

## Supporting Information

### Tunable Janus Colloidal Photonic Crystal Supraballs with Dual Photonic Band Gaps

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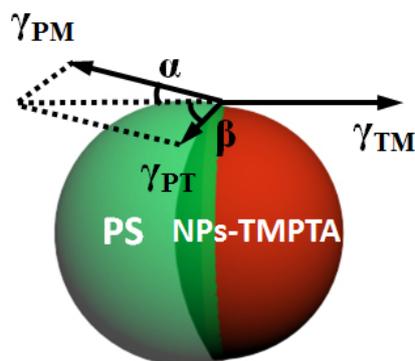
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#### Supplementary Movies

**Movie S1** Rolling-over of CPC Janus supraballs Controlled under Magnetic field.wmv

**Movie S2** Rotation of CPC Janus supraballs Controlled under Magnetic field.wmv

#### Supplementary Figures



**Figure S1** Schematic illustration of the balancing of the interfacial tensions between triple phases and the formation of CPC supraparticles.

The geometry of the biphase droplet arises from the minimization of the interfacial free energies, which follows the rules of Young-Dupre's equations [Equations (1) and (2)] and is presented in Figure S2.

$$\gamma_{TM} = \gamma_{PM} \cos \alpha + \gamma_{PT} \cos \beta \quad (1)$$

$$\gamma_{MP} \sin \alpha = \gamma_{PT} \sin \beta \quad (2)$$

where  $\gamma_{PT}$  means the interfacial tension between the PS solution and NPs-TMPTA,  $\gamma_{PM}$  means the interfacial tension between the PS solution and methylsilicone oil,  $\gamma_{TM}$  means the interfacial tension between TMPTA and methylsilicone oil, and  $\alpha$  and  $\beta$  are the contact angles depicted in Figure S2. Through adjusting  $\gamma_{TM}$ ,  $\gamma_{PM}$  and  $\gamma_{PT}$ , we could calculate  $\alpha$  and  $\beta$  and obtain stable biphasic droplets with predictable geometries.

To gain further insight into the self-assembly behavior of the biphasic droplets, the spreading coefficient (S) showing the spread behavior of the liquid phase was necessary to be investigated [Equation (3)].

$$S_a = \gamma_{bc} - (\gamma_{ab} + \gamma_{ac}) \quad (3)$$

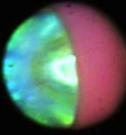
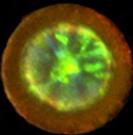
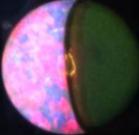
where  $\gamma_{bc}$  means the interfacial tension between phase b and c.

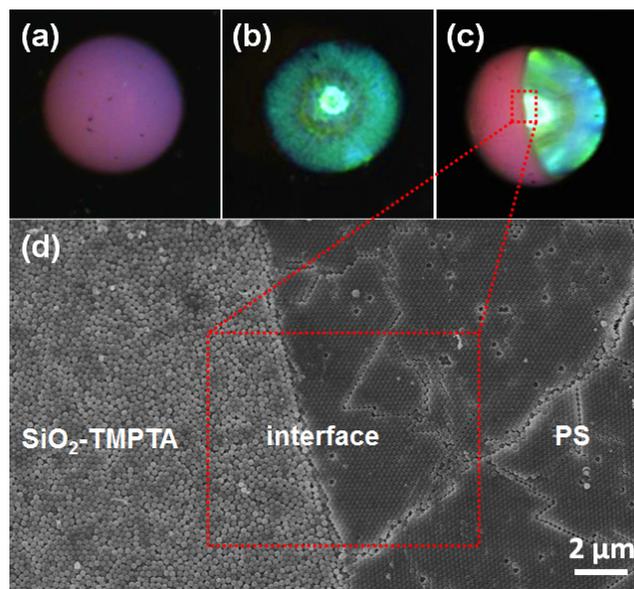
As listed in Table S1. When  $S_T < 0$ , NPs-TMPTA covered a part of PS droplet surface to achieve a CPC Janus supraparticles as Group 1 and 3 displayed. When  $S_T > 0$  NPs-TMPTA spread entirely across the whole PS spherical phase to generate a core-shell structure as Group 2 showed.

**Table S1** Three groups of interfacial tension for fabrication of CPC supraparticles

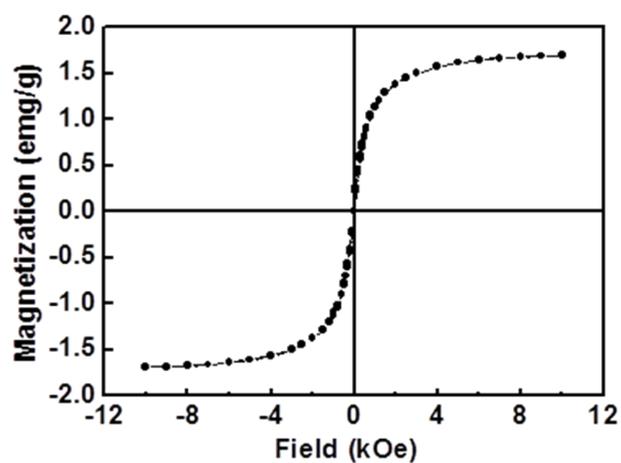
Group	Triton X-100 (wt% in PS)	$\gamma_{PT}$	$\gamma_{PM}$	$\gamma_{TM}$	$S_M$	$S_T$	$\alpha$		$\beta$		model	experimental
							(mN/m)		(°)			
1	0.4	1.0	4.2	3.7	-6.9	-0.5	12.6	113.5				
2	0	10.8	20.9	3.7	-13.8	6.4	Null	Null	Null			
3	0.5	0.9	3.6	4.1	-6.8	-1.4	11.2	50.8				

In Group 1 and 2, the two phases are PS solutions and SiO<sub>2</sub>-TMPTA. In Group 3, the two phases are PS solutions and Fe<sub>3</sub>O<sub>4</sub>-TMPTA. The models in Table S1 are theoretically simulated according to the interfacial values by using Surface Evolver software.<sup>1</sup> For Group 2, because of the null of values of  $\alpha$  and  $\beta$  according to Young-Dupre's equations, the simulation is invalid for Group 2.

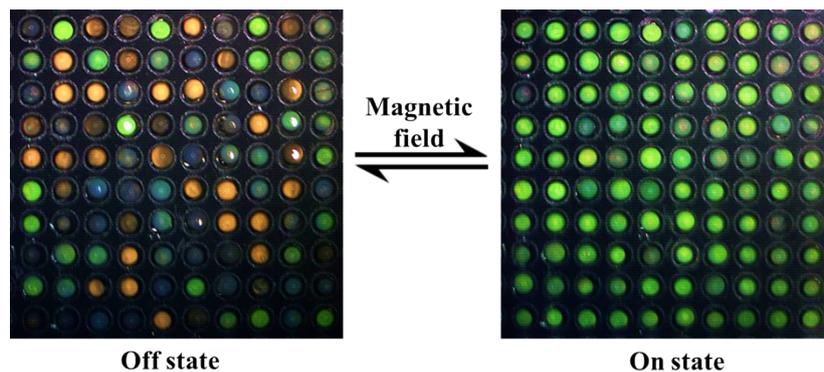
	Group 1	Group 2	Group 3
Simulation model		<p>Null</p>	
Experimental result			



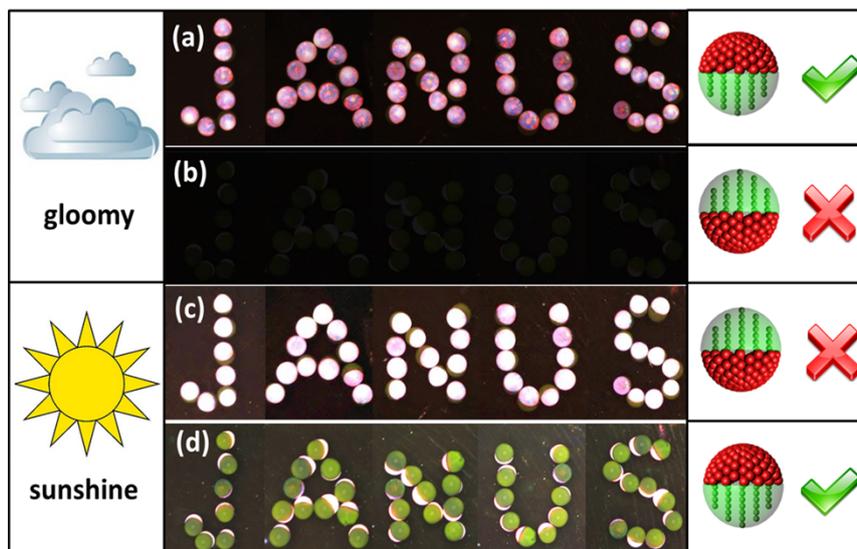
**Figure S2** Optical microscope images of the PS/SiO<sub>2</sub>-TMPTA JSs: (a) SiO<sub>2</sub>-TMPTA hemispheres, (b) PS hemispheres and (c) interface of the biphasic Janus supraballs and (d) corresponding SEM images.



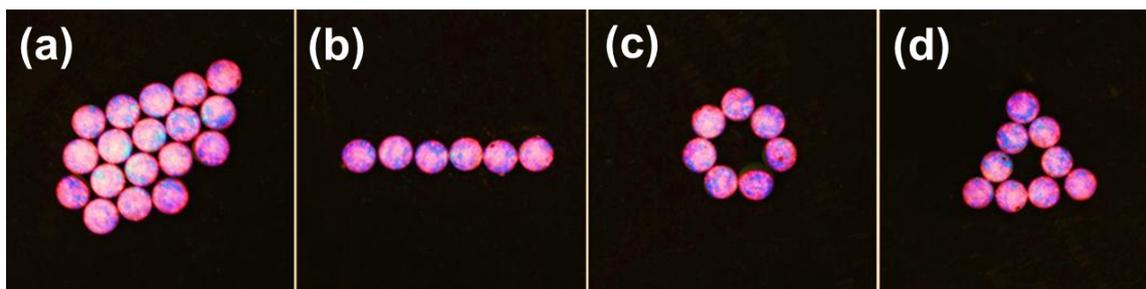
**Figure S3** Field dependence of the magnetization for dried PS/Fe<sub>3</sub>O<sub>4</sub>-TMPTA JSs at 300 K.



**Figure S4** Rotation control of single-phase  $\text{Fe}_3\text{O}_4$ -TMPTA supraballs. Without adding magnetic field, the supraballs arranged disorderly in the template holes, while adding a vertically magnetic field, all of the supraballs rotate to the same orientation with green color upside.



**Figure S5** Photographs taken under weak light intensity when J-2 supraballs were controlled by external magnetic field for rolling-over to (a) PS hemispheres upside and (b)  $\text{Fe}_3\text{O}_4$ -TMPTA hemispheres upside. Photographs taken under strong light intensity when Janus supraballs were controlled by external magnetic field for rotation to (c) PS hemispheres upside and (d)  $\text{Fe}_3\text{O}_4$ -TMPTA hemispheres upside.



**Figure S6** Magnetic-controllable patterning of PS/Fe<sub>3</sub>O<sub>4</sub> JSs (a) array, (b) line, (c) circle and (d) triangle.

The self-assembly behavior of these Janus particles were affected by magnetic field of different orientation or intensities. The self-assembly behavior of these Janus particles were affected by magnetic field of different orientation or intensities. As shown in Figure S6a, close-packed assembly were formed when large bar permanent magnet were posited underneath. The J-2 JSs arranged into a line along the edge of the big circle magnet in Figure S6b. For Figure S6c, a mini cylindrical magnet was placed under the particles, so the particles arranged into a small round. In Figure S6d, a triangle pattern was formed randomly when we paralleled moved the underlaying large bar magnet.

## References

1 <http://www.susqu.edu/facstaff/b/brakke/evolver/evolver.html>