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Entropically Stabilized Colloidal Crystals Hold Entropy in Collective Modes[1] JAMES ANTONAGLIA, GREG VAN ANDERS, SHARON GLOTZER, Univ of Michigan - Ann Arbor — Ordered structures can be stabilized by entropy if the system has more ordered microstates available than disordered ones. However, "locating" the entropy in an ordered system is challenging because entropic ordering is necessarily a collective effort emerging from the interactions of large numbers of particles. Yet, we can characterize these crystals using simple traditional tools, because entropically stabilized crystals exhibit collective motion and effective stiffness. For a two-dimensional system of hard hexagons, we calculate the dispersion relations of both vibrational and librational collective modes. We find the librational mode is gapped, and the gap provides an emergent, macroscopic, and density-dependent length scale. We quantify the entropic contribution of each collective mode and find that below this length scale, the dominant entropic contributions are librational, and above this length scale, vibrations dominate. This length scale diverges in the high-density limit, so entropy is found predominantly in libration near dense packing.

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