Supporting Information

Moiré and honeycomb lattices through self-assembly of hard-core/soft-shell microgels: Experiment and simulation

Kirsten Volk,[a] *Florian Deißenbeck,*[b] *Suvendu Mandal,*[b] *Hartmut Löwen,*[b,*] *Matthias Karg*[a,*]

[a]Institut für Physikalische Chemie I: Kolloide und Nanooptik, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany

[b]Institut für Theoretische Physik II: Weiche Materie, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany

* E-Mail: hlowen@thphy.uni-duesseldorf.de; karg@hhu.de

Void space in single hexagonal monolayer

Figure S1 illustrates the void space in between three neighboring particles of a hexagonal monolayer with a center-to-center nearest neighbor distance $d_{c\text{-}c}$. The yellow circles illustrate the rigid cores of the core/shell particles with the green surrounding circles being representative of the soft, deformable hydrogel shells. In the dry state the core/shell particles have a diameter d_{dry}. The void can accommodate a spherical particle with a footprint diameter d_{void}.

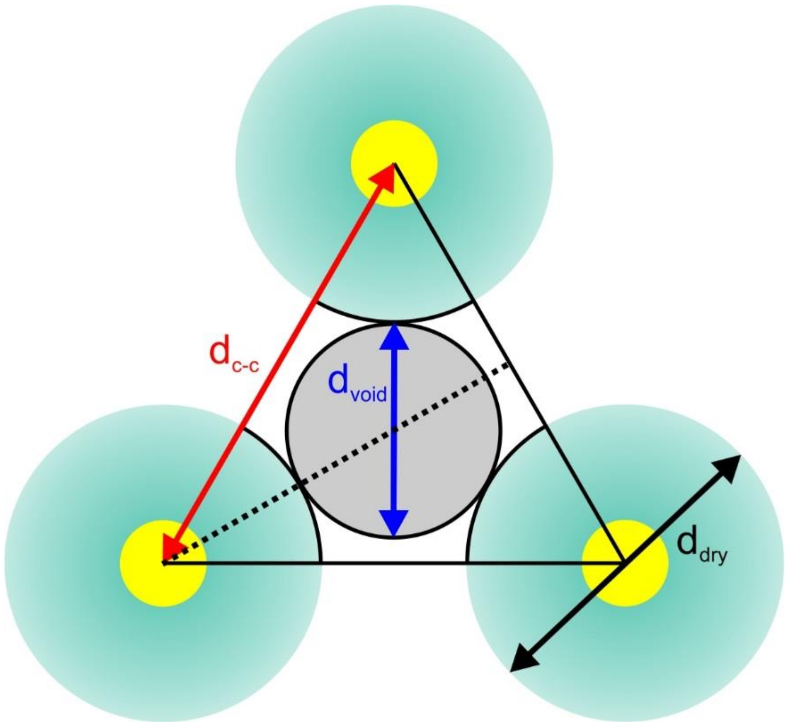

Figure S1: Schematic depiction of void space with the diameter d_{void} in the center of three dry core/shell particles with a diameter d_{dry} and a center-to-center distance of $d_{c\text{-}c}$.

The length l of the dashed line (altitude) in figure S1 is:

$$l = \frac{\sqrt{3}}{2} d_{c-c} \qquad \textbf{(S1)}$$

The center of the grey circle divides l in a 2:1 ratio. Correspondingly the radius of the grey circle r_{void} = ½ d_{dry} is $\frac{2}{3}l$ minus the dry core/shell particle radius r_{dry} = ½ d_{dry}. Thus:

$$r_{void} = \frac{2}{3}\frac{\sqrt{3}}{2} d_{c-c} - r_{dry} = \frac{1}{\sqrt{3}} d_{c-c} - r_{dry} \qquad \textbf{(S2)}$$

The void diameter is then given by:

$$d_{void} = \frac{2}{\sqrt{3}} d_{c-c} - d_{dry} \qquad \textbf{(S3)}$$

Hexagonal sublattices in honeycomb and Moiré lattices

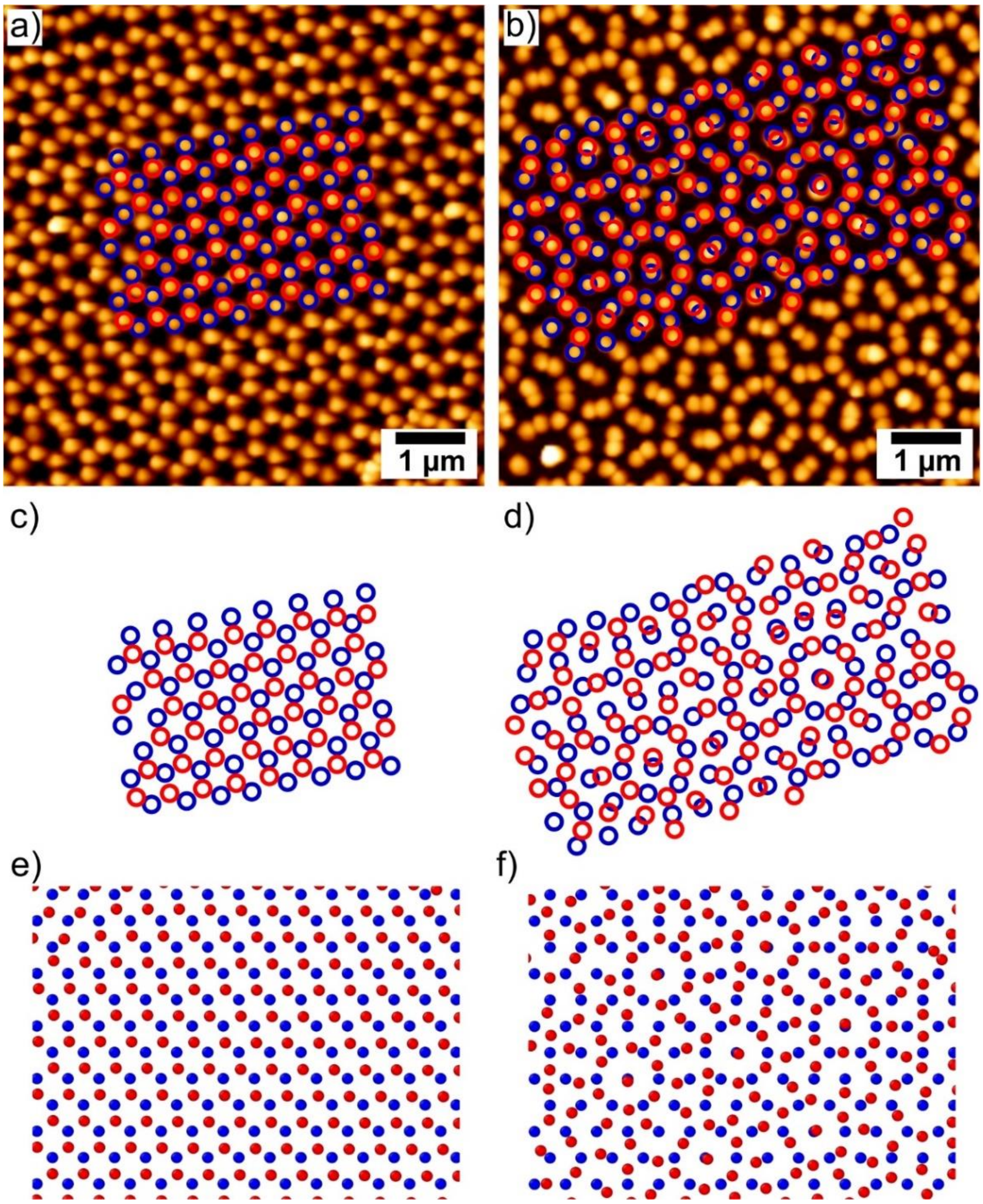

Figure S2. Visualisation of the two hexagonal sublattices (blue and red circles) in AFM images of (a) the honeycomb microstructure and (b) the Moiré lattice. In (c) and (d) the indicated sublattices are shown without the underlying AFM image for better visibility. (e) and (f) show the two sublattices from BD simulations.

Influence of rotation angle on Moirè lattices

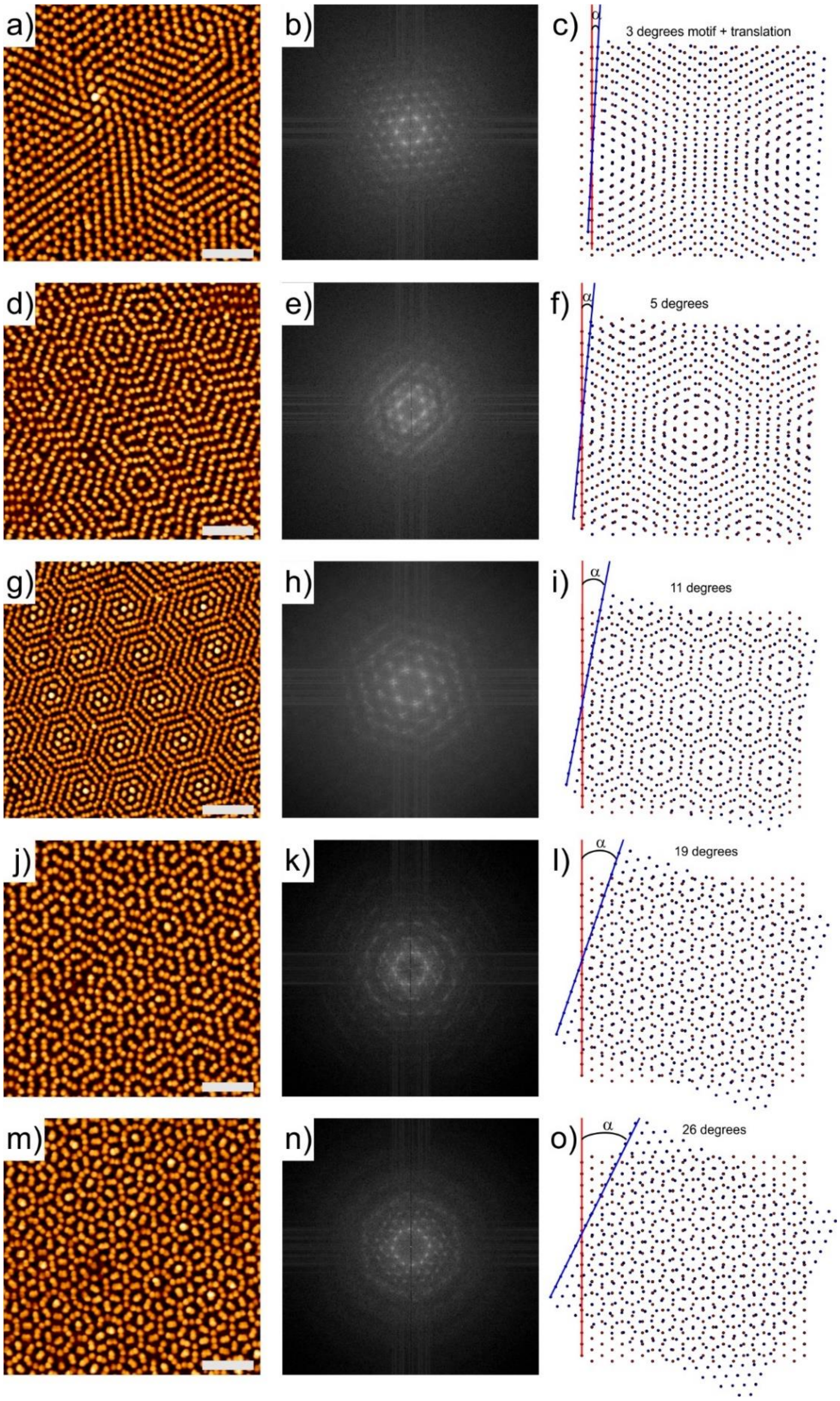

Figure S3: Characterisation of different Moiré motifs: AFM height images (a, c, g, j, m) and respective FFTs (b, e, h, k, n) and sketches of two overlaid perfect hexagonal patterns with angle of rotation α matching the experiment to give Moiré motifs (c, f, I, l, o).

Cross-section analysis of Moiré lattices

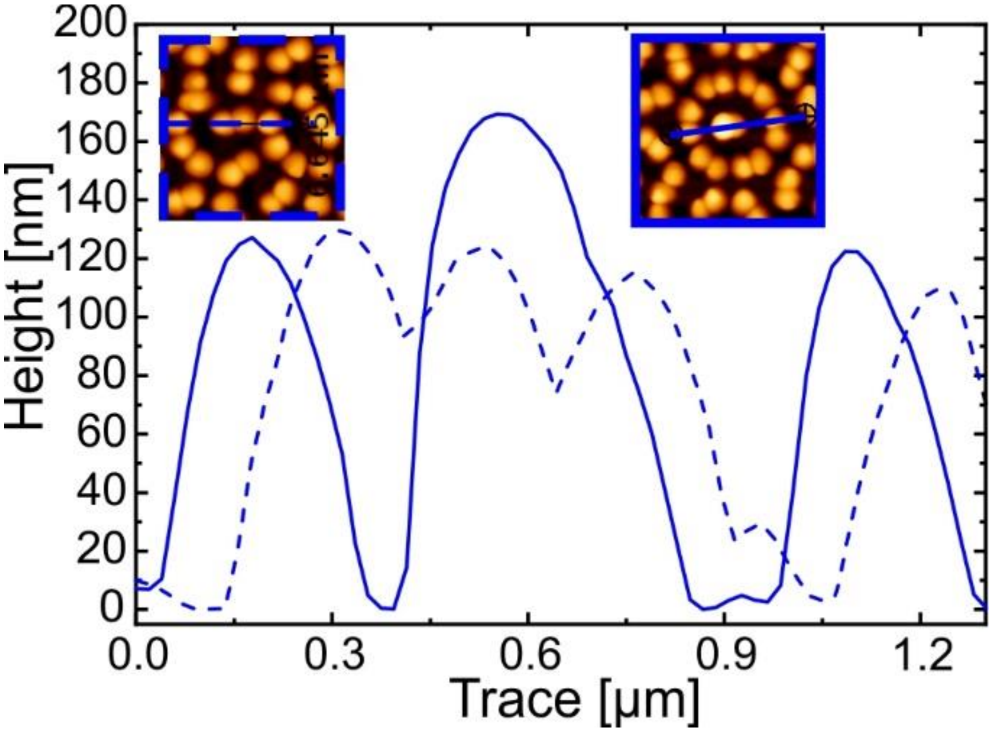

Figure S4: Cross-section analysis of an experimental Moiré structure along different trajectories. For the dashed line all particles sit next to each other in a single plane. The solid line cross-section crosses a position where two particles are deposited on nearly the same spot and thus the particle from the second deposited layer sits on top of the underlying particle from the first monolayer. In the latter case the upper particle is sterically hindered to settle into the underlying monolayer plane. Thus the cross-section shows a significantly increased total height at this particular position. Insets depict the corresponding AFM height images with the traces used for the cross-sections.