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Is a hierarchical dynamics the best route to the self-assembly of a hierarchical structure? THOMAS HAXTON, STEPHEN WHITELAM, Lawrence Berkeley National Laboratory — Mimicking nature's ability to assemble functional hierarchical materials will require understanding how to promote the self-assembly of structure on multiple lengthscales while avoiding kinetic traps. We use computer simulation to study the self-assembly of a simple hierarchical structure, a square lattice whose repeat unit is a tetramer. Although the target material is organized hierarchically, it self-assembles most reliably when its assembly pathway consists of the sequential addition of monomers to a single structure. Hierarchical assembly pathways via dimer and tetramer intermediates result in lower yield, because these intermediates tend to associate in ways incompatible with the target structure. In addition, assembly via tetramers results in the formation of incomplete building blocks (trimers) that cannot combine to form the target crystal. We use analytic theory to relate assembly pathways to the underlying thermodynamics, identifying two principles for optimal assembly: 1) make the free energy gap between the target phase and the most stable fluid phase comparable to the thermal energy, and 2) ensure that no other dense phases (liquids or close-packed solids of monomers or oligomers) or fluids of incomplete building blocks fall within this gap.

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