Nucleation and growth processes by molecular simulations
Application to molecular and colloidal crystals

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Project description: Crystals, composed of a regular and periodic arrangement of either atoms/molecules or nanoparticles, are ubiquitous in Nature but are also an important component in many field of applications, from optics to energy storage to construction materials. Understanding and controlling their formation is thus of paramount importance. It proceeds through the nucleation of small nuclei which further grow/crystallize to lead to the final crystalline material. The nucleation, thus, plays a crucial role in the control of the microscopic properties (size, purity, structure, morphology), which directly affect the physical chemistry of the crystalline material formed at the macroscopic scale. However, this process is still far from being well understood and is a field of intensive research. This is because experiments still struggle to characterize crystal nucleation, which happens on exceedingly small/short length/time scales (ns/nm). Conversely, molecular simulations, which could indeed provide invaluable insight, are hampered by the fact that nucleation is a rare event, as seconds, or days or even weeks are typically needed for a crystalline nucleus to reach its critical size and proceed toward crystallization. Thus, the development and use of enhanced sampling techniques are needed to tackle the time scale problem via molecular simulations. In the INTERFACES Department, an important research effort is put on the nucleation and growth of solid crystalline materials and thin films of oxides for applications in catalysis, energy storage, construction materials, photonics and environment see e.g [1-3]. At present, it is the subject of concern in five PhD thesis (Lina Bouzouaid, Estelle Poupelloz, Antoine Patt, thèse Simon Kimber, Guillaume Bareigts), several master internships, one ANR project on gas hydrates and one European consortium project on cementitious hydrates. The main goal of the PhD project will be to develop original simulation techniques based on Monte-Carlo and Molecular simulations to study the nucleation and growth of crystals and to deployed them on either gas hydrates, cementitious hydrates or colloidal crystals for which we have a large set of experimental data. The work will benefit from the recent and original development of enhanced sampling techniques allowing the calculation of the Gibbs free energy and surface free energy[4-6] as well as from a large set of numerical tools to follow the nucleation and growth process at a microscopic scale [7].

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