Electronic Supplementary Material (ESI) for Soft Matter.

AFM Methods

A monolayer of 5.2 nm Au-NPs (purchased from Ocean Nanotech) was formed by depositing 10 µL of 1 mg/mL suspension onto a 100µL de-ionized water droplet over a carbon grid with 2 µm diameter holes (Quantifoil 657-200-CU from Ted Pella) on a Teflon substrate. At the air-water interface, the Au-NPs self-assemble into an ordered monolayer. Eventually, the water droplet evaporates and the Au-NP monolayer is deposited onto the grid, suspending over the holes and creating free-standing membranes of particles. The grid was then transferred to a 15 mm metal specimen disc for atomic force microscopy (AFM) measurements of the membranes' mechanical properties. AFM was performed using an Asylum Cypher ES atomic force microscope. Tapping-mode AFM imaging was used to locate the fully suspended membranes and then contact-mode AFM was used to perform force curve measurements. For these, the AFM tip was lowered to the center of the membrane, increasing the van der Waals interaction force F, which was measured along with the resulting indentation δ caused in the membrane.

For a two-dimensional sheet such as a nanoparticle monolayer, these force curves have been shown to fit to the equation $F = \sigma^{2D}(\pi)\delta + E^{2D}\left(\frac{q^3}{R^2}\right)\delta^3$.[1, 2] Here, σ^{2D} is the prestress of the membrane, set into the particles by factors such as particle synthesis and sample preparation. The Young's modulus can be extracted from the cubic term. In this term, R is the membrane radius, E^{2D} is the 2-D Young's modulus, and q is a function of the Poisson ratio, which for Au-NP thin sheets has been measured directly.[3] Once the cubic coefficient is fitted and the E^{2D} is extracted, the traditional 1-D Young's modulus can be obtained by dividing E^{2D} by the height h of the sample. In this case h is the diameter of the particle core (5.2 nm) plus two dodecanethiol ligand lengths (1.7 nm each), pictured as standing straight out from both the top and bottom of a particle. 16 force curves from 13 membranes on this sample were analyzed, and the Young's modulus was measured to be E = 2.8 ± 0.4 GPa. The uncertainty given here is the standard deviation of moduli extracted from each force curve fit individually.

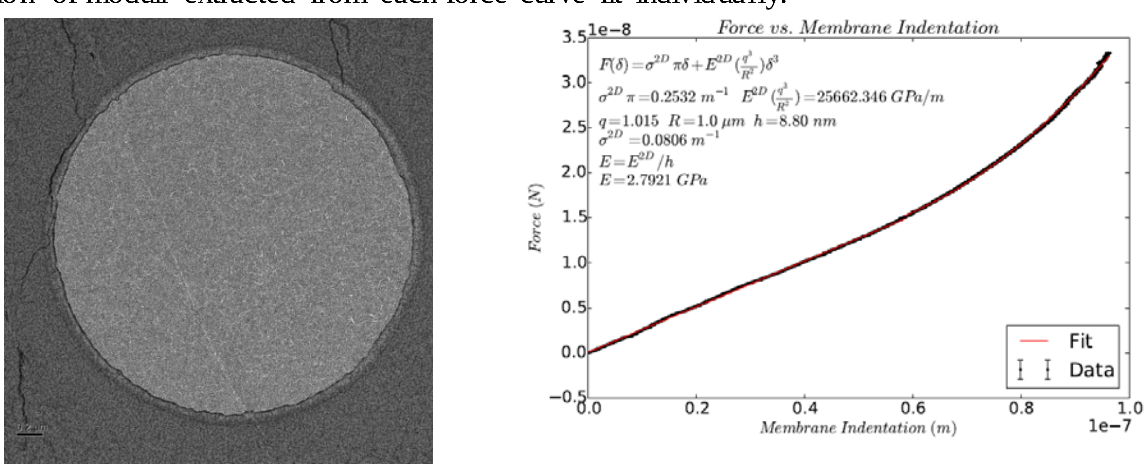

Fig. 1: a) A TEM image of an Au-NP monolayer suspended over an AFM grid hole. b) A force curve created from the average of the 16 different force curves collected from the sample, fitted to the above-mentioned equation to extract the average Young's moduli of the data.

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[1] C. Lee, X. D. Wei, J. W. Kysar, J. Hone, *Science* 2008, **321**, 385-388.
[2] J. He, P. Kanjanaboos, N. L. Frazer, A. Weis, X. M. Lin, H. M. Jaeger, *Small* 2010, **6**, 1449-1456.
[3] P. Kanjanaboos, A. Joshi-Imre, X. M. Lin, H. M. Jaeger, *Nano Lett.* 2011, **11**, 2567-2571.