Reconfigurable self-assembly through chiral control of interfacial tension,
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Recommended with a Commentary by Efi Efrati, James Franck Institute, The University of Chicago.

Chiral self-assembly, the spontaneous formation of structures from constituents which have some notion of twist, poses great theoretical and experimental challenges. On the one hand uniform and isotropic helical order (obtained by homogenous and isotropic relative twisting of adjacent particles) is incompatible with Euclidean three-dimensional geometry and therefore gives rise to geometric frustration[1-3]. On the other hand, literally, the broken symmetry under reflections is expected to manifest in the assembled structure, yet the specific handedness of it is not determined solely by the handedness of its constituents[4,5]. In the paper recommended here the authors succeed in harnessing the geometric frustration to their advantage by controlling colloidal membrane interfacial tension through chiral interactions. This results and in a wide variety of reconfigurable chiral structures, and paves a path for a better understanding and precise control of chiral self-assembly.

Virus replication processes, perfected over billions of years for high yield and high fidelity, allow the mass production of identical rod like colloidal particles of desired geometry and mechanical properties. The authors use fd-virus, a thin and chiral rod-like bacteriophage, and Dextran, a non-adsorbing polymer, to induce depletion interaction between the rod-like viruses that together form one-rod-thick colloidal rafts. The chiral interaction between the rods is temperature dependent and can be tuned all the way down to zero[6]. The control over the attractive interactions and intrinsic chirality allows the generation and stabilization of a wide variety of reconfigurable structures ranging from simple disks through ring-like twisted ribbons to doubly twisted ribbons and starfish membranes.

Unlike lipid bilayers, the liquid colloid monolayers constructed in this work do not form vesicles and have a stable exposed edge[7]. The authors have demonstrated that the rods' orientation in this exposed edge is in the plane of the membrane (perpendicular to their direction in the bulk) allowing the rod-polymer interface to curve in both directions, thus reducing its area. In the absence of chiral interactions between the rods the orientation rotation of the rods between the bulk and boundary is driven by the reduction of interfacial area at the expense of some elastic energy due to the relative twist of the rods. In this case both right and left handed rod orientation rotations are observed. When chiral interactions are present only one preferred orientation rotation is observed. In this case it is energetically favorable for neighboring rods to adopt a specific relative twist, and the parallel packing of rods, as appears in the bulk of the monolayer, becomes frustrated. This renders the formation of boundaries favorable, and leads to the chiral control of

interfacial tension. For sufficiently strong chiral interactions circular shaped monolayers become unstable and are replaced by twisted ribbons.

For low chirality-interaction-strength circular disks are formed. The rods' orientation twists in the preferred handedness along radial lines. This twist penetrates only a finite depth away from the boundary[8]. As chiral interactions become more dominant the disks are gradually replaced by helical ribbons whose width is given by the twist penetration depth. This twist, directed perpendicularly to the boundary along the traverse direction of the helical ribbons, is expected to possess the same handedness as the radial twist in the disk. As in general helical ribbons display opposite twists along their length and width this implies that the authors have also demonstrated a path for the generation of say left handed helicoidal ribbons from right handedly stacking rods. While not stated explicitly in the text, this achievement makes another crucial contribution to our ability to understand and control chiral self-assembly.
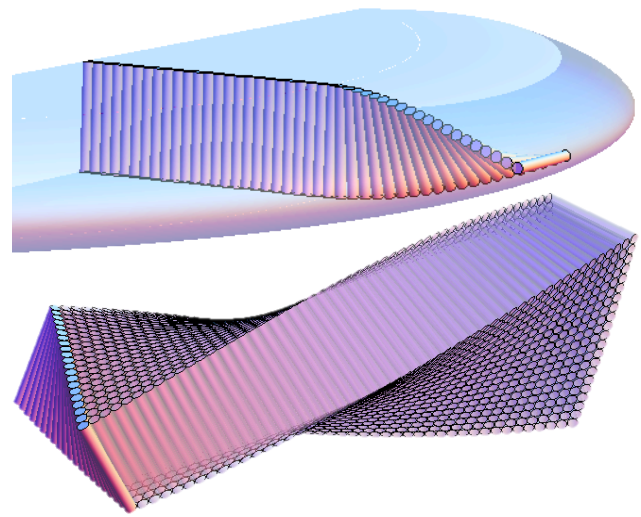
(Top) Illustration of a right handed radial twist perpendicular to the disk boundary allowing the interface to curve in both direction.
(Bottom) Illustration of a right handed twist perpendicular to the strip boundary, oriented along the width of the strip (solid cylinders along the left end). This right handed traverse arrangement gives rise to a left handed longitudinal structure by external surface minimization.

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