ADVANCED MATERIALS

# Supporting Information

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# Harnessing Localized Ridges for High-Aspect-Ratio Hierarchical Patterns with Dynamic Tunability and Multifunctionality

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**Supplementary Information for**

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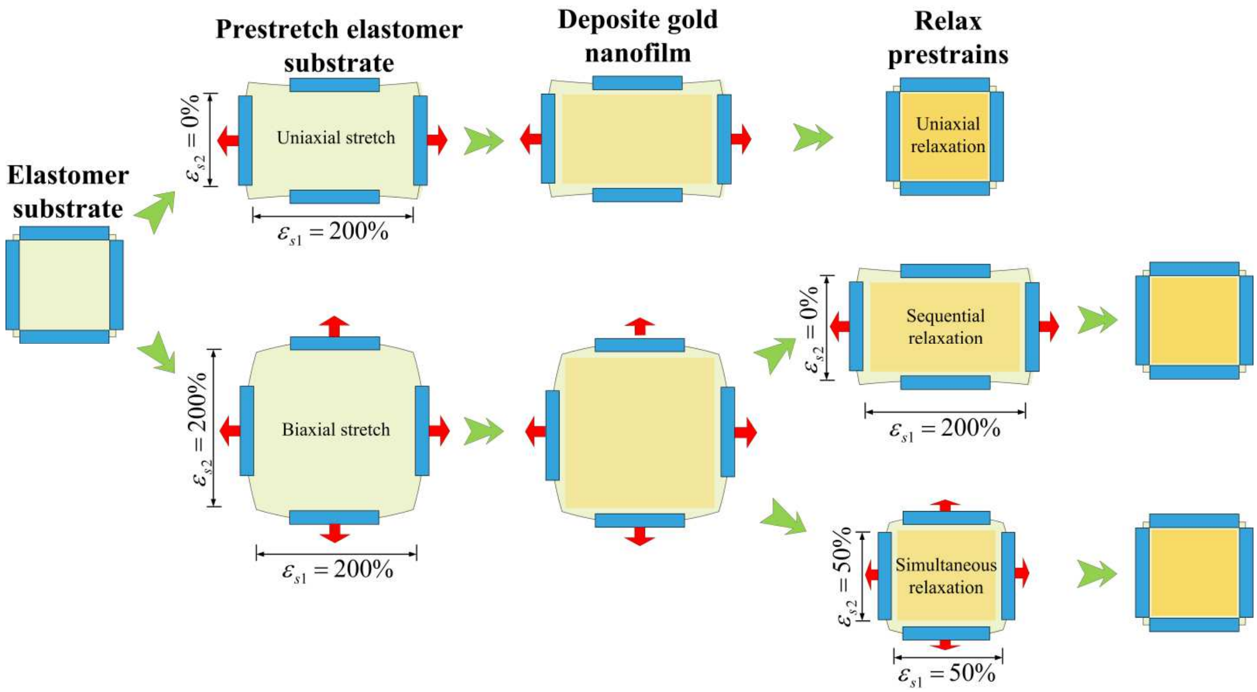


**Figure S1. Schematic illustration of the fabrication process for the hierarchical surface patterns.**

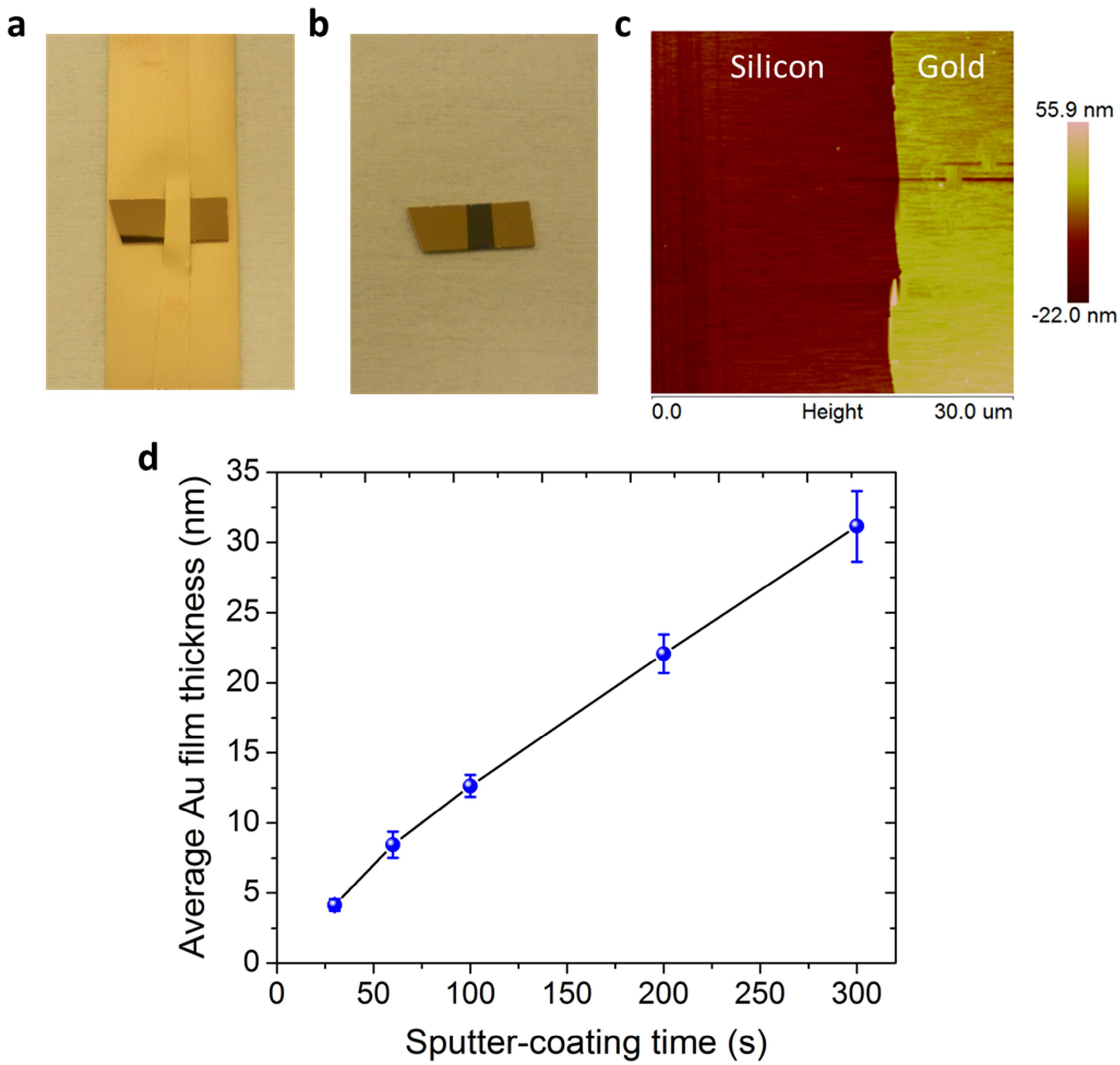


**Figure S2. Measurement of the thicknesses of gold films as a function of sputter-coating time.** We measure the thicknesses of gold films under various sputter-coating times using atomic force microscope (AFM). Before sputter-coating, we partially cover the milled silicon wafer with a small piece of tape (**a**), then sputter the sample with specified time and peel off the tape from the silicon wafer (**b**). Finally, the surface topography of the partially-coated silicon wafer is measured by AFM (**c**). The thicknesses of nano-films under different sputter-coating time (**d**) are then obtained from the AFM images by the software NanoScope Analysis. For each deposition time, we test three different samples and measure three times for each sample, and then use the average values as the measured film thickness.

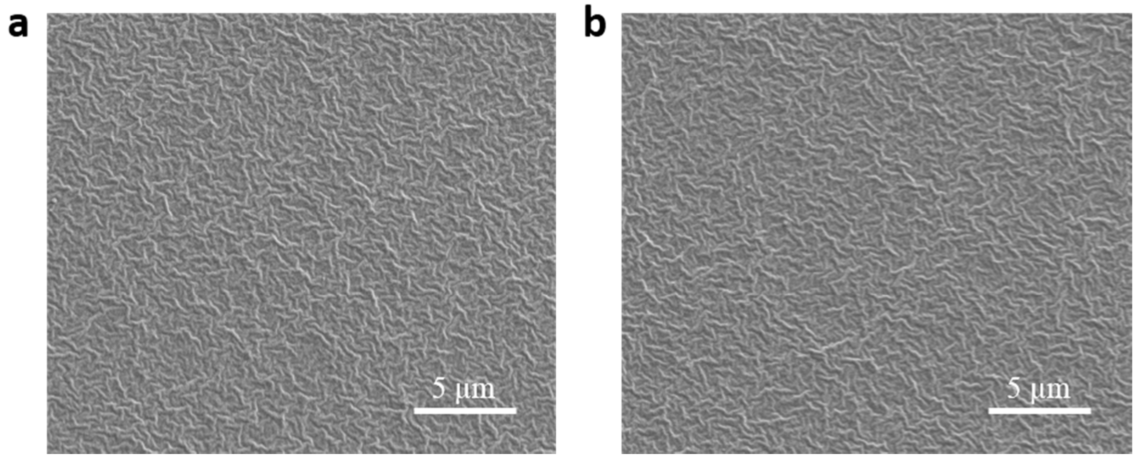


**Figure S3. The wavelength of the first-level wrinkles is independent of the gold film thickness:** SEM images of the first-level wrinkles on gold films with thicknesses of 4.0±0.4 nm (**a**) and 13±1 nm (**b**).

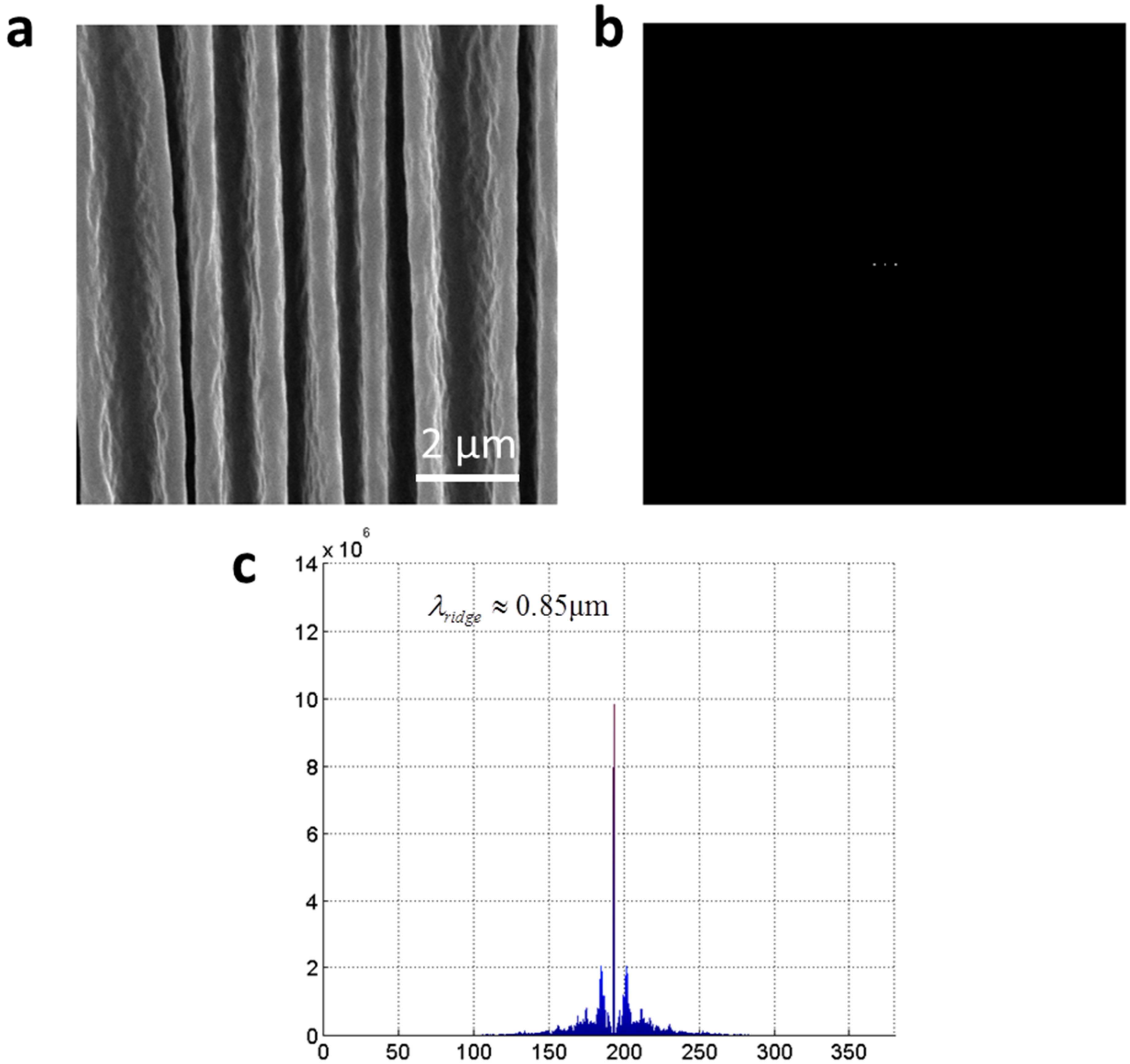


**Figure S4. Measurement of the wavelength of the ridged pattern:** (**a**) SEM images of ridge pattern formed by fully relaxation of a gold film (13±1 nm) on uniaxially pre-strained elastomer with $\varepsilon_{pre1} = 200\%$ . The SEM images are analyzed by 2D fast Fourier transform (2D FFT) using MATLAB (MathWorks Inc., Natick, MA, USA). The two symmetric bright spots shown in the 2D FFT pattern (**b**) denote the characteristic frequency of the images in frequency domain, which indicates that the wavelength of the patterns in (**a**) has a good uniformity. The main wavelength of the patterns can be determined from the peak values in the power spectrum (**c**) based on the relationship between frequency and space domain.[22, s1, s2]

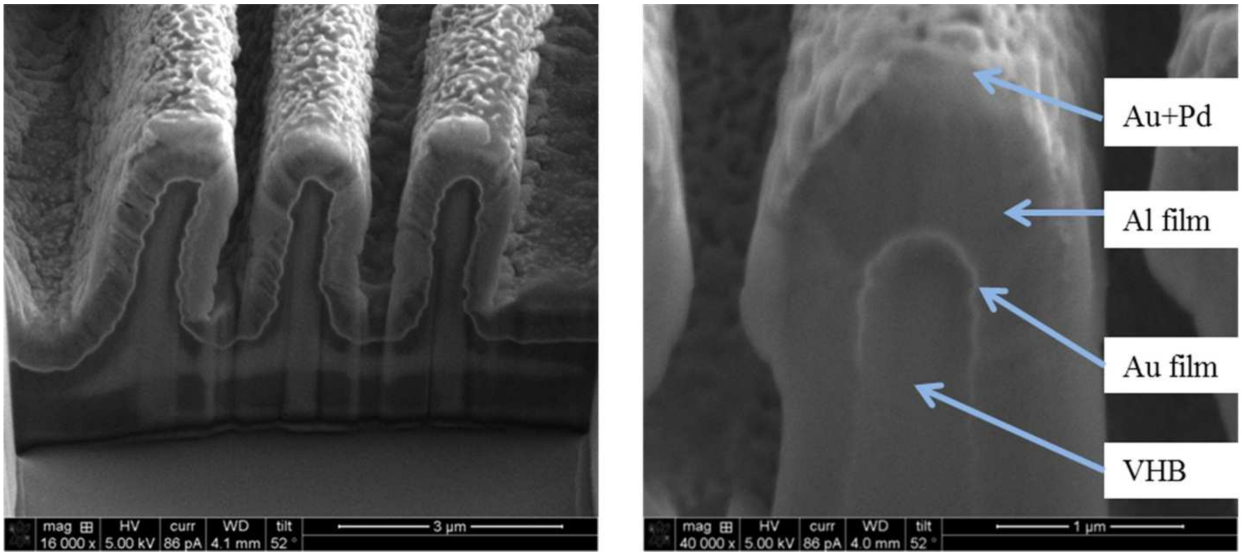


**Figure S5. Measurement of the amplitude of the ridge pattern.** SEM images of sectioned ridges of gold film (31±2 nm) on fully relaxed substrate with uniaxial pre-strain of 250%. The ridged sample is sectioned by Focused Ion Beam (FIB) milling.[s3] Before sectioning the sample, we deposit 500nm aluminum (Al) followed by 50nm mixture of gold and palladium (Pb) on top of the ridged film to prevent potential damage of the film from gallium ion irradiation. Thereafter, the cross-sectional profiles are recorded by the high-resolution SEM built in FIB-system. Since the SEM images are taken from a tilted sample with an angle of 52º, the true height of the ridge $L$ are related to the measured value from the SEM image $l$ via $L = l / \sin\left(52^{\circ}\right)$.[s4] Furthermore, from a total of 5 different samples with substrate pre-strains of 200% and 250%, we find that the substrates all deform together with the gold films at ridges.

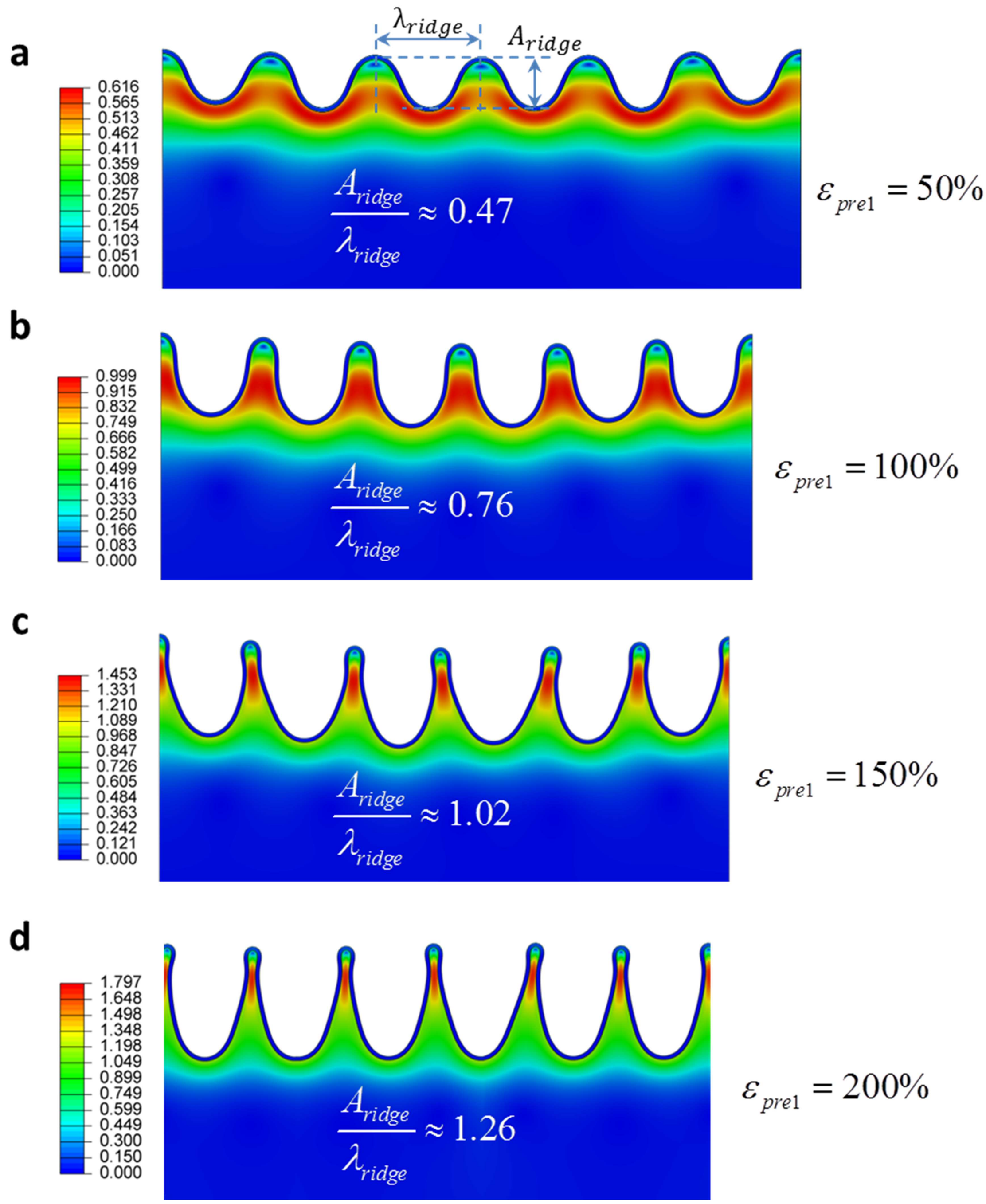


**Figure S6. Localized ridges on fully relaxed substrates with uniaxial pre-strains of 50%, 100%, 150% and 200% calculated by finite-element model.** The shear modulus ratio between the film and substrate is 1000. The contour represents the maximum in-plane principal strain.

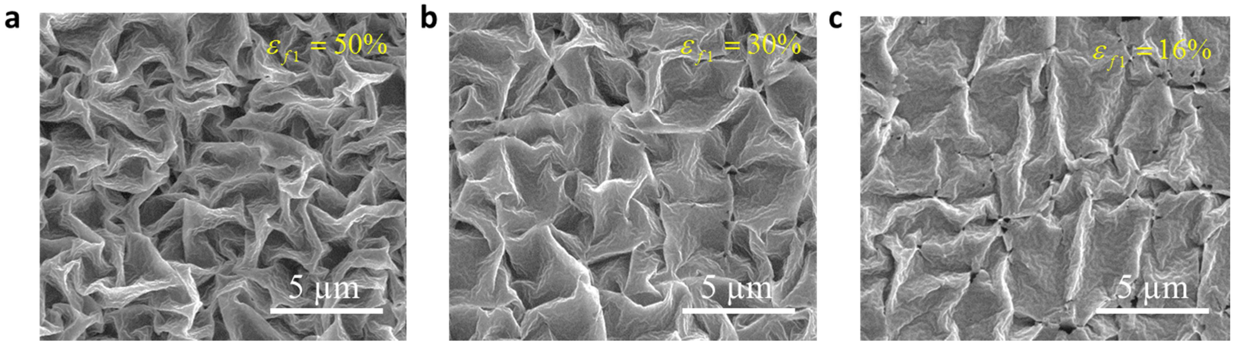


**Figure S7. Evolution of hierarchical pattern on fully relaxed substrate being stretched back:** SEM images of hierarchical pattern of gold film under nominal biaxial compressive strains of 50%, 30% and 16%.

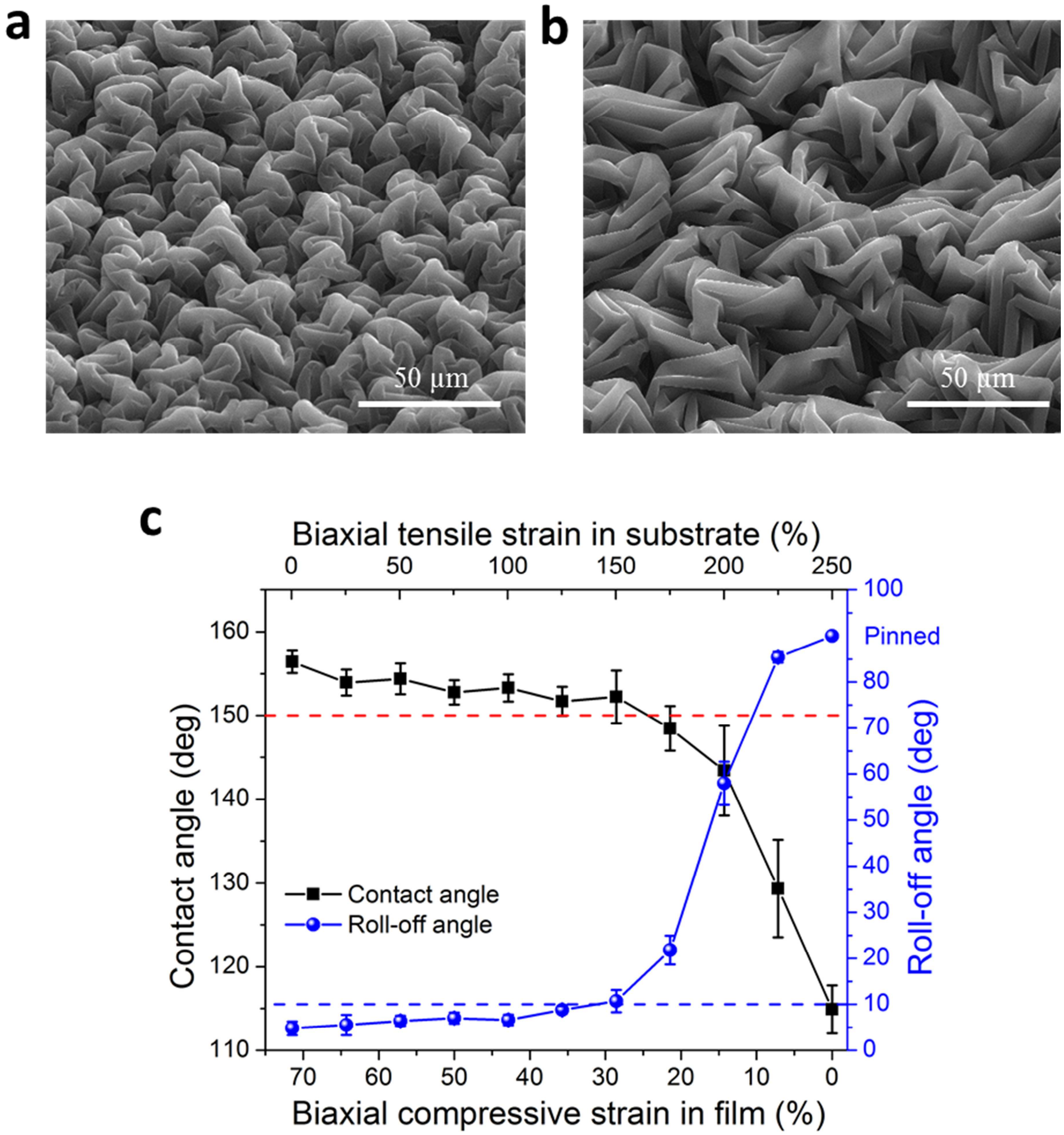


**Figure S8. Super-hydrophobicity and tunable wettability of the hierarchical patterns of silver or aluminum film on biaxially pre-strained substrates with simultaneous relaxation:** A thin layer of silver or aluminum film (~15 nm) is coated by E-beam Metal Evaporator (Kurt Lesker PVD 75) on bi-axially pre-strained elastomer with $\varepsilon_{pre1} = \varepsilon_{pre2} = 250\%$ , and the as-deposited silver sample is then modified with saturated vapor of (heptadecafluoro-1,1,2,2-

tetrahydrodecyl) trichlorosilane for 2 hours. SEM images of hierarchical pattern of silver film (**a**) and aluminum film (**b**) on fully relaxed substrates. The static contact and roll-off angles of water droplet on the silver film as functions of the biaxial compressive strain in the film or tensile strain in the substrate (**c**).

## Additional Reference

[S1] R. C. Gonzalez, R. E. Woods, S. L. Eddins, Digital image processing using MATLAB, Gatesmark Publishing Knoxville , 2009.
[S2] D.A. Kunz , P. Feicht , S. Gödrich , H. Thurn , G. Papastavrou , A. Fery , * and J. Breu, Adv. Mater. 2013, 25, 1337.
[S3] J.-Y. Sun, S. Xia, M.-W. Moon, K. H. Oh, K.-S. Kim, Proceedings of the Royal Society A: Mathematical, Physical and Engineering Science 2012, 468, 932.
[S4] JEOL Ltd, A Guide to Scanning Microscope Observation, 1.

**Video S1.** Water drops rolling off from the hierarchical surface pattern at a tilting angle of 4°**.** The hierarchical pattern is generated by simultaneously relaxing a biaxially pre-strained substrate ($\varepsilon_{pre1} = \varepsilon_{pre2} = 250\ \%$) with sputter-coated gold film ($H_f = 13 \pm 1$ nm).