.

FIG. 2. (a) 0.3-ML Co coverage on Au(111) (1600×1600 Å²); 2-ML-high polygonal Co islands nucleate at the kinks of the Au(111) zigzag reconstruction. (b) Height variation along the line indicated in (a).

FIG. 4. (a) 3-ML Co coverage on Au(111) (1600×800 Å²). At this coverage the Co layer is already contiguous. (b) 7-ML Co coverage (3200×1600 Å²) showing a persisting regular array of Co islands related to the initial nucleation pattern.
Self-Assembly: Nucleation-3

**Co pillars on Au (111)**

![Diagram showing layers of InAs, GaAs, Bi-layers, and Au substrate.](image)

**FIG. 1.** Principle of multilayered systems of self-organizing dots. (a) Strain energy minimization induces vertical self-assembly. (b) Presence of dots from successive layers [4]. (b) Presence of dots in the image is due to the interlayer-spacer thickness is sufficiently reduced.


**FIG. 2.** The pillar-growth process is illustrated by STM views of the top surface. For steps (a)–(c), a cross-section view of real data is provided, along with a schematic view. (a) After deposition of a nominal thickness of 0.2 AL of Co on Au(111) at 300 K. The main STM image is 300 × 300 nm. The cross-section reveals that the dots are 2 AL high. (b) After deposition of Au up to the completion of the fourth atomic layer (step 2 in the text). The STM image is 60 × 35 nm. The hollows are about 0.06 nm deep, which is consistent with twice the height difference between a bulk hcp Co(0001) AL (0.205 nm) and an fcc Au(111) AL (0.235 nm). (c) After deposition of the second layer of Co dots (step 3 in the text). The STM image is 65 × 25 nm. The hollows are about 0.12 nm deep, i.e., 4 times the height difference between a Co and a Au atomic layer, suggesting that the dots are now 4 AL high. (d) After 20 AL of Co have been stacked (300 × 300 nm STM image). A self-organized array of pillars of nearly pure Co has been formed, with pillars 3 nm in diameter and 5.5 nm high.
Self-Assembly: Reduction-1

FePt: Solution phase metal salt reduction
Reduction of Pt-acetylacetonate to Pt
Decomposition of Fe(CO)$_5$ to Fe

(A) TEM micrograph of a 3D assembly of 6-nm as-synthesized Fe$_{50}$Pt$_{50}$ particles deposited from a hexane/octane dispersion onto a SiO-coated copper grid. (B) TEM micrograph of a 3D assembly of 6-nm Fe$_{50}$Pt$_{50}$ sample after replacing oleic acid/oleyl amine with hexanoic acid/hexylamine. (C) HRSEM image of a ~180-nm-thick, 4-nm Fe$_{52}$Pt$_{48}$ nanocrystal assembly annealed at 560°C for 30 min under 1 atm of N$_2$ gas. (D) High-resolution TEM image of 4-nm Fe$_{52}$Pt$_{48}$ nanocrystals annealed at 560°C for 30 min on a SiO-coated copper grid.

Self-Assembly: Reduction -2

(A) SEM image of self-assembled Co-nanocrystal superlattice device. (B) Transmission electron microscope image of a Co-nanocrystal superlattice before annealing, shows a ~4-nm interparticle distance (nanocrystal diameter is 10 nm). (C) Co-nanocrystal superlattice after annealing (~2-nm interparticle distance).