Séminaire

Lundi 15 juin 2026 à 14h30

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Freezing of Polydisperse Hard Spheres into Complex Crystal Phases

Hard spheres are arguably the most fundamental model system used in studies of colloidal self-assembly. As a result, we have extensive knowledge of their phase behavior and dynamics - at least for systems consisting only of one or two sizes of sphere. However, the situation becomes more complex once we mix a wide range of particle sizes. In reality, colloidal particles are never truly monodisperse: they always show at least a small variation in their sizes. In computer simulations, this polydispersity is often ignored for the sake of convenience: monodisperse systems avoid the need to choose a detailed distribution of particle characteristics, have simpler phase behavior, and are easier to simulate. However, polydispersity can have a profound effect on the phase diagram. Even small amounts of polydispersity are known to significantly shift phase boundaries [1], and simulations suggest the emergence of complex crystal structures at high polydispersity [2], replacing the simple face-centered cubic (FCC) structure that is stable for single-sized spheres.

Unfortunately, existing computational methods to determine equilibrium phase diagrams for polydisperse systems are cumbersome, and often rely on significant approximations. In this talk I will describe a new simulation approach that we recently developed for locating fluid-crystal coexistences in polydisperse mixtures [3]. Specifically, we make use of direct coexistence simulations in the semigrand-canonical ensemble to determine the freezing line of polydisperse hard spheres with a Gaussian size distribution. We show that with increasing polydispersity, the FCC phase is replaced by more complex crystal structures, resembling binary or ternary phases. This work sheds new light on the freezing behavior of polydisperse hard spheres, and provides a convenient route for the exploration of other polydisperse systems in the future.

[1] M. Fasolo and P. Sollich, Physical Review Letters **91**, 068301 (2003).

[2] P. K. Bommineni, N. R. Varela-Rosales, M. Klement, and M. Engel, Physical Review Letters, **122**, 128005 (2019).

[3] A. Castagnède, L. Filion, and F. Smallenburg, Journal Of Chemical Physics **163**, 134121 (2025).

Les personnes souhaitant rencontrer l'orateur sont priées de prendre contact avec Jean Farago.