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Title: **Thermodynamics of structure-forming systems**

Abstract

Structure-forming systems are ubiquitous in nature, ranging from atoms building molecules to self-assembly of colloidal amphibolic particles. The understanding of the underlying thermodynamics of such systems remains an important problem. Here, we derive the entropy for structure-forming systems that differs from Boltzmann-Gibbs entropy by a term that explicitly captures clustered states. For large systems and low concentrations the approach is equivalent to the grand-canonical ensemble; for small systems we find significant deviations. We derive the detailed fluctuation theorem and Crooks' work fluctuation theorem for structure-forming systems. The connection to the theory of particle self-assembly is discussed. We apply the results to several physical systems.
We present the phase diagram for patchy particles described by the Kern-Frenkel potential. We show that the Curie-Weiss model with molecule structures exhibits a first-order phase transition.