

## Electronic Supplementary Information

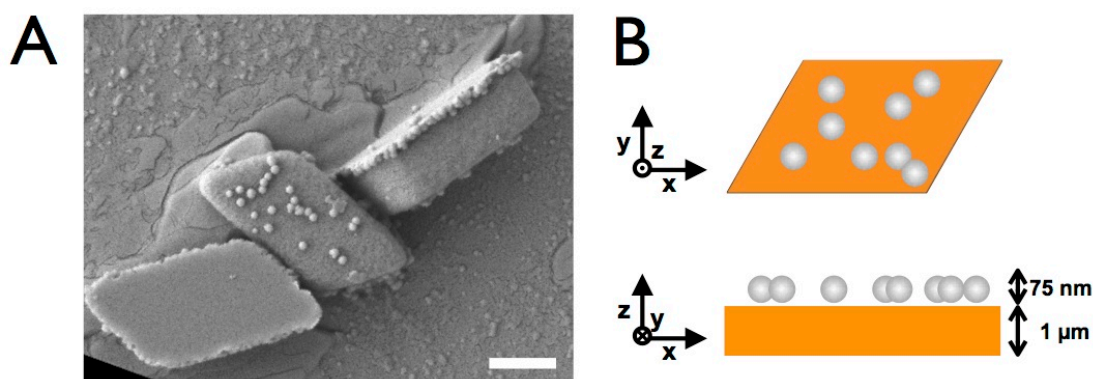
### Controlling Enantiomeric Populations in Fluctuating Brownian Monolayers of Chiral Colloids

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#### Selectively controlling facial roughness of lithographic platelets

Controlling particle roughness selectively only on either the upper or the lower surfaces of lithographic platelets, potentially useful for creating anisotropic facial interactions by depletion effects, can be achieved in several ways. For instance, monodisperse silica spheres (diameter  $\approx 75$  nm, Nissan Chemical Industries Ltd., 30.5%<sub>w</sub> in isopropyl alcohol (IPA)) can be selectively bonded to the top faces of the parallelograms.<sup>1</sup> After lithography, just before lift-off, we spin-coat (Cookson Electronics Equipment, model G3P-8) the developed wafers that have arrays of parallelograms (see Figure 1, main text) with 2.5 mL of 0.5%<sub>w</sub> silica dispersed in IPA at 1000 rpm for 4 s with an acceleration of 300 rpm/s, then 3000 rpm for 40 s with acceleration 500 rpm/s, decelerating to 0 rpm using 500 rpm/s. After spin-coating, we place the wafers on a hot plate at 95 °C for 90 s to promote residual IPA evaporation and adhesion of the silica nanospheres to the SU-8 particles' faces. We lift-off the particles using the same procedure as for the bare SU-8 particles. Figure S1 shows a typical SEM image of L-parallelograms having larger silica roughness only on their top faces. Alternatively, a controlled roughness can be selectively imparted to the bottom surfaces of the platelets by incorporating nanospheres into the release layer or depositing nanospheres on the release layer prior to SU-8 coating; some of these nanospheres make contact with the SU-8 layer and sparsely roughen the bottom faces after lithographic optical crosslinking. Imparting a higher roughness only to the bottom surfaces of platelets can also be used to suppress depletion attractions between the bottom faces and the wall compared to the smoother top faces and the wall. Thus, facial surface-roughness modifications can also be used to control the chirality of platelets in a monolayer above a wall in the presence of a depletion agent.



**Figure S1.** Silica spheres ( $\approx 75$  nm diameter) are selectively deposited onto the top surfaces of L-parallelograms to increase surface roughness on those faces. Panel **A**: SEM image of silica-caused roughness only on the top faces of SU-8 parallelogram platelets. Scale bar is 1  $\mu$ m. Panel **B**: schematic illustrations (top and side views) of a microscale parallelogram platelet that has only its top face roughened by nanospheres.

#### References

1. K. Zhao, T. G. Mason, *Phys. Rev. Lett.* 2007, **99**, 268301.