Does classical nucleation theory provide an accurate description for nucleation in metals?

Summary

Nucleation is the first step that occurs when a new phase of material grows from a supercooled or supersaturated phase. This phenomena is typically understood using a theory called classical nucleation theory. In this theory the driving force for nucleation is the lowering of energy that accompanies the transition from a metastable to stable phase. However, this transition is resisted because of the energetic penalty that is associated with forming an interface between the two phases. Recently, new methods based on metadynamics for extracting this energetic barrier have been developed [1,2,3,4]. These methods have been used to study the transition between liquid and solid Lennard Jones. We have thus demonstrated that classical nucleation theory correctly predicts the shape of the crystalline nucleus [4]. The aim of this project is to apply these ideas to study nucleation in a more realistic system.

Project

Nucleation is an important component of many technological and natural processes, including metal casting, the assembly of microtubules in cells and the formation of water droplets and ice crystals in the atmosphere. In spite of its generic importance, however, our understanding of nucleation kinetics is somewhat unsatisfactory, even for the simplest cases of homogenous nucleation. The main reason for this is that the experimental determination of nucleation rates is challenging. Many of the available analytic techniques can only be used to perform ex situ analysis, which cannot fully capture the dynamic processes that take place during nucleation.

A considerable number of studies in the last two decades have used computer simulation to study homogeneous nucleation. Much of this computational effort has concentrated on investigating how free energy changes with cluster size and in using the curve obtained to determine the height and position of the nucleation barrier. Our recent work [1], demonstrated that there are problems in this approach because it is typically only possible to investigate small nuclei in simulations. One must therefore extrapolate any curve obtained from simulation to find the height of the nucleation barrier. This extrapolation is problematic because the parameters for these nucleation curves are highly correlated. On thus sees large discrepancies between the barrier heights obtained from independent simulations of the same system because of statistical noise.

A better approach for studying nucleation is thus to extract the surface excess free energy for the interface between the solid and the liquid from simulations of a planar interface between the two phases [2,3]. If multiple simulations of different possible interfaces are performed a Wulf construction can then be used to extract the shape of the nucleus [4]. We have shown that statistical noise has a much smaller effect on the parameters extracted from such simulations. Furthermore, we have also shown that the shapes of the nuclei that are observed in such simulations are fully consistent with the shapes that would be predicted based on classical nucleation theory [4].

Thus far we have only applied these methods on a model system of Lennard Jones particles. The aim of this project would be to use these models to study nucleation in models of more realistic materials such as metals.

Bibliography

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