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Large scale MD simulations of nucleation from vapor

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Nucleation theory from the vapor phase plays an important role in origin of cosmic dust particles. The widely used classical nucleation theory gives a macroscopic description of homogeneous nucleation. However it predicts nucleation rates which disagree by several orders of magnitude from experimental measurements and also from numerical results from molecular dynamics (MD) and Monte Carlo simulations[1-6].

We present first results of large scale molecular dynamics simulations of homogeneous nucleation. We performed MD simulations of nucleation from vapor for systems of 10^9 Lennard-Jones (L-J) type molecules in order to test nucleation theories. The MD simulations were done with the LAMMPS code (:Large-scale Atomic/Molecular Massively Parallel Simulator) on up to 32'768 cpus on the HERMIT and SuperMUC supercomputers. We obtain a PRACE award (Partnership for Acvanced Computing in Europe) of 36 million cph hours on Hermit at HLRS. Simulations were done for wide ranges of the initial supersaturation ratio and temperature (T =0.3-1.0 e/k) where e and k are the binding energy of the L-J potential and the Boltzmann constant, respectively.

Here we present the results of the nucleation rate form these simulations and compare them to two widely used nucleation models, the modified classical nucleation theory (MCNT) and the semiphenomenological (SP) model [1]. At low temperatures MCNT significantly underestimates the nucleation rates (by up to 10^9) and at T=1.0 e/k it overestimates the nucleation rates by up to 10^5. The SP model matches the nucleation rates and the cluster size distributions found in previous MD simulations at higher supersaturations quite well [4,6]. However at the lower supersaturations probed here the SP model predictions differ from the MD results by large factors (up to 10^{3-5}). The reason of differences would come from that the correction by the second virial coefficient is not enough because the critical clusters are larger for lower supersaturations. We will also present MD results on cluster size distributions, free energy functions, and sticking probabilities.

1. A. Dillmann and G. E. A. Meier, J. Chem.Phys.94, 3872 (1991).
2. D. W. Oxtoby, J. Phys: Condens. Matter, 4, 7627 (1992).
3. K. Yasuoka and M. Matsumoto, J. Chem. Phys. 109, 8451 (1998).
4. K. K. Tanaka, H. Tanaka, K. Kawamura, and K. Nakazawa, J. Chem. Phys. 122, 184514 (2005).
5. J. Merikanto, E. Zapadinsky, A. Lauri, I. Napari, and H. Vehkamaki, J. Chem. Phys. 12, 104303 (2007).
6. K. K. Tanaka, H. Tanaka, K. Kawamura, and T. Yamamoto, J. Chem. Phys. 134, 204313, (2011).

Keywords: nucleation, crystallization, MD simulation, billion molecules, LAMMPS

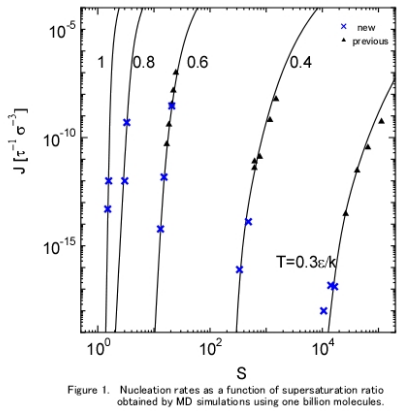

Figure 1. Nucleation rates as a function of supersaturation ratio obtained by MD simulations using one billion molecules.