Nonequilibrium Molecular Dynamics Simulation
of the Thermocapillary Effect

Molekulardynamische Nichtgleichgewichtssimulation
des thermokapillaren Effekts

Figures

Vom Fachbereich Maschinenbau an der Technischen Universität Darmstadt
zur Erlangung des akademischen Grades eines Doktor-Ingenieurs (Dr.-Ing.)
genehmigte DISSERTATION

vorgelegt von
Dipl.-Ing. Holger Andreas Maier
aus Alzenau

Darmstadt 2011
D17
15 Figures

Figure 1  Liquid-liquid phase diagram of a binary water / 1-Butanol mixture at ambient pressure. 15-5
Figure 2  Thermocapillary convection in a physical experiment and its reproduction in a numerical experiment. 15-5
Figure 3  Scheme of our procedure to study the thermocapillary effect in a "numerical experiment" instead of the "physical experiment". 15-6
Figure 4  Fundamental approach to study phenomena on the microscopic level in the classical limit. 15-6
Figure 5  Hexagonal convection pattern of the Marangoni-Bénard instability. 15-7
Figure 6  Typical experimental setup for the study of thermal creep and molecular gas flows. 15-7
Figure 7  Local surface and volume forces on a macroscopically small subvolume $\Delta V$. 15-7
Figure 8  Stress tensor elements for three mutually perpendicular and macroscopically small cut surfaces $\delta S_j$. 15-7
Figure 9  Two different microscopic definitions of the kinetic contribution to the stress vector associated with the cut surface $\delta S_j$. 15-7
Figure 10  Diffuse and specular reflection at a gas adsorption layer. 15-8
Figure 11  Sketch of a typical BD NEMD simulation system with indication of the perturbed "boundary regions". 15-8
Figure 12  Setup of the simulation system with the expected convection roll cells driven by the thermocapillary effect. 15-9
Figure 13  Basic sketch of the equilibrium interfacial systems simulated. 15-10
Figure 14  Basic sketch of the nonequilibrium interfacial systems simulated. 15-10
Figure 15  Dimensionless $T^*\rho^a$-diagram of the saturation curve of pure one-centre LJ particle substances. 15-10
Figure 16  Typical hyperbolic tangent partial density profiles (top) at a liquid-liquid interface in a mixture of the substances ArA and ArB. 15-11
Figure 17  Local observables obtained for different spatial resolutions in separate $NVT$ simulations of the first heterophasic equilibrium interfacial system. 15-13
Figure 18  Comparison of the local observables obtained by using slabs in the two first nonequilibrium one-phase systems. 15-15
Figure 19  Locations and orientations of selected confined cut-surfaces used in the determination of the local stresses. 15-15
Figure 20  Sketch of an equilibrium interfacial system together with the subvolumes for which the local observables are computed. 15-16
Figure 21  Sketch of a nonequilibrium interfacial system together with the subvolumes for which the local observables are computed. 15-16
Figure 22  Production simulations of the different systems in chronological order. 15-19
Figure 23  Comparison of selected local observables in different production simulations of the first heterophasic equilibrium interfacial system. 15-23
Figure 24  Sequence of the preparational and production simulations of the first systems.................... 15-24
Figure 25  Selected snapshots of the demixing in the equilibration simulation e-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120 (second preparation route)................................................................. 15-26
Figure 26  Local densities determined over three 10ns long successive segments of the equilibration simulation e-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120 (second preparation route) .... 15-27
Figure 27  Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140................................................... 15-28
Figure 28  Comparison of selected local observables in separate production simulations of the first heterophase nonequilibrium interfacial system.................................................................... 15-32
Figure 29  Preparation of an initial phase point from cut-outs of phase points in Buhns NPT simulations............................................................................................................................ 15-33
Figure 30  Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (first preparation route) .. 15-34
Figure 31  Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (second preparation route)...................................................... 15-35
Figure 32  Selected snapshots of the demixing in the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (third preparation route)......................................................... 15-37
Figure 33  Local observables determined over three 10ns long successive segments of the steadying simulation N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (third preparation route) 15-38
Figure 34  Detailed distributions of the local observables in the first heterophase nonequilibrium interfacial system and its corresponding ones ................................................................. 15-49
Figure 35  Spatially averaged distributions of the local observables in the first heterophase nonequilibrium interfacial system and its corresponding ones ............................................. 15-63
Figure 36  Excerpt of the instantaneous heat fluxes coupled into or out of the hot thermostated region and the numbers of the particles residing in it in N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140 ........................................................................ 15-63
Figure 37  Consistency of the local observables in the first heterophase nonequilibrium interfacial system ......................................................................................................................... 15-64
Figure 38  Comparison of the summarised local observables in the corresponding first heterophase interfacial systems ........................................................................................................ 15-66
Figure 39  Comparison of the summarised local observables in several heterophase nonequilibrium interfacial systems with different cut-off radii ......................................................... 15-72
Figure 40  Comparison of the summarised local observables in several heterophase nonequilibrium interfacial systems with different ArB particle masses.................................................. 15-78
Figure 41  Comparison of the summarised local observables in several heterophase nonequilibrium interfacial systems with strict and loose thermostats .................................................. 15-82
Figure 42  Comparison of the summarised local observables in the first homo- and heterophase nonequilibrium interfacial systems .................................................................................. 15-87
Figure 43  Comparison of the mutual solubilities in the Ar1Ar2 and in the Ar5Ar5 mixture as function of the temperature .......................................................... 15-88
Figure 44  Comparison of the thermocapillary convection in Ar5Ar5 systems with different z-dimensions ................................................................. 15-91
Figure 45  Comparison of the thermocapillary convection in Ar5Ar5 systems with different y-dimensions .................................................................................................................. 15-93
Figure 46 Comparison of the thermocapillary convection in Ar$5$Ar$5$ systems with different y- and z-dimensions ................................................................. 15-94

Figure 47 Comparison of the local com velocities in Ar$5$Ar$5$ nonequilibrium interfacial systems that differ only in the temperature difference between the thermostated regions .................. 15-95

Figure 48 Section of the Ar$5$Ar$5$ pxT diagram at $x_{Ar} = 0.5$ with indication of the global pressures and temperatures in selected systems ...................................................................................... 15-95

Figure 49 Comparison of the thermocapillary convection in several Ar$5$Ar$5$ systems that differ in the global temperatures and pressures ........................................................................................................... 15-97

Figure 50 Comparison of the thermocapillary convection in several Ar$5$Ar$5$ systems that differ only in the values of the mixing parameter $\xi$ ............................................................................................................. 15-98

Figure 51 Cut of the Ar$5$Ar$5$ pxT diagram at $x_{Ar} = 0.5$ with indication of the global pressures and temperatures in the systems with different mixing parameter values $\xi$ ............................................................................................................. 15-98

Figure 52 Comparison of the thermocapillary convection in Ar$5$Ar$5$ systems with different particle masses, $m_{ArA}$ and $m_{ArB}$ .................................................................................................................................................. 15-99

Figure 53 Cut of the Ar$5$Ar$5$ pxT diagram at $x_{Ar} = 0.5$ with indication of the global pressures and temperatures in the systems with different particle masses, $m_{ArA}$ and $m_{ArB}$ .................................................................................................................................................. 15-99

Figure 54 Detailed analysis of the density distribution in the homophasic nonequilibrium interfacial systems .......................................................................................................................................................... 15-101

Figure 55 Equilibrium density distribution to expect in a homophasic nonequilibrium interfacial system; ........................................................... ................................................................. 15-102

Figure 56 Example of the density distribution to expect in a nonequilibrium interfacial system based on its related equilibrium interfacial systems; ............................................................................................................. 15-104

Figure 57 Phase diagram indicating the global states in several differently composed but otherwise comparable nonequilibrium one-phase systems ................................................................. 15-104

Figure 58 Concentration dependence of the different density y-gradients and of the Soret coefficients in otherwise similar nonequilibrium one-phase systems ............................................................................................................. 15-104

Figure 59 Particle specific com velocities in selected nonequilibrium one-phase and interfacial systems .................................................................................................................................................. 15-108

Figure 60 Relation between the central particle specific com y-velocities, densities and density y-gradients at the centers between the thermostated regions in a homophasic nonequilibrium interfacial system .......................................................................................................................................................... 15-110

Figure 61 Distributions of selected stress tensor elements using the first homophasic nonequilibrium interfacial system as an example ............................................................................................................. 15-111

Figure 62 Comparison of the local volume y-forces obtained from the normal stresses, $S_{yz}$, and from the shear stresses, $S_{yz}$, in the first homophasic nonequilibrium interfacial system .................. 15-112

Figure 63 Shear stresses, $S_{yz}$, as function of $x$ in the nonequilibrium interfacial system, N-Ar$5$Ar$5$-0.6-1.0-13384-14096-4.74x8.00x37.6-100-140 .......................................................................................................................................... 15-112

Figure 64 Local densities in the unary nonequilibrium liquid-vapour system ............................................................................................................. 15-118

Figure 65 Comparison of the local observables in several unary nonequilibrium liquid-vapour systems with different average system temperatures ............................................................................................................. 15-121

Figure 66 Purely repulsive external force field of the type "ff1" in the first solid-liquid interfacial system ............................................................................................................................................................................. 15-121

Figure 67 Local observables in the first solid-liquid interfacial system with a purely repulsive force field of the type "ff1" ............................................................................................................................................................................. 15-127
Figure 68  Intensity of the convection as function of the slope in the first solid-liquid system with an external force field of the type "ff1" ........................................................................................................ 15-128

Figure 69  Repulsive external force field of the type "ff2" in the unary nonequilibrium interfacial reference system .......................................................................................................................... 15-128

Figure 70  Local observables in the unary nonequilibrium interfacial system with a repulsive force field of the type "ff2", N-Ar1-1.0-6870-4.74x8.00x9.40-100-140-ff2-1.0-3.0-1 ................................................ 15-134

Figure 71  Intensity of the thermocapillary convection in several unary nonequilibrium interfacial systems that differ only in the slope of the external force field "ff2" ................................................................ 15-135

Figure 72  Attractive external force field of the type "ff3" in the unary nonequilibrium interfacial reference system, N-Ar5-1.0-6870-4.74x8.00x9.40-100-140-ff3-1.0-3.0-10 ......................................................... 15-135

Figure 73  Local observables in the unary nonequilibrium interfacial system with attractive force field of the type "ff3", N-Ar5-1.0-6870-4.74x8.00x9.40-100-140-ff3 .......................................................... 15-141

Figure 74  Intensity of the thermocapillary convection in several unary nonequilibrium interfacial systems that differ only in the slope of the external force field "ff3" .................................................. 15-142

Figure 75  Attractive-repulsive external force field of the type "ff4" in the unary nonequilibrium interfacial reference system ................................................................................................................. 15-142

Figure 76  Local observables in the unary nonequilibrium interfacial system with an attractive-repulsive force field of the type "ff4", N-Ar1-1.0-6870-4.74x8.00x9.40-100-140-ff4-1.0-3.0-1 ......................................................... 15-148

Figure 77  PT-phase diagram of water .................................................................................................................. 15-149

Figure 78  PVT-phase diagram of water .................................................................................................................. 15-149

Figure 79  PT-phase diagram of the Ar5Ar5 mixture with equimolar composition ............................................. 15-150

Figure 80  Qualitative Pxt-phase diagram of the Ar5Ar5 mixture ................................................................. 15-150

Figure 81  PV-data as obtained in several EMD simulations of an equimolar Ar5Ar5 mixture at different temperatures .................................................................................................................. 15-150

Figure 82  Examples of unusual stable configurations found in the liquid-liquid-vapour region of the Ar5Ar5 mixture .................................................................................................................. 15-151

Figure 83  Example of the concentration dependence of different thermodynamic properties related to the mixing behaviour of the Ar5Ar5 mixture .......................................................... 15-152

Figure 84  Examples of "butterfly" diagrams of the Ar5Ar5 mixture ............................................................. 15-153

Figure 85  Snap shot of a configuration in the two-dimensional nonequilibrium interfacial system............. 15-153

Figure 86  Local observables in the two-dimensional nonequilibrium interfacial system .......................... 15-156
We show the data from experiments and from thermodynamic calculations using the NRTL equation \cite{sorensen1979}.

Figure 1  Liquid-liquid phase diagram of a binary water / 1-Butanol mixture at ambient pressure

(a) Physical experiment: Thermocapillary convection in a binary system of two partially miscible liquids (green and yellow) at different bulk phase temperatures $T_+$ and $T_-$ \cite{oertel2002}; Temperature fluctuations in the interfacial region ignite the thermocapillary convection that selfsustains by dragging new hot liquid from the bulk phases to the interface where the latter gives off the heat to the counterflow in the opposite phase and returns into the hot bulk phase. Under certain system conditions the thermocapillary convection becomes stationary and temperature differences along the interface can be observed between the places where the convection approaches ($T_H$) and leaves ($T_C$). Note, gravity is necessary to keep the interface flat but buoyancy driven convection shall be negligible.

(b) Numerical experiment: reproduction of the physical experiment in MD simulation with simplifications mentioned in the text; The hatching represents the interfacial region. In order to comply with the periodic system boundaries the thermocapillary system is mirrored along the $xy$- and the $xz$-plane.

Figure 2  Thermocapillary convection in a physical experiment and its reproduction in a numerical experiment
Figure 3  Scheme of our procedure to study the thermocapillary effect in a "numerical experiment" instead of the "physical experiment".

Figure 4  Fundamental approach to study phenomena on the microscopic level in the classical limit.
Figure 5  Hexagonal convection pattern of the Marangoni-Bénard instability

Figure 6  Typical experimental setup for the study of thermal creep and molecular gas flows

Figure 7  Local surface and volume forces on a macroscopically small subvolume \( \Delta V \)

Figure 8  Stress tensor elements for three mutually perpendicular and macroscopically small cut surfaces \( \delta S_j \)

a) definition, that considers only the particles crossing the cut surface

b) definition, that considers all particles in the vicinity of the cut surface

Figure 9  Two different microscopic definitions of the kinetic contribution to the stress vector associated with the cut surface \( \delta S_j \)

Both sketches show the same exemplary particle arrangements at two subsequent time steps. Different particles contribute to the stress vector depending on the definition. We highlight them in black compared to the grey ones.
Figure 10  Diffuse and specular reflection at a gas adsorption layer

Figure 11  Sketch of a typical BD NEMD simulation system with indication of the perturbed "boundary regions"
In the "intermediate regions" between them, the non-physical behaviour in the perturbation fades
Figure 12  Setup of the simulation system with the expected convection roll cells driven by the thermocapillary effect.

The grey and white circles represent the particles of the two species, generally called ArA and ArB here. The liquid-liquid interfaces are oriented parallel to the xy-plane. The regions parallel to the yz-plane represent the "thermostated regions". The hot one carries a horizontal and the cold one a diagonal hatching.
Figure 13  Basic sketch of the equilibrium interfacial systems simulated.

Figure 14  Basic sketch of the nonequilibrium interfacial systems simulated.
It indicates additionally the boundary or thermostated regions.

Figure 15  Dimensionless $T^\star \rho^\star$-diagram of the saturation curve of pure one-centre LJ particle substances.
The diagram shows additionally the reduced temperature-density ranges in the mixtures simulated in this work.
Figure 16  Typical hyperbolic tangent partial density profiles (top) at a liquid-liquid interface in a mixture of the substances ArA and ArB
We show additionally the related interatomic particle force and potential of mean force profiles (middle) as well as the related interatomic volume forces (bottom). The ArA rich phase resides on the left side and the ArB rich phase on the right side. The interfacial width related to the ArA partial density profile $\omega_{ArA}$ is indicated by the arrow.
a) Densities

b) Temperatures
c) Com Velocities

Figure 17  Local observables obtained for different spatial resolutions in separate NVT simulations of the first heterophase equilibrium interfacial system. We divide it into different numbers of slab-shaped subvolumes in the analysis. Nondetermined results occur for a subdivision into 576 subvolumes since some of them are not populated by particles at every time step.
1) **Densities**

2) **Temperatures**
Figure 18  Comparison of the local observables obtained by using slabs in the two first nonequilibrium one-phase systems
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140
and N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140

Figure 19  Locations and orientations of selected confined cut-surfaces used in the determination of the local stresses
Figure 20  Sketch of an equilibrium interfacial system together with the subvolumes for which the local observables are computed. The subvolumes lumped together into the ArA or ArB phases are indicated in different shades of grey. The subvolumes considered in the calculation of the P-averages in each phase are additionally labelled with the letter "P".

Figure 21  Sketch of a nonequilibrium interfacial system together with the subvolumes for which the local observables are computed. The subvolumes lumped together into the ArA or ArB phases are indicated in different shades of grey. The subvolumes considered in the calculation of the C, M, or H-averages in each phase are labelled with the letters "C", "M", or "H".
a) EMD simulations of heterophasic interfacial systems

b) EMD simulations of homophasic interfacial systems
### c) NEMD simulations of heterophasic interfacial systems

- **20ns**
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140\(\uparrow\)75

- **5ns**
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-a\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-b\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-c\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-d\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-e\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-f\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-g\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-h\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-i\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-j\(\uparrow\)75
  - N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-k\(\uparrow\)75

- **10ns**
  - N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140\(\uparrow\)75
  - N-Ar1Ar3-0.6-1.0-6797-22-4.74x8.00x9.40-100-140\(\uparrow\)75
  - N-Ar1Ar3-0.6-1.0-20-7089-4.74x8.00x9.40-100-140\(\uparrow\)75

- **10ns**
  - N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140\(\uparrow\)75
  - N-Ar1Ar4-U.1-b-YY\(\uparrow\)75
  - N-Ar1Ar4-0.6-1.0-19-7098-4.74x8.00x9.40-100-140\(\uparrow\)75

---

15-18
d) NEMD simulations of homophasic interfacial and their corresponding one-phase systems

Figure 22 Production simulations of the different systems in chronological order
We give additionally the lengths of the preparational simulations. Bold symbols indicate the altered simulation parameters.
a) Density profiles
b) Temperature profiles
Figure 23  Comparison of selected local observables in different production simulations of the first heterophase equilibrium interfacial system
E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber,
E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-nvt,
and E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40—71.5;
They start from different initial phase points obtained through the first, second or third preparation route.
Figure 24  Sequence of the preparational and production simulations of the first systems
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 and E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120
Figure 25  Selected snapshots of the demixing in the equilibration simulation e-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120 (second preparation route)
**a) Densities**

Figure 26  Local densities determined over three 10ns long successive segments of the equilibration simulation e-Ar1Ar2-0.6-10-3346-3524-4.74x8.00x9.40-120 (second preparation route)

Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.
Figure 27  Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-6795-24-47.4x80.0x94.0-100-140
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.
a) Density profiles
b) Temperature profiles
c) Com velocity components

Figure 28  Comparison of selected local observables in separate production simulations of the first heterophase nonequilibrium interfacial system
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1,
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r2,
and N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r3
They start from different initial phase points obtained through the first, second or third preparation route.
We link the different cut-outs together in $y$-direction to obtain a phase point that resembles one to be found in the first nonequilibrium interfacial system. The figure shows additionally the numbers of particles found in the different cut-outs.

Figure 29  Preparation of an initial phase point from cut-outs of phase points in Buhn's NPT simulations
a) Densities

b) Temperatures

Figure 30  Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-47.4x80.0x94.0-100-140 (first preparation route)
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.
Figure 31  Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-47.4x80.0x94.0-100-140 (second preparation route)
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.

a) Densities

b) Temperatures
Figure 32  Selected snapshots of the demixing in the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (third preparation route)
a) Densities

b) Temperatures

Figure 33  Local observables determined over three 10ns long successive segments of the steadying simulation N-Ar1Ar2-0.6-1.0-3346-3524-47.4x80.0x94.0-100-140 (third preparation route)
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.
a) Densities
b) ArA partial densities
c) ArB partial densities
d) Temperatures
e) Com y-velocities
f) Com $z$-velocities
g) Interatomic volume $y$-forces
h) Interatomic volume \(z\)-forces
i) Interatomic ArA particle $y$-forces

j) Interatomic ArB particle $y$-forces
k) Interatomic ArA particle z-forces

l) Interatomic ArB particle z-forces
Figure 34  Detailed distributions of the local observables in the first heterophasic nonequilibrium interfacial system and its corresponding ones

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-ber,
E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber,
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140,
and N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140
a) Densities
b) Temperatures
c) Com velocities
**d) Volume forces**
e) Particle forces
f) Normal stresses

Figure 35  Spatially averaged distributions of the local observables in the first heterophase nonequilibrium interfacial system and its corresponding ones

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-ber,
E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber,
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140,
and N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140

We plot additionally the fitted hyperbolic tangent density z-profiles in the interfacial systems.

Figure 36  Excerpt of the instantaneous heat fluxes coupled into or out of the hot thermostated region and the numbers of the particles residing in it in N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140

15-63
a) I-averages of the interatomic and kinetic volume $y$-forces as well as their sum as function of $z$

b) I-averages of the interatomic and kinetic particle $y$-forces as well as their sum as function of $z$

c) Particle $z$-forces obtained in the simulation compared to those computed from the partial density profiles by using the BGY equation using the absolute $y$-coordinates of $|y|=0.4\,\text{nm}$ and of $|y|=3.6\,\text{nm}$ as an example

Figure 37 Consistency of the local observables in the first heterophasic nonequilibrium interfacial system N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1
a) Densities

b) Temperatures
c) Volume forces

d) Particle forces

Figure 38  Comparison of the summarised local observables in the corresponding first heterophasic interfacial systems
E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120 and N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140
a) Densities

\[ \Delta \rho(z) = \frac{\rho(z) - \rho_0}{\rho_0} \]

\[ \frac{\Delta \rho(z)}{y} = \frac{\rho(z) - \rho_0}{\rho_0 y} \]

\[ \Delta \rho(y) = \frac{\rho(y) - \rho_0}{\rho_0} \]

\[ \frac{\Delta \rho(y)}{y} = \frac{\rho(y) - \rho_0}{\rho_0 y} \]

\[ \Delta \rho(z) = \frac{\rho(z) - \rho_0}{\rho_0} \]

\[ \frac{\Delta \rho(z)}{y} = \frac{\rho(z) - \rho_0}{\rho_0 y} \]

\[ \Delta \rho(y) = \frac{\rho(y) - \rho_0}{\rho_0} \]

\[ \frac{\Delta \rho(y)}{y} = \frac{\rho(y) - \rho_0}{\rho_0 y} \]
b) Temperatures
c) Com velocities
d) Volume forces

\[ \sigma_{ij}(\text{[Pa/Nm}^2]) \]

\[ z \text{ [nm]} \]

e) Particle forces

\[ \sigma_{ij}(\text{[Pa/Nm}^2]) \]

\[ z \text{ [nm]} \]
f) Normal stresses

Figure 39  Comparison of the summarised local observables in several heterophasic nonequilibrium interfacial systems with different cut-off radii

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1,
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140,
and N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140

We include also their corresponding systems in the comparison.
a) Densities
b) Temperatures
c) Com velocities

MeansLocYZComVelYZ.epsi

MeansLocYComVelZ.epsi

MeansLocZComVelZ.epsi

SigmaLocYComVelZ.epsi

SigmaLocZComVelZ.epsi

15-76
d) Volume forces

e) Particle forces
f) Normal stresses

Figure 40  Comparison of the summarised local observables in several heterophase nonequilibrium interfacial systems with different ArB particle masses
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140, N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140, N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140;
We include also their corresponding systems in the comparison.
a) Densities