

The Role of Curvature in Vapor-Liquid Nucleation

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Nucleation is of fundamental interest in science and technology, and has been widely investigated by experiments, theory, and simulations.^[1] Classical nucleation theory is still the most widely used theoretical tool to predict nucleation rates, however, its predictions are often orders of magnitude away from simulations and experiments.^[2] One of the crudest assumptions of classical nucleation theory is the neglect of curvature dependence in the surface tension and the nonequilibrium transfer properties. In our work, we discuss the role of curvature in vapor-liquid nucleation. We show that the surface tension of a small bubble/droplet deviates significantly from that of the planar interface,^[3] and that the heat and mass transfer coefficients of the bubble/droplet are highly curvature dependent.^[4] By expanding the properties in the curvature, we derive a consistent description of the steady-state non-isothermal nucleation rate using nonequilibrium thermodynamics.^[5] The coefficients in the curvature expansions are obtained by combining density functional theory with nonequilibrium molecular dynamics. Results will be presented first for the Lennard-Jones fluid as a benchmark example, and then for water. By comparing the results to classical nucleation theory and experiments, new and important insight will be given on how curvature affects nucleation rates.

References

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