Supplementary Information (SI) for Nanoscale.
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# Supporting Information (SI)

# Abrasive-free chemical-mechanical planarization (CMP) of gold for thin film nano-patterning

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# A. Pad conditioning

Two types of pad conditioning were used prior to CMP:

- *Low* conditioning: 30 s polishing of a dummy, without use of a diamond tool.
- *Moderate* conditioning: 24 s conditioning with a diamond tool.

Figure S1 shows a minor difference in material removal rate (MRR, slopes of the curves) between the two conditioning methods for a common set of CMP parameters: 26 nm/min at low conditioning and 30 nm/min at moderate conditioning. Though the difference is negligeable, moderate conditioning was nevertheless used in the standard process flow to maximize stability.

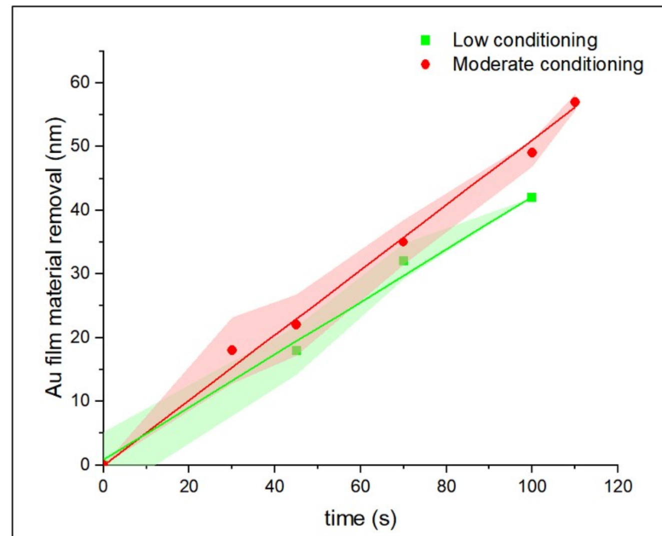


*Figure S1: Au film thickness removed during CMP following the two conditioning methods (CMP parameters: 1/200, 4.3 psi, 30 rpm, 50 mL/min, RT).*

## B. CMP experimental parameter ranges for experiments in section "*Unstructured thin film CMP and material removal mechanism*"

| Varied parameter | Dilution ratio | Pressure (psi) | Platten rotation (rpm) | Slurry flow (mL/min) | Temperature of the slurry (°C) | Referring |
|---|---|---|---|---|---|---|
| Dilution | - | 4.35 | 30 | 37.5 | 20 | Figure 2a |
| Pressure | 1/100 | - | 30 | 37.5 | 20 | Figure 2b |
| Platten rotation | 1/100 | 4.35 | - | 37.5 | 20 | Figure 2b |
| Slurry flow | 1/100 | 4.35 | 30 | - | 20 | Figure 2b |
| Pressure | 1/200 | - | 30 | 50 | 20 | Figure 2b |
| Platten rotation | 1/200 | 4.35 | - | 50 | 20 | Figure 2b |
| Slurry flow | 1/200 | 4.35 | 30 | - | 20 | Figure 2b |
| Temperature | 1/200 | 4.35 | 30 | 50 | - | Table 1 |

*Table S1: CMP parameters*

| Parameter | Measurement uncertainty |
|---|---|
| Au film thickness | ±10% of remaining film thickness |
| Pad pressure | ±0.25 psi |
| Platen rotation speed | ±1 rpm |
| Slurry flow rate | ±2 mL/min |
| MRR | Difference between maximum and minimum slope of the polishing curve ~ ±10 nm/min |

*Table S2: CMP parameter uncertainties*

# C. CMP experimental parameters and their effect on the MRR (unstructured Au film)

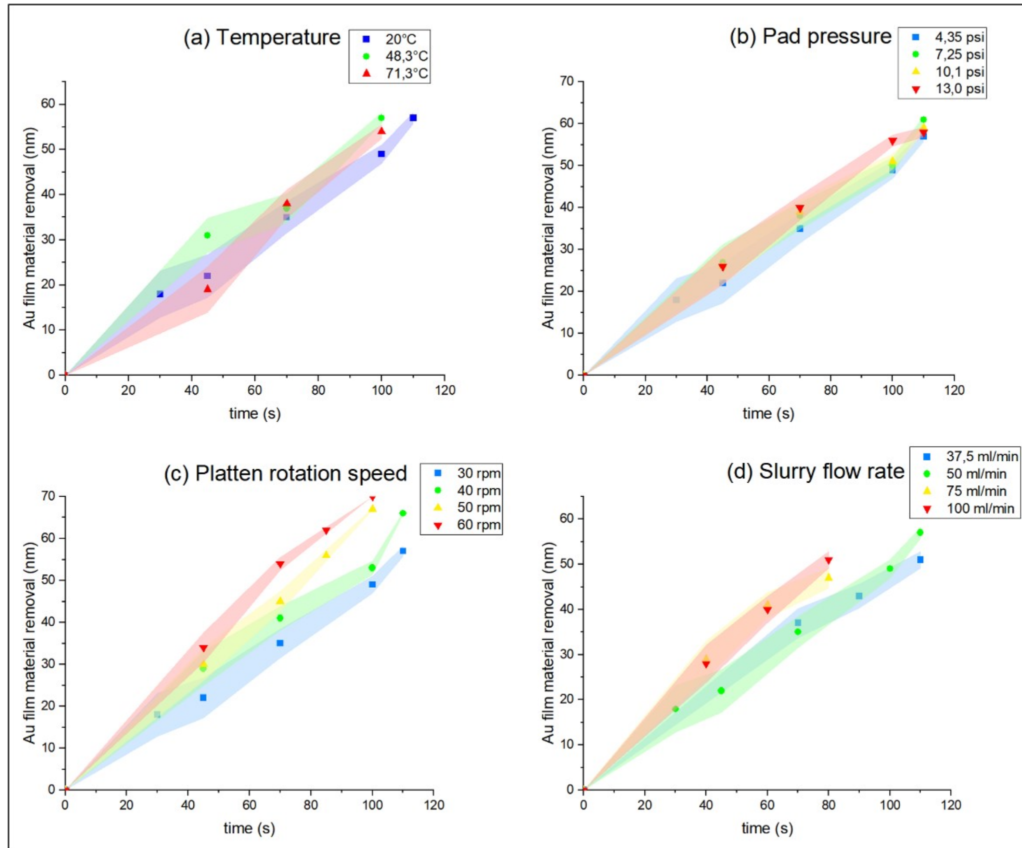


*Figure S2: Au film removal during CMP for different (a) temperatures, (b) applied pressures, (c) platen rotation speeds and (d) slurry flow rates, at 1/200 dilution ratio.*

# D. Effect of polishing time on macroscopic defects and scratches

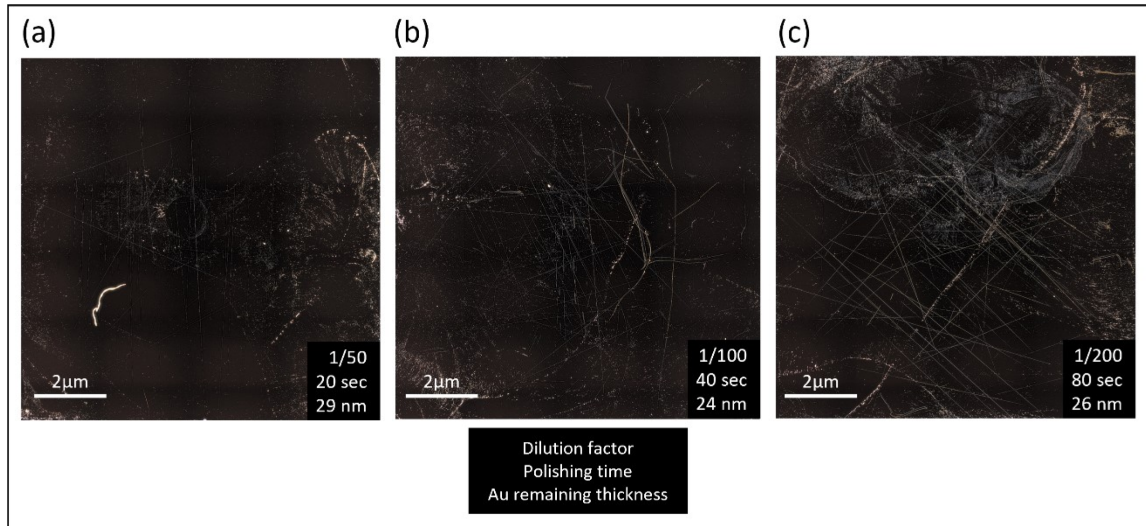


*Figure S3: Macroscopic surface defects viewed by optical microscopy after CMP to reduce Au film thickness to ~20-30 nm, at different polishing time and slurry dilution factor (a:1/50 – 20 sec; b:1/100 – 40 sec; c:1/200 – 80 sec). CMP parameters: 4.35 psi, 30 rpm, 50 mL/min, RT.*

## E. Au CMP damascene in silica

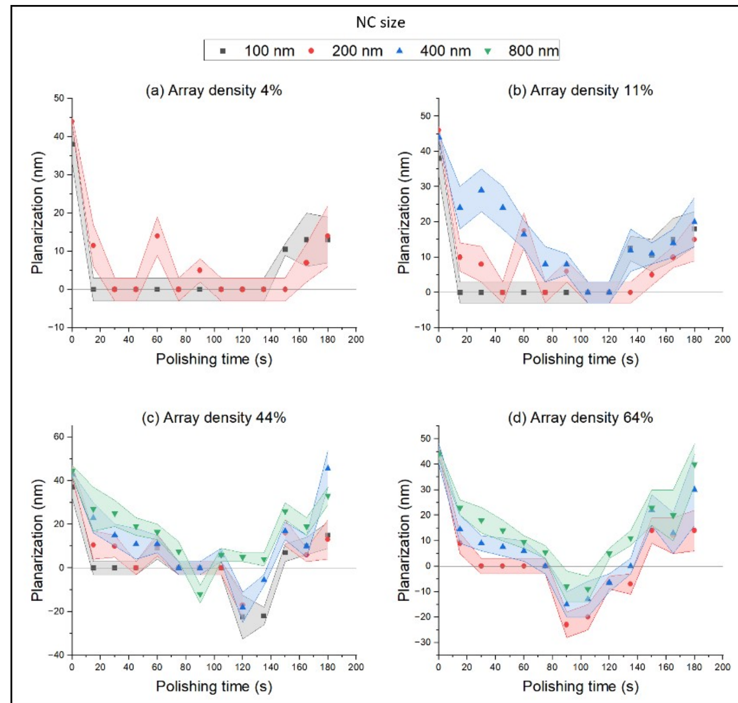


*Figure S4: Planarization curves for various cube sizes (100 nm – 800 nm) and array densities (a) 4% (b) 11% (c) 44% and (d) 64% at fixed CMP parameters (1/200, 4.3 psi, 30 rpm, 75 mL/min, RT).*

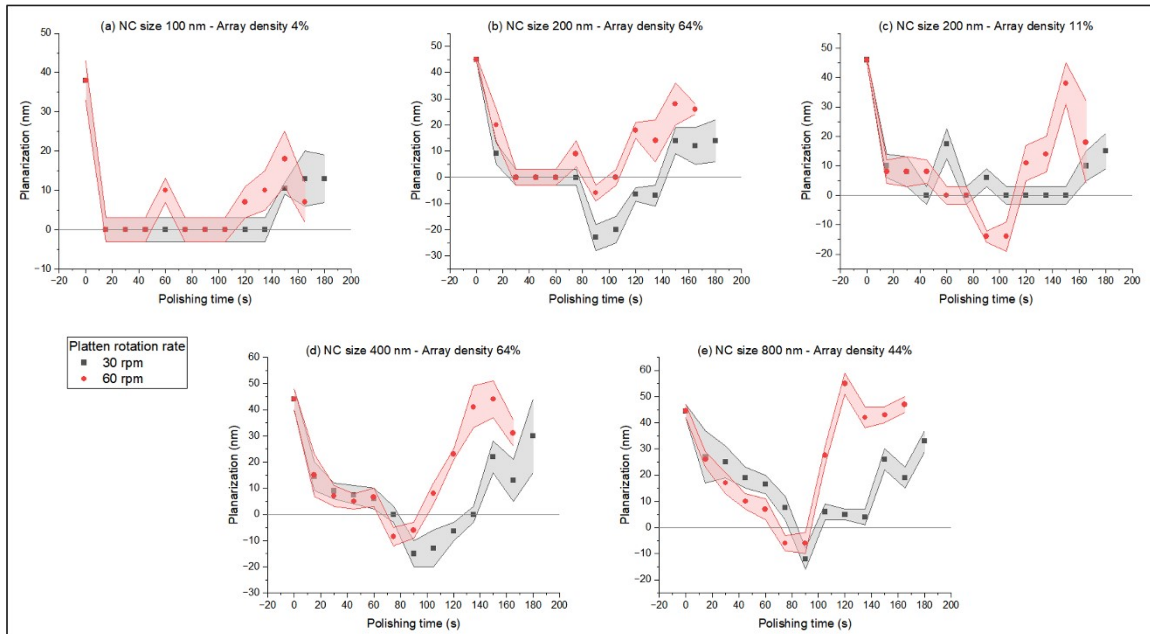


*Figure S5: Planarization curves for two different pad rotation speeds (30 and 60 rpm) for various cube sizes and array densities. Other CMP parameters are 1/200, 4.3 psi, 75 mL/min, RT.*

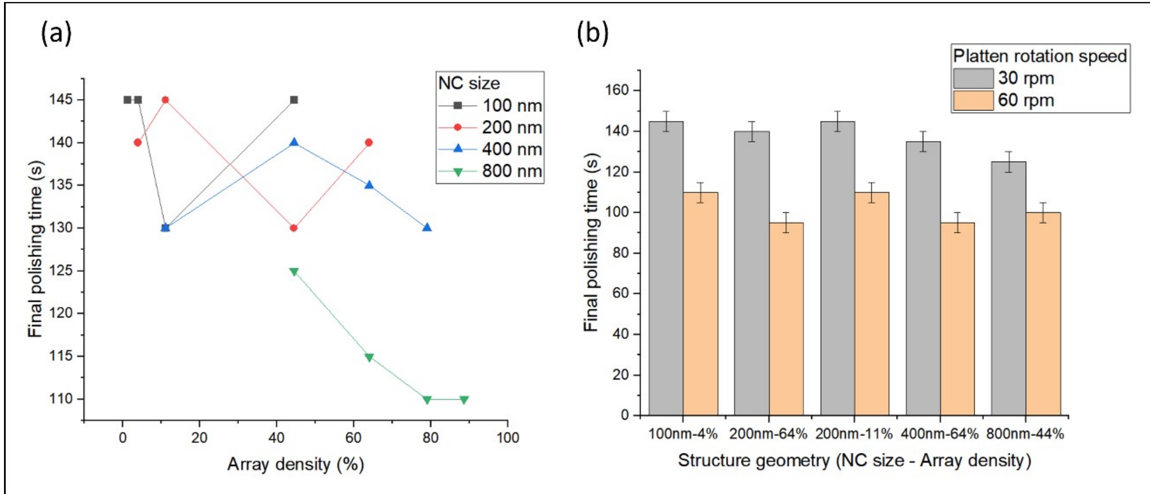


*Figure S6: Polishing time over a range of cube sizes and array densities: (a) for a specific case of fixed CMP parameters and (b) for two different platten rotation speeds.*

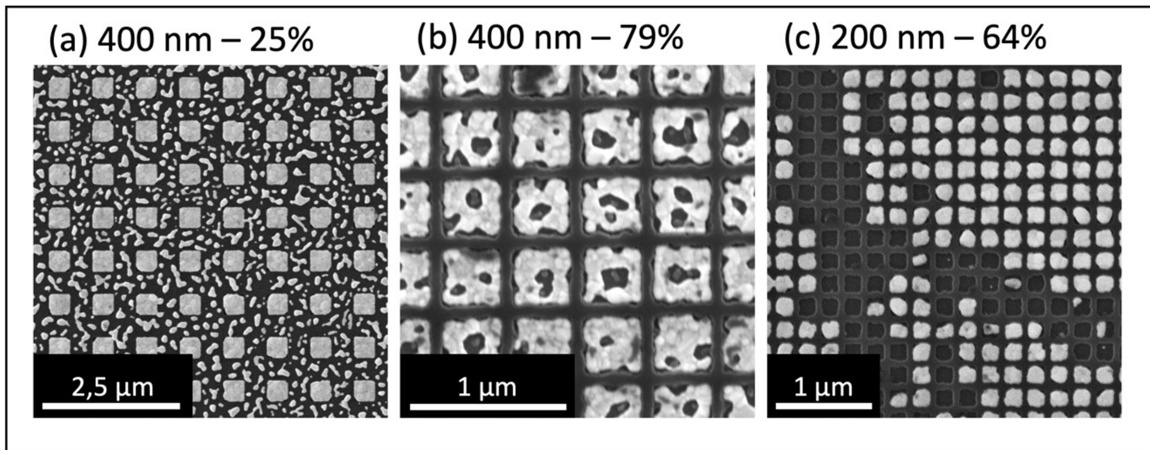


*Figure S7: SEM images of gold nanocubes (NC) embedded in $SiO_2$ revealed by abrasive-less CMP damascene fabrication for various device geometries to illustrate typical defects that may occur during development and parameter optimization : a) residual Au on the $SiO_2$ surface between NCs due to under-polishing; b) excessive dishing of the NCs and/or delamination at the center of the wells due to slight over-polishing; c) NC delamination due to excessive over-polishing and/or sub-optimal Au deposition parameters.*

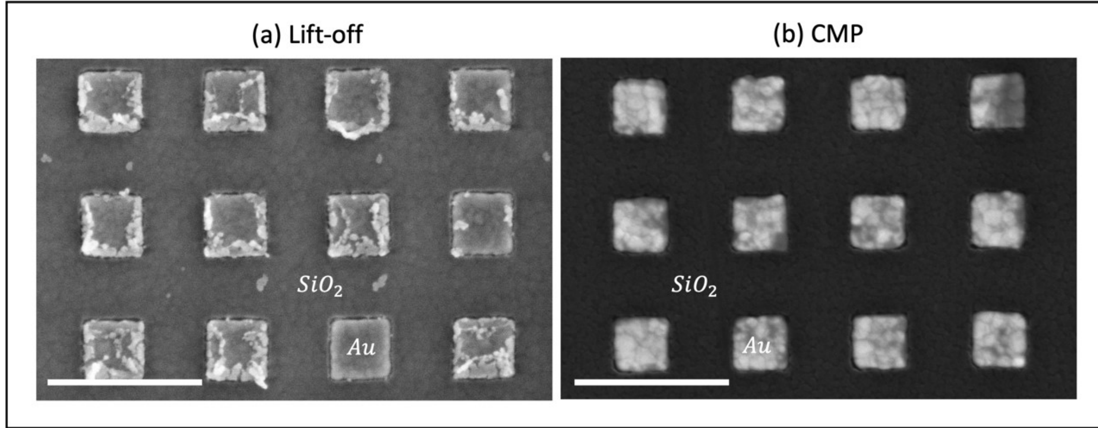


*Figure S8: SEM surface images of gold NC embedded in $SiO_2$ fabricated by (a) lift-off and (b) abrasive-less CMP. Formation of collar defects above the NC made by lift-off, and absence of collar defects with the CMP technique. Scale bar 500 nm.*

# F. Electromagnetics numerical modeling of nanostructured plasmonic array over mirror

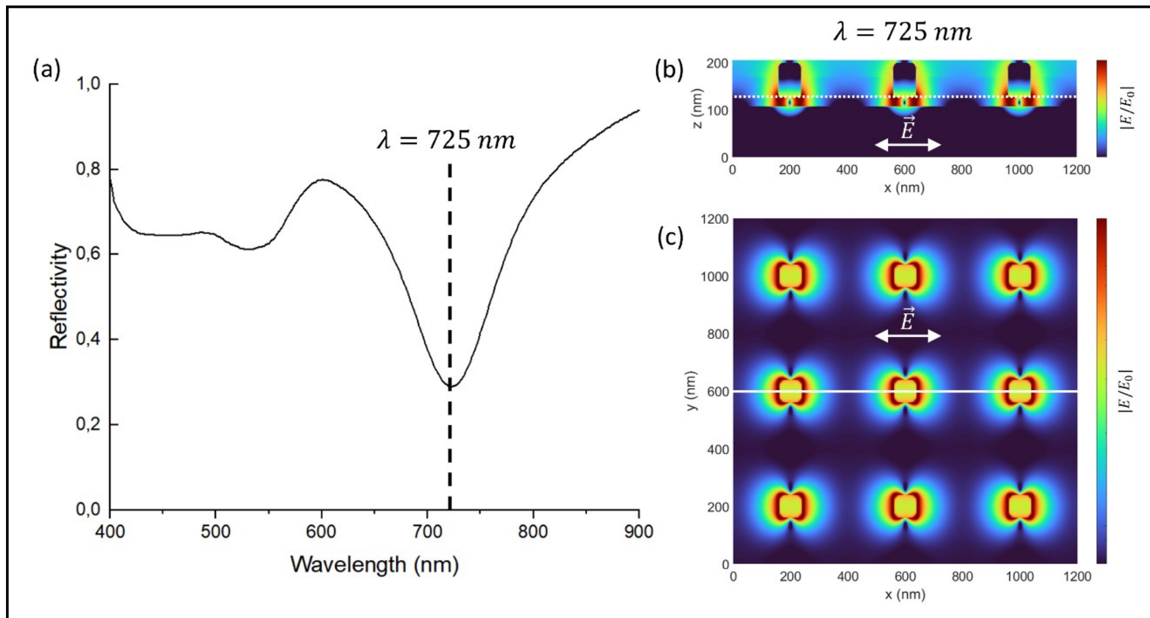


*Figure S9: Electromagnetic simulation of NC array over Au mirror (400 nm pitch and 100 nm cube size) (a) normalized reflection in far-field under normal incidence excitation. (b-c) Electric field amplitude distribution (b) in the cross-section plane and (c) in the array plane in the gap at the base of the NC at 725 nm wavelength. Polarization of electric field is represented as white double arrowed line. Cross section position is given by the continuous white line in array plane map, and array plane position is denoted by the dotted line on cross section map.*