Electronic Supplementary Material (ESI) for Nanoscale Advances.


# Shaping graphene with optical forging: from single blister to complex 3D structures

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## Reproducibility

Precise control of laser exposure combined with piezo scanning allow us to fabricate reproductible patterns with great precision. In Figure S1 matrices of blisters formed upon irradiation with 80 pJ pulse energy and different irradiation times are presented. Blisters in each matrix resemble each other. These examples show the possibility of scaling up 3D direct writing process. However, one should bear in mind that for high quality patterning steady system is required.

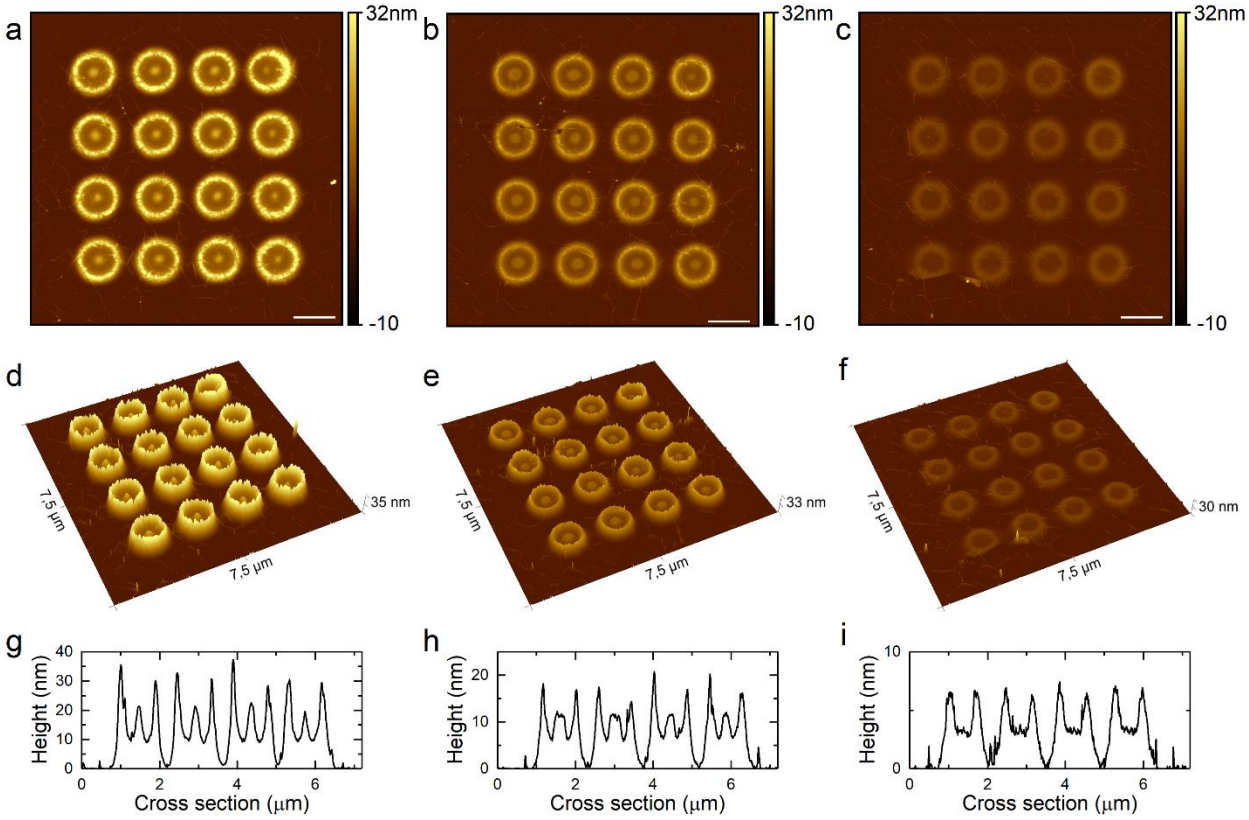


Figure S1. AFM (a, b, c) height images and (d, e, f) 3D view images of matrixes of blisters prepared with 80 pJ pulse energy, 1.5 µm separation between consecutive laser spots, and constant irradiation time: (a, d) 180, (b, e) 60 and (c, f) 10 s per spot. Scale bar 1 µm. Cross section measured through the central part of blisters in third row for dose: (g) 180, (h) 60 and (i) 10 s per spot.

**Lateral cross section of forged blisters**

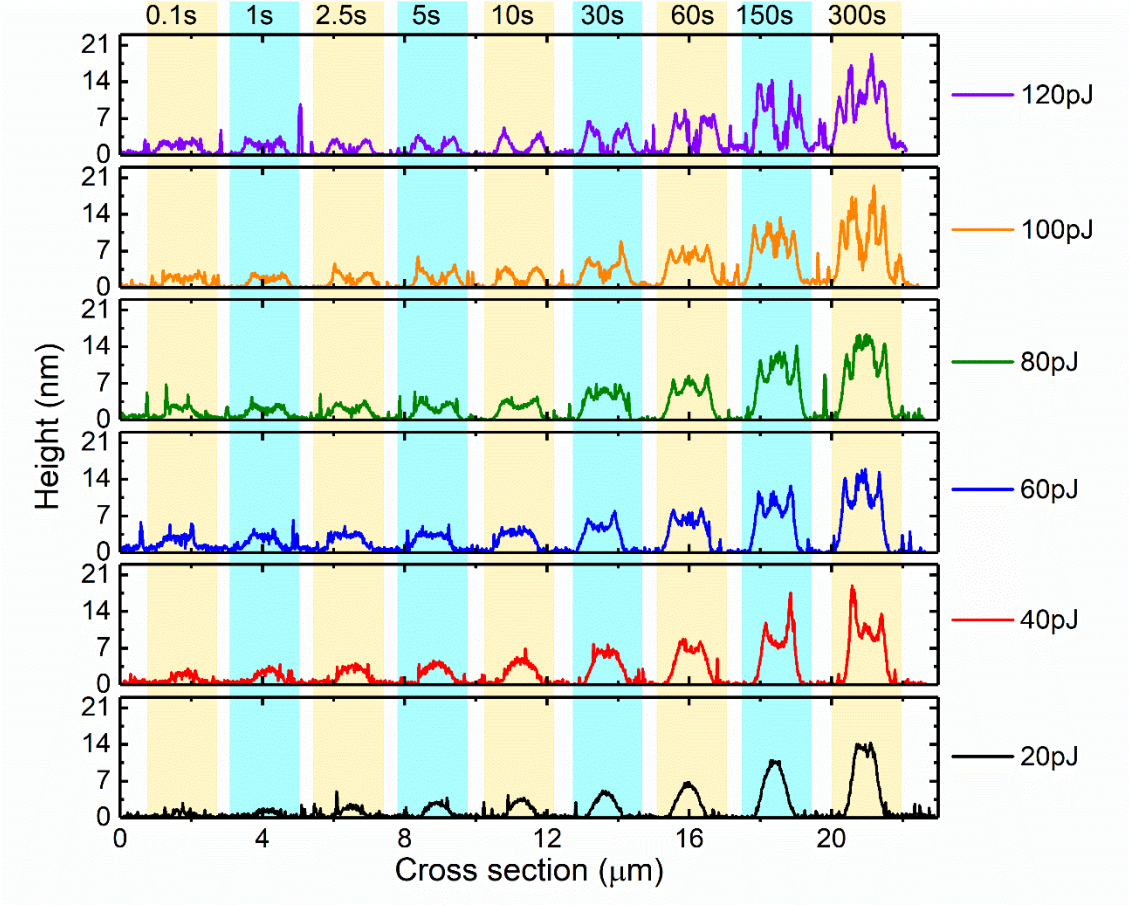


Figure S2. Cross section taken from AFM height image of blisters presented in Figure 1a.

## Raman spectroscopy studies

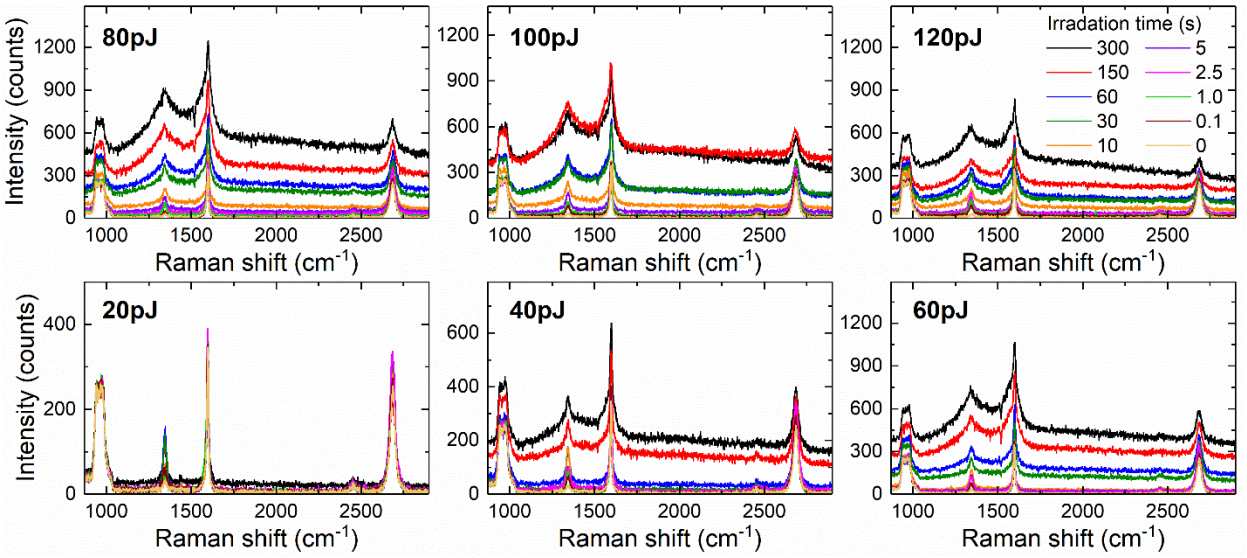


Figure S3. Raman spectra collected for blisters with different irradiation times, grouped by used pulse energies. Excitation wavelength: 515 nm.

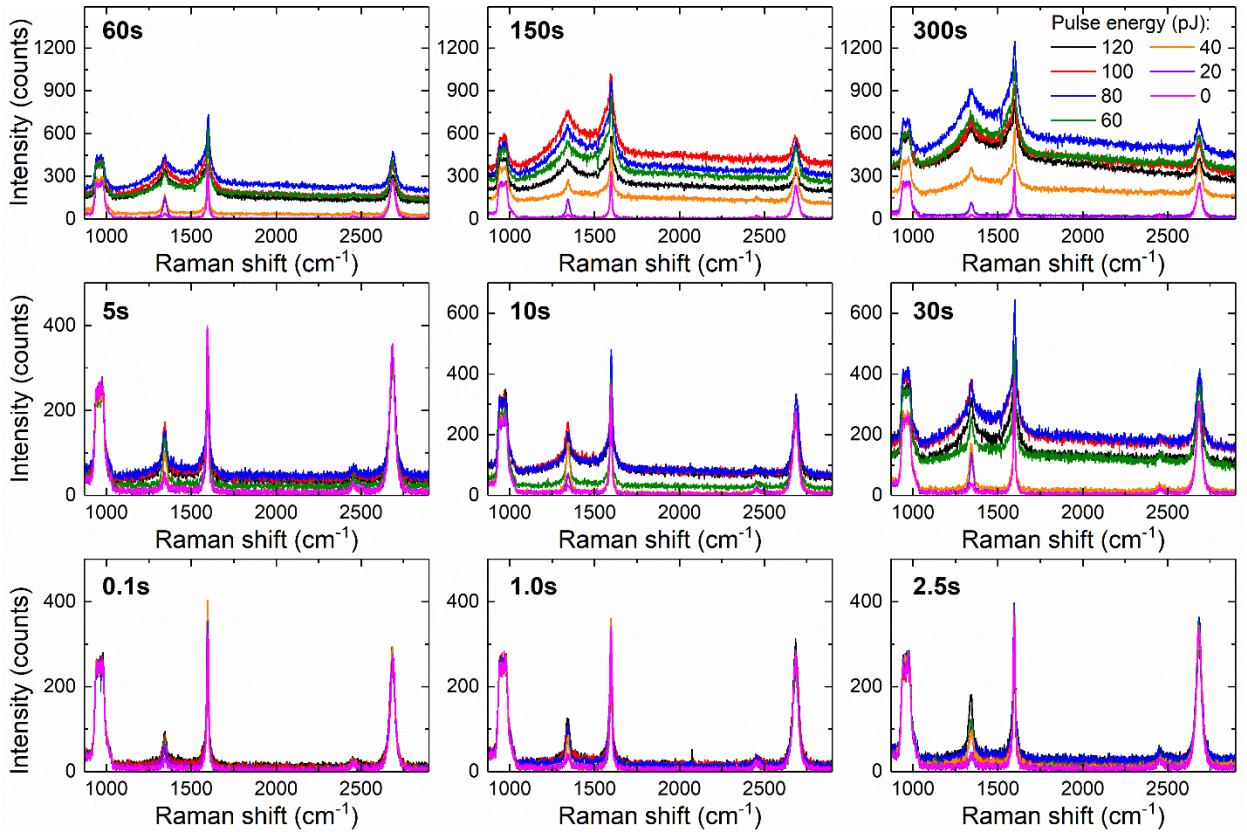


Figure S4. Raman spectra collected for blisters produced with various pulse energies, grouped by used irradiation time. Excitation wavelength: 515 nm.

## Oxygen plasma etching

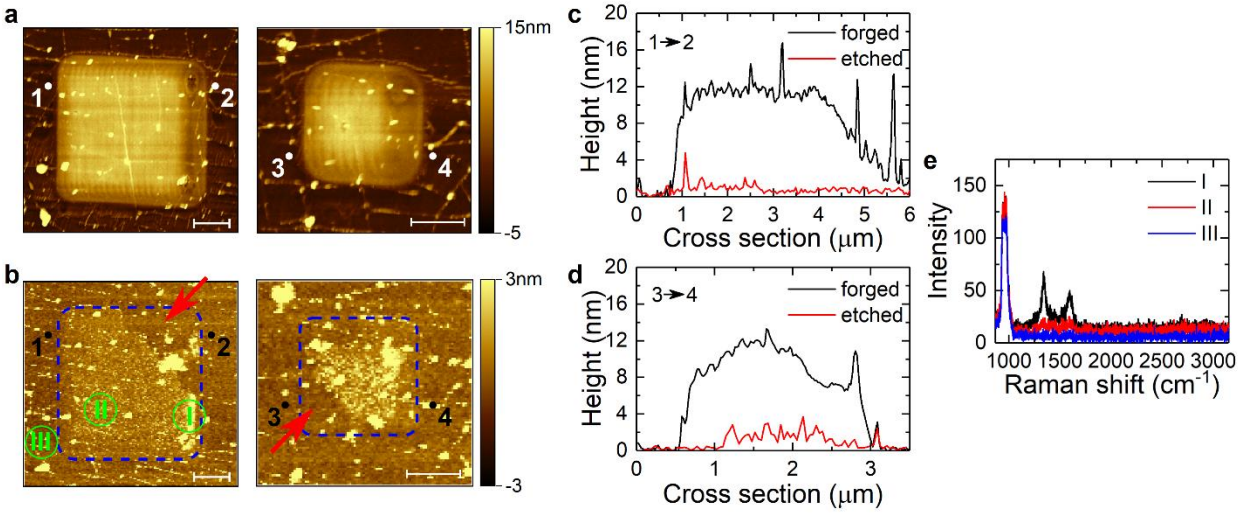


Figure S5. AFM images of optically forged squares (a) before and (b) after $O_2$ plasma etching. Squares were prepared with 60pJ and 1s irradiation time per spot. Scale bar 1 μm. Red arrows point completely removed optically forged features, and intact substrate surface. Blue dashed frame is added to guide eye to the contour of forged square. Cross section of forged blisters before and after etching taken between points (c) 1-2 and (d) 3-4. (e) Raman spectra collected after $O_2$ plasma etching in the places indicated at (b), showing a presence of small amount of the amorphous carbon.

## AFM channels

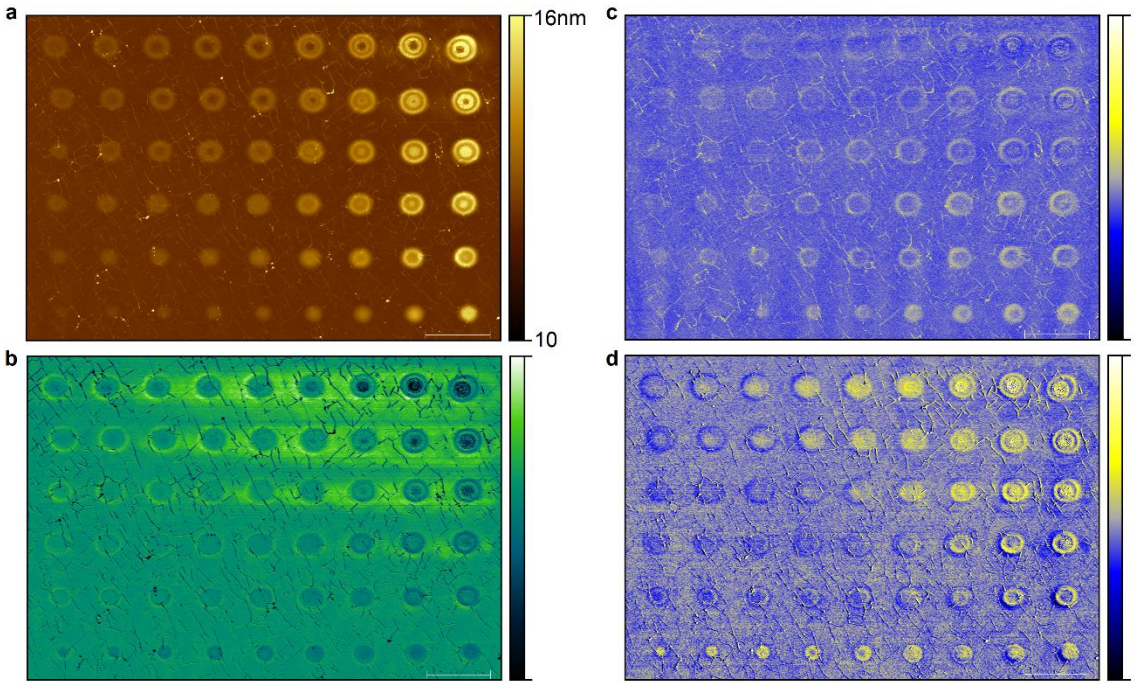


Figure S6. (a) Topography height image of blisters and corresponding (b) adhesion, (c) deformation, and (d) elastic modulus (DMT) channels. Scale bar 3 μm.

## Tetramer and pentamer

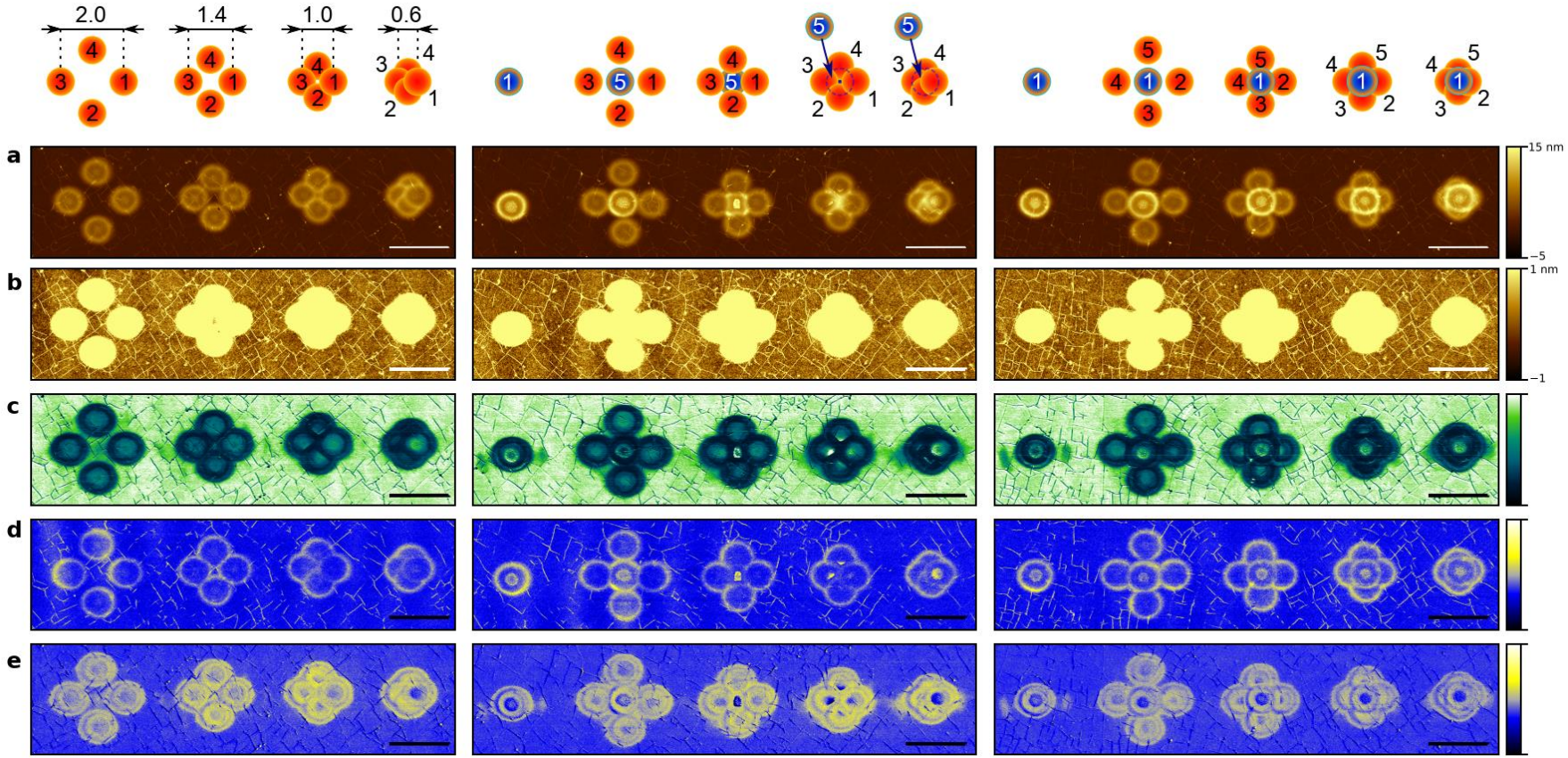


Figure S7. Schemes of complex patterning, with marked order of irradiation. On the left and center, first blisters with 10 s exposure per spot (orange circles) were prepared. On right blister with longer exposure was irradiated first (60 s, blue circle). All blisters were created using 60pJ pulse energy. Topography AFM images of obtained patterns with full (a) and reduced (b) color scale, and corresponding adhesion (c), deformation (d) and DMT modulus (e) channels. Scale bar 2 μm.

## Complex structures patterning

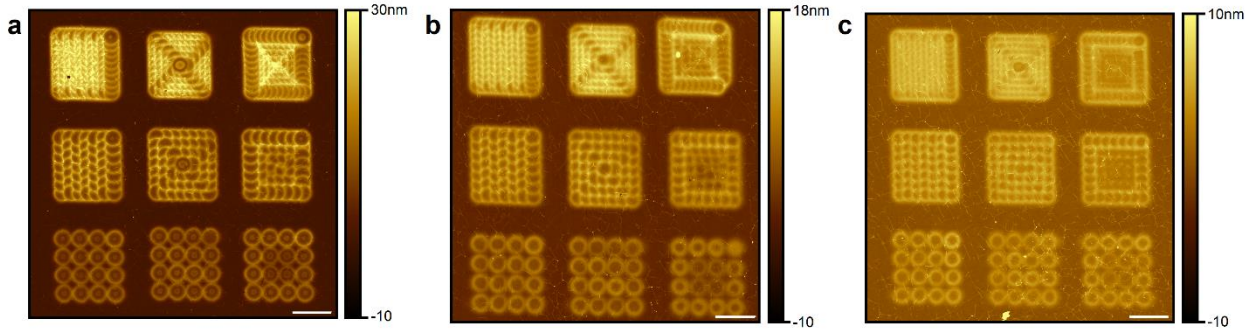


Figure 8. AFM height images of patterns prepared with 100 pJ pulse energy and (a) 30 s, (b) 10 s and (c) 2.5 s irradiation time per spot. Patterns were prepared with 0.3 µm (top row), 0.5 µm (middle row) and 1.0 µm (bottom row) separation distance between the laser spots. Left, central and right columns correspond to the bidirectional, inside-out and outside-in pathways, respectively. Scale bar 1 µm.

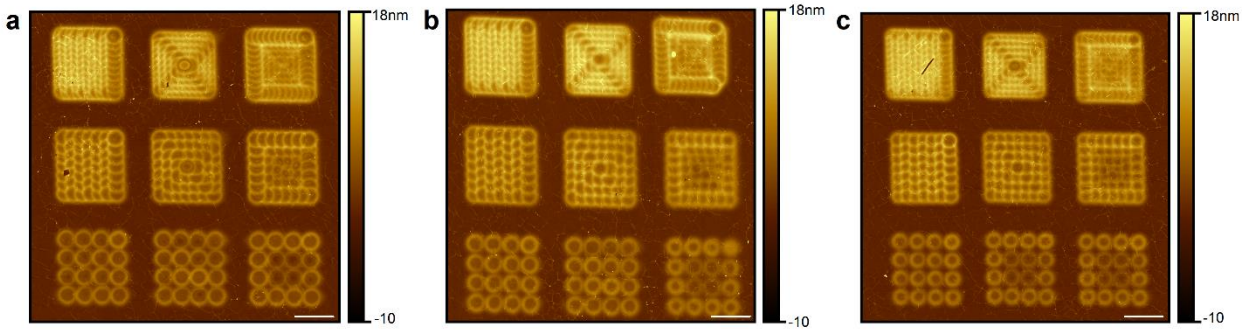


Figure 9. AFM height images of patterns prepared with 10 s irradiation time per spot and (a) 120 pJ, (b) 100 pJ and (c) 80 pJ pulse energy. Patterns were prepared with 0.3 µm (top row), 0.5 µm (middle row) and 1.0 µm (bottom row) separation distance between the laser spots. Left, central and right columns correspond to the bidirectional, inside-out and outside-in pathways, respectively. Scale bar 1 µm.