Supplementary Information

Orientation Correlation Function (OCF)

Figure 1 Difference in decay rates for self-assembled rectangles in a dense fluid and a solid.

The OCF is calculated from the particle orientation, \( \theta_n \), where \( n \) takes into account the symmetry of the system. Given the underlying symmetry of the system, the \( \theta_4 \) order parameter is calculated:

\[
\theta_4 (\vec{r}_j) = e^{i4\theta_j}
\]

This order parameter can then be used to calculate the Orientation Correlation Function, OCF:

\[
OCF(r) = \frac{1}{N_r} \sum_{i=1}^{N} \theta_4 (\vec{r}_i) \cdot \theta_4^* (\vec{r}_j), \ r - dr < |r_{ij}| < r + dr
\]

This function can be used to determine whether the system is in a fluid or solid state, based on the functional form of the decay: a solid will have a power-law decay while a fluid will decay exponentially, as shown in fig. 1.
Figure 2 At constant $n_k = 4$, the amplitude is varied to determine its effect on the bonding well. As amplitude increases, the width and depth of the well increases showing the overall increase in the DEF. The well associated with the random domino tiling is also eliminated as $A$ increases.

Figure 3 At constant $A = 0.14$, the wavenumber is varied to determine its effect on the bonding well. At $A = 0.14$, even $n_k$ have greater effect on the depth of the bonding well than odd $n_k$, but neither has much effect on the well corresponding to the random domino tiling.
Figure 4 At constant $A = 0.28$, the wavenumber is varied to determine its effect on the bonding well. At $A = 0.28$ both even and odd $n_k$ for $n_k > 1$ increase the depth of the bonding well. Odd $n_k$ is not effective at eliminating the well associated with the random domino tiling. $n_k = 2, 4$ eliminate the well corresponding to the random domino tiling, which returns for $n_k = 6, 10$. 
Figure 5 Snapshots showing the best assemblies of equilibrium simulation frames for (a) rectangles ($\beta L^2P = 9.6$) and allophilic rectangles with (b) $n_k = 1$, $A = 0.14$, $\beta L^2P = 10.2$, (c) $n_k = 3$, $A = 0.14$, $\beta L^2P = 12.0$, (d) $n_k = 2$, $A = 0.28$, $\beta L^2P = 13.0$, (e) $n_k = 3$, $A = 0.28$, $\beta L^2P = 13.6$, (f) $n_k = 4$, $A = 0.57$, $\beta L^2P = 8.8$, (g) $n_k = 5$, $A = 0.28$, $\beta L^2P = 11.4$, (h) $n_k = 6$, $A = 0.28$, $\beta L^2P = 10.0$, (i) $n_k = 10$, $A = 0.28$, $\beta L^2P = 9.6$. Wavenumbers increase left to right, while amplitude increases top to bottom. Bonded shapes are colored as in Figure 2 in the main paper; otherwise, they are colored grey. Shapes that improve assembly of the square lattice relative to rectangles are indicated by a green surrounding box.