Understanding crystallization from solution and at interfaces with ab-initio machine-learning models

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Crystallization is a process of key importance for many modern technologies, including climate change mitigation via carbon mineralization and the manufacturing of pharmaceuticals, and it also plays a central role in geological, planetary, and climate sciences. In spite of its importance, the microscopic crystallization mechanisms of many systems are still today unknown due to the short time and length scales involved in the process. Molecular dynamics simulations driven by machine-learning models based on ab-initio quantum-mechanical theory offer a unique opportunity to simulate crystallization from first principles and with unprecedented accuracy. These state-of-the-art models capture phenomena such as polarization and bond forming/breaking which are often inaccessible to traditional empirical force fields. Here, we show that using the Deep Potential Molecular Dynamics (DeePMD) methodology [1] we can tackle a broad range of crystallization scenarios including crystallization from the melt, from solution, and at interfaces. We train models based on density-functional theory (DFT) calculations for complex systems with up to five chemical elements, and with the capacity to simultaneously describe ions in aqueous solutions and complex water/surface interactions. The presentation is structured as follows. First, we discuss recent calculations of ab initio ice nucleation rates from supercooled water using a model based on the SCAN DFT functional which gives remarkable agreement with experiment [2]. Then, we present new insights about the feldspar/water interface (Figure 1) with an eye towards understanding the very high ice nucleation activity of K-rich feldspars [3]. Lastly, we describe recent efforts to train models able to describe nucleation and growth of minerals from aqueous solution.

References

[1] Zhang, Han, Wang, Car, and E, Phys. Rev. Lett. 120, 143001 (2018)
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Figures

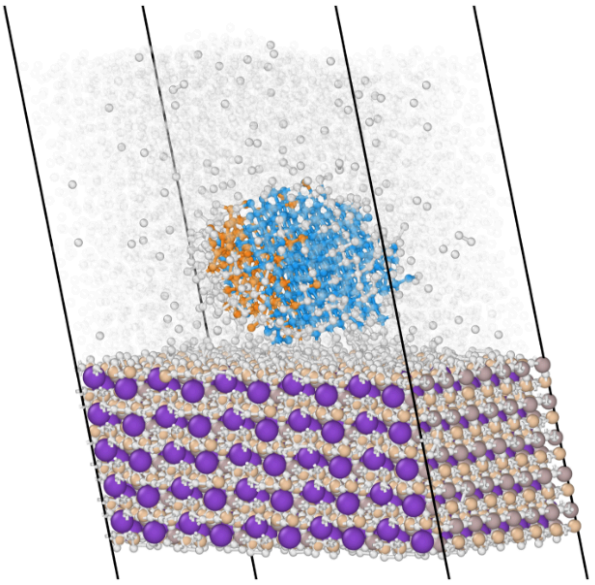

Figure 1. Ice cluster at the (001) surface of microcline feldspar in contact with water at 280 K (27 K of supercooling). K, Al, and Si atoms are shown as purple, gray, and brown spheres, respectively, with their corresponding van der Waals radii. O atoms with environments compatible with ice I_h and I_c are shown in orange and blue, respectively. O atoms with other environments are shown as white spheres. In order to easily visualize the solid cluster, we display with high transparency O atoms with displacement magnitudes larger than 3 Å over the previous 1 ns of simulation. For clarity, H atoms are not shown. Bonds are displayed between O atoms with interatomic distances smaller than 3 Å.