Exploiting non-equilibrium phase separation for self-assembly
Supporting Information

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1 Assembly of star and wedges at constant pressure

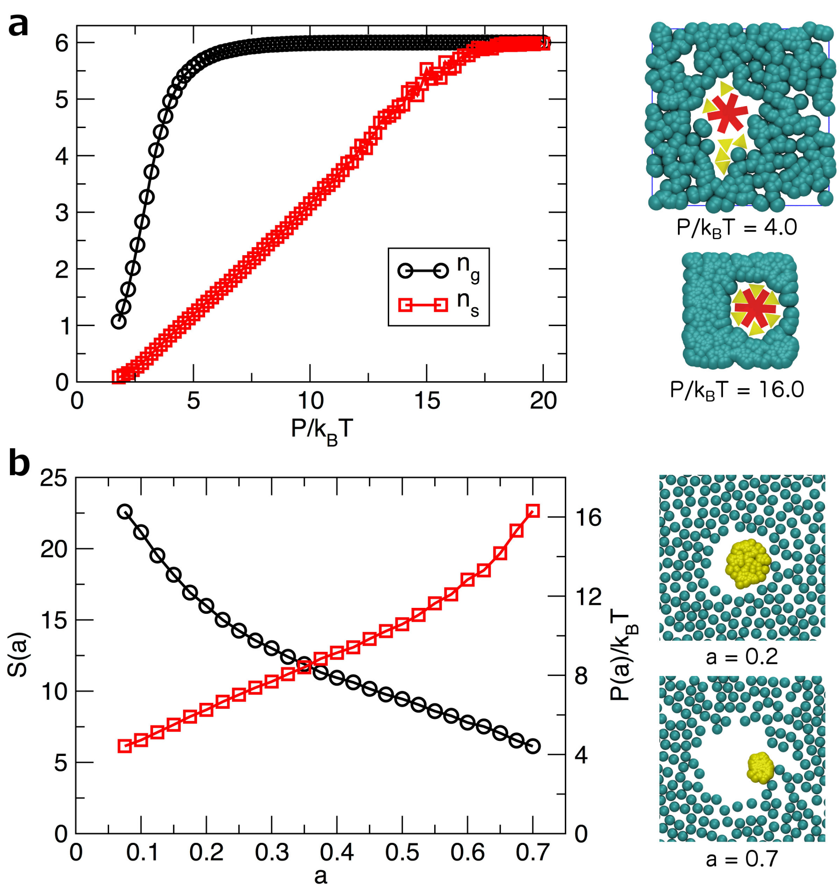
To probe the assembly behavior of the star-and-wedges at equilibrium conditions, we performed constant pressure Monte Carlo simulations with standard volume updates [1]. The simulated systems comprised the seven polyhedral shapes (with size mismatch between wedges and pockets of the star) and 439 ideal gas particles of diameter σ that serve as a pressure bath. Ideal gas particles do not have any mutual interactions but exclude area in interactions with star and wedges. These interactions favor demixing of ideal gas particles and star/wedges at high density or pressure, as illustrated in Supporting Figure 1a. Measures of the degree of aggregation and assembly increase monotonically with pressure.

2 Pressure effected by phase segregation

We define the effective (surface) pressure that is exerted by a driven group of particles on an undriven, segregated group of particles by measuring the area occupied by a collection of segregated ideal gas particles. To this end, we performed Monte Carlo simulations of a system of 399 hard discs and 100 ideal gas particles. Discs undergo the biased "shaking" dynamics described in the main manuscript with $\tau = 1200$. (The size of the simulation box was chosen to match the area accessible to shaking discs to that in simulations of the star and wedges system at a packing fraction of 0.65.) For amplitudes larger than approximately $a = 0.05$, ideal gas particles form a segregated dense cluster. To measure the area S occupied by the cluster, we define a grid of square cells with edge length 1.042σ; S is defined as the total area of all cells that are occupied by at least one ideal gas particle. We then define the effective pressure using the ideal gas equation of state, $P/k_{\mathrm{B}}T = N/S$. Time averages of area $S(a)$ and pressure $P(a)$ are plotted as a function of the amplitude of shaking in Supporting Figure 1b.

References

[1] Frenkel, D. & Smit, B. *Understanding Molecular Simulation* (Academic Press, San Diego, 2002), 2 edn.



SUPPORTING FIG. 1: (**a**) Average values of aggregated wedges n_{g} and assembled wedges n_{s} as a function of pressure in Monte Carlo simulations using ideal gas particles as a pressure bath. Snapshots show typical configurations at low and high pressure. (**b**) Area $S(a)$ (black circles) occupied by a cluster of 100 ideal gas particles in a bath of driven hard discs, as a function of amplitude a. Red squares indicate the corresponding effective pressure $P(a)/k_{\mathrm{B}}T$ calculated from the ideal gas equation of state. Snapshots show typical configurations at two different amplitudes. (Ideal gas particles are colored yellow.)