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Engineering the Entropic Bond for Inverse Design of Colloidal Assemblies

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Abstract:
Entropy, information, and order are important concepts in many fields, relevant for materials to machines, for biology to economics. Entropy is typically associated with disorder; yet, the counterintuitive notion that a thermodynamic system of hard particles might - due solely to entropy - spontaneously assemble from a fluid phase into an ordered crystal was first predicted in the mid-20th century, first for rods, and then spheres. The ordering of colloids by entropy maximization upon crowding is now well established, producing colloidal crystals possessing order and complexity beyond anything previously imagined. We show how entropic bonding can be quantified, discuss the notion of the entropic bond in the context of traditional chemical bonds, and show how the entropic bond may be *engineered* despite its statistical, emergent nature. We present a new thermodynamic approach to the inverse design of colloidal matter we call Digital Alchemy, and demonstrate its use in obtaining colloidal crystals with arbitrary complexity optimized for structure and properties.