

# InSiDE

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## Innovatives Supercomputing in Deutschland



# Innovative HPC Methods and Application to Highly Scalable Molecular Simulation

Molecular simulation allows entering regions beyond the domain of classical continuum approaches. For various application fields, like process engineering, material sciences, bio- and nanotechnology, atomistic scale simulations based on Molecular Dynamics (MD) and Monte-Carlo (MC) are regarded as a key to substantial further progress. Despite this well-known potential of molecular methods for industrial research and development, there is still limited usage due to the extreme computing time needed. On the other hand, computer hardware is continuously more powerful, still following Moore's law. In contrast to the frequency increase battle of the past, the actual processor development focuses on parallelism and manycore architectures are supposed to deliver a power-efficient performance gain.

Running a simulation on a large number of homogeneous (e.g. x86) or even heterogeneous (e.g. CellBE) cores, which are connected in a hierarchical structure over a CPU, a node or the whole system, requires an adaptation of the code to get a maximized performance. Parallelism also comes to fore on the core itself, supporting SIMD instructions or vectorization. This also has to be taken into account during code development. Massively parallel systems with a large number of processing elements also boost the importance of load balancing issues. Furthermore, software engineering aspects are important: Efficient code still needs to be readable and expandable to be open for future needs.

## Project Goals

The Lehrstuhl für Thermodynamik (LTD) of Technische Universität Kaiserslautern, the Lehrstuhl für Thermodynamik und Energietechnik (ThEt) of Universität Paderborn, the Lehrstuhl für Informatik mit Schwerpunkt Wissenschaftliches Rechnen (SCCS) of Technische Universität München, the High Performance Computing Center Stuttgart (HLRS) and the Fraunhofer Institut für Techno- und Wirtschaftsmathematik (ITWM) joined to meet the challenge of creating and improving highly scalable numerical molecular simulation software. The software packages will be optimized for massively parallel manycore systems featuring a hybrid OpenMP+MPI parallelization, but also addressing new architectures, like the Cell Broadband Engine. Dynamic load balancing algorithms for parallelization based on spatial decomposition are developed to deal with the very heterogeneous particle distributions which are typical for the application domain. These algorithms use e.g. kD-trees, space filling curves as well as graph based approaches to optimize the domain decomposition for equally distributed load, but also try to minimize communication. Concurrently software development based on Partitioned Global Address Space (PGAS) languages will re-implement already existing functionality and provide novel tools, which are a foundation for future work.

## Simulation Software

In the center of the project are the two simulation packages called ms2 and ls1, which are the result of preliminary

work of a subset of the IMEMO project partners, being enhanced during the project.

ms2 is a software package to get new insights into a large range of thermo-physical properties of arbitrary rigid molecules mixtures. The program, initiated in 2001 at the Universität Stuttgart, was already successfully used for three Industrial Fluid Property Simulation Challenges ([www.ifpsc.org](http://www.ifpsc.org)), allowing for a title of champions in 2007 [1]. ms2 implements MD as well as MC simulation with several classical ensembles to determine static, entropic and transport properties. The reasonably object oriented Fortran90 code is parallelized for distributed memory architectures using MPI and with all relevant loops being vectorized, it is also suitable for vector machines.

ls1 is a MD simulation tool for time dependent nano-scale processes for arbitrary rigid molecule mixtures with a focus on large systems which aims to close the gap between the nanoscopic and microscopic scale. The C++ code was initiated in 2005 at the Universität Stuttgart to study nucleation and flow phenomena and targets massively-parallel distributed memory architectures using MPI.

## Applications

Some exemplary applications of these software packages for an atomistic numerical simulation will give a short overview about the direction of the ongoing work. Taking a set of molecular models for 78 real fluids and their binary mixtures as a basis [2,3], the fluid phase coexistence was studied for ternary mixtures using the ms2 program. The phase separation properties for the ternary system consisting of methane, ethane, and carbon dioxide are shown in

Figure 1 for a temperature of 230 K and a pressure of 4.65 MPa, which is near the triple point temperature of carbon dioxide (217 K) and above the critical temperature of methane (191 K). Ethane and carbon dioxide were modeled using the quadrupolar two-center Lennard-Jones potential, which predicts the azeotropy for this subsystem [2], and methane was represented by the Lennard-Jones (LJ) potential. The agreement with experimental data [5] is excellent. A systematic comparison with respect to all 33 ternary mixtures for which experimental data are available confirms that this is generally the case [4].

Vapor to liquid nucleation for systems containing up to ten million particles was studied using the ls1 program. In a closed system, the nucleation process is instationary, since the vapor is continuously depleted by the aggregation of monomers to small droplets. This leads to a high uncertainty for the nucleation rate obtained from canonical ensemble MD simulation [6,7]. The precision can be significantly increased by a new molecular simulation method for

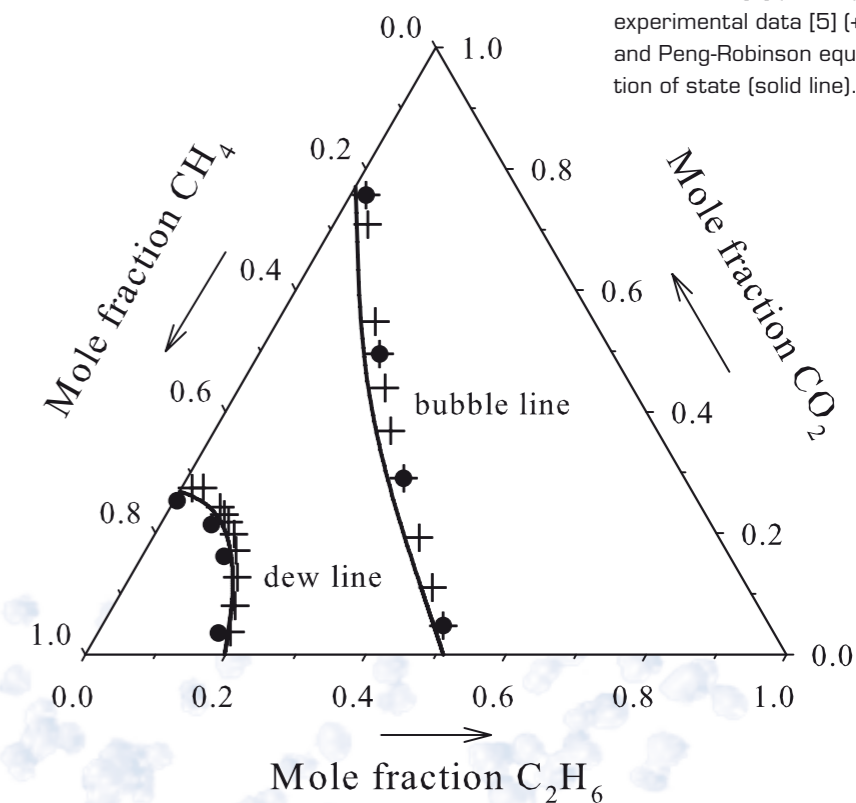


Figure 1: Ternary vapor-liquid equilibrium phase diagram for the mixture  $\text{CH}_4 + \text{CO}_2 + \text{C}_2\text{H}_6$  at 230 K and 4.65 MPa: simulation data [4] (bullets), experimental data [5] (+), and Peng-Robinson equation of state (solid line).

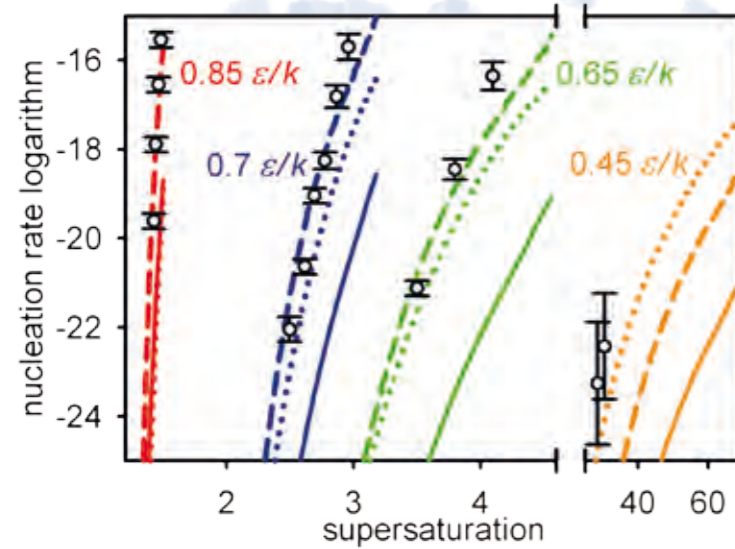
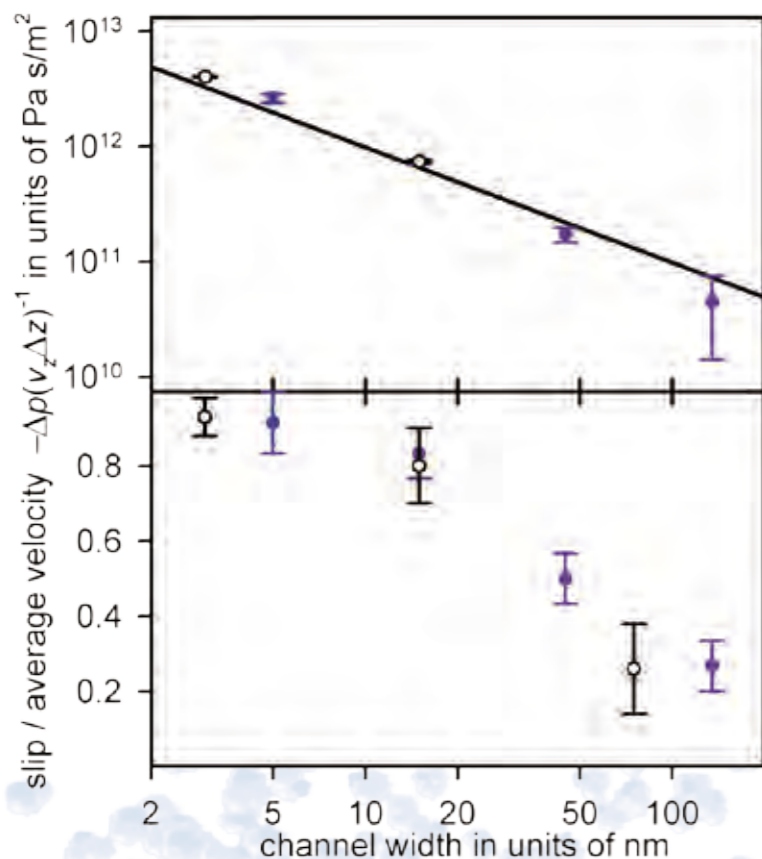


Figure 2: Nucleation rate over supersaturation for the t. s. LJ fluid at  $T = 0.45, 0.65, 0.7,$  and  $0.85 \epsilon/k_B$ , wherein  $\epsilon$  is the energy parameter of the t. s. LJ model, according to CNT (solid line), the SPC modification [7] (dashed line), as well as the HCSF (dotted line) in comparison with present simulation results (circles)

Figure 3: Pressure drop  $\delta p$  in terms of the flow velocity  $v_z$  and the channel length  $\delta z$  (top) as well as slip velocity in terms of  $v_z$  (bottom), for Poiseuille flow of liquid methane at  $T = 166.3$  K as well as  $v_z = 10$  (circles) and  $30$  m/s (bullets), as a function of the channel diameter, compared with Darcy's law (solid line)



sampling both nucleation kinetics and steady-state properties of the nucleating supersaturated vapor. The idea behind this approach is to simulate the production of droplets up to a given size for a specified metastable state. This is achieved by extending MD simulation in the grand canonical ensemble with an "intelligent being" that continuously removes all large droplets: McDonald's demon [8]. A series of simulations for the truncated and shifted LJ (t. s. LJ) fluid, an accurate model for noble gases and methane, shows that the classical nucleation theory (CNT) underpredicts the nucleation rate by two orders of magnitude [Fig. 2]. The Hale critical scaling formalism (HCSF) [9], which was introduced to explain experimental data on low-temperature nucleation, leads to better agreement for the t. s. LJ fluid near and below the triple point temperature ( $T_3 = 0.65 \epsilon / k_B$ ). This proves for low temperatures that the validity of HCSF extends up to extremely high nucleation rates which are experimentally inaccessible but can be studied by molecular simulation.

Due to their anisotropy, graphite nanochannels and carbon nanotubes are of particular interest. On the nanometer length scale, continuum approaches like the Navier-Stokes equation break down [10]. It is known for Poiseuille flow with a channel width below 2 nm that the slip velocity, which is often neglected, reaches almost the same magnitude as the flow velocity [11].

The Is1 program was applied to the Poiseuille flow of methane, represented by the t. s. LJ fluid, through a graphite channel, where a rescaled version of the Tersoff [12] potential was used for the carbon atoms. The interaction between fluid molecules and wall atoms was also modeled by the LJ potential [13].

As figure 3 shows, a transition from high to low slip velocities occurs at a channel diameter of about 40 nm. Darcy's law, stating that the pressure drop is inversely proportional to the cross-sectional area of the channel, is valid down to the molecular length scale.

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## Partners

High Performance Computing Center Stuttgart (HLRS) | Lehrstuhl für Thermodynamik und Energietechnik (ThEt), Universität Paderborn | Lehrstuhl für Thermodynamik (LTD), Technische Universität Kaiserslautern | Lehrstuhl für Informatik mit Schwerpunkt Wissenschaftliches Rechnen (SCCS), Technische Universität München | Institut für Techno- und Wirtschaftsmathematik (ITWM), Fraunhofer Gesellschaft, Kaiserslautern

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BASF SE | Bayer Technology Services GmbH (BTS) | Evonik Industries AG | IBM Deutschland Entwicklung GmbH

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