Effect of Interaction Potential on Crystal Nucleation Kinetics for Lennard-Jones-like Particles

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Abstract: Crystal nucleation phenomena have practical relevance to pharmaceutical production, climate science, and oil transportation. Molecular simulations are a vital tool for uncovering the microscopic mechanism of this process, but requires interaction models that accurately capture the behavior of the system under study. The structural, thermodynamic, and kinetic properties of materials and fluids are the result of the interaction potentials between atoms. Molecular dynamics allows us to probe the effects of varying potential parameters, which is not possible with experiments. Understanding these effects is key to developing force fields that accurately model these systems of interest. Our goal is to examine the effects on nucleation kinetics of softening the repulsion and attraction of Lennard-Jones-like potentials by modifying the repulsive exponent, *n*. Crystal nucleation rates are calculated using forward flux sampling for two potentials, where $n = 12$ and $n = 7$, and compared within the framework of classical nucleation theory (CNT). We find that the nucleation rates are nearly identical at the same undercooling. This is explained by calculating the thermodynamic properties of the liquid and crystal that are used in CNT, including the bulk solid-liquid free energy difference, density, and interfacial free energy. Though the bulk free energy differences and densities differ between the two potentials, the cancellation of these factors causes the estimated free energy barriers and rates of nucleation to be equal. The net effect of decreasing the repulsive exponent therefore appears to be an increase in density. A comparison of nucleation mechanisms is also made for the two potentials.