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Free energy of cluster formation and a new scaling relation for the nucleation rate

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Recently we performed molecular dynamics (MD) simulations of homogeneous nucleation from vapor for systems of (1-8) $\times$ 10^9 Lennard-Jones atoms [Diemand etal. J. Chem. Phys. 139, 074309 (2013)]. The very large MD simulations allow us to determine the formation free energy of clusters accurately over a wide range of cluster sizes, for the first time. This is now possible because such large simulations allow for very precise measurements of the cluster size distribution in the steady state nucleation regime. The peaks of the free energy curves give critical cluster sizes, which agree well with independent estimates based on the nucleation theorem. Using these results, we derive an analytical formula and a new scaling relation for nucleation rates: ln J' / η is scaled by ln S / η, where the supersaturation ratio is S, η is the dimensionless surface energy, and J' is a dimensionless nucleation rate. This relation can be derived using the free energy of cluster formation at equilibrium which corresponds to the surface energy required to form the vapor-liquid interface. At low temperatures (below the triple point), we find that the surface energy divided by that of the classical nucleation theory does not depend on temperature, which leads to the scaling relation and implies a constant, positive Tolman length equal to half of the mean inter-particle separation in the liquid phase.

Keywords: nucleation, molecular dynamics simulation, nucleation rate, scaling, free energy of cluster formation