

Supporting information section

1. Monolayer Particle Interaction Apparatus (MPIA)

The MPIA consisted of a Langmuir trough (Riegler & Kirstein GmbH, Potsdam, Germany) and a force measuring unit, see supplementary Fig. 1. The Langmuir trough was coated with poly(tetrafluoroethylene) (PTFE) and was equipped with two PTFE barriers that compressed around the centre of the trough. The temperature of the subphase was maintained at $20.0 \pm 0.1^\circ\text{C}$ by circulating thermostatted water through the base of the trough (C25P, ThermoHaake, Karlsruhe, Germany). The base of the film balance contained an exchangeable glass window that allowed the colloid probe to be attached to the force measurement unit, which was below the Langmuir trough.

In order to measure the forces, a colloid probe was attached to the cantilever holder (PEEK, MPIP, Germany), which was passed through a small hole that was in the glass window in the base of the Langmuir trough. Water did not leak from the trough when the cantilever holder was moved up and down in a force measurement, due to the presence of an o-ring (Viton-Seal $5 \times 9 \times 2$, DFV-Dichtungen, Berlin, Germany) on the inside of the hole. The lower end of the cantilever holder was clamped to a closed loop piezo translation stage (model P-753.21C, $25 \mu\text{m}$ range, Physik Instrumente, Karlsruhe, Germany). The cantilever was mounted at an angle of 21° on the cantilever holder, ensuring that only the tip of the cantilever, i.e., the particle that was glued to the cantilever, was brought in contact with the air/water interface. The distance between the colloid probe and the monolayer was changed by moving the cantilever holder up and down in the z -direction by applying a voltage to the digital piezo controller (model E-661.CP). The forces between the colloid probe and the monolayer were measured in the

aqueous subphase by measuring the deflection (Δx) of the cantilever as a function of piezo position with a laser beam ($\lambda = 658 \text{ nm}$, $P_{out} \leq 5 \text{ mV}$, FKP-620-23, Schäfter + Kirchoff, Hamburg, Germany) that was reflected from the under-side of the gold coated cantilever onto a position sensing detector (PSD, L2L20SP, Laser Components, Olching, Germany). The detector voltage signal was converted to cantilever deflection in nanometers in the following way. A mica plate that was glued to a glass substrate was placed across the top sides of the Langmuir trough, which was filled with water. The probe was brought into contact with the mica and a minimum of 100 force curves were recorded. The constant compliance region of those force curves were determined, i.e., the region where the probe was in constant contact with the mica. The slope values of that region were averaged and the resulting value was used to convert the deflection signal (Δx) from volts to nanometers for the force curves measured between the monolayer and the probe. The force (F) between the monolayer and the probe was then calculated using Hooke's law, $F = k\Delta x$, where k is the spring constant of the cantilever. Zero force was defined at large cantilever-substrate separations, where no surface forces were acting on the cantilever.

Supplementary Fig. 1. Schematic diagram (A) and photo (B) of the Monolayer Particle Interaction Apparatus (MPIA).

