Supplementary Information

Self-Assembly of Archimedean Tilings with Enthalpically and Entropically Patchy Polygons

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Calculation of entropic potential of mean force and torque. We computed the tendency for particles to align entropically *via* the entropic potential of mean force and torque.[40] We considered a pair of polygons in a sea of other polygons, and denote nearby relative positions and orientations of the pair by $\Delta\xi_1$ and $\Delta\xi_2$. The free energy difference for the sea particles between these two states is given by the logarithm of the ratio of the number of microstates available to the sea when the pair is in the configuration $\Delta\xi_1$ to the number of microstates when it is in the configuration $\Delta\xi_2$, which we denote by $\Omega\,(\Delta\xi_1)$ and $\Omega\,(\Delta\xi_2)$ respectively. To evaluate this ratio we note that some microstates in $\Omega(\Delta\xi_1)$ will also be in $\Omega\,(\Delta\xi_2)$. The number of such states will be proportional to the probability that a randomly selected state in $\Omega\,(\Delta\xi_1)$ is also in $\Omega(\Delta\xi_2)$. To determine this probability we fix a pair of tiles at $\Delta\xi_1$, and compute the probability that a trial move to $\Delta\xi_2$ is accepted $p(\Delta\xi_1 \rightarrow \Delta\xi_2)$. We also determine the probability $p(\Delta\xi_2 \rightarrow \Delta\xi_1)$ of the reverse move from $\Delta\xi_2$ to $\Delta\xi_1$. In addition we must also take into account the difference in infinitesimal volumes available to the pair at a given relative position and orientation. The PMFT difference is then given by

$$e^{-\beta(F_{12}(\Delta\xi_1)-F_{12}(\Delta\xi_2))} = \frac{J(\Delta\xi_1)}{J(\Delta\xi_2)}\frac{p(\Delta\xi_1 \rightarrow \Delta\xi_2)}{p(\Delta\xi_2 \rightarrow \Delta\xi_1)}$$

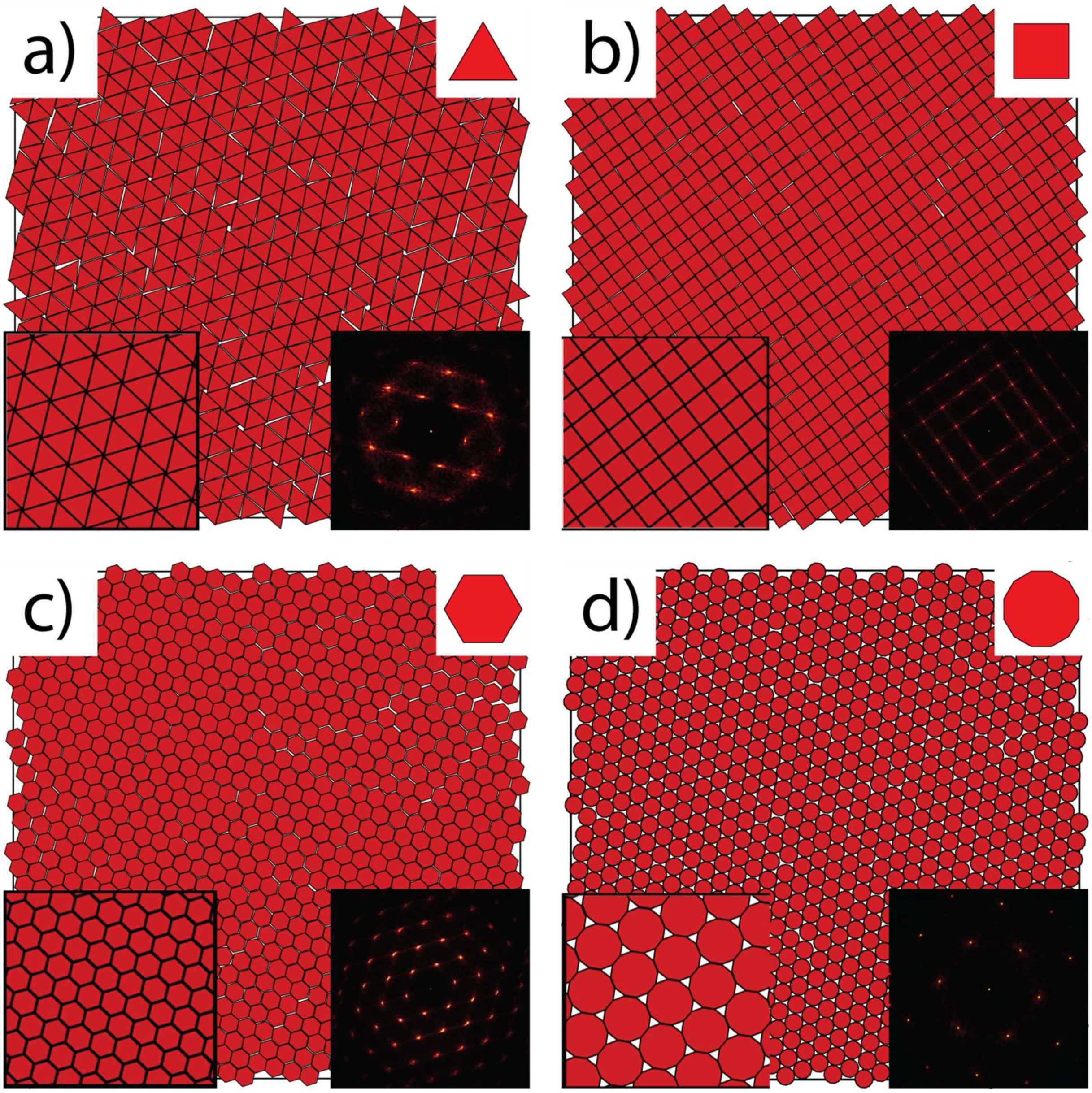

Figure S1. Representative snapshots of sections of larger simulations for the (3^6), (4^4), (6^3), and (3.12^2) ATs self-assembled with excluded volume interactions only. Insets show the nanoplate, a diffraction pattern of the snapshots, and a compressed close-up. (a) Triangles self-assemble the (3^6) tiling at a packing fraction of 0.90, (b) squares self-assemble the (4^4) tiling at a packing fraction of 0.94, (c) hexagons self-assemble the (6^3) tiling at a packing fraction of ~ 0.93, and (d) dodecagons self-assemble the (3.12^2) tiling at a packing fraction of ~ 0.85.