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Very large two-dimensional superlattice domain of monodisperse gold nanoparticles by self-assembly

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Details of the 2D self-assembly process

As shown in Fig. S1, gold nanoparticles with a special property for 2D self-assembly float to the air-toluene interface of a toluene droplet and form a monolayer film. The floating is immediate if the toluene droplet is larger than the hexane droplet initially containing gold nanoparticles (Scheme 1 in the main text) and slower if the hexane droplet is of size similar to or larger than the toluene droplet [1]. Hexane evaporates ~4 times faster than toluene. A nearby water droplet is pushed to the toluene droplet that surrounds it. Then, gold nanoparticles are transferred from the air-toluene interface to the air-water interface. The color of the gold nanoparticle monolayer film is red-purple at the air-toluene interface and gold nanoparticles might be in the 2D liquid state, meaning that nanoparticles are not close-packed and there are some thermal movements of individual nanoparticles at the 2D interface between air and toluene. In contrast, the color is blue-purple at the air-water interface, meaning close-packed gold nanoparticles that might be in the 2D solid state. Fragmented 2D plates of gold nanoparticles in Fig. S2 further support the hypothesis of the 2D solid state at the air-water interface. While individual microspheres can be spatially resolved in real-time by optical microscopy for phase transition between 2D liquid and solid [2], no electron or X-ray microscopy is yet available for resolving individual nanoparticles' thermal motions at the air-liquid interface of a liquid droplet. Instead, we plan to do some grazing-incidence small angle X-ray scattering (GISAXS) experiments to verify the 2D liquid or solid states of gold nanoparticles at the air-liquid interfaces. For scanning electron microscopy of gold nanoparticle superlattice domains, we put a water droplet on a silicon piece of 5x5 mm^2 area, a toluene droplet around the water droplet, and a hexane droplet with gold nanoparticles in it. After evaporation of hexane, toluene, and water in this order, the monolayer film of gold nanoparticles is very gently deposited onto the silicon substrate.

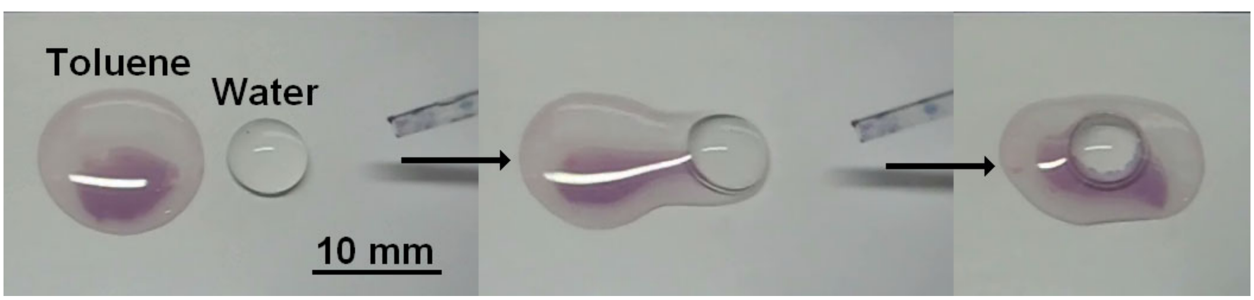

Fig. 1S Gold nanoparticles of red-purple color and in 2D liquid-like state at the air-toluene interface are transferred to the air-water interface where they are of blue-purple color and in 2D solid-like state (images taken from a real-time movie).

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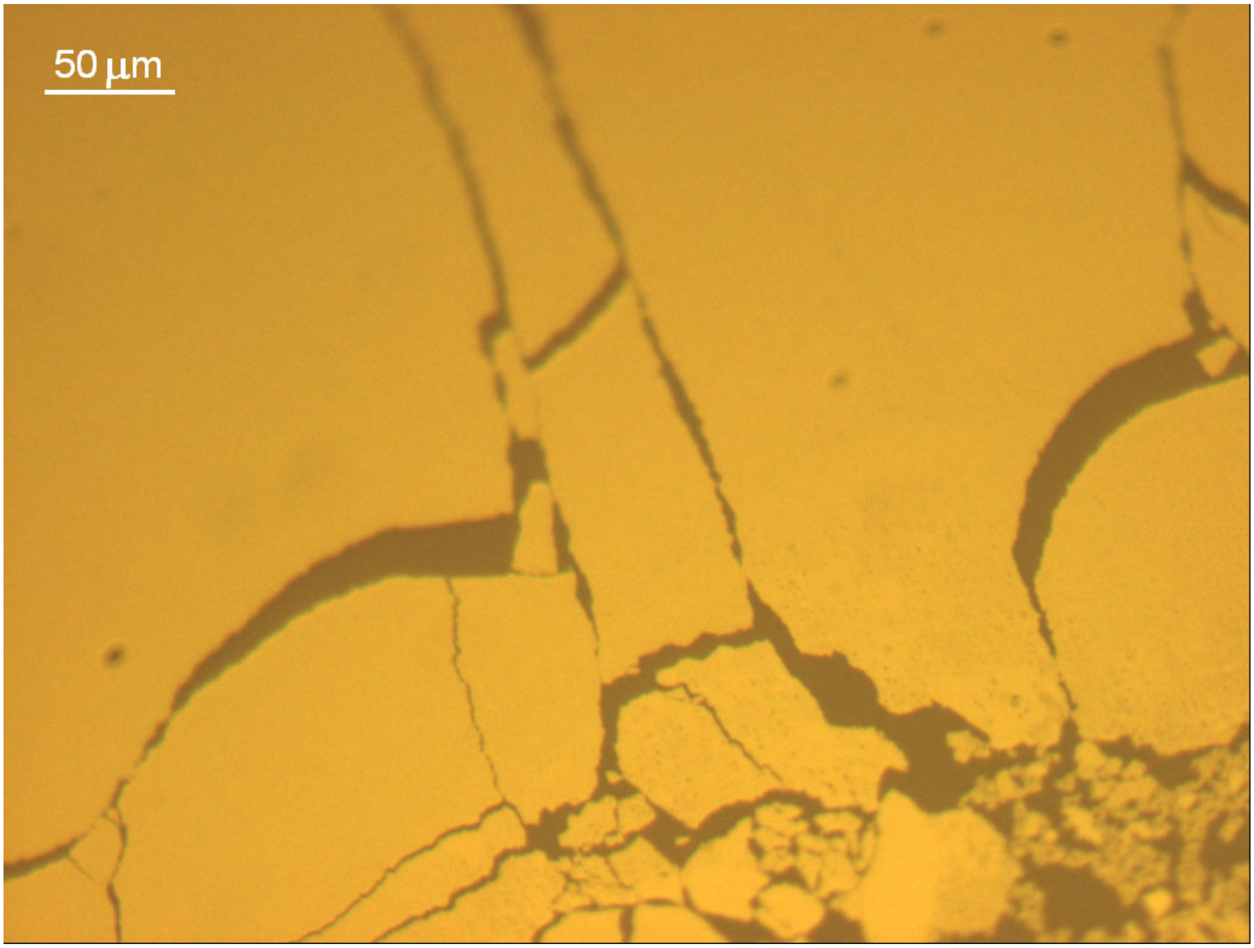

Fig. 2S Fragmented 2D plates of gold nanoparticles at the air-water interface.

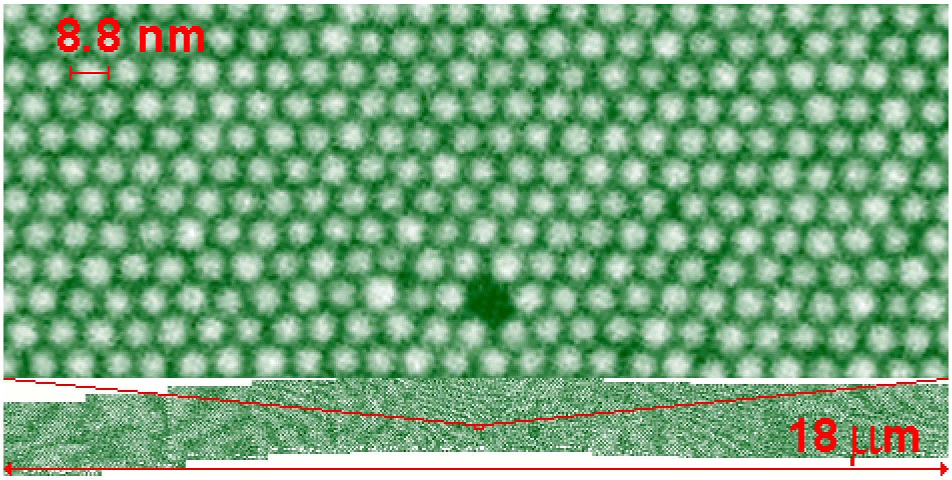

Fig. 3S Whole SEM image of the 2D superlattice domain with spatially resolved individual gold nanoparticles and defects such as single vacancy and dislocation, available as ESI to be downloaded (19500 x 2037 pixels, 33 Mbyte).

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Movie of Fig. 1S and the whole image of gold nanoparticles in Fig. 1d

Fig. 1S are images taken from a real-time movie (4.5 Mbyte, mpg format) that is also available as ESI in addition to the whole SEM image (19500 x 2037 pixels, 33 Mbyte, gif format) of the 2D superlattice domain of 18 μm long with spatially resolved, 360,000 individual nanoparticles and defects such as single vacancy and dislocation (Fig. 3S). If the ESI gif image file (33 Mbyte) is so large that there are some difficulties in downloading and watching it, a 7-min video for zooming into and panning the huge image is available at YouTube (search keyword: EahLab, "http://www.youtube.com/watch?v=vLudYhwXyAM").

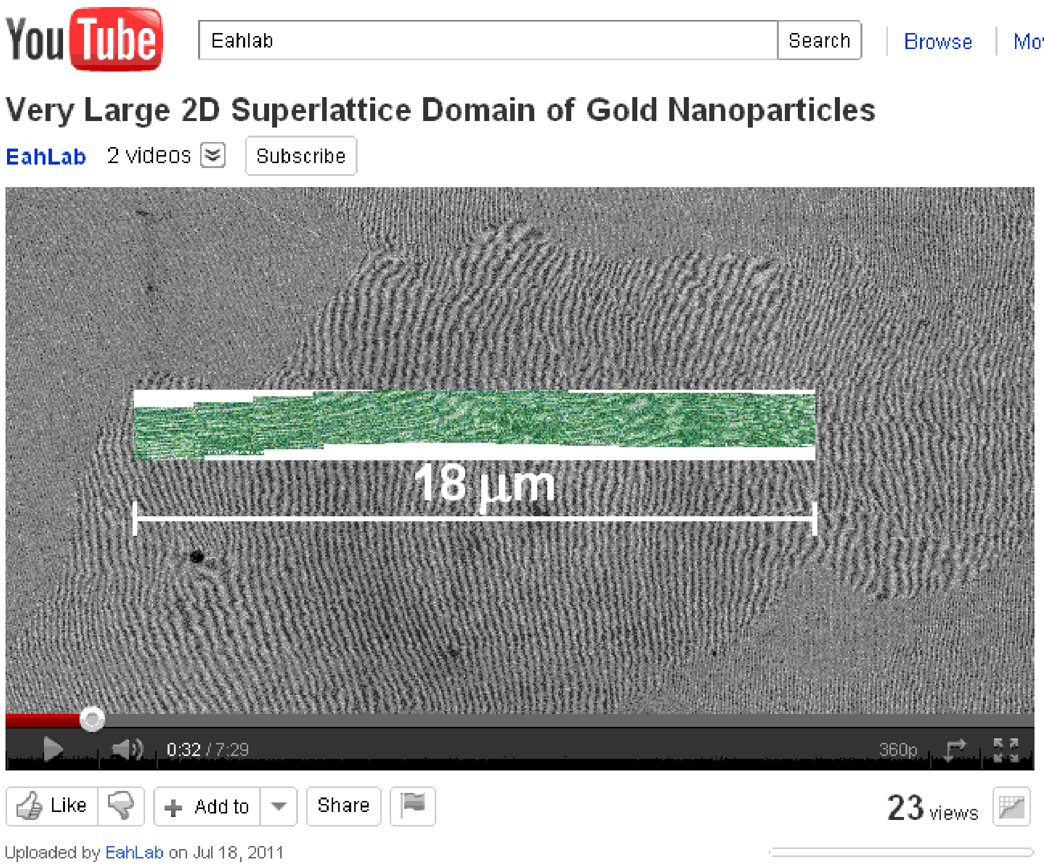

Fig. 4S 7-min video for zooming into and panning the huge image in Fig. 3S, available at YouTube (search keyword: EahLab, "http://www.youtube.com/watch?v=vLudYhwXyAM").

References

[1] M. N. Martin. J. I. Basham, P. Chando and S.-K. Eah, "Charged gold nanoparticles in non-polar solvents: 10-min synthesis and 2D self-assembly", *Langmuir*, 2010, **26**, 7410.
[2] G. Meng and V. N. Manoharan, "Phase transitions of colloidal continents" (private communication).