Understanding crystallization with ab initio machine learning simulation

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Crystallization is a process of key importance for many modern technologies, such as the manufacturing of pharmaceuticals, and it also plays a central role in geological, planetary, and climate sciences. Over the years, molecular dynamics simulations have provided key insights into the microscopic mechanisms underlying crystal nucleation and growth. The recent advent of machine learning (ML) interatomic potentials based on large datasets of *ab initio* electronic-structure calculations has improved the accuracy and predictive power of these simulations, and provided access to much larger system sizes and longer simulation times. Moreover, ML potentials have greatly expanded the scope of molecular simulations by providing access to reactive crystallization processes.

In this talk, I will discuss the use of this technique to drive large-scale, highly-accurate and reactive molecular dynamics simulations of crystallization. I will illustrate this approach with several examples. First, I will show that we can leverage this tool to compute homogeneous ice nucleation rates from first principles which are in remarkable agreement with experiment [1]. Furthermore, I will discuss some results about the formation of ice on feldspar, the most important ice nucleating particle in the atmosphere [2]. Finally, I will discuss the application of this technique to study the crystallization of calcium carbonate from aqueous solution, a process where reactivity plays an essential yet poorly understood role [3]. Taken together, these results show the great promise of machine learning as a tool to bridge time and length scales, and to provide insight into complex crystallization phenomena, which were thought to be out of reach for molecular simulation.

References

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