

# LARGE-SCALE, LONG-TIME MOLECULAR DYNAMICS SIMULATION OF CRYSTAL GROWTH

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## EXECUTIVE SUMMARY

In order to discover the fundamental principles underlying the process of crystal growth we study soft-matter systems that self-assemble into a plethora of different structure types. Our open-source simulation package HOOMD-blue [1], which is designed for GPU architectures, offers molecular dynamics [2] as well as hard-particle Monte Carlo simulation techniques [3] and grants us access to the workings of a large range of systems with various interactions. We are simulating the growth of a variety of crystal structures from the fluid. By comparing the growth of simple and complex or even aperiodic structures, we aim for a universal understanding of the formation of ordered structures from disordered fluids. We anticipate that our studies will impact multiple fields of chemistry, physics, materials science, chemical engineering, geology, pharmaceutical sciences, and any disciplines in which crystal formation, nucleation and growth, and/or the emergence of order from disorder are of interest.

## INTRODUCTION

Crystal growth is still a largely obscure process that researchers have been trying to clarify for decades [4]. According to classical theories, crystals grow from the liquid in an atom-by-atom fashion. This model seems to work well enough for simple crystal structures, which consist of particles or atoms of only one kind that all have the same crystallographic environment in the final crystal. However, if the ensuing crystal structure will contain dozens, hundreds, or thousands of atoms in a unit cell—or if it is aperiodic, as are quasicrystals, meaning that no unit cell exists at all—how does each atom find its position in the crystal?

While the observation of crystal growth in experiments has been, until very recently, an almost impossible undertaking in atomic systems and remains challenging on the soft-matter length scale, molecular dynamics simulation can grant access to this phenomenon on adjustable time and length scales. By simulating crystal structures of varying complexities with HOOMD-blue and observing particle attachment and particle environments prior to and after their attachment to a growing crystal, we can identify characteristic growth modes and compare structural motifs in the fluid and crystalline phases.

## METHODS & RESULTS

In previous studies, we identified different crystal structures that occur in systems growing from an oscillating pair potential [5], as well as a Lennard-Jones Gauss potential. We are now investigating crystal growth with atomistic resolution in order to resolve the relationship between the complexity of a crystal structure and its growth mechanism.

We are performing molecular dynamics simulations of the growth of simple crystal structures. We are investigating the densest sphere packings: the

cubic close packing (*ccp*, also: *cF4*-Cu structure type) and the hexagonal close packing (*hcp*, also: *hP2*-Mg structure type), as well as the body-centered cubic packing (*cI2*-W structure type), the simple cubic structure (*cP1*-Po), a simple hexagonal structure (*hP1* structure type), a simple chiral cubic structure (*cP4*-Li structure type), and a compressed version of the hexagonal close packing (*hP2*) with different resulting particle environments.

For all of those structures we have now determined the respective crystallization temperatures and extracted snapshots of a newly formed crystal nucleus at that temperature. These snapshots will be used for seeded growth simulations conducted at the crystallization temperature, in order to (a) bypass nucleation, which is a rare event and an interesting mechanism that should be studied separately, and (b) avoid multiple nucleation events, which would make the data much harder to interpret. These seeded simulations can then be successively scaled up to systems containing many millions of particles in order to reach a regime where size-effects of the crystallite play a vanishing role.

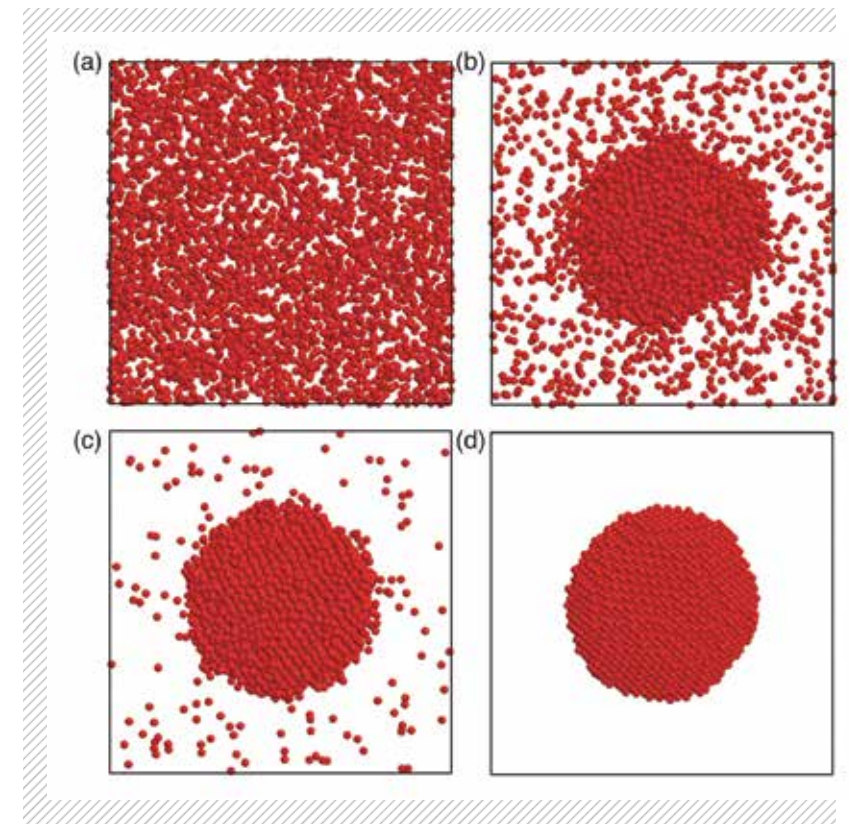
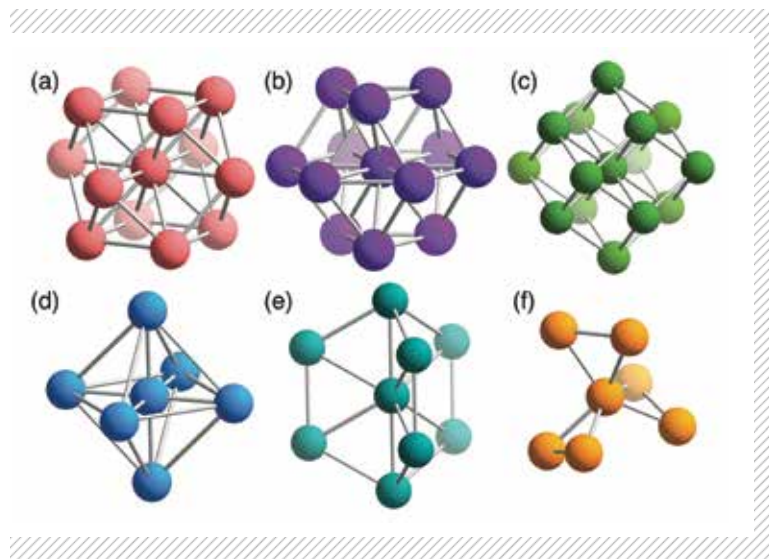
In the following months, increasingly complex, periodic crystal structures will be simulated, including different Frank-Kasper phases, clathrates, and other, previously unreported structures. Furthermore, the growth of an icosahedral quasicrystal [5] will be simulated and analyzed. Consequently we will be able to better understand the difference between the growth of an aperiodic and a periodic crystal structure, or otherwise describe how both can be regarded as variations of the same principle mechanism.

By tuning the parameters of the isotropic pair potential that we are using, we can access phase regions that exhibit crystal growth from either the fluid or the gas phase. Contrasting both regimes will help us understand how universal the observed motifs of crystal growth truly are.

## WHY BLUE WATERS

Blue Waters is the **only** NSF-funded system that offers large-scale access to GPUs. GPUs can accelerate soft matter simulations by more than an order of magnitude vs. CPU-based calculations, and therefore drastically improve the turn-around for research. The performance of the HOOMD-blue code was previously benchmarked with Blue Waters [6], demonstrating its ability to scale on up to 1,024 nodes. Blue Waters allows us to run the

**FIGURE 1:** Coordination polyhedra of the simple crystal structures grown with attractive isotropic pair potentials. (a) *cF4*-Cu, (b) *hP2*-Mg, (c) *cI2*-W, (d) *cP1*-Po, (e) *hP2* (compressed), (f) *cP4*-Li.



**FIGURE 2:** Simulation snapshots of (a) the fluid phase, (b) the emerging liquid droplet, (c) the forming crystallite, and (d) the final crystal, at successively decreasing temperatures.

## NEXT GENERATION WORK

Within the framework of this project, we are focusing on the exploration of crystal growth. The nucleation of a crystalline seed is a physically distinct process that requires far larger compute resources to explore, as it is a rare event that needs to be probed with statistical methods and hence a large number of very large sample runs. We envision that studying the nucleation of diverse crystal structures from various initial states would become feasible with the next-generation Track-1 computing architecture.