Supporting Information

Ordered Particle Arrays via a Langmuir Transfer Process: Access to Any Two-Dimensional Bravais Lattice

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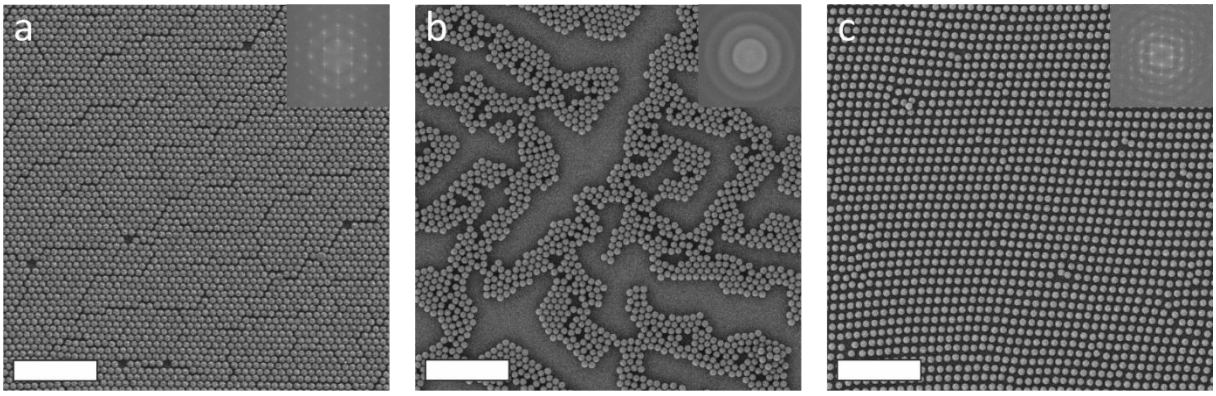

Figure S1. SEM images of particle monolayers on hydrophilic and hydrophobic substrates. The insets show the corresponding fast Fourier transform (FFT). (a) Particles on a hydrophilic glass substrate (*hcp* structure). (b) Particles on a hydrophobic substrate without thermal fixation (collapsed line structure). (c) Particles on a hydrophobic substrate with thermal fixation (non-close-packed oblique structure). Scale bars are 5 µm.

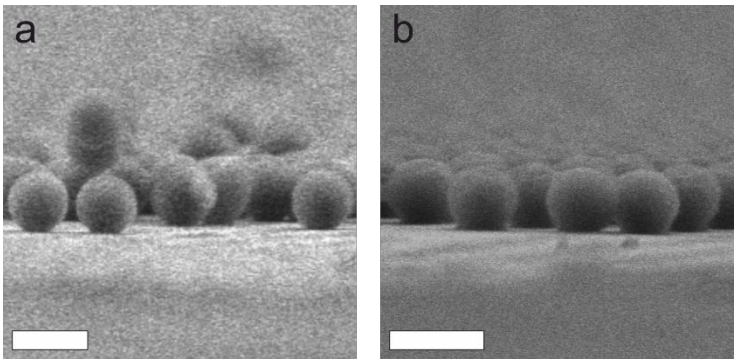

Figure S2. Side view SEM images of (a) non-immobilized and (b) immobilized particles. Without thermal annealing, the particles can be viewed as hard spheres which feature a minimum contact area on the substrate. Upon drying of the monolayer, these particles are subject to capillary forces that lead to a shift to new particle positions. Thermal annealing of the particles above their glass transition temperature T_g leads to an increase of the contact area on the substrate. In the viscous state, the particles fuse onto the substrate and become immobilized at their positions even under capillary stress. Scale bars are 500 nm.

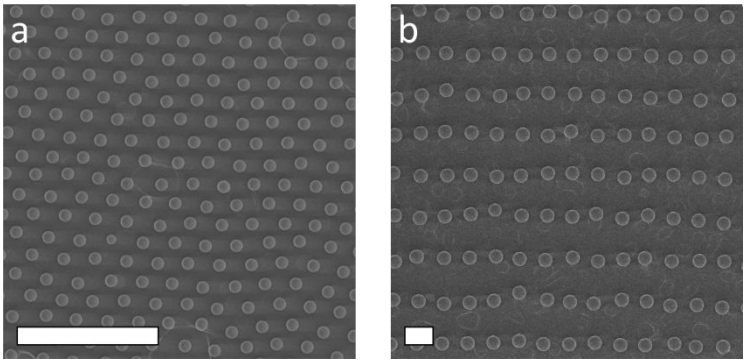

Figure S3. SEM images of particle monolayers immobilized on hydrophobic substrates with moderate glass transition temperature. PS Particles (T_g ≈ 105 °C) with a diameter of (a) 453 nm and (b) 2560 nm thermally fixated on spin-coated *P(nBA-co-MMA) layer* (T_g ≈ 55 °C). Scale bars are 5 µm.

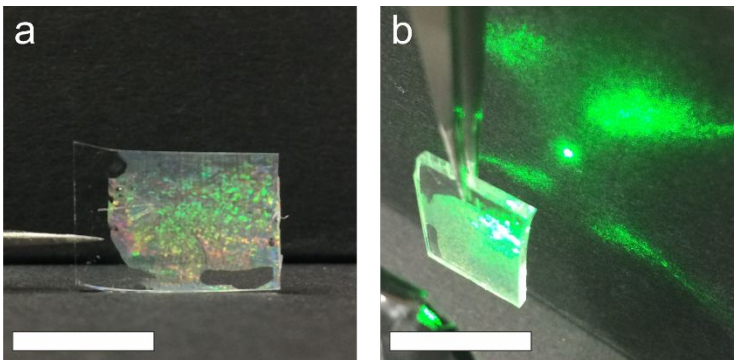

Figure S4. Immobilized particle monolayer. (a) Photograph of an immobilized particle monolayer on a hydrophobic substrate. The iridescent color preserved after drying of the monolayer indicate an intact positional arrangement of the particles. (b) Laser diffraction of an immobilized particle monolayer. The four-fold symmetric peaks confirm the presence of an extended particle domain with square symmetry. Scale bars are 1 cm.

Calculation of δ, α and S

Experimentally found structures can be assigned to specific stretching parameters δ, α and S. For this the stretched vectors $\overrightarrow{a'}$ and $\overrightarrow{b'}$ with the coordinates x_a', y_a' and x_b', y_b' have to be extracted from SEM images. The coordinates are inserted into the following two equations to calculate α and S. In order to determine the right parameters, the results of both $\overrightarrow{a'}$ and $\overrightarrow{b'}$ have to be screened with respect to δ.

$$\alpha = arccot \frac{y' - y(\delta)}{x' - x(\delta)} \quad \text{(S1)}$$

$$S = \frac{x' - x(\delta) \cdot cos^2 \alpha + y(\delta) \cdot sin \alpha \cdot cos \alpha}{x(\delta) \cdot sin^2 \alpha + y(\delta) \cdot sin \alpha \cdot cos \alpha} \quad \text{(S2)}$$

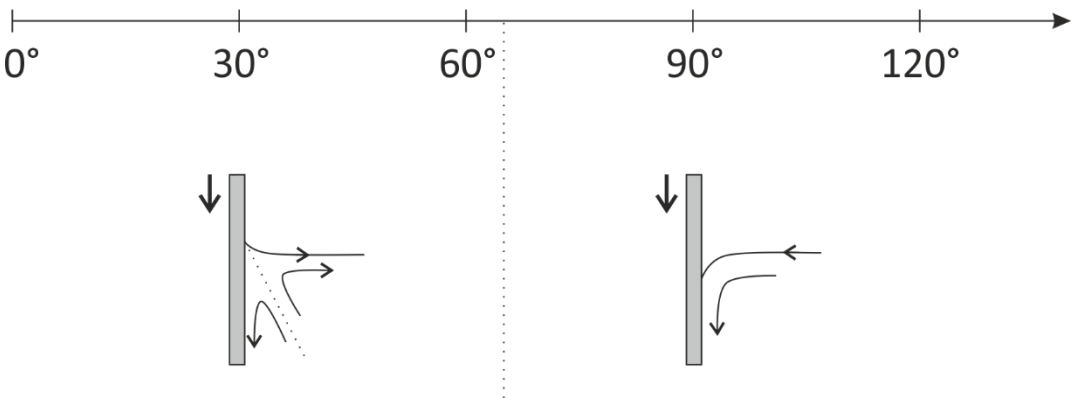

Figure S5. Streamline profiles upon immersion of a solid substrate at different contact angles. (29) The dotted line indicates the critical angle θ_c below which an X-type is impossible.

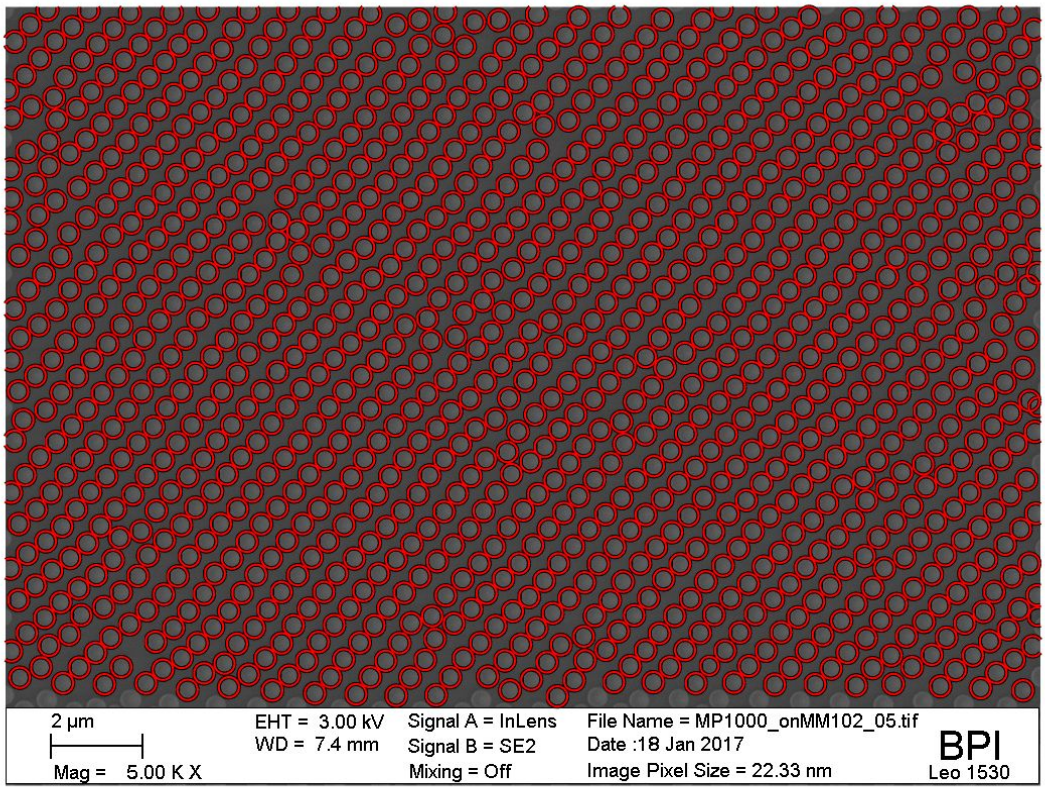

Figure S6. SEM image of a stretched monodomain. The red circles indicate the particles found by MATLAB's circle detection function after optimizing the search parameters manually.

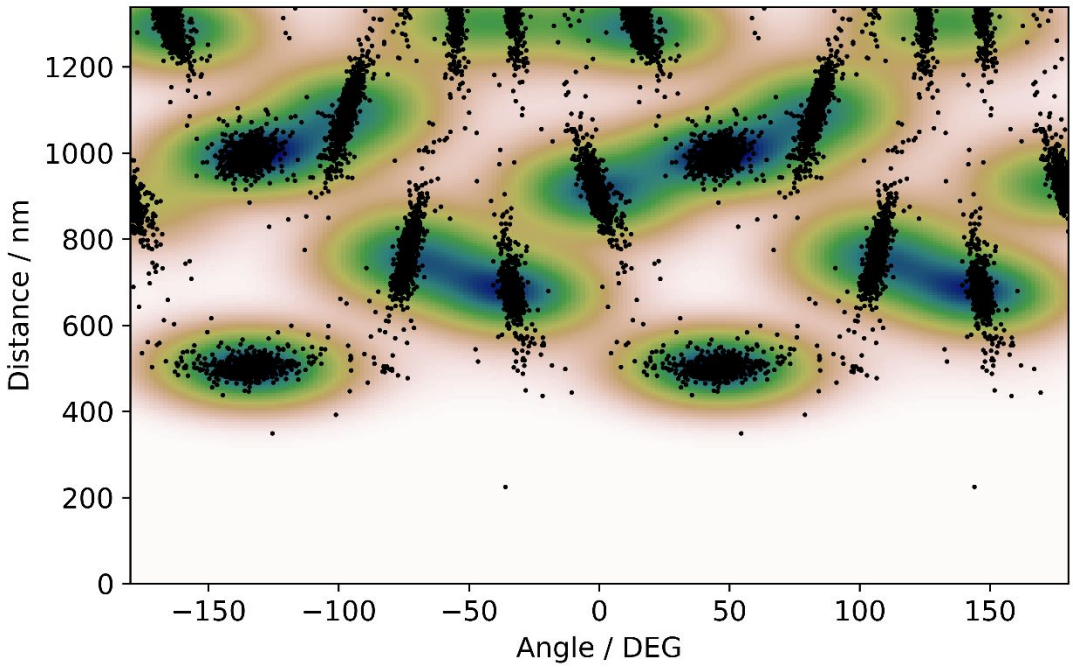

Figure S7. 2D Histogram of the distance between particle pairs and the corresponding angle with the x-axis for the SEM image in Figure S6. Each black dot represents an inter-particle connection. A Gaussian kernel density estimator is used to calculate the smooth distribution function (color). Local maxima in the distribution, with small interparticle distances, represent possible candidates for the stretched basis vectors $\overrightarrow{a'}$ and $\overrightarrow{b'}$ in polar coordinates.