Electronic Supplementary Information

Supracolloidal helices from soft Janus particles by tuning particle softness

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Figure S1: Distance dependence of the anisotropic attractive potential U_{ij} for different α_{ij}^R while keeping the adhesion energy $G = 2.0 \ k_B T$ and $\theta_i = \theta_j = 0^\circ$.



Figure S2: Excess pressure obtained from a series of simulations for the systems with increasing values of $\alpha_{ij}^R = 396$, 1596, 9996, and 39996 in the repulsive potential $U_{ij} = \frac{\alpha_{ij}^R}{2} \left(1 - \frac{r_{ij}}{r_c}\right)^2$, all the relations between the pressure and the number density can be fitted well by a parabola function $p = \rho k_B T + \kappa \alpha_{ij}^R \rho^2$ ($\kappa = 0.101 \pm 0.001$).



Figure S3: Supercolloidal helices self-assembled from Janus particles for different α_{ij}^A and β while keeping $\alpha_{ij}^R = 396$, $\nu = 0.5$, and $\phi = 5\%$. (a) Single helices at $\alpha_{ij}^A = 308$ ($G \approx 17.00 \ k_B T$), $\beta = 115^{\circ}$. (b) Double helices at $\alpha_{ij}^A = 242$ ($G \approx 11.50 \ k_B T$), $\beta = 120^{\circ}$.



Figure S4: The effect of the concentration of Janus particles ϕ on the formation of Bernal spirals as illustrated in Fig.2d. (a) $\phi = 1\%$. (b) $\phi = 2.5\%$. (c) $\phi = 5\%$. (d) $\phi = 7.5\%$.



Figure S5: (a) Self-assembly diagram of soft Janus particles in the G- β space at $\alpha_{ij}^R = 9996$ (E = 0.667 MPa), $\nu = 0.25$, and $\phi = 5\%$. Blue triangles, red spheres, and green squares denote the states with predominantly (b) micellar clusters, (c) Bernal spirals, and (d) the arrested state, respectively.



Figure S6: (a) Self-assembly diagram of soft Janus particles in the G- β space at $\alpha_{ij}^R = 1596$ (E = 0.116 Mpa), $\nu = 0.25$, and $\phi = 5\%$. Blue triangles, red spheres, and green squares denote the states with predominantly (b) micellar clusters, (c) Bernal spirals, and (d) the arrested state, respectively.



Figure S7: (a) Distribution S(n) of the number of nearest neighbors per Janus particle n, and (b) the helical order parameter Q_h for different α_{ij}^R while keeping adhesion energy $G = 55.00k_BT$, $\beta = 80^\circ$, $\nu = 0.25$ and $\phi = 5\%$.



Figure S8: The radial distribution function g(r) for typical self-assembled helical structures in Fig. 3.

Figure S9: Relative distributions of neighboring particles in typical self-assembled structures in Fig.2. (a) Distribution S(n) of the number of nearest neighbors per Janus particle n. (b) Distribution $P(\cos \gamma)$ of $\cos \gamma = \mathbf{n}_i \cdot \mathbf{n}_j$ for all pairs of contacting Janus particles.

Figure S10: Typical snapshots in the self-assembling process of single helices. (a) 0τ . (b) $4.0 \times 10^2 \tau$. (c) $2.0 \times 10^3 \tau$. (d) $3.2 \times 10^4 \tau$.

Figure S11: Typical snapshots in the self-assembling process of double helices. (a) 0τ . (b) $4.0 \times 10^2 \tau$. (c) $2.0 \times 10^3 \tau$. (d) $3.2 \times 10^4 \tau$.