Electronic Supplementary Material (ESI) for Soft Matter.
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Electronic Supporting Information

Patterning of colloids into spirals via confined drying

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1. Synthesis of hematite ellipsoids:

To synthesize hematite ellipsoids, 9.24 g of iron (III) perchlorate, $Fe(ClO_4)_3$ (Alfa aesar, India), 1.2 g of urea, $CO(NH_2)_2$ (Merck, India) and 0.125 g sodium di-hydrogen phosphate, NaH_2PO_4 (Merck, India) are added to 200 ml Milli-Q water taken in a piranha (70% H_2SO_4 and 30% H_2O_2 by volume) cleaned Pyrex bottle. Thereafter, it is kept in an oven preheated to 100°C for 24 h. After the completion of the reaction, the product is cooled overnight. The particles in the reaction product are separated by centrifugation at a fixed 6000 rpm for ~ 30 min and further washed multiple times with de-ionized water until a clear supernatant is obtained. The synthesized particles are highly mono-disperse as confirmed from the high resolution scanning electron microscopy (HR-SEM) image and are found to have an aspect ratio $\alpha = 4.72 \pm 0.42$ as shown in Fig. 1(a) in the manuscript. Finally, pH of the hematite ellipsoid dispersion is adjusted to ~ 2 by adding aqueous nitric acid (HNO_3). Under these pH condition, the particles are highly charged with a zeta potential of +40 mV (measured in 0.0001 M $NaCl$) i.e., the dispersion are charge stabilized and do not show any gravity settling.

2. Spiral deposit patterns on the top and bottom substrates:

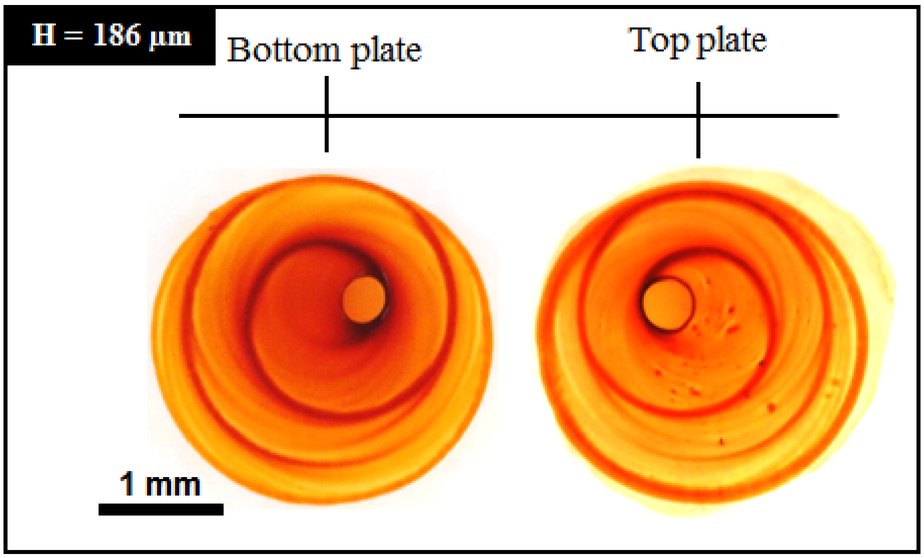

Figure S1: Final particulate deposit left on both top and bottom substrate by drying of dispersion containing hematite ellipsoids at $\phi \sim 3wt\%$ concentration with fixed confinement spacing of H $\sim$ 186 μm.

3. Spatial distribution of particles in spiral patterns:

The patterns obtained by drying of dispersion at $\phi \sim 3$ $wt\%$, the deposit height near the near the core of the spiral at $\psi = 0$ is highest and away from the core along the spiral turn with increasing ψ from 0 to π, the height of the deposit decreases, where ψ is an angle the arc of spiral spans. As shown in Fig. S2 the accumulation of particle decreases from the core to the edge of the dried deposits. It is also evident from the color intensity in the microscopy images that in the spacing between the spiral turns, the deposit consists of relatively few particles.

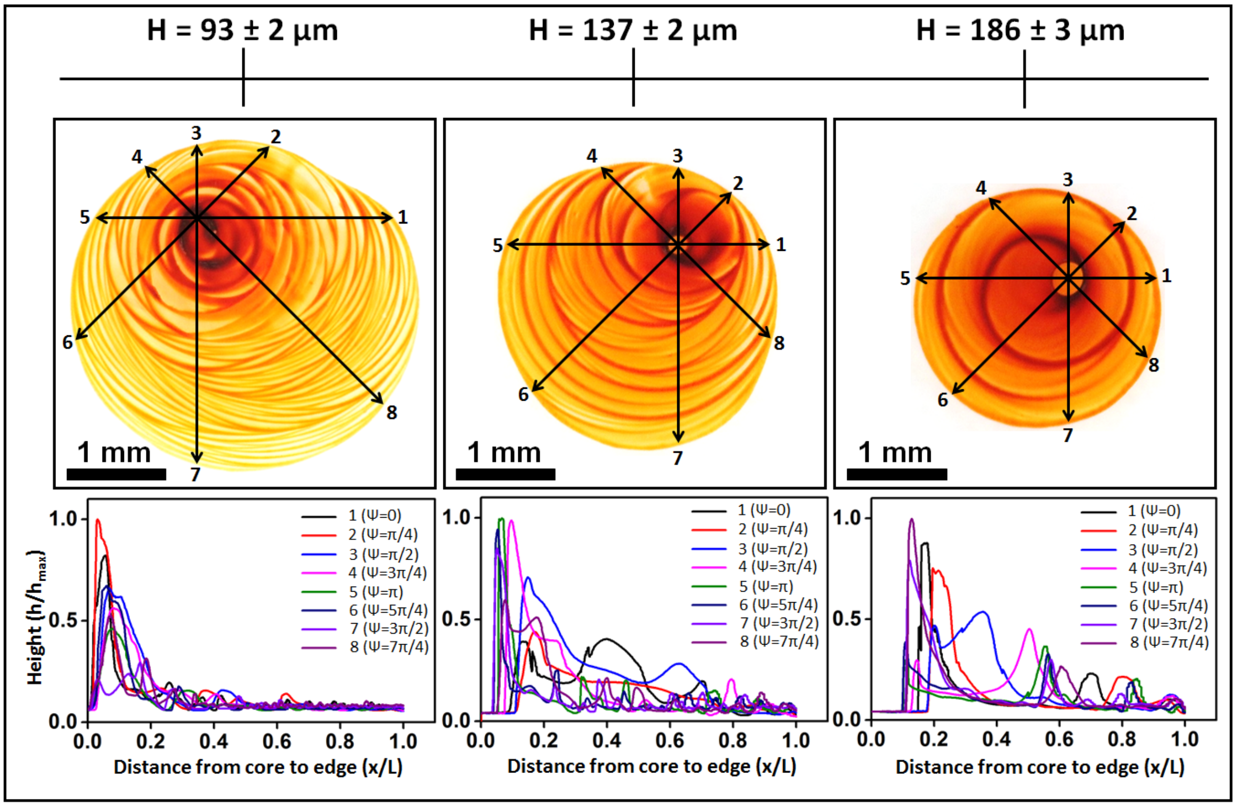

Figure S2: The spatial distribution of particles in the spiral deposit. The height profile measurement confirms that the accumulation of particles near the core of the spiral is significantly large and away from core to the edge of deposit, the height decreases.

4. Influence of substrate wettability:

To investigate the influence of substrate wettability on the formation spiral patterns, identical evaporation experiments are performed by considering drying of dispersions confined between two hydrophobic substrates. The commercially available paraffin film stuck on a glass is used as substrate. The equilibrium contact angle of water drop on these substrates is measured to be $\theta_e = 104 \pm 3°$. The dispersion confined between such substrates take the shape of a convex nodoid as shown schematically in Fig. S3 (a). The final deposit patterns obtained on these substrates are also found to be spiral as shown in Fig. S3 (b). During the early

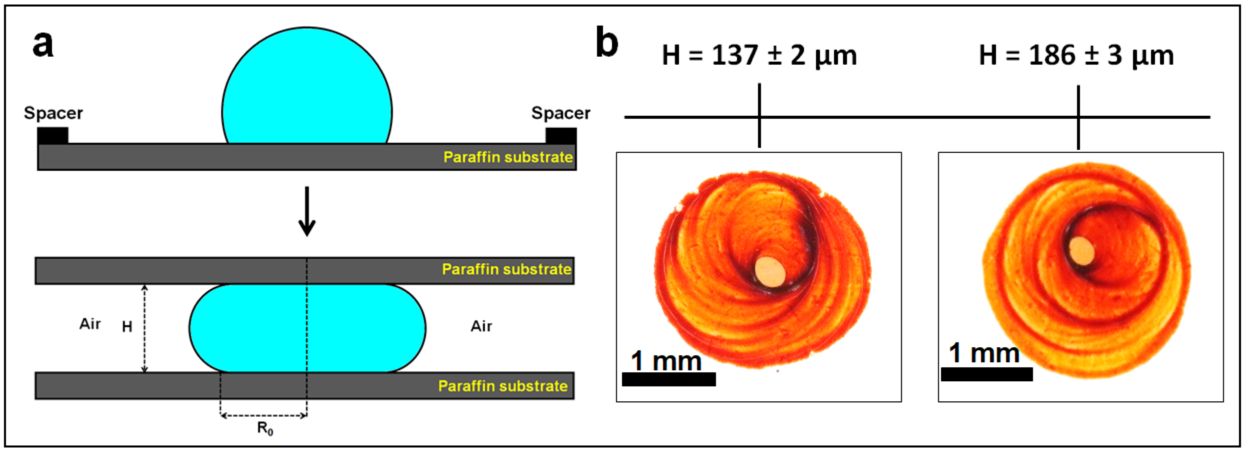

Figure S3: (a) Schematic representation of the drying of dispersion confined between two parallel hydrophobic substrates. (b) Optical microscopy images of final dried deposits obtained upon complete evaporation of the solvent show spiral deposits patterns irrespective of the different confinement spacing at fixed particle concentration of ~ 3 $wt\%$.

period of drying, the dispersion is pinned at the contact line on the both top and bottom paraffin substrate. However, due to the inward receding motion of the liquid-vapour interface as a result of solvent evaporation, the shape of the dispersion eventually becomes catenoid (or concave nodoid). Thus as a result of similar drying kinetics that occurs at later part of drying period finally results in similar spiral deposit patters even when the dispersions confined between hydrophobic substrates are dried.