The mystery of the discrepancy between experiments and simulations in hard sphere crystallization

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A set of hard spheres is arguably the simplest system that shows a fluid-to-crystal transition [1]. This system can be studied numerically with either Monte Carlo or Molecular Dynamics simulations [1,2] and experimentally with suspensions of colloidal particles stabilised against aggregation [3]. Experiments and simulations are consistent in the equilibrium freezing and melting densities [3]. However, experimental measures of the nucleation rate are not in good agreement with numerical calculations [2,4]. This issue has been investigated by several groups [5,6] and the latest conclusion is that "the discrepancy still persists" [6]. We have re-examined the crystal nucleation rate in a wide density range with a novel powerful approach called seeding [7]. Indeed, the discrepancy between experiments and simulations seems to be large for low densities (of tens of orders of magnitude for the nucleation rate). However, we propose a possible explanation for such discrepancy.

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