Supporting Information

Chain Stiffness-Induced Entropy Effects Mediate Interfacial Assembly of Janus Nanoparticles in Block Copolymers: from Interfacial Nanostructures to Optical Responses

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The files include:

Figures S1-S9

SI-1. Relation between persistence length and K_a

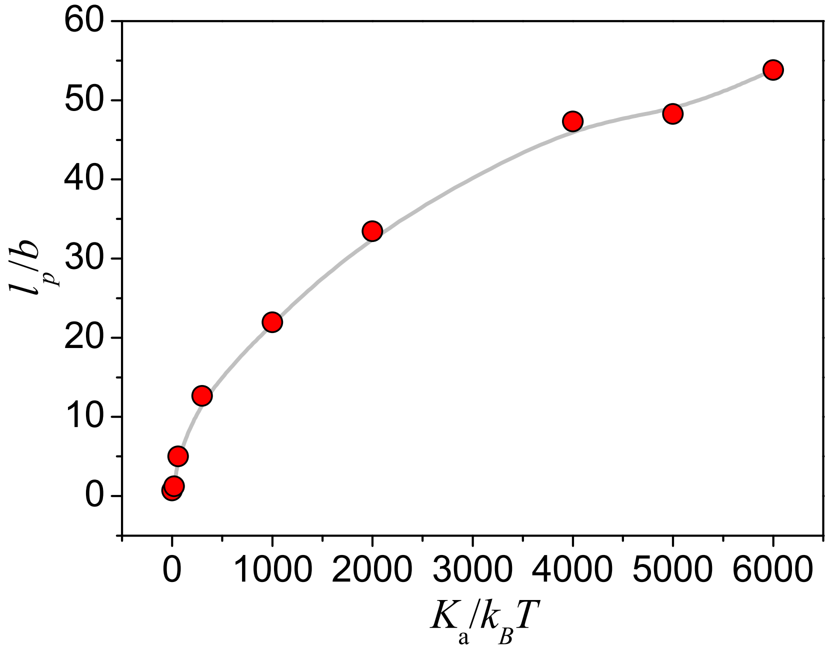

Figure S1. Persistence length of the polymer chain, l_p, as a function of K_a. Here b=0.5r_c is the equilibrium bond length. The line is to guide the eye. Here l_p is calculated through systematical DPD simulations of a single polymer chain.[1] The relationship between K_a and l_p indicates a detailed correlation between our simulations results and the possible experimental systems.

Ref.1: Micka, U.; Kremer, K. *J. Phys.*: *Condens. Matter* **1996**, *8*, 9463-9470.

SI-2. The detailed discretization forms of Eqs.3 and 4 in the main text.

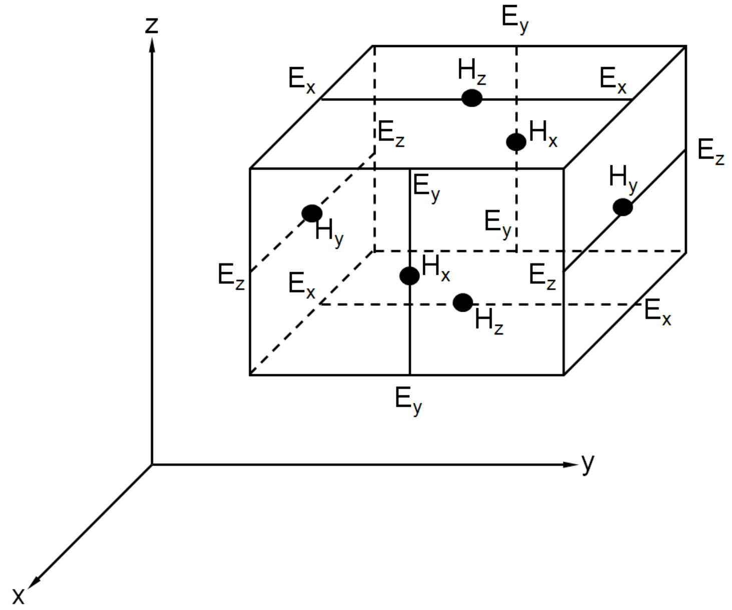

Figure S2. Positions of the field components within in Yee cell.

Following Yee's notation,[2] we assume that the grid point of the space is defined as (i, j, k) with coordinates $(i\delta_x, j\delta_y, k\delta_z)$, where $\delta = \delta_x = \delta_y = \delta_z$, is the cubic cell size, and δ_t is the time increment. Making the substitution

$$\widetilde{E} = \left(\frac{\varepsilon_0}{\mu_0}\right)^{1/2} E \tag{S1}$$

A set of finite-difference equations is obtained as follows:

$$\begin{aligned}
&\widetilde{E}_x^{n+1}(i+1/2, j, k) \\
&\quad = CA_x(i+1/2, j, k)\widetilde{E}_x^{n}(i+1/2, j, k) \\
&\quad + CB_x(i+1/2, j, k)[H_z^{n+1/2}(i+1/2, j+1/2, k) \\
&\quad - H_z^{n+1/2}(i+1/2, j-1/2, k) \\
&\quad + H_y^{n+1/2}(i+1/2, j, k-1/2) \\
&\quad - H_y^{n+1/2}(i+1/2, j, k+1/2)]
\end{aligned} \tag{S2}$$

$$
\begin{aligned}
H_x^{n+1/2}&\left(i, j+1/2, k+1/2\right) \\
&= H_x^{n-1/2}\left(i, j+1/2, k+1/2\right) \\
&+ RC[\widetilde{E}_y\left(i, j+1/2, k+1\right) \\
&- \widetilde{E}_y^n\left(i, j+1/2, k\right) \\
&+ \widetilde{E}_z^n\left(i, j, k+1/2\right) \\
&- \widetilde{E}_z^n\left(i, j+1, k+1/2\right)]
\end{aligned}
\tag{S3}
$$

$$
CA_x\left(i+1/2, j, k\right) = \frac{2\varepsilon_x\left(i+1/2, j, k\right) - \sigma_x\left(i+1/2, j, k\right)\delta_t}{2\varepsilon_x\left(i+1/2, j, k\right) + \sigma_x\left(i+1/2, j, k\right)\delta_t} \tag{S4}
$$

$$
CB_x\left(i, j, k+1/2\right) = \frac{2\varepsilon_0 RC}{2\varepsilon_z\left(i, j, k+1/2\right) + \sigma_z\left(i, j, k+1/2\right)\delta_t} \tag{S5}
$$

$$
RC = \frac{\delta_t}{\delta\left(\mu_0\varepsilon_0\right)^{1/2}} = 0.5 \tag{S6}
$$

Ref.2: Yee, S. K. *IEEE Trans*. *Antennas Prop*. **1966**, *AP-14*, 302-307.

SI-3. Additional simulation results

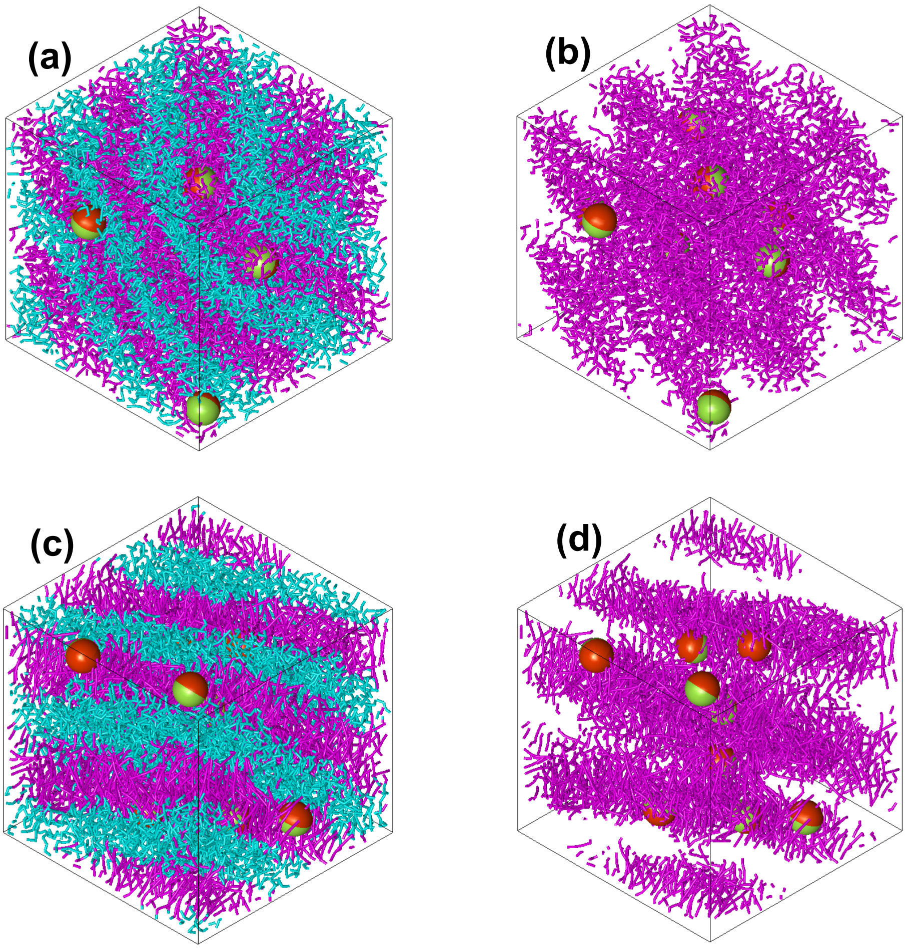

Figure S3. Comparison between the self-assembled structures of Janus nanoparticles in AB diblock copolymers with flexible (a, b) and rodlike (c, d) block B, where (a, b) K_a=0k_BT, and (c, d) K_a=1000k_BT. In (b) and (d), the segment of flexible block A is removed for clarity.

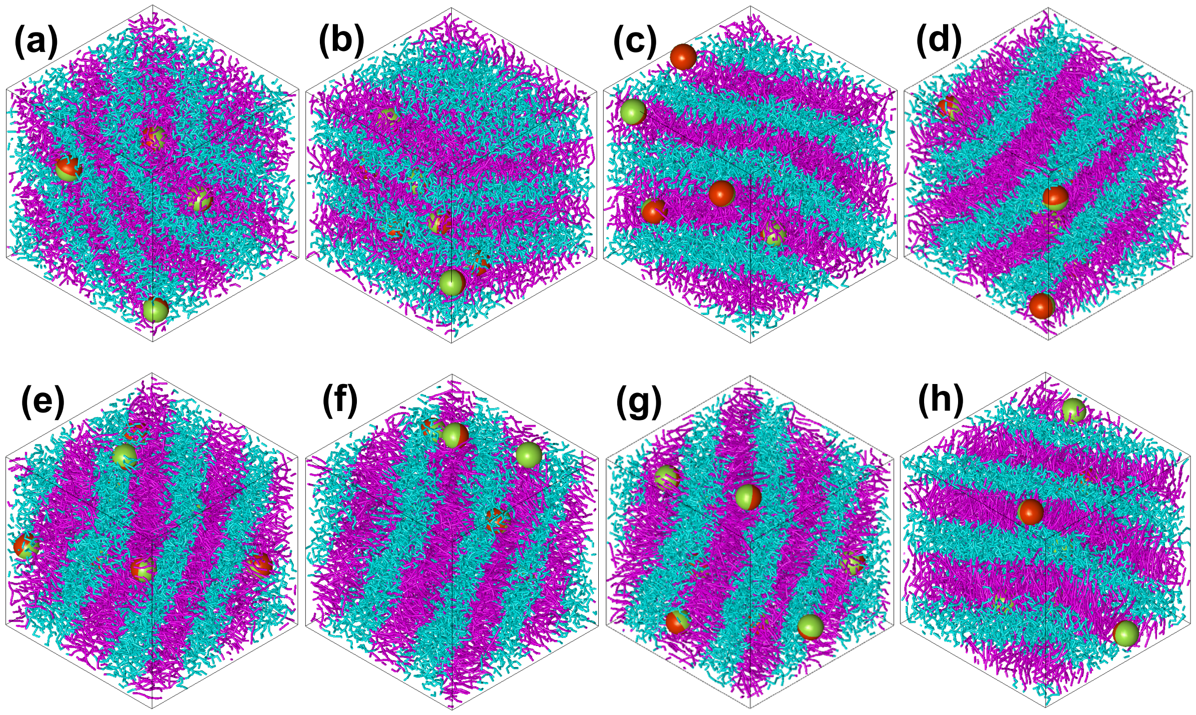

Figure S4. Self-assembly of Janus nanoparticles in AB diblock copolymers with flexible A block but stiff B block of a series of stiffness: (a) K_a=0k_BT, (b) K_a=20 k_BT, (c) K_a=40k_BT, (d) K_a=80k_BT, (e) K_a=100k_BT, (f) K_a=300 k_BT, (g) K_a=700k_BT, and (h) K_a=900k_BT.

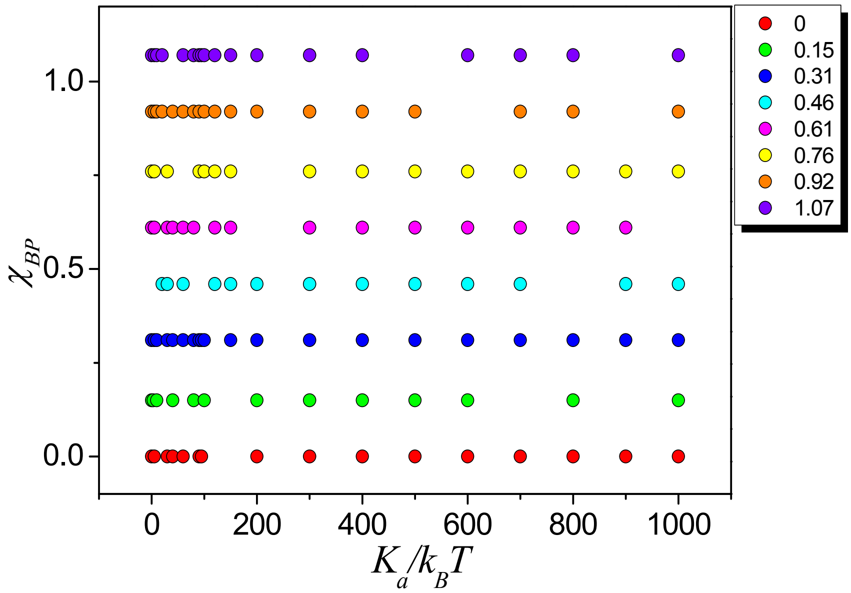

Figure S5. Points where independent runs were performed to generate the contour map of Figure 3.

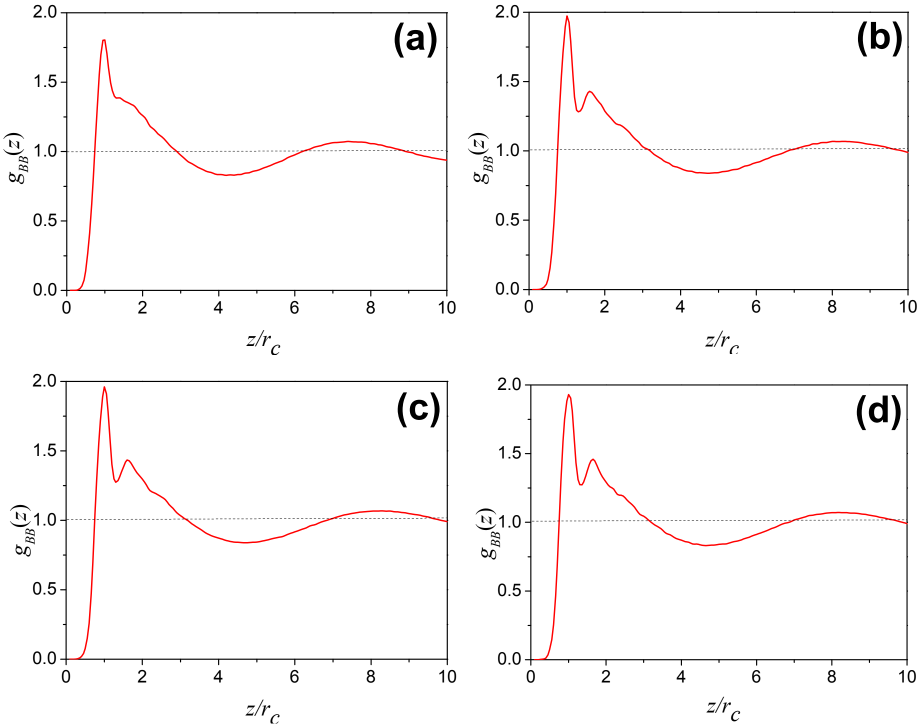

Figure S6. Pair correlation function between B-block beads for various stiffness of block B, $g_{BB}(z)$: (a) K_a=0k_BT, (b) K_a=60 k_BT, (c) K_a=100k_BT, and (d) K_a=400k_BT

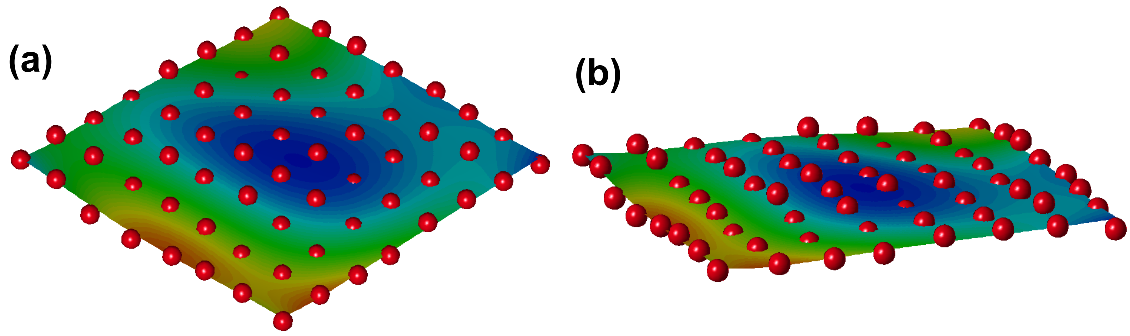

Figure S7. The position correlation between the interfacial DPD beads and the best-fitted surface of the phase interface from different views.

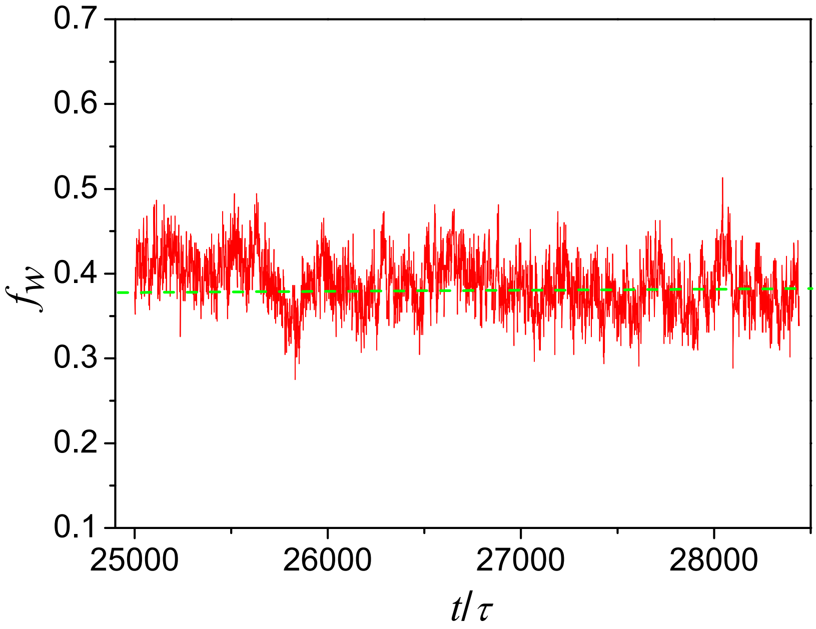

Figure S8. Temporal evolution of wrapping degree, f_w, after the formation of lamella structure for the stiff B block with K_a=100k_BT.

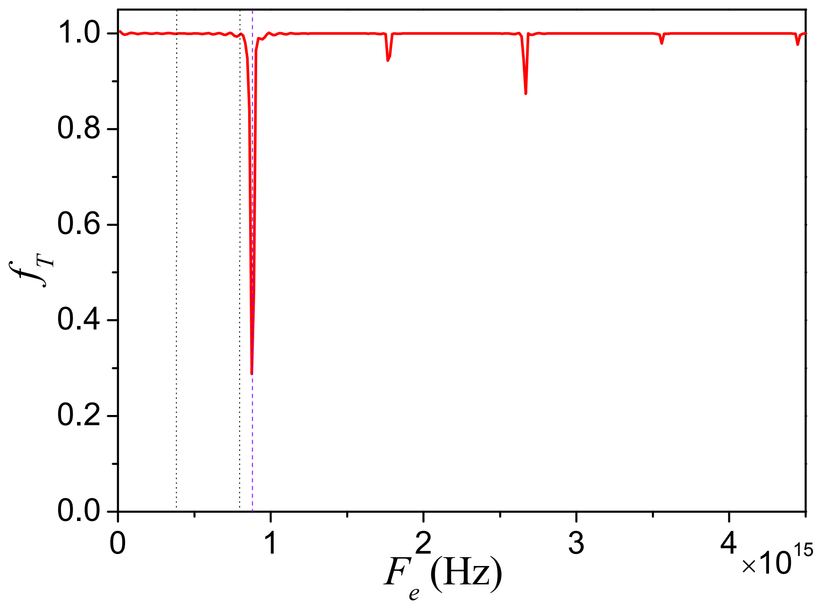

Figure S9. Transmittance spectra from the FDTD simulation of symmetry diblock copolymers. The dotted vertical lines mark the frequency range of visible spectrum. The violet dashed line denotes the principal frequency of the transmittance spectra.