doi:10.1038/nature09713

Supplementary Figure 1

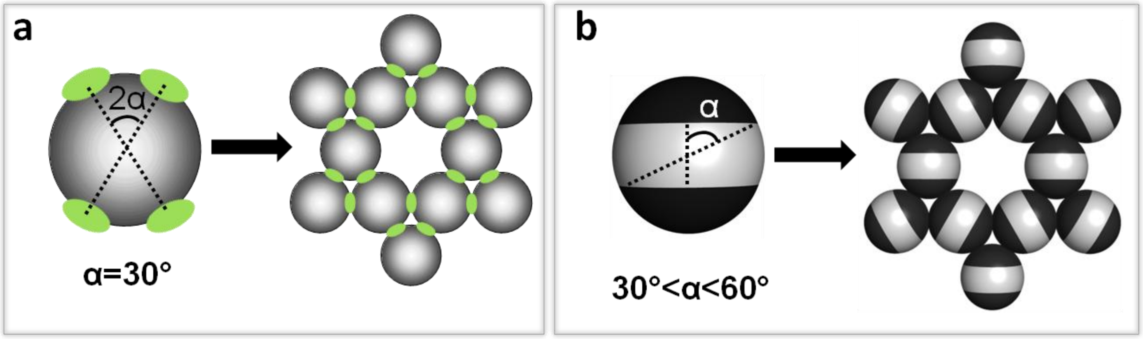

Figure legend:

Kagome lattice assembled alternatively from direct or convergent assembly of colloidal spheres. **(a)** Direct assembly requires four point-like patches (green) distributed precisely on the equator of the sphere at needed angles, $\alpha = 30°$, serving as directional attractive points to glue particles into the Kagome lattice pattern. **(b)** Convergent assembly starts with spheres decorated at opposing poles with attractive patches (black) that subtend an angle that allows contact with up to two but not three neighboring spheres, if $30° < \alpha < 60°$. Note that this angular range of patch is based on considering the size of the geometrical patch alone, while in real experiments additional factors would also come into play. Thermal motion allows the spheres to rotate with respect to one another at fixed separation over a wide range of subtended angle and this convergent assembly scheme results in Kagome lattice.

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Supplementary Figure 2

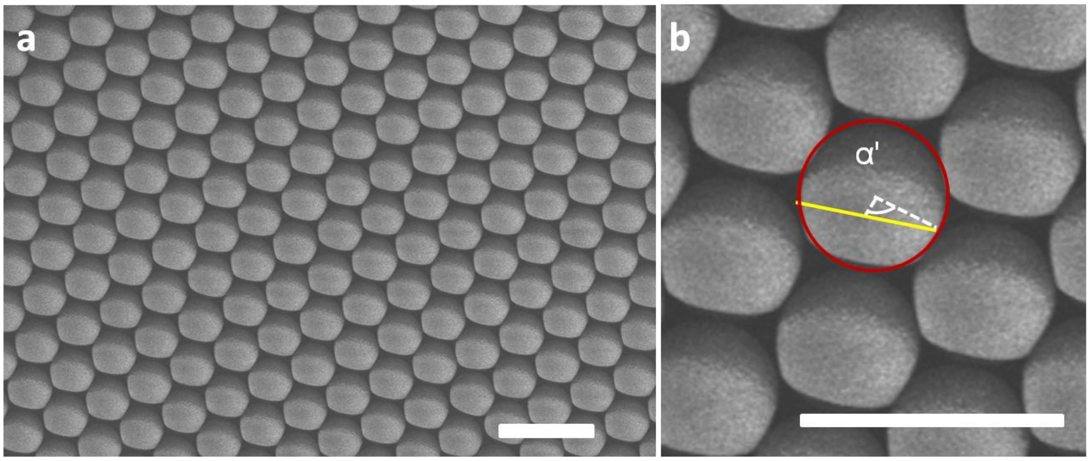

Figure legend:

Scanning electron microscope (SEM) images of triblock Janus latex spheres produced by glancing-angle deposition of gold coatings. **(a)** A top view normal to the monolayer of spheres on a silica wafer after gold deposition. The gold patches (brighter regions of the SEM micrographs) are slimmer on one axis, wider on the other. **(b)** An enlarged view of the resulting elongated-shaped gold patches. The two-dimensional opening angle along the long axis (yellow line), is $\alpha' = (65 \pm 4)$ º; here, the long axis is what matters as the system selects particle-particle orientations that maximize hydrophobic contacts. This measured size of the hydrophobic patch is slightly larger than what is solely dictated by the geometry needed for hydrophobic contact; the entropy introduced by having a slightly larger patch, hence rotational degeneracy, appears to stabilize the lattice. The electrostatic repulsion from the adjacent repulsive middle bands shrinks further the allowed range for contact[23]. We are aware that the opening angle depends not solely on the glancing angle but also on the direction of deposition relative to the axes of the two-dimensional crystal, not everywhere the same owing to grain boundaries in the monolayer crystal[16]. However, this effect does not change the opening angle if measured also along the longer axis.

Scale bar is 2 µm.

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Supplementary Figure 3

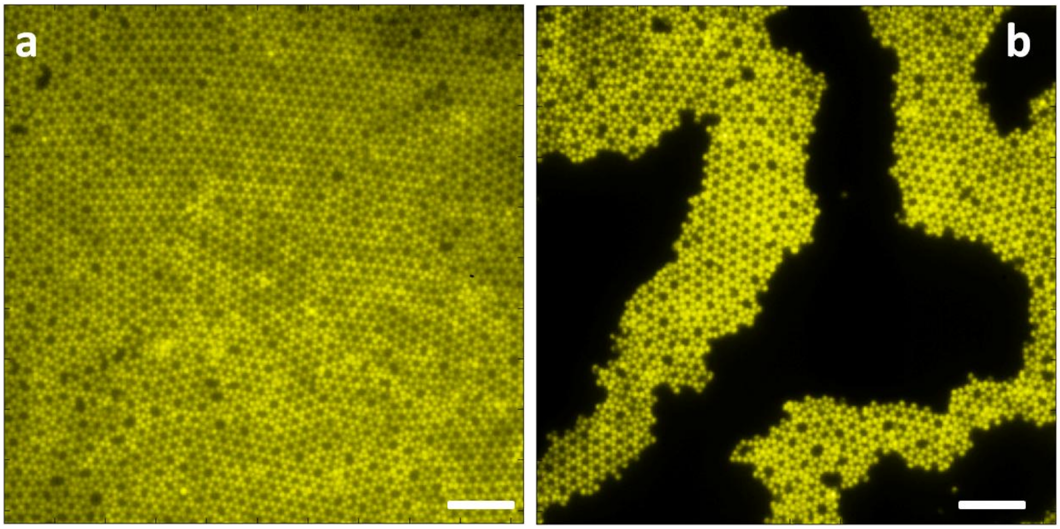

Figure legend:

Long-range order of the Kagome lattice self-assembled from triblock Janus spheres. **(a)** When the particle concentration is high, crystalline grains of different orientations impinge to form a polycrystalline structure. **(b)** When the particle concentration is sub-monolayer, the Kagome lattice likewise forms. Note the clear boundary between crystalline regions and solution (black background). Scale bar is 10 μm.

Supplementary Movie Legends:

Supplementary Movie 1: This movie file shows the breathing and vibration of a complete Kagome lattice assembled from triblock Janus spheres in aqueous 3.5 mM NaCl. The movie is acquired at 5 fr/s and is displayed at 10 fr/s. The view size is 24.6 ×24.6 μm.

Supplementary Movie 2: This movie file shows the early stage of the assembly, up to an elapsed time of 140 sec, after NaCl (final concentration is 3.5 mM) is added to a sedimented suspension in deionized water. The system evolves from discrete particles, to strings of particles, then evolves triangular nodes from distortion of strings, and finally the triangular nodes branch and weave into webs when ends of branches meet. In this movie file, dynamic events of interest are highlighted by phrases, circles and arrows (white). The movie is acquired at 10 fr/s and is displayed at 20 fr/s. The view size is 31.6 ×31.6 μm.

Supplementary Movie 3: This movie shows the later stage of the assembly, in the period 2.9 to 3.1 hr, after NaCl (final concentration is 3.5 mM) is added to a sedimented suspension in deionized water. As time passes, the number of particles per unit area is constant but increasing order develops. Here particles in a crystalline environment are labeled in blue circles and the proliferation of crystalline regions is highlighted by blue lines connecting particles and their nearest neighbors, showing the progressive development of Kagome lattice order. Red lines denote suboptimal bonding from Delaunay triangulation . The movie is acquired at 3.3 fr/s and only one of every five

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acquired frames is included in the movie. The movie is displayed at 6.7 fr/s. The view size is 10.4×10.9 μm.