Crystallization of Lennard-Jones nanodroplets: from near melting to deeply supercooled

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ABSTRACT: We carry out molecular dynamics and Monte Carlo simulations to characterize nucleation in liquid clusters of 600 Lennard-Jones particles over a broad range of temperatures. We find that Classical Nucleation Theory (CNT) predicts the rate quite well, even when employing simple modelling of crystallite shape, chemical potential, surface tension and particle attachment rate, down to the temperature where the droplet loses metastability and crystallization proceeds through growth-limited nucleation. Below this crossover temperature, the nucleation rate is controlled by particle attachment rates and is still described by CNT, but with thermodynamic quantities that appear to be "frozen in" to values at the crossover temperature. We use the formalism of mean first-passage times to determine the rate and to reconstruct free energy profiles, which agree at higher temperatures with those obtained through Monte Carlo. Discrepancy arises when twinned structures with five-fold symmetry provide a competing free energy pathway out of the region of critically-sized embryos. We find that crystallization begins with hcp-fcc stacked precritical nuclei and differentiation to various end structures occurs when these embryos are critical or post-critical in size. We find that the bulk melting temperature controls the rate, even thought the solid-liquid coexistence temperature for the droplet is significantly lower. Additionally, we find that the anisotropy of critical embryos grows at low temperature, but largely follows the same size dependence of anisotropy for embryos taken from a single temperature near coexistence.

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