Electronic Supplementary Material (ESI) for Soft Matter.
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Supplementary Information for "Melting and solid-solid transitions of two-dimensional crystals composed of Janus spheres"

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1 Simulation details

The anisotropic attraction slightly distorts the triangular lattice so that the unit cell contains two particles located at $(0,0)$ and $(a_x/2, a'_y)$ (Fig. 2a inset). The measured $a_x = 1.060$, $a_y = 1.870$ and $a'_y = 0.9997$ are determined from a perfect lattice at zero temperature with the minimum mean potential energy of the particles

$$U = -U_{12}/2 + \sum_{\substack{i=1,2 \\ |\boldsymbol{r}_i - \boldsymbol{r}_j| < r_{\text{cutoff}}}} U_{ij}/2,$$

where the subscript $i = 1, 2$ are the two particles in a unit cell.

We change the pseudorandom number generator in the LAMMPS code to the 64-bit Mersenne Twist algorithm (MT19937-64)[1, 2], which has a better performance in generating the thermal noise term in Langevin dynamics. We use the unit quaternions algorithm [3, 4, 5, 6] to represent the orientations of the particles and the Velocity Verlet algorithm[4, 5] to integrate the equations of motion. In each simulation, the system is slowly heated from $T = 0.01$ and is well equilibrated at each temperature step. The duration at each temperature is at least 5×10^7 dt and extended to 2×10^8 dt near the phase transition point. The time step is d$t = 0.002$.

2 Voronoi diagrams

The Voronoi diagrams in Fig. S1 show thermally activated defects such as dislocations and disclinations. A dislocation is a pair of neighboring particles with 5- and 7-nearest neighbors, respectively. A disclination is an isolated non-6-neighbor particle. A free dislocation can disrupt the quasi-long-range translational order and result in a crystal-to-hexatic phase transition, while a free disclination can disrupt quasi-long-range orientational order and result in a hexatic-to-liquid transition. However, dislocations

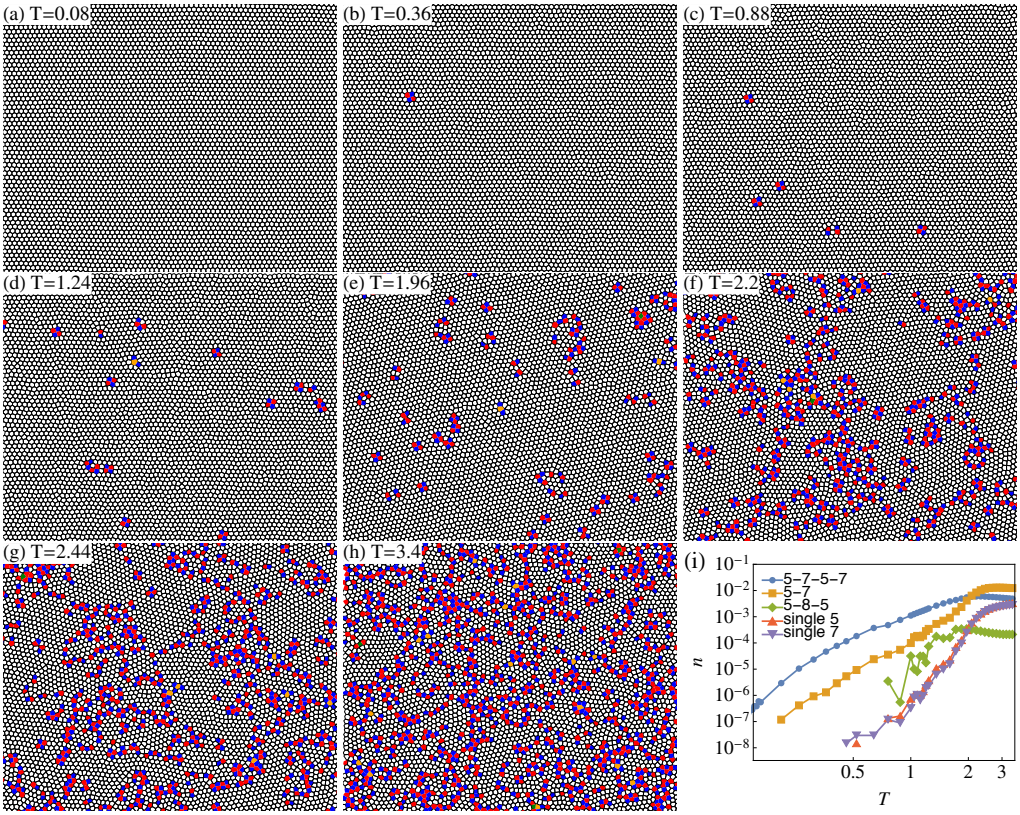

Figure S1: (a–h) Voronoi diagrams of a subarea at different temperatures. Quadrilateral: green; Pentagon: blue; Hexagon: white; Heptagon: red; Octagon: orange. (i) The densities of dislocation pairs (5-7-5-7 quartets), dislocations (5-7 pairs), 5-8-5 defects, 5-neighbor defects and 7-neighbor defects.

and disclinations are often not "free"; e.g., two separated dislocations with opposite Burger vectors are canceled, and the global order of the crystal phase remains intact. Whether dislocations and disclinations are free can hardly be identified; hence, the Voronoi diagrams only provide a rough estimate of the solid-hexatic and hexatic-liquid transition points. Figure S1 shows that the crystal has almost no defects at $T \lesssim 0.88$. At $0.36 \lesssim T \lesssim 0.88$, the 5-7-5-7 quadrupoles are not regarded as defects since their Burger vectors are zero. At $T \lesssim 0.36$, even quadrupoles do not form (Fig. S1a), indicating that the triangular lattice is nearly perfect. At $0.88 \lesssim T \lesssim 1.8$, the dislocation density is low, and their Burgers vectors are mainly canceled. In the high-temperature regime $1.8 < T < 2.5$, the triangular lattice melts in two steps, with the hexatic phase at $1.84 < T < 2.32$ (Fig. S1e-f) and liquid phase at $T > 2.32$ (Fig. S1g-h).

3 Movies

Movie 1: Stripe domains drift randomly under thermal fluctuations at $T = 0.061$.
Movie 2: The nucleation and growth of stripe domains in the solid-solid transition at $T = 0.061$.

References

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