Supporting Information

Robust Pattern Transfer of Nanoimprinted Features for Sub-5 nm Fabrication

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1. Nanoimprint mold preparation

200 mm Si wafers with 100 nm-thick diamond-like carbon (DLC) films, grown by PECVD, were obtained from Dr. Christopher Jahnes (IBM T. J. Watson Research Center, Yorktown Heights, NY). The wafers were diced into small substrates, ~ 2" x 1". The substrates were rinsed with acetone and isopropanol and dried with a nitrogen streamprior to the e-beam resist spinning. HSQ (Fox 12, Dow Corning), diluted in MIBK (1/4 g/g) was applied on the substrates by spin coating with a spinning rate of 6000 rpm for 1 min., resulting in HSQ films with a thickness of 20 nm (the thickness was calibrated on HSQ coated Si substrates test samples by null ellipsometry (Rudolf III)).

Patterning was done using an FEI XL 30 Sirion scanning electron microscope equipped with a Nabity NPGS pattern generator. Exposure was done at 30 keV with a probe current of 25 pA for the highest resolution features. 5 μm x 5 μm arrays of dots separated by 200 nm in vertical direction and by 100 nm in horizontal direction were written with a range of point exposure doses from 11 fC to 18 fC. The development was done with Microposit LDD-26W Developer (Rohm and Haas Electronic Materials) for 2 min., followed by a DI water rinse and drying in nitrogen.

The oxygen plasma etching was done in Oxford PlasmaLab 80 etcher with the following conditions: chamber pressure of 28 mTorr; oxygen flow rate of 100 sccm; and RF power of 150 W. The etching time was 25 sec., resulting in a final DLC thickness of 50 nm, as monitored by null ellipsometry. The HSQ mask was removed after the etching by dipping the substrate in buffer oxide etchant solution (1:6) for 1 min., followed by a DI water rinse and drying in nitrogen.

An anti-adhesion coating was applied by plasma fluorination, using the Oxford PlasmaLab 80 tool, with the following parameters: C_4F_8 flow of 100 sccm; chamber pressure of 88 mTorr; RF power of 100W, and process time of 30 sec.

2. Preparation of substrates for nanoimprint

100 mm Si wafers with a thermally grown 300 nm silicon dioxide layer were cleaved to 1"x1" pieces, which were rinses with acetone and isopropanol and dried with nitrogen prior the resist spinning. 35K PMMA (MR-I 100nm, Microresist Technolohgy, GmbH) , diluted in anisol (2/1 v/v), was spun onto the substrates at a spinning rate of 4000 rpm, followed by baking of the substrates on hot plate (180^oC, 5 min.).

3. SEM analysis

All SEM images were taken using a Hitachi 4700 scanning electron microscope, operated in ultra-high-resolution mode, with an electron accelerated energy of 10 KeV, working distance of 5 mm, mixed (secondary + back-scatter) detector and chamber vacuum level of $1X10^{-3}$ Pa.

The size analysis of the imaged structures was done using "ImageJ" software. For the calculation of the average size of imaged objects, at least 8 different measurements were taken in each case, and the mean value and the standard deviation (1σ) were calculated.

For cross-section SEM images, the Si (100) samples were prepared by cleaving the substrates along a crystalline direction. First, a scratch parallel to the crystalline direction was made close to the patterned area in order to define the cleavage location, using a Karl Suss RA120M wafer scriber. Then the substrate was carefully cleaved along the direction of the initial scribe, with the cleave crossing through the patterned area.

High temperature annealing of AuPd nanodots

Annealing of samples at 450 ^{o}C was carried out on a hot plate, covered with a pyrex funnel, which was connected to a pipe with constant nitrogen flow. For temperatures higher than 450 ^{o}C, the annealing was done in a quartz furnace under a constant nitrogen flow. In all cases, the annealed substrates were cooled to room temperature by natural cooling under constant nitrogen flow prior their removal from the plate/furnace.

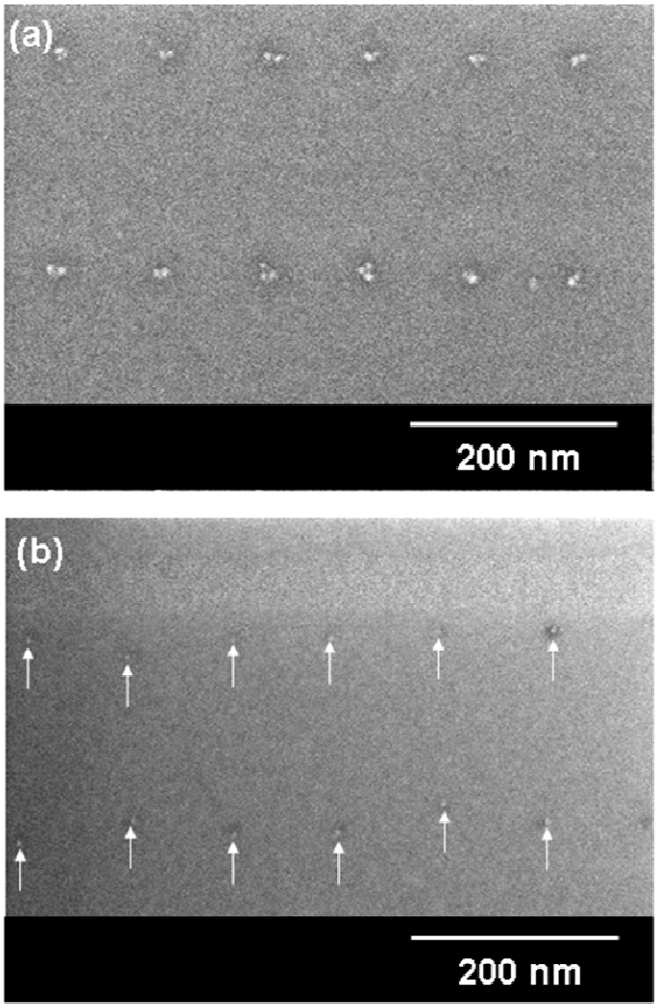

Fig S1. (a) Nanodots made of 0.5 nm Ti/ 0.8 nm AuPd, and annealed at 450 ^{o}C. Multiple nanoparticles with random sizes can be clearly seen in the original dot areas. (b) The same dots, annealed at 800 ^{o}C. The migration of the dots from their original position is shown by arrows.