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Entropically-Driven Destabilization of Nanoparticle Crystals SANAT KUMAR, Columbia Univ — Detailed computer simulations show that polymer-induced depletion forces can cause nanoparticles (NPs) in the athermal limit to crystallize, but only for short polymers at melt-like densities. For long chains, this depletion-induced attraction is in competition with the entropy loss associated with confining a polymer chain within the cavities in the NP crystal. For chains larger than these voids, these crystals are unstable and the NPs form an aggregated, but non-crystalline structure. The experimental results of Mackay et al. [Science 311:1740, 2006], who find immiscibility for chains smaller than the NP radii but miscibility for larger chains, thus probably reflect this physics, which we show to only be important when the osmotic pressure driving the polymer chains into the NP crystal is relatively large.

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