Supporting Information

Field-Directed Assembly of Patchy Anisotropic Microparticles with Defined Shape

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1. Fabrication process of anisotropic patchy particles

Spin coating was performed following the procedures provided by MicroChem, Inc (http://microchem.com/pdf/SU8_2-25.pdf). A detailed description of the process for fabricating anisotropic patchy particles is provided below.



Fig. S1The fabrication process for generating anisotropic patchy particles. Five steps are used for the fabrication of anisotropic, patchy microparticles: (A-B) spin coat a Si wafer with photoresist (C) expose the resist to a patterned photomask containing desired design (D) develop uncross-linked resist (E) deposit metal and (F) release by applying moderate shear forces.

Si wafers were thoroughly washed with a series of acetone and methanol. Wafers were then dried with a stream of N_2 gas. Wafers were placed on a hot plate at 95°C for 5 min to fully dry. Wafers were then placed on the center of a spinner and covered with at least 3 mL of SU-8 photoresist with the following programmed run (the following methods were for 10 µm thick photoresist layers using SU-8 10; please refer to the MicroChem manual (see above) for exact methods for achieving different heights):

- Ramp (500 rpm at 100 rpm for 5 sec)
- High spin (3,000 rpm at a ramp of 300 rpm for 30 sec)

• Stop (0 rpm at a ramp of 1000 rpm for 0 sec)

The spin coated wafers were then soft baked at 65°C for 120 sec and then at 95°C for 300 sec. The wafers were then immediately loaded into the MA6 Mask Aligner (Karl Suss, Garching, Germany), vacuum locked, and exposed to an energy dosage of ~150 mJ cm⁻² UV light at a wavelength of 365 nm. Wafers were immediately post-baked on the wafer at 65°C for 60 sec then again at 95°C for 120 sec. The exposed wafers were then completely submerged in SU-8 developer (MicroChem) for 120 sec, rinsed with isopropanol, and dried with a stream of N₂ gas. Finally, as described in the manuscript, particles were deposited with evaporated metal and removed from the substrate through the application of gentle shear forces applied via a rubber policeman (Fig. S2).



Fig. S2 Scanning electron micrographs of various anisotropic microparticles with different coatings. This figure includes micrographs of (a) uncoated cylinders (height = 25 μ m, dia. = 10 μ m), (b) top-coated cylinders (height = 25 μ m, dia. = 10 μ m), (c) side-coated cylinders (height = 25 μ m, dia. = 10 μ m), (d) uncoated cuboids (height = 25 μ m, dia. = 10 μ m), (e) uncoated cubes (10 μ m), (f) an uncoated cube (10 μ m), (g) uncoated cuboids (height = 25 μ m, dia. = 10 μ m), (h) uncoated hexagons(height = 10 μ m, dia. = 10 μ m), and (i) uncoated hexagons (height = 10 μ m, dia. = 10 μ m). Scale bars on micrographs are 20 μ m unless otherwise noted.

2. Videos of DEP and MAP assembly

Note: the field direction in the videos is rotated 90° *from the field direction shown in the manuscript.*

Video 1: Particle interactions of top-coated cubes at low frequencies (2 kHz)

This video shows the effect of various field strengths on DEP assembly dynamics of topcoated cubes. Applied field strengths of 3.33, 6.67, 10.0, and 13.3 kV m⁻¹ are shown. At the lowest field strength shown (3.33 kV m^{-1}), particles exhibit a slow response to the applied field in comparison to the higher field strengths analyzed. At field strengths of 6.67 kV m⁻¹, particles assembled without flipping, suggesting kinetic trapping. At field strengths of 13.3 kV m⁻¹, particles are seen to flip to align in a uniform orientation and then assemble.

Video 2: Particle interactions of top-coated cubes at high frequencies (20 kHz)

This video shows the flipping behavior of top-coated cubes at high frequencies (20 kHz). As discussed in the manuscript, dark or opaque particles refer to an orientation where the metalcoated region is in the viewing plane and transparent particles refer to an orientation where the metal-coated region is not in the viewing plane. At high frequencies, the particles are seen to pivot then align, roll toward a uniform orientation then assemble, and also assemble with the metallic faces both in the viewing plane and normal to the viewing plane. Please refer to the manuscript for a discussion on the behaviors of top-coated cubes at various field strengths and frequencies during DEP assembly.

3. Model parameters

Table S1. Assumed properties of materials used in $COMSOL^{TM}$ modeling. The relative permittivity of SU-8 was considered to be independent of the frequency of the applied AC-field and hence a fixed dielectric constant value was of 4.2 used for the simulations.

Material	Conductance (S/m)	Relative Permittivity	Relative Magnetic Permeability (Hx10 ⁶ /m)
Metal coating	Au $(4x10^7)$	Au (1x10 ⁹)	Ni (10 or 20)
SU-8	-	4.2	1
Counter ionic later	0.2	78	-
Water	$4 \mathrm{x} 10^{-4}$	78	1
Ferrofluid	-	-	14.9

4. Configurational energies of single-side coated particles in magnetic fields

We estimated the energies of close-packed and zigzagged configurations formed in different ferrofluid concentrations via magnetophoresis. As seen in Table S2, we obtain the same energy for the zigzagged and close-packed structures in water. The difference between the observed morphology of zigzagged and close-packed structures in water may therefore not be due to differences in energy, but may be due to the entropy, as the zigzagged patterns may have additional degrees of freedom. As the ferrofluid concentration is increased, on the other hand, close-packed structures have lower energy. **Table S2.**Configurational energies (units: 10^{-7} J) of zigzagged and close-packed structuresat various ferrofluid concentrations.

	Water	2 vol.% ff	4 vol.% ff
Zigzagged	0.2463	0.347	0.451
Close-packed	0.2463	0.316	0.378

5. Author contributions

C.W.S. developed the method for fabricating anisotropic shaped patchy microparticles and worked with J.L. on their synthesis and characterization. S.Z. performed the DEP assembly experiments with the assistance of C.W.S. and B.B., following discussion and directions by O.D.V. COMSOL modeling for DEP and MAP assembly was performed by B.B. and Y.Y., respectively. Y.Y. performed the MAP assembly experiments with the assistance of C.W.S. The manuscript was written by C.W.S., B.B.Y., O.D.V., and G.P.L. All authors jointly discussed the results and commented on the manuscript.