Supporting Information for: Macroscopic chiral symmetry breaking in monolayers of achiral nonconvex platelets

Nikoletta Pakalidou,[1] David L. Cheung,[2] Andrew J. Masters,[1] and Carlos Avendano[1,*]

[1] *School of Chemical Engineering and Analytical Science, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK*

[2] *School of Chemistry, National University of Ireland, Galway, University Road, Galway, Ireland*

S1. EQUATIONS OF STATE OF NONCONVEX POLYGONS

In addition to the phase behaviour of nonconvex hexagonal and square platelets presented in the main text, we analyse the effect of the in-plane rotational symmetry of nonconvex polygons with three- and five-fold symmetries and curvature $k = 1/3$. The phase diagrams for these systems are shown in Figures S1 and S2.

We discuss first the phase behaviour of nonconvex pentagons. The results, which are shown in Figure S1, are similar to the results previously reported for perfect pentagons with straight edges [1]. Nonconvex pentagons exhibit two phase transitions. The first transition is from an isotropic (Iso) state to a hexagonal rotator (RHx) phase in which the particles arrange in a hexagonal lattice with random orientations. The second transformation is from a RHx phase to a distorted hexagonal (Hx) phase. In this Hx phase the five-fold orientational order parameter Φ_5 is negligible for all packing fractions explored (results not shown), but Φ_{10} approaches unity which shows that the Hx phase is formed by alternating rows of oppositely pointing particles (see Figure 2(b) in the main text).

The results for nonconvex triangles are shown in Figure S2. Contrary to what it is observed in the simulations of perfect triangles studied by Gantapara and co-workers [2], where particles form a triatic phase, nonconvex triangular platelets with curvature $k = 1/3$ exhibit a transition from a low-density isotropic phase to a striped (achiral) triangular (Tr) phase characterised by alternating rows of oppositely oriented particles (see Figure 2(d) in the main text).

Finally, based on the same symmetry arguments used for squares, pentagons, and hexagons, one would expect that non-convex platelets with 7-, 8-, 9-, 10-, and 11-fold symmetries cannot form chiral crystals as these structures form hexagonal crystals. This is indeed confirmed by analysing the close-packed configurations of heptagons and octagons shown in Figure S3(a,b). The case of dodecagons, however, is different. Analysis of the close-packed configurations of dodecagons with $k = 1/3$, as shown in Figure S3(c), reveals the formation of a chiral structure. In this case the symmetry of the platelets is a multiple of the number of nearest neighbours in the crystal, thus half of the edges can be paired promoting a similar angular displacement as it was observed in the case of hexagons and squares.

S2. GEOMETRICAL MECHANISM OF MACROSCOPIC CHIRAL SYMMETRY BREAKING

As shown in references [3–5], certain buckling instabilities in 2D low-density cellular materials can induce macroscopic chiral symmetry breaking (CSB). These structures resemble the structures observed in nonconvex hexagons and squares. When these instabilities are comprised only of half sinusoids, neither square nor hexagonal cellular materials exhibit macroscopic CSB (see Figures S4(a,c)). However, when the instabilities are comprised of complete sine waves, the system exhibits macroscopic CSB through the induction of a rotation of the walls of the cellular materials in the same direction (see S4(b,d)). In our systems of curved nonconvex platelets we do not have such buckling instabilities. However, the pairing of the curved edges of a given platelet with the edges of neighbouring particles induces a similar rotation mechanism as in cellular materials that leads to the appearance of macroscopic CSB. This induced rotation in nonconvex platelets is sketched in Figure S5.

In the case of triangular cellular materials, buckling instabilities can induce chiral rotation when the triangles are arranged in an edge-to-edge configuration as shown in Figure S6(a). However, the most efficient packing of nonconvex triangles is when they arrange in a vertex-to-edge configuration for which geometrical frustration prevents the rotation of the particles in the same direction as shown in Figure S6(b).

* carlos.avendano@manchester.ac.uk

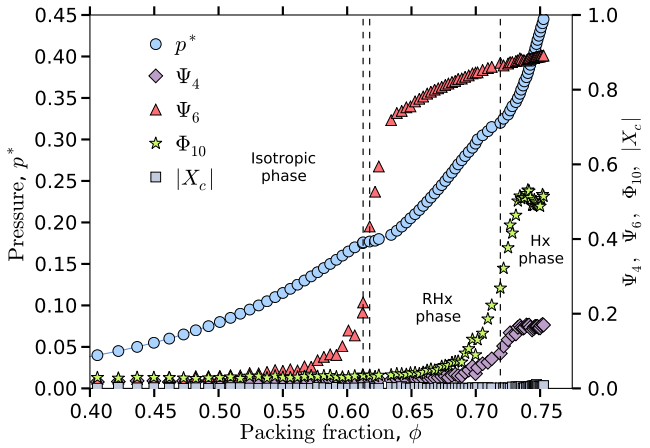

FIG. S1. Pressure-packing fraction ($p^* - \phi$) phase diagram for nonconvex pentagon platelets with curvature $k = 1/3$. The dependence of the global bond-order parameters Ψ_4 and Ψ_6, the orientational order parameter Φ_{10}, and the chiral order parameter X_c on the packing fraction ϕ is indicated.

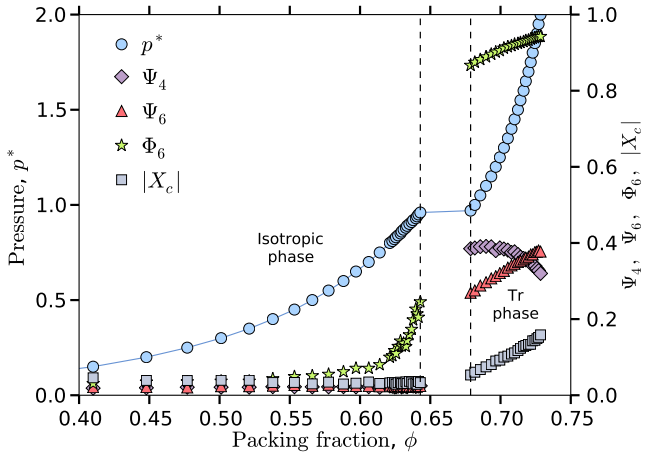

FIG. S2. Pressure-packing fraction ($p^* - \phi$) phase diagram for nonconvex triangular platelets with curvature $k = 1/3$. The dependence of the global bond-order parameters Ψ_4 and Ψ_6, the orientational order parameter Φ_6, and the chiral order parameter X_c on the packing fraction ϕ is indicated.

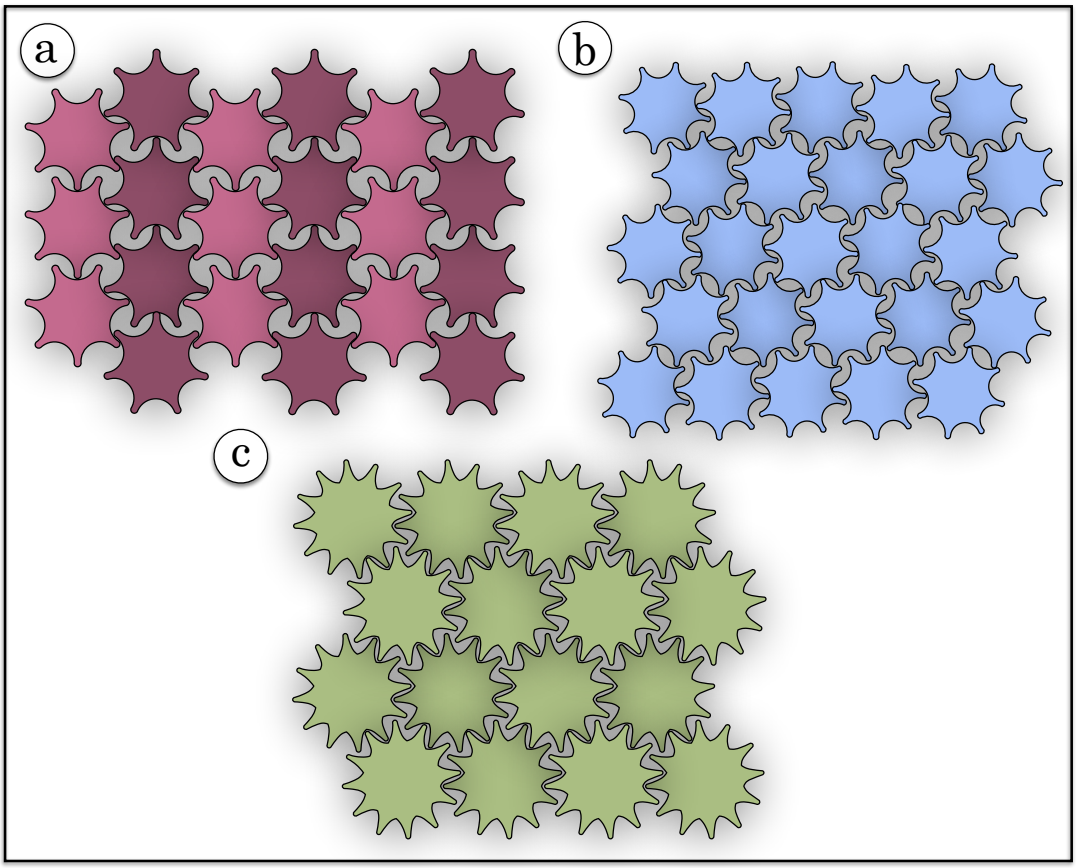

FIG. S3. Representative configurations of close-packed microstructures observed in monolayers formed by nonconvex platelets with curvature $k = 1/3$ obtained using the pressure-annealing method of Filion et al. [6] The configurations correspond to crystals of (a) heptagons, (b) octagons, and (c) dodecagons. Only dodecagonal platelets form a chiral structure.

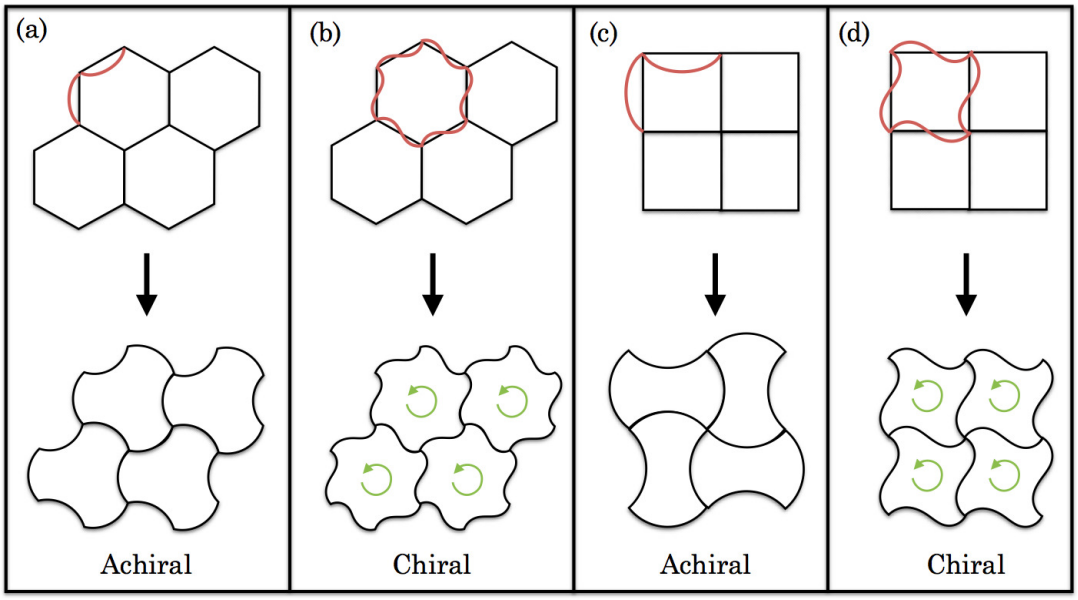

FIG. S4. Buckling instabilities in two-dimensional cellular materials with hexagonal (a,b) and square (c,d) symmetries. Figures (a) and (c) correspond to instabilities comprising of half sinusoids showing the formation of achiral configurations. Figures (b) and (d) correspond to instabilities comprised of complete sinusoids that allow the structure to rotate in a common direction to form chiral configurations.

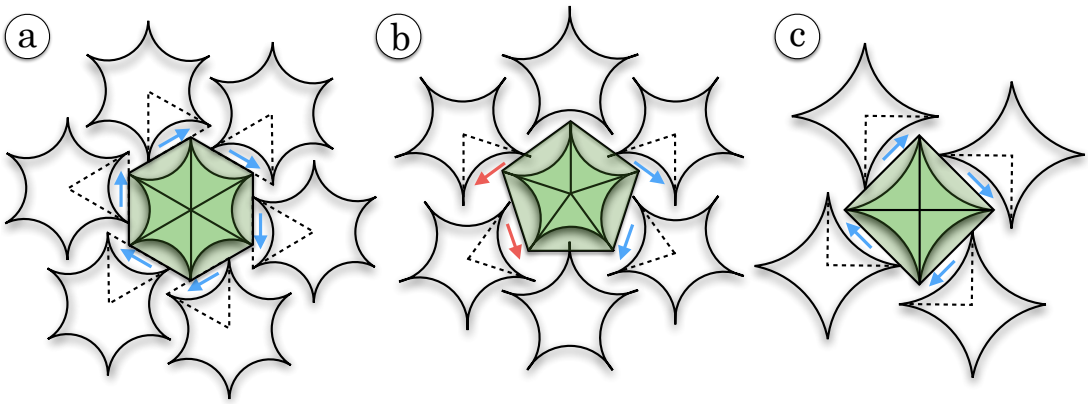

FIG. S5. Sketch of geometrical configurations in a close-packed configuration of nonconvex polygonal platelets. The configurations correspond to (a) hexagons, (b) pentagons, and (c) square platelets. Only hexagonal and square platelets form configurations in which all the edges of a central particle can be paired with edges of neighbouring particles. This complete pairing of the edges allows a rotation of the system in a common direction allowing the formation of chiral phases. Conversely, pentagons exhibit geometrical frustration that forbids the rotation of the particles in a common direction.

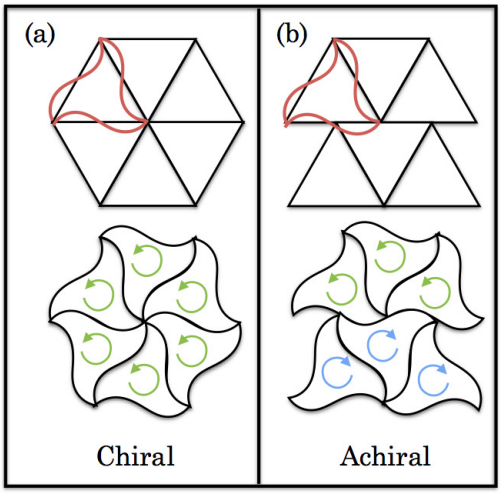

FIG. S6. Buckling instabilities in two-dimensional cellular materials with triangular geometry. Figure (a) corresponds to a edge-to-edge triangular structure, while Figure (b) corresponds to a vertex-to-edge triangular structure. Instabilities comprising of complete sine functions are shown for both systems, showing that only system (a) can form a chiral structure. The instabilities in structure (b) are geometrically frustrated.

[1] T. Schilling, S. Pronk, B. Mulder, and D. Frenkel, Phys. Rev. E **71**, 036138 (2005).
[2] A. P. Gantapara, W. Qi, and M. Dijkstra, Soft Matter **11**, 8684 (2015).
[3] S. H. Kang, S. Shan, W. L. Noorduin, M. Khan, J. Aizenberg, and K. Bertoldi, Adv. Mater. **25**, 3380 (2013).
[4] S. H. Kang, S. Shan, A. Košmrlj, W. L. Noorduin, S. Shian, J. C. Weaver, D. R. Clarke, and K. Bertoldi, Phys. Rev. Lett. **112**, 098701 (2014).
[5] B. Haghpanah, J. Papadopoulos, D. Mousanezhad, H. Nayeb-Hashemi, and A. Vaziri, Proc. R. Soc. A-Math. Phys. Eng. Sci. **470**, 20130856 (2014).
[6] L. Filion, M. Marechal, B. van Oorschot, D. Pelt, F. Smallenburg, and M. Dijkstra, Phys. Rev. Lett. **103**, 188302 (2009).