Supplementary Material (ESI) for *Soft Matter*

Supplementary Methods

Production of Fibronectin Fibrils by Manual Pulling of a Drop of Protein Solution

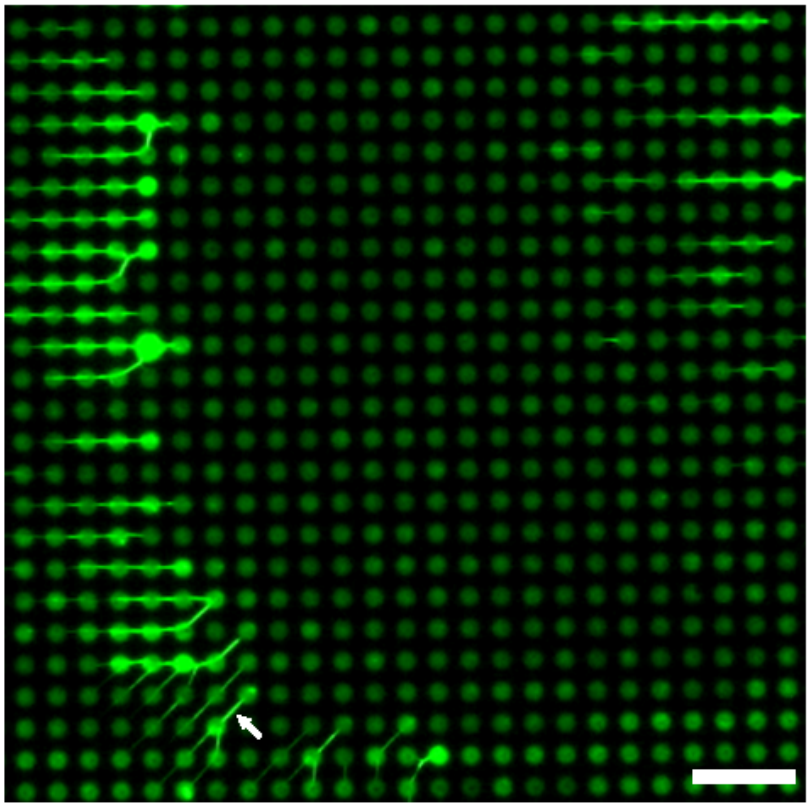
A drop (10 µl) of FN-ATTO 488 (200 µg ml^{-1} in PBS) was deposited onto a PDMS micropillar array using a micropipette. Capillary forces exerted by the micropipette tip were used to pull the drop into one direction at a speed of approximately 50 mm min^{-1}.

Western Blot analysis of ECM preparations

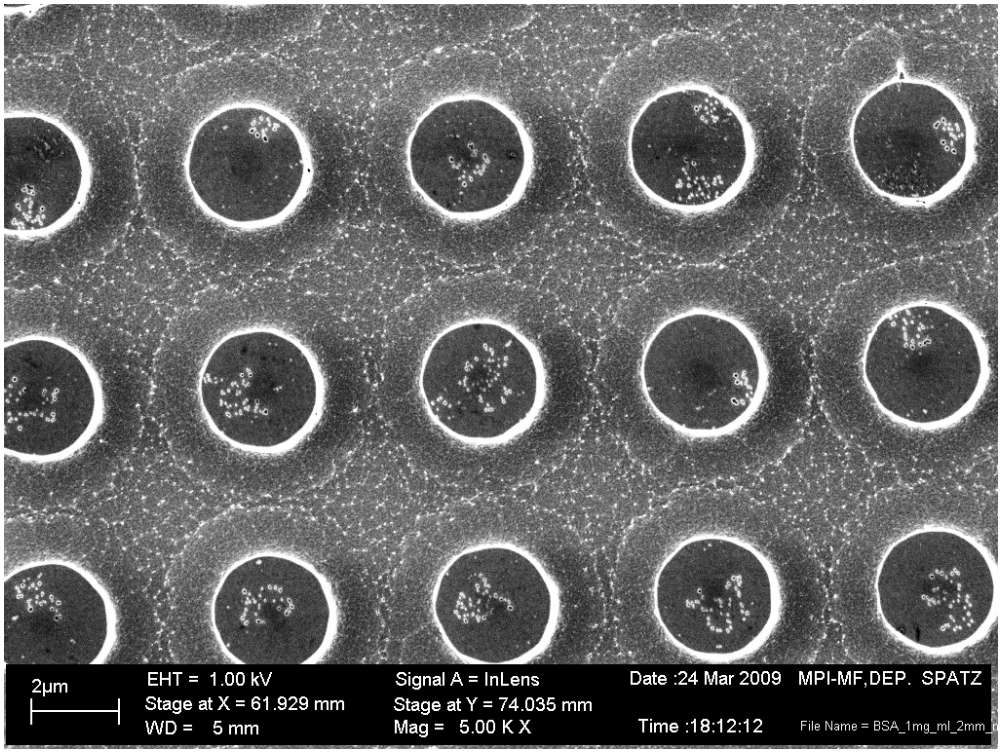
Electrophoretic separation and western blotting of ECM proteins under reducing conditions was performed using 4-20% polyacrylamide gradient gels according to the manufacturer's protocol (#25244, Pierce). After staining in water containing acetic acid (5 % v/v) and Ponceau S (0.1 % v/v), nitrocellulose membranes were photographed and de-stained in UltraPure water. Primary antibodies against LN (#L9393, SIGMA-Aldrich), COL I (#C2456, SIGMA-Aldrich) and FN (#F0791, SIGMA-Aldrich and #AB2033, Chemicon) were diluted 1:2000 in blocking buffer and incubated over night at room temperature. Secondary antibodies (#A11001 and #A21245, Invitrogen) were diluted 1:2000 in blocking buffer and incubated for 1 h. Samples were rinsed with Tris buffered saline (TBS) and imaged on an upright microscope (AxioImager, Carl Zeiss) using a 10x water dipping objective.

Production of Actin Nanofibrils

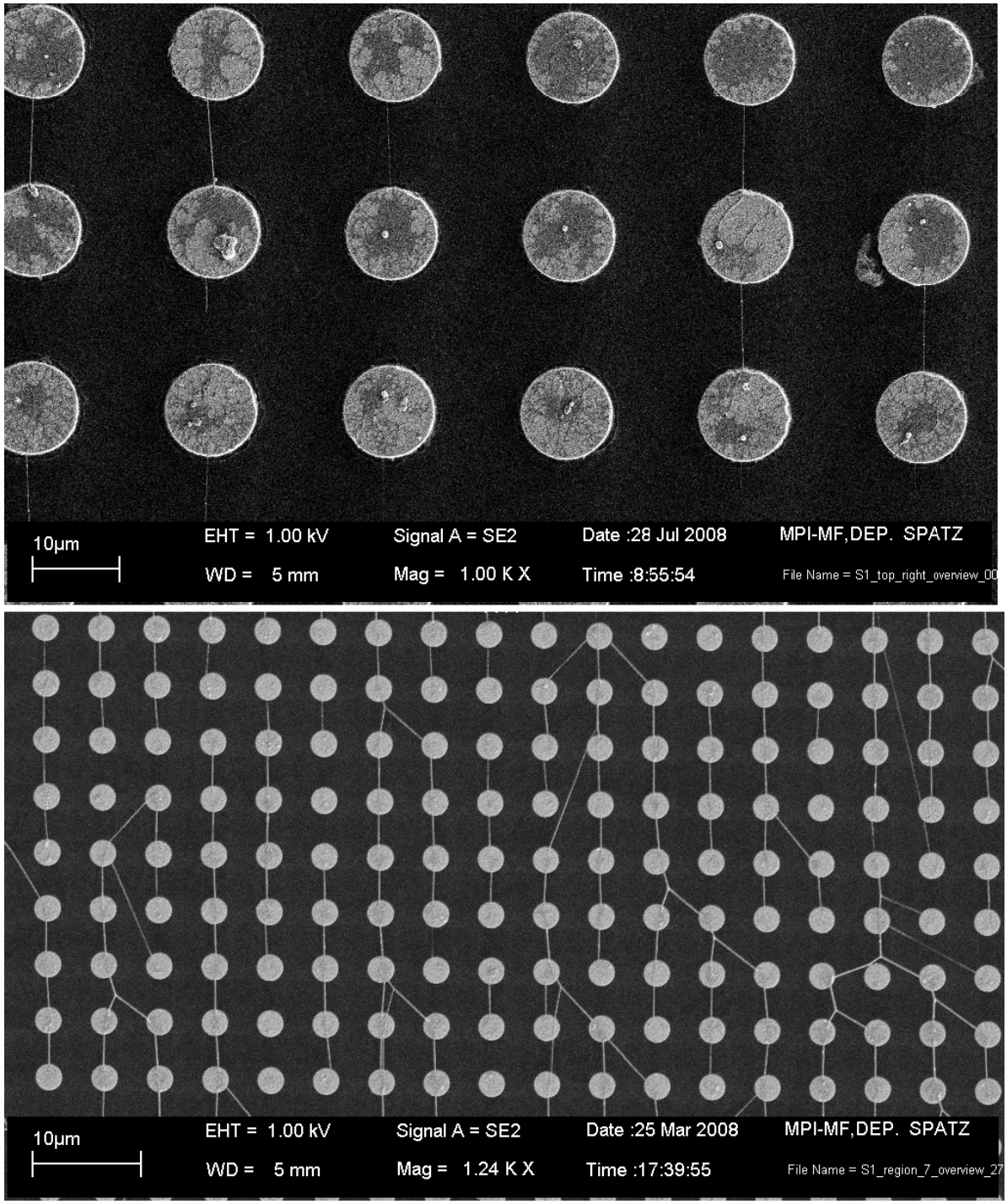
Actin monomers were applied at a concentration of 0.3 mg ml^{-1} in G-buffer (2 mM Tris, 0.2 mM $CaCl_2$, 0.2 mM ATP, 0.2 mM dithiothreitol, 0.005% NaN_3, pH 8.0). Nanofibrils formed by de-wetting at a pulling speed of 2 mm min^{-1}.



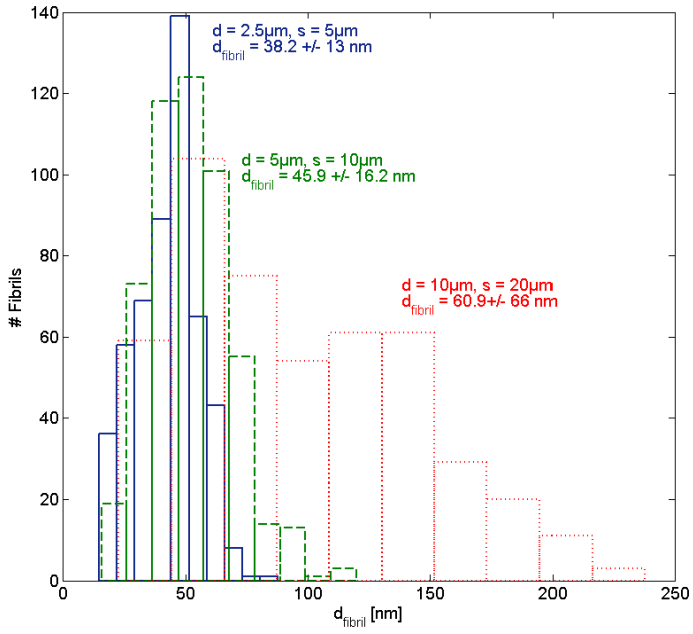
Supplementary Figure 1. Fibronectin fibrils produced by manually pulling a drop of FN over a poly(dimethylsiloxane) (PDMS) micropillar array. The fibrils at the curved area of the moving meniscus point towards the center of the droplet (white arrow). Scale bar: 30 µm.



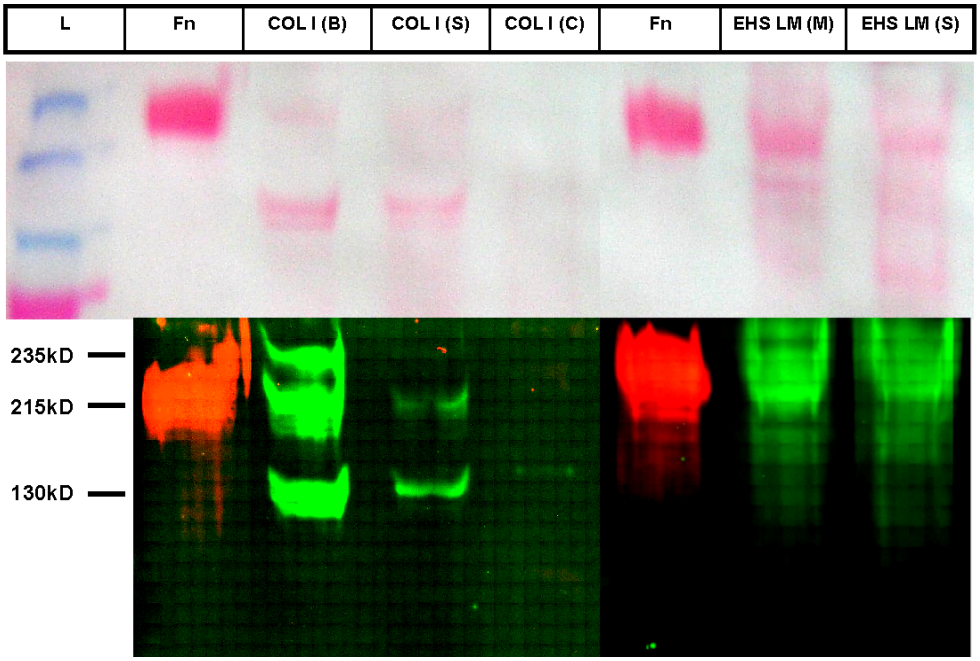
Supplementary Figure 2. Bovine serum albumin (BSA), even when employed at concentrations of 1 mg ml^{-1}, does not yield nanofibrils at a pulling speed of 2 mm min^{-1}.



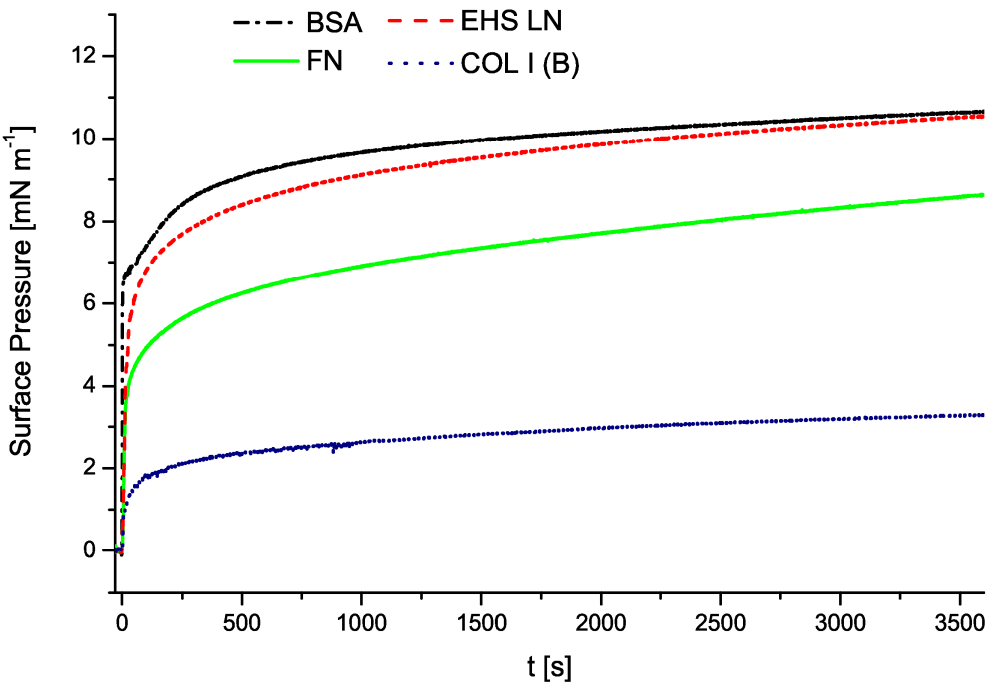
Supplementary Figure 3. TOP: A two-fold increase in FN protein concentration and 2-fold reduction of pulling speed leads to higher amount of irregular fibrils (white arrows). BOTTOM: Likewise, using 2-fold lower than optimal protein concentrations and 2-fold increased pulling speeds, FN nanofibrillar arrays show defects. FN was used at a concentration of 25 µg ml^{-1} (TOP) and 12.5 µg ml^{-1} (BOTTOM), respectively. In both experiments, the pulling speed was 10 mm min^{-1}. Note that the optimal protein concentration and pulling speed vary as a function of pillar geometry as shown in Fig. 2.



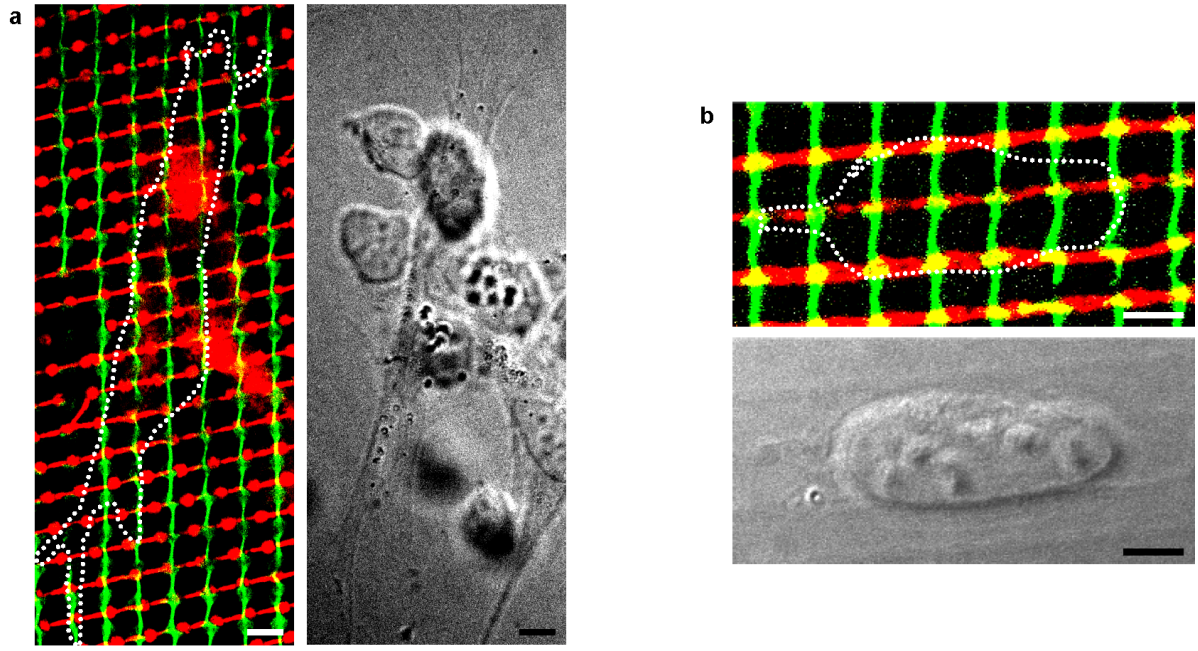
Supplementary Figure 4. Fibronectin nanofibril diameter is a function of pillar geometry. When using a FN concentration of 25 µg ml^{-1} and a pulling speed of 10 mm min^{-1} on different array geometries, the distribution of FN nanofibril diameters widens for pillar diameters smaller (blue) and larger (red) than 5 µm. Additionally, as shown in Supplementary Fig. 3, the coverage decreases on arrays with larger pillar diameter and cross-connections form on arrays with smaller pillar diameter.



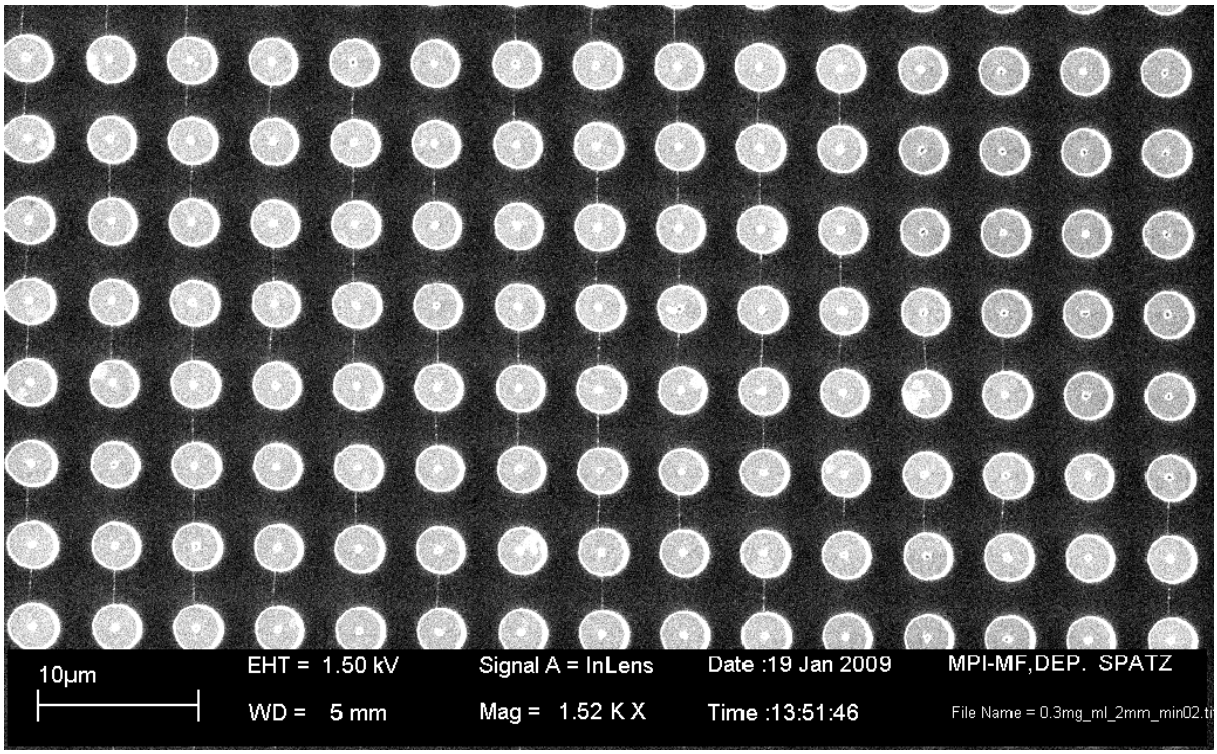
Supplementary Figure 5. Western Blot analysis of the protein preparations used in the experiments. TOP: Ponceau S stain containing the molecular weight ladder (L). BOTTOM: COL I (S) (green, left hand side) showed reduced amounts COL I alpha chains. No cross-contamination of either LN (green, right hand side) or COL I preparation with FN (red) could be detected using our immunofluorescence assay. Each lane was loaded with 2.67 µg of protein.



Supplementary Figure 6. The change in surface pressure as depicted in Fig. 4 was recorded over a time course of 1h. After a significant increase in surface pressure within 300 seconds, only slight changes in surface pressure occur.



Supplementary Figure 7. Differential cell adhesion on arrays with altered nanofibril spacing. (a) HFF cells align with FN nanofibrils on arrays with nanofibril spacings of 2.5 µm. (b) SH SY5Y cells show elongation along LN nanofibrils on crossed arrays with the same geometry as in (a). Scale bar. 5 µm.



Supplementary Figure 8. Actin nanofibrils formed after de-wetting of a silicon micropillar array with d = 2.5 µm pillar diameter and s = 5 µm center-to-center distance. While the nanofibril diameters are uniform, array coverage was not optimal in this preliminary experiment.