Surface energy of bubbles evaluated by large-scale molecular dynamics simulations of homogeneous bubble nucleation

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Bubble nucleation in liquid is a liquid-to-vapor transition phenomenon, and plays an important role in many areas of science and technology. Recetnly we presented large-scale molecular dynamics simulations of homogeneous bubble (liquid-to-vapor) nucleation with LAMMPS [1]. The simulations contain half a billion Lennard-Jones atoms and cover up to 56 million time steps. The unprecedented size of the simulated volumes allows us to resolve the nucleation and growth of many bubbles per run in simple direct micro-canonical simulations while the ambient pressure and temperature remain almost constant. It is widely believed that classical nucleation theory (CNT) generally underestimates bubble nucleation rates by very large factors. We showed that the measured rates agree well with the CNT within two orders of magnitude in the superheated boiling regime (positive ambient pressure), while the CNT prediction underestimates the nucleation rates significantly in the cavitation regime (lower temperatures and negative pressures) [1,2]. We revisited classical nucleation theory (CNT) for the homogeneous bubble nucleation rate and improve the classical formula using a correct prefactor in the nucleation rate [3]. Most of the previous theoretical studies have used the constant prefactor determined by the bubble growth due to the evaporation process from the bubble surface. However, the growth of bubbles is also regulated by the thermal conduction, the viscosity, and the inertia of liquid motion. These effects can decrease the prefactor significantly, especially when the liquid pressure is much smaller than the equilibrium one. The deviation in the nucleation rate between the improved formula and the CNT can be as large as several orders of magnitude. Our improved, accurate prefactor and recent advances in molecular dynamics simulations and laboratory experiments for argon bubble nucleation enable us to precisely constrain the free energy barrier for bubble nucleation. Tanaka et al. [4] showed that the simple expression including the Tolman correction to the surface tension with a small Tolman length leads to good agreements with the recent MD simulations. But it is also important to compare with other models. The model of surface tension based on the Helfrich expansion is interesting to consider [5], although it includes some additional parameters. Recently, Wilhelmsen et al. [5] obtained values for the parameters in the Helfrich expansion for various temperatures and cutoff radii in the case of a Lennard-Jones liquid using density functional theory (DFT) calculations. The validity of the Helfrich expansion for nanosized bubbles has not yet been fully clarified [5]. We tested the Helfrich expansion model for various bubble sizes and temperatures, using results for surface tension from our MD simulations, and found an agreement stronger than that the pure Tolman description offers.

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