2D “SOAP”-ASSEMBLY OF NANOPARTICLES VIA COLLOID-INDUCED CONDENSATION OF MIXED LANGMUIR MONOLAYERS OF FATTY SURFACTANTS

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Figure S-1. UV-Vis absorbance spectrum of 1-layer quartz-supported film of SA/ODA monolayer with adsorbed AuNP's deposited onto solid support after adsorption completion; surface pressure of LB transferring was of 1.5 mN/m.

Figure S-2. Stability isotherms for SA/ODA monolayers at different initial surface pressures: (1) 20 mN/m; and (2) 10 mN/m; the film-covered surface area was held constant. The horizontal dashed lines and vertical arrows indicate the relative decrease of surface pressure $\Delta\pi = \pi_{\text{compressed}} - \pi_{\text{equilibrium}}$.

Figure S-3. In situ UV-vis absorbance spectra of single-component SA monolayer initially compressed up to 30 mN/m on gold hydrosol at constant surface area; time is indicated by a vertical arrow.

Figure S-4. Plots of SPB intensity maximum vs time for ODA (black) and SA/ODA (red) monolayers on gold hydrosol. For corresponding spectra, see Figure 2a and 2b, respectively (main text).

Figure S-5 Schematics illustrating the rearrangement of ODA monolayer during spreading and further equilibration on a surface of gold colloidal solution. (a, b) Adsorption of AuNPs to freshly spread drops of ODA solution on hydrosol surface after solvent evaporation; (a) top view and (b) side view. This initial stage of monolayer formation corresponds to line 1 with intensity maximum at 660nm in the spectrum in Fig. 2a. (c, d) Spreading of ODA with adsorbed particles over the available surface area along the gradients of concentration and surface tension until uniform distribution of a material on a surface is achieved; (c) top view, and (d) side view. This stage corresponds to a blue-shift of SPB accompanied with an increase of intensity in

Fig. 2a. (e, f) Aggregation of ODA-coated nanoparticles through Van der Waals interactions (top views). This stage corresponds to a reversed red-shift of SPB in Fig.2a

Figure S-6. SEM image of 1-layer solid supported film of ODA monolayer with adsorbed AuNP's deposited onto silicon wafer at surface pressure of 14-15 mN/m.

Figure S-7. In situ UV-vis absorbance spectra of compressed SA/ODA monolayer with constant surface area on a surface of gold hydrosol. Spectra 1 and 2 correspond to surface pressures of 0 mN/m (before compression) and 20 mN/m (after compression). Time is indicated by a vertical arrow; the spectra were recorded every 20 minutes. The dashed lines with horizontal arrows illustrate the red shifts of SPB.

Figure S-8. In situ UV-vis absorbance spectra of compressed SA/ODA monolayer on gold hydrosol. Spectra 1 and 2 correspond to surface pressures of 0 mN/m (before compression) and 10 mN/m (after compression). Time is indicated by a vertical arrow; the spectra were recorded every 20 minutes. The dashed lines with horizontal arrows illustrate the red shifts of SPB.

Figure S-9. Large-scale SEM microphotographs correspond to Fig.6 in the main text. Two different morphologies were found within the same one-layer SA/ODA film deposited after the adsorption completion from a surface of gold hydrosol onto silicon wafer; surface pressure of LB transferring was of 1.5 mN/m.

Figure S-10. SEM microphotographs of one-layer SA/ODA films deposited after the adsorption completion from a surface of gold hydrosol onto freshly prepared surfaces of (a) graphite and (b) mica; the surface pressure of LB transferring was of 1.3-1.5 mN/m.

Figure S-11. SEM microphotographs illustrate the presence of both types of morphologies in 2D “soap”-gels transferred onto freshly prepared surfaces of (b, c) mica and (a, d) graphite; the surface pressure of LB transferring was of 1.3-1.5 mN/m.

Figure S-12. (a) AFM image of the surface of 5-layers of 2D soap-gel sequentially deposited onto silicon wafer at 30 mN/m after the recompression, and (b) corresponding large-scale SEM microphotograph; the portion of the film was preliminary removed by the AFM tip.

Figure S-13. AFM image of nanoparticles assemblies stabilized with SA/ODA and deposited onto quartz plate after recompression to 30 mN/m. (i) – nanoparticle cluster, (ii) – patch of the pure organic phase. Grey-scales correspond to 45 nm (a) and 140 nm (b)

Figure S-14. TEM micrographs of crystallites in one-layer film formed under continuous compression and deposited onto silica surface at 30mN/m.

Figure S-15. TEM microphotograph and corresponding SAED patterns for (a, b) gold hydrosol and (c, d) AuNP-free crystallites formed in one-layer film, which was exposed to gold hydrosol at constant surface pressure of 30 mN/m.

Figure S-16. (a) TEM micrograph of the dendrite "leg". The sample shows the portion of nanoparticle-covered crystalline phase originally deposited onto silica and re-transferred on formvar-coated TEM grid. (b) AFM image of dendrite surface in the 9-layer film. Greyscale corresponds to 120 nm.

Figure S-17. Dark and bright field optical microphotographs of dendrite-like crystallites formed in the gold-supported film depending on the number of deposited layers: a) 3-layer, b) 5-layer, c) 7-layer, d) 9-layer and e) 19- layer film.

Figure S-18. Optical image of crystallites in 11-layer film of separated 2D “soap”-gel deposited at constant lateral pressure of 30 mN/m onto gold polycrystalline support

Figure S-19. (a) Optical image of the AFM investigated area of the sample. (b) Corresponding AFM scan of the dendrite-like crystallite.

Figure S-19. In situ UV-vis absorbance spectra of initially compressed to 30 mN/m SA/ODA (mixing ratio 1:1) monolayer on gold hydrosol at constant surface area. Time is indicated by a vertical arrow; the dashed lines with horizontal arrows illustrate the red shifts of SPB.