

Supporting Information:

Experimental section:

10 wt% aqueous suspension of silica spheres of sizes of 296, 519, and 1050 nm and polystyrene (PS) spheres of 164, 210, 270, 350, 496, and 925 nm were purchased from Microparticles GmbH, Germany. The water in all experiments was prepared using a three stage Millipore Milli-Q Plus 185 purification system and had a resistivity higher than 18.2 M Ω cm.

The suspensions were firstly diluted with water and then centrifuged, followed by decanting the supernatants, and redispersed in water. The repetition of this process for 3 times allowed removal of free ions in the colloidal suspensions. Note that all containers of colloidal suspensions were plastic to avoid the introduction of ions. The centrifugation also enabled us to concentrate the suspensions. The sphere concentrations were adjusted in the range of 5 wt% - 52 wt%. When the sphere concentrations were higher than 50 wt%, the suspensions exhibited brilliant reflective color, suggesting that the crystallization of spheres occurred inside.

Copper rings of 1 cm in diameter were cleaned consecutively by chloroform, ethanol, and water. They were dipped in to the colloidal suspension and withdrawn by hand, and thin suspension films were enclosed by the copper rings, suspended at the air/water/air interface. Afterwards, the suspension films were dried under air and at the ambient temperature and humidity. The drying period was ranged from tens of minutes to several hours, depending on the sphere concentrations; the higher the sphere concentration, the quicker the drying process. After complete drying, solid colloidal crystal films were obtained, which showed strong iridescence. Note that we were not able to reproducibly form freestanding colloidal crystals using spheres with

sizes of larger than 1000 nm, especially silica ones, at the air/water/air interface; no freestanding film was formed by drying at the elevated temperature.

In order to form curved freestanding colloidal crystal films, the colloidal suspensions of 52 wt% were dropped on Teflon blocks. We used carefully pipettes to blow air into the suspension drops to form hemispherical bubbles. The subsequent drying at the ambient condition for about 30 min left behind iridescent hemispherical freestanding crystal films.

SEM images were recorded with a Gemini LEO 1550 instrument operated at 3 kV. The samples were not sputtered by thin Au films prior to SEM measurements in order to rule out the adhesion effect of the Au coating on the mechanical properties of the freestanding films. SEM imaging of the cross-sections of broken films was used to determine the film thickness.

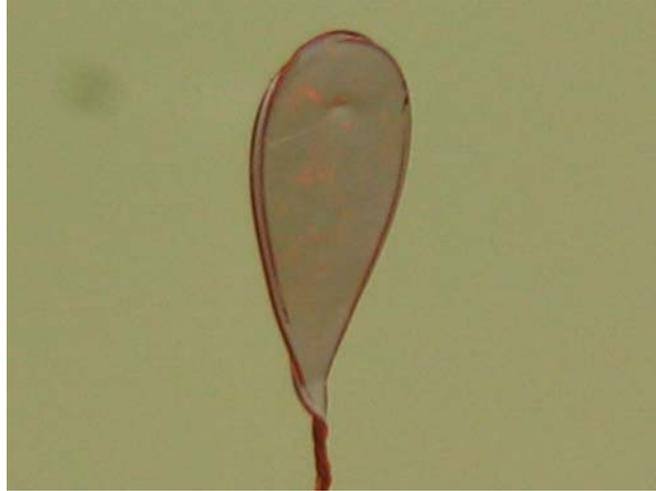


Figure S1. Photograph of a free-standing colloidal crystal film obtained by dipping a copper ring into a 10% suspension of 470 nm PS spheres, followed by drying at ambient temperature.

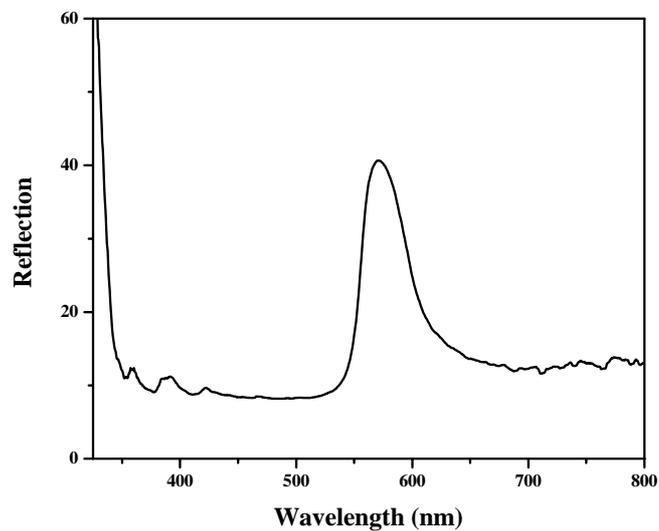


Figure S2. Reflection spectrum of the free-standing colloidal crystal film of 270 nm PS spheres derived from self-assembly at the air/water/air interface.

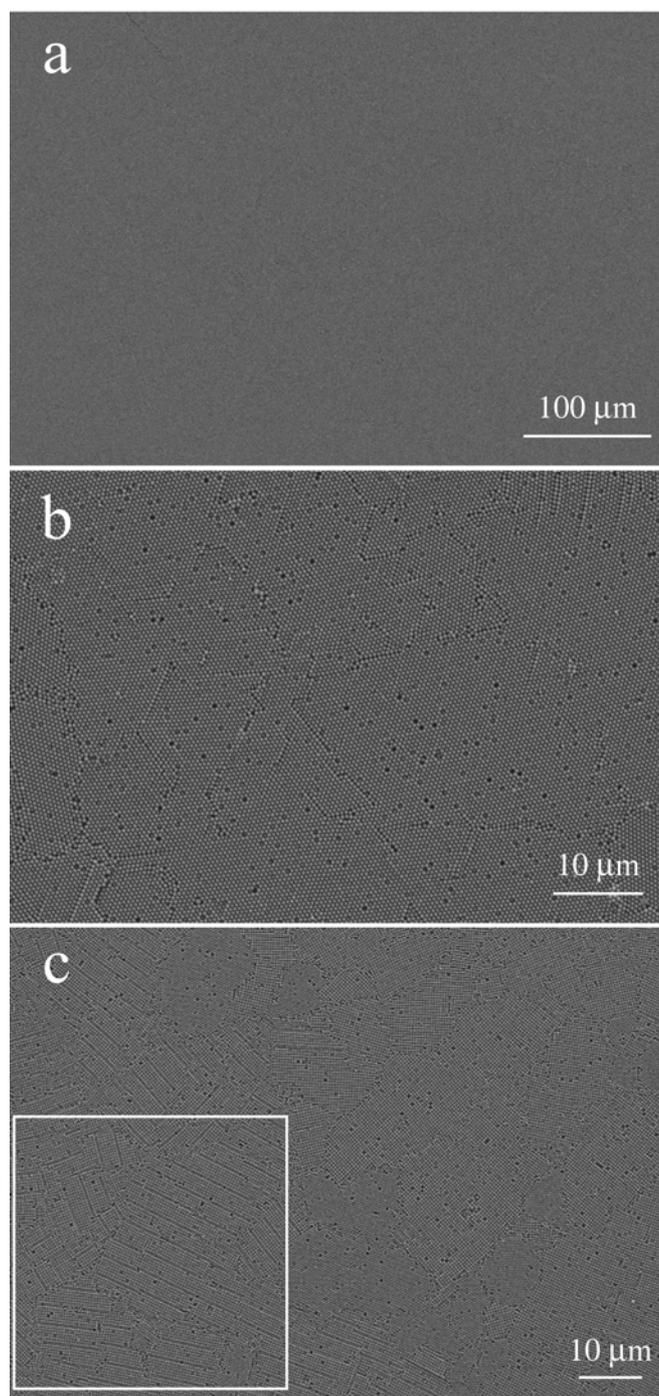


Figure S3. Low (a) and high (b) magnification SEM images of the curved free-standing colloidal crystal film obtained by blowing air into a droplet of the concentrated suspension of 470 nm PS spheres laid on a plastic substrate. (c) SEM image of cubic packed domains formed in the film. The white square highlights the new type of cubic packing structure.