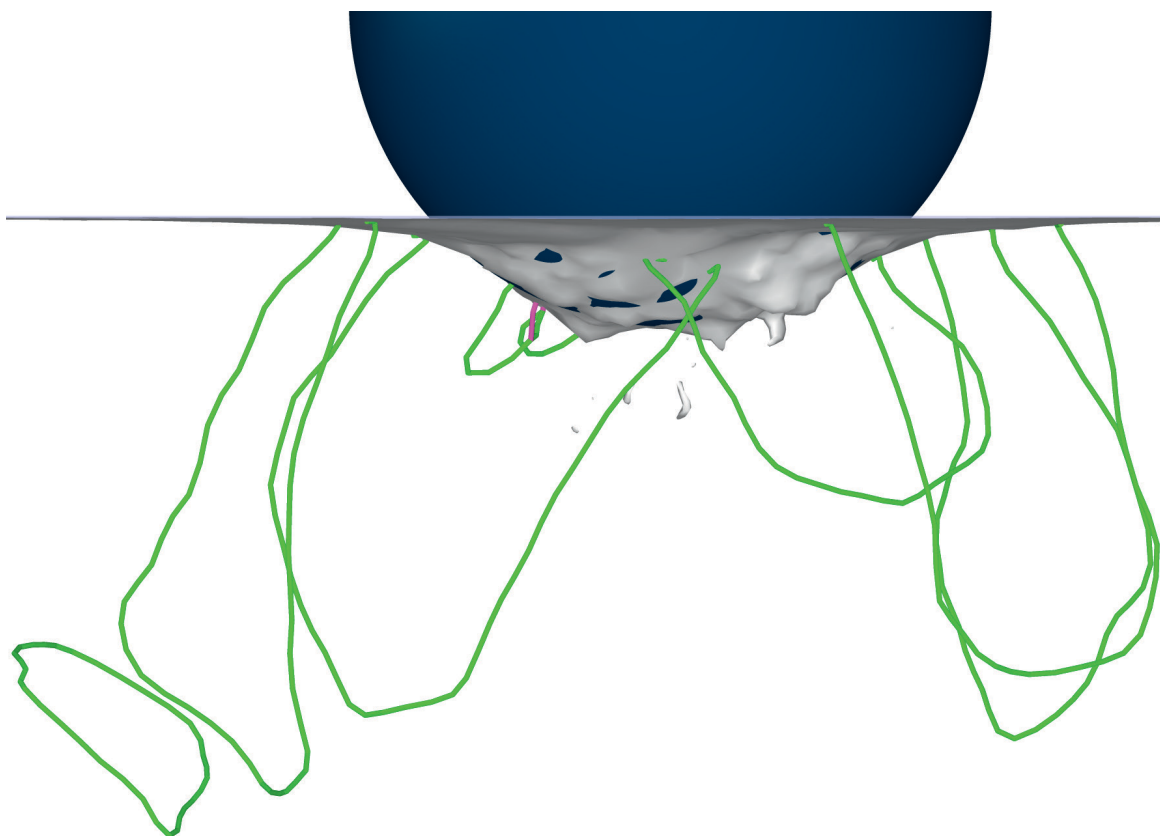




Adaptive-precision interatomic potentials for large-scale atomistic simulations

David Immel

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Abstract

Large-scale atomistic simulations rely on interatomic potentials providing an efficient representation of atomic energies and forces. Modern machine-learning (ML) potentials provide the most precise representation compared to electronic structure calculations while traditional empirical potentials provide a less precise, but computationally much faster representation and, thus, allow simulations of larger systems. In the present work, a novel method is developed to combine a traditional and a ML potential to a multi-resolution description, leading to an adaptive-precision interatomic potential (APIP) with an optimum of performance and precision in large complex atomistic systems. This new method is, in contrast to previous methods, appropriate for microcanonical ensembles as both momentum and energy are conserved by the energy-based coupling approach. The required precision is determined per atom by a local structure analysis and updated automatically during simulation. The approach is implemented as APIP package for the molecular-dynamics simulator LAMMPS and includes a per-atom computational load estimation so that a load-balancer can prevent computational-time problems arising due to the atom dependent force-calculation times, which makes APIP suitable for large-scale atomistic simulations.

The approach is demonstrated in nanoindentation simulations for both the prototypical face-centred cubic metal copper and the body-centred cubic metal tungsten: a computationally efficient embedded atom method (EAM) potential is combined with a precise but computationally less efficient ML potential based on the atomic cluster expansion (ACE) into an adaptive-precision interatomic potential tailored for the nanoindentation. The numerically more expensive ACE potential is employed selectively only in regions of the computational cell where high precision is required. The comparison with pure EAM and pure ACE simulations shows that for copper, all potentials yield similar dislocation morphologies under the indenter with only small quantitative differences. In contrast, markedly different plasticity mechanisms are observed for tungsten in simulations performed with the central-force EAM potential compared to results obtained using the many-body ACE potential, that is able to describe accurately angular characteristics of bonding. All ACE-specific mechanisms are reproduced in the APIP nanoindentation simulations, however, with a significant speedup of 20-30 times compared to the pure ACE simulations. Hence, adaptive-precision interatomic potentials overcome the performance gap between the precise ACE and the fast EAM potential by combining the advantages of both potentials.

Finally, the novel energy and momentum conserving and dynamically self-adapting coupling approach is further improved by local averaging to a conservative adaptive-precision potential, which, by design, guarantees both energy and momentum conservation and is also included in the APIP LAMMPS package. As the force precision improves with the averaging radius, force-coupling approaches, which usually cannot define a potential energy, are not required any more in order to obtain accurate forces on specific atoms. Hence, conservative adaptive-precision potentials can be used regularly in the future – without the currently existing problems caused by the violation of conservativity. By coupling a fast EAM tungsten potential to a highly accurate ACE tungsten potential, the conservation properties are verified numerically and it is shown that one can achieve – dependent on both the potential and the atomistic system – a speedup of one or two orders of magnitude compared to a pure ACE simulation.

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Nomenclature

μ VT	grand canonical
AA	all-atom
AA	all-atom
ACE	atomic cluster expansion
acf	autocorrelation function
ALL	a loadbalancing library
AP	adaptive precision
APA	adaptive-precision atomic cluster expansion
APIP	adaptive-precision interatomic potential
bcc	body-centred cubic
BDA	bcc defect analysis
CCDF	complementary cumulative distribution function
CG	coarse-grained
CNA	common neighbour analysis
CNP	common neighbourhood parameter
CSP	centro-symmetry parameter
Cu	copper
DFT	density functional theory
DFT	density functional theory
DXA	dislocation extraction algorithm of OVITO
fcc	face-centred cubic
FP	fast potential
IMOMM	integrated molecular orbital molecular mechanics
LAMMPS	Large-scale Atomic/Molecular Massively Parallel Simulator
Lennard-Jones	LJ
LOTF	learn-on-the-fly
MD	molecular dynamics
MD	molecular dynamics
ML	machine learning

MM	molecular mechanics
NPT	isothermal–isobaric
NVE	microcanonical
NVT	canonical
PAP	permuted adaptive partitioning
PP	precise potential
QM	quantum mechanics
QM	quantum mechanics
QM/MM	quantum mechanics/molecular mechanics
RCB	recursive coordinate bisectioning
RMSE	root mean square error
SG	staggered grid
std	standard deviation
tabGAP	tabulated Gaussian approximation potential
k_B	Boltzmann constant
$\bar{\Delta}_i^{\text{B,ACE}}$	derivation of an atom i from a reference configuration calculated with the potential ACE and averaged in the force cutoff of atom i
β	rescaling factor for relative momenta of the local thermostat
$\Delta \mathbf{F}_i^{\text{lt}}$	effective force difference of atom i due to the local thermostat
$\Delta \mathbf{v}$	velocity change
$\Delta_i^{\text{B,ACE}}$	derivation of an atom i from a reference configuration calculated with the potential ACE
ΔE_i	potential energy difference of atom i at the end of a time step between the conservative reference system and the system with updated switching parameters
ΔF	force precision
ΔH_i	total energy difference of atom i at the end of a time step between the conservative reference system and the system with updated switching parameters
Δt	timestep
Δt_E	duration of the NVT equilibration of a surface
Δt_{mom}	duration after which the momentum is set to zero in the equilibration
Δt_{RCB}	interval after which the RCB load balancer is called
ΔT_{resc}	kinetic energy change due to rescaling
Δt_{SG}	interval after which the staggered-grid load balancer is called
ΔT_i	kinetic energy difference of atom i at the end of a time step between the conservative reference system and the system with updated switching parameters
$\Delta \lambda_{\text{min}}$	minimum step size for a switching parameter
Δ^E	positive constant used for the energy-based weighting policy of pacemaker
Δ^F	positive constant used for the energy-based weighting policy of pacemaker

δ_o^{tol}	tolerance of the observable o used in the loss function of atomicrex
Δ_{coeff}	penalty term in a loss function dependent on the current parameters
$\Delta_{\text{high}}^{\text{B,ACE}}$	threshold $\Delta_{\text{high}}^{\text{B}}$ calculated with the potential ACE
$\Delta_{\text{high}}^{\text{B}}$	threshold for Δ_i^{B}
$\Delta_{\text{low}}^{\text{B,ACE}}$	threshold $\Delta_{\text{low}}^{\text{B}}$ calculated with the potential ACE
$\Delta_{\text{low}}^{\text{B}}$	threshold for Δ_i^{B}
Δ_{rad}	penalty term in a loss function dependent on the current parameters
Δ_i^{B}	deviation of an atom i from a reference configuration
δ_p	tolerance of the pressure during equilibration
δ_T	tolerance of the temperature during equilibration
$\Delta_{\text{high},\epsilon}^{\text{B,ACE } 50}$	derivation of an atom i from a reference configuration calculated with the potential ACE, differs only by ϵ from $\Delta_{\text{low},\epsilon}^{\text{B,ACE } 50}$
$\Delta_{\text{low},\epsilon}^{\text{B,ACE } 50}$	derivation of an atom i from a reference configuration calculated with the potential ACE, differs only by ϵ from $\Delta_{\text{high},\epsilon}^{\text{B,ACE } 50}$
δ_{ij}	Kronecker δ
ϵ	small positive number
γ^{L}	damping constant of a Langevin thermostat
γ_p^{NH}	damping parameter of the pressure according to Nose-Hoover
γ_T^{NH}	damping parameter of the temperature according to Nose-Hoover
$\hat{\mathbf{r}}$	unit vector of $\mathbf{r}$
$\hat{r}$	unit vector in the direction of $\mathbf{r}$
κ	relative force weight
κ^{DFT}	relative force weight calculated automatically for DFT reference data in pacemaker
κ^{MD}	relative force weight calculated automatically for MD reference data in pacemaker
$\lambda_i^{\text{B,ACE}}$	switching parameter of atom i calculated using cluster basis functions of the potential ACE
λ_α	switching parameter of molecule α
λ_i^{B}	switching parameter of atom i calculated using cluster basis functions
λ_i^{c}	constant switching parameter used for the calculation of ΔH_i
λ_i^{d}	dynamic switching parameter used for the calculation of ΔH_i
λ_i^{EF}	switching parameter of the first energy- and force-weighting approach
λ_i^{FPP}	switching parameter of molecule i in AdResS
$\lambda_{0,i}$	switching parameter calculated with the time averaged CSP
$\lambda_{\alpha\alpha'}$	switching parameter of a pair of molecules
$\lambda_{\text{avg,msz},i}$	time averaged switching parameter with minimum step size
$\lambda_{\text{avg},i}$	time averaged switching parameter
$\lambda_{\text{min},i}$	switching parameter that includes a spatial switching zone
$\mathcal{F}$	nonlinear embedding function used in the atomic cluster expansion

$\mathcal{L}$	loss function minimised to fit a potential
$\mathcal{L}^{\text{crex}}$	loss function of atomicrex
$\mathcal{L}_{\text{ace}}^{\text{DFT}}$	loss function implemented in pacemaker
$\mathcal{L}_{\text{ace}}^{\text{MD}}$	loss function developed for pacemaker which includes potential energy reference values for individual atoms
$\mathcal{N}_i^{\text{imp}}$	set of nearest neighbours used in the CSP calculation according to LAMMPS
$\mathcal{O}$	set of scalar observables used for the fitting with atomicrex
$\mathcal{S}$	set of structures used for the fitting of a fast potential
$\mathcal{S}^{\text{ACE},f}$	reference dataset containing MD data which is used to fit a fast ACE potential
$\mathcal{S}^{\text{EAM,Cu}}$	set of structures used for the optimisation of the copper EAM potential
∇_i	nabla operator of atom i
Ω_i	set of atoms in the neighbourhood of atom i including atom i
$\Omega_{\lambda,i}$	set of atoms within a distance of $r_{\lambda,\text{hi}}$ to atom i
$\omega_{in^{\ell}l^{\ell}m^{\ell}}^{\text{B},f}$	weight of a derivative, calculated with cluster basis functions of a fast ACE potential
$\omega_{in^{\ell}l^{\ell}m^{\ell}}^{\text{L}}$	weight of the derivative of the switching parameter, calculated with cluster basis functions of a fast ACE potential
$\omega_{in^{\text{p}}l^{\text{p}}m^{\text{p}}}^{\text{A},\text{p}}$	weight of a derivative, calculated with many-body basis functions of a precise ACE potential
ω_{inlm}^{A}	weight of a derivative, calculated with many-body basis functions
ω_{inlm}^{B}	weight of a derivative, calculated with cluster basis functions
ω_{inlm}^{B}	weight of a derivative, calculated in an atomic cluster expansion in the basis $\mathcal{B}$
Φ	pair potential used for an EAM potential
$\phi_{nlm}(\mathbf{r})$	pair basis function of ACE
β	non-spatial constant used in the descriptor calculation
$\mathbf{d}^{(0)}$	reference descriptor corresponding to an atomic configuration that can be computed by the fast model
$\mathbf{d}_i$	vectorial descriptor of atom i whose length is used to calculate a differentiable switching parameter
$\mathbf{F}_{si}^{\text{pred}}$	force on atom i in structure s predicted with the current set of parameters during the minimisation of a loss function
$\mathbf{F}_{si}^{\text{targ}}$	force of atom i in structure s which is used as reference value
$\mathbf{F}_{\text{ind}}(\mathbf{r}_i)$	force between atom i and the indenter
$\mathbf{F}_i$	force on atom i
$\mathbf{F}_i^{(f)}$	force on atom i according to a fast model
$\mathbf{F}_i^{(p)}$	force on atom i according to a precise model
$\mathbf{F}_i^{\text{APA}}$	force on atom k according to an APA potential
$\mathbf{F}_i^{\text{AP}}$	force on atom i according to an adaptive-precision potential
$\mathbf{F}_i^{\text{CAP,ACE}}$	force on atom i according to an ACE potential as precise part of a conservative adaptive precision potential

- $\mathbf{F}_i^{\text{CAP,EAM}}$ force on atom i according to an EAM potential as fast part of a conservative adaptive precision potential
- $\mathbf{F}_i^{\text{CAP}}$ potential energy of atom i described by a conservative adaptive precision potential
- $\mathbf{F}_i^{\text{c}}$ constant force used for the calculation of ΔH_i
- $\mathbf{F}_i^{\text{d}}$ dynamic force used for the calculation of ΔH_i
- $\mathbf{F}_i^{\text{EF}}$ force on atom i according to the first coupling approach
- $\mathbf{F}_i^{\text{FM}}$ force on atom i calculated according to force mixing
- $\mathbf{F}_i^{\text{FPA}}$ force on atom i according to a per-atom force-weighting approach
- $\mathbf{F}_i^{\text{HS}}$ force on atom i according to the hot spot method
- $\mathbf{F}_i^{\text{L}}$ force on atom i from a Langevin thermostat
- $\mathbf{F}_i^{\text{MM}}$ force on atom i according to a molecular mechanics model
- $\mathbf{F}_i^{\text{QM}}$ quantum mechanical force on atom i
- $\mathbf{F}_i^{\text{T,FPA}}$ thermodynamic force on atom i according to a per-atom force-weighting approach
- $\mathbf{F}_i^{(\nabla\lambda)}$ force on atom i caused by the differentiation of the switching parameter of a conservative adaptive-precision potential
- $\mathbf{F}_i^{(\nabla\lambda,\text{CSP})}$ force on atom i caused by the differentiation of the CSP of the switching parameter of a conservative adaptive-precision potential
- $\mathbf{F}_i^{(\nabla\lambda,w)}$ force on atom i caused by the differentiation of w^{d} of the switching parameter of a conservative adaptive-precision potential
- $\mathbf{F}_k^{\text{ACE,A}}$ force calculated with an atomic cluster expansion in the basis $\mathcal{B}$
- $\mathbf{F}_k^{\text{ACE,A}}$ force calculated with an atomic cluster expansion in the basis of many-body basis functions
- $\mathbf{F}_k^{\text{ACE,B}}$ force calculated with an atomic cluster expansion in the basis of cluster basis functions
- $\mathbf{F}_{\alpha i \alpha' j}^{\text{inter,AA}}$ force on atom i of molecule α due to atom j of molecule α' according to the intermolecular all-atom potential of in H-AdResS
- $\mathbf{F}_{\alpha i \alpha'}^{\text{inter,CG}}$ force on atom i of molecule α due to molecule α' according to the intermolecular coarse-grained potential of H-AdResS
- $\mathbf{F}_{\alpha i}^{\text{drift}}$ drift force on atom i of molecule α in H-AdResS
- $\mathbf{F}_{\alpha i}^{\text{H-AdResS}}$ force on atom i of molecule α according to the unmodified H-AdResS Hamiltonian
- $\mathbf{F}_{\alpha i}^{\text{intra}}$ force on atom i of molecule α according to the intramolecular potential of H-AdResS
- $\mathbf{F}_{ij}^{\text{at}}$ force between molecules i and j calculated in an all-atom system
- $\mathbf{f}_{ij}^{\text{CAP}}$ force contribution of the atom pair ij caused by the differentiation of a conservative adaptive-precision potential
- $\mathbf{F}_{ij}^{\text{cg}}$ force between molecules i and j calculated using a coarse-grained model
- $\mathbf{f}_{ij}^{(\nabla\lambda)}$ force contribution of the atom pair ij caused by the differentiation of the switching parameter of a conservative adaptive-precision potential
- $\mathbf{f}_{ij}^{(\nabla\lambda,\text{CSP})}$ force contribution of the atom pair ij caused by the differentiation of the CSP of a conservative adaptive-precision potential
- $\mathbf{f}_{ij}^{(\nabla\lambda,w)}$ force contribution of the atom pair ij caused by the differentiation of w^{d} of a conservative adaptive-precision potential
- $\mathbf{f}_{ik}^{\text{ACE}}$ force contribution of the atom pair ik of an ACE potential

$\mathbf{f}_{ik}^{\text{EAM}}$	force component of EAM
$\mathbf{f}_{ki}^{\lambda}$	force contribution of the atom pair ik of the gradient of the switching function to an adaptive-precision force
$\mathbf{f}_{ki}^{(\text{f})}$	force contribution of the atom pair ik of a fast potential
$\mathbf{f}_{ki}^{(\text{p})}$	force contribution of the atom pair ik of a precise potential
$\mathbf{f}_{ki}^{\text{APA}}$	force component of the potential APA calculated on atom i
$\mathbf{f}_{ki}^{\text{AP}}$	force contribution of the atom pair ik to an adaptive-precision force
$\mathbf{f}_{ki}^{\text{L,f}}$	force component calculated from the gradient of the switching parameter of an APA potential with the fast potential 'f' on atom i
$\mathbf{f}_{ki}^{\mathcal{B},\text{ACE}}$	force component of ACE calculated in a cluster expansion of the basis $\mathcal{B}$ on atom i
$\mathbf{g}_i$	differentiable vectorial atomic quantity of atom i that is locally averaged to calculate a differentiable descriptor $\mathbf{d}_i$
P_{ind}	indentation load
$\mathbf{p}_i$	momentum of atom i
$\mathbf{p}_{\alpha i}$	momentum of atom i of molecule α
$\mathbf{p}_{\text{rel},j}$	momentum of atom j relative to a group of atoms required for the local thermostat
$\mathbf{p}_{j,\text{resc}}$	momentum of atom j after rescaling through the local thermostat
$\mathbf{r}$	distance vector
$\mathbf{r}^{\text{d}}$	distance between two atoms
$\mathbf{r}_{\text{ind}}$	position of the centre of a spherical indenter in a nanoindentation
$\mathbf{r}_{\text{ind},0}$	initial position of the centre of a spherical indenter in a nanoindentation
$\mathbf{r}_i$	atomic position of atom i
$\mathbf{r}_{i,j,\text{ref}}$	initial reference vectors used for the original definition
$\mathbf{r}_{j,i}$	vector from atom j to atom i
$\mathbf{r}_{ji}$	vector from atom j to atom i
$\mathbf{v}_{\text{cm}}$	centre-of-mass velocity of rescaled atoms
$\mathbf{v}_{\text{ind}}$	velocity of the indenter in a nanoindentation
$\mathbf{v}_i$	velocity of atom i
$\mathbf{v}_i^{\text{resc}}$	velocity of atom i after the application of the local thermostat
$\mathbf{v}_i^{\text{unsc}}$	velocity of atom i before the application of the local thermostat
$\mathbf{x}_i$	position of atom i
ρ_i^{atom}	load of atom i used in the load balancing algorithm
$\rho_p^{(\mathcal{X})}$	average work per particle of calculation $\mathcal{X}$ on processor p
ρ_{klm}^{dxa}	dislocation density of klm dislocations
σ	standard deviation
σ^2	variance
τ_{atom}	computational time per atom and timestep
$\tau_{\text{system}}^{\text{PP}}$	estimated calculation time of the system calculated with the precise potential

τ_p	total force computation time of processor p
$\tau_p^{(\mathcal{X})}$	measured calculation time of calculation $\mathcal{X}$ on processor p
CSP_{hi}	higher threshold of the CSP used to calculate a switching parameter
CSP_{lo}	lower threshold of the CSP used to calculate a switching parameter
CSP_i	centro-symmetry parameter of atom i
$\text{CSP}_i^{\text{list}}$	differentiable centro-symmetry parameter
$\text{CSP}_i^{\text{ref}}$	CSP calculated with initial reference vectors
$\text{CSP}_{\text{avg},i}$	time averaged CSP
$\Theta(x)$	Heaviside step function
$\Theta(x)$	Heaviside step function
Θ_{ij}^{in}	1 if j is an atom in the CSP pair-list of atom i and 0 otherwise
Θ_{ij}^{ngh}	atom, that was at the begin of the simulation on the opposite side of i than j
$\tilde{c}_{\mathbf{n}lm}^{(p,K)}$	expansion coefficient of a many-body basis function
$\tilde{E}_i$	term used for the calculation of conservative adaptive precision forces
$\tilde{w}_{\mathbf{n}l}^{(K,B)}$	weight of the cluster basis function $B_{\mathbf{n}l}^{(K,B)}$ calculated with the variance instead of standard deviation
$\varphi_i^{(p)}$	atomic property number p of atom i given by an atomic cluster expansion
$\varphi_i^{(p,A)}$	atomic property p expanded in many-body basis functions
$\varphi_i^{(p,B)}$	atomic property p expanded in cluster basis functions
ξ	embedding function of an EAM potential
$\xi^{\text{Cu,Fit}}$	optimised embedding function of the EAM copper potential
ξ^{Cu}	embedding function of the original copper EAM potential
$\xi_0^{\text{Cu,Fit}}$	energy offset in the embedding function of copper
$\zeta(r_{ij})$	contribution of the atom pair ij to the electron charge density of atoms i and j
ζ_i	electron charge density of atom i
a_0^{fcc}	lattice parameter at 0 K in an fcc lattice
a_0^{fcc}	lattice parameter of a fcc lattice
A_c	projected contact area
a_c	contact radius
a_{pz}	radius of the plastic zone
A_o	scalar property of the observable o used in the loss function of atomicrex
$A_{\mathbf{n}lm}^{(K)}$	many-body basis function of ACE
$A_{\mathbf{n}l\ell m\ell}$	atomic base of a fast ACE potential
$A_{\mathbf{n}p\ell p m p}$	atomic base of a precise ACE potential
$A_{\mathbf{n}lm}$	atomic base of ACE
$B_{\mathbf{n}l\ell}^{(K,\text{ref})}$	reference values of the cluster basis function in the reference environment to which the fast ACE potential is fitted

$B_{\mathbf{ml}}^{(K)}$	cluster basis function of ACE
$B_{\mathbf{ml}}^{(K^f)}$	cluster basis function with a small basis size of a fast ACE potential
C_{11}	elastic constant
C_{12}	elastic constant
C_{44}	elastic constant
$c_{\mathbf{nl}}^{(p,K)}$	independent expansion coefficient of a cluster basis function
$c_{\text{rot},\mathbf{ml}}^{(K)}$	generalised Clebsch-Gordan coefficient
d_{hi}	threshold of the descriptor d_i above which atoms are calculated with the precise model
d_{lo}	threshold of the descriptor d_i below which atoms are calculated with the fast model
d_i	descriptor of atom i used to calculate a differentiable switching parameter
E	total potential energy of all atoms
E^{IMOMM}	potential energy of the IMOMM-approach
$E_{\text{precise}}^{\text{PAP}}$	potential energy of atoms in the precise region of PAP
$E_{\text{QM}}^{\text{PAP}}$	total quantum mechanical energy according to PAP
$E_{\text{precise},ij}^{\text{PAP}}$	potential energy of the precise system plus atoms i and j calculated with QM
$E_{\text{precise},i}^{\text{PAP}}$	potential energy of the precise system plus atom i calculated with QM
$E_{\text{struct},s}^{\text{pred}}$	total potential energy of a structure s predicted with the current set of parameters during the minimisation of a loss function
$E_{\text{struct},s}^{\text{targ}}$	total potential energy of a structure which is used as reference value
E^*	indentation modulus
E_{coh}	cohesive energy
E_{ext}	energy of an external field
$E_{\text{min}}^{\text{targ}}$	approximation of the ground state of the potential energy in pacemaker using E_{si}^{targ}
$E_{\text{MM}}^{\text{precise}}$	potential energy of the precise subsystem calculated with molecular mechanics
$E_{\text{MM}}^{\text{sys}}$	potential energy of a system calculated with molecular mechanics
$E_{\text{QM}}^{\text{precise}}$	potential energy of the precise subsystem calculated with quantum mechanics
$E_{\text{struct},\text{min}}^{\text{targ}}$	approximation of the ground state of the potential energy in pacemaker
E_i	potential energy of atom i
$E_i^{(f)}$	potential energy of an atom i calculated with a fast potential
$E_i^{(p)}$	potential energy of an atom i calculated with a precise potential
$E_i^{\text{ACE,FS}}$	potential energy of atom i described by ACE with a Finnis-Sinclair embedding function
$E_i^{\text{ACE,f}}$	potential energy of atom i calculated with a fast ACE potential
$E_i^{\text{ACE,p}}$	potential energy of atom i calculated with a precise ACE potential
E_i^{ACE}	potential energy of atom i described by ACE
E_i^{APA}	potential energy of atom i calculated with the potential APA
E_i^{AP}	potential energy of atom i according to an adaptive-precision potential

E_i^{CAP}	potential energy of atom i described by a conservative adaptive precision potential
E_i^{EAM}	potential energy of atom i according to an EAM potential
E_i^{EF}	potential energy of atom i according to the first coupling approach
E_i^{PAP}	quantum mechanical interaction of switching atom i with the spherical precise region of PAP
E_{ij}^{pair}	potential energy of the atom pair ij
E_{ij}^{PAP}	quantum mechanical interaction of the switching atoms i and j
E_i^{pair}	potential energy of atom i according to a pair potential
E_{si}^{pred}	potential energy of atom i in structure s predicted with the current set of parameters during the minimisation of a loss function
E_{si}^{targ}	potential energy of atom i in structure s which is used as reference value
$f^{(\text{cut})}$	radial function
$f^{(\text{trans})}(x, x_{\text{lo}}, x_{\text{hi}})$	transition function that maps $x \in \mathbb{R}$ to $[0, 1]$
f_{norm}	constant used to rescale a force
f_{pz}	material-dependent factor
$f_{ki\alpha}^{\text{A,ACE}}$	spatial component α of $f_{ki}^{\text{A,ACE}}$
g_i	differentiable atomic quantity of atom i that is locally averaged to calculate a differentiable descriptor d_i
H	Hamiltonian
H	total energy of a system
h	indentation depth
H^{FEC}	free energy compensation term of H-AdResS
$H^{\text{H-AdResS}}$	Hamiltonian of H-AdResS
$H_{\alpha}^{\text{inter,FEC}}$	correction of the intermolecular potential of atom α
I	imbalance of the load in a parallel simulation
I^{thr}	user-defined imbalance threshold above which the load-balancer is used
k_{ind}	force constant of an indenter in a nanoindentation
l_{dxa}	dislocation length of klm dislocations
$m_{\alpha i}$	mass of atom i of molecule α
N^{imp}	number of nearest neighbours used in the CSP calculation according to LAMMPS
N^{PAP}	number of atoms in the switching zone of PAP
N_{buffer}	additional neighbours used to identify the nearest neighbour pairs during the CSP-calculation
N_{struct}	number of structures
N_i^{ngh}	number of neighbours of atom i within the force cutoff
N_p^{aa}	number of atoms administered by processor p
$N_p^{(\mathcal{X})}$	number of calculations of calculation $\mathcal{X}$ on processor p
N_s^{atoms}	number of atoms in structure s
$N_{\lambda, \text{avg}}$	number of timesteps used to average a switching parameter

$N_{\text{CSP,avg}}$	number of averaged timesteps
P	number of properties that $\mathcal{F}$ depends on
P_{H}	Hertzian load
p_{m}	mean contact pressure
r_{cut}	cutoff radius of an interatomic potential
$r_{\text{cut}}^{\text{pot}}$	cutoff radius of the interatomic potential 'pot'
R_{ind}	radius of an indenter in a nanoindentation
$r_{\text{w}}^{\text{cut}}$	cutoff distance beyond which $w^{\text{d}}(r)$ is zero
$r_{\lambda,\text{hi}}$	distance which is used as upper threshold to include a spatial switching zone in a switching parameter
$r_{\lambda,\text{lo}}$	distance which is used as lower threshold to include a spatial switching zone in a switching parameter
R_{nl}	radial basis functions of ACE
S	speedup of an adaptive-precision simulation compared to a constant precision simulation of the precise potential
S^{est}	on-the-fly estimated speedup of an adaptive-precision potential
T	temperature
t	time
T^{ref}	temperature of the reference environment to which a fast ACE potential is fitted
$t^{(\text{d})}$	overhead when a conservative adaptive-precision potential is computed for the whole system
$t^{(\text{f})}$	computing time of the whole system with the fast model
$t^{(\text{p})}$	computing time of the whole system with the precise model
T_{targ}	target temperature of a thermostat
T_i	kinetic energy of atom i
T_i	kinetic energy of atom i
V^{dxa}	effective volume of the plastic zone
V^{inter}	total potential energy of an intermolecular potential
$V_{\alpha}^{\text{inter,AA}}$	potential energy of molecule α calculated with an intermolecular potential using all atoms
$V_{\alpha}^{\text{inter,CG}}$	potential energy of molecule α calculated with an intermolecular potential using coarse-grained particles
$V_{\alpha}^{\text{intra}}$	potential energy of molecule α calculated with an intramolecular potential
V_{ind}	potential of the indenter in a nanoindentation
V_{ind}	volume that is displaced by the indenter
$V_{\alpha\alpha'}^{\text{inter,AA}}$	all-atom potential of a pair of atoms i and j from molecules α and α'
$V_{\alpha\alpha'}^{\text{inter,CG}}$	intermolecular potential of a pair of molecules
w^{B}	scaling factor for $w_{\mathbf{n}^{\text{f}}\mathbf{t}^{\text{f}}}^{(K^{\text{f}},\text{B})}$
$w^{\text{d}}(r)$	differentiable weighting function used for the descriptor calculation
$w_{\text{norm}}^{\text{E}}$	constant that ensures the normalisation of w_s^{E}

$w_{\text{norm}}^{\text{F}}$	constant that ensures the normalisation of w_{si}^{F}
$w_{\text{norm}}^{\text{E,MD}}$	constant that ensures the normalisation of $w_{si}^{\text{E,EB}}$
$w_s^{\text{E,EB}}$	weight of the energy of structure s according to the energy-based weighting policy
w_s^{E}	weight of the energy of structure s
$w_{\mathbf{n}^{\text{f}}\mathbf{t}^{\text{f}}}^{(\text{K}^{\text{f}},\text{B})}$	weights of a cluster basis function difference
$w_{i,j_{\text{pot}}}^{\text{EF}}$	weight of the potential j_{pot} on atom i
$w_{si}^{\text{E,EB}}$	weight of the potential energy of atom i in structure s according to the energy-based weighting policy
w_{si}^{E}	weight of the potential energy of atom i in structure s
$w_{si}^{\text{F,EB}}$	weight of the force of atom i in structure s according to the energy-based weighting policy
w_{si}^{F}	weight of the force of atom i in structure s
x_{fpm}	fraction of atoms that are calculated with the precise model
Y_l^m	spherical harmonics
Z_{ref}	reference nuclear charge number
Z_i	nuclear charge number of atom i

Chapter 1

Introduction

Computer simulations of atomistic systems have developed into a powerful tool used in physics, chemistry and materials science[1–11]. The key quantity in the simulation of atomistic systems is the potential energy of the system, from which one can calculate the forces on atoms. The potential energy is most accurately calculated using quantum mechanics (QM). However, efficiently calculating the electronic structure with density functional theory (DFT)[5] is only possible for small systems of hundreds or thousands of atoms[12, 13].

However, larger systems with magnitudes more atoms are required for certain simulations to be more realistic – like for the investigation of plastic deformation[14–16] or melting[17–19]. One rarely can use first-principle methods and solve the Schrödinger equation for such large systems, but has to use faster methods. Interatomic potentials neglect the electronic structure and calculate the potential energy of a system directly from the atomic positions and the chemical species, which allows the simulation of millions of atoms[1].

Traditional empirical interatomic potentials assume a functional form of the potential with few parameters, that are fitted to reproduce experimental measurements of interest[20–22]. A well-known example is the Lennard-Jones (LJ) potential[20], which is a pair potential with one attractive and one repulsive term. Lennard-Jones optimised the parameters until the famous 12-6 potential for Argon was published in 1931[20, 23], which developed as a commonly used benchmark potential and is still in use nowadays[24, 25]. Over time, the proposed functional form of interatomic potentials has become more and more complicated[26, 27] and can nowadays include even QM-principles[28, 29]. The advent of DFT influenced the development of interatomic potentials: while calculating millions of atoms is difficult due to non-linear scaling with the system size[30], DFT can calculate many small systems of hundreds of atoms. Thus, the experimental reference values have been partially or completely replaced by DFT calculations[26, 31, 32].

In recent years, machine learning (ML) influenced the development of interatomic potentials, following a different approach than traditional empirical potentials. Rather than proposing a fixed functional form – like Lennard-Jones did – ML potentials can use a more general form with orders of magnitude more parameters, as all these parameters can be learned from a huge training dataset of DFT-reference values. In the 1990s, the first ML-potentials directly calculated the potential energy of the whole system[33–35], incapable of dealing with a different number of atoms than they were trained to. Since 2007, ML-potentials are able to calculate large systems of variable sizes through two assumptions: first, like for empirical potentials, the potential energy of a system is given by the sum over the potential energy of all atoms and, second, the potential energy of an atom can be calculated from the local environment of this atom, described by descriptors that are invariant under rotation, translation and permutation of atoms[36]. Based on this idea, many ML potentials have been developed[37–40], which provide a DFT-like accuracy, that remarkably increased compared to the accuracy of traditional empirical potentials[4, 41–43]: The accuracy of the interactions between particles in long time simulations has been limited in the past due to the approximative nature of the classical force-field description. With ab-initio molecular dynamics (MD)[44] limited to small systems, ML potentials have received large attention for translating the accuracy of DFT calculations into large MD simulations[45–51].

In summary, there is a variety of different interatomic potentials – traditional ones with rather fixed functional form and modern ML potentials with a more general form and magnitudes more parameters. Therefore, selecting an approach for a potential for a simulation has become increasingly difficult over time. The choice of the potential-approach depends primarily on the phenomena and material, which need to be simulated. For example, the bonding in transition metals has both a nearly free electron and directional covalent type, whose strengths depends on the filling of the

d-band and determines in which lattice a transition metal crystallises in[52]. With a full d-band, like for face-centred cubic (fcc) copper (Cu), the bonding is metallic and can be described with sufficient accuracy by many-body central force approaches (such as the embedded atom model (EAM))[53]. In contrast, the partially filled d-bands of body-centred cubic (bcc) transition metals – such as tungsten (W) – cause also directional bonds, that cannot be described properly by central-force models but require explicit descriptions of the directional (and thus angular dependent) bonds[52, 54–60]. Such angular-dependent bonds are well described by modern many-body ML potentials – like the recently developed atomic cluster expansion[40](ACE), which is a state-of-the-art ML potential[4, 61–65].

The ongoing modelling of more accurate potentials comes at the cost of computational time: most ML potentials remain several orders of magnitude slower than the simple empirical potentials[4, 62, 66]. Since the length and time scale, which can be covered in a simulation, is strongly dependent on the cost of energy- and force-computation[67], the evaluation time of the interatomic potential is a crucial factor. With computational resources being limited, there is a conflict of interest between the accuracy of the potential on the one hand and time and length scale of the simulation on the other hand. Therefore, making interatomic potentials more efficient and decreasing their required computing time is an essential research objective. Thus, there are approaches, which do not aim for the highest accuracy possible, but use simpler and faster descriptors[68, 69], fewer descriptors[70]¹ or both [71] to speedup ML potentials. However, modifying the whole potential in such a way decreases the accuracy in the whole simulated system – which always affects the decisive atoms. Instead, one can save computing time by reducing the accuracy in a more targeted approach.

Some simulations, like the ones of phase transitions or amorphous materials, require the highest accuracy in the whole system. However, the latter is not true for all systems: Large-scale MD simulations of nanoindentations [72, 73] can provide valuable insight into microscopic mechanical behaviour and reveal underlying mechanisms that govern initial stages of plastic deformation: when the indenter penetrates into a surface, the material first deforms elastically and then, after reaching the yield stress, continues with irreversible plastic deformation, where the affected volume can be described by the elastic and plastic zone, respectively[74]. Slip, mediated by dislocations, and twinning as fundamental mechanisms of plastic deformation[75] can be directly observed and tracked in atomistic simulations of nanoindentations[76], but cannot be directly observed in experiments. The simulation of these phenomena requires the highest accuracy: The used interatomic potential is decisive to get trustworthy results in a nanoindentation simulation since especially atomic-level details vary strongly dependent on the used interatomic potential[24, 77–79]. Furthermore, the length scale of nanoindentations is challenging[80]: too small systems are problematic as the elastic and plastic zone should not be influenced by the system size[81, 82] and, once nucleated and gliding away from the indentation point, dislocations should not interact across a periodic boundary. Therefore, large systems with millions of atoms are required to simulate nanoindentations[83–85]. The majority of the system serves as material background and needs to maintain elastic fields and provide space for dislocations to glide. Thus, this majority of the system does not require the highest accuracy possible and one can save computational time by decreasing the accuracy in this part of the system[86–89].

There is a wide range of applications, which do not require the highest accuracy in the whole system. In materials science, apart from the discussed nanoindentations, e.g., dynamical crack propagation requires only a precise simulation of the crack tip. In chemistry, e.g., catalytic reactions at surfaces or interfaces require a precise treatment of reacting atoms and surface atoms, but not for atoms far below the surface or far away from reaction centres. In physics, surface-growth processes, for example for semiconductors, require a precise treatment of the surface, but with evolving growth and agglomeration of many new atoms, the atoms belonging to the original surface layer become less important and can be calculated with less precision, i.e., they do not contribute actively any more to the physical growth process.

For all these applications, one can maintain the required accuracy while improving computational efficiency by coupling different interaction models. Typically, a more accurate but computationally demanding model is only used for a small portion of the simulated system, in which the decisive processes take place. An efficient but less accurate model is then applied for the remaining parts of the simulated system, in which only small variations are expected (for instance maintaining a long range elastic field or temperature gradients). Traditionally regions for precise and fast models are identified before the start of a simulation and the challenge is a sound description of the interface between the two regions [90–98], e.g., to prevent phonon distraction or reflection at this interface.

¹An optimisation of hyperparameters in Ref. [70] can increase the precision while maintaining the cost. Alternatively, like the comparison of REF-ACE and XPOT-ACE-4F shows, one can decrease the accuracy slightly (for the worst test by a factor of 1.07 and 1.72 for forces and energies) for a speedup by the factor of 5.

However, this prior region identification requires knowledge of expected key atomistic events before the start of the simulation, while for many simulations it is desirable that regions of fast or precise description appear or vanish dynamically as dictated by the trajectories of the atoms. A well-known and widely applied example is coupling of QM approaches with molecular mechanics (MM), which originated almost 50 years ago [90]. Current QM/MM implementations are able to adjust both regions during simulations based on various criteria [99, 100]. On a coarser level, e.g., in soft matter [6, 93, 95, 101–103], an adaptive description has been introduced to reduce the number of degrees of freedom in a system to speed up the computations. On even larger scales, coupling between classical atomistic simulations and continuum simulations (like material point method [104] or finite element simulations [105]) are performed. The commonality of the given examples is that one can separate different subsystems, i.e., one subsystem which has to be described with high precision because of high sensitivity of results on the underlying method, and another subsystem, which serves as a material background, which is not strongly perturbed and can be described by cheaper, less accurate methods. Since the perturbation state of a material might show temporal and spatial dependence, such a distinction between subsystems ideally works dynamically and adaptively.

Such a coupling is not restricted to two completely different methods, but can be also used within all-atom (AA) simulations without decreasing the number of degrees of freedom, i.e., the coupling can be done also for two interatomic potentials with different levels of accuracy and computational cost [91, 92, 106, 107]. However, there are only few approaches [108], that couple two interatomic potentials in way, which is appropriate for a microcanonical ensemble, since usually either energy, momentum or both are not conserved. The present work develops such an energy- and momentum conserving approach to speed up large-scale atomistic simulations by automatically adjusting the precision locally dependent on the atomic configuration.

In a first attempt, the possibility of conservative adaptive-precision ML potentials is explored by creating a conservative adaptive-precision ACE potential: Starting from a precise ACE potential, a faster ACE potential is created by using fewer descriptors, which is then coupled to the original ACE potential. However, the considerable performance gap of 2-3 magnitudes between the evaluation of traditional empirical and modern ML potentials makes a coupling of a fast empirical and a precise ML potential most promising. Therefore, an energy- and momentum conserving framework for such a coupling of empirical- and ML-potentials is developed based on the learnings from the ACE-ACE coupling. The coupling of empirical- and ML-potentials is demonstrated by coupling a fast EAM and a precise ACE potential. EAM can accurately describe fcc Cu, but fails in describing bcc W due to the previously discussed directional bonding of W. Therefore, fcc Cu and bcc W are ideal to validate and test the newly developed EAM-ACE coupling method. Simulations of nanoindentations are – as discussed previously – a prototypical application for adaptive-precision methods. Thus, nanoindentation simulations of fcc Cu and bcc W are performed with adaptive-precision EAM-ACE potentials and compared with nanoindentations, that are purely simulated with EAM and ACE, respectively.

Finally, the novel energy- and momentum-conserving, dynamically self adapting coupling approach for MD simulations, that couples fast and precise interatomic potentials, is further improved to a conservative adaptive-precision potential. Thereby, conservative adaptive-precision potentials can be regularly used in the future – without the currently existing problems caused by the violation of conservativity – saving a significant amount of energy and paving the way to more resource-responsible computing.

These new coupling approaches have the potential to be applied in the wide range of previously discussed applications in physics, chemistry and materials science, which are currently restricted to either limited global accuracy or to small system sizes due to computational efficiency. The Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [1] is a modularly designed open-source MD code, that is widely used [14, 68, 74, 77, 109] and whose capabilities have been extended over the years by many community contributions [4, 110–112]. All of the previously outlined use cases can be simulated with LAMMPS [113–119]. Therefore, the new coupling approaches are implemented as adaptive-precision interatomic potential (APIP)-package for the LAMMPS code [1]. Thereby, the achievements presented in the present work are available to the community – to resolve the conflict of interest between high accuracy and more realistic time- and length-scales of simulations.

The present work is organised as follows: in Chapter 2, the few existing coupling approaches for two interatomic potentials are discussed, resulting in the new coupling approach for two interatomic potentials, that is appropriate to simulate a microcanonical ensemble. The developed coupling approach is tested to couple two ML-based ACE potentials of different precision and cost in Chapter 3. As traditional empirical potentials are much faster than the used fast ACE potential, a

framework for the coupling of empirical and ML potentials is presented in Chapter 4, which is used to couple EAM and ACE Cu potentials. Computational performance and accuracy of the EAM-ACE coupling approach are demonstrated in Chapter 5 in nanoindentations of fcc Cu and bcc W. Finally, a method to achieve both, energy and force precision, for conservative adaptive-precision potentials is developed in Chapter 6. The underlying APIP-package for LAMMPS is presented in Chapter 7. Chapter 8 concludes with a summary of the present work and an outlook on further opportunities for improvement.

Chapter 2

Coupling mechanisms

The Hamiltonian H of a classical system gives the total energy of a system and consists of the potential energy E_i and the kinetic energy T_i of all atoms i , i.e.

$$H = \sum_i (T_i + E_i) . \quad (2.1)$$

An interatomic potential calculates the potential energy E_i of an atom i . Here, it is assumed that the calculation depends only on the atomic positions $\mathbf{r}_i$. More complicated descriptions can include, amongst others, charges and magnetism. In the absence of long-range interaction like Coulomb interaction, an interaction is short-range. For short-range interactions, atoms i and j do not interact if they are further apart than the cutoff radius r_{cut} . Therefore, the potential energy E_i of an atom i depends only on the distances $\mathbf{r}_{ji} = \mathbf{r}_j - \mathbf{r}_i$ to all atoms j within the force cutoff of atom i . The force $\mathbf{F}_i$ on an atom i is calculated from the total potential energy E as

$$\mathbf{F}_i = -\nabla_i E = -\nabla_i \sum_j E_j , \quad (2.2)$$

where ∇ is the nabla-operator. With the atomic force $\mathbf{F}_i$ one can integrate the equations of motion over time, i.e., propagate a given set of atoms in time.

There are many interatomic potentials[20–22, 37–40, 120–123] with a varying degree of complexity. Therefore, one can select two interatomic potentials, where the first interatomic potential is more precise and the second interatomic potential faster. With the first potential, one can calculate the precise potential energy $E_i^{(\text{p})}$. With the second potential, one can calculate faster the potential energy $E_i^{(\text{f})}$ which is however less precise than $E_i^{(\text{p})}$. The corresponding forces $\mathbf{F}_i^{(\text{p})}$ and $\mathbf{F}_i^{(\text{f})}$ are then calculated according to Eq. (2.2). When the high precision of the first potential is only required for a subset of all atoms in a system, an adaptive-precision approach which couples the precise and the fast potential becomes promising. Using the faster second potential for all atoms that do not require the high precision would save computational time. However, the question of how to couple two models of different precision and cost in a way that allows a precision change of subsystems at runtime cannot be answered conclusively.

The most precise and expensive interatomic potentials, machine-learning (ML) potentials, are rather new. Therefore, the range of precision and cost which is available within interatomic potentials has grown recently. As this range is the sole motivation for the coupling, there are few approaches for the coupling of two interatomic potentials. In contrast, the coupling between two different methods is a long used technique. Quantum mechanics (QM) has been coupled to molecular mechanics (MM) for almost 50 years[90]. Meanwhile, adaptive precision adjustments are also included in hybrid QM/MM simulations[99, 100]. As the MM region of a QM/MM simulation is described by interatomic potentials, the adaptive-precision QM/MM couplings are of interest for the development of a coupling method for two interatomic potentials. On the other hand, interatomic potentials are also coupled to faster methods. The simulation of coarse-grained (CG) particles can be coupled with all-atom (AA) simulations with adaptive-precision[6, 93, 124–126].

In this chapter, approaches for the coupling of two interatomic descriptions of different precision and cost are discussed, where QM/MM and AA/CG approaches are included due to the partial usage of interatomic potentials. The coupling approaches can be categorised into three groups: an approach can be based on the weighting of energies, forces or both. Weighting both energies and forces is discussed in Section 2.1. Coupling approaches based on the forces are discussed in Section 2.2 and energy-based approaches in Section 2.3. The consequences of the presented approaches for a coupling of two interatomic potentials are discussed in Section 2.4.

2.1 Weighting energies and forces (all-atom/all-atom)

This first approach is suggested in Ref. [107] and used to couple a fast Tersoff potential with a precise ReaxFF reactive potential to simulate crack propagation in silicon. Each potential j_{pot} is weighted per atom i with a weighting factor $w_{i,j_{\text{pot}}}^{\text{EF}}$. The weights are normalised through [107]

$$\sum_{j_{\text{pot}}} w_{i,j_{\text{pot}}}^{\text{EF}} = 1, \quad (2.3)$$

where $w_{i,j_{\text{pot}}}^{\text{EF}} \in [0, 1]$ applies. For the coupling of two potentials follows $w_{i,1}^{\text{EF}} = 1 - w_{i,0}^{\text{EF}}$. Thus, the switching parameter $\lambda_i^{\text{EF}} = w_{i,0}^{\text{EF}}$ is introduced, that determines the used potential for atom i . The potential energy E_i^{EF} and the atomic force $\mathbf{F}_i^{\text{EF}}$ according to this approach are given by [107]

$$E_i^{\text{EF}} = E_i^{(\text{f})} \lambda_i^{\text{EF}} + E_i^{(\text{p})} (1 - \lambda_i^{\text{EF}}), \quad (2.4)$$

$$\mathbf{F}_i^{\text{EF}} = \mathbf{F}_i^{(\text{f})} \lambda_i^{\text{EF}} + \mathbf{F}_i^{(\text{p})} (1 - \lambda_i^{\text{EF}}). \quad (2.5)$$

The switching parameter λ_i^{EF} is calculated based on the strain energy as the coupled potentials are similar for low strains but deviate for large ones.

For this first approach, $\mathbf{F}_i^{\text{EF}} \neq -\nabla_i \sum_j E_j^{\text{EF}}$ applies instead of Eq. (2.2). Thus, this potential is not conservative and violates energy conservation. Therefore, this coupling approach requires a thermostat and one cannot simulate a microcanonical ensemble. The force on atom i with $\lambda_i^{\text{EF}} = 1$ is according to Eq. (2.2) $\mathbf{F}_i^{(\text{f})}$. For Newton's third law to apply, the force of all neighbours j of the considered atom i needs to be $\mathbf{F}_j^{(\text{f})}$, i.e., $\lambda_j^{\text{EF}} = 1$ needs to apply for all neighbours j of i . As the neighbouring atoms j have also neighbours, this argument can be repeated until $\lambda_i^{\text{EF}} = 1$ applies for all atoms i and thus the fast potential is not coupled any more to the precise potential. Therefore, using a switching parameter λ_i^{EF} to weight both force and potential energy of atom i according to Eqs. (2.4) and (2.5) violates both Newton's third law and momentum conservation. For consistency, either the energy or force can be weighted by a factor [94].

2.2 Weighting forces

The force is, in contrast to the potential energy, entering directly into the integration of motion. Thus, several approaches [6, 92, 99, 106, 124] are based on a weighting of forces. A mixing approach per atom for two interatomic potentials is presented in Section 2.2.1. An approach for a AA/CG coupling based on pairs is given in Section 2.2.2. Finally, a QM/MM approach based on a weighting per atom is presented in Section 2.2.3.

2.2.1 Per atom (all-atom/all-atom)

An approach to weight forces per atom is used in Ref. [92] for a liquid water system. A precise interatomic potential is used in a first region and a fast interatomic potential is used in a second region. There is a switching region with $\lambda_i \in (0, 1)$ between both regions. The force is given as [92]

$$\mathbf{F}_i^{\text{FPA}} = \lambda_i \mathbf{F}_i^{(\text{p})} + (1 - \lambda_i) \mathbf{F}_i^{(\text{f})} + \mathbf{F}_i^{\text{L}} + \mathbf{F}_i^{\text{T,FPA}}, \quad (2.6)$$

where $\mathbf{F}_i^{\text{L}}$ is a Langevin force and $\mathbf{F}_i^{\text{T,FPA}}$ is a thermodynamic force. The different definitions of the precise and the fast interatomic potential cause a pressure imbalance on the transition region. The from this pressure difference resulting error of the density profile in the transition region is minimised by the thermodynamic force $\mathbf{F}_i^{\text{T,FPA}}$. This approach approximates the grand-canonical ensemble.

This approach does not comment on how to calculate the potential energy. To integrate the equations of motion over time in a MD simulation one only needs the forces on all particles, but the potential energy is not required and can therefore be undefined. As the grand-canonical is approximated and the potential energy is undefined, this approach does not conserve the energy. The thermodynamic force $\mathbf{F}_i^{\text{T,FPA}}$ depends on the position $\mathbf{r}_i$ of atom i rather than a position difference $\mathbf{r}_{ij}$ of two atoms and thus neither momentum conservation nor Newton's third law apply. The interpolated force $\lambda_i \mathbf{F}_i^{(\text{p})} + (1 - \lambda_i) \mathbf{F}_i^{(\text{f})}$ does not conserve the momentum like previously discussed in Section 2.1 for Eq. (2.5) which is similar to Eq. (2.6).

2.2.2 Per pair (all-atom/coarse-grained)

The force based AdResS-approach is presented in Ref. [6, 124] and used in a system of liquid molecules. A coarse-grained region is coupled via a transition zone with $\lambda_i^{\text{FPP}} \in (0, 1)$ to an all-atom-region, where the all-atom region is described by a pair potential. Thus, one can calculate the force $\mathbf{F}_{ij}^{\text{at}}$ between two molecules i and j from the pair potential. Furthermore, one can derive a coarse-grained pair force $\mathbf{F}_{ij}^{\text{cg}}$. Between the all-atom and the coarse-grained region is a switching region in which $\lambda_i \in (0, 1)$ applies and thus atoms are described by a combination of both interaction models. The force according to this approach is [124]

$$\mathbf{F}_{ij}^{\text{FPP}} = \lambda_i^{\text{FPP}} \lambda_j^{\text{FPP}} \mathbf{F}_{ij}^{\text{at}} + (1 - \lambda_i^{\text{FPP}} \lambda_j^{\text{FPP}}) \mathbf{F}_{ij}^{\text{cg}}, \quad (2.7)$$

where $\lambda_i^{\text{FPP}} \in [0, 1]$ is the switching parameter of molecule i . Newton's third law applies for the pair potentials, i.e., $\mathbf{F}_{ij}^{\text{at}} = -\mathbf{F}_{ji}^{\text{at}}$ and $\mathbf{F}_{ij}^{\text{cg}} = -\mathbf{F}_{ji}^{\text{cg}}$ applies. Therefore, Eq. (2.7) satisfies Newton's third law and thus conserves the momentum as $\mathbf{F}_{ij}^{\text{FPP}} = -\mathbf{F}_{ji}^{\text{FPP}}$ applies by design. However, this approach does not result in energy conservation, since the underlying potential cannot be reconstructed from the weighted forces (cf. Eq. (2.7))[6]. Thus, this approach uses a thermostat to simulate the grand canonical ensemble. As a thermostat can conserve the momentum, this approach is in principle momentum conserving.

2.2.3 Per atom (quantum mechanics/molecular mechanics)

The coupling of quantum mechanics and molecular mechanics (QM/MM) was introduced in Ref. [90]. One can define the potential energy according to the integrated molecular orbital molecular mechanics (IMOMM) scheme as [127]

$$E^{\text{IMOMM}} = E_{\text{MM}}^{\text{sys}} + E_{\text{QM}}^{\text{precise}} - E_{\text{MM}}^{\text{precise}}, \quad (2.8)$$

where $E_{\text{MM}}^{\text{sys}}$ is the potential energy of the system calculated with molecular mechanics and the potential energy of the precisely treated subsystem is $E_{\text{QM}}^{\text{precise}}$ when calculated with quantum mechanics and $E_{\text{MM}}^{\text{precise}}$ when calculated with molecular mechanics. Double counting interactions is prevented by the subtraction of $E_{\text{MM}}^{\text{precise}}$.

The IMOMM energy is reused in the hot spot approach in Ref. [99]. The hot spot approach uses the quantum mechanical energies in a spherical region, i.e., the hot spot. The MM energies are used according to Eq. (2.8) for all interactions which are not covered by the QM-calculation of the hot spot. Calculating the energy according to Eq. (2.8) provides the forces $\mathbf{F}_i^{\text{MM}}$ according to Eq. (2.2) and $\mathbf{F}_i^{\text{QM}}$ dependent on the quantum mechanical calculation. The forces are weighted like in Eq. (2.5), namely as

$$\mathbf{F}_i^{\text{HS}} = \lambda_i \mathbf{F}_i^{\text{QM}} + (1 - \lambda_i) \mathbf{F}_i^{\text{MM}}, \quad (2.9)$$

where the switching parameter λ_i is 1 at the centre of the hot spot and decays dependent on the distance to the centre of the hot spot in a small switching region from 1 and reaches 0 at the radius of the hot spot. The in the force $\mathbf{F}_i^{\text{HS}}$ (cf. Eq. (2.9)) introduced switching function does not follow from E^{IMOMM} (cf. Eq. (2.8)). Therefore, the hot spot method does not conserve the energy and is used to simulate a canonical ensemble. Furthermore, the hot spot method does not conserve the momentum for the in Section 2.1 discussed reason.

2.3 Weighting Energies

Approaches based on weighting of energies have been taken in Refs. [93, 100, 125, 126, 128]. An per-atom weighting approach for AA/CG coupling is described in Section 2.3.1 and an pair-weighting approach in Section 2.3.2. A QM/MM approach based on the weighting of clusters is discussed in Section 2.3.3.

2.3.1 Per atom (all-atom/coarse-grained)

H-AdResS as described in Ref. [93] is used to simulate a microcanonical ensemble for molecular liquids, where there is a coarse-grained region and an all-atom region. In the switching region in between, the switching parameter λ changes continuously from 0 to 1 dependent on the position of a molecule α . The aim is to generate a thermodynamic equilibrium between the coarse-grained and the all-atom region, where the ansatz ensures that pressure or density are equal in the whole system.

The interaction between atoms is described by the intramolecular potential $V_{\alpha}^{\text{intra}}$ for atoms of the same molecule and by the intermolecular pair potentials $V_{\alpha}^{\text{inter,AA}}$ or $V_{\alpha}^{\text{inter,CG}}$ dependent on the switching parameter λ_{α} for atoms of different molecules. The Hamiltonian (cf. Eq. (2.1)) is given as [93]

$$H^{\text{H-AdResS}} = \sum_{\alpha i} \frac{\mathbf{p}_{\alpha i}^2}{2m_{\alpha i}} + \sum_{\alpha} (\lambda_{\alpha} V_{\alpha}^{\text{inter,AA}} + (1 - \lambda_{\alpha}) V_{\alpha}^{\text{inter,CG}} + V_{\alpha}^{\text{intra}} + H^{\text{FEC}}(\lambda_{\alpha})) , \quad (2.10)$$

where $\mathbf{p}_{\alpha i}$ and $m_{\alpha i}$ are mass and momentum of atom i of molecule α and $H^{\text{FEC}}(\lambda_{\alpha})$ is a free energy compensation term that depends on the switching parameter. The free energy compensation term is best understood after calculating the forces following from this Hamiltonian. The force contains the contributions $\mathbf{F}_{\alpha i \alpha' j}^{\text{inter,AA}}$ from the intermolecular all-atom potential, $\mathbf{F}_{\alpha i \alpha' j}^{\text{inter,CG}}$ from the intermolecular coarse-grained potential and $\mathbf{F}_{\alpha i}^{\text{intra}}$ from the intramolecular potential. The force is given as [93]

$$\mathbf{F}_{\alpha i}^{\text{H-AdResS}} = \sum_{\alpha'} \left(\frac{\lambda_{\alpha} + \lambda_{\alpha'}}{2} \sum_j \mathbf{F}_{\alpha i \alpha' j}^{\text{inter,AA}} + \left(1 - \frac{\lambda_{\alpha} + \lambda_{\alpha'}}{2} \right) \mathbf{F}_{\alpha i \alpha' j}^{\text{inter,CG}} \right) + \mathbf{F}_{\alpha i}^{\text{intra}} - \mathbf{F}_{\alpha i}^{\text{drift}} , \quad (2.11)$$

where the drift force $\mathbf{F}_{\alpha i}^{\text{drift}}$ is given by

$$\mathbf{F}_{\alpha i}^{\text{drift}} = (V_{\alpha}^{\text{inter,AA}} - V_{\alpha}^{\text{inter,CG}}) \nabla_{\alpha i} \lambda_{\alpha} - \left. \frac{dH^{\text{FEC}}(\lambda_{\alpha})}{d\lambda} \right|_{\lambda=\lambda_{\alpha}} . \quad (2.12)$$

The first term of the drift force is the gradient of the switching function weighted with the difference of the potential of coarse-grained and all-atom potential of molecule α . This force term only depends on the position of a single molecule and thus violates both momentum conservation and Newton's third law. Therefore, this term of the drift force induces pressure and density changes in the system. To minimise this drift, the free energy correction $H^{\text{FEC}}(\lambda_{\alpha})$ was introduced in the Hamiltonian (Eq. (2.12)). H^{FEC} is a time-independent function of the switching parameter and thus of the position of a molecule α . The free energy compensation term is calculated with Kirkwood's thermodynamic integration (cf. Ref. [129]). The correction term is measured in a fluid where the switching parameter is constant for all molecules and does not depend on the position.

2.3.2 Per pair (all-atom/coarse-grained)

Ref. [125] was published earlier than H-AdResS (cf. Section 2.3.1) and contains a similar approach. The switching parameter $\lambda_{\alpha\alpha'}$ is calculated per atom pair. Therefore, $V_{\alpha}^{\text{inter,CG}}$ and $V_{\alpha}^{\text{inter,AA}}$ are not given per atom but per pair, i.e., by $V_{\alpha\alpha'}^{\text{inter,CG}}$ and $V_{\alpha\alpha'}^{\text{inter,AA}}$. The total intermolecular potential is given by

$$V^{\text{inter}} = \sum_{\alpha} \left(\sum_{\alpha' > \alpha} \left(\lambda_{\alpha\alpha'} V_{\alpha\alpha'}^{\text{inter,CG}} + (1 - \lambda_{\alpha\alpha'}) \sum_{ij} V_{\alpha i \alpha' j}^{\text{inter,AA}} \right) + H_{\alpha}^{\text{inter,FEC}} \right) , \quad (2.13)$$

where $H_{\alpha}^{\text{inter,FEC}}$ is a correction per atom. In contrast to H-AdResS, the drift force (cf. Eq. (2.12)) that follows from the gradient of the switching function is not calculated but neglected due to the correction term $H_{\alpha}^{\text{inter,FEC}}$. The correction term is calculated at runtime through thermodynamic integration and ensures energy conservation.

Therefore, this method does not require a prior calculation of a correction in a system with constant- λ like H-AdResS. The force calculated from Eq. (2.13) under neglect of the gradient of the switching parameter is, apart from a slightly different calculation of $\lambda_{\alpha\alpha'}$, the force used in AdResS (cf. Eq. (2.7)) [126]. Therefore, the used forces conserve the momentum and Newton's third law applies. In contrast to AdResS, the thermostat is only used in the switching zone between the AA and the CG region. Hence, this approach does not simulate a canonical ensemble. However, as the forces determine the dynamics and the book-keeping of energy-changes does not influence the used forces, this approach also does not simulate a microcanonical ensemble.

2.3.3 Per cluster (quantum mechanics/molecular mechanics)

The permuted adaptive partitioning (PAP) approach suggested in Ref. [100] uses a spherical precise region with $\lambda = 1$, a switching region in which λ decays to 0 and an outer molecular mechanics

region in which $\lambda = 0$ applies. The potential energy of atoms in the precise region is $E_{\text{precise}}^{\text{PAP}}$. One can only compute the total potential energy of a system in a density functional theory (DFT) calculation but no potential energies of single atoms. Adding the atom i in the switching zone to the quantum mechanical calculation gives the energy $E_{\text{precise},i}^{\text{PAP}}$. The potential energy of atom i which interacts quantum mechanically with the spherical precise region is then calculated as

$$E_i^{\text{PAP}} = E_{\text{precise},i}^{\text{PAP}} - E_{\text{precise}}^{\text{PAP}}. \quad (2.14)$$

The quantum mechanical potential energy of the atom pair i, j is calculated analogously as

$$E_{ij}^{\text{PAP}} = E_{\text{precise},ij}^{\text{PAP}} - E_i^{\text{PAP}} - E_j^{\text{PAP}} - E_{\text{precise}}^{\text{PAP}}, \quad (2.15)$$

where $E_{\text{precise},ij}^{\text{PAP}}$ denotes the potential energy of the precise spherical system to which atoms i and j have been added. The by Eqs. (2.14) and (2.15) introduced mechanism to calculate the quantum mechanical potential energy of a cluster of k atoms can be done up an order of N^{PAP} , where N^{PAP} is the number of atoms in the switching zone. The total quantum mechanical energy is then given by weighting the energy contributions with the corresponding switching parameter of the to the precise calculation added atoms, i.e.

$$E_{\text{QM}}^{\text{PAP}} = E_{\text{precise}}^{\text{PAP}} + \sum_{i=1}^{N^{\text{PAP}}} \lambda_i E_i^{\text{PAP}} + \sum_{i=1}^{N^{\text{PAP}}-1} \sum_{j=i+1}^{N^{\text{PAP}}} \lambda_i \lambda_j E_{ij}^{\text{PAP}} + \dots \quad (2.16)$$

where contributions of to the orders 3 to N^{PAP} are indicated by the dots. The energy $E_{\text{QM}}^{\text{PAP}}$ is then used as $E_{\text{QM}}^{\text{precise}}$ in the IMOMM model (cf. Eq. (2.8)) from which the total potential energy of the system is computed. Ref. [100] does not explain the force calculation of PAP¹, but states that PAP is equivalent to the ONIOM-XS method for one atom in the switching zone and ONIOM-XS[130] calculates the forces using the gradient of the switching parameter according to Eq. (2.1). Whether the momentum is conserved depends on the implementation: when the centre of the spherical precise region is just a constant, there is a force on each atom in the switching zone that depends only on the position of this atom relative to the centre of the spherical region without any force that acts on another atom via Newton's third law like discussed in Section 2.3.1 for H-AdResS. When the centre of the spherical precise region is time dependent and calculated only from atomic positions, Newton's third law and momentum conservation apply; in this case this approach also conserves the energy. The combinatoric complexity with $2^{N^{\text{PAP}}}$ QM-calculations for the calculation of Eq. (2.16) is the main disadvantage of this method.

The PAP-approach is adapted in Ref. [126] for a mixed atomistic and coarse-grained system. As the AA/CG-approach uses Lagrangian mechanics, one cannot use a standard symplectic integrator like velocity Verlet[131] but has to use an ad hoc one developed within the approach.

2.4 Conservative approach

The presented coupling approaches are summarised in Table 2.1 and the following discussion is visualised in Fig. 2.1. One can, in principle, reconstruct a Hamiltonian from an adaptive-precision force-approach², in particular for pair potentials. Thus, the forces according to Eqs. (2.7) and (2.11) are similar, apart from the differently calculated switching parameter λ_{ij} and the drift force following from the difference of the approaches. However, as one needs to solve integrals, such a reconstruction of a Hamiltonian is not straightforward and may not be possible at all, in particular for many-body potentials (and forces) that are common for modern ML-potentials. Furthermore, an adaptive switching parameter, that we aim for, complicates the reconstruction of the Hamiltonian even more compared to the time independent switching parameter of H-AdResS

¹Ref. [100] presents a second coupling method that sorts the atoms in the switching zone to improve (but not solve) the poor scaling with the number of atoms in the buffer zone. The force calculation is explained for the second coupling method and includes the gradient of the switching parameter.

²The force on an atom is calculated according to Eq. (2.2), but can be simplified for a pair potential $E^{\text{pair}}(r)$: the potential energy of an atom i is given by $E_i = \frac{1}{2} \sum_j E^{\text{pair}}(r_{ij})$ and the force by $\mathbf{F}_i = \sum_j E^{\text{pair}}(r_{ij}) \frac{\mathbf{r}_{ij}}{r_{ij}}$, where one can calculate the pair force $F^{\text{pair}}(r)$ from the pair potential, i.e. $F^{\text{pair}}(r) = -\frac{\partial E^{\text{pair}}(r)}{\partial r}$, and vice versa the potential energy $E^{\text{pair}}(r) = \int_r^\infty F^{\text{pair}}(r') dr'$ from the pair force. Thus, one can try to reconstruct an adaptive-precision pair energy from an adaptive-precision pair potential like $F^{\text{pair}}(r) = \lambda(r) F^{\text{pair,fast}}(r) + (1 - \lambda(r)) F^{\text{pair,precise}}(r)$ by solving the one-dimensional integral. However, to get a well defined potential energy for use in a MD simulation, one wants an analytical (rather than a numerical) solution of the integral, which might not exist. Furthermore, modern ML-potentials are no pair potentials and thus, one cannot simplify the calculation to a one-dimensional integral.

Table 2.1: Comparison of the presented coupling approaches.

approach section	system	weighting		conservation		ensemble
		E/F	per	energy	momentum	
2.1	AA/AA	E, F	atom	no	no	NVT
2.2.1	AA/AA	F	atom	no	no ^a	μ VT
2.2.2	AA/CG	F	pair	no	yes	μ VT
2.2.3	QM/MM	F	atom	no	no	NVT
2.3.1	AA/CG	E	atom	yes ^b	no	NVE
2.3.2	AA/CG	E	pair	(yes) ^c	yes	none ^d
2.3.3	QM/MM	E	cluster	yes ^e	yes	NVE

^aThe thermodynamic force on an atom depends only on the position of this atom but not on any other atom.

^bThis approach requires a previous simulation with a constant λ .

^cThe tracked energy changes do not influence the trajectory of particles.

^dThe number of particles and the volume are conserved. However, the temperature is not conserved as a thermostat is only used in a subsystem and the energy is only conserved by book-keeping, that does not influence the forces.

^eThe cluster weighting QM/MM coupling method involves the calculation of $2^{N^{\text{PAP}}}$ precise QM systems, that vary slightly, and is, therefore, quite expensive.

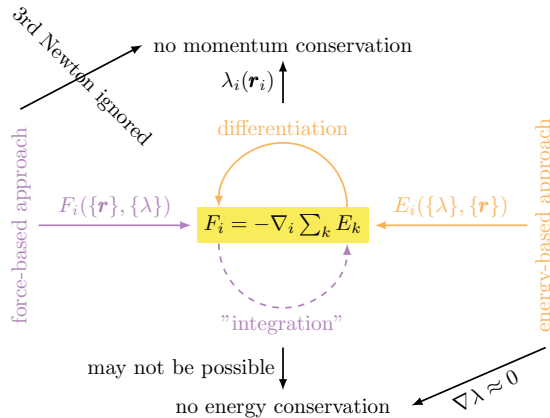


Figure 2.1: Visualisation of the advantages and disadvantages of a force-based and and energy-based coupling approaches as discussed in Section 2.4.

and AdResS. Hence, force-based coupling approaches do not conserve the energy as one cannot simply reconstruct the Hamiltonian from a force ansatz. Instead, these approaches aim for the canonical or grand canonical ensemble (cf. Section 2.2). In contrast, one can, in principle, conserve the energy with energy-based coupling approaches (cf. Section 2.3) as the calculation of the force according to Eq. (2.2) requires only the calculation of a gradient and no integration. The present work aims for a coupling mechanism of two interatomic potentials in a microcanonical ensemble. Therefore, an energy-based approach is used here.

The energy-based approaches weight the energy per atom (cf. Eq. (2.10)), pair (cf. Eq. (2.13)) and cluster (cf. Eq. (2.16)). The weighting per atom i is possible for interatomic potentials as the potential energy E_i can be calculated for every interatomic potential, in contrast to DFT-calculations that provide only the potential energy $\sum_i E_i$ of the whole system.

When the potential energy is given by a pair potential, i.e., $E_i^{\text{pair}} = \sum_j E_{ij}^{\text{pair}}(r_{ij})$, one can introduce a coupling in the potential energy E_{ij}^{pair} of an atom pair like in Eq. (2.13). One has to ensure that one does not describe the atomic interaction of two atoms with both the precise and the fast potential to avoid double counting, i.e., one needs to weight the fast calculated contribution with λ and the precisely calculated contribution with $(1 - \lambda)$. However, only the least precise interatomic potentials are pair-potentials. The most precise potentials are no pair potentials, but the natural candidates for the precise interatomic potential in an AA/AA coupling. Therefore, one cannot identify a summand E_{ij}^{pair} in precise interatomic potentials. Instead, one has to compute a whole subsystem using the precise potential, add an atom and compute the whole subsystem again to isolate the potential energy contributions of the added atom like in Section 2.3.3. Although this is possible, this induces a huge overhead as one needs to compute the potential energy of a subsystem not only once but multiple times. The QM/MM approach with $2^{N^{\text{PAP}}}$ QM-calculations for N^{PAP} atoms in the switching zone could be optimised as one can compute the potential energy of single atoms with an interatomic potential, but a huge administrative and combinatoric overhead would remain.

Weighting the energy per pair is a natural choice for pair-potentials, but impractical for precise many-body potentials. Weighting the energy per cluster requires a huge administrative overhead with multiple calculations of the same atom. Weighting the energy per particle is a natural choice for many-body potentials as they calculate the potential energy per atom and, thus, the approach taken in the present work.

To conserve the momentum and avoid breaking Newton's third law, one has to be cautious when defining the switching parameter. The according to Eq. (2.2) calculated force on an atom in a time-independent switching zone depends due to the gradient of the switching parameter on the position of this atom within the switching region (cf. Eq. (2.12)). The momentum of the considered atom might even change when there are no other atoms. Therefore, an over time constant switching zone violates the momentum conservation. Such a violation of momentum conservation is prevented by an automatically calculated switching parameter which depends on distances r_{ij} between atoms. Thereby, the switching zone is also adaptive to a changed configuration in the system. Hence, an adaptive-precision potential that conserves energy and momentum naturally is given by

$$E_i^{\text{AP}}(\{\mathbf{r}_{ij}\}) = \lambda_i(\{\mathbf{r}_{ij}\})E_i^{(\text{f})}(\{\mathbf{r}_{ij}\}) + (1 - \lambda_i(\{\mathbf{r}_{ij}\}))E_i^{(\text{p})}(\{\mathbf{r}_{ij}\}). \quad (2.17)$$

The adaptive-precision Hamiltonian is then given by Eq. (2.1) and the force is calculated according to Eq. (2.2).

To minimise the in Section 2.2.1 occurred problems due to different definitions of the fast and the precise potential, the fast potentials should be as similar as possible to the precise potential for all cases in which the fast potential is used.

Chapter 3

Adaptive-precision ACE potential

An approach to couple two interatomic potentials of different precision and cost was developed in Chapter 2. Whenever it is used, the fast potential should be as similar as possible to the precise potential. The atomic cluster expansion (ACE)[40] is a precise and modern machine-learning potential. Both precision and computational time of an ACE potential depend on the number of atomic clusters which are expanded. By decreasing this number of clusters of an existing potential and optimising the remaining parameters of the potential for a specific reference environment, one can create a faster potential which depends on the same approach like the precise one. Therefore, two ACE potentials of different precision and cost can serve as an example for the energy-based coupling approach in Eq. (2.17).

Hence, this chapter presents an adaptive-precision ACE(APA) potential which combines a fast ACE potential and a precise ACE potential. First, ACE is described in Section 3.1. In Section 3.2 is shown how one can create a fast ACE potential and calculate the switching parameter λ_i for the coupling. The developed model is then applied to create an APA copper potential in Section 3.3. Section 3.4 concludes this chapter with a discussion of findings and remaining optimisation possibilities of this APA approach.

3.1 Atomic cluster expansion

In this section, the atomic cluster expansion is described according to Refs. [4, 40], starting with the potential energy calculation and followed by the force calculation and notes on the implementation. This description is limited to the aspects of the atomic cluster expansion that are relevant for this dissertation. Therefore, the basis that is used for the expansion depends only on the atomic positions. One can add dependencies on atomic species, magnetism, charge and other variables[132].

3.1.1 Energy

The atomic energy for an atom i according to the ACE-model is given by a general nonlinear embedding function $\mathcal{F}$ which depends on P atomic properties $(\varphi_i^{(1)}, \dots, \varphi_i^{(P)})$, namely

$$E_i^{\text{ACE}} = \mathcal{F}(\varphi_i^{(p)}). \quad (3.1)$$

With a Finnis-Sinclair[133] (FS) approach one gets the energy

$$E_i^{\text{ACE,FS}} = \varphi_i^{(1)} + \sqrt{\varphi_i^{(2)}} \quad (3.2)$$

which is used for the copper (Cu) potential in Ref. [4]. Each atomic property $\varphi_i^{(p)}$ of atom i is expanded in a basis of atomic clusters. To do so, one needs a set of orthogonal and complete pair basis functions $\phi_{nlm}(\mathbf{r})$. The pair-basis functions[40]

$$\phi_{nlm}(\mathbf{r}) = \sqrt{4\pi} R_{nl}(r) Y_l^m(\hat{r}) \quad (3.3)$$

are a combination of a radial basis function $R_{nl}(r)$ and a spherical harmonics $Y_l^m(\hat{r})$, where $\hat{r} = \mathbf{r}/r$ is a unit vector. The atomic base A_{inlm} is defined as the projection of the pair basis functions ϕ_{nlm} on the atomic density and given as[40]

$$A_{inlm} = \sum_j \phi_{nlm}(\mathbf{r}_{ji}), \quad (3.4)$$

where j denotes neighbouring atoms and $\mathbf{r}_{ji} = \mathbf{r}_j - \mathbf{r}_i$ the distance vector between two atoms. One can define the many-body basis functions $A_{inlm}^{(K)}$ as[4]

$$A_{inlm}^{(K)} = \prod_{k=1}^{(K)} A_{in_k l_k m_k}, \quad (3.5)$$

where the indices $\mathbf{n} = (n_1, \dots, n_K)$, $\mathbf{l} = (l_1, \dots, l_K)$ and $\mathbf{m} = (m_1, \dots, m_K)$ describe which atomic basis functions are multiplied. A quantity $\varphi_i^{(p)}$ expanded in these many-body basis functions is then given as

$$\varphi_i^{(p,A)} = \sum_{Knlm} \tilde{c}_{nlm}^{(p,K)} A_{inlm}^{(K)}, \quad (3.6)$$

where $\tilde{c}_{nlm}^{(p,K)}$ are expansion coefficients. The atomic energy is in the absence of an external field invariant with respect to rotation of the global coordinate system. For the expansion in $A_{inlm}^{(K)}$ Eq. (3.6) the expansion coefficients $\tilde{c}_{nlm}^{(p,K)}$ need to ensure rotational invariance and are therefore no free model parameters. To include the rotational invariance in the base, the many-body basis functions are combined to cluster basis functions $B_{inl}^{(K)}$. A cluster basis function is given as

$$B_{inl}^{(K)} = \sum_{\mathbf{m}} c_{\text{rot},\mathbf{m}l}^{(K)} A_{inlm}^{(K)}, \quad (3.7)$$

where $c_{\text{rot},\mathbf{m}l}^{(K)}$ are generalised Clebsch-Gordan coefficients. The calculation of $c_{\text{rot},\mathbf{m}l}^{(K)}$ depends on K . The limits of the sum over $\mathbf{m} = (m_1, \dots, m_K)$ depend on $\mathbf{l}$ and K . The reader is referred to Ref. [40] for details of both sum limits and the calculation of the rotation coefficient. To make it a little less abstract, for $K = 3$ applies

$$B_{inl}^{(3)} = \sum_{m_1=-l_1}^{l_1} \sum_{m_2=-l_2}^{l_2} \sum_{m_3=-l_3}^{l_3} c_{\text{rot},m_1 m_2 m_3 l_1 l_2 l_3}^{(3)} \prod_{k=1}^3 A_{in_k l_k m_k}, \quad (3.8)$$

where

$$c_{\text{rot},m_1 m_2 m_3 l_1 l_2 l_3}^{(3)} = \begin{pmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \quad (3.9)$$

are Wigner $3j$ symbols. A quantity $\varphi_i^{(p)}$ expanded in cluster basis functions is given as

$$\varphi_i^{(p,B)} = \sum_{Knl} c_{nl}^{(p,K)} B_{inl}^{(K)}, \quad (3.10)$$

where $c_{nl}^{(p,K)}$ are independent expansion coefficients that are fitted to training data[134]. The cluster basis functions fulfil fundamental translation, rotation, inversion, and permutation invariance[135].

3.1.2 Force

When the energy E_i is described by an atomic cluster expansion (cf. Eq. (3.1)) in the cluster basis functions $B_{inl}^{(K)}$ (cf. Eq. (3.10)), the force on atom k is calculated according to Eq. (2.2) as

$$\begin{aligned} \mathbf{F}_k^{\text{ACE,B}} &= - \sum_i \nabla_k \mathcal{F}(\varphi_i^{(p,B)}) \\ &= - \sum_i \sum_p \frac{\partial \mathcal{F}}{\partial \varphi_i^{(p,B)}} \sum_{Knl} c_{nl}^{(p,K)} \sum_{nlm} \frac{\partial B_{inl}^{(K)}}{\partial A_{inlm}} \nabla_k A_{inlm} \\ &= - \sum_i \sum_{nlm} \omega_{inlm}^{\text{B}} \nabla_k A_{inlm}, \end{aligned} \quad (3.11)$$

where we shorten the equation by introducing

$$\omega_{inlm}^{\text{B}} = \sum_p \frac{\partial \mathcal{F}}{\partial \varphi_i^{(p,B)}} \sum_{Knl} c_{nl}^{(p,K)} \frac{\partial B_{inl}^{(K)}}{\partial A_{inlm}}. \quad (3.12)$$

One can calculate the force in a similar way when the many-body functions $A_{inlm}^{(K)}$ are used as basis for the atomic cluster expansion, namely as,

$$\mathbf{F}_k^{\text{ACE,A}} = - \sum_i \sum_{nlm} \omega_{inlm}^{\text{A}} \nabla_k A_{inlm}, \quad (3.13)$$

where

$$\omega_{inlm}^A = \sum_p \frac{\partial \mathcal{F}}{\partial \varphi_i^{(p,A)}} \sum_{Knlm} \tilde{c}_{nlm}^{(p,K)} \prod_{k=1, k \neq t}^K A_{in_k t_k m_k} \delta_{nn_t} \delta_{ll_t} \delta_{mm_t}. \quad (3.14)$$

As Eqs. (3.11) and (3.13) have the same form and just use another ω_{inlm}^B dependent on the basis $\mathcal{B}$ used for atomic cluster expansion, one can write the force according to the ACE model as

$$\mathbf{F}_k^{\text{ACE}, \mathcal{B}} = - \sum_i \sum_{nlm} \omega_{inlm}^B \nabla_k A_{inlm}. \quad (3.15)$$

The gradient of the atomic base is required to calculate Eq. (3.15) and given as

$$\begin{aligned} \nabla_k A_{inlm} &= \nabla_k \sum_j \phi_{nlm}(\mathbf{r}_{ji}) \\ &= \sum_j \nabla_j \phi_{nlm}(\mathbf{r}_{ji}) \delta_{kj} + \sum_j \nabla_i \phi_{nlm}(\mathbf{r}_{ji}) \delta_{ki} \\ &= \nabla_k \phi_{nlm}(\mathbf{r}_{ki}) - \sum_j \nabla_j \phi_{nlm}(\mathbf{r}_{ji}) \delta_{ki}, \end{aligned} \quad (3.16)$$

where δ_{ij} is the Kronecker δ and

$$\nabla_i \phi_{nlm}(\mathbf{r}_{ji}) = -\nabla_j \phi_{nlm}(\mathbf{r}_{ji}) \quad (3.17)$$

is used. Using the gradient of the atomic base Eq. (3.16) allows to write the atomic force Eq. (3.15) as

$$\begin{aligned} \mathbf{F}_k^{\text{ACE}, \mathcal{B}} &= - \sum_i \sum_{nlm} \omega_{inlm}^B \nabla_k \phi_{nlm}(\mathbf{r}_{ki}) + \sum_i \sum_{nlm} \sum_j \omega_{inlm}^B \nabla_j \phi_{nlm}(\mathbf{r}_{ji}) \delta_{ki} \\ &= - \sum_j \sum_{nlm} \omega_{jnml}^B \nabla_k \phi_{nlm}(\mathbf{r}_{kj}) + \sum_{nlm} \sum_j \omega_{knlm}^B \nabla_j \phi_{nlm}(\mathbf{r}_{jk}). \end{aligned} \quad (3.18)$$

Introducing the force component

$$\mathbf{f}_{ki}^{\mathcal{B}, \text{ACE}} = \sum_{nlm} \omega_{inlm}^B \nabla_k \phi_{nlm}(\mathbf{r}_{ki}) \quad (3.19)$$

allows to write the force Eq. (3.18) as

$$\mathbf{F}_k^{\text{ACE}, \mathcal{B}} = \sum_i \left(\mathbf{f}_{ik}^{\mathcal{B}, \text{ACE}} - \mathbf{f}_{ki}^{\mathcal{B}, \text{ACE}} \right). \quad (3.20)$$

3.1.3 Implementation

The atomic cluster expansion is implemented in LAMMPS[1] as presented in [4, 135]. The computation of $\varphi_i^{(p)}$ is faster when it is expanded in $A_{inlm}^{(K)}$ Eq. (3.6) than in $B_{int}^{(K)}$ Eq. (3.10) as less sum evaluations are required[134]. Therefore, the atomic cluster expansion was in LAMMPS initially only implemented in the basis of the many-body functions $A_{inlm}^{(K)}$ [4]. The atomic cluster expansion in the basis of the cluster basis functions was added later with the introduction of an active learning strategy for ACE[135].

In LAMMPS, one computes the atomic cluster expansion for one atom after the other. The computation for an atom i includes the computation of its potential energy E_i^{ACE} and the force components $\mathbf{f}_{ki}^{\mathcal{B}, \text{ACE}}$ for all neighbouring atoms k . Thus, the force $\mathbf{F}_i^{\text{ACE}, \mathcal{B}}$ is not computed directly with the energy, but added up through the iteration over all atoms as shown in Algorithm 1.

3.1.4 Fitting an ACE potential

The tool `pacemaker`[134] allows to fit an ACE potential. The parameters of an ACE potential are fitted by minimising a loss function $\mathcal{L}$. Density functional theory (DFT) calculations of many different structures are used as reference to fit a precise ACE potential. The DFT calculation of a structure provides the total potential energy $E_{\text{struct}, s}^{\text{targ}}$ of this structure s and the forces $\mathbf{F}_{si}^{\text{targ}}$ for all N_s^{atoms} atoms i of structure s . This target quantities are compared with $E_{\text{struct}, s}^{\text{pred}}$ and $\mathbf{F}_{si}^{\text{targ}}$ which are calculated using the current parameters of the ACE potential for the same structure s . The

Algorithm 1 Computation of the ACE energies E_i^{ACE} and forces $\mathbf{F}_i^{\text{ACE},\mathcal{B}}$ for all atoms i according to Eqs. (3.1) and (3.20).

```

for all atoms  $i$  do
   $\mathbf{F}[i] \leftarrow \mathbf{0}$ 
end for
for all atoms  $i$  do
  compute atom  $i$  including  $E_i^{\text{ACE}}$  and  $\mathbf{f}_{ki}^{\mathcal{B},\text{ACE}}$  ▷ The computation takes place here.
   $E[i] \leftarrow E_i^{\text{ACE}}$  ▷ previously computed according to Eq. (3.1)
  for all neighbours  $k$  of  $i$  do
     $\mathbf{f} \leftarrow \mathbf{f}_{ki}^{\mathcal{B},\text{ACE}}$  ▷ previously computed according to Eq. (3.19)
     $\mathbf{F}[k] -= \mathbf{f}$  ▷ cf. Eq. (3.20)
     $\mathbf{F}[i] += \mathbf{f}$  ▷ cf. Eq. (3.20)
  end for
end for

```

loss function is minimised by adjusting the parameters of the ACE potential. The loss function implemented in **pacemaker** is

$$\begin{aligned} \mathcal{L}_{\text{ace}}^{\text{DFT}} = & (1 - \kappa) \sum_{s=1}^{N_{\text{struct}}} w_s^{\text{E}} \left(\frac{E_{\text{struct},s}^{\text{pred}} - E_{\text{struct},s}^{\text{targ}}}{N_s^{\text{atoms}}} \right)^2 \\ & + \kappa \sum_{s=1}^{N_{\text{struct}}} \sum_{i=1}^{N_s^{\text{atoms}}} w_{si}^{\text{F}} \left(\mathbf{F}_{si}^{\text{pred}} - \mathbf{F}_{si}^{\text{targ}} \right)^2 + \Delta_{\text{coeff}} + \Delta_{\text{rad}}, \end{aligned} \quad (3.21)$$

where w_s^{E} , w_{si}^{F} and κ are weights per structure, atom and quantity, N_{struct} denotes the number of structures and Δ_{coeff} and Δ_{rad} are penalty terms which are dependent on the current values of the parameters and explained in Ref. [134]. The weights per atom and structure are normalised through

$$1 = \sum_{s=1}^{N_{\text{struct}}} w_s^{\text{E}} = \sum_{s=1}^{N_{\text{struct}}} \sum_{i=1}^{N_s^{\text{atoms}}} w_{si}^{\text{F}}. \quad (3.22)$$

Therefore, one can use the relative force weight $\kappa \in [0, 1]$ in the loss function independent of the number of used structures N_{struct} . The simplest way to fulfil the weight norm Eq. (3.22) is with a uniform (U) weighting policy, i.e., $w_s^{\text{E,U}} = 1/N_{\text{struct}}$ and $w_{si}^{\text{F,U}} = w_s^{\text{E,U}}/N_s^{\text{atoms}}$. The other in **pacemaker** implemented weighting policy are energy-based (EB) weights. Overweighting of the ground state is prevented by using the energy-based per structure weights, which are given as

$$w_s^{\text{E,EB}} = w_{\text{norm}}^{\text{E}} / \left(\frac{E_{\text{struct},s}^{\text{targ}}}{N_s^{\text{atoms}}} - E_{\text{struct,min}}^{\text{targ}} + \Delta^{\text{E}} \right)^2, \quad (3.23)$$

where the constant $w_{\text{norm}}^{\text{E}}$ ensures that Eq. (3.22) applies, $\Delta^{\text{E}} \approx 1 \text{ eV}$ is a positive constant and

$$E_{\text{struct,min}}^{\text{targ}} = \min \left(\frac{E_{\text{struct},s}^{\text{targ}}}{N_s^{\text{atoms}}} \right) \quad (3.24)$$

approximates the ground state. Overweighting the force of atoms with zero force is prevented in a similar way, namely by using

$$w_{si}^{\text{F,EB}} = w_{\text{norm}}^{\text{F}} w_s^{\text{E,EB}} / ((\mathbf{F}_{si}^{\text{targ}})^2 + \Delta^{\text{F}}), \quad (3.25)$$

where the constant $w_{\text{norm}}^{\text{F}}$ ensures that Eq. (3.22) applies and $\Delta^{\text{F}} \approx 1 \text{ eV}^2 \text{ \AA}^{-2}$ is a positive constant. One can automatically calculate the relative force-weight κ by using the variance σ^2 of the reference data, namely

$$\kappa^{\text{DFT}} = \sigma^2(E_{\text{struct},s}^{\text{targ}}/N_s^{\text{atoms}}) / (\sigma^2(E_{\text{struct},s}^{\text{targ}}/N_s^{\text{atoms}}) + \sigma^2(\mathbf{F}_{si}^{\text{targ}})). \quad (3.26)$$

3.2 Adaptive-precision ACE model

A precise ACE potential is fitted to a broad range of structures and is thus also applicable for this broad range of structures. However, to achieve a good precision for all of these structure one needs hundreds of basis functions for one potential[4]. Therefore, a precise ACE potential is expensive to compute. The evaluation speed of an ACE potential depends on the number of used cluster basis functions. Thus, one can create a fast ACE potentials by using far less basis functions. By simply reducing the number of basis functions one would only reduce the precision of the potential. However, by narrowing the broad range of reference structures, one can achieve a good precision in the vicinity of a reference atomic environment. One can for example create a fast ACE copper potential which is precise for face-centred cubic (fcc) copper at room temperature. This fast ACE potential can be used to speedup simulations of the precise ACE potential: one can use the fast potential for atoms which are well described by it and use the precise but expensive ACE potential only for the remaining atoms. Therefore, one needs a detection mechanism which measures for each atom the deviation of its atomic environment from the reference configuration to which the fast potential is fitted to ensure that the fast potential is only used for appropriate atoms.

We assume the existence of two ACE potentials here, where the precise (p) one has much more basis functions than the fast (f) ACE potential which only works in the vicinity of a known reference environment. The combination of the potentials is made per atom according to the energy-based approach in Eq. (2.17), i.e.

$$E_i^{\text{APA}} = \lambda_i^{\text{B}} E_i^{\text{ACE,f}} + (1 - \lambda_i^{\text{B}}) E_i^{\text{ACE,p}}, \quad (3.27)$$

where $E_i^{\text{ACE,f}}$ and $E_i^{\text{ACE,p}}$ are the potential energy according to the fast and the precise ACE potential and $\lambda_i^{\text{B}} \in [0, 1]$ is an auxiliary function that decides based on a detection mechanism which precision is used on an atom i .

The creation of a fast ACE potential suited for one reference environment is described in Section 3.2.1. The detection mechanism which measures the deviation of an atomic environment from the reference configuration of the fast potential is described in Section 3.2.2. The switching parameter λ_i^{B} is then described in Section 3.2.3. Afterwards, the calculation of energies and forces is discussed in Section 3.2.4. The implementation of energy and force calculation of an adaptive-precision ACE potential is presented in Section 3.2.5.

3.2.1 Fitting a fast ACE potential

To fit a fast ACE potential which is similar to a precise ACE potential in the vicinity of a reference atomic environment, one needs reference data to which one can fit the fast ACE potential. To make the fast potential and the precise one as similar as possible in the reference environment, one should not fit the fast ACE potential to DFT reference data, but to reference data calculated directly with the precise ACE potential.

In order to fit the most accurate ACE potential possible, one needs the most precise reference data, i.e., one needs DFT reference data. Hence, only DFT data are normally used as reference data to fit an ACE potential. DFT reference calculations provide only the total potential energy of a structure s . A molecular dynamics (MD) simulation provides also the atomic energy E_i^{ACE} for all atoms i .

The gradient of an energy surface is described by the forces. Under the assumption that the gradients are known everywhere, knowing the value of the energy surface at one point, i.e., per structure, is enough to correctly adjust the energy of this surface. However, if the gradient, i.e., the forces, is not known everywhere it needs to be interpolated between known values, where this interpolation reduces the precision. Thus, providing the energies per atom rather than per structure increases the energy precision as less interpolations of the forces are required.

An ACE potential is fitted to DFT reference data, as described in Section 3.1.4, using the loss function $\mathcal{L}_{\text{ace}}^{\text{DFT}}$ according to Eq. (3.21). $\mathcal{L}_{\text{ace}}^{\text{DFT}}$ only includes the reference energy $E_{\text{struct},s}^{\text{targ}}$ of a whole structure s . However, as the reference energy E_{si}^{targ} is available in MD calculations, one can use E_{si}^{targ} instead of $E_{\text{struct},s}^{\text{targ}}$ to increase the quality of the reference data and thus the precision of the fit. One can calculate E_{si}^{targ} in a MD simulation with the precise ACE potential of few thousand atoms at the target temperature. Therefore, one can fit the fast ACE potential to MD data simulated with the precise ACE potential at the target temperature of the reference environment.

Including the atomic energy in the loss function $\mathcal{L}_{\text{ace}}^{\text{DFT}}$ Eq. (3.21) gives the loss function

$$\begin{aligned} \mathcal{L}_{\text{ace}}^{\text{MD}} = & (1 - \kappa) \sum_{s=1}^{N_{\text{struct}}} \sum_{i=1}^{N_s^{\text{atoms}}} w_{si}^{\text{E}} \left(E_{si}^{\text{pred}} - E_{si}^{\text{targ}} \right)^2 \\ & + \kappa \sum_{s=1}^{N_{\text{struct}}} \sum_{i=1}^{N_s^{\text{atoms}}} w_{si}^{\text{F}} \left(\mathbf{F}_{si}^{\text{pred}} - \mathbf{F}_{si}^{\text{targ}} \right)^2 + \Delta_{\text{coeff}} + \Delta_{\text{rad}}, \end{aligned} \quad (3.28)$$

where w_{si}^{E} denotes a weight per force which is normalised according to

$$1 = \sum_{s=1}^{N_{\text{struct}}} \sum_{i=1}^{N_s^{\text{atoms}}} w_{si}^{\text{E}}. \quad (3.29)$$

The energy-based weight $w_{si}^{\text{E,EB}}$ is adapted from Eq. (3.23) and given as

$$w_{si}^{\text{E,EB}} = w_{\text{norm}}^{\text{E,MD}} w_s^{\text{E,EB}} / \left(E_{si}^{\text{targ}} - E_{\min}^{\text{targ}} + \Delta^{\text{E}} \right)^2, \quad (3.30)$$

where the constant $w_{\text{norm}}^{\text{E,MD}}$ ensures that Eq. (3.29) applies, $w_s^{\text{E,EB}}$ is given according to Eq. (3.23) and

$$E_{\min}^{\text{targ}} = \min(E_{si}^{\text{targ}}) \quad (3.31)$$

approximates the ground state similar to Eq. (3.24). The automatically calculated relative force weight can be calculated using the variance of the atomic energy, namely

$$\kappa^{\text{MD}} = \sigma^2(E_{si}^{\text{targ}}) / \left(\sigma^2(E_{si}^{\text{targ}}) + \sigma^2(\mathbf{F}_{si}) \right). \quad (3.32)$$

3.2.2 Detection mechanism

Just like a cluster expanded property, the deviation Δ_i^{B} of an atom from the reference configuration, to which the fast potential has been fitted, must not change due to translation and rotation of the global coordinate system. Therefore, the cluster basis functions $B_{i\mathbf{n}^i\mathbf{t}^i}^{(K^i)}$ of the fast ACE potential are a natural choice to define Δ_i^{B} . Thereby one introduces overhead as the evaluation of a cluster expanded property in the $A_{i\mathbf{n}^i\mathbf{t}^i\mathbf{m}^i}^{(K^i)}$ many-body basis is faster than in the $B_{i\mathbf{n}^i\mathbf{t}^i}^{(K^i)}$ cluster basis function basis. We define the deviation from the reference configuration as

$$\Delta_i^{\text{B}} = \sum_{K^i\mathbf{n}^i\mathbf{t}^i} w_{\mathbf{n}^i\mathbf{t}^i}^{(K^i,\text{B})} \left(B_{i\mathbf{n}^i\mathbf{t}^i}^{(K^i)} - B_{\mathbf{n}^i\mathbf{t}^i}^{(K^i,\text{ref})} \right)^2, \quad (3.33)$$

where $B_{\mathbf{n}^i\mathbf{t}^i}^{(K^i,\text{ref})}$ are the reference values of the cluster basis function in the reference environment to which the fast ACE potential is fitted and $w_{\mathbf{n}^i\mathbf{t}^i}^{(K^i,\text{B})}$ are weights. Because atoms thermally fluctuate at finite temperatures, so do the cluster basis functions, where the magnitude of these fluctuations is not the same for all cluster basis functions. Therefore, one needs the weights, which were introduced in Eq. (3.33). One needs to compensate for the different magnitudes of the fluctuations of the cluster basis functions. This magnitude of the fluctuation can be measured by the standard deviation (std) of the corresponding cluster basis function at the temperature T^{ref} at which one wants to perform a simulation. One can measure this standard deviation in multiple timesteps of a small system of few thousand atoms at the temperature T^{ref} . The weights are defined as

$$w_{\mathbf{n}^i\mathbf{t}^i}^{(K^i,\text{B})} = \frac{w^{\text{B}}}{\text{std}(B_{i\mathbf{n}^i\mathbf{t}^i}^{(K^i)}) \Big|_{T=T^{\text{ref}}}}, \quad (3.34)$$

where w^{B} is a scaling factor which is calculated according to

$$1 = \sum_i \Delta_i^{\text{B}} \Big|_{T=T^{\text{ref}}} \quad (3.35)$$

for the data that are used to calculate the standard deviation. Thus, the weights are calculated with data measured in a single small reference calculation. The expected deviation Δ_i^{B} is 1 due to Eq. (3.35). Therefore, $\Delta_i^{\text{B}} \gg 1$ indicates that this atom i is in an environment that is not similar to the reference environment and should not be described with the fast ACE potential. Using the variance instead of the standard deviation in Eq. (3.34) would be the natural choice as the squared differences of cluster basis functions are used in Eq. (3.33), but the variance has proven to be less precise as shown in Appendix A.

3.2.3 Switching parameter

The deviation Δ_i^B from the reference configuration can be used to calculate the switching parameter λ_i^B as

$$\lambda_i^B = f^{(\text{trans})}(\Delta_i^B, \Delta_{\text{low}}^B, \Delta_{\text{high}}^B), \quad (3.36)$$

where Δ_{low}^B and Δ_{high}^B are thresholds and the transition function $f^{(\text{trans})}$ is given by

$$f^{(\text{trans})}(x, x_{\text{lo}}, x_{\text{hi}}) = \begin{cases} 1 & \text{for } x \leq x_{\text{lo}}, \\ 0.5 + 0.9375 \frac{x_{\text{hi}} + x_{\text{lo}} - 2x}{x_{\text{hi}} - x_{\text{lo}}} - 0.625 \left(\frac{x_{\text{hi}} + x_{\text{lo}} - 2x}{x_{\text{hi}} - x_{\text{lo}}} \right)^3 & \text{for } x_{\text{lo}} < x < x_{\text{hi}}, \\ + 0.1875 \left(\frac{x_{\text{hi}} + x_{\text{lo}} - 2x}{x_{\text{hi}} - x_{\text{lo}}} \right)^5 & \\ 0 & \text{for } x_{\text{hi}} \leq x, \end{cases} \quad (3.37)$$

$f^{(\text{trans})}$ is taken from Ref.[134] as the first and second derivative of $f^{(\text{trans})}$ are smooth at x_{lo} and x_{hi} . $\lambda_i^B = 1$ applies for all atoms with $\Delta_i^B \leq \Delta_{\text{low}}^B$ which can be described with the fast ACE potential. The use of a continuous switching function for $\Delta_i^B \in (\Delta_{\text{low}}^B, \Delta_{\text{high}}^B)$ allows a smooth transition from the fast description with $\lambda_i^B = 1$ to the precise one with $\lambda_i^B = 0$ for all atoms with $\Delta_i^B \geq \Delta_{\text{high}}^B$ and vice versa for all atoms.

3.2.4 Adaptive-precision ACE equations

The adaptive-precision ACE energy E_i^{APA} is given according to Eq. (3.27) as $E_i^{\text{APA}} = \lambda_i^B E_i^{\text{ACE,f}} + (1 - \lambda_i^B) E_i^{\text{ACE,p}}$. The calculation of E_i^{APA} implies the calculation of the switching parameter λ_i^B Eq. (3.36) with the deviation Δ_i^B Eq. (3.33) from the reference configuration. As the calculation of the deviation of the reference configuration requires the cluster basis functions of the fast potential, this basis is also used to calculate the potential energy $E_i^{\text{ACE,f}}$ of the fast potential. In contrast, the many-body function basis is used for the calculation of the precise energy $E_i^{\text{ACE,p}}$ as this basis allows a faster computation. The adaptive-precision ACE force $F_k^{\text{APA}} = -\nabla_k \sum_i E_i^{\text{APA}}$ is then calculated similar to the ACE force in Eqs. (3.11) and (3.13) as

$$\begin{aligned} F_k^{\text{APA}} &= -\nabla_k \sum_i \left(\lambda_i^B E_i^{\text{ACE,f}} + (1 - \lambda_i^B) E_i^{\text{ACE,p}} \right) \\ &= -\sum_i \left(\left(E_i^{\text{ACE,f}} - E_i^{\text{ACE,p}} \right) (\nabla_k \lambda_i^B) + \lambda_i^B (\nabla_k E_i^{\text{ACE,f}}) + (1 - \lambda_i^B) (\nabla_k E_i^{\text{ACE,p}}) \right) \\ &= -\sum_i \left(\left(E_i^{\text{ACE,f}} - E_i^{\text{ACE,p}} \right) \frac{\partial \lambda_i^B}{\partial \Delta_i^B} \sum_{K^\dagger \mathbf{n}^\dagger l^\dagger m^\dagger} \frac{\partial \Delta_i^B}{\partial B_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{(K^\dagger)}} \sum_{\mathbf{n}^\dagger l^\dagger m^\dagger} \frac{\partial B_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{(K^\dagger)}}{\partial A_{\mathbf{in}^\dagger l^\dagger m^\dagger}} \nabla_k A_{\mathbf{in}^\dagger l^\dagger m^\dagger} \right. \\ &\quad \left. + \sum_{\mathbf{n}^\dagger l^\dagger m^\dagger} \lambda_i^B \omega_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{\text{B,f}} \nabla_k A_{\mathbf{in}^\dagger l^\dagger m^\dagger} + \sum_{\mathbf{n}^\dagger l^\dagger m^\dagger} (1 - \lambda_i^B) \omega_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{\text{A,p}} \nabla_k A_{\mathbf{in}^\dagger l^\dagger m^\dagger} \right) \\ &= -\sum_i \left(\sum_{\mathbf{n}^\dagger l^\dagger m^\dagger} \left(\left(E_i^{\text{ACE,f}} - E_i^{\text{ACE,p}} \right) \omega_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{\text{L}} + \lambda_i^B \omega_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{\text{B,f}} \right) \nabla_k A_{\mathbf{in}^\dagger l^\dagger m^\dagger} \right. \\ &\quad \left. + \sum_{\mathbf{n}^\dagger l^\dagger m^\dagger} (1 - \lambda_i^B) \omega_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{\text{A,p}} \nabla_k A_{\mathbf{in}^\dagger l^\dagger m^\dagger} \right), \end{aligned} \quad (3.38)$$

where the factor

$$\omega_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{\text{L}} = \frac{\partial \lambda_i^B}{\partial \Delta_i^B} \sum_{K^\dagger \mathbf{n}^\dagger l^\dagger m^\dagger} \frac{\partial \Delta_i^B}{\partial B_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{(K^\dagger)}} \frac{\partial B_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{(K^\dagger)}}{\partial A_{\mathbf{in}^\dagger l^\dagger m^\dagger}} \quad (3.39)$$

is introduced to shorten the equation. To calculate $\omega_{\mathbf{in}^\dagger l^\dagger m^\dagger}^{\text{L}}$ according to Eq. (3.39), one needs

$$\frac{\partial \lambda_i^B}{\partial \Delta_i^B} = \begin{cases} 0 & \text{for } \lambda_i^B \in \{0, 1\}, \\ \frac{1.875}{\Delta_{\text{high}}^B - \Delta_{\text{low}}^B} \left(-1 + \left(1 - 2 \frac{\Delta_i^B - \Delta_{\text{low}}^B}{\Delta_{\text{high}}^B - \Delta_{\text{low}}^B} \right)^2 \right. & \text{for } \lambda_i^B \in (0, 1), \\ \left. - \left(1 - 2 \frac{\Delta_i^B - \Delta_{\text{low}}^B}{\Delta_{\text{high}}^B - \Delta_{\text{low}}^B} \right)^4 \right) & \end{cases} \quad (3.40)$$

which is calculated according to Eqs. (3.36) and (3.37) and

$$\frac{\partial \Delta_i^B}{\partial B_{in'l^f m^f}^{(K^f)}} = 2w_{n'l^f}^{(K^f, B)} \left(B_{in'l^f m^f}^{(K^f)} - B_{n'l^f}^{(K^f, \text{ref})} \right). \quad (3.41)$$

which is calculated according to Eq. (3.33). The adaptive-precision ACE force according to Eq. (3.38) can be further calculated by inserting $\nabla_k A_{inlm}$ according to Eq. (3.16). The thereby introduced sums over neighbouring atoms are solved according to Eq. (3.18), i.e., like in the calculation of the ACE-force. The described calculation is used to define

$$\begin{aligned} \mathbf{f}_{ki}^{\text{APA}} &= \left(E_i^{\text{ACE, f}} - E_i^{\text{ACE, p}} \right) \sum_{n^f l^f m^f} \omega_{in^f l^f m^f}^L \nabla_k \phi_{n^f l^f m^f}(\mathbf{r}_{ki}) + \lambda_i^B \sum_{n^f l^f m^f} \omega_{in^f l^f m^f}^{B, f} \nabla_k \phi_{n^f l^f m^f}(\mathbf{r}_{ki}) \\ &\quad + (1 - \lambda_i^B) \sum_{n^p l^p m^p} \omega_{in^p l^p m^p}^{A, p} \nabla_k \phi_{n^p l^p m^p}(\mathbf{r}_{ki}) \\ &= \left(E_i^{\text{ACE, f}} - E_i^{\text{ACE, p}} \right) \mathbf{f}_{ki}^{L, f} + \lambda_i^B \mathbf{f}_{ki}^{B, f} + (1 - \lambda_i^B) \mathbf{f}_{ki}^{A, p}, \end{aligned} \quad (3.42)$$

where

$$\mathbf{f}_{ki}^{L, f} = \sum_{n^f l^f m^f} \omega_{in^f l^f m^f}^L \nabla_k \phi_{n^f l^f m^f}(\mathbf{r}_{ki}). \quad (3.43)$$

is introduced analogue to Eq. (3.19) in the ACE-force calculation. Note, that $\mathbf{f}_{ki}^{B, f}$ and $\mathbf{f}_{ki}^{A, p}$ are given according to Eq. (3.19) for the fast atomic cluster expansion in $B_{in'l^f m^f}^{(K^f)}$ and for the precise atomic cluster expansion in $A_{in^p l^p m^p}^{(K^p)}$. The adaptive-precision ACE (APA) force is then given as

$$\mathbf{F}_k^{\text{APA}} = \sum_i (\mathbf{f}_{ik}^{\text{APA}} - \mathbf{f}_{ki}^{\text{APA}}). \quad (3.44)$$

3.2.5 Implementation

The aim is to save computation time through the simplification of the computation for all atoms i which are in an environment similar to the reference environment to which the fast potential is fitted. Therefore, it is crucial to prevent the computation of the precise atomic cluster expansion for atoms that do not need it. The adaptive-precision ACE energy is calculated according to Eq. (3.27) as $E_i^{\text{APA}} = \lambda_i^B E_i^{\text{ACE, f}} + (1 - \lambda_i^B) E_i^{\text{ACE, p}}$. Therefore, one needs to compute λ_i^B for an atom i to decide afterwards which atomic energy is required. The precise energy $E_i^{\text{ACE, p}}$ is not required for $E_i^{\text{APA}}|_{\lambda_i^B=1} = E_i^{\text{ACE, f}}$, i.e., $E_i^{\text{ACE, p}}$ does not contribute to the total energy of the system for $\lambda_i^B = 1$. Therefore, the computation of the precise potential is also not required to obtain the atomic forces, i.e., one can skip the calculation of the precise atomic cluster expansion for all atoms i with $\lambda_i^B = 1$. The latter is also visible in the equations: $\lambda_i^B = 1$ implies according to Eq. (3.40) $\frac{\partial \lambda_i^B}{\partial \Delta_i^B}|_{\lambda_i^B=1} = 0$ which implies $\omega_{in^f l^f m^f}^L|_{\lambda_i^B=1} = 0$. Thus, the force contribution $\mathbf{f}_{ki}^{\text{APA}}|_{\lambda_i^B=1}$ according to Eq. (3.42) simplifies to $\mathbf{f}_{ki}^{\text{APA}}|_{\lambda_i^B=1} = \mathbf{f}_{ki}^{B, f}$. Similarly, one only needs the precise atomic cluster expansion for atoms i with $\lambda_i^B = 0$ as $E_i^{\text{APA}}|_{\lambda_i^B=0} = E_i^{\text{ACE, p}}$ and $\mathbf{f}_{ki}^{\text{APA}}|_{\lambda_i^B=0} = \mathbf{f}_{ki}^{A, p}$ apply. Therefore, one only needs to compute both atomic cluster expansions for atoms i with $\lambda_i^B \in (0, 1)$. Hence, case distinctions depending on λ_i^B avoid unrequired computations as shown in Algorithm 2.

The computation of the precise atomic cluster expansion for an atom i gives $\mathbf{f}_{ki}^{A, p}$ and $E_i^{\text{ACE, p}}$ which can be weighted accordingly afterwards to compute the adaptive-precision ACE energy and forces. Therefore, one can use the existing ACE code for the computation of the precise atomic cluster expansion. However, the code to compute the fast atomic cluster expansion in the cluster basis functions $B_{in'l^f m^f}^{(K^f)}$ needs to be extended as the switching parameter λ_i^B also depends on the cluster basis functions. Once the cluster basis functions $B_{in'l^f m^f}^{(K^f)}$ are computed for an atom i , one can compute λ_i^B according to Eqs. (3.33) and (3.36). $\omega_{in^f l^f m^f}^L$ and $\omega_{in^f l^f m^f}^{B, f}$ can be computed simultaneously as both depend on $\frac{\partial B_{inlm}^{(K)}}{\partial A_{inlm}}$ (cf. Eqs. (3.12) and (3.39)). Furthermore, $\mathbf{f}_{ki}^{B, f}$ and $\mathbf{f}_{ki}^{L, f}$ can be computed simultaneously as both depend on $\nabla_k \phi_{nlm}(\mathbf{r}_{ki})$ and a previously computed ω (cf. Eqs. (3.19) and (3.43)). Case distinctions depending on λ_i^B allow only required quantities to be computed as shown in Algorithm 3, i.e., one can abort the computation of the fast atomic cluster expansion once $\lambda_i^B = 0$ has been computed and compute $\mathbf{f}_{ki}^{L, f}$ only for $\lambda_i^B \in (0, 1)$.

Algorithm 2 Computation of the ACE energies E_i^{APA} and forces $\mathbf{F}_i^{\text{APA}}$ for all atoms i according to Eqs. (3.27) and (3.44). This algorithm is an extension of Algorithm 1 which includes the adaptive-precision.

```

for all atoms  $i$  do
   $\mathbf{F}[i] \leftarrow \mathbf{0}$ 
   $E[i] \leftarrow 0$ 
end for
for all atoms  $i$  do
  compute fast ACE for atom  $i$  ▷ Use modified ACE code (cf. Algorithm 3).
   $\lambda \leftarrow \lambda_i^{\text{B}}$  computed in fast atomic cluster expansion
  if  $\lambda \neq 1$  then ▷ Compute precise ACE only if required and save thereby computational time.
    compute precise atomic cluster expansion for atom  $i$  ▷ use unmodified ACE code
  end if
  if  $\lambda \neq 1$  then ▷ Otherwise  $E_i^{\text{ACE,p}}$  has not been computed.
     $E[i] += (1 - \lambda)E_i^{\text{ACE,p}}$ 
  end if
  if  $\lambda \neq 0$  then ▷ Otherwise  $E_i^{\text{ACE,f}}$  has not been computed.
     $E[i] += \lambda E_i^{\text{ACE,f}}$ 
  end if
  for all neighbours  $k$  of  $i$  do
     $\mathbf{f} \leftarrow \mathbf{0}$ 
    if  $\lambda \neq 1$  then ▷ Otherwise  $\mathbf{f}_{ki}^{\text{A,p}}$  has not been computed.
       $\mathbf{f} += (1 - \lambda)\mathbf{f}_{ki}^{\text{A,p}}$ 
    end if
    if  $\lambda \neq 0$  then ▷ Otherwise  $\mathbf{f}_{ki}^{\text{B,f}}$  has not been computed.
       $\mathbf{f} += \lambda \mathbf{f}_{ki}^{\text{B,f}}$ 
    end if
    if  $\lambda \in (0, 1)$  then ▷ Otherwise  $\mathbf{f}_{ki}^{\text{L,f}}$  has not been computed.
       $\mathbf{f} += (E_i^{\text{ACE,f}} - E_i^{\text{ACE,p}})\mathbf{f}_{ki}^{\text{L,f}}$ 
    end if
     $\mathbf{F}[k] -= \mathbf{f}$  ▷ cf. Eq. (3.44)
     $\mathbf{F}[i] += \mathbf{f}$  ▷ cf. Eq. (3.44)
  end for
end for

```

3.3 Adaptive-precision ACE copper potential

In this section, two adaptive-precision ACE (APA) potentials are created according to the in Section 3.2 developed method. The ACE copper potential from Ref. [4] uses a basis of 742 many-body basis functions and is thus denoted as 'ACE 742'. Due to the large basis ACE 742 is well suited as precise ACE potential for an APA potential. Therefore, copper (Cu) is used as demonstrator material, with defect-free fcc copper at room temperature as reference environment that should be described by a fast ACE potential. In Section 3.3.1, two faster ACE potentials with a basis of 10 and 50 cluster basis functions – referred to as 'ACE 10' and 'ACE 50' – are fitted to be used as fast ACE potentials. ACE 10 is designed to investigate the coupling of two ACE potentials in the boundary case of a comparable very fast ACE potential, while ACE 50 is used to investigate the coupling mechanism with a medium-precision ACE potential. Thus, the force cutoff r_{cut} , after which there is no interaction between two atoms¹, of ACE 742 is also used for ACE 50, i.e., $r_{\text{cut}}^{\text{ACE 742}} = r_{\text{cut}}^{\text{ACE 50}} = 7.4 \text{ \AA}$. In contrast, the default cutoff radius of **pacemaker** of $r_{\text{cut}}^{\text{ACE 10}} = 6.0 \text{ \AA}$ is used for ACE 10 to make this potential as fast as possible. Afterwards, the calculation of the deviation Δ_i^{B} according to Eq. (3.33) and the switching parameter λ_i^{B} according to Eq. (3.36) are tested for both fast ACE potentials in a snapshot of a nanoindentation in Section 3.3.2 and an adaptive-precision ACE copper potential is defined. The by this potential achieved precision and computational time are presented and discussed in Sections 3.3.3 and 3.3.4. Section 3.4 concludes with learnings from the creation of an adaptive-precision ACE potential and further optimisation options of precision and computational time.

¹No interaction means $R_{nl}(r) = 0$ for $r \geq r_{\text{cut}}$ (cf. Eq. (3.3)).

Algorithm 3 Computation of the fast atomic cluster expansion in $B_{in^t l^t m^t}^{(K^t)}$ for an adaptive-precision ACE potential.

```

compute many-body basis functions  $A_{in^t l^t m^t}^{(K^t)}$                                 ▷ according to Eq. (3.5)
compute cluster basis functions  $B_{in^t l^t m^t}^{(K^t)}$                                 ▷ according to Eq. (3.7)
compute  $\Delta_i^B$                                                                     ▷ according to Eq. (3.33)
compute  $\lambda_i^B$                                                                     ▷ according to Eq. (3.36)
if  $\lambda_i^B = 0$  then
    end computation
else if  $\lambda_i^B = 1$  then
    compute  $\omega_{in^t l^t m^t}^{B,f}$                                                     ▷ according to Eq. (3.12)
    compute  $f_{ki}^{B,f}$                                                             ▷ according to Eq. (3.19)
else
    compute  $\omega_{in^t l^t m^t}^L$  and  $\omega_{in^t l^t m^t}^{B,f}$                                 ▷ according to Eqs. (3.12) and (3.39)
    compute  $f_{ki}^{B,f}$  and  $f_{ki}^{L,f}$                                             ▷ according to Eqs. (3.19) and (3.43)
end if

```

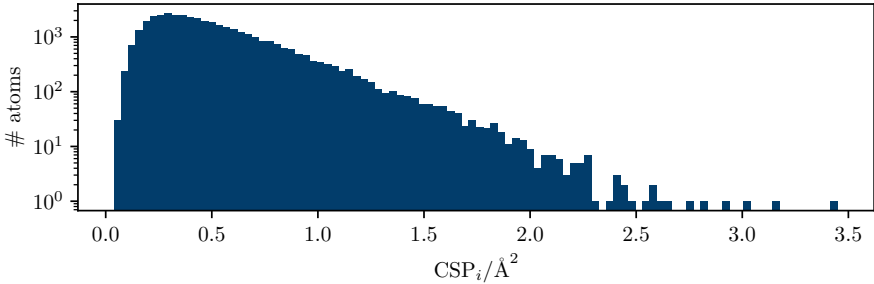


Figure 3.1: Histogram of the centro-symmetry parameter CSP_i of all atoms i in the reference MD structures which are used to fit the fast ACE potentials.

3.3.1 Fitting fast ACE potentials

Normally, an ACE potential is fitted to DFT data in order to obtain the highest possible precision as described in Section 3.1.4. In contrast, here a fast ACE potential should be as similar as possible to the precise ACE potential. Therefore, data of a simulation with the precise potential of fcc Cu at room temperature are used as reference data as described in Section 3.2.1.

The reference MD simulation is initialised with 12^3 fcc unit cells of Cu at 0 K, where the used lattice constant of $3.6496 \text{ \AA}$ was previously measured with the precise ACE potential in a NPT ensemble with zero pressure at room temperature. The atoms are simulated with periodic boundary conditions in all three dimensions and a timestep of 1 fs in a NVT ensemble until the average temperature of the last 1 ps corresponds within a tolerance of 0.1 K to the target temperature of 292 K, i.e., room temperature. The NVT ensemble is realised by an Langevin thermostat whose damping constant is 10 ps. Afterwards, a NVE ensemble is simulated for 50 ps and a snapshot containing the potential energy and force of all atoms is written every 10 ps. Thereby, the reference MD data set $\mathcal{S}^{\text{ACE},f}$ contains 6 structures with 6912 atoms.

The centro-symmetry parameter (CSP)[76] detects defects and surfaces and can be used to analyse the reference structures. The histogram of CSP_i of all atoms i contained in the reference structures is shown in Fig. 3.1. The histogram shows a peak at $0.3 \text{ \AA}^2$, where the number of atoms per bin decays about exponentially for a larger CSP. The largest centro-symmetry parameter included in the reference structures is about $3.5 \text{ \AA}^2$. All atoms contained in the reference MD data are only thermally fluctuating. There are no defects included.

An ACE potential is fitted by minimising a loss function $\mathcal{L}$. The loss function $\mathcal{L}_{\text{ace}}^{\text{DFT}}$ (cf. Eq. (3.21)) is used to fit against DFT data. The loss function $\mathcal{L}_{\text{ace}}^{\text{MD}}$ (cf. Eq. (3.28)) can be used to fit against MD data which contain the potential energy of every single atom rather than only the total potential energy of a structure. The loss functions $\mathcal{L}_{\text{ace}}^{\text{MD}}$ and $\mathcal{L}_{\text{ace}}^{\text{DFT}}$ contain the relative

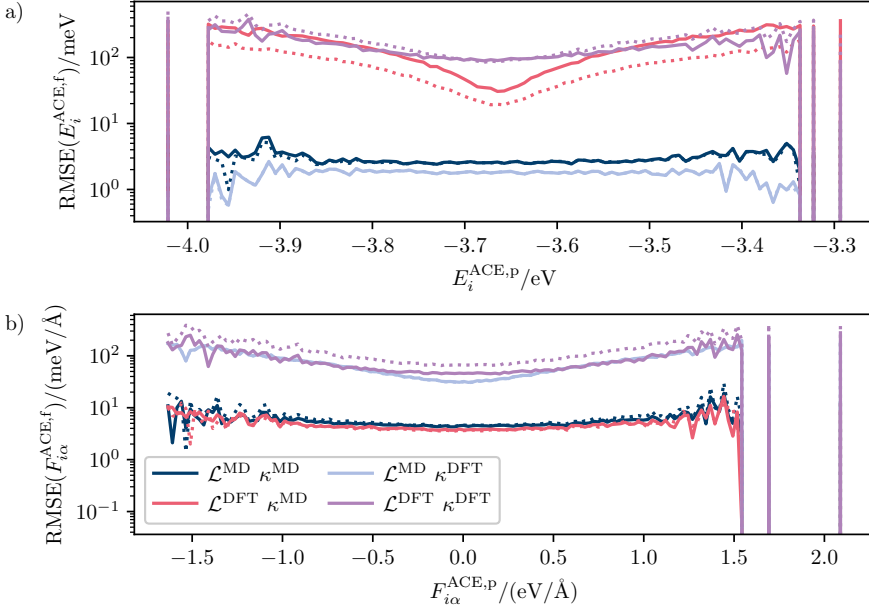


Figure 3.2: Energy and force precision for the training data $\mathcal{S}^{\text{ACE},f}$ achieved in the fitting process of a fast ACE potential which uses a basis of 50 cluster basis functions. The root mean square error (RMSE) is shown for a) potential energies E_i and b) forces components $F_{i\alpha}$. Atoms are sorted into bins dependent on a) $E_i^{\text{ACE},p}$ and b) $F_{i\alpha}^{\text{ACE},p}$ and the RMSE is calculated per bin after the minimisation of the loss function $\mathcal{L}$. The colours correspond to the in the legend given loss function $\mathcal{L}$ (cf. Eqs. (3.21) and (3.28)) and relative force weight κ (cf. Eqs. (3.26) and (3.32)). The solid lines correspond to a uniform weighting policy, where the dotted lines correspond to an energy-based weighting policy.

force weight κ which is a user-determined parameter. An automatic calculation is implemented in `pacemaker`, namely κ^{DFT} Eq. (3.26) which can be adapted also for MD reference structures to κ^{MD} Eq. (3.32). Here, $\kappa^{\text{DFT}}|_{\mathcal{S}^{\text{ACE},f}} = 1.2 \times 10^{-7}$ and $\kappa^{\text{MD}}|_{\mathcal{S}^{\text{ACE},f}} = 8.8 \times 10^{-2}$ apply. Furthermore, one can assign an equal weight to all atoms or use an energy-based weighting policy as described in Sections 3.1.4 and 3.2.1.

The in the minimisation process of ACE 50 for the training data $\mathcal{S}^{\text{ACE},f}$ achieved precision of potential energies and forces is shown in Fig. 3.2 for both loss functions $\mathcal{L}^{\text{MD}}$ and $\mathcal{L}^{\text{DFT}}$, where each loss function is used with κ^{MD} , κ^{DFT} , uniform weighting policy and energy-based weighting policy. At first, the fits which use an uniform weighting policy are analysed in the following. Figure 3.2 shows that $\mathcal{L}^{\text{DFT}}$ with κ^{DFT} works neither for forces nor for energies compared to the other loss functions. Using $\mathcal{L}^{\text{DFT}}$ with κ^{MD} instead of κ^{DFT} , i.e., a larger relative force weight, improves the force precision to 4.01 meV/Å but changes the energy precision only marginally. Using $\mathcal{L}^{\text{MD}}$ instead of $\mathcal{L}^{\text{DFT}}$ with κ^{DFT} improves the energy precision to 1.82 meV, as potential energies are used as target values per atom rather than per structure, but changes the force precision only marginally. The difference between $\mathcal{L}^{\text{MD}}$ and $\mathcal{L}^{\text{DFT}}$ in the achieved force precision shows the most essential improvement in the fitting process. For $\mathcal{L}^{\text{DFT}}$, one needs a set of many small structures with different total potential energies to achieve a good energy precision, but one has not many different structures in this case as the fast ACE potential should only work in the vicinity of the reference environment. For $\mathcal{L}^{\text{MD}}$, however, few similar structures generated in a single MD simulation are sufficient to achieve a good energy precision. Using $\mathcal{L}^{\text{MD}}$ with κ^{MD} combines both previously discussed improvements of $\mathcal{L}^{\text{DFT}}$ with κ^{DFT} : both the force precision of 4.69 meV/Å and the energy precision of 2.60 meV are good. The by $\mathcal{L}^{\text{MD}}$ with κ^{MD} achieved energy-precision is slightly worse than the force precision achieved with κ^{DFT} instead of κ^{MD} as $\kappa^{\text{MD}} > \kappa^{\text{DFT}}$ applies, i.e., using κ^{DFT} increases the relative energy weight $(1 - \kappa)$ and thus the energy precision. Using $\mathcal{L}^{\text{MD}}$ instead of $\mathcal{L}^{\text{DFT}}$ with κ^{MD} slightly decreases the force precision as achieving the same energy

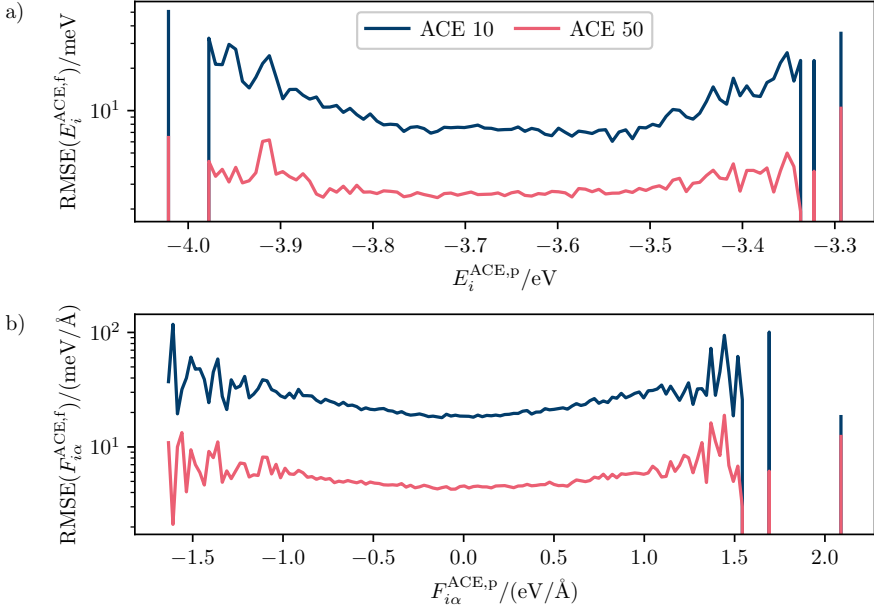


Figure 3.3: Energy and force precision for the training data $\mathcal{S}^{\text{ACE},f}$ achieved in the fitting process of a fast ACE potentials ACE 10 and ACE 50 with the loss function $\mathcal{L}^{\text{MD}}$ with κ^{MD} and a uniform weighting policy. The root mean square error (RMSE) is shown for a) potential energies E_i and b) forces components $F_{i\alpha}$. Atoms are sorted into bins dependent on a) $E_i^{\text{ACE},p}$ and b) $F_{i\alpha}^{\text{ACE},p}$ and the RMSE is calculated per bin after the minimisation of the loss function.

contribution to the loss function is easier with energy reference values per structure in $\mathcal{L}^{\text{DFT}}$ than with energy reference values per atom in $\mathcal{L}^{\text{MD}}$. As the energy contribution to the loss function $\mathcal{L}^{\text{DFT}}$ is more easily minimised, the loss minimisation algorithm more concentrates on the force contribution to the loss function and thereby achieves a better force precision. This discussion shows that increasing the force precision too much decreases the energy precision and vice versa. Using $\mathcal{L}^{\text{MD}}$ with κ^{MD} is a good compromise between energy and force precision and increases the precision compared to $\mathcal{L}^{\text{DFT}}$ with κ^{DFT} for about one order of magnitude.

The default parameters of **pacemaker** were used for the energy-based weighting policy. The previous analysis is also valid for the energy-based weighting policy although the achieved accuracies are slightly different (cf. dotted and solid lines in Fig. 3.2). For $\mathcal{L}^{\text{MD}}$ with κ^{MD} , which gives both precise energies and forces, there are no relevant differences in the achieved precision between the weighting policies. Thus, $\mathcal{L}^{\text{MD}}$ with κ^{MD} and the uniform weighting policy are used to construct an adaptive-precision ACE potential. The potential ACE 10 which only uses 10 instead of 50 cluster basis functions and has a smaller force cutoff than ACE 50 is also fitted with this loss function. ACE 10 has an energy precision of 7.76 meV and a force precision of 20.61 meV/Å in the reference data, where the precision dependent on the value of force and energy is shown in Fig. 3.3. Thus, ACE 10 is about 3 to 4 times less precise than ACE 50.

The phonon spectra of both fitted ACE potentials are in good agreement with the phonon spectra of ACE 742 as shown in Fig. 3.4. Scalar properties calculated with the fitted fast ACE potentials are in Table 3.1 compared to the values calculated with the precise ACE 742 potential. The lattice constant and cohesive energy of the fast potentials are in good agreement with the lattice constant of ACE 742. Although not fitted, the vacancy formation energy of ACE 50 and ACE 742 are similar, but the vacancy formation energy of ACE 10 is negative. Therefore, using ACE 10 near vacancies needs to be prevented. The surface energies of ACE 742 are larger than the surface energies of the fast ACE potentials, the surface energy of (111) surfaces is for ACE 10 even negative. Thus, one should not use ACE 10 at surfaces. In contrast, the interstitial formation energies are although not fitted in relatively good agreement. The elastic constants calculated

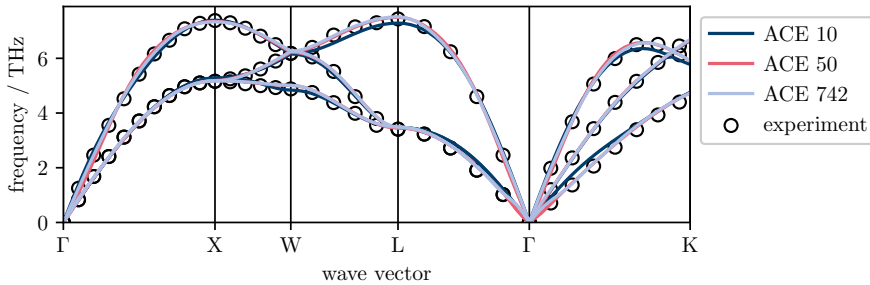


Figure 3.4: Phonon spectra calculated with ASE[136] of the fitted ACE potentials compared to the precise ACE 742 potential and experimental values[137] measured by neutron diffraction at the temperature 80 K.

Table 3.1: Comparison of quantities calculated with the fitted fast ACE potentials ACE 10 and ACE 50, the precise potential ACE 742, DFT and experimental reference values.

	ACE 10	ACE 50	ACE 742	DFT	Exp.
lattice parameter a_0^{fcc} / Å	3.6977	3.6278	3.6309		
cohesive energy / eV	-3.7064	-3.6997	-3.6995		
vacancy formation energy / eV	-1.2843	1.0818	1.1285	1.07[4]	1.27 [138]
surface energy 111 / Jm ⁻²	-1.0093	0.2112	1.3566	1.36[4]	
surface energy 100 / Jm ⁻²	0.3574	0.1335	1.5111	1.51[4]	
surface energy 110 / Jm ⁻²	0.1132	0.4651	1.5889	1.57[4]	
interstitial formation energy / eV					
100-dumbbell	3.1629	3.0092	3.1025	3.10[139]	2.8-4.2[140]
octahedral	3.3772	3.2419	3.3392	3.35[139]	
tetrahedral	3.3932	3.5392	3.6217	3.64[139]	
elastic constant C_{11} / GPa	158.9	142.3	173.8	177[4]	177 [141]
elastic constant C_{12} / GPa	108.1	84.0	120.5	132[4]	125 [141]
elastic constant C_{44} / GPa	67.8	81.9	77.8	82[4]	81 [141]

with the fast potentials are smaller than the ones calculated with ACE 742, but are reasonable. Note, that the fast potentials are fitted for usage at room temperature, but the properties shown in Table 3.1 are defined at 0 K. Hence, the effect of slightly smaller elastic constants might be in practise less relevant when the potentials are used only in the vicinity of the reference environment. In conclusion, the fast ACE potentials can extrapolate reasonable interstitial formation energies but the computed surface energies are wrong as no surfaces are included in the reference dataset $\mathcal{S}^{\text{ACE},f}$.

3.3.2 Detection mechanism and switching function

The detection mechanism is described in Section 3.2.2. The deviation Δ_i^B of the reference environment is calculated according to Eq. (3.33) as squared deviation of all cluster basis functions $B_{\mathbf{n}^i\mathbf{l}^i}^{(K^f)}$ from the value $B_{\mathbf{n}^i\mathbf{l}^i}^{(K^f,\text{ref})}$ in the reference environment. This squared deviation is weighted according to Eq. (3.34) with the expected standard deviation of a cluster basis function. This standard deviation is measured in the structures of reference dataset $\mathcal{S}^{\text{ACE},s}$ which was used to fit the fast ACE potentials. One could measure the reference values $B_{\mathbf{n}^i\mathbf{l}^i}^{(K^f,\text{ref})}$ also in $\mathcal{S}^{\text{ACE},s}$, but the copper atoms fluctuate at room temperature only around their equilibrium positions. Therefore, the reference values of the cluster basis functions are measured in an undisturbed fcc lattice at 0 K, where the same lattice constant is used that was used to construct $\mathcal{S}^{\text{ACE},s}$. As Δ_i^B depends on cluster basis functions, it depends on an ACE potential. The histogram of Δ_i^B in $\mathcal{S}^{\text{ACE},s}$ is shown in Fig. 3.5. One sees the normalisation to 1 according to Eq. (3.35), where the median of Δ_i^B is at 0.6. The maximum of the histogram is at $\Delta_i^B \approx 0.2$ and the number of atoms per bin decays until about 12 for ACE 50 and until about 11 for ACE 10, where there are few outliers in both cases.

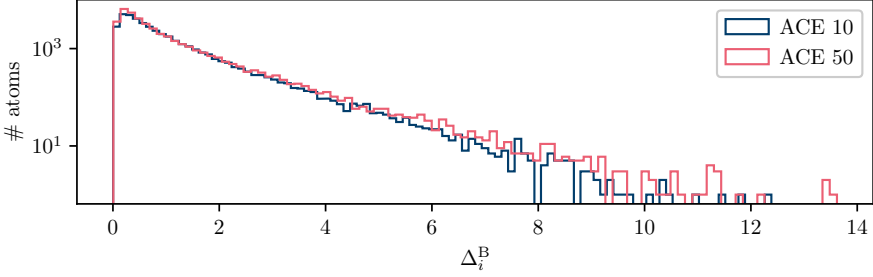


Figure 3.5: Histogram of the deviation Δ_i^B of the reference environment calculated according to Eq. (3.33) in the reference dataset $\mathcal{S}^{\text{ACE},s}$ that was used to fit the fast ACE potentials ACE 10 and ACE 50. The deviation Δ_i^B depends on cluster basis functions and therefore also on the used ACE potential. The histogram is given for the fast ACE potentials ACE 10 and ACE 50.

Therefore, atoms with a deviation up to $\Delta_{\text{high}}^{\text{B,ACE } 10} = 11$ respectively $\Delta_{\text{high}}^{\text{B,ACE } 50} = 12$ are expected in the vicinity of the reference environment.

In a nanoindentation an indenter is moved into a material to characterise the hardness of the material. Dislocations nucleate due to the indentation and may glide through the system and interact with each other. To simulate these effects one needs a large system, which is why the length scale of nanoindentations is challenging[80]. As primarily the surface and dislocated atoms are of interest rather than the remaining atoms, which serve as material background, a nanoindentation might profit from the usage of an adaptive-precision potential. Therefore, a nanoindentation is used as test system in the following.

The deviation Δ_i^B is visualised in Fig. 3.6 for a snapshot of a nanoindentation of a (100) copper surface. The nanoindentation is simulated at room temperature and thus one can use the previously fitted fast ACE potentials to calculate Δ_i^B . As the nanoindentation snapshot contains a surface and dislocations it serves here as test system for the deviation calculation. The dislocations are identified and visualised by the dislocation extraction algorithm (DXA)[142] of OVITO[143]. The deviation Δ_i^B calculated with ACE 10 is referred to as $\Delta_i^{\text{B,ACE } 10}$ and shown in Fig. 3.6a. The first two layers of atoms at the surface have a deviation $\Delta_i^{\text{B,ACE } 10}$ larger than $\Delta_{\text{high}}^{\text{B,ACE } 10}$. Out of the perspective of atoms in the third atom layer, the surface atoms are 3.6 Å away and the next non-existing atom-layer would be in a distance of 5.4 Å, which is near the force cutoff $r_{\text{cut}}^{\text{ACE } 10} = 6.0$ Å. Thus, the first two atom layers have a large enough $\Delta_i^{\text{B,ACE } 10}$ that can prevent the usage of the ACE 10 energies and forces and the third atom layer is far enough away from the surface so that the evaluation of ACE 10 seems appropriate. However, $\Delta_i^{\text{B,ACE } 10}$ has near dislocations primarily the for this temperature in bulk expected deviation from the reference structure and thus fails to detect dislocations, i.e., there are almost no atoms shown near the dislocation lines in Fig. 3.6a. Therefore, one can use an adaptive-precision ACE potential based on ACE 10 only in simulations without defects.

The deviation Δ_i^B calculated with ACE 50 is referred to as $\Delta_i^{\text{B,ACE } 50}$ and shown in Fig. 3.6b. Due to the larger force cutoff of $r_{\text{cut}}^{\text{ACE } 50} = 7.4$ Å, $\Delta_i^{\text{B,ACE } 50}$ is larger than expected for bulk in the first three atom layers at the surface. The nearest due to the surface missing atom is 7.2 Å away for atoms in the fourth atom layer and thus near the force cutoff. Hence, the most relevant atoms at the surface have a sufficiently large $\Delta_i^{\text{B,ACE } 50}$ to prevent the incorrect extrapolation of ACE 50 at the (100) surface. Furthermore, $\Delta_i^{\text{B,ACE } 50}$ is larger than expected in defect-free bulk near dislocation lines, i.e., most of the atoms located between dislocation lines in Fig. 3.6b are shown as $\Delta_i^{\text{B,ACE } 50} > \Delta_{\text{high}}^{\text{B,ACE } 50}$ applies. Thus, the visualisation of $\Delta_i^{\text{B,ACE } 50}$ looks promising, in contrast to the visualisation of $\Delta_i^{\text{B,ACE } 10}$. There are single atoms with $\Delta_i^{\text{B,ACE } 50} > \Delta_{\text{high}}^{\text{B,ACE } 50}$ which are neither near a dislocation nor near the surface as visualised in Fig. 3.6b. Treating this atoms with the precise ACE potential is not required and costs computational time. This atoms are detected as $\Delta_i^{\text{B,ACE } 50}$ is selected according to Fig. 3.5 to include nearly all, but not all, values of $\Delta_i^{\text{B,ACE } 50}$ in the reference dataset $\mathcal{S}^{\text{ACE},f}$. However, one should not increase $\Delta_{\text{high}}^{\text{B,ACE } 50}$ to detect less bulk atoms as this would also increase the risk of incorrect extrapolation.

The switching parameter λ_i^B is calculated according to Eq. (3.36). Besides Δ_i^B and Δ_{high}^B , one

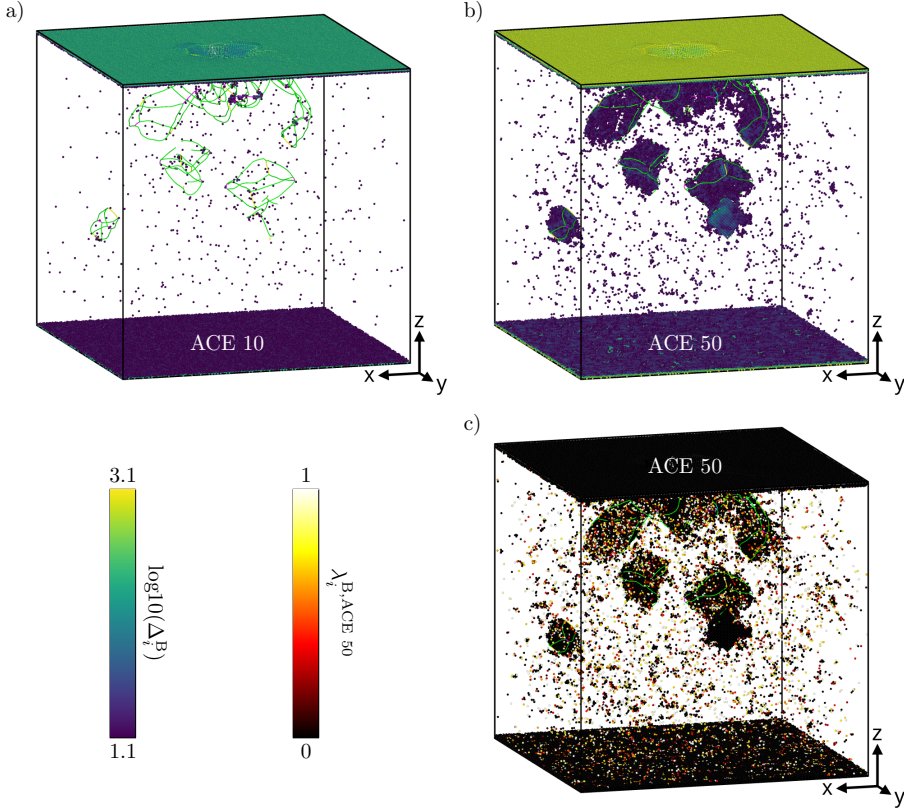


Figure 3.6: Visualisations of an at room temperature simulated nanoindentation of a (100) copper surface for an indentation depth of 20 Å. The coloured lines are dislocation lines which are identified by the dislocation extraction algorithm (DXA)[142] of OVITO[143]. Additionally, all atoms i with a deviation Δ_i^B of the reference environment (cf. Eq. (3.33)) larger than Δ_{high}^B , i.e., all atoms with a Δ_i^B that is larger than expected for bulk copper at 292 K, are shown in a) for $\Delta_i^{B,ACE 10}$ and in b) for $\Delta_i^{B,ACE 50}$. The atoms are coloured according to Δ_i^B . $\Delta_i^{B,ACE 10}$ does not detect dislocations, but $\Delta_i^{B,ACE 50}$ does. c) The Atoms are coloured dependent on $\lambda_i^{B,ACE 50}$ according to Eq. (3.36), where all atoms i with $\lambda_i^{B,ACE 50} = 1$ are not shown.

also needs the parameter Δ_{low}^B which determines the transition zone in which atoms are partially calculated with the fast and partially calculated with the precise ACE potential (cf. Eq. (3.27)). All atoms with $\Delta_{\text{high}}^B < \Delta_i^B$ need to be calculated with the precise potential to prevent an incorrect extrapolation of the fast ACE potential as these values of Δ_i^B are not present in the reference data (cf. Fig. 3.5). Thus, one needs a value of Δ_i smaller than Δ_{high}^B from which atoms can be transferred into a more precise calculation or less precise but faster calculation in case of a decreasing Δ_i^B . If Δ_{low}^B is too small, too many atoms are calculated precisely. If Δ_{high}^B is too large, the transformation between precise and fast is not smooth. $\Delta_{\text{low}}^{B,ACE 50} = 10$ is selected as there is a minimum in the histogram of Δ_i^B as shown in Fig. 3.5 and this selection prevents too much atoms in the transition zone. Thereby, the parametrisation of $\lambda_i^{B,ACE 50}$ calculated from $\Delta_i^{B,ACE 50}$ is complete and one can use this switching function to compute the adaptive-precision potential APA 50 which is given according to Eq. (3.27) as

$$E_i^{\text{APA 50}} = \lambda_i^{B,ACE 50} E_i^{\text{ACE 50}} + (1 - \lambda_i^{B,ACE 50}) E_i^{\text{ACE 742}}. \quad (3.45)$$

The switching parameter $\lambda_i^{B,ACE 50}$ is visualised in Fig. 3.6c, where atoms with $\lambda_i^{B,ACE 50} = 1$, i.e., fast calculated atoms, are not shown. There are thermally fluctuating atoms in the transition zone with $\lambda_i^{B,ACE 50} \in (0, 1)$, but the majority of the atoms is not shown and therefore calculated fast.

3.3.3 Precision

The main objective of an adaptive-precision ACE potential is a reduction of the computational time of a precise potential while keeping the high precision of the precise potential. The adaptive-precision ACE potential APA 50 was developed in Sections 3.3.1 and 3.3.2. As the precise potential energy of ACE 742 is used near dislocations and at the surface, one can test the precision of the developed APA 50 potential in a snapshot of a nanoindentation. The achieved precision is shown in Section 3.3.3.1. Afterwards the achieved precision is explained in Section 3.3.3.2. Based on this explanation, further optimisation possibilities are presented in Section 3.3.3.3.

3.3.3.1 Demonstration

In this section, the in Section 3.3 created potential APA 50 is tested in snapshots of a nanoindentation to evaluate its precision and applicability in a realistic scenario with 4 million atoms. A nanoindentation can serve as a test system as there is initially only a surface and defects which should not be described with the fast ACE 50 potential develop during the nanoindentation like shown in Fig. 3.7. Initially, $\lambda_i^{\text{B,ACE } 50} = 0$, which implies according to Eq. (3.45) the usage of the precise energy of the ACE 742 potential for this atom i , applies only for the surface and few thermally fluctuating atoms in the bulk like shown in Fig. 3.7a. For the small indentation depths in Fig. 3.7b-c, there are no dislocations yet, but there is a growing structure of neighbouring atoms i below the indenter with $\lambda_i^{\text{B,ACE } 50} = 0$. This is reasonable as the first dislocations nucleate below the surface[144]. With ongoing indentation, dislocations (cf. Fig. 3.7d) and prismatic dislocation loops (cf. Fig. 3.7e-i) nucleate, where in both cases there are atoms with $\lambda_i^{\text{B,ACE } 50}$ next to the dislocations. Therefore, the detection through $\Delta_i^{\text{B,ACE } 50}$ of atoms which cannot be described by the energy of ACE 50 looks reasonable not only for dislocations, but also for the onset of plasticity before the dislocation nucleation.

The accuracy achieved is analysed in Fig. 3.8 for the snapshot at 20 Å indentation depth which is shown in Figs. 3.6b-c and 3.7i. Figure 3.8a shows the achieved precision dependent on $\Delta_i^{\text{B,ACE } 50}$. APA 50 achieves for $\lambda_i^{\text{B,ACE } 50} = 1$ with 2.65 meV the same energy precision like in the training dataset $\mathcal{S}^{\text{ACE,f}}$ with 2.60 meV. Hence, the detection of a similar environment with $\Delta_i^{\text{B,ACE } 50}$ works. The negligible energy precision difference shows that the energies are not incorrectly extrapolated. The energy precision of APA 50 is worst in the first quintile of the transition zone with 4.23 meV but increases for larger values of $\Delta_i^{\text{B,ACE } 50}$ as the fraction of the used precise energy $E_i^{\text{ACE } 742}$ increases (cf. Eq. (3.45)) until the precise energies are used exactly for $\lambda_i^{\text{B,ACE } 50} = 0$. The energy precision of ACE 50 for atoms with $\lambda_i^{\text{B,ACE } 50} \in (0, 1)$ is 4.6 meV still good and decreases only significantly for atoms with $\lambda_i^{\text{B,ACE } 50} = 0$ to 293.65 meV in the last quintile. This shows, that the choice of the thresholds $\Delta_{\text{low}}^{\text{B,ACE } 50}$ and $\Delta_{\text{high}}^{\text{B,ACE } 50}$ for Eq. (3.36) was appropriate.

The force precision is shown in Fig. 3.8b dependent on $\Delta_i^{\text{B,ACE } 50}$. The force precision of ACE 50 is with 9.69 meV/Å in the first four quintiles of $\lambda_i^{\text{B,ACE } 50} = 1$ about a factor of 2 worse than in the reference data $\mathcal{S}^{\text{ACE,f}}$, where the force precision achieved is 4.69 meV/Å. The force precision decreases to 25.89 meV/Å and 130.56 meV/Å in the last quintile of $\lambda_i^{\text{B,ACE } 50} = 1$ and $\lambda_i^{\text{B,ACE } 50} \in (0, 1)$. The force precision of ACE 50 is in the transition range of $\lambda_i^{\text{B,ACE } 50} \in (0, 1)$ by a factor of 13 worse than in the first four quintiles of $\lambda_i^{\text{B,ACE } 50}$. This shows, that the thresholds $\Delta_{\text{low}}^{\text{B,ACE } 50}$ and $\Delta_{\text{high}}^{\text{B,ACE } 50}$ for Eq. (3.36) are too high out of a force-precision perspective although the thresholds are appropriate to achieve a good energy precision (cf. Fig. 3.8a). The force-precision of APA 50 is good for $\lambda_i^{\text{B,ACE } 50} = 1$, worst in the third quintile of the transition range with 66.35 meV/Å and on average 24.80 meV/Å for $\lambda_i^{\text{B,ACE } 50} = 0$. Thus, APA 50 more precisely reproduces the precise ACE 742 forces in all quintiles than ACE 50. It seems reasonable to use APA 50 as the energy-precision is good and the worst force precision affects only few atoms in the transition range of $\lambda_i^{\text{B,ACE } 50} \in (0, 1)$.

All surface atoms are reliably detected by $\Delta_i^{\text{B,ACE } 50}$, where the value of $\Delta_i^{\text{B,ACE } 50}$ is a few hundred as shown in Fig. 3.6. Apart from surface atoms, one has no intuition what $\Delta_i^{\text{B,ACE } 50}$ means physically. Therefore, the precision is shown in Fig. 3.8c-d also dependent on the centro-symmetry parameter which can be interpreted physically. The plot is separated into thermally fluctuation atoms, dislocations and surface atoms. As surface atoms are detected exactly by $\Delta_i^{\text{B,ACE } 50}$ and thus the exact values of the ACE 742 potential energy are used, one can see in Fig. 3.8c that there are atoms which are falsely identified as surface atoms. The same happens for dislocations. However, one can see that both energy and force precision of APA 50 are in particular bad for dislocations. Hence, the partially bad force precision of APA 50 seems less negligible as dislocations

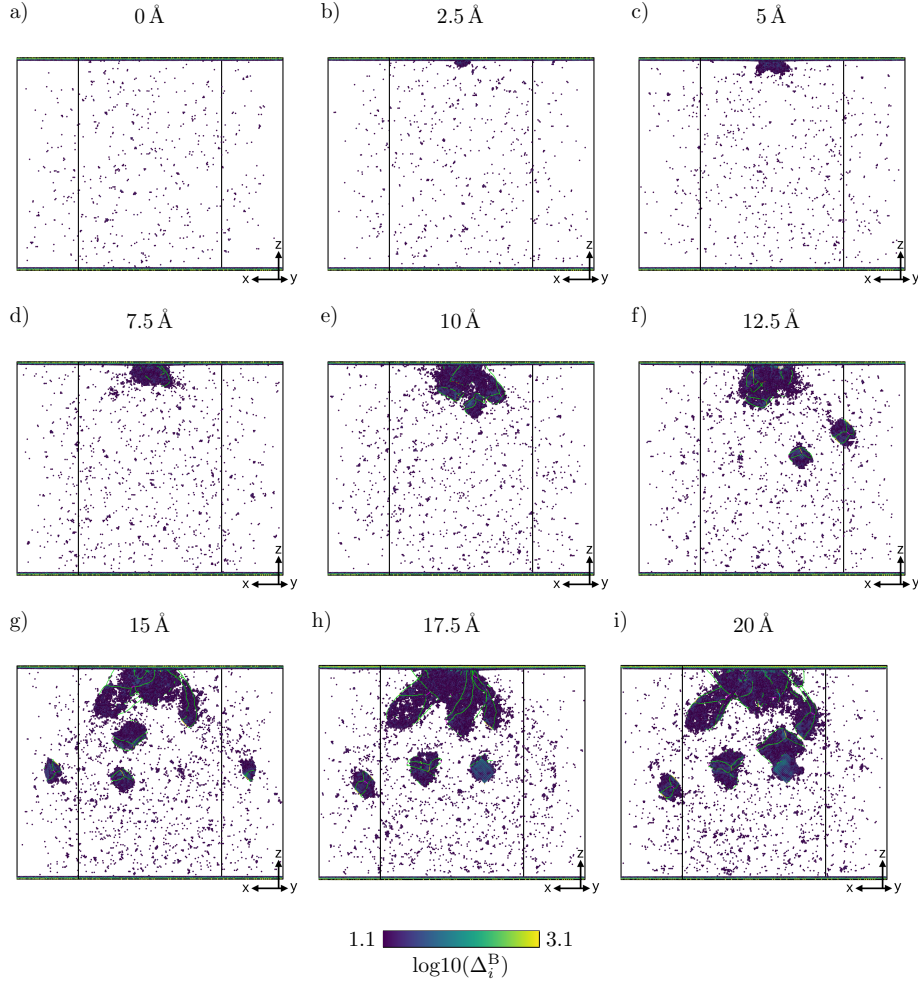


Figure 3.7: Visualisations of an at room temperature simulated nanoindentation of a (100) copper surface. The coloured lines are dislocation lines which are identified by the dislocation extraction algorithm (DXA)[142] of OVITO[143]. Additionally, all atoms i with $\lambda_i^{\text{B,ACE } 50} = 0$, i.e., all atoms which require a precise calculation, are shown. The atoms are coloured according to the deviation Δ_i^B of the reference environment according to Eq. (3.33). The visualisations are for different indentation depths from a) 0 Å to i) 20 Å as indicated above each visualisation.

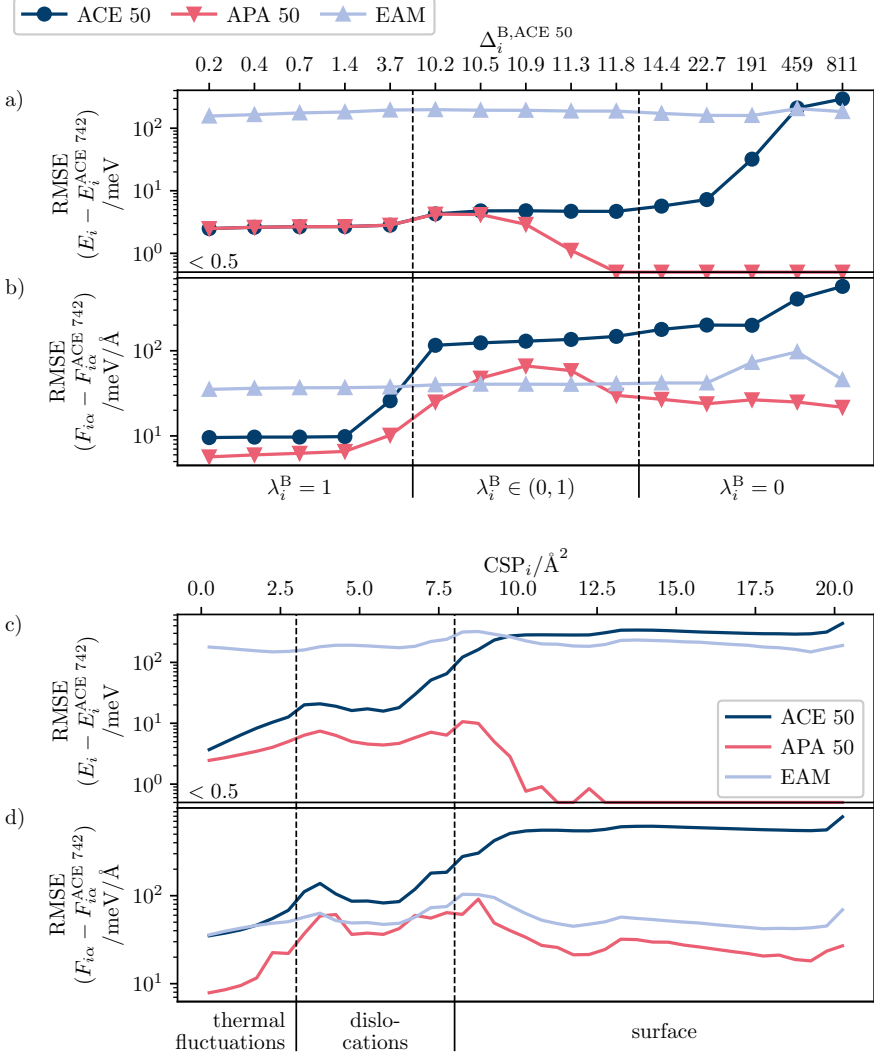


Figure 3.8: Analysis of the energy and force precision achieved of APA 50 for different subgroups of atoms. The analysis is done for the in Fig. 3.6 visualised snapshot of a nanoindentation of (100) copper at room temperature in an indentation depth of 20 Å. a-b) The atoms are separated in three groups dependent on $\lambda_i^{\text{B,ACE 50}}$: the first group consists of all atoms i with $\lambda_i^{\text{B,ACE 50}} = 1$, i.e., all atoms for which the fast calculated energy $E_i^{\text{ACE 50}}$ is used in APA 50. The second group consists of all atoms with $\lambda_i^{\text{B,ACE 50}} \in (0, 1)$ and the third group consists of all atoms with $\lambda_i^{\text{B,ACE 50}} = 0$. Each group is separated in quintiles dependent on $\Delta_i^{\text{B,ACE 50}}$. The RMSE is calculated per quintile for a) potential energies and b) forces. The mean $\Delta_i^{\text{B,ACE 50}}$ per quintile is given on the top axis. c-d) The atoms are sorted into bins of width $0.5 \text{ \AA}^{-2}$ dependent on the centro-symmetry parameter (CSP) and the RMSE is calculated per bin for c) potential energies and d) forces. As the CSP detect asymmetries that are characteristic for dislocations and surface atoms, the corresponding ranges of the CSP are shown on the bottom axis although this detection is not exact and there are few falsely detected atoms. For comparison the RMSE of ACE 50 and the EAM2 potential of Ref. [145] are also given.

Table 3.2: Comparison of Pearson correlation coefficients(PCC), root mean squared error (RMSE) and mean of the in Fig. 3.9 visualised $f_{k\alpha}^{A,ACE\ 50}$ and $f_{k\alpha}^{A,ACE\ 742}$.

neighbour shell	1	2	3	4	5	6	7	8
$\langle r_{ki} \rangle_{ki} / \text{\AA}$	2.58	3.65	4.47	5.16	5.77	6.33	6.83	7.30
$\langle f_{k\alpha}^{A,ACE\ 50} \rangle_{k\alpha} / \text{eV \AA}^{-1}$	2.1e-1	1.9e-2	5.7e-2	1.8e-2	1.7e-3	3.5e-3	1.1e-3	6.0e-5
$\langle f_{k\alpha}^{A,ACE\ 742} \rangle_{k\alpha} / \text{eV \AA}^{-1}$	2.1e-1	2.1e-2	5.6e-2	1.8e-2	1.7e-3	2.6e-3	9.9e-4	1.5e-4
$\text{RMSE}(f_{k\alpha}^{A,ACE\ 50} - f_{k\alpha}^{A,ACE\ 742}) / \text{eV \AA}^{-1}$	7.4e-3	4.0e-3	3.7e-3	1.5e-3	1.4e-3	1.1e-3	3.7e-4	2.1e-4
$\text{PCC}(f_{k\alpha}^{A,ACE\ 50}, f_{k\alpha}^{A,ACE\ 742})$	1.00	0.99	1.00	1.00	0.87	0.99	0.96	0.84

are of interest in most simulations and should be described with high precision.

3.3.3.2 Discussion

Figure 3.8 shows that the RMSE of APA 50 compared to ACE 742 is up to 10.6 meV and 4.2 meV for the potential energies when analysed dependent on CSP and $\Delta_i^{B,ACE\ 50}$. Thus, the energy-precision is quite good. The RMSE of the forces is up to 91.5 meV/\AA and 66.3 meV/\AA when analysed dependent on CSP and $\Delta_i^{B,ACE\ 50}$. For comparison, the RMSE achieved in the fitting of ACE 50 for the reference dataset $\mathcal{S}^{ACE,f}$ is 2.6 meV for the potential energy and 4.7 meV/\AA for forces. Thus, the precision of APA 50 in the nanoindentation snapshot is up to a factor of 19.5 and 2.3 for forces and energies worse than the precision of ACE 50 in $\mathcal{S}^{ACE,f}$. Ideally, this factors should be near 1 as ACE 50 is only used in the vicinity of the reference environment to which it was fitted. However, this comparison shows that the energy-precision of APA 50 is quite good, but the force-precision is not. The adaptive-precision ACE force on an atom k is given according to Eqs. (3.42) and (3.44) as

$$\mathbf{F}_k^{APA\ 50} = \sum_i (\mathbf{f}_{ik}^{APA\ 50} - \mathbf{f}_{ki}^{APA\ 50}), \quad (3.46)$$

where the force contributions are

$$\mathbf{f}_{ki}^{APA\ 50} = (E_i^{ACE\ 50} - E_i^{ACE\ 742}) \mathbf{f}_{ki}^{L,ACE\ 50} + \lambda_i^{B,ACE\ 50} \mathbf{f}_{ki}^{B,ACE\ 50} + (1 - \lambda_i^{B,ACE\ 50}) \mathbf{f}_{ki}^{A,ACE\ 742}. \quad (3.47)$$

In contrast, the precise force is given as

$$\mathbf{F}_k^{ACE\ 742} = \sum_i (\mathbf{f}_{ik}^{A,ACE\ 742} - \mathbf{f}_{ki}^{A,ACE\ 742}). \quad (3.48)$$

Comparing Eqs. (3.46) and (3.48) shows that a force difference between APA 50 and ACE 742 originates in a difference between the force contributions $\mathbf{f}_{ki}^{APA\ 50}$ and $\mathbf{f}_{ki}^{A,ACE\ 742}$. The force contribution difference is

$$\mathbf{f}_{ki}^{APA\ 50} - \mathbf{f}_{ki}^{A,ACE\ 742} = (E_i^{ACE\ 50} - E_i^{ACE\ 742}) \mathbf{f}_{ki}^{L,ACE\ 50} + \lambda_i^{B,ACE\ 50} (\mathbf{f}_{ki}^{B,ACE\ 50} - \mathbf{f}_{ki}^{A,ACE\ 742}). \quad (3.49)$$

Thus, a force contribution difference is according to Eq. (3.49) caused either by $(\mathbf{f}_{ki}^{B,ACE\ 50} - \mathbf{f}_{ki}^{A,ACE\ 742})$ or $(E_i^{ACE\ 50} - E_i^{ACE\ 742}) \mathbf{f}_{ki}^{L,ACE\ 50}$. In Eq. (3.49), the difference of $\mathbf{f}_{ki}^{B,ACE\ 50}$ and $\mathbf{f}_{ki}^{A,ACE\ 742}$ is weighted with λ_i and thus only applies for neighbours i whose potential energy is calculated with ACE 50, i.e., for all atoms whose environment is similar to the reference environment. In the reference dataset $\mathcal{S}^{ACE,f}$, the difference of $\mathbf{f}_{ki}^{A,ACE\ 50}$ and $\mathbf{f}_{ki}^{A,ACE\ 742}$ is small as visualised in Fig. 3.9. The Pearson correlation coefficient (PCC) is about 1.00 in the first four neighbour shells and between 0.84 and 0.99 in the next four neighbour shells and atoms that are further away are outside of the cutoff radius as shown in Table 3.2. Thus, there is a difference between $\mathbf{f}_{ki}^{A,ACE\ 50}$ and $\mathbf{f}_{ki}^{A,ACE\ 742}$, in particular for atoms in the fifths and eights neighbour shell. Therefore, it is expected that the force precision decreases when force contributions calculated with ACE 50 and ACE 742 are combined as in Eq. (3.46).

Such a combination of force contributions occurs for all atoms k , in whose force cutoff the switching parameter is not equal for all atoms. An example is the (100) Cu surface as not all atoms in the force cutoff of surface atoms have a switching parameter of 0 (cf. Section 3.3.2): only three

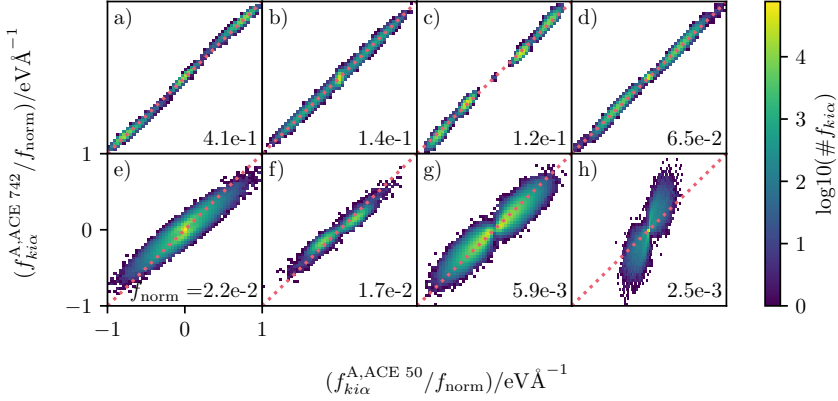


Figure 3.9: Comparison of $f_{ki}^{A,ACE 50}$ and $f_{ki}^{A,ACE 742}$ calculated according to Eq. (3.19). The comparison is done for one structure of the reference dataset $\mathcal{S}^{ACE,f}$. The three scalar components $f_{ki\alpha}^{A,ACE}$ are used rather than the vector $\mathbf{f}_{ki}^{A,ACE}$. The force components are compared per neighbour shell in ascending order from a) to h), i.e., k is in the first neighbour shell of i in a) and in the eighth neighbour shell in h). For better visualisation, the force components are normed with f_{norm} per neighbour shell, where f_{norm} is given in the lower right corner of each subplot. Thereby, the axis range is $[-1,1]$ for all subplots and only annotated in e). The dotted line shows where $f_{ki\alpha}^{A,ACE 50} = f_{ki\alpha}^{A,ACE 742}$ applies.

atom layers at the surface have $\lambda_i^{B,ACE 50} = 0$, but the fourth and fifth atom layer are also in the force cutoff of surface atoms, i.e., about 20 neighbouring atoms of the fifths to eights neighbour shell of surface atoms have a switching parameter of 1. To quantify the by this mechanism expected force error, one can use $RMSE(f_{ki\alpha}^{A,ACE 50} - f_{ki\alpha}^{A,ACE 742})$ which is shown in Table 3.2 for a snapshot of the reference data $\mathcal{S}^{ACE,f}$. As first approximation, the RMSE per atom is about $0.76 \text{ meV}\text{\AA}^{-1}$ for atoms in the fifths to eights neighbour shell. As about 20 neighbours k in these neighbour shells of surface atoms i have a switching parameter of 1. As both $f_{ki\alpha}$ and $f_{ik\alpha}$ influence $\mathbf{F}_i^{APA}$ (cf. Eq. (3.46)), one expects, in first approximation, a force error of about $20 \times 2 \times 0.76 \text{ meV}\text{\AA}^{-1} = 30.4 \text{ meV}\text{\AA}^{-1}$ for surface atoms. Hence, the RMSE of surface atoms with $CSP_i \geq 11$ in Fig. 3.8d is explained by force contributions of neighbouring atoms with a switching parameter of 1, i.e., atoms which are in the vicinity of the reference environment. The same force-precision decreasing mechanism also occurs near dislocations, but cannot be estimated just as easily. Ideally, the switching parameter increases from 0 to 1 over few atom layers to minimise the effect discussed instead of the abrupt change from 0 at one atom layer to 1 at the next atom layer like at the discussed surface.

The second term which causes a force contribution difference in Eq. (3.49) and thus decreases the force precision is $(E_i^{ACE 50} - E_i^{ACE 742}) \mathbf{f}_{ki}^{L,ACE 50} \cdot \mathbf{f}_{ki}^{L,ACE 50}$. $\mathbf{f}_{ki}^{L,ACE 50}$ is calculated according to Eq. (3.43), where $\mathbf{f}_{ki}^{L,ACE 50} = 0$ applies for $\lambda_i^{B,ACE 50} \in \{0, 1\}$ according to Eqs. (3.39) and (3.40). Thus, to investigate the influence of on the force precision one can select

$$\Delta_{high,\epsilon}^{B,ACE 50} = \Delta_{low,\epsilon}^{B,ACE 50} + \epsilon, \quad (3.50)$$

where ϵ is a small positive number that prevents the occurrence of $\lambda_{\epsilon,i}^{B,ACE 50} \in (0, 1)$ and thus also the occurrence of $\mathbf{f}_{ki}^{L,ACE 50} \neq 0$. Thereby, the potential energy of atoms can jump unphysically between two timesteps, but it is instructive to investigate the influence of $\mathbf{f}_{ki}^{L,ACE 50}$ on the force precision. The thresholds $\Delta_{low}^{B,ACE 50} = 10$ and $\Delta_{low}^{B,ACE 50} = 12$ are used in the APA 50 potential. Therefore, APA 50 is used with $\Delta_{low,\epsilon}^{B,ACE 50} = 10$ and $\Delta_{low,\epsilon}^{B,ACE 50} = 12$ and $\Delta_{high,\epsilon}^{B,ACE 50}$ according to Eq. (3.50) rather than the default thresholds for $\Delta_i^{B,ACE 50}$ in Fig. 3.10 for the same snapshot that is analysed in Fig. 3.8. The difference of the RMSE of the forces between both values of $\Delta_{low,\epsilon}^{B,ACE 50}$ is less than $3.6 \text{ meV}\text{\AA}^{-1}$ as shown in Fig. 3.10b, where the worst RMSE is $22.6 \text{ meV}\text{\AA}^{-1}$. In contrast, when the default values of the thresholds of APA 50 are used, the worst RMSE of the forces is with $66.4 \text{ meV}\text{\AA}^{-1}$ three times larger. The mean RMSE decrease when the transition-range width of ϵ is used is $1.2 \text{ meV}\text{\AA}^{-1}$, $24.2 \text{ meV}\text{\AA}^{-1}$ and $8.2 \text{ meV}\text{\AA}^{-1}$ for atoms i with $\lambda_i^{B,ACE 50} = 1$,

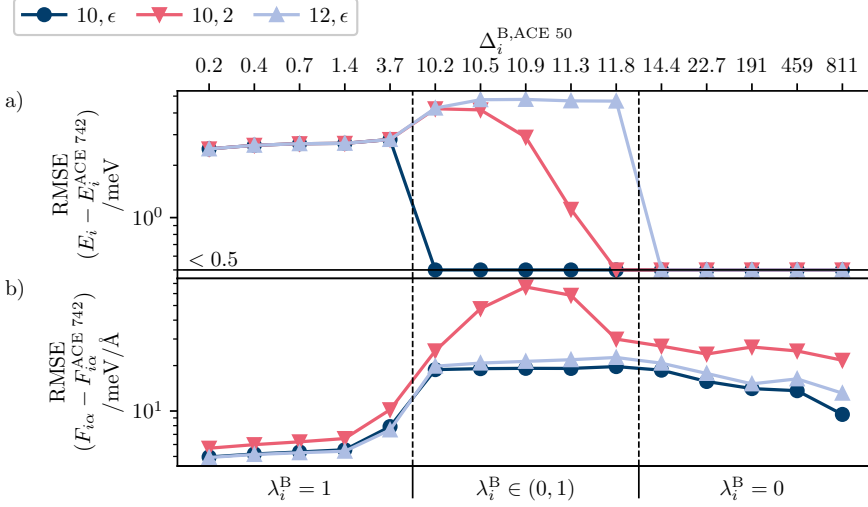


Figure 3.10: Analysis of the energy and force precision achieved of APA 50 dependent on the width of the transition range $\Delta_{\text{high}}^B - \Delta_{\text{low}}^B$ (cf. Eq. (3.36)). The analysis is done for the in Fig. 3.6 visualised snapshot of a nanoindentation of (100) copper at room temperature in an indentation depth of 20 Å. The atoms are separated in three groups dependent on $\Delta_i^{B,ACE 50}$: the groups consists of all atoms i with $\Delta_i^{B,ACE 50} \leq 10$, $\Delta_i^{B,ACE 50} \in (10, 12)$ and $\Delta_i^{B,ACE 50} \geq 12$, i.e., the groups correspond to $\lambda_i^{B,ACE 50} = 1$, $\lambda_i^{B,ACE 50} \in (0, 1)$ and $\lambda_i^{B,ACE 50} = 0$ with the thresholds defined and demonstrated for APA 50 in Sections 3.3 and 3.3.3.1. The ranges of the switching parameter are given on the bottom axis. Each group is separated in quintiles dependent on $\Delta_i^{B,ACE 50}$. The RMSE is calculated per quintile for a) potential energies and b) forces. The mean $\Delta_i^{B,ACE 50}$ per quintile is given on the top axis. The used values of Δ_{low}^B and $\Delta_{\text{high}}^B - \Delta_{\text{low}}^B$ are given in the legend, where the width ϵ is defined in Eq. (3.50).

$\lambda_i^{B,ACE 50} \in (0, 1)$ and $\lambda_i^{B,ACE 50} = 0$. Thus, the precision decrease caused by $\mathbf{f}_{ki}^{L,ACE 50}$ affects most the atoms i with $\lambda_i^{B,ACE 50} \in (0, 1)$, but is also non-negligible for the remaining quintiles analysed in Fig. 3.10b, i.e., $\mathbf{f}_{ki}^{L,ACE 50}$ also decreases the force-precision for all neighbouring atoms k within the force-cutoff of atom i to a non-negligible extent. This comparison has shown that, the calculation of $\mathbf{f}_{ki}^{L,ACE 50}$ and thus the calculation of $(\nabla_k \lambda_i^B)$ in Eq. (3.38) decreases the force precision of atoms i with $\lambda_i^{B,ACE 50} \in (0, 1)$ in a problematic extent. However, a smooth transition from the fast to the precise description and vice versa of an atom is essential to avoid the discontinuity of the potential energy over time.

In conclusion, one cannot expect the force precision which is achieved in the reference dataset $\mathcal{S}^{ACE,f}$ during the fit of ACE 50 also for APA 50 for two reasons. Firstly, the force precision decreases when the total force $F_k^{APA 50}$ on an atom k is calculated from both force contributions $\mathbf{f}_{ki}^{B,ACE 50}$ of ACE 50 and force contributions $\mathbf{f}_{ki}^{A,ACE 742}$ of ACE 742. Secondly, the force precision decreases for all atoms in whose force cutoff is an atom k with $\lambda_k^{B,ACE 50} \in (0, 1)$ due the calculation of $\mathbf{f}_{ik}^{L,ACE 50}$.

3.3.3.3 Optimisation potential

As the force contribution of the gradient of the switching parameter is weighted with $(E_i^{ACE 50} - E_i^{ACE 742})$ (cf. Eqs. (3.38) and (3.42)), one can reduce the influence of this precision-decreasing quantity by decreasing the potential energy difference $(E_i^{ACE 50} - E_i^{ACE 742})$. The energy precision of ACE 50 decreases when $\Delta_i^{B,ACE 50}$ increases as shown in Fig. 3.11a. Therefore, using smaller thresholds Δ_{low}^B and Δ_{high}^B decreases the influence of $\mathbf{f}_{ik}^{L,ACE 50}$. Decreasing the thresholds $\Delta_{\text{low}}^{B,ACE 50}$ and $\Delta_{\text{high}}^{B,ACE 50}$ by 7 to 3 and 5 prevents the peak of the RMSE of the forces between both thresholds as shown in Fig. 3.11b. However, the force precision analysed dependent on the centro-symmetry

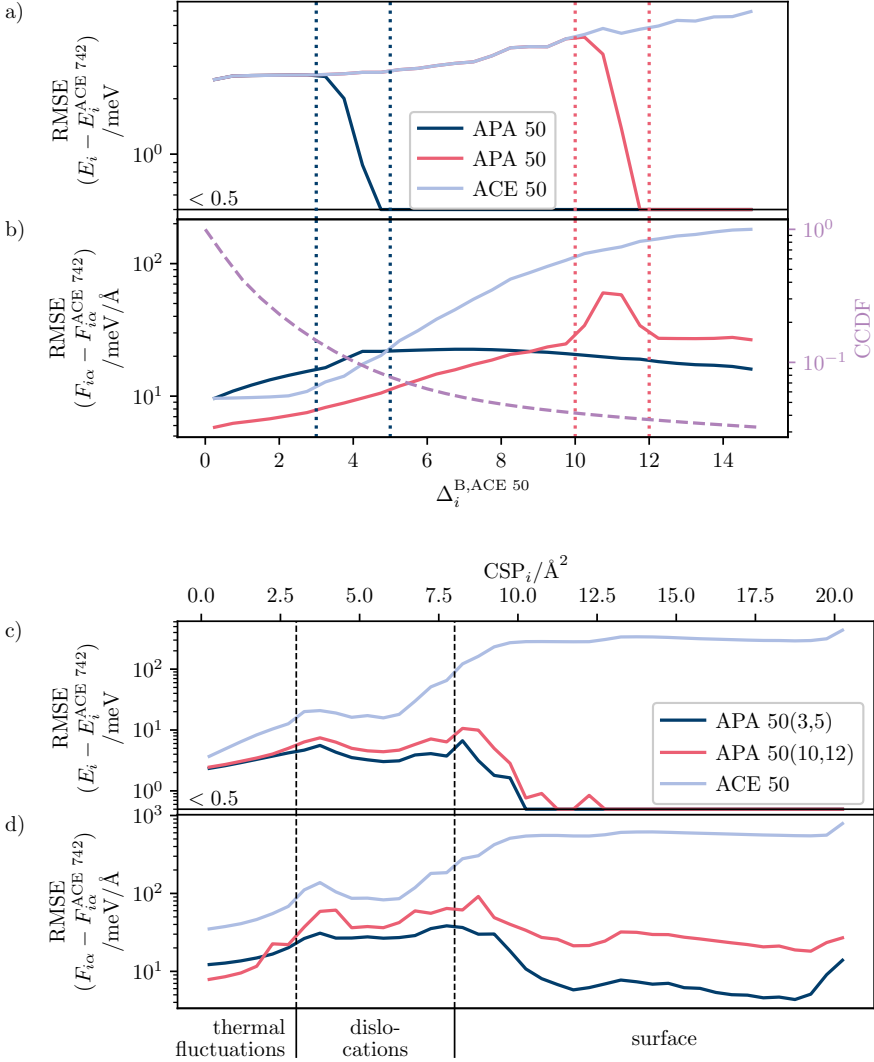


Figure 3.11: Analysis of the energy and force precision achieved of APA 50 dependent on the thresholds $\Delta_{\text{low}}^{\text{B,ACE 50}}$ and $\Delta_{\text{high}}^{\text{B,ACE 50}}$ (cf. Eq. (3.36)). The analysis is done for the in Fig. 3.6 visualised snapshot of a nanoindentation of (100) copper at room temperature in an indentation depth of 20 Å. a-b) The atoms are sorted into bins of width 0.5 dependent on $\Delta_i^{\text{B,ACE 50}}$ and the RMSE is calculated per bin for a) potential energies and b) forces. The shown range of $\Delta_i^{\text{B,ACE 50}}$ is truncated at 15 to allow the visualisation with the chosen bin width. The varied $\Delta_{\text{low}}^{\text{B,ACE 50}}$ and $\Delta_{\text{high}}^{\text{B,ACE 50}}$ are drawn as vertical dotted lines in the colour of the corresponding APA graph. Furthermore, the complementary cumulative distribution function (CCDF) is shown as dashed graph in b) to indicate the fraction of the system which requires the computation of ACE 742. c-d) The atoms are sorted into bins of width $0.5 \text{ \AA}^{-2}$ dependent on the centro-symmetry parameter (CSP) and the RMSE is calculated per bin for c) potential energies and d) forces. As the CSP detect asymmetries that are characteristic for dislocations and surface atoms, the corresponding ranges of the CSP are shown on the bottom axis. The precision of ACE 50 is shown for comparison.

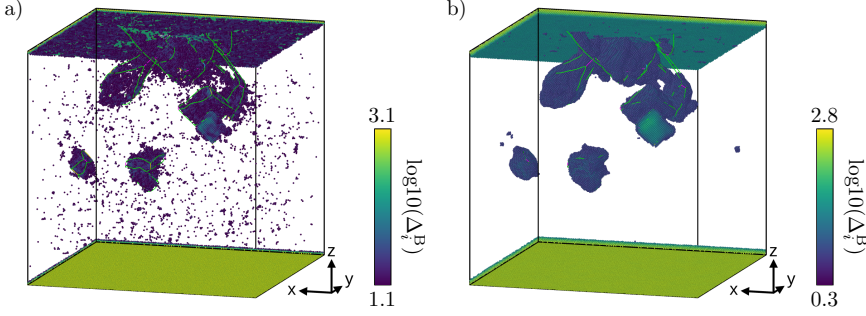


Figure 3.12: Visualisations of an at room temperature simulated nanoindentation of a (100) copper surface for an indentation depth of 20 Å. The coloured lines are dislocation lines which are identified by the dislocation extraction algorithm (DXA)[142] of OVITO[143]. a) Additionally, all atoms i with $\Delta_i^{\text{B,ACE } 50} \geq \Delta_{\text{high}}^{\text{B,ACE } 50}$ (cf. Eq. (3.33)) are shown and coloured according to $\Delta_i^{\text{B,ACE } 50}$. b) Additionally, all atoms i with $\bar{\Delta}_i^{\text{B,ACE } 50} \geq 6.0$ (cf. Eq. (3.51)) are shown and coloured according to $\bar{\Delta}_i^{\text{B,ACE } 50}$.

parameter (CSP) only decreases from $91.5 \text{ meV}\text{\AA}^{-1}$ to $38.4 \text{ meV}\text{\AA}^{-1}$ for the least precise bin as shown in Fig. 3.11d, where the force precision is in particular bad for dislocations. Therefore, simply increasing the number of ACE 742 evaluations by decreasing the thresholds $\Delta_{\text{low}}^{\text{B}}$ and $\Delta_{\text{high}}^{\text{B}}$ does not solve the problem of the partially bad force precision. Furthermore, the fraction of atoms that require the computation of the expensive atomic cluster expansion of ACE 742, is increased by the factor of 3.5 to 15% as shown in Fig. 3.11b.

Although the influence of the gradient of the switching function, i.e., the influence of $\mathbf{f}_{ik}^{\text{L,ACE } 50}$, was significantly decreased by decreasing the thresholds $\Delta_{\text{low}}^{\text{B,ACE } 50}$ and $\Delta_{\text{high}}^{\text{B,ACE } 50}$, the force precision is still in particular bad for dislocations. As visualised in Fig. 3.6b-c, $\Delta_i^{\text{B,ACE } 50}$ is above the defined thresholds for atoms within dislocation patterns, but not for atoms outside of the dislocation patterns, i.e., the dislocation lines are clearly visible. Thus, there are many atoms i in the force cutoff of dislocations k for which $\lambda_i^{\text{B,ACE } 50} = 1$ applies and which decrease the force precision of atom k (cf. Eq. (3.49)). Therefore, to increase the force precision of dislocations k , one needs to increase the fraction of neighbouring atoms i which have $\lambda_i^{\text{B,ACE } 50}$. Simply reducing the thresholds $\Delta_{\text{low}}^{\text{B,ACE } 50}$ and $\Delta_{\text{high}}^{\text{B,ACE } 50}$ is not enough as shown in Fig. 3.11d. Therefore, the computation of the switching parameter itself, i.e., Eq. (3.36), needs to be modified.

Instead of the deviation $\Delta_i^{\text{B,ACE } 50}$ of the reference environment, one could use the average deviation in the force cutoff of atom i which is given as

$$\bar{\Delta}_i^{\text{B,ACE } 50} = \frac{1}{N_i^{\text{ngb}}} \sum_{j \in \{k | r_{ik} \leq r_{\text{cut}}^{\text{ACE } 50}\}} \Delta_j^{\text{B,ACE } 50}, \quad (3.51)$$

where N_i^{ngb} denotes the number of atoms within the force cutoff of atom i , i.e., the number of atoms in the set over which the sum is calculated. The deviation $\Delta_i^{\text{B,ACE } 50}$ fluctuates for bulk atoms and $\Delta_i^{\text{B,ACE } 50} > \Delta_{\text{high}}^{\text{B,ACE}}$ applies by design (cf. Section 3.3.2) also far away from dislocations and the surface as visualised in Fig. 3.12a. The usage of the local average $\bar{\Delta}_i^{\text{B,ACE } 50}$ allows to average out single atoms with $\Delta_i^{\text{B,ACE } 50} > \Delta_{\text{high}}^{\text{B,ACE}}$. Furthermore, when $\Delta_i^{\text{B,ACE } 50} > \Delta_{\text{high}}^{\text{B,ACE}}$ applies for few atoms i next to each other, these atoms can influence $\bar{\Delta}_j^{\text{B,ACE } 50}$ and thus the switching parameter of common neighbours j . Both advantages are visible in Fig. 3.12b. Some dislocation lines are not visible when $\bar{\Delta}_i^{\text{B,ACE } 50}$ is used and hence the fraction of precisely described atoms in the force cutoff of dislocations has improved. However, there are still dislocation lines visible. Thus, the suggested modification of the switching parameter would increase the force precision of dislocations, but still needs to be optimised. The drawback of the suggested modification is the increased computation cost as the calculation of $(\nabla_k \lambda_i^{\text{B}})$ is far more complicated when the local average of the deviation $\Delta_i^{\text{B,ACE } 50}$ is used.

As the force contribution of the gradient of the switching function decreases the force precision, one could try to neglect this force contribution. However, assuming $(\nabla_k \lambda_i^{\text{B}}) \approx 0$ in Eq. (3.38)

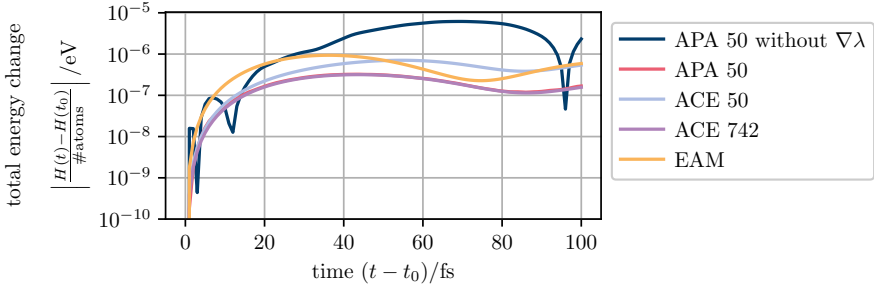


Figure 3.13: Change of the total energy H over time dependent on the used interatomic potential. The system at the time t_0 is similar to the nanoindentation visualised in Fig. 3.6, but contains only 1.26 million instead of 4 million atoms. The system is simulated in a NVE ensemble, in which the indenter was removed for this simulation to ensure energy conservation. The EAM2 potential of Ref. [145] is given for comparison.

implies that the potential would no longer be conservative. The total energy of a system is given as

$$H = \sum_i (T_i + E_i) + E_{\text{ext}}, \quad (3.52)$$

where T_i denotes the kinetic energy of an atom i and E_{ext} the energy of possibly existing external fields. The energy conservation for a simulation starting from the analysed snapshot of the nanoindentation without the indenter is shown in Fig. 3.13 for different potentials. For the non-conservative APA 50 potential in which the gradient of the switching parameter is neglected, the maximum energy change is 6.6 times larger in than for the conservative potentials, where only 100 fs, i.e., 100 timesteps, were simulated. This small test shows that one cannot simply neglect the gradient of the switching parameter. To neglect this gradient, the switching parameter needs to change adiabatically slow. Furthermore, an energy correction to ensure energy conservation exactly would be required.

To summarise, one can increase the force-precision near dislocations and surfaces by modifying the switching parameter (cf. Eq. (3.51)) or decreasing the thresholds $\Delta_{\text{low}}^{\text{B,ACE } 50}$ and $\Delta_{\text{high}}^{\text{B,ACE } 50}$, but both optimisations increase the computational cost. Furthermore, neglecting the gradient of the switching parameter would improve the force-precision, but would require corrections in order to conserve the energy.

3.3.4 Computing time

The precision of APA 50 which was developed in Sections 3.3.1 and 3.3.2, was analysed in Section 3.3.3. The main objective of an adaptive-precision ACE potential is a reduction of the computational time which would be required with a precise potential, i.e., to achieve a speedup compared to the precise potential. Therefore, the computational time of the 1.26 million atom simulation visualised in Fig. 3.13 is analysed in the following, where the achieved speedup is shown in Section 3.3.4.1 and further improvements are discussed in Section 3.3.4.2.

3.3.4.1 Demonstration

The required core-h of the energy-conservation test (cf. Fig. 3.13), i.e., a simulation of 1.26 million atoms for 100 timesteps, is shown in Fig. 3.14. Although there was no load balancing, the slowest core takes in the imbalanced simulation on average only 1.08 times more time than the average core. Therefore, load imbalances are negligible due to the small amount of used cores. The speedup of ACE 10 and ACE 50 compared to the precise ACE 742 simulation is 19.0 and 3.3. Thus, the fitted fast ACE potentials are faster than the precise ACE 742 potential. The fast ACE potentials are not applicable for dislocations and surfaces as they incorrectly extrapolate both potential energies and forces (cf. Fig. 3.8). APA 50 prevents this incorrect extrapolations although it does not reach the same force-precision like ACE 50 in the reference data $\mathcal{S}^{\text{ACE},f}$ for all atoms as discussed previously (cf. Fig. 3.8). The speedup of APA 50 in the 4 million atom simulation with dislocations and surface atoms is 2.2 compared to ACE 742. The APA 10 potential combines ACE 10 and ACE 742

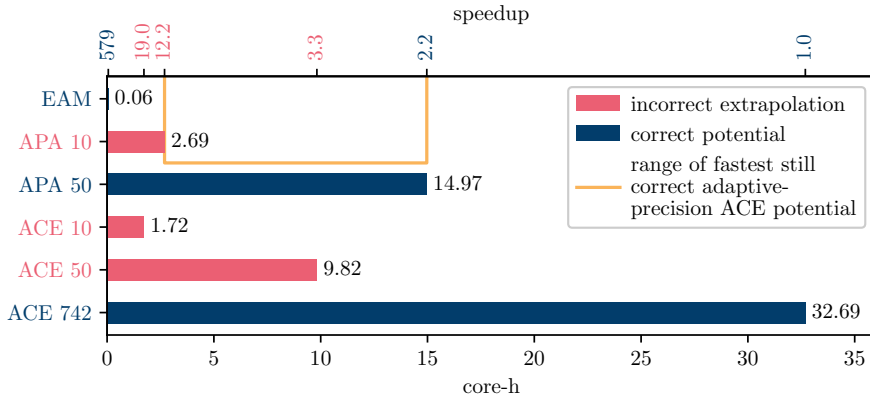


Figure 3.14: Required core-h of the energy-conservation test visualised in Fig. 3.13 as bar chart. The speedup of each simulation compared to the precise ACE 742 simulation is given on the top axis. The bars of potentials that are incorrectly extrapolated in this simulation are coloured red and the remaining bars are coloured blue. Furthermore, the horizontal range, i.e., core-h and speedup, of the fastest APA potential that does not include incorrect extrapolations is indicated in orange. The EAM2 potential of Ref. [145] and APA 10 are given for comparison, where the thresholds $\Delta_{\text{low}}^{\text{B,ACE } 10}$ and $\Delta_{\text{high}}^{\text{B,ACE } 10}$ of APA 10 are set according to the same criterion like for APA 50 based on Fig. 3.5. The simulations were run with LAMMPS on six cores of the Intel(R) Core(TM) i7-8700 CPU @ 3.20GHz.

via a switching parameter based on $\Delta_i^{\text{B,ACE } 10}$, where the thresholds $\Delta_{\text{low}}^{\text{B,ACE } 10}$ and $\Delta_{\text{high}}^{\text{B,ACE } 10}$ are set according to the same criterion like for APA 50 based on Fig. 3.5. As the detection of atoms which need to be described with ACE 742 does not work with $\Delta_i^{\text{B,ACE } 10}$ (cf. Fig. 3.6), the speedup of 12.2 of APA 10 compared to ACE 742 only shows a limitation of the presented adaptive-precision ACE method. Thus, the fastest APA potential which does not incorrectly extrapolates needs to use more than 10 and less than 50 cluster basis functions as basis. Therefore, one can expect from the APA method a maximum speedup between 12.2 and 2.2 for this case study.

In the APA 50 simulation 8.6% and 8.4% of the atoms have $\lambda_i^{\text{B,ACE } 50} < 1$ in the first and last timestep, i.e., about 8.5% of the atoms require the calculation of the precise but expensive ACE 742 potential. Therefore, this system is a realistic example which can benefit from the reduced computational time by an APA potential. With a fraction of 8.5% ACE 742 evaluations the computational time of APA 50 is ideally $(32.69 \times 0.085 + 9.82 \times 0.915)$ core-h = 11.76 core-h, where the computational times of ACE 742 and ACE 50 are used (cf. Fig. 3.14). The difference of 3.21 core-h to the computational time measured for APA 50 is caused by the calculation of detection mechanism. The computation of $\Delta_i^{\text{B,ACE } 50}$ causes overhead as the required basis of cluster basis functions is slower to evaluate than the basis of many-body functions used for ACE 50 (cf. Section 3.1.3).

3.3.4.2 Optimisation potential

In the energy-conservation test simulated with APA 50, about 8.5% of the atoms required a precise computation. With this fraction of precise evaluations, the highest possible speedup is $1/0.085 \approx 11.8$. The by APA 50 achieved speedup is however only 2.2 as thereby about 5 times smaller. This shows that the speedup is not only limited by the fraction of the system that requires a precise calculation, but also by the computation time of fast potential and detection mechanism. In order to optimise the speedup of an adaptive-precision potential, one should use both a faster potential than ACE 50 for the not precisely calculated atoms and a faster computable switching parameter than λ_i^{B} based on $\Delta_i^{\text{B,ACE } 50}$. The EAM potential used in the energy-conservation test is 579 times faster than ACE 742, where ACE 50 is only 3.3 times faster than ACE 742. Therefore, using an EAM potential as fast potential rather than an ACE potential would be beneficial.

3.4 Conclusion

In this chapter, the atomic cluster expansion (ACE) was summarised first in Section 3.1. Based on ACE, an adaptive-precision ACE (APA) potential was developed in Section 3.2 and the APA 50 copper potential was created as demonstration in Section 3.3. The by APA 50 achieved precision was discussed in Section 3.3.3 and the achieved speedup in Section 3.3.4, where optimisation potential is given in both cases.

One can decrease the basis size of an ACE potential to speedup the computation of an ACE potential. By narrowing the broad range of reference structures, one can keep the precision in the vicinity of a reference atomic environment at a reduced computation cost. One can then use the potential energy of the fast ACE potential in the vicinity of the reference environment and only use the original potential which is precise but expensive for the remaining atoms (cf. Eq. (3.27)).

The ACE copper potential of Ref. [4] was used as precise potential and denoted as ACE 742 due to the size of its basis. To fit a fast ACE potential which is as similar as possible to ACE 742 in the vicinity of a reference environment, one can use MD reference data calculated with the precise ACE potential instead of DFT reference data. Thereby, the reference data contains also potential energies per atom which are not available in DFT data. The use of this potential energies per atom as reference instead of the total potential energy of the structure (cf. Eqs. (3.21) and (3.28)) improves the fitting of a fast ACE potential (cf. Fig. 3.2). Two fast ACE potentials were fitted, i.e., ACE 10 and ACE 50, where the number denotes the number of cluster basis functions which are used as basis for the corresponding potential. The calculation of basic properties with the fitted fast ACE potentials shows that the potentials only work in the vicinity of the reference environment. Vacancies and surfaces are described badly by the fitted potentials (cf. Table 3.1).

The cluster basis functions (cf. Eq. (3.7)) are used to measure the deviation Δ_i^B of a configuration from the reference configuration (cf. Eq. (3.33)). This deviation is used to calculate the switching parameter λ_i^B according to Eq. (3.36). Atoms whose deviation from the reference environment is present in the MD reference dataset $\mathcal{S}^{\text{ACE},f}$ can be described by the potential energy of the fast ACE potential. Thereby, one can save computation time for these fast calculated atoms as the calculation of the precise and expensive atomic cluster expansion is not required. The switching parameter ensures that the potential energy of an atom i is described only with the precise potential energy when the deviation Δ_i^B from the reference environment is larger than in the reference dataset to which the fast potential was fitted (cf. Section 3.3.2), i.e., the extrapolation of the potential energy of the fast potential is prevented.

Testing the developed deviation-calculation mechanism in a snapshot of a nanoindentation (cf. Fig. 3.6) showed that $\Delta_i^{\text{B},\text{ACE } 10}$ does increase at the surface, but does not increase near dislocations. Therefore, one cannot use the adaptive-precision ACE potential APA 10 which combines ACE 10 and ACE 742 for structures that contain dislocations. However, one can use APA 50 which combines ACE 50 and ACE 742 according to Eq. (3.45) as $\Delta_i^{\text{B},\text{ACE } 50}$ increases both near dislocations and surfaces. The comparison of $\Delta_i^{\text{B},\text{ACE } 10}$ and $\Delta_i^{\text{B},\text{ACE } 50}$ shows that the detection of unknown environments with Δ_i^B improves with the size of the used basis and that the basis can be too small.

A snapshot of a nanoindentation served as test system for an adaptive-precision ACE potential in Section 3.3.3.1. As this snapshot contains dislocations, the APA 50 potential was used. The precision achieved of APA 50 in this nanoindentation snapshot is 10.6 meV for potential energies and 91.5 meVÅ⁻¹ for forces, where the force precision is worst for dislocations but better at the surface or for thermally fluctuating atoms (cf. Fig. 3.8c-d). The analysis in Section 3.3.3.2 shows that the force components $\mathbf{f}_{ki}$ (cf. Eq. (3.19)) of ACE 50 and ACE 742 are similar, but not equal (cf. Fig. 3.9). Therefore, the force-precision decreases, when force contributions of ACE 50 and ACE 742 are combined to the total force $F_k^{\text{APA } 50}$ (cf. Eqs. (3.46) and (3.47)). The force on atom k depends on the switching parameters λ_i^B of all atoms i within the force cutoff of atom k . Therefore, one can increase the precision of an adaptive precision potential for an atom k by decreasing the switching parameters of neighbouring atoms i as discussed in Section 3.3.3.3. Furthermore, the calculation of $(\nabla_k \lambda_i^B)$ in Eq. (3.38) decreases the force precision. Hence, neglecting this gradient of the switching parameter would improve the force precision but also violate energy conservation (cf. Fig. 3.13). Therefore, one would need to apply an energy correction to compensate for the change of the total energy H (cf. Section 3.3.3.3).

A simulation of 1.26 million copper atoms for 100 fs starting from a nanoindentation snapshot served as test to analyse the speedup compared to a precise simulation with ACE 742 (cf. Fig. 3.14). The speedup of APA 50 is 2.2, where about 8.5% of the atoms required the computation of the expensive ACE 742 potential. The speedup could be improved by further reducing the basis size of the fast ACE potential, but the optimisation possibilities are limited as a basis size of 10 with a

speedup of 12.2 is too small and results in incorrect extrapolations (cf. Section 3.3.4.1). Note, that the speedup depends on the fraction of not required ACE 742 computations and therefore strongly depends on the system. However, one could improve the speedup of an adaptive-precision potential by using not a fast ACE potential but a faster potential. This speedup optimisation would require a replacement of Δ_i^B in the switching parameter by a faster computable quantity that does not rely on a fast ACE potential as discussed in Section 3.3.4.2.

EAM is 579 times faster than ACE 742 while ACE 10 and ACE 50 are only 19.0 and 3.3 times faster than ACE 742 (cf. Fig. 3.14). The precision of EAM in the nanoindentation snapshot is 321.9 meV for the potential energy and 103.8 meV Å for forces. Thus, EAM is 30.3 and 1.1 times less precise for potential energies and forces than APA 50 compared to ACE 742. As forces dominate the dynamics and the EAM potential is 263 times faster than APA 50 (cf. Fig. 3.14), one can directly use a fast but less precise potential like EAM and does not need an adaptive-precision ACE potential. However, the construction and analysis of the adaptive-precision ACE potential has shown five improvement options.

1. The fast potential, which is combined with a precise potential, should be as fast as possible in order to optimise the achievable speedup.
2. Detecting atoms, whose environment is not similar to the reference environment, should be as fast as possible in order to optimise the achievable speedup.
3. The switching parameter should be 0 in the force cutoff of atoms that require a precise calculation. Thus, the switching parameter requires an improved calculation method.
4. The switching parameter should change continuously in space from 0 to 1 over few atoms to minimise the force-precision loss introduced by combining force contributions of two different potentials.
5. The computation of the gradient of the switching parameter decreases the force precision and should thus be prevented. Hence, neglecting this force contribution and compensate for the thereby introduced energy-conservation error would improve the force precision.

These findings can be used to construct a both faster and more precise adaptive-precision model.

Chapter 4

Adaptive-precision EAM-ACE coupling

The present chapter is based on my paper *Adaptive-precision potentials for large-scale atomistic simulations*[146].

There is still a considerable performance gap between the evaluation of classical fields and ML potentials, e.g., the force calculation for a classical EAM potential and the ML-based ACE method differ by a factor of 100-1000[4]. The 100 fs-simulation of 1.26 million copper atoms in Fig. 3.14 served as example with a core-h difference of the factor 579. Therefore accessibility of time and length scales of systems is limited by the performance gap between classical force fields and modern ML potentials which results in a conflict between system size and precision. This conflict of objectives also worked out in Section 3.3.3.3 where all precision increasing optimisations would also increase the required computational time of the adaptive-precision ACE potential.

With the advent of ML potentials, coupling of different interaction models shows strong potential for combining a high accurate description in small-to-medium sized subsystems with acceptable computational costs and a low accurate description in large subsystems, opening the path to long time and large length scale simulations including high accuracy, where it is needed. Such a coupling between a ML-potential and a classical force-field description has been proposed in Ref. [92] using a force-mixing approach to simulate a grand-canonical ensemble in the thermodynamic limit.

In contrast, in the present work, we use an energy-mixing approach which is consistent with a Hamiltonian description and allows, in principle, the simulation of a microcanonical ensemble. In so doing we introduce a method to couple a precise ML-potential for subregions of atoms of interest and a fast traditional potential for the remaining system components to overcome the conflict between system size and precision for classical atomistic simulations. The atoms of interest are thereby automatically detected by a customisable detection mechanism and therefore the method works autonomously and self adaptively in space and time. To further reduce the execution time we implemented the method into the parallel simulation engine LAMMPS[1].

Due to the change of energy and force description in the two regions (high-/low-accuracy), a spatial zone is usually introduced, which provides a smooth transition from one into the other region[94, 107]. Two ACE potentials with the same force cutoff and described by the same underlying equations are very similar as shown in Fig. 3.9. Such a spatial zone is even required for the coupling of two such similar potentials as discussed in Section 3.3.3.3. Therefore, we introduce such a spatial zone in the calculation of the switching parameter. The energy-based coupling approach according to Eq. (2.17)) is used as force-based approaches cannot conserve the energy (cf. Section 2.4).

The present chapter is organised as follows: as we aim for a coupling of a traditional with a ML potential, the embedded atom model (EAM) is summarised as an exemplary traditional potential in Section 4.1 and compared with ACE (cf. Section 3.1) as exemplary ML potential. Then, we introduce our adaptive-precision model for the combination of a fast, traditional potential with a precise but expensive one in Section 4.2. Section 4.3 shows the application of the introduced adaptive-precision mechanism, i.e., an EAM potential is combined with an ACE potential for copper. Finally, we demonstrate in Section 4.4 the capabilities in terms of precision and efficiency of the adaptive-precision copper potential for a nanoindentation of 4×10^6 atoms calculated for 100 ps with LAMMPS[1]. In Section 4.5 we conclude.

Table 4.1: Comparison of Pearson correlation coefficients(PCC), root mean squared error (RMSE) and mean of the in Fig. 4.1 visualised $f_{ki\alpha}^{\text{EAM}}$ and $f_{ki\alpha}^{\text{A,ACE 742}}$. The used EAM potential is EAM2 of Ref. [145].

neighbour shell	1	2	3	4	5	6	7	8
$\langle r_{ki} \rangle_{ki} / \text{\AA}$	2.56	3.62	4.43	5.12	5.72	6.27	6.76	7.23
$\langle f_{ki\alpha}^{\text{EAM}} \rangle_{ki\alpha} / \text{eV \AA}^{-1}$	4.7e-2	1.6e-2	7.0e-3	8.9e-4	2.0e-7	0	0	0
$\langle f_{ki\alpha}^{\text{A,ACE 742}} \rangle_{ki\alpha} / \text{eV \AA}^{-1}$	2.1e-1	1.9e-2	5.9e-2	2.0e-2	1.7e-3	2.7e-3	1.2e-3	2.4e-4
RMSE($f_{ki\alpha}^{\text{EAM}}$ $- f_{ki\alpha}^{\text{A,ACE 742}}) / \text{eV \AA}^{-1}$	3.0e-1	5.6e-2	7.1e-2	2.4e-2	2.7e-3	2.9e-3	1.4e-3	4.8e-4
PCC($f_{ki\alpha}^{\text{EAM}}, f_{ki\alpha}^{\text{A,ACE 742}}$)	-0.63	-0.75	-0.99	0.58	0.22	0.00	0.00	0.00

4.1 Embedded atom model

The embedded atom model (EAM) is a fast, classical interatomic potential. In Section 4.1.1, a more detailed description of the potential and force calculation is provided. The forces calculated with a copper EAM potential are compared in Section 4.1.2 to the ones calculated with an ACE potential.

4.1.1 Energy and forces

The energy of an atom i described with the embedded atom model (EAM)[53] is given by

$$E_i^{\text{EAM}} = \xi(\zeta_i) + \frac{1}{2} \sum_{j \neq i} \Phi(r_{ij}), \quad (4.1)$$

where ξ is the embedding function, Φ a pair potential and the electron charge density ζ_i is given as

$$\zeta_i = \sum_{j \neq i} \zeta(r_{ij}), \quad (4.2)$$

where $\zeta(r_{ij})$ is the contribution to the electron charge density. Thus, an EAM potential is defined by three functions: 1. embedding function $\xi(\zeta_i)$, 2. charge density $\zeta(r)$ and 3. pair potential $\Phi(r)$. These three functions are parametrised differently for each EAM potential and even their proposed form varies between different EAM potentials for the same element (cf. Refs. [147, 148]).

The force is given according to Eq. (2.2) and can be calculated analogously with the equivalent of Eqs. (3.16) and (3.17), i.e.

$$\mathbf{F}_k^{\text{EAM}} = \sum_i (\mathbf{f}_{ik}^{\text{EAM}} - \mathbf{f}_{ki}^{\text{EAM}}), \quad (4.3)$$

where the force component

$$\mathbf{f}_{ki}^{\text{EAM}} = \left(\frac{\partial \xi}{\partial \zeta} \bigg|_{\zeta=\zeta_i} \frac{\partial \zeta}{\partial r} \bigg|_{r=r_{ki}} + \frac{1}{2} \frac{\partial \Phi}{\partial r} \bigg|_{r=r_{ki}} \right) \hat{\mathbf{r}}_{ki} \quad (4.4)$$

is introduced to shorten the equation.

4.1.2 Comparison with ACE

In this section, the force contributions of EAM and ACE are compared for two copper potentials. For simplicity, the for the comparison used structures contain only bulk atoms and no defects or surfaces. The ACE 742 potential of Ref. [4] that was used in Section 3.3 as precise potential is used as ACE potential. The EAM2 potential of Ref. [145] is used as EAM potential as it is widely used[149–152].

The RMSE of the force components $F_{i\alpha}$ is dependent on the value of $F_{i\alpha}$ between $26 \text{ meV \AA}^{-1}$ and $176 \text{ meV \AA}^{-1}$, where the large RMSE occurs for large forces that only rarely occur (cf. Fig. 4.1i). Thus, 91% of the force components $F_{i\alpha}$ of ACE and EAM differ from each other by less than $50 \text{ meV \AA}^{-1}$. The force of an adaptive-precision potential is, however, according to Eq. (3.42), given by the force contributions $\mathbf{f}_{ki}$ of first the precise potential, second the fast potential and third the gradient of the switching parameter. Hence, the force contributions $f_{ki\alpha}$ of EAM and ACE

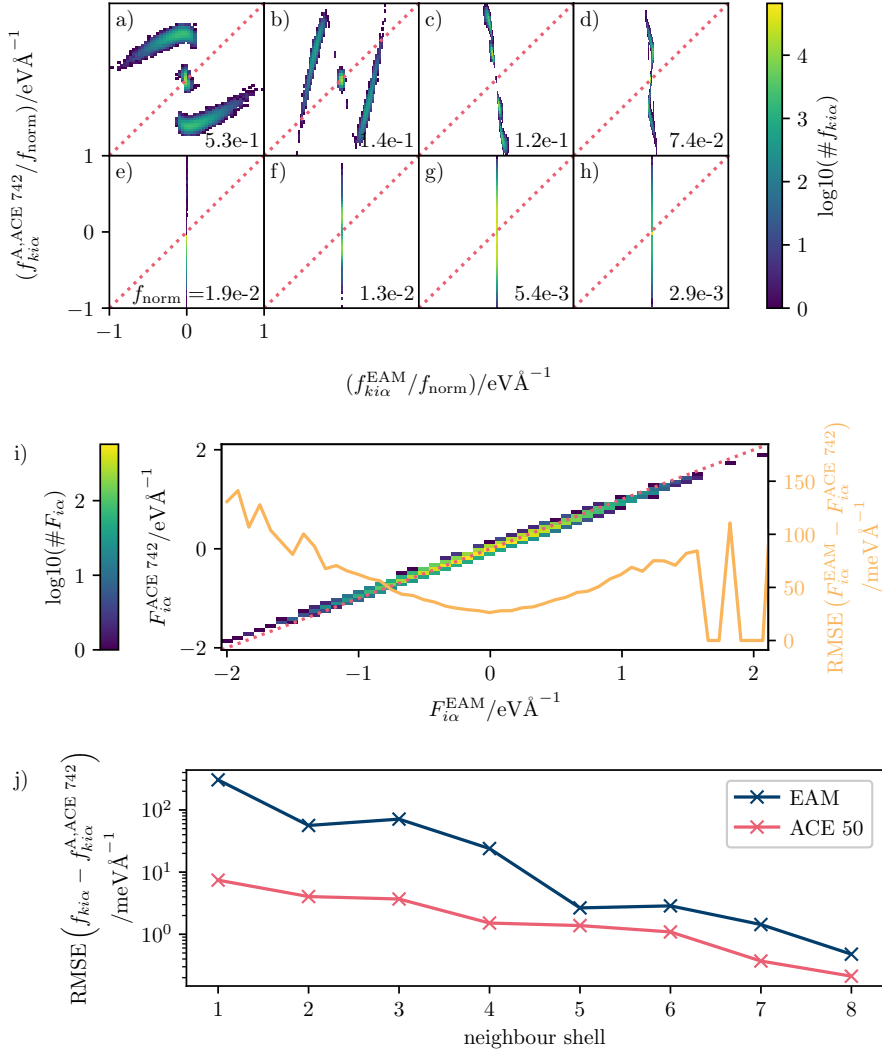


Figure 4.1: a-h) Comparison of f_{ki}^{EAM} (cf. Eq. (4.4)) calculated with a copper EAM potential and $f_{ki}^{\text{ACE 742}}$ (cf. Eq. (3.19)) calculated with ACE 742. The comparison is done for one structure of the reference dataset $\mathcal{S}^{\text{EAM,Cu}}$ (cf. Section 4.3.1). The three scalar components $f_{ki\alpha}$ are used rather than the vector $\mathbf{f}_{ki}$. The force components are compared per neighbour shell in ascending order from a) to h), i.e., k is in the first neighbour shell of i in a) and in the eighth neighbour shell in h). For better visualisation, the force components are normalized with f_{norm} per neighbour shell, where f_{norm} is given in the lower right corner of each subplot. Thereby, the axis range is $[-1, 1]$ for all subplots and only annotated in e). The dotted line shows where $f_{ki\alpha}^{\text{EAM}} = f_{ki\alpha}^{\text{ACE 742}}$ applies. i) 2D-histogram of the force components $F_{i\alpha}^{\text{EAM}}$ and $F_{i\alpha}^{\text{ACE 742}}$, where the colourbar is shown next to the plot. The RMSE is calculated per $F_{i\alpha}^{\text{EAM}}$ -bin and shown on the right-hand vertical axis for comparison. The dotted line indicates equality of EAM and ACE force component. j) The RMSE of the pair force contributions $f_{ki\alpha}$ of EAM and ACE 50 compared to ACE 742 is shown dependent on the neighbour shell of the atom pair ki . The potential EAM2 of Ref. [145] is used as copper EAM potential for all subplots.

are compared in Fig. 4.1a-h. The EAM potential has a force cutoff of $5.5 \text{ \AA}$ in contrast to $7.4 \text{ \AA}$ of ACE 742. Thus, the force contributions of neighbours necessarily differ from the fifth to the eighth neighbour shell (cf. average distance $\langle r_{ki} \rangle_{ki}$ per shell in Table 4.1), i.e., $\mathbf{f}_{ki}^{\text{EAM}} = \mathbf{0}$ while $\mathbf{f}_{ki}^{\text{ACE 742}} \neq \mathbf{0}$. Due to the different force cutoff, one expects that the force components differ also in the last common neighbour shell of both potentials, i.e., the fifth shell, to compensate for the missing outer neighbour shells. While this expectation is fulfilled for two ACE potentials with different force cutoffs, the force contributions differ completely also in the first four neighbour shells for EAM and ACE. ACE and EAM potential are described by completely different force equations and built a very similar force $\mathbf{F}_{i\alpha}$ (cf. Fig. 4.1i) by different force contributions (cf. Fig. 4.1a-h). The RMSE of the force contributions of EAM compared to ACE 742 decays from $304.8 \text{ meV \AA}^{-1}$ in the first neighbour shell to $0.5 \text{ meV \AA}^{-1}$ in the eighth neighbour shell as visualised in Fig. 4.1j. In contrast, the RMSE of the force contributions of ACE 50 (cf. Section 3.3.1) compared to ACE 742 decays from $7.4 \text{ meV \AA}^{-1}$ in the first neighbour shell to $0.2 \text{ meV \AA}^{-1}$ in the eighth neighbour shell (cf. Fig. 4.1j). Therefore, the coupling of EAM and ACE requires, in contrast to the coupling of two ACE potentials, a spatial switching zone in which the switching parameter smoothly changes from 0 to 1. The force on an atom i of an adaptive-precision potential depends on the switching parameters λ_j of all atoms j in the force cutoff, as discussed in Section 3.3.3.2. Thus, a spatial zone needs also ensure that all $\mathbf{f}_{ki}^{\text{EAM}}$ of the first four neighbour shells with a RMSE of $304.8 \text{ meV \AA}^{-1}$ to $23.9 \text{ meV \AA}^{-1}$ are not used for the computation of forces that require a high precision. Therefore, the spatial zone needs to keep the switching parameter of 0 for about $5.5 \text{ \AA}$ and start the smooth transition to a switching parameter of 1 afterwards.

4.2 Adaptive-precision coupling method

Our model for a coupling of a fast traditional interatomic potential with a precise ML potential is presented in the following. First, the representation of energy and forces is given in Section 4.2.1. The construction of the switching parameter, which determines the used potential per atom, is discussed in Section 4.2.2 and the integration of the equations of motion in Section 4.2.3. With adaptive-precision and dynamic precision-selection in a parallel simulation comes the need for dynamic load balancing as the computational time changes over magnitudes between atoms. Therefore, we present our dynamic load-balancing method in Section 4.2.4. Finally, in Section 4.2.5 is shown how a timestep of a MD simulation works with the presented method.

4.2.1 Representation of energy and forces

The total energy of the system is given as $H = \sum_i (T_i + E_i) + E_{\text{ext}}$ (cf. Eq. (2.1)), where T_i is the kinetic and E_i the potential energy of atom i and E_{ext} are possibly existing external fields. The energy of our adaptive-precision approach combines a precise and a fast energy, $E_i^{(\text{p})}$ and $E_i^{(\text{f})}$, per atom i . We will use the atomic cluster expansion (ACE)[40] as precise and the embedded atom model (EAM)[53] as fast interatomic potential. Precise and fast energies are combined with a continuous switching parameter $\lambda_i \in [0, 1]$ per atom i (cf. Eq. (2.17)), namely to the adaptive-precision potential energy

$$E_i^{\text{AP}} = \lambda_i E_i^{(\text{f})} + (1 - \lambda_i) E_i^{(\text{p})}. \quad (4.5)$$

During the course of a simulation the energy will be switched automatically from fast to precise or vice versa as required by the local atomic environment. The switching parameter λ_i needs to change continuously to prevent shocks by instant energy changes. One can save computation time by using the adaptive-precision energy E_i instead of the precise energy $E_i^{(\text{p})}$ since the precise calculation is only required for a subset of atoms in the adaptive-precision calculation. One needs to make sure that the energies $E_i^{(\text{f})}$ and $E_i^{(\text{p})}$ are as similar as possible for the atoms, which are calculated with the fast potential, to prevent a systematic energy change due to the change of the switching parameter. The adaptive-precision energy according to Eq. (4.5) gives the total potential energy $\sum_k E_k^{\text{AP}} = \sum_k (\lambda_k E_k^{(\text{f})} + (1 - \lambda_k) E_k^{(\text{p})})$ from where the adaptive-precision force $\mathbf{F}_i^{\text{AP}} = -\nabla_i \sum_k E_k^{\text{AP}}$ can be calculated as

$$\mathbf{F}_k^{\text{AP}} = \sum_i \left(-\lambda_i (\nabla_k E_i^{(\text{f})}) - (1 - \lambda_i) (\nabla_k E_i^{(\text{p})}) + (\nabla_k \lambda_i) (E_i^{(\text{p})} - E_i^{(\text{f})}) \right). \quad (4.6)$$

The force can be analogously calculated to Eqs. (3.38), (3.42) and (3.44) as

$$\mathbf{F}_k^{\text{AP}} = \sum_i (\mathbf{f}_{ik}^{\text{AP}} - \mathbf{f}_{ki}^{\text{AP}}), \quad (4.7)$$

where the force contributions are given as

$$\mathbf{f}_{ki}^{\text{AP}} = \left(E_i^{(\text{f})} - E_i^{(\text{p})} \right) \mathbf{f}_{ki}^{\lambda} + \lambda_i \mathbf{f}_{ki}^{(\text{f})} + (1 - \lambda_i) \mathbf{f}_{ki}^{(\text{p})}, \quad (4.8)$$

where $\mathbf{f}_{ki}^{\lambda}$ depends on the calculation mechanism of the switching parameter. For the coupling of a fast EAM and a precise ACE potential, the force contributions $\mathbf{f}_{ki}^{(\text{f})}$ are given by Eq. (4.4) and the force contributions $\mathbf{f}_{ki}^{(\text{p})}$ are given by Eq. (3.19).

It is important to note that $\lambda_i = 0$ is not the only requirement for a precise force on atom i since the force F_i depends according to Eq. (4.6) on the switching parameters λ_k of all atoms k within the force cutoffs of the interatomic potentials. Two potentials created according to different models (such as EAM and ACE) have qualitatively different mechanisms to construct a quantitatively only slightly different force (cf. Fig. 4.1). Therefore, a smooth transition zone between fast and precise energy description is essential to smoothly transfer the force-construction mechanism of the precise potential into the one of the fast potential.

4.2.2 Adiabatic switching

The switching function λ changes the energy-precision (cf. Eq. (4.5)) and thereby also the force-precision during simulation dependent on the atomic environment. This change of precision is ideally quasi-adiabatic to prevent perturbations of the system. The switching function itself has generally no physical meaning, it is only an auxiliary function to determine the precision. However, the force on atom i depends according to Eq. (4.6) on $(\nabla_i \lambda_k)$ and thus not only on the value of the switching parameter but also on the calculation mechanism. Therefore, an adiabatically slow change of the switching parameter is important to minimise the unphysical force contribution of $(\nabla_i \lambda_k)$.

The switching function needs a detection mechanism for particles that require a precise calculation. This detection mechanism strongly depends on the simulation. The centro-symmetry parameter[76] (CSP) detects, e.g., defects and surface atoms where the common neighbour analysis (CNA)[153] can characterise grain boundary structures[154]. The common neighbourhood parameter(CNP)[155] combines CNA and CSP. CSP, CNA and CNP are implemented in LAMMPS[156, pages 1828, 1843 and 1844] and therefore accessible for a wider community. Depending on the underlying physical system and simulation, other distinctions are possible. To detect, for example, stress-related events, the von-Mises stress can be used as an indicator. For dynamic simulations of atom bombardment like in Ref. [157] the particle velocity can be used and in simulations of damages, like for crack propagation, the strain energy can serve as indicator[107]. For a coating like in Ref. [158] one can include only atoms of the same element like the central atom i in the calculation of CSP_i to make the interface look like a surface.

The switching parameters $\{\lambda_k\}$ of all atoms k within the force cutoff of atom i determine the force-precision of atom i as discussed in Section 4.2.1. Therefore, when an atom i is detected for a precise calculation, the switching function needs to change also the switching parameters λ_k of the neighbouring atoms k to ensure a precise force calculation of atom i (cf. spatial switching zone in Section 4.1.2).

To detect defects and surfaces, the CSP serves here as detection mechanism. However, it can be changed to other criteria without big implementation overhead. The calculation of the CSP is discussed in Section 4.2.2.1. Afterwards, the construction of our switching function, taking into account the requirements discussed, is described in Section 4.2.2.2.

4.2.2.1 Centro-symmetry parameter

The centro-symmetry parameter (CSP) is given as

$$\text{CSP}_i(t) = \sum_{j=1}^{N/2} (\mathbf{r}_{i,j}(t) + \mathbf{r}_{i,j+N/2}(t))^2, \quad (4.9)$$

where N is the number of nearest neighbours and j and $j + N/2$ correspond to a pair of opposite nearest neighbours of the central atom i .

To calculate the CSP according to Ref. [76], initially reference vectors $\mathbf{r}_{i,j,\text{ref}}$ from the central atom i to its closest neighbouring atoms j are determined in an undistorted lattice. In the distorted lattice, one uses the distorted vectors $\mathbf{r}_{i,j}$ closest in distance to the undistorted reference vectors to calculate the centro-symmetry parameter $\text{CSP}_i^{\text{ref}} = \sum_{j=1}^{N/2} (\mathbf{r}_{i,j} + \mathbf{r}_{i,j+N/2})^2$, where j and $j + N/2$ correspond to a pair of opposite nearest neighbours of the central atom i . The set of distorted vectors used to calculate $\text{CSP}_i^{\text{ref}}$ may contain a neighbour multiple times and non-nearest neighbours.

LAMMPS[156, p. 1828] uses a different definition for the centro-symmetry parameter CSP^{imp} which does not require a reference system. At first, one searches the set $\mathcal{N}_i^{\text{imp}}$ of the N^{imp} nearest neighbouring atoms of the central atom i , where N^{imp} is the number of nearest neighbours in the undistorted lattice. Afterwards, the $N^{\text{imp}}/2$ pairs with the smallest $|\mathbf{r}_{i,j_1} + \mathbf{r}_{i,j_2}|$ with $j_1, j_2 \in \mathcal{N}_i^{\text{imp}}$ are used to calculate the centro-symmetry parameter. This definition may also contain duplicates, but relevant non-nearest neighbours may be neglected. This neglect of non-nearest neighbours is relevant for the computed CSP^{imp} of tungsten in a bcc lattice at 300 K like shown in Fig. 4.2. The effect of this neglect is reduced by searching $N + N_{\text{buffer}}$ rather than N nearest neighbours for $\mathcal{N}_i$. Thus, we calculate CSP^{imp} according to the LAMMPS-definition, but use $N + N_{\text{buffer}}$ atoms for the identification of the $N/2$ relevant opposite-neighbour pairs denoted as $j, j + N/2$.

The switching parameter needs to detect atoms for the precise calculation precisely to prevent too many evaluations of the precise and expensive potential. This requirement is fulfilled by the CSP since only neighbouring atoms similar to the expected neighbours in an undistorted lattice are used for the calculation. Missing or displaced atoms in the second neighbour-shell of the central atom do not influence the CSP. Hence, the nearest neighbour-shell of a defect, vacancy or surface atom is detected, but the second-nearest neighbour-shell is not. The latter behaviour might not be useful when low angle symmetric tilt grain boundaries, for example, are of interest as only a subset of atoms may be detected[159]. However, one can use the CSP to detect some grain boundaries[3].

4.2.2.2 Construction of the switching function

The centro-symmetry parameter according to Eq. (4.9) detects defects and surface atoms, which need to be calculated by the precise potential. Therefore, we use the CSP as starting point to construct the switching function for a nanoindentation. The centro-symmetry parameter fluctuates over time as atoms fluctuate dependent on their temperature. Since the thermal fluctuations of atoms should not be detected, we introduce a moving average

$$\text{CSP}_{\text{avg},i}(t) = \frac{1}{N_{\text{CSP,avg}}} \sum_{n=1}^{N_{\text{CSP,avg}}} \text{CSP}_i(t - n\Delta t) \quad (4.10)$$

of $N_{\text{CSP,avg}}$ timesteps Δt . The time-averaged centro-symmetry parameter can be used to calculate a switching parameter $\lambda_{0,i}$ as

$$\lambda_{0,i}(t) = f^{(\text{cut})} \left(\frac{\text{CSP}_{\text{avg},i}(t) - \text{CSP}_{\text{lo}}}{\text{CSP}_{\text{hi}} - \text{CSP}_{\text{lo}}} \right), \quad (4.11)$$

where we use the radial function

$$f^{(\text{cut})}(x) = \begin{cases} 1 & \text{for } x \leq 0, \\ 0.5 + 0.9375y - 0.625y^3 + 0.1875y^5 \text{ with } y = 1 - 2x & \text{for } 0 < x < 1, \\ 0 & \text{for } 1 \leq x. \end{cases} \quad (4.12)$$

$f^{(\text{cut})}$ is taken from Ref.[134] as the first and second derivative of $f^{(\text{cut})}$ are smooth at 0 and 1. $\lambda_{0,i}(t) = 1$ applies for all atoms with $\text{CSP}_{\text{avg},i}(t) \leq \text{CSP}_{\text{lo}}$. The use of a continuous switching

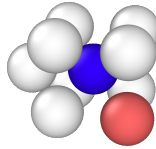


Figure 4.2: Snapshot of bcc tungsten at 300 K. The eight nearest neighbours (white) of the central atom (blue) do not contain all neighbouring atoms required to calculate the centro-symmetry parameter CSP^{ref} . The 9th-nearest neighbour (red) is missing. Thus, one needs to evaluate more than the 8 nearest neighbours for the CSP^{imp} -calculation.

function for $\text{CSP}_{\text{avg},i} \in [\text{CSP}_{\text{lo}}, \text{CSP}_{\text{hi}}]$ ensures a smooth transition from the fast interatomic potential to the precise one and vice versa for all atoms.

One can change the argument of $f^{(\text{cut})}$ in Eq. (4.11) to use a different detection mechanism and use the following definitions of the switching function as presented without further changes. The argument must be 0 for atoms to be calculated exactly and 1 for atoms to be calculated quickly.

Since the force on an atom depends on all switching parameters within its force cutoff as discussed in Section 4.2.1, we introduce

$$\lambda_{\min,i}(t) = \min \left(\left\{ 1 - (1 - \lambda_{0,j}(t)) f^{(\text{cut})} \left(\frac{r_{ij}(t) - r_{\lambda,\text{lo}}}{r_{\lambda,\text{hi}} - r_{\lambda,\text{lo}}} \right) : j \in \Omega_{\lambda,i} \right\} \right), \quad (4.13)$$

where $\Omega_{\lambda,i} = \{j : r_{ij} < r_{\lambda,\text{hi}}\}$ is the set of neighbouring atoms. As $\lambda_{0,i} \in [0, 1]$ applies, $\lambda_{\min,i} \in [0, 1]$ follows. For both calculation and understanding of $\lambda_{\min,i}(t)$ it is useful to start with $\lambda_{0,i}(t)$ as initial value for the minimum search. Atoms j that were not detected for a precise calculation have $\lambda_{0,j}(t) = 1$. Their contribution to the minimum search is 1 independent of r_{ij} and thus negligible. Only neighbours j that were detected for a precise calculation and thus have $\lambda_{0,j}(t) < 1$ and $\lambda_{0,i}(t)$ can be the searched minimum. Atoms with $\lambda_{0,i}(t) = 0$ set $\lambda_{\min,j}(t) = 0$ for all neighbouring atoms j with $r_{ij} \leq r_{\lambda,\text{lo}}$ and may set $\lambda_{\min,j} \in (0, 1)$ for neighbouring atoms with $r_{ij} \in (r_{\lambda,\text{lo}}, r_{\lambda,\text{hi}})$. Thus, Eq. (4.13) decreases the switching parameter for neighbours of atoms, which need to be calculated precisely, to ensure a higher fraction of precise force contributions for these atoms, i.e., Eq. (4.13) introduces the in Section 4.1.2 discussed spatial switching zone.

Since the radial function $f^{(\text{cut})}(r_{ij})$ used in Eq. (4.13) also fluctuates due to atomic fluctuations, we use a moving average

$$\lambda_{\text{avg},i}(t) = \frac{1}{N_{\lambda,\text{avg}}} \sum_{n=1}^{N_{\lambda,\text{avg}}} \lambda_{\min,i}(t - n\Delta t) \quad (4.14)$$

of $N_{\lambda,\text{avg}}$ timesteps. To prevent unnecessary fluctuations of the switching function further, we introduce a minimum step size $\Delta\lambda_{\min}$ and neglect changes smaller than this step size in form of

$$\lambda_{\text{avg,msz},i}(t) = \begin{cases} \lambda_{\text{avg},i}(t) & \text{for } |\lambda_{\text{avg},i}(t) - \lambda_{\text{avg,msz},i}(t - \Delta t)| \geq \Delta\lambda_{\min} \\ & \text{or } \lambda_{\text{avg},i}(t) \in \{0, 1\}, \\ \lambda_{\text{avg,msz},i}(t - \Delta t) & \text{otherwise.} \end{cases} \quad (4.15)$$

When an atom is detected for a precise calculation according to $\lambda_{\min,i}(t)$ it takes $N_{\lambda,\text{avg}}$ timesteps until $\lambda_{\text{avg},i}$ is changed completely due to the time average. Therefore, a change of $\lambda_{\text{avg},i}$ in the order of magnitude of $1/N_{\lambda,\text{avg}}$ is expected. Changes of $\lambda_{\text{avg,msz},i}$ smaller than $1/N_{\lambda,\text{avg}}$ can be neglected without disturbing the smooth transition from the fast interatomic potential to the precise one or vice versa in a relevant way. Thus, the use of an appropriately small $\Delta\lambda_{\min}$ prevents only λ changes due to atomic fluctuations. We use $\lambda_{\text{avg,msz},i}$ according to Eq. (4.15) as switching function for energy- and force-calculations as the CSP precisely detects defects and surface atoms and the switching function ensures precise forces on these detected atoms.

4.2.3 Integration of motion

An adaptive-precision interatomic potential is given by Eq. (4.5), where the switching parameter can be given by Eq. (4.15). How to time-integrate the equations of motion of particles described by such a potential is addressed in Section 4.2.3.1. Afterwards, an approach that avoids the unphysical force-contribution of the gradient of the switching parameter during the time integration is presented in Section 4.2.3.2.

4.2.3.1 Velocity-Verlet integration

With a known velocity $\mathbf{v}_i(t)$, position $\mathbf{x}_i(t)$, force $\mathbf{F}_i(t)$ and switching parameter $\lambda_i(t)$ of the last timestep t , one can propagate an atom in time. A timestep of Δt according to the velocity-Verlet

integration[131] updated for the use with an adaptive-precision potential is given as,

$$1. \quad \mathbf{v}_i \left(t + \frac{\Delta t}{2} \right) = \mathbf{v}_i(t) + \frac{\mathbf{F}_i(t)}{m_i} \frac{\Delta t}{2} \quad (4.16a)$$

$$\mathbf{x}_i(t + \Delta t) = \mathbf{x}_i(t) + \mathbf{v}_i \left(t + \frac{\Delta t}{2} \right) \Delta t \quad (4.16b)$$

$$2. \quad \lambda_i(t + \Delta t) \text{ is calculated from } \lambda_i(t) \text{ and } \{\mathbf{x}(t + \Delta t)\} \quad (4.16c)$$

$$3. \quad \mathbf{F}_i(t + \Delta t) = \mathbf{F}_i(\{\mathbf{x}_i(t + \Delta t)\}, \{\lambda_i(t + \Delta t)\}) \quad (4.16d)$$

$$E_i(t + \Delta t) = E_i(\{\mathbf{x}_i(t + \Delta t)\}, \lambda_i(t + \Delta t)) \quad (4.16e)$$

$$4. \quad \mathbf{v}_i(t + \Delta t) = \mathbf{v}_i \left(t + \frac{\Delta t}{2} \right) + \frac{\mathbf{F}_i(t + \Delta t)}{m_i} \frac{\Delta t}{2}, \quad (4.16f)$$

where step 2 in Eq. (4.16) is new and required due to the adaptive-precision potential. The first velocity and position calculations for $\mathbf{v}_i(t + \Delta t/2)$ and $\mathbf{x}_i(t + \Delta t)$ use the given $\lambda_i(t)$. A new lambda $\lambda_i(t + \Delta t)$ has to be calculated in the second step of the integration since the positions of the particles have changed and thus also their switching parameter. Therefore, the afterwards calculated quantities $\mathbf{F}_i(t + \Delta t)$, $E_i(t + \Delta t)$ and $\mathbf{v}_i(t + \Delta t)$ depend on the new calculated lambda $\lambda_i(t + \Delta t)$.

4.2.3.2 Avoiding unphysical force-contributions by $(\nabla_i \lambda_k)$ with a local thermostat

The force calculation according to Eq. (4.16d) requires according to Eq. (4.6) the knowledge of $(\nabla_i \lambda_k)$. The force contribution through this gradient of the switching function should be small as a slowly changing switching function is used. Such a slowly changing switching function prevents uncontrollably large forces caused by a fluctuating switching parameter. However, as discussed in Section 4.2.2, the switching function has generally no physical meaning, but only influences the required precision. Furthermore, the need to change the switching parameter adiabatically slow implies the usage of time averages whose gradient cannot be calculated consistently. As this gradient of a time averaged quantity cannot be calculated, we need to neglect $(\nabla_i \lambda_k)$ in Eq. (4.6). Thereby, this approach is not conservative any more and would violate energy conservation if we did not compensate for the error as described in the following.

The adaptive-precision potential is conservative for a constant set of $\{\lambda_i\}$ as the calculation of $\nabla_i \lambda_k$ is not required in this case. Thus, a second system, whose atoms have compared to the real system identical atomic positions and identical but constant switching parameters, can be imagined at any timestep and is referred to as conservative reference system. This conservative reference system offers the possibility of neglecting the unphysical force contribution of $(\nabla_i \lambda_k)$ in an energy and momentum-conserving way. We want to stress that if λ is kept constant per atom over time we have a conservative dynamics and therefore conserve energy and momentum naturally. Thus, we can use the system with constant switching parameters as reference system for each atom to apply a momentum-conserving correction for energy conservation which is described in the following. Note that the presented correction approach is independent of the calculation of λ and can be used for any switching function.

The energy difference ΔH_i of atom i at the end of a time step between the conservative reference system and the system with updated switching parameters is caused by the change of the switching parameter. To calculate this energy error ΔH_i , one has to finish the calculation of an integration step for both sets of switching parameters. As the forces of the reference system and the dynamic- λ system depend on the same atomic positions, one can easily calculate both forces during the force calculation by just summing up two forces weighted with the corresponding switching parameter. This energy difference affects both the kinetic energy change ΔT_i and the potential energy change ΔE_i , namely

$$\Delta H_i = \Delta T_i + \Delta E_i. \quad (4.17)$$

The potential energy change ΔE_i affects only atoms whose switching parameter has changed (cf. Eq. (4.5)). The kinetic energy change ΔT_i affects atoms, where the switching parameter has changed in the force cutoff (cf. Eq. (4.6)). ΔH_i is the energy difference of atom i at the end of a time step between the conservative reference system with constant switching parameters and the system with updated switching parameters. Therefore, one has to finish the calculation of an integration step for both sets of switching parameters. The quantities of the conservative reference system calculated with the constant switching parameter $\lambda_i^c(t + \Delta t) = \lambda_i(t)$ are denoted with a 'c' written as superscript. In contrast, the quantities calculated with the dynamically updated switching parameter $\lambda_i^d(t + \Delta t)$ are denoted with a 'd' written as superscript. The force on an

atom i , which is calculated using the updated switching parameter under neglect of $\nabla_i \lambda_k$ in Eq. (4.6), does not conserve the energy and is given by

$$\mathbf{F}_i^d = \sum_k \left(-\lambda_k^d (\nabla_i E_k^{(f)}) - (1 - \lambda_k^d) (\nabla_i E_k^{(p)}) \right). \quad (4.18)$$

With the same force equation like Eq. (4.18) but λ_k^c instead of λ_k^d , we get the conservative reference force $\mathbf{F}_i^c$. Since the form of the equation is the same and both forces depend on the same atomic positions $\mathbf{x}_i(t + \Delta t)$, one can easily calculate both $\mathbf{F}_i^d$ and $\mathbf{F}_i^c$ during the force calculation by just summing up two forces, where only the only difference is the used switching parameter. Therefore, calculating two forces instead of one increases the force calculation time only slightly, rather than doubling it.

To measure the violation of energy conservation (cf. Eq. (4.17) by neglecting $\nabla_i \lambda_k$ and using $\mathbf{F}_i^d$ according to Eq. (4.18), we calculate both the potential energy difference

$$\Delta E_i(t + \Delta t) = E_i^c(t + \Delta t) - E_i^d(t + \Delta t) \quad (4.19)$$

and the kinetic energy difference

$$\Delta T_i(t + \Delta t) = (|\mathbf{v}_i^c(t + \Delta t)|^2 - |\mathbf{v}_i^d(t + \Delta t)|^2) \frac{m_i}{2} \quad (4.20)$$

at the end of the timestep. The potential energy difference is given as

$$\Delta E_i(t + \Delta t) = (\lambda_i^c(t + \Delta t) - \lambda_i^d(t + \Delta t)) \left(E_i^{(f)}(t + \Delta t) - E_i^{(p)}(t + \Delta t) \right). \quad (4.21)$$

Note that Eq. (4.21) only uses already calculated potential energies and does not introduce further overhead. The switching parameter is known for all atoms and the computation of Eq. (4.21) does not require the knowledge of potential energies of atoms i with $\lambda_i^c(t + \Delta t) = \lambda_i^d(t + \Delta t)$. The kinetic energy difference is given as

$$\begin{aligned} \Delta T_i(t + \Delta t) = \sum_{\alpha=1}^3 \left(\frac{(\Delta t)^2}{8m_i} \left((F_{i\alpha}^c(t + \Delta t))^2 - (F_{i\alpha}^d(t + \Delta t))^2 \right) \right. \\ \left. + \left(F_{i\alpha}^c(t + \Delta t) - F_{i\alpha}^d(t + \Delta t) \right) v_{i\alpha} \left(t + \frac{\Delta t}{2} \right) \frac{\Delta t}{2} \right), \end{aligned} \quad (4.22)$$

where α denotes the spatial component of a vector. It is important to use only the forces of the interatomic potential as $\mathbf{F}_i^c$ and $\mathbf{F}_i^d$ and to neglect any present external forces since such additional forces may not be energy- and momentum-conserving.

We want to add or remove energy to compensate for the work ΔH_i performed on the system when the switching parameters $\{\lambda_i\}$ are changed. Since one cannot simply change the potential energy in a predictable way, we change the kinetic energy of particles. The energy errors are calculated per particle i and should also be corrected locally. Therefore, we need a local thermostat which changes the kinetic energy of atoms by the measured energy error ΔH_i . The local thermostat rescales the momenta of a group of atoms Ω_i , which are part of the neighbour list of atom i , relative to the centre-of-mass velocity by the factor $\beta(\Omega_i, \Delta H_i)$. The rescaling conserves the momentum as the rescaling is relative to the centre-of-mass velocity.

Coupling particles to a local heat bath through collisions of two particles according to Lowe-Andersen[160], for example, is therefore impractical since the direction of the momentum change of the two particles is in the direction of the relative distance of both particles. This predefined direction of a momentum change for two particles may result in very high and unphysical forces when the relative momentum of the two considered particles is not parallel to the direction of the momentum change. Thus, one needs to apply a correction to the set Ω_i of atoms rather than to a pair of atoms to distribute the additional force over multiple atoms. The additional force, that is required to change the kinetic energy of a group of atoms by ΔH_i , is minimal when the momenta of all particles are rescaled since the direction of force and momentum change is equal in this case. However, such a rescaling would violate momentum conservation. Thus, we need to rescale with respect to the centre-of-mass velocity

$$\mathbf{v}_{\text{cm}}(\Omega_i) = \left(\sum_{j \in \Omega_i} m_j \mathbf{v}_j \right) / \left(\sum_{j \in \Omega_i} m_j \right) \quad (4.23)$$

of the rescaled atoms. We want to rescale only the relative momentum

$$\mathbf{p}_{\text{rel},j}(\Omega_i) = \mathbf{p}_j(\Omega_i) - m_j \mathbf{v}_{\text{cm}}(\Omega_i) \quad (4.24)$$

of all particles $j \in \Omega_i$ by the factor $\beta(\Omega_i)$, namely

$$\mathbf{p}_{j,\text{resc}}(\Omega_i) = m_j \mathbf{v}_{\text{cm}}(\Omega_i) + \beta(\Omega_i) \mathbf{p}_{\text{rel},j}(\Omega_i). \quad (4.25)$$

The total relative momentum $\sum_{j \in \Omega_i} \mathbf{p}_{\text{rel},j} = \mathbf{0}$ vanishes and the rescaling according to Eq. (4.25) therefore conserves the momentum. The kinetic energy change in Ω_i due to the rescaling is

$$\Delta T_{\text{resc}}(\Omega_i) = (\beta(\Omega_i)^2 - 1) \sum_{j \in \Omega_i} \frac{1}{2m_j} p_{\text{rel},j}(\Omega_i)^2. \quad (4.26)$$

To correct the detected energy error ΔH_i according to Eq. (4.17), we need to select $\beta(\Omega_i)$ to satisfy $\Delta H_i = \Delta T_{\text{resc}}(\Omega_i)$, i.e.,

$$\Delta H_i = (\beta(\Omega_i)^2 - 1) \sum_{j \in \Omega_i} \frac{1}{2m_j} p_{\text{rel},j}(\Omega_i)^2. \quad (4.27)$$

As Eq. (4.27) is a quadratic equation in β , there are two solutions, from which we select

$$\beta(\Omega_i) = \sqrt{\frac{\Delta H_i}{\sum_{j \in \Omega_i} \frac{1}{2m_j} p_{\text{rel},j}(\Omega_i)^2} + 1} \quad (4.28)$$

as $\beta(\Omega_i) = -\sqrt{\dots}$ would alter the direction of the momenta for $\Delta H_i = 0$. Velocity rescaling with β according to Eq. (4.28) conserves the momentum for $\Delta H_i = 0$.

It is important to note that the rescaling factor β depends on the momentum before the rescaling. Therefore, our method presents challenges in terms of parallelisation: atoms near a domain boundary are in neighbour lists of particles, that are administered by different processors. The rescaled velocities of a set of atoms near a domain boundary need to be known also on the neighbouring domains in order to rescale velocities on the neighbouring domain with the correct velocities of particles, i.e., one needs to communicate velocities after every rescaling. To prevent this expensive communication, we only rescale locally administered particles but no ghost particles: we use a random selection of locally administered particles of the neighbour list of particle i and the particle i itself as set Ω_i . Thereby, the energy correction by rescaling relative momenta can be applied per processor without communication for all particles with an energy error $\Delta H_i \neq 0$.

The maximum possible decrease of the relative momentum per particle is to decrease the relative momentum for all particles $j \in \Omega_i$ to 0 kgm/s with $\beta(\Omega_i) = 0$. Thus, according to Eq. (4.28) energy errors $\Delta H_i < -\sum_{j \in \Omega_i} p_{\text{rel},j}(\Omega_i)^2 / (2m_j)$ cannot be corrected as they correspond to a negative radicand in Eq. (4.28) and imply $\beta \notin \mathbb{R}$. Hence, one needs to include more particles in the rescaling in this case. The occurrence of negative radicands depends on the simulation setup and the combined potentials. We did not observe negative radicands at room temperature as will be discussed in Section 4.3.2.3. Simulations at a temperature of 0 K could be more challenging as there is no kinetic energy in the system in this case. However, there are no thermal fluctuations of atoms at this temperature and the atoms have known distances to their neighbours. Differences from this reference configuration, that is simulated with a fast potential, can be far more easily detected than at room temperature. Therefore, the fast potential can be far more precise for small deviations of the known 0 K-configuration and ΔE_i (cf. Eq. (4.21)) should be negligible. Thus, the amount of energy, that needs to be changed through rescaling at 0 K is far smaller compared to a simulation at room temperature. Hence, one can expect that simulations near 0 K are possible without the occurrence of negative radicands in Eq. (4.28).

Concrete prevention strategies and workarounds in LAMMPS for negative radicands are discussed in the following. Energy errors $\Delta H_i < -\sum_{j \in \Omega_i} p_{\text{rel},j}(\Omega_i)^2 / (2m_j)$ cannot be corrected by the local thermostat as they correspond to a negative radicand in Eq. (4.28) and imply $\beta \notin \mathbb{R}$. Hence, one needs a larger neighbour list to include more atoms in the set Ω_i in case of a negative radicand. As a dynamic neighbour-list cutoff is not implemented in LAMMPS, one needs to use the last restart file to run the simulation with a larger neighbour-list cutoff. Requesting a larger neighbour-list cutoff as precautionary measure is possible, but would reduce the performance of the whole simulation rather than just of a small subset of timesteps. As we use a parallel-computing approach including a load-balancing scheme problems due to the domain geometry are possible. When the load balancer reduces the size of a domain, it reduces thereby also the number of local

particles which are available for rescaling. Thus, in case of a negative radicand one could also restart the simulation with temporary disabled load balancing to avoid the existence of small domains. For details regarding the load balancing see Section 4.2.4.

In summary we note that the presented rescaling approach fulfils momentum conservation as only relative momenta are rescaled. Furthermore, the rescaling approach results in energy conservation for the forces according to Eq. (4.18). One can then use $\mathbf{F}_i^d(t + \Delta t)$ (cf. Eq. (4.18)) as $\mathbf{F}_i(t + \Delta t)$ (cf. Eq. (4.16)) and the rescaled velocities as $\mathbf{v}_i(t + \Delta t)$ at the begin of the next integration step. Note that the application of this local thermostat contains essentially two stochastic components. Firstly, the selection of neighbouring atoms that are rescaled is stochastic, namely Ω_i . Secondly, the direction of the effective force is the direction of the relative momentum $\mathbf{p}_{\text{rel},j}(\Omega_i)$ (cf. Eq. (4.25)) and thus stochastic.

This local thermostat is a tool to correct locally a known energy gain or loss of an atom i during the integration step by adjusting the kinetic energy in the local environment of this atom. We exactly know the energy error due to the neglect of $\nabla_i \lambda_k$ in Eq. (4.6) and correct this error to conserve the energy within numerical precision. The integrator changes the velocity of particles of mass m according to

$$\Delta v = \Delta t \frac{F}{m} \quad (4.29)$$

due to a force F . One can understand the energy-error correction as additional force which is just used at the end of a timestep, but not in the next integrator step. The application of this local thermostat contains a stochastic component as direction of the effectively applied force is in direction of the relative momentum and thus stochastic. Note that this local thermostat is not considered to provide a canonical ensemble. It can be considered as an energy correction, but not primarily as temperature correction, i.e., the objective function is not the temperature, but the energy. Thus, non-equilibrium simulations, non-energy conserving simulations like an canonical ensemble and simulations with external forces can be performed with the local thermostat.

4.2.4 Load balancing

Time integrating the equations of motion for a set of atoms described by a short-range interatomic potential requires per atom only information from neighbouring atoms within a cutoff distance. Thus, LAMMPS uses distributed-memory parallelism via MPI[1]. The simulation box is divided into disjoint domains which cover the whole simulation box according to a spatial domain-decomposition approach[161]. Each MPI task, in the following denoted as processor, is assigned to one domain and administers all particles located in this domain. Our adaptive-precision potential requires a highly different computational time per atom dependent on the used potential which causes load-imbalances between the domains. Hence, we present a load-balancing approach to dynamically change the domain sizes during a simulation.

We calculate the centro-symmetry parameter $\text{CSP}_i(t)$ according to Eq. (4.9) and the switching parameter λ according to Eq. (4.13) within the force-calculation routine to include both into the load balancing of the force computation. Thus, we perform four independent calculations during the force-calculation routine, whose total duration needs to be balanced: the calculation of 1. the fast potential (FP), 2. the precise potential (PP), 3. CSP_i (CSP) and 4. $\lambda_{\min,i}$ from $\lambda_{0,j}$ (λ). Since all four calculations are required only for a subset of particles and the precise potential can be 100 times slower¹ than the precise potential, the code inherently needs load balancing.

For all four force calculations to be executed independently one after the other on all processors, it is important that none of the calculations contains communication with other processors as communication is a synchronisation point within a force calculation and may include high additional waiting times. Thus, avoiding communication during the force calculation is essential to allow effective load balancing. The EAM calculation in LAMMPS includes communication of the derivative of the embedding function and the electron density within the force calculation. In order to avoid this communication during the force calculation, we compute these two quantities on all processors which require them. This results in a double calculations of these two quantities for atoms near domain boundaries, but this cost is acceptable in our case to achieve proper load balancing. The ACE calculation did not require any adjustments due to communication prevention within the force calculation, since there is no communication within the force computation function of the ACE implementation of LAMMPS.

We assume per processor p and calculation $\mathcal{X} \in \{\text{FP}, \text{PP}, \text{CSP}, \lambda\}$ an average work per particle

$$\rho_p^{(\mathcal{X})} = \tau_p^{(\mathcal{X})} / N_p^{(\mathcal{X})}, \quad (4.30)$$

¹Time measurements for all four calculations are shown in Fig. 4.9.

where the total duration $\tau_p^{(\mathcal{X})}$ and the number of calculations $N_p^{(\mathcal{X})}$ is measured within the force calculation of $\mathcal{X}$. We rescale the work $\rho_p^{(\mathcal{X})}$ with a constant factor per processor p in order to match the total work of our model with the also measured total calculation time τ_p of the processor. Thereby, one can assign an individual load ρ_i^{atom} to atom i with possible contributions of $\rho_{p(i)}^{\text{FP}}$, $\rho_{p(i)}^{\text{PP}}$, $\rho_{p(i)}^{\text{CSP}}$ and $\rho_{p(i)}^{\lambda}$. The load ρ_i^{atom} is used as input for a load-balancing algorithm². We use a staggered grid[163] which is used for particle simulations[164] and which is implemented in the load-balancing library ALL[162].

The processor p with the highest work restricts the speed of a simulation since faster processors need to wait due to communication and synchronisation. Thus, the imbalance, as defined in LAMMPS[156, p. 1317],

$$I = \frac{\max_p(\tau_p)}{\langle \tau_p \rangle_p} \quad (4.31)$$

of the force calculation time τ_p gives a measure for the quality of the load balancing, where an imbalance of 1 corresponds to a perfectly balanced system. Thus, load balancing is only used when the imbalance is worse than a user-defined threshold I^{thr} .

Note, that the measured force-calculation times can be used to estimate on-the-fly the calculation time $\tau_{\text{system}}^{\text{PP}}$ that it would take to calculate the entire system with the precise potential. As a system, which is completely calculated with the precise potential, can be balanced by a load balancer, we neglect load imbalances for the estimation of the calculation time $\tau_{\text{system}}^{\text{PP}}$. Therefore, $\tau_{\text{system}}^{\text{PP}}$ is given as

$$\tau_{\text{system}}^{\text{PP}} = \langle \rho_p^{\text{PP}} N_p^{\text{aa}} \rangle_p \quad (4.32)$$

where N_p^{aa} denotes the number of atoms administered by processor p . The estimated speedup S^{est} of the adaptive-precision potential is then

$$S^{\text{est}} = \frac{\tau_{\text{system}}^{\text{PP}}}{\max_p(\tau_p)}. \quad (4.33)$$

For the adaptive-precision to be beneficial, $S^{\text{est}} > 1$ must apply. Therefore, the on-the-fly speedup estimation can be used to detect on-the-fly whether the calculation time, which is saved by simplified calculations, outweighs the overhead of calculating detection mechanism and switching parameter, i.e., whether $S^{\text{est}} > 1$ applies. For $S^{\text{est}} \leq 1$ one can then switch off the adaptive-precision potential and use the precise potential instead.

4.2.5 Implementation

Algorithm 4 shows how a timestep works for an energy conserving MD simulation using the adaptive-precision framework that was presented in Section 4.2. The velocity-Verlet integration (cf. Ref. [131]) gives the basic structure and was already implemented in LAMMPS[1]. All required adjustments due to our adaptive-precision method are inserted into this basic structure. The computation of $\lambda_{\min,i}$ according to Eq. (4.13) can be expensive, as it depends – like a potential energy itself – on the atomic distances r_{ij} to neighbouring atoms j . Thus, this computation is done within the force-calculation routine, for which the computational time can be measured and balanced due to the absence of communication barriers. For the same reason, the computation of the centro-symmetry parameter (CSP) is done within the force-calculation routine. The remaining compute steps are inserted based on their dependencies into the algorithm.

4.3 Adaptive-precision EAM-ACE copper potential

In this section, an ACE potential and an EAM potential are combined to an adaptive-precision copper potential according to Eq. (4.5) with the switching function λ , which is described in Section 4.2.2. The atomic cluster expansion potential (cf. Section 3.1) of Ref. [4] is used as precise potential as it is a modern ML-potential with a good representation of its DFT reference data. We use the EAM2 potential of Ref. [145], which is described in Section B.1.1, as starting point for the fast potential since it is widely used[149–152]. First, the EAM potential is optimised in Section 4.3.1. Then, the parameters of the coupled optimised EAM- and the ACE-potential are selected and discussed in Section 4.3.2.

²The load-balancing library ALL[162] supports a histogram-based approach and a local approach to balance a staggered grid. The local approach only uses communication with neighbouring processors and prevents global communication and only requires the sum of ρ_i of all particles i on a processor as input. However, we use the histogram-based approach as this approach resulted in a better balanced system for the problems under study. Using the local balancing approach might be beneficial for other systems under study.

Algorithm 4 MD simulation with the developed adaptive-precision potential. The **violet** steps are required for all MD simulations. The **red** steps are the executed one after the other without communication in between. The computational time τ_p is measured per processor p for these **red** steps and balanced with the load-balancer.

```

for all timesteps  $t$  do
  update  $\mathbf{v}_i$  according to Eq. (4.16a)
  update  $\mathbf{x}_i$  according to Eq. (4.16b)
  update history of  $\lambda_{\min,i}$ 
  compute  $\lambda_{\text{avg},i}$  according to Eq. (4.14)
  compute  $\lambda_{\text{avg,msz},i}$  according to Eq. (4.15)
  compute  $\lambda_{0,i}$  according to Eq. (4.11)
  compute imbalance  $I$  according to Eq. (4.31)
  if balancing timestep and  $I > I^{\text{thr}}$  then
    apply load balancing as described in Section 4.2.4
  end if
  exchange atom information with neighbouring processors
  compute fast potential for the corresponding atoms
  compute precise potential for the corresponding atoms
  compute CSP for the corresponding atoms according to Eq. (4.10)
  update the history of CSP $i$ 
  compute  $\lambda_{\min,i}$  for the corresponding atoms according to Eq. (4.13)
  exchange force information with neighbouring processors
  compute energy error  $\Delta H_i$  according to Eq. (4.17)
  update  $\mathbf{v}_i$  according to Eq. (4.16f)
  apply local thermostat to correct  $\Delta H_i$  as described in Section 4.2.3.2
end for

```

4.3.1 Optimising the EAM potential

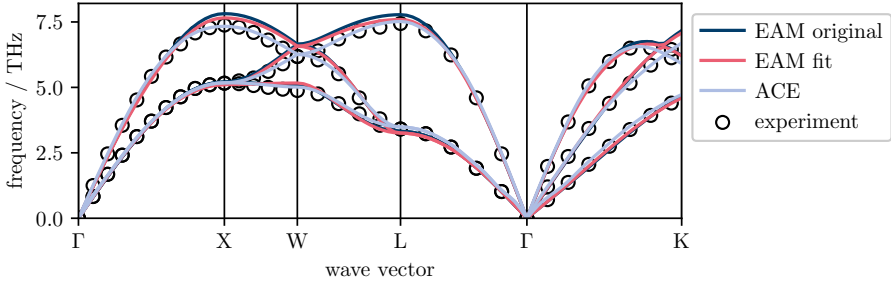
We need to optimise the EAM potential to minimise systematic energy differences between the used EAM and ACE potentials. Furthermore, the optimisation gives the possibility to improve the forces as well. The EAM potential is fitted for applications with surfaces, vacancies and interstitials, which are detected for a precise calculation, and, therefore, do not need to be described by the fast and less accurate EAM potential. There might be a systematic energy offset between EAM and ACE energies, as both potentials were fitted to different reference data. Such an energy offset can be corrected by adding a constant energy $\xi_0^{\text{Cu,Fit}}$ to the energy of the EAM potential. This energy offset correction does not affect the EAM forces as the derivative of an added constant is zero. Specifically, we introduce the constant energy $\xi_0^{\text{Cu,Fit}}$ to the original embedding function $\xi^{\text{Cu}}(x)$ (Eq. (B.4)), namely $\xi^{\text{Cu,Fit}}(x) = \xi^{\text{Cu}}(x) + \xi_0^{\text{Cu,Fit}}$ and use $\xi^{\text{Cu,Fit}}(x)$ instead of $\xi^{\text{Cu}}(x)$. The set of parameters $\mathcal{A}^{\text{Cu}}$ (cf. Section B.1.1) is optimised by minimising a loss function $\mathcal{L}^{\text{crex}}$ with **atomicrex**[165]. The loss function includes atomic energies E_{si} and forces F_{si} of different structures s as well as the scalar properties A_o . We use the lattice parameter a_0^{fcc} , the cohesive energy and elastic constants as scalar properties. The target values denoted with 'targ' are the corresponding values calculated with the highly accurate ACE potential. The predicted values denoted with 'pred' are calculated with the current set of parameters $\mathcal{A}$. The differences between predicted and target values are per quantity weighted with a tolerance δ^{tol} . The used target values and tolerances of

Table 4.2: Target values with tolerance used in the loss function Eq. (4.34) for the optimisation of the EAM potential.

bulk property	target value	tolerance δ^{tol}
lattice parameter a_0^{fcc}	3.6309 Å	0.001 Å
cohesive energy E_{coh}	-3.6995 eV/atom	0.01 eV/atom
bulk modulus B	138.2 GPa	1 GPa
elastic constant C_{11}	173.8 GPa	1 GPa
elastic constant C_{12}	120.5 GPa	1 GPa
elastic constant C_{44}	77.8 GPa	1 GPa
force F_{md}	MD simulation	0.01 eV Å ⁻¹
atomic energy E_{md}	MD simulation	0.01 eV

Table 4.3: Comparison of quantities calculated with EAM, optimised EAM and ACE with DFT and experimental reference values.

	ACE	EAM original	EAM fit	DFT	Exp.
lattice parameter a_0^{fcc} / Å	3.6309	3.6149	3.6305		
cohesive energy / eV	-3.6995	-3.5579	-3.6957		
vacancy formation energy / eV	1.1285	1.2735	1.0589	1.07[4]	1.27 [138]
surface energy (111) / Jm ⁻²	1.3566	1.2443	0.8802	1.36[4]	
surface energy (100) / Jm ⁻²	1.5111	1.3515	0.9765	1.51[4]	
surface energy (110) / Jm ⁻²	1.5889	1.4822	1.1225	1.57[4]	
interstitial formation energy / eV					
100-dumbbell	3.1025	3.0809	2.9662	3.10[139]	2.8-4.2[140]
octahedral	3.3392	3.2559	3.1303	3.35[139]	
tetrahedral	3.6217	3.5655	3.4240	3.64[139]	
elastic constant C_{11} / GPa	173.8	171.5	171.3	177[4]	177 [141]
elastic constant C_{12} / GPa	120.5	124.2	120.1	132[4]	125 [141]
elastic constant C_{44} / GPa	77.8	76.2	80.6	82[4]	81 [141]

**Figure 4.3:** Phonon spectra calculated with ASE[136] of the fitted EAM potential, original EAM potential, the ACE potential and experimental values[137] measured by neutron diffraction at the temperature 80 K.

the loss function are listed in Table 4.2. The loss function to be minimised is

$$\mathcal{L}^{\text{crex}} = \sum_{s \in \mathcal{S}} \left\langle \left(\frac{E_{si}^{\text{targ}} - E_{si}^{\text{pred}}}{\delta_{E_{\text{md}}}^{\text{tol}}} \right)^2 + \left(\frac{\|\mathbf{F}_{si}^{\text{targ}} - \mathbf{F}_{si}^{\text{pred}}\|}{\delta_{F_{\text{md}}}^{\text{tol}}} \right)^2 \right\rangle_i + \sum_{o \in \mathcal{O}} \left(\frac{A_o^{\text{targ}} - A_o^{\text{pred}}}{\delta_o^{\text{tol}}} \right)^2, \quad (4.34)$$

with the set of scalar observables $\mathcal{O}$ and the set of structures $\mathcal{S}$. We use two snapshots of a NVE simulation of copper with $10 \times 10 \times 10$ unit cells at 300 K and periodic boundary conditions as structures $\mathcal{S}^{\text{EAM,Cu}}$ for the fit. These structures are sufficient since we start with an already tested and validated EAM potential and the optimised EAM potential is for use only with atomic configurations similar to the used structures. We start the fitting with the parameters of EAM2 from Ref. [145] and $\xi_0^{\text{Cu,Fit}} = 0$ eV. In a first run, only the energy offset $\xi_0^{\text{Cu,Fit}}$ is fitted. Afterwards all parameters including $\xi_0^{\text{Cu,Fit}}$ are fitted. The optimised parameters are listed in Section B.1.1 in Table B.1. Scalar properties calculated with the optimised EAM potential are shown in Table 4.3. Lattice constant and cohesive energy of the optimised EAM potential match through the optimisation. The elastic constants of ACE are in very good agreement with both EAM and optimised EAM potential. The phonon spectra are compared in Fig. 4.3 and are in good agreement.

4.3.2 Parameter selection

The parameter selection of the combined EAM and ACE potential is discussed in this section. The parameters of the hybrid potential are listed in Table B.2. We refer to the adaptive-precision potential with this parameter set as Hyb1. These parameters are used in 1. the mechanism used

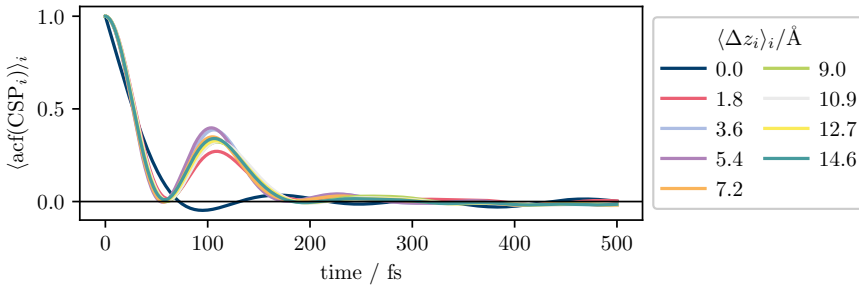


Figure 4.4: Mean autocorrelation functions of the centro-symmetry parameter CSP_i according to Eq. (4.9). Each shown graph is the mean of the autocorrelation functions (acf) of one atom layer in z -direction. The mean distance $\langle \Delta z_i \rangle_i$ of the corresponding atom layer to the (100)-copper surface is given in the legend. The autocorrelation functions are calculated from data of a 5 ps simulation with an adaptive-precision potential.

to detect atoms that require a precise calculation, 2. in the switching function and 3. in the local thermostat that is used to avoid unphysical force contributions by $(\nabla_i \lambda_k)$. The parameters regarding the centro-symmetry parameter are covered in Section 4.3.2.1, the ones of the switching function in Section 4.3.2.2 and the parameters of the local thermostat are discussed in Section 4.3.2.3.

4.3.2.1 Centro-symmetry parameter

N_{buffer} is used to ensure all relevant neighbouring atoms are used in the calculation of the centro-symmetry parameter according to Eq. (4.9). For copper, we have not observed the case of unexpected high centro-symmetry parameters we discussed in Fig. 4.2 for tungsten. Therefore, we use $N_{\text{buffer}} = 0$. An unexpected high CSP is less likely for copper as the distance in lattice constants between the first and second neighbour shell is for bcc with $1 - \sqrt{3}/4 \approx 0.13$ smaller than for fcc with $1 - \sqrt{1}/2 \approx 0.29$.

The centro-symmetry parameter of an atom i fluctuates due to atomic fluctuations. To suppress this fluctuations, we use a time average of $N_{\text{CSP,avg}}$ timesteps in Eq. (4.10). To determine $N_{\text{CSP,avg}}$, we measured autocorrelation functions (acf) for 5 ps near a (100)-copper surface as shown in Fig. 4.4 as mean per (100) atom layer dependent on the distance to the surface. The acf of the CSP has a peak at about 110 fs for all layers apart from the surface layer. As the CSP depends on the local environment, a different acf for the surface atoms is expected. Since the time averaging of the centro-symmetry parameter is used to average out thermal fluctuations, we want to average the CSP up to the peak of the acf of non-surface atoms at 110 fs. With the used timestep $\Delta t = 1$ fs follows $N_{\text{CSP,avg}} = 110$.

The distribution of the centro-symmetry parameter CSP_i in the training data is used to set the thresholds CSP_{lo} and CSP_{hi} for Eq. (4.11). The fast potential is optimised for usage at a target temperature with MD data of the precise and expensive potential. The training data contain only thermal fluctuations of atoms with observed $\text{CSP}_i \in [0 \text{ \AA}^2, 3 \text{ \AA}^2]$. Therefore, we distinguish between a small CSP of the thermal fluctuations and all other atoms with a higher CSP. As atoms with $\text{CSP}_{\text{avg},i} \geq \text{CSP}_{\text{hi}}$ are detected for a precise calculation according to Eq. (4.11), we select $\text{CSP}_{\text{hi}} = 3 \text{ \AA}^2$. Thereby, we do not detect atoms with expected thermal fluctuations but all other atoms. We use $\text{CSP}_{\text{lo}} = 2.5 \text{ \AA}^2$ since the difference of $0.5 \text{ \AA}^2$ between the thresholds allows a smooth transition for atoms from the fast to the precise potential. One might use a slightly different value CSP_{lo} , but far lower values limit the possible performance gain since thermally fluctuating atoms are treated partially with the precise potential. It is essential to treat the majority of the thermally fluctuating atoms completely with EAM as one can only save computational time when ACE is not evaluated for most atoms at all.

4.3.2.2 Switching function

The cutoff radii $r_{\lambda,\text{lo}}$ and $r_{\lambda,\text{hi}}$ are used in the calculation of $\lambda_{\text{min},i}$ according to Eq. (4.13) to decrease the switching parameters for neighbouring atoms of atoms, which require a precise calculation.

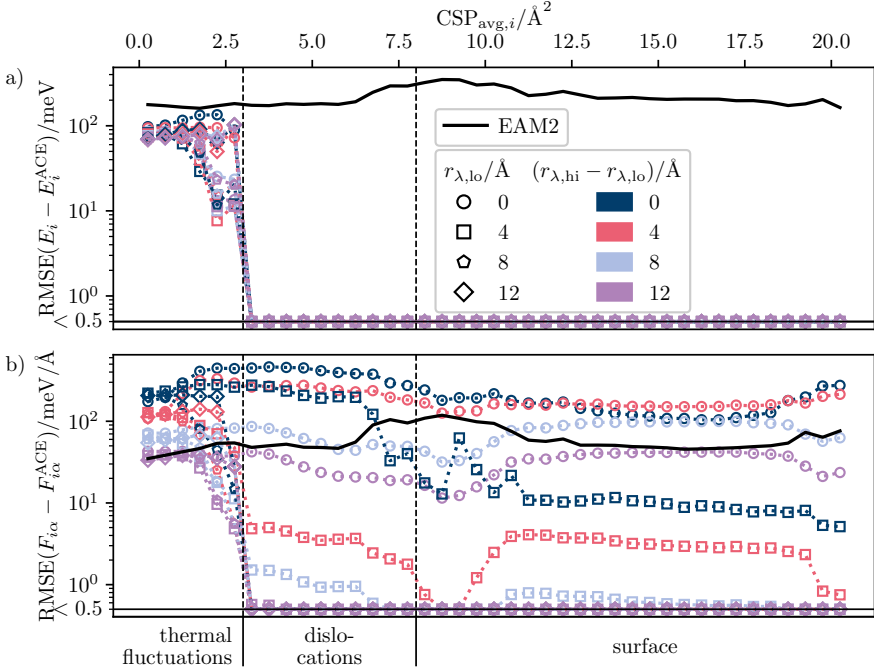


Figure 4.5: Root mean square error (RMSE) of the adaptive-precision potential compared with the precise ACE potential for a snapshot of a nanoindentation dependent on $r_{\lambda,\text{lo}}$ and $r_{\lambda,\text{hi}}$. All atoms whose forces are influenced by both EAM and ACE are grouped dependent on $\text{CSP}_{\text{avg},i}$ and the RMSE is calculated per group. Atoms with pure EAM or ACE force-contributions are not considered since their energies and forces are good. The RMSE of ACE compared to EAM2 of [145] is shown as reference. The cutoff $r_{\lambda,\text{lo}}$ is represented by different symbols and the width $r_{\lambda,\text{hi}} - r_{\lambda,\text{lo}}$ of the switching zone is colour-coded.

The force on an atom depends on all switching parameters within the force cutoff. Thus, $r_{\lambda,\text{lo}}$ and $r_{\lambda,\text{hi}}$ increase the force precision for precisely calculated atoms. To determine the cutoff radii $r_{\lambda,\text{lo}}$ and $r_{\lambda,\text{hi}}$, we calculate energies and forces according to Eqs. (4.5) and (4.18) for one timestep with $\lambda_{\text{min},i}$ as switching function under neglect of $(\nabla_i \lambda_k)$ as discussed in Section 4.2.3. However, we do not apply a local thermostat to prevent random influence in this parameter study and calculate only one timestep. We use a snapshot of a nanoindentation including dislocations. The energy error compared to the precise ACE energy as shown in Fig. 4.5a vanishes by design for precisely calculated atoms independently of the varied parameters since it depends only on $\lambda_{\text{min},i}$ of the corresponding precisely calculated atom i . The force error compared to the precise ACE forces is shown in Fig. 4.5b and depends for the case of thermal fluctuations mainly on the width $r_{\lambda,\text{hi}} - r_{\lambda,\text{lo}}$ of the switching zone between precise and fast atoms but is independent of $r_{\lambda,\text{lo}}$. The force error for thermal fluctuations can be larger than for the EAM reference force since only the total forces rather than the force contributions of EAM and ACE are fitted as shown in Fig. 4.1. Therefore, a switching zone is needed to change smoothly between EAM and ACE atoms. The larger the switching zone, the smaller the force error on thermal fluctuations, but the more computationally expensive the calculation becomes since more precise calculations are required. We selected a switching zone width of $r_{\lambda,\text{hi}} - r_{\lambda,\text{lo}} = 8 \text{ \AA}$. The force error for precisely calculated atoms depends primarily on $r_{\lambda,\text{lo}}$. We selected $r_{\lambda,\text{lo}} = 4 \text{ \AA}$. With these selected values, $f^{\text{(cut)}} \left(\frac{r_{\text{cut}}^{\text{EAM,Cu}} - r_{\lambda,\text{lo}}}{r_{\lambda,\text{hi}} - r_{\lambda,\text{lo}}} \right) \approx 0.98$ follows (cf. Eq. (4.12)), i.e., the spatial transition zone ensures, according to Eq. (4.13), that there are almost no force contributions by the EAM potential for atoms that are detected for a precise calculation (cf. Eq. (4.8)). This corresponds to the conclusion of the analysis of the force contributions $\mathbf{f}_{ik}$ in Section 4.1.2.

The switching parameter $\lambda_{\min,i}$ according to Eq. (4.13) of atom i fluctuates as it depends on the distance r_{ij} to neighbouring atoms j , which require a precise calculation. Thus, we use a time average of $N_{\lambda,\text{avg}}$ timesteps in Eq. (4.14) to average out these fluctuations of the switching parameter. As this is the same motivation like for the time averaging of the centro-symmetry parameter in Eq. (4.10), we use the same number $N_{\lambda,\text{avg}} = N_{\text{CSP,avg}} = 110$ of averaged timesteps.

4.3.2.3 Local thermostat

An energy error ΔH_i according to Eq. (4.17) of atom i is corrected by rescaling momenta of the set of random neighbouring atoms Ω_i according to Eq. (4.28). The distribution of ΔH_i in Fig. 4.6a for a snapshot of a copper nanoindentation at 20 Å indentation depth and at a temperature of 300 K shows that energy corrections need to be applied up to an order of magnitude of 1 meV due to changes of the potential energy. The energy difference between ACE and EAM is in the order of 100 meV according to Fig. 4.5a and the minimum change of the switching parameter is $\Delta\lambda_{\min} = 1/N_{\lambda,\text{avg}} \approx 10^{-2}$. Thus, the energy fluctuations due to the change of λ_i in the order of 1 meV are expected. The kinetic energy T_i of an atom at 300 K is $T_i = (3/2)k_{\text{B}}T \approx 39$ meV, where k_{B} is the Boltzmann constant and T the temperature. The kinetic energy changes only slightly with an appropriate timestep and the influence of a λ update on the forces and therefore on the kinetic energy change is even lower. Therefore, the contribution of the kinetic energy to the energy error is magnitudes smaller than the potential-energy contribution as shown in Fig. 4.6a. Nevertheless, a kinetic energy change due to rescaling in the order of 1 meV is not negligible compared to an average kinetic energy of 39 meV at 300 K. Hence, we need to distribute the effect of rescaling onto a larger number of particles in order to minimise effects on single atoms. In simulations we use a maximum number $|\Omega_i| = 800$ of rescaled neighbours. The velocity change due to rescaling is shown in Fig. 4.6b for the snapshot of the nanoindentation. The absolute velocity change $|v_{i\alpha}^{\text{unsc}} - v_{i\alpha}^{\text{resc}}|$ is smaller than 0.1% $v_{i\alpha}^{\text{unsc}}$ for $v_{i\alpha}^{\text{unsc}} \geq 0.1$ Å/ps and smaller than 0.1 mÅ/ps for about $v_{i\alpha}^{\text{unsc}} \leq 0.1$ Å/ps. Thus, the velocity changes are small but nevertheless may influence the dynamics of the system. To characterise this effect further, we translate the velocity changes into effective forces. Velocities are updated by the integrator according to $\Delta\mathbf{v} = (\mathbf{F}/m)\Delta t$ (cf. Eqs. (4.16a) and (4.16f)). Hence, the additional force contribution is given as

$$\Delta F_{i\alpha}^{\text{lt}} = (v_{i\alpha}^{\text{unsc}} - v_{i\alpha}^{\text{resc}})m_i/\Delta t \quad (4.35)$$

for all particles i and spatial dimensions α . The force $\Delta F_{i\alpha}^{\text{lt}}$ according to Eq. (4.35), however, is a result of the neglect of $\nabla_i\lambda_k$ in Eq. (4.6) as discussed in Section 4.2.3. The velocity updates in Fig. 4.6b correspond to the theoretical forces in Fig. 4.6c. These forces, including rescaling, on precisely calculated atoms are within a tolerance up to 10 meV/Å compared to the precise ACE forces. The potential energy E_i depends, according to Eq. (4.5), only on the switching parameter λ_i of atom i . Therefore, the precision of the potential energies E_i is unaffected by the rescaling.

4.4 Demonstration

We calculated a nanoindentation with 4 million atoms and a (100)-surface at 300 K for 100 ps on JURECA-DC[166] with the adaptive-precision copper potential Hyb1 (cf. Section 4.3 and Table B.2). The achieved precision is analysed in Section 4.4.1 and the saved computation time compared to a full ACE simulation in Section 4.4.2.

4.4.1 Precision

The force difference

$$\Delta F = \text{RMSE}(F_{i\alpha}^{\text{Hyb1}} + \Delta F_{i\alpha}^{\text{lt}} - F_{i\alpha}^{\text{ACE}}) \quad (4.36)$$

of rescaled atoms, including the theoretical rescaling force $F_{i\alpha}^{\text{lt}}$ according to Eq. (4.35), of the adaptive-precision potential Hyb1 compared to ACE forces during the nanoindentation is shown in Fig. 4.7a. The force difference ΔF^{Hyb1} is calculated from force components $F_{i\alpha}$ of all atoms i in all three spatial dimensions α . For defects and surface atoms, the forces are within a tolerance up to 10 meV/Å compared to the precise ACE forces for most cases while the exact potential energy of ACE is used according to Eq. (4.5).

We changed single parameters of the parameter set Hyb1 and performed simulations with otherwise identical parameters to perform a sensitivity analysis on results of the selected parameters. The influence of the changed parameters is shown in Fig. 4.7b. Increasing the thresholds CSP_{lo} and CSP_{hi} by $0.5 \text{ \AA}^2$ decreases the force precision for thermally fluctuating atoms and dislocations

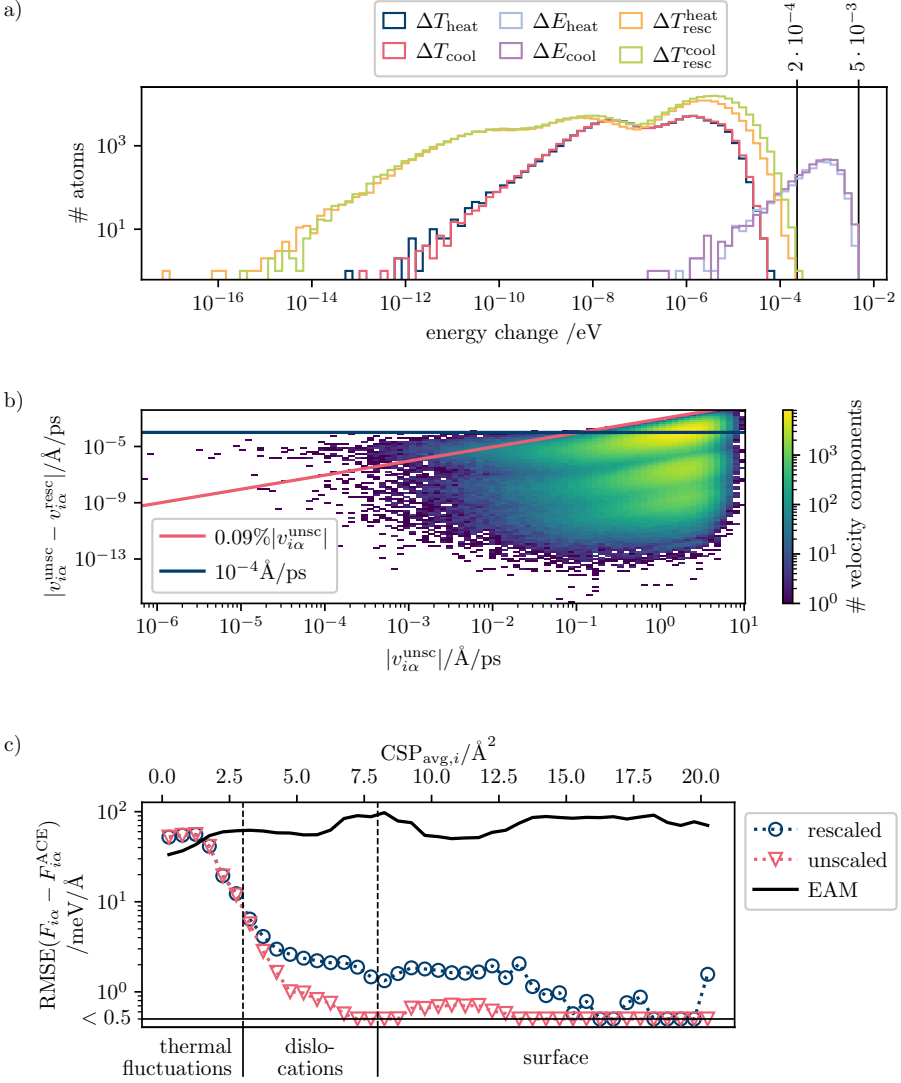


Figure 4.6: Rescaling-related quantities of a nanoindentation at 300 K and 20 Å indentation depth. a) Histogram of potential energy change ΔE_i according to Eq. (4.21), kinetic energy change ΔT_i according to Eq. (4.22) and kinetic energy change ΔT_{resc} due to the applied rescaling according to Eq. (4.25) separated for positive values denoted with 'heat' and negative values denoted with 'cool'. b) 2D-Histogram of the velocity $|v_{i\alpha}^{\text{unsc}}|$ before the rescaling step compared with the velocity change $|v_{i\alpha}^{\text{unsc}} - v_{i\alpha}^{\text{resc}}|$ during the rescaling step, where i denotes a particle and α a spatial dimension. c) Root mean square error (RMSE) of different forces compared with the precise ACE potential. All rescaled atoms are grouped dependent on $\text{CSP}_{\text{avg},i}$ and the RMSE is calculated per group. The applied forces according to Eq. (4.18) are denoted with 'unscaled'. The graph denoted with 'rescaled' corresponds to the applied forces plus theoretical forces according to Eq. (4.35) due to the rescaling. The forces of the optimised EAM potential used for the adaptive-precision potential are denoted with 'EAM' and given as reference.

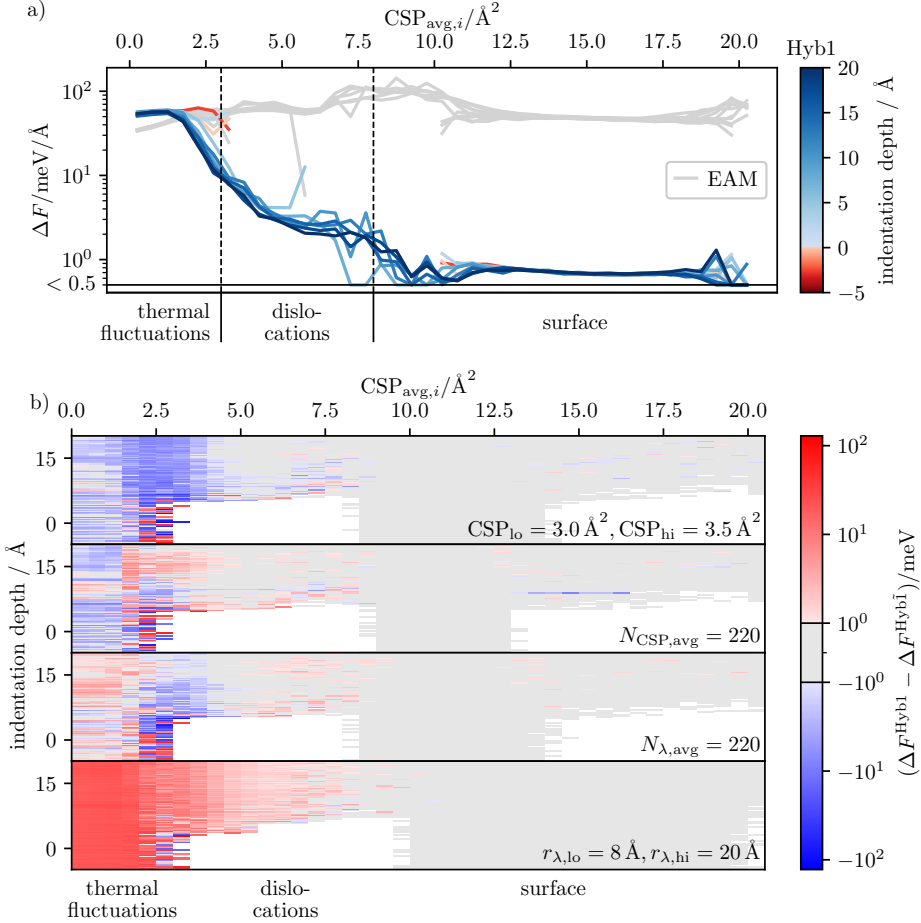


Figure 4.7: a) Force error ΔF^{Hyb1} (cf. Eq. (4.36)) of rescaled atoms dependent on $\text{CSP}_{\text{avg},i}$ and indentation depth of a nanoindentation. b) Force error ΔF^{Hyb1} compared with the force error $\Delta F^{\text{Hyb1}\tilde{}}$ of another simulation with the parameter set $\text{Hyb1}\tilde{}$, where the parameters in the lower right corner of the corresponding plot are different than in Hyb1 . The force error difference $\Delta F^{\text{Hyb1}} - \Delta F^{\text{Hyb1}\tilde{}}$ is colour-coded, where blue and red corresponds to a smaller force error of Hyb1 and $\text{Hyb1}\tilde{}$ respectively. The simulations for a) and the lowest plot of b) are calculated in a cubic system with 100^3 unit cells, 68^3 unit cells are used for the remaining simulations.

with $\text{CSP}_{\text{avg},i} < 4.5 \text{ \AA}^2$. The decreased force precision for the dislocations is expected as these are the atoms which are simulated with the fast potential due to the increased CSP thresholds. Thus, the only advantage of increasing the CSP thresholds is performance. Doubling the averaging time of the centro-symmetry parameter $N_{\text{CSP,avg}}$ decreases the force precision on thermally fluctuating atoms. The force precision of dislocations increases in most snapshots. As dislocations are in contrast to the surface not stationary, increasing the CSP averaging time may reduce the detection possibility for moving dislocations. Thus, one can adjust $N_{\text{CSP,avg}}$ dependent on the expected dislocation dynamics. Doubling the averaging time of the switching parameter $N_{\lambda,\text{avg}}$ decreases the force precision of dislocations with $\text{CSP}_{\text{avg},i} < 4 \text{ \AA}^2$, but increases the force precision of dislocations with a larger time-averaged CSP. The force precision for dislocations decreases with an increasing $\text{CSP}_{\text{avg},i}$ according to Fig. 4.7a and this effect is enhanced by doubling $N_{\lambda,\text{avg}}$. Thus, increasing $N_{\lambda,\text{avg}}$ is not beneficial. Increasing the cutoffs $r_{\lambda,\text{lo}}$ and $r_{\lambda,\text{hi}}$ of $\lambda_{\text{min},i}$ by $4 \text{ \AA}$ and $8 \text{ \AA}$ respectively, increases the number of precise calculations and thus the force precision for all atoms apart from surface atoms which are already treated with the precise potential. The only disadvantage is the increased computational time due to more precise calculations. Therefore, the cutoffs of $\lambda_{\text{min},i}$ can be adjusted dependent on the performance requirements.

4.4.2 Computational efficiency

The reason to use the adaptive-precision EAM-ACE potential is to save computational time, which is why the computational efficiency is analysed in the following. First, the imbalance of the work is investigated in Section 4.4.2.1. The computational time of the force calculations is analysed in Section 4.4.2.2. Finally, the achieved speedup is presented in Section 4.4.2.3.

4.4.2.1 Imbalance

The processor with the highest work restricts the speed of a simulation since faster processors need to wait due to communication and synchronisation. One can quantify this effect with the imbalance I (cf. Eq. (4.31)), where $I = 1$ corresponds to a perfectly balanced system and $I > 1$ indicates imbalance. The mean imbalance in the nanoindentation is 1.41 with a standard deviation of 0.26. Dynamic load-balancing of an adaptive-precision potential is challenging as the work distribution changes drastically when the precision of atoms is switched from fast to precise or vice versa as discussed in the following.

We use a nanoindentation to discuss the challenges of dynamic load-balancing of a simulation with an adaptive-precision potential in the following. The numbers of precise and fast calculations per processor are shown in Fig. 4.8a as 2D-histogram of all load-balancing steps, where load-balancing is done as described in Section 4.2.4. The 2D-histogram shows that balancing few precise calculations on some processors with much more fast calculations on other processors works. More precisely, the domains with the fewest fast calculations perform on average 568 precise calculations, where the domains with the fewest precise calculations perform on average 44245 fast calculations.

To analyse dynamic load balancing further and include also non-performant load-balancing strategies, we only use the start of the equilibration instead of a whole nanoindentation. The required core-hours per timestep during the equilibration of a surface in a NVT ensemble and the imbalance are shown in Fig. 4.8b for different load-balancing strategies. Our load-balancing method applying a staggered grid (SG) and estimating the work per particle with the four force-calculation times $\rho_p^{(\mathcal{X})}$ (cf. Eq. (4.30)) results in an imbalance of 1.88 when used every 100 timesteps. Not estimating the work per particle but per processor p with only one time measurement of the total force-calculation time τ_p means that the actual load distribution on the processor is unknown to the load balancer. Thus, load balancing with only one average work ρ_p (cf. Eq. (4.30)) per processor using the recursive coordinate bisectioning (RCB) method of LAMMPS results in an imbalance of 6.17. The imbalance difference between SG and RCB demonstrates the benefit of using one average work $\rho_p^{(\mathcal{X})}$ per individual force calculation $\mathcal{X}$. The imbalance 1 of a perfectly balanced system is not reached since the system is dynamic and changes the workload in each timestep.

An atom i in a bulk of only thermally fluctuating atoms can change its value $\lambda_{0,i}$ to a value smaller 1 due to a spontaneous fluctuation which might be reversed in one of the next timesteps and therefore produces workload fluctuations due to changes of the neighbouring $\lambda_{\text{min},j}$ parameters according to Eq. (4.13). More concrete, the atom changes the switching parameter of all neighbouring atoms within the cutoff $r_{\lambda,\text{hi}}$ to a value smaller 1 which implies a precise and expensive ACE calculation. Within the cutoff $r_{\lambda,\text{hi}}^{\text{Hby1}} = 12 \text{ \AA}$ are 626 atoms at 0 K which is more than the 568 precise calculations for the processors with the fewest fast calculations in Fig. 4.8a. Thus, one

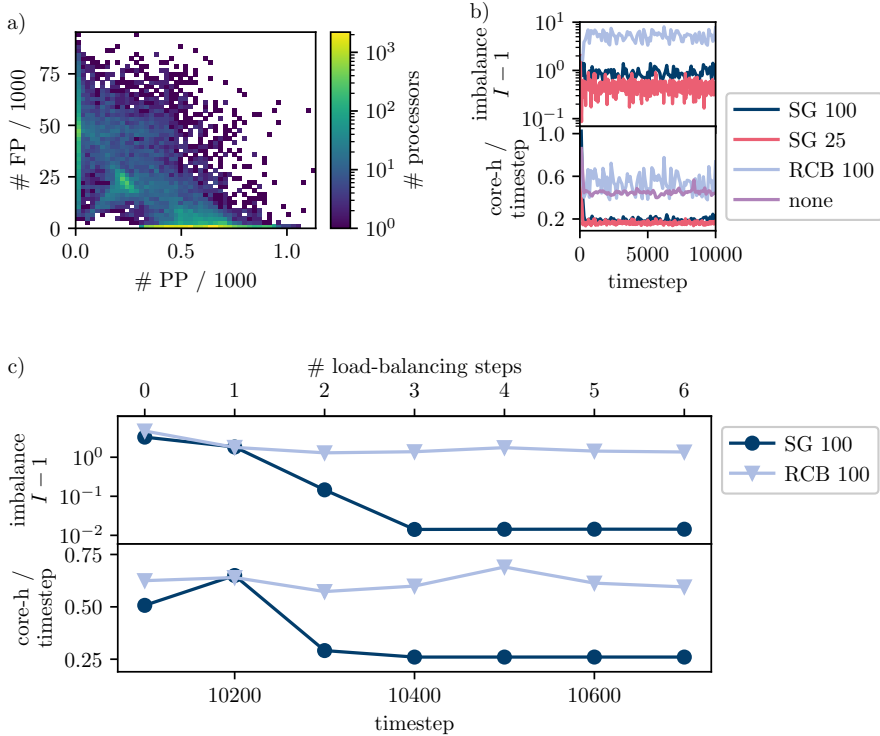


Figure 4.8: a) Number of atoms per processor which require the calculation of the fast potential (FP) and the precise potential (PP) for all load-balancing steps of a nanoindentation in a cubic system of 100^3 unit cells and load balancing with a staggered grid domain decomposition. b) Imbalance I according to Eq. (4.31) and core-h per timestep during equilibration of a surface in a NVT ensemble compared for different load-balancing methods. The staggered grid method (SG) uses per processor p the work $\rho_p^{(\mathcal{X})}$ of all four force calculations $\mathcal{X}$, where the recursive coordinate bisectioning (RCB) method only uses one work ρ_p per processor and is thus worse balanced. The number behind the load balancing method in the legend is the number of timesteps after which the corresponding load balancer is called. c) Load balancing in the system of b) but with static atoms only to test the behaviour of the load balancers.

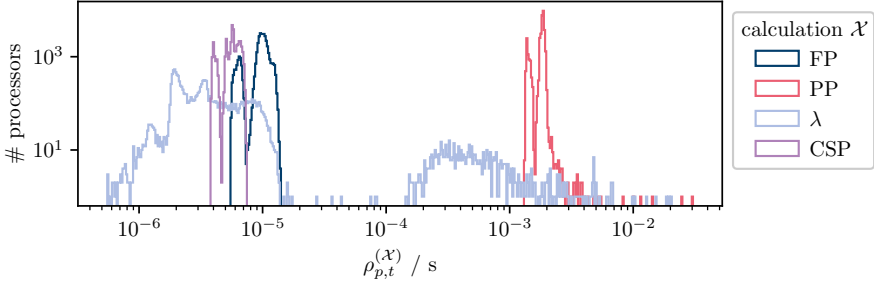


Figure 4.9: Histogram of the average work $\rho_{p,t}^{(\mathcal{X})}$ for all load-balancing steps of a nanoindentation in a cubic system of 100^3 unit cells and load balancing with a staggered grid domain decomposition.

atom with $\lambda_{\min,i} < 1$ can double the work of its processor and one cannot expect an imbalance of 1 according to Eq. (4.31) for such a dynamic system. However, the staggered-grid load balancer reaches with 1.47 a better imbalance when called every 25 timesteps since it can better follow the dynamics of the system. When we freeze all atoms in the NVT equilibration of the surface, we get a static force calculation after $N_{\lambda,\text{avg}}^{\text{Hyb1}} + N_{\text{CSP,avg}}^{\text{Hyb1}} = 220$ timesteps. The SG method reduces the imbalance of this static system up to 1.02 as shown in Fig. 4.8c. Hence, the staggered-grid-load balancer works, but the system dynamics is challenging. In contrast, the RCB-load balancer cannot balance the static system due to the missing per atom and calculation $\mathcal{X}$ force-calculation times $\rho_p^{(\mathcal{X})}$ (cf. Eq. (4.30)).

4.4.2.2 Calculation specific computational time

The average work per atom for the four subprocesses is $\langle \rho_{p,t}^{\text{FP}} \rangle_{p,t} = 9.6 \mu\text{s}$, $\langle \rho_{p,t}^{\text{PP}} \rangle_{p,t} = 1.7 \text{ ms}$, $\langle \rho_{p,t}^{\lambda} \rangle_{p,t} = 48.6 \mu\text{s}$ and $\langle \rho_{p,t}^{\text{CSP}} \rangle_{p,t} = 5.6 \mu\text{s}$, where the processor-dependency is discussed in the following.

Histograms of the work $\rho_p^{(\mathcal{X})}$ according to Eq. (4.30) used at all load balancing steps of a nanoindentation calculated on 384 processors on JURECA-DC[166] are shown in Fig. 4.9. $\rho_{p,t}^{\lambda}$ with $\langle \rho_{p,t}^{\lambda} \rangle_{p,t} = 48.6 \mu\text{s}$ is distributed over four orders of magnitude since the calculation time required on atom i depends also on $\lambda_{0,j}$ of neighbouring atoms. Since the number of atoms with $\lambda_{0,i} \neq 1$ should be small compared with the number of all atoms, we iterate over all atoms with $\lambda_{0,i} \neq 1$ and calculate $\lambda_{\min,j}$ for all neighbours j of i . As we search a minimum switching parameter $\lambda_{\min,j}$ per neighbour, we can abort the calculation for $\lambda_{\min,j} \leq \lambda_{0,i} \neq 1$ without calculating r_{ij} and $f^{(\text{cut})}$. As the calculation of $\lambda_{\min,j}$ is only required on the processor which administrates particle j it is not required to compute the parameter $\lambda_{\min}$ for ghost particles. Thus, the work caused by an atom i depends on the neighbouring atoms and is not constant for all processors. The histograms show $\langle \rho_{p,t}^{\text{FP}} \rangle_{p,t} = 9.6 \mu\text{s}$, $\langle \rho_{p,t}^{\text{PP}} \rangle_{p,t} = 1.7 \text{ ms}$ and $\langle \rho_{p,t}^{\text{CSP}} \rangle_{p,t} = 5.6 \mu\text{s}$, where there are two peaks next to each other since the calculation times are dependent on the number of neighbours which is about a factor of two smaller for surface atoms. Therefore, one cannot use one constant processor-independent time per atom. The histograms show the need to measure the required work per atom per processor. Furthermore, the histograms show that the fraction of surface atoms is non-negligible. The atoms at the surface are detected by the centro-symmetry parameter and thus require a precise calculation and hence the domains administered by a processor are small. Note that excluding surface atoms from the precise calculation is possible by comparing the CSP to a given reference configuration, for example initial or equilibrium configuration, instead of using the absolute value of the CSP as input for the switching function in Eq. (4.11).

4.4.2.3 Speedup

The motivation to use an adaptive-precision potential is to save computation time compared to a precise potential while preserving accuracy in regions of the simulation where it is required. The nanoindentation with 4 million atoms calculated for 100 ps on JURECA-DC[166] with the adaptive-precision potential Hyb1 (cf. Table B.2) shows a speedup of 11.3 compared to a full ACE simulation like visualised in Fig. 4.10a. We calculated the surface and dislocations, which developed during the nanoindentation like visualised in Fig. 4.10b, precisely with the ACE-potential

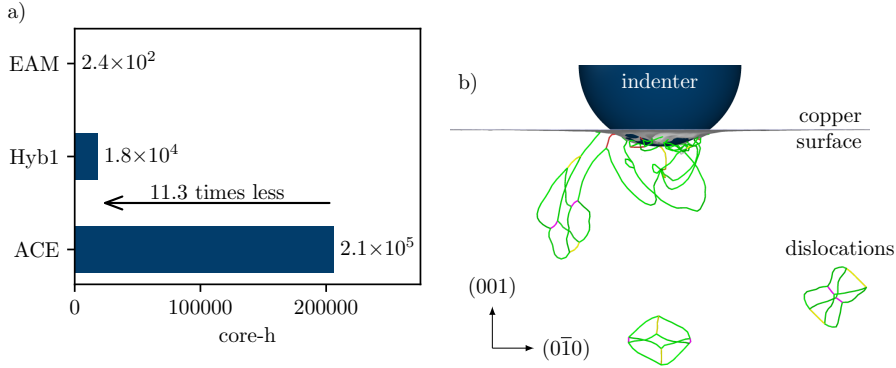


Figure 4.10: a) Total computation time of a nanoindentation with 4 million atoms (100^3 unit cells) simulated for 100 ps with the adaptive-precision potential Hyb1 compared to ACE and EAM nanoindentations. EAM, hybrid and ACE simulations are calculated on 128, 384 and 2048 cores of JURECA-DC[166] respectively. b) Visualisation of the dislocation line defects which occur in a nanoindentation. The visualisation is done with OVITO[143] and the dislocations are identified by the dislocation analysis[142] of OVITO.

and saved computation times for the remaining atoms by using the fast EAM-potential. Note that the amount of saved computation time strongly depends on the number of not required evaluations of the precise and expensive potential as discussed in the following using the equilibration of the nanoindentation as example.

A nanoindentation with 4 million atoms calculated on JURECA-DC[166] with the adaptive-precision potential Hyb1 (cf. Table B.2) requires 11 and 13 times less core-h for equilibration and nanoindentation itself like visualised in Fig. 4.11. The amount of saved computation time depends on the system itself. Dislocations develop during a nanoindentation and therefore there are more precisely calculated atoms at the end of a nanoindentation than at the begin or during the equilibration of the surface. Hence, one requires 59 and 33 times less core-h for the simulations with periodic boundaries in all spatial directions since there are no precisely calculated atoms at the surface. One saves even computation time for all simulation parts for Hyb2 where we increased the cutoffs $r_{\lambda,lo} = 8 \text{ \AA}$ and $r_{\lambda,hi} = 20 \text{ \AA}$ and used the remaining parameters of Hyb1.

4.5 Conclusion

We introduced a method which allows to compute adaptive-precision interatomic potentials to speedup large-scale atomistic simulations with a region or atoms of special interest. The atoms of special interest are automatically detected by a customisable detection mechanism and simulated with the precise potential while the fast potential is used for the remaining atoms. The energy model (Eq. (4.5)) ensures precise energies for the detected atoms while the switching function (cf. Section 4.2.2) is used to get also precise forces. We presented a load-balancing method with calculation specific per-atom works per processor using a staggered grid to prevent the load-balancing issues arising by combining two potentials of different cost. We demonstrated the presented adaptive-precision method by creating the copper potential Hyb1 (Table B.2) from an EAM and an ACE potential. We explained how the fast EAM potential is optimised to match bulk properties with the precise ACE potential.

A nanoindentation with 4 million atoms calculated for 100 ps was used to demonstrate the capabilities of the method. The achieved accuracy of Hyb1 for the detected atoms of interest during the nanoindentation is for forces $10 \text{ meV/\AA}$ while the exact potential energies of the ACE potential are obtained by design. The nanoindentation showed a speedup of 11.3 compared to a full ACE simulation.

Note that the amount of computation time that can be saved heavily depends on the number of not required precise calculations and therefore on the simulation setup and the detection mechanism for atoms of interest. Hence, the expected speedup cannot be generalised. Our adaptive-precision method includes overhead in form of the detection mechanism and the calculation of the switching

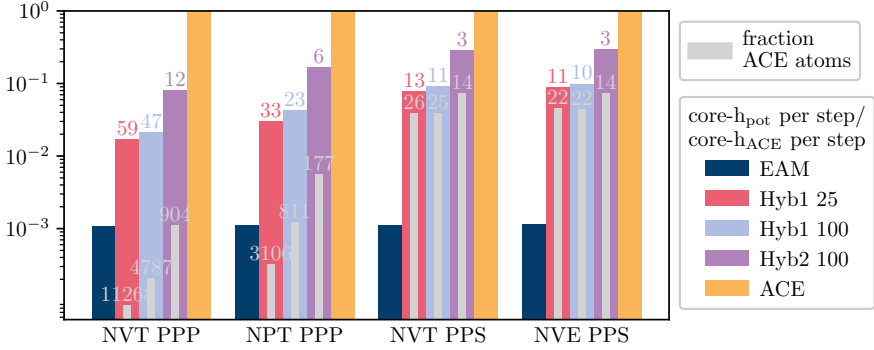


Figure 4.11: Total computation time of nanoindentations with 4 million atoms (100^3 unit cells) simulated with adaptive-precision potentials compared to ACE and EAM nanoindentations. The number behind the potential in the legend is the number of timesteps after which the staggered-grid load balancer is called. EAM, hybrid and ACE simulations are calculated on 128, 384 and 2048 cores of JURECA-DC respectively. The fraction of used core-h compared to ACE is given on top of the bars. The approximated fraction of ACE atoms is given in grey to visualise the overhead induced by the method. The times are given for all four parts of a simulation separately. NVT and NPT ensembles are simulated with periodic boundaries (PPP) to prepare a surface (PPS). The surface is equilibrated in an NVT ensemble. The nanoindentation itself is calculated in an NVE ensemble.

parameter. Therefore, in both limits, when all or no atoms require a precise calculation, using an adaptive-precision potential is not beneficial. Between both limits, however, there will be a range in the fraction of precise atoms, where the adaptive-precision potential becomes beneficial like demonstrated with the nanoindentations in which the majority of precise calculations could be accelerated. Since the force-calculation time of the precise potential is measured to enable dynamic load-balancing, one can, as discussed in Section 4.2.4, decide on-the-fly whether or not to apply the adaptive precision, i.e., whether the overhead dominates the computational benefit. In this case it is a simple switch to compute all atoms in the simulation via the accurate potential.

The force on an atom depends according to Eq. (4.6) on the switching parameters of all neighbouring atoms. Therefore, we use a spatial switching-zone to ensure a smooth transition of the potential from the precisely to the fast calculated atoms. The size of this spatial switching zone, $8 \text{ \AA}$ or $12 \text{ \AA}$ in this paper, should be small in comparison to the system size. Otherwise, a single atom which is identified for a precise potential description would cause the computation of a majority of the system atoms with the precise potential, since atoms in the transition zone require the computation of both the precise and fast potential in order to be weighted according to Eqs. (4.5) and (4.6). As mentioned earlier, an optimisation of computational time would then suggest to apply the precise potential for the complete system, in order to reduce or avoid computational overhead. This case study shows that an adaptive-precision potential is best suited for large-scale atomistic simulations where a decision can be made, which atoms require a high precision description. In many simulations, a large speedup can be achieved then by skipping most of the precise calculations. Furthermore, an adaptive-precision potential allows to embed precisely calculated atoms of interest at low cost into millions of quickly calculated atoms and thereby allows larger simulations, for which nanoindentations serve here as an example. The dislocation loops nucleate below the indenter and glide away from there (cf. Fig. 4.10b). This type of system, considered on a longer time scale, definitely requires a very big system, since dislocation loops evolve in each spatial dimension and should not artificially interact with themselves via periodic boundary conditions, which is quite likely if the system size is too small.

As discussed, our method needs a detection mechanism for atoms of interest which need to be calculated precisely. We selected the centro-symmetry parameter (CSP) as detection mechanism as we were interested in defects and surface atoms for the nanoindentation demonstration. However, one can replace this detection mechanism easily dependent on the own needs as discussed in Section 4.2.2 where we suggest detection mechanisms for grain-boundary structures, coatings,

interfaces and cracks, among others. However, our proposed dynamical adjustment of the group of atoms is not appropriate for spontaneous phenomena that cannot be anticipated by the detection mechanism as one cannot ensure a smooth transition from the fast to the precise potential which would result in artefacts.

The application of an adaptive-precision potential for other situations where the highest precision is required only locally but not globally like interfaces, cracks and grain boundaries is straightforward while the generalisation of the detection mechanism for non-crystalline systems like amorphous solids is a natural next question. Automated training of the fast potential would improve the usability of adaptive-precision potentials. The present work is based on a CPU-version of our method and, therefore, could be improved by the usage of GPUs.

Chapter 5

Nanoindentation

The application of the adaptive-precision method, that was presented in Chapter 4, in large-scale nanoindentations is published in my paper *Nanoindentation simulations for copper and tungsten with adaptive-precision potentials* [167], on which the present chapter is based.

Large-scale molecular dynamics (MD) simulations of nanoindentations [72, 73] can provide valuable insights into microscopic mechanical behaviour of metallic materials and reveal underlying mechanisms that govern initial stages of plastic deformation. To obtain trustworthy results it is essential to describe the atomic interactions with accurate and transferable interatomic potentials [73]. For free-electron metals with close-packed face-centred cubic (fcc) crystal structures, many-body central force approaches, such as the embedded atom method [53] or the Finnis-Sinclair potentials [133], usually provide sufficient accuracy. However, many transition metals with partially filled d-bands cannot be described properly by such central-force models [52, 54–60]. These metals, crystallising in hexagonal close-packed (hcp) or body-centred cubic (bcc) structures, require explicit descriptions of their angular-dependent interactions.

Two prototypical elements representing these families of metals are fcc copper and bcc tungsten. Understanding mechanical properties of both metals at the nanoscale is highly relevant due to ever increasing miniaturisation. The refractory metal W is also considered as plasma facing material (PFM) for next generation fusion reactors [168–170]. As PFMs need to sustain extreme operating conditions, understanding changes of their mechanical properties due to formation of large number of crystal defects under irradiation is of utmost importance. Even though nanoindentation simulations have already provided valuable mechanistic insights into microscale plasticity for both Cu [16, 83, 171, 172] and W [2, 77, 84, 85, 173, 174], further investigations and validations with improved interatomic potentials are still needed, especially for bcc W.

In recent years, machine-learning (ML) potentials proved to be able to deliver an accuracy close to that of first-principles electronic-structure methods at a fraction of their computational cost [4, 175–178]. Even though there exist ML potentials for Cu [4] and W [69, 179–182], their application to large-scale nanoindentation simulations is still limited. One contributing reason is that most ML potentials remain several orders of magnitude slower than the simple empirical potentials [4, 62, 66].

A viable way to overcome the performance gap between ML and simple empirical potentials is via the adaptive-precision (AP) method [146], that was developed and presented in Chapter 4. In the AP method, the potential energy is a weighted average of the combined potentials depending on a switching parameter. The switching parameter is calculated by a customisable detection mechanism so the assignment of atoms to a particular region can be done in an automatised way. Thus, the method works autonomously and self adaptively in space and time. An important feature of the AP method is that the time integration of the equations of motion maintains the conservation of energy and momentum.

In this chapter, we apply the AP method to combine the speed of EAM potentials and the accuracy of ACE potentials in large-scale nanoindentation simulations. Furthermore, we extend the original AP approach to account properly for homogeneous nucleation of dislocations that takes place below the indenter in regions of highest shear stress [144]. These events marking the onset of plasticity need to be simulated with the accurate ACE potential. We also perform EAM-only and ACE-only simulations for comparison to assess the usage of the AP method.

The present chapter is organised as follows: first, the setup of the nanoindentations is described in Section 5.1.1. Afterwards, the used interatomic potentials are presented in Section 5.1.2, including the new region-of-interest description for an adaptive-precision potential. We simulated nanoindentations with EAM potentials, adaptive-precision EAM-ACE potentials and ACE po-

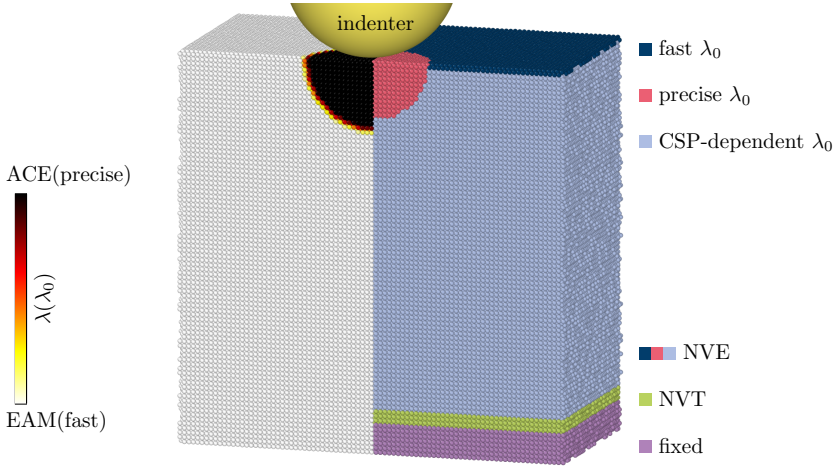


Figure 5.1: Cut through the system along the (100) plane below the indenter. Right side: the top atoms (■) are simulated in a NVE ensemble, the bottom atoms are fixed (■), and the atoms in between are simulated in a NVT ensemble (■). For simulations with an adaptive-precision potential, the parameter λ_0 is used to calculate the switching parameter λ which is required to calculate the potential energy according to Eq. (4.5). To ensure a precise simulation, $\lambda_0 = 0$ is set after the equilibration for all atoms which are in a hemisphere (■) whose centre is on the surface of the material under the centre of the indenter. The centro-symmetry parameter (CSP) is used to calculate λ_0 for most atoms (■) to dynamically adjust the precision. To save the computation time, $\lambda_0 = 1$ is set at the surface outside of the hemisphere (■) to prevent the detection of the surface by the CSP. Left side: initially used switching parameter λ calculated from λ_0 . A region of interest below the indenter is calculated precisely while the remaining atoms are calculated quickly.

tentials. The nanoindentations are analysed and the influence of the potential is investigated in Section 5.2. Section 5.3 concludes with a comparison between the Cu and W simulation results.

5.1 Methods

This section describes how we simulated the nanoindentations (Section 5.1.1) and the used inter-atomic potentials (Section 5.1.2).

5.1.1 Nanoindentation

The MD simulations were performed using the LAMMPS software package [1]. One of common approaches in the nanoindentation simulations is to separate the simulation block into three parts [14, 77, 84], as visualised in Fig. 5.1. The bottom part is kept fixed to prevent rotation and translation of the sample, the central part is simulated using the NVT ensemble to dissipate heat generated by the indentation, and the top part is simulated using the NVE ensemble. The spherical rigid indenter was described by the following potential [76]

$$V_{\text{ind}}(\mathbf{r}) = \frac{k_{\text{ind}}}{3} \Theta(R_{\text{ind}} - |\mathbf{r}_{\text{ind}} - \mathbf{r}|)(R_{\text{ind}} - |\mathbf{r}_{\text{ind}} - \mathbf{r}|)^3, \quad (5.1)$$

where k_{ind} is the force constant, $\mathbf{r}_{\text{ind}}(t)$ the centre of the indenter, R_{ind} the indenter radius, and $\Theta(x)$ the Heaviside step function. In the beginning of the simulation, the indenter and surface are not in contact and the centre of the indenter is located at $\mathbf{r}_{\text{ind},0}$. The indenter is then gradually inserted into the sample with a constant velocity $\mathbf{v}_{\text{ind}}$ such that its position changes as $\mathbf{r}_{\text{ind}}(t) = \mathbf{r}_{\text{ind},0} - \mathbf{v}_{\text{ind}}t$.

We use a relaxation techniques adapted from Refs. [14, 183]. At first, a perfect lattice is created at 0 K. The perfect lattice is simulated with periodic boundary conditions in all dimensions as NVT ensemble with a Langevin thermostat with the damping constant γ^L until the temperature is within the tolerance δ_T to the target temperature T_{targ} . In the second step, a NPT ensemble is

Table 5.1: Parameters used during the three steps of equilibration and during the nanoindentation as described in Section 5.1.1.

property	copper			tungsten		
	EAM	AP	ACE	EAM	AP	ACE
step 1: NVT periodic boundaries						
γ^L/ps	1	1	1	1	1	1
δ_T/K	0.2	0.2	0.2	0.2	0.2	0.2
step 2: NPT periodic boundaries						
δ_p/bar	10	10	10	10	10	10
$\gamma_T^{\text{NH}}/\text{ps}$	0.1	0.1	0.1	0.1	0.1	0.1
$\gamma_p^{\text{NH}}/\text{ps}$	1	1	1	1	1	1
$p_{\text{targ}}/\text{bar}$	0	0	0	0	0	0
step 3: NVT surface						
$\Delta t_E/\text{ps}$	50	50	50	100	2	100
γ^L/ps	10	10	10	0.1	0.1	0.1
$\Delta t_{\text{mom}}/\text{ps}$	1	1	1	1	1	1
step 4: nanoindentation						
$\gamma_T^{\text{NH}}/\text{ps}$	0.1	0.1	0.1	0.1	0.1	0.1
all steps						
T_{targ}/K	292	292	292	300	300	300
$\Delta t/\text{ps}$	0.001	0.001	0.001	0.001	0.001	0.001
$\Delta t_{\text{RCB}}/\text{ps}$	0.1	-	0.1	0.1	-	0.1
$\Delta t_{\text{SG}}/\text{ps}$	-	0.025	-	-	0.025	-

simulated according to Nose-Hoover with damping parameters γ_T^{NH} and γ_p^{NH} for temperature and pressure until target temperature and target pressure p_{targ} are within the tolerance δ_T and δ_p of the target values. At this target temperature and pressure, the surface is created at the top of the simulation box in the z direction and the open boundary conditions are used instead. The bottom atoms in the z direction are frozen at their current positions and their velocity is set to zero and not updated any more. In a third simulation of the duration Δt_E a NVT ensemble with a Langevin thermostat is applied. The thermostat damps the pressure waves of the created surface. The total momentum is set to zero repeatedly after the time interval Δt_{mom} since it is not conserved by the Langevin thermostat. All equilibration steps and the nanoindentation itself are simulated with the timestep Δt .

Dynamic load balancing is essential for the adaptive-precision simulations as the time required to calculate forces and energies for a particle heavily depends on the used interatomic potential. We use a staggered grid[163] as spatial domain-decomposition[161] which is balanced by ALL[162] as described in Section 4.2.4. For the EAM and ACE simulation, we use the recursive coordinate bisectioning (RCB)[184] from LAMMPS which balances LAMMPS' tiled domain layout according to the measured force-calculation time. The load balancer is called after every time interval Δt_{RCB} or Δt_{SG} if the system is imbalanced enough.

All mentioned parameters are listed in Table 5.1 for the copper and tungsten simulations with all three interatomic potentials.

5.1.2 Interatomic potentials

The used EAM, ACE and adaptive-precision EAM-ACE potentials are described in the following. Details regarding the used potentials are provided in Appendix B.

5.1.2.1 Embedded atom model

In the embedded atom model (EAM; cf. Section 4.1) potentials used in this chapter, the energy of an atom i is given according to Eq. (4.1) by an embedding function ξ , an electron charge density ζ and a pair potential Φ . For Cu, we used the EAM2 parametrisations of Ref. [145], which has been thoroughly tested and is widely used [149–152]. For W, we used the EAM2 potential from Ref. [148], which was developed for simulating radiation defects and dislocations and is widely used[77, 185–187].

Table 5.2: Parameter values used in the copper nanoindentation simulations.

parameter	value
surface orientation	(100)
temperature	292 K
simulation box size	$364.2 \text{ \AA} \times 364.2 \text{ \AA} \times 363.2 \text{ \AA}$
number of atoms	4000000
height of fixed region	10 $\text{\AA}$
height of NVT region	10 $\text{\AA}$
timestep Δt	1 fs
indenter radius R_{ind}	60 $\text{\AA}$
indenter velocity $\mathbf{v}_{\text{ind}}$	$\{0, 0, 25\} \text{ m/s}$
indenter force constant k_{ind}	$10 \text{ eV \AA}^{-3}$
maximum indentation depth	20 $\text{\AA}$
initial indentation depth	-5 $\text{\AA}$

5.1.2.2 Atomic cluster expansion

The employed atomic cluster expansion (ACE; for details see Section 3.1) potentials used a combination of linear and square-root expansions that were shown to give the best accuracy and computational performance [4]. The potential energy of an atom i described by ACE is given according to Eq. (3.2), where an expanded property $\varphi_i^{(p)}$ is given by Eq. (3.6). For Cu, we used the ACE parametrisation developed in Ref. [4], which is widely used [188–190], for example, in dislocation simulations [191]. For W, we used an ACE parametrisation trained on an extensive DFT dataset of bulk structures as well as defects. Further information of this potential is provided in Section B.2.2.

5.1.2.3 Adaptive-precision potential

The atomic energy of the AP potential, which combines the fast EAM and the precise ACE as described in Chapter 4, is given by Eq. (4.5), i.e., the switching parameter $\lambda_i \in [0, 1]$ decides whether ACE ($\lambda_i = 0$), EAM ($\lambda_i = 1$) or an linear interpolation between EAM and ACE ($\lambda_i \in (0, 1)$) is used to describe the potential energy of an atom i . We used the switching parameter based on the centro-symmetry parameter (CSP) [76], which was presented in Section 4.2.2.2 to detect atoms that need to be treated with the more accurate potential. In addition, we fixed the value of λ_i in specific regions of the simulation block, as visualised in Fig. 5.1. Since the first dislocations nucleate primarily in the regions of highest shear stress near the indenter surface [144], the region below the indenter is always simulated with the ACE potential. In practice, we set the initial value of $\lambda_{0,i} = 0$ in a hemisphere of radius 40 $\text{\AA}$ whose centre is at the contact point where the indenter first touches the surface. Additionally, to prevent the CSP from detecting surface atoms, we set $\lambda_{0,i} = 1$ for surface atoms outside of the ACE hemisphere. For the remaining atoms, $\lambda_{0,i}$ is calculated during the simulation based on the CSP according to Eq. (4.11). The switching parameter $\lambda_i(\{\lambda_{0,j}\})$ is calculated according to Eqs. (4.13) to (4.15). Using this setup, we minimise the amount of the computationally more expensive ACE calculations. λ_i is recalculated and updated dynamically during a simulation as described in Section 4.2.3. For Cu, we use the parametrisation Hyb1 (cf. Table B.2).

5.2 Results

The copper nanoindentations are compared in Section 5.2.1, followed by the comparison for tungsten in Section 5.2.2. Finally, the computational efficiency is discussed in Section 5.2.3.

5.2.1 Copper

Before starting the nanoindentation simulation, we equilibrate the $\text{Cu}_{(100)}$ surface as described in Section 5.1.1. The nanoindentation simulation is then performed using the NVE ensemble, but a thin layer of atoms at the bottom of the simulation box is thermostated and the very bottom layers are kept fixed to dissipate heat generated during the indentation and to prevent translation and rotation of the sample, as visualised in Fig. 5.1. The fundamental parameters of the simulation are summarised in Table 5.2.

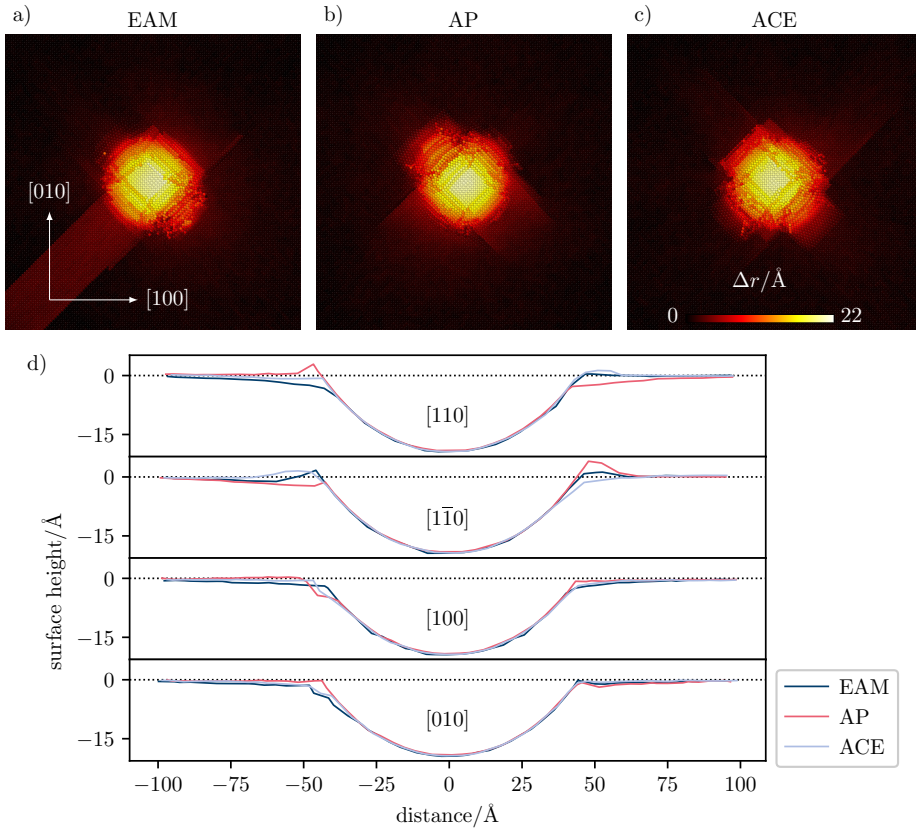


Figure 5.2: a-c) Displacement Δr of the copper atoms at 20 Å indentation depth compared with -5 Å indentation depth. d) Cross sections through the centre of the supercell below the indenter along different directions at 20 Å indentation depth.

The displacement of the surface atoms which developed during the nanoindentations with the three potentials is visualised in Fig. 5.2(a-c). The height of the surface on a line through the contact point of surface and indenter is shown for cross sections along the $\langle 110 \rangle$ and $\langle 100 \rangle$ directions in Fig. 5.2(d). We observe small pile-ups in the $\langle 110 \rangle$ directions, similarly to Ref. [16], and no pile-up in the $\langle 100 \rangle$ directions.

The force between an atom i and the indenter is given as $\mathbf{F}_{\text{ind}}(\mathbf{r}_i) = -\nabla_{|\mathbf{r}_{\text{ind}} - \mathbf{r}|} V_{\text{ind}}(\mathbf{r}_i)$ with V_{ind} according to Eq. (5.1). The indentation load

$$\mathbf{P}_{\text{ind}} = \sum_i \mathbf{F}_{\text{ind}}(\mathbf{r}_i) \cdot \frac{\mathbf{v}_{\text{ind}}}{v_{\text{ind}}} \quad (5.2)$$

is given as total force on all atoms in the indentation direction $[00\bar{1}]$ and automatically calculated during the simulation. The Hertzian analysis [79, 192, 193] is applied to evaluate the Hertzian load P_H on the surface as a function of the indentation depth h as

$$P_H = \frac{4}{3} E^* R_{\text{ind}}^{1/2} h^{3/2}, \quad (5.3)$$

where R_{ind} is the indenter radius and E^* the indentation modulus. This formula does not consider elastic anisotropy of materials, but it holds for spherical indenters with a modified indentation modulus [193–195]. The Hertz fit allows to detect the onset of plastic deformation. Both the simulated load P_{ind} and the fitted analytical solution P_H are shown in Fig. 5.3(a). The fits give $E_{\text{EAM}}^* = 115.9$ GPa, $E_{\text{AP}}^* = 116.1$ GPa and $E_{\text{ACE}}^* = 112.0$ GPa and are thus similar for all potentials.

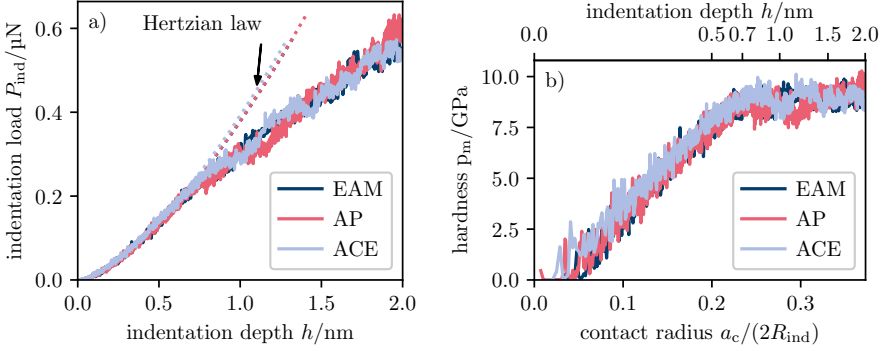


Figure 5.3: a) Indentation load P_{ind} and elastic Hertzian load P_H according to Eqs. (5.2) and (5.3) computed for the copper nanoindentations. b) Mean contact pressure or hardness p_m according to Eq. (5.4) dependent on the contact radius a_c according to Eq. (5.5). The contact radius is normalised with the radius R_{ind} of the spherical indenter.

The hardness or mean contact pressure p_m on the surface is

$$p_m = \frac{P_{\text{ind}}}{A_c} = \frac{P_{\text{ind}}}{\pi a_c^2}, \quad (5.4)$$

where A_c is the projected contact area with the contact radius a_c . There are different models for the contact radius [16, 172, 196]. We use the contact radius

$$a_c = \sqrt{h(2R_{\text{ind}} - h)} \quad (5.5)$$

as this model was applied in nanoindentation simulations of Cu [16] and W [84] with spherical rigid indenters and thus enables a seamless comparison of the results with the literature. The mean contact pressure p_m is shown in Fig. 5.3(b). It increases linearly until plastic deformation sets in at $a/(2R_{\text{ind}}) \approx 0.23$ and reaches a steady state for larger contact radii. This behaviour is independent of the interatomic potential used, as expected from Refs. [77, 196]. The average hardness at the steady state is 8.9 GPa with a standard deviation up to 0.5 GPa. The hardness reported for Cu₍₁₀₀₎ in Ref. [16] is 11.7(14) GPa and thus somewhat higher, but the indenter velocity was two times faster than in the present work; as the hardness increases with the indenter velocity [197, 198], a larger hardness magnitude is therefore expected.

The dislocation extraction algorithm (DXA) [142] implemented in OVITO [143] was used to calculate the dislocation length l_{klm} of dislocations of type klm . For the fcc lattice, DXA identifies dislocations with the Burgers vectors $1/6\langle 112 \rangle$, $1/6\langle 110 \rangle$, $1/3\langle 100 \rangle$, $1/2\langle 110 \rangle$, $1/3\langle 111 \rangle$ and other Burgers vectors. Dislocation networks observed in the simulation for the indentation depth $h/a_c \approx 0.34$ are shown in Fig. 5.4.

Prismatic dislocation loops nucleate as in Ref. [16] for all potentials. The prismatic dislocation loops glide in the directions $[10\bar{1}]$, $[\bar{1}01]$, $[01\bar{1}]$ and $[0\bar{1}1]$. Five dislocation loops nucleate during the simulation for EAM while four nucleate for the AP potential and ACE.

The dislocation density

$$\rho_{klm}^{\text{dxa}} = l_{klm}/V^{\text{dxa}} \quad (5.6)$$

is calculated within the effective volume V^{dxa} of the plastic zone. The radius a_{pz} of the plastic zone is given by

$$a_{\text{pz}} = f_{\text{pz}} a_c, \quad (5.7)$$

where the factor $f_{\text{pz}} \in [0, 3.5]$ depends on the material [199]. For Cu, we used $f_{\text{pz}} = 1.9$ [200]. Thus, the effective volume of the plastic zone is given by

$$V^{\text{dxa}} = 2\pi a_{\text{pz}}^3/3 - V_{\text{ind}}, \quad (5.8)$$

where the volume V_{ind} displaced by the indenter, without considering sink-in and pile-up effects, is given as [77, 201].

$$V_{\text{ind}} = \pi h^2 (R_{\text{ind}} - h)/3. \quad (5.9)$$

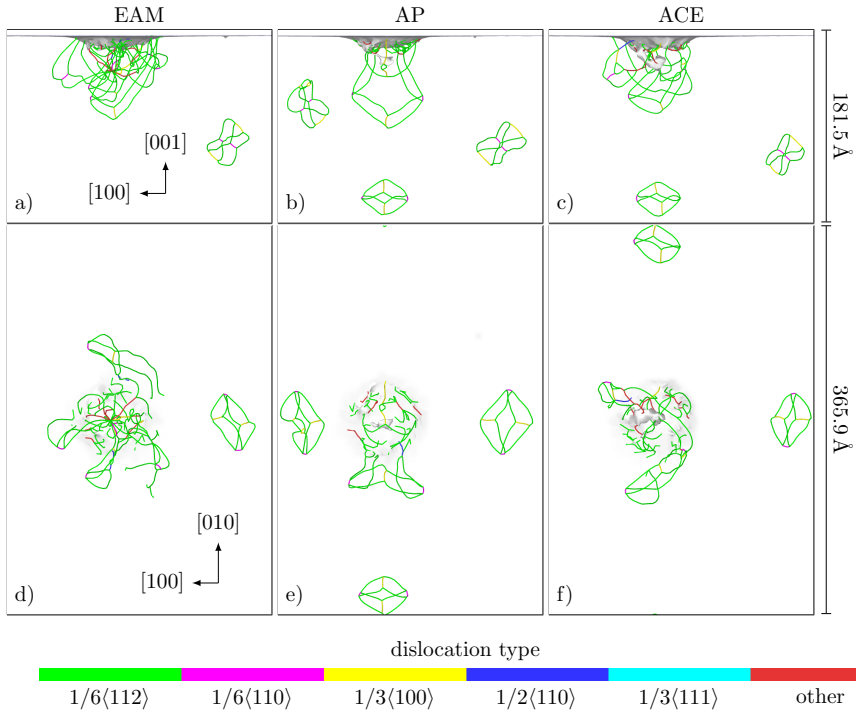


Figure 5.4: Dislocation lines in Cu identified by the DXA analysis of OVITO for a normalised indentation depth of $h/a_c \approx 0.34$. The dislocation lines are colour-coded according to their identified dislocation type. A scale bar is given on the right side for every row.

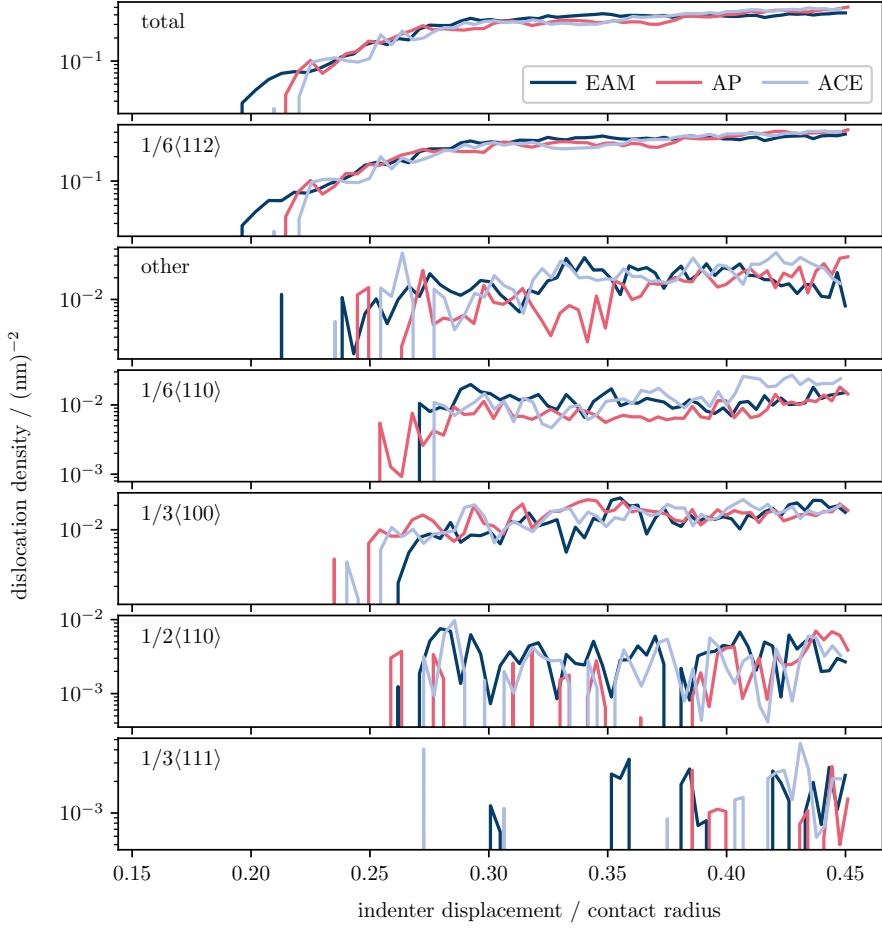


Figure 5.5: Dislocation density according to Eq. (5.6) for nanoindentations of $\text{Cu}_{(100)}$. The dislocation density is given for all by the DXA of OVITO identified dislocation types. Furthermore, the total type-independent dislocation density is shown.

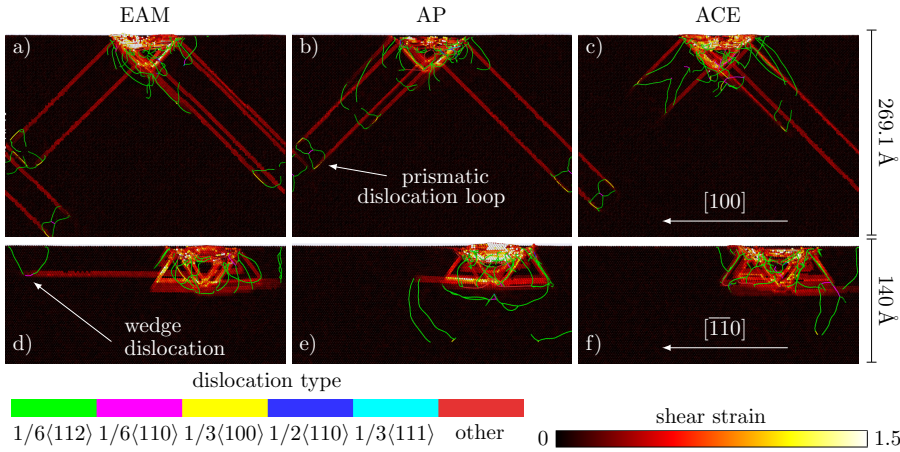


Figure 5.6: The copper atoms are colour-coded according to the shear strain evaluated using OVITO in the a-c) (010) and d-f) ($\bar{1}\bar{1}0$) plane through the contact point for a normalised indentation depth of $h/a_c \approx 0.45$. The dislocation lines computed with the DXA analysis of OVITO are shown for 35 Å in the normal direction of the corresponding plane and colour-coded according to their dislocation type. A scale bar is given on the right side for every row.

The densities of different dislocation types as a function of the indentation depth are shown in Fig. 5.5. As expected, the first nucleated dislocation has the Burgers vector $1/6\langle 112 \rangle$ for all potentials, but the nucleation occurs earlier for EAM than for the AP potential and ACE. We observe all dislocation types being nucleated for all potentials.

The shear strain was evaluated using OVITO according to the method described in Ref. [202], where the initial configuration was used as a reference. The shear strain is visualised at the end of the simulations in Fig. 5.6. The red slip traces in Fig. 5.6(a-c) in the $[10\bar{1}]$ and $[\bar{1}0\bar{1}]$ directions with a shear strain of about 0.3 mark the glide of prismatic dislocation loops. Furthermore, a wedge-shaped dislocation develops close the surface in the EAM simulation (see Fig. 5.6d) as in Ref. [83] and glides in the $[\bar{1}\bar{1}0]$ direction. This leads $1/2[\bar{1}\bar{1}0]$ atomic displacements which are visible at the surface in Fig. 5.2a.

5.2.2 Tungsten

The nanoindentation simulation for W was carried out in equivalent manner as that for Cu. The parameters of the simulation are given in Table 5.3.

The displacement of the surface atoms between initial and maximum indentation depth is

Table 5.3: Parameters and the used values in the tungsten nanoindentations performed with molecular dynamics simulations.

parameter	value
surface orientation	(100)
temperature	300 K
initial box size	$273.1 \text{ Å} \times 273.1 \text{ Å} \times 271.9 \text{ Å}$
number of atoms	1272112
height fixed region	20 Å
height NVT region	10 Å
timestep Δt	1 fs
indenter radius R_{ind}	60 Å [77]
indenter velocity $\mathbf{v}_{\text{ind}}$	$\{0, 0, 20\} \text{ m/s}$ [77]
indenter force constant k_{ind}	236 eV Å^{-3} [77]
maximum indentation depth	20.8 Å
initial indentation depth	-4.2 Å

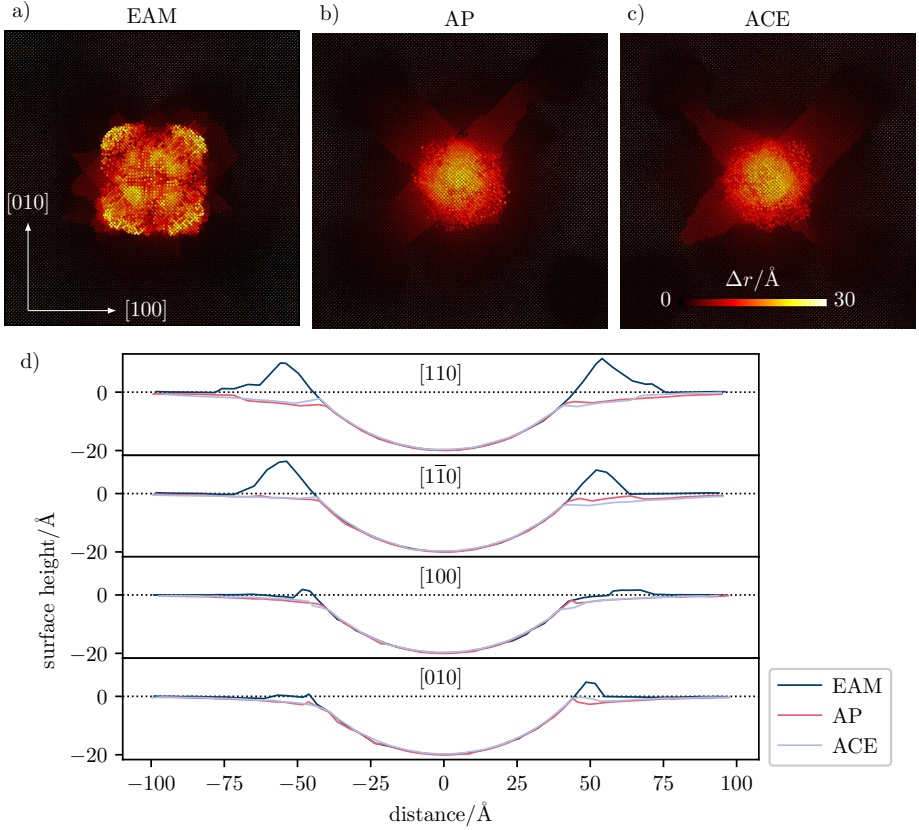


Figure 5.7: a-c) Displacement Δr of the W atoms at 20.8 Å indentation depth compared with -4.2 Å indentation depth. d) Cross sections through the centre of the supercell below the indenter along different directions at 20.8 Å indentation depth.

visualised in Fig. 5.7(a-c). In contrast to Cu, the observed surface pattern from the EAM simulation differs markedly from those of the AP and ACE simulations. For EAM, marked pile-ups are observed along the $\langle 110 \rangle$ directions while for AP and ACE the surfaces around the indenter remain almost flat. The variations of the surface height are plotted for different cross sections through the contact point of surface and indenter in Fig. 5.7(d). EAM shows a pile-up of about 10.2(13) Å in the $\langle 110 \rangle$ directions while small depressions develop in the AP and ACE simulations.

The results can be compared with those of other nanoindentation simulations [77, 203] that used the same rigid spherical indenter and the same indentation velocity. In these studies, several interatomic potentials, including a tabulated Gaussian approximation potential (tabGAP) [69], were systematically compared. The tabGAP potential is an ML potential [37] which contains up to three-body descriptors and was extensively tested for various defects in W [69]. Similar to our study, pile-ups were found for the EAM potential while surface depressions were predicted by tabGAP at 30 Å indentation depth [203].

The Hertzian loads P_H fitted to the indentation loads P_{ind} according to Eq. (5.3) are plotted in Fig. 5.8(a). The obtained indentation moduli of 519.0 GPa for EAM, 458.2 GPa for AP and 452.9 GPa for ACE confirm the consistency of AP and ACE simulations.

The hardness calculated according to Eq. (5.4) is shown in Fig. 5.8(b). The sample deforms elastically until $h \approx 8.3$ Å, 7.0 Å and 7.4 Å for EAM, AP and ACE simulations, respectively. The hardness values in the steady state of plastic deformation are 44.3(26) GPa for EAM, 35.6(30) GPa for the AP potential and 35.1(41) GPa for ACE. The pile-up, which develops in the EAM simulation, increases the contact radius while the surface depression in the AP and ACE simulations reduces the contact radius. Neither of these effects is incorporated in the analytical model of the contact

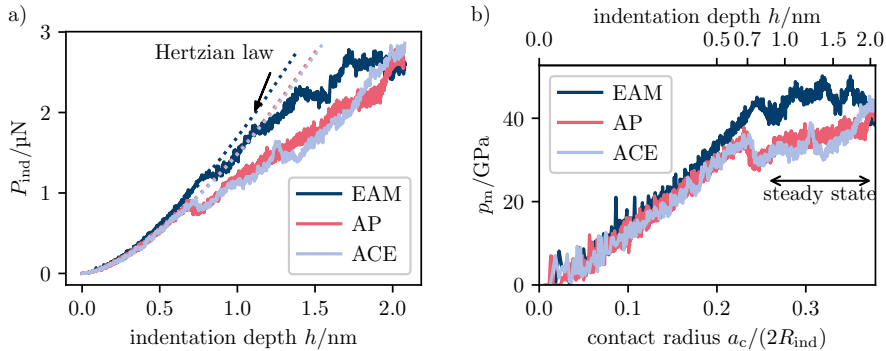


Figure 5.8: a) Indentation load P_{ind} and elastic Hertzian load P_H according to Eqs. (5.2) and (5.3) computed for the W nanoindentations. b) Mean contact pressure or hardness p_m according to Eq. (5.4) dependent on the contact radius a_c according to Eq. (5.5). The contact radius is normalised by the radius R_{ind} of the spherical indenter.

radius in Eq. (5.5). Thus, the hardness is overestimated in the EAM simulation compared to the AP and ACE simulations, which is consistent with the observed values. The hardness values are comparable with the value of 37.8 GPa reported in Ref. [84] for another EAM potential [204], where the indentation velocity of 50 m/s and the force constant of $10 \text{ eV} \text{ \AA}^{-3}$ were applied.

A convenient tool to identify defects in bcc materials is the bcc defect analysis (BDA) [205], which has been employed for inspection of several nanoindentation simulations [77, 206]. The BDA identification is based on common neighbour analysis [153], coordination number and centrosymmetry parameter. The algorithm can identify surface atoms, atoms next to a vacancy, screw and non-screw dislocations, $\{110\}$ planar faults and twin boundaries. The screw dislocations and twin boundaries cannot be distinguished by BDA, but the line character of screw dislocations and planar character of twin boundaries can be discerned easily by a visual inspection. Since deformation twinning may compete with dislocation-mediated plasticity in W nanocrystals [207], BDA provides a complementary view to the detection of dislocation lines with DXA. Both DXA and BDA visualisations for the formalised indentation depth $h/a_c \approx 0.41$ are shown in Fig. 5.9. Furthermore, the DXA and BDA results for the maximum indentation depth $h/a_c \approx 0.62$ are shown in Fig. 5.10.

For EAM, two $1/2\langle 111 \rangle$ dislocation loops are visible in Fig. 5.9(a), as they emerge from the highly distorted region below the indenter. In contrast, a deformation twin develops initially in both AP and ACE simulations. The twin structures grow in the $\langle 111 \rangle$ directions on the $\{211\}$ habit planes. Eventually, the twin propagation stops and $1/2\langle 111 \rangle$ dislocation loops start to nucleate from the twin tip, as visible in Fig. 5.9(b). This process likely occurs by coalescence of three $1/6\langle 111 \rangle$ twinning dislocations and leads to gradual retraction of the twin and transformation of the twinned region into a dislocation network. At the maximum indentation depth, as shown in Fig. 5.10(b,c), only dislocation loops are present below the indenter. Such incipient nanocontact plasticity via nucleation, propagation and annihilation of twins has been reported in simulations of bcc Ta [208]. A similar outcome was also observed in the recent tabGAP simulations [77], but some twins were still present at the maximum indentation depth of 30 Å while all twins transformed into dislocation lines in our simulations. The initial twinning deformation was not observed for any other classical potentials used in Ref. [77].

Dislocation lines identified by the DXA and BDA approaches are shown in Fig. 5.10 for the maximum indentation depth of $h/a_c \approx 0.62$. While DXA can distinguish bcc dislocations with the $1/2\langle 111 \rangle$, $\langle 100 \rangle$ and $\langle 110 \rangle$ Burgers vectors, BDA can resolve only the $1/2\langle 111 \rangle$ screw and non-screw types. The dislocation network in EAM simulations differs markedly from those in AP and ACE simulations. In the latter simulations, one can almost exclusively see dislocations with the shortest $1/2\langle 111 \rangle$ Burgers vector. The dislocation half-loops emanating from the indented region consist of long segments of predominantly screw character connected by a curved mixed/edge segment. The presence of long screws is expected due to their large Peierls stress and thermally-activated motion. The half-loops propagate away from the indenter and their interaction leads in some cases to formation of prismatic dislocation loops, as observed also in Ref. [77]. These loops continue to

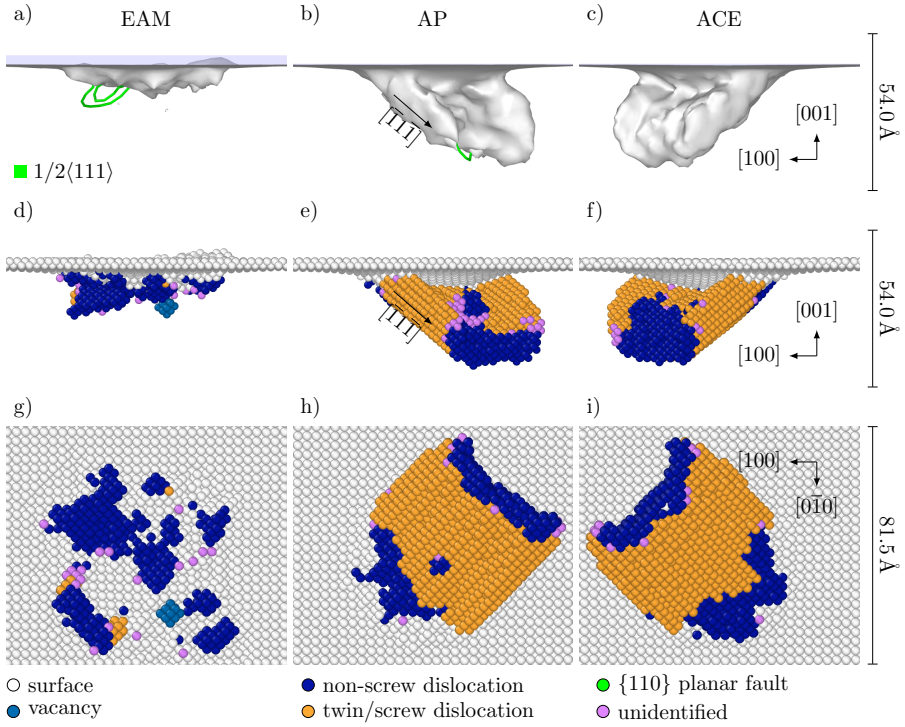


Figure 5.9: Visualisation of dislocation lines using the DXA algorithm of OVITO (a-c) and bcc defects identified by BDA (d-i) for a normalised indentation depth of $h/a_c \approx 0.41$. In BDA, the atoms are colour-coded according to the identified defect type while perfect bulk atoms are not shown. A scale bar is given on the right side for every row.

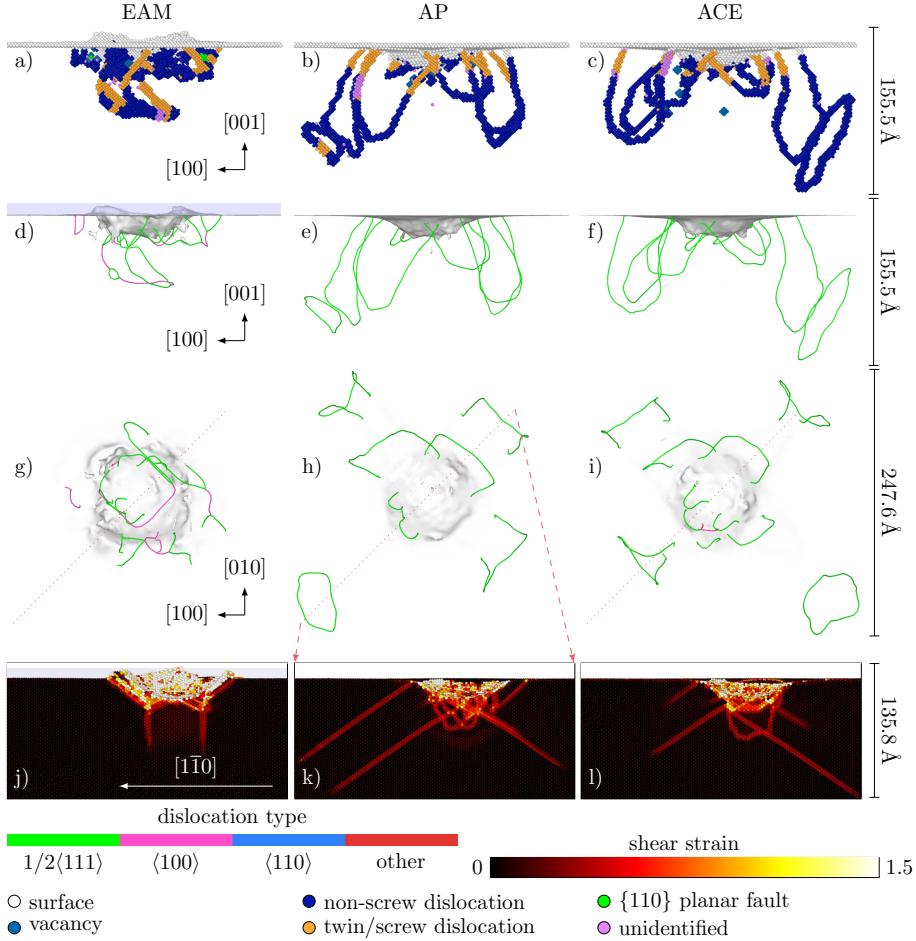


Figure 5.10: a-c) Visualisation of the atoms detected by the bcc defect analysis for the maximum normalised indentation depth of $h/a_c \approx 0.62$. d-i) Visualisation of the dislocation lines identified by the dislocation extraction algorithm viewed from different perspectives. The dotted line visualises the (110) plane. j-l) A cut in this plane through the system is used to visualise the by OVITO calculated shear strain. Scale bars are given on the right side of each row. The visualisations in the first column are from the EAM simulation, the second column contains the ones of the adaptive-precision (AP) simulation and the ACE simulation is shown in the third column.

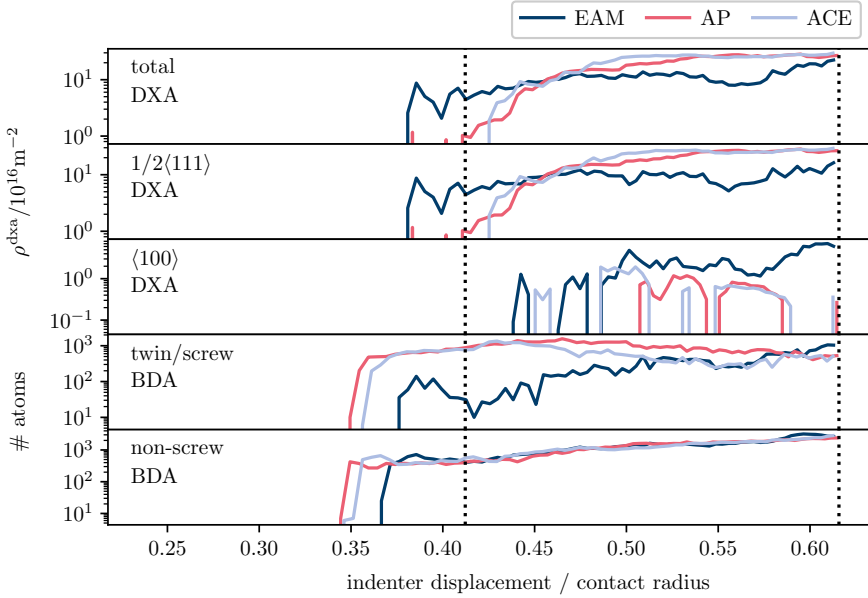


Figure 5.11: Dislocation density evaluated using DXA and BDA analyses for nanoindentations of $W_{(100)}$. Since BDA cannot distinguish twins and screw dislocations, only their sum is given. The dashed vertical lines mark the indentation depths analysed in Figs. 5.9 and 5.10.

glide in the $\langle 111 \rangle$ directions as evidenced by the slip lines in Fig. 5.10(k,l).

In contrast, there are no extended dislocation segments or $\langle 111 \rangle$ loops observed in the EAM simulation, as shown in Fig. 5.10(d). Instead, the dislocation network is concentrated primarily right under the indenter with significant portion of $\langle 100 \rangle$ dislocations that glide in the $[00\bar{1}]$ direction normal to the surface, as visible on the slip traces in Fig. 5.10(j). This observation is consistent with results from Ref. [77], where $\langle 100 \rangle$ shear loops, which nucleated below the indenter and glided in the $[00\bar{1}]$ direction, were observed in simulations that used the same EAM potential and indenter but a larger indentation depth.

The dislocation density is calculated according to Eq. (5.6). For this we need the radius a_{pz} of the plastic zone according to Eq. (5.7), where the material-dependent factor $f_{pz} = 1.9$ is used for W [77]. The dislocation density as function of the indentation depth is shown in Fig. 5.11. Furthermore, the plot also includes defect analysis using the BDA algorithm. The number of twins/screw dislocations has a maximum at 0.47 and 0.43 for the AP and ACE simulations, respectively. This is consistent with the visual analysis discussed above, since twins gradually transform to $1/2\langle 111 \rangle$ dislocations. Thus, Fig. 5.11 confirms the transition from initial twin-mediated plasticity to dislocation-mediated plasticity in the AP and ACE simulations. For EAM, there is only dislocation-mediated plasticity. Furthermore, one can clearly see the different mechanisms of dislocation-mediated plasticity between EAM simulation and ACE/AP simulations. The density of $\langle 100 \rangle$ dislocations remains high during the course of the EAM simulation while these dislocations are only intermittent in the ACE/AP simulations.

5.2.3 Computational efficiency

The nanoindentation simulations of copper (see Section 5.2.1) and tungsten (see Section 5.2.2) were carried out on the JURECA-DC supercomputer [166]. For copper, the speedup of the AP nanoindentation simulation compared to that performed with ACE only is about 21, as shown in Fig. 5.12a. The fixing of the switching parameter in the zones of interest, as described in Section 5.1.2.3, results in a speedup of 1.9 of the AP simulation compared to the AP simulation in the previous chapter (cf. Fig. 4.10a). For tungsten, the speedup of the AP simulation compared to

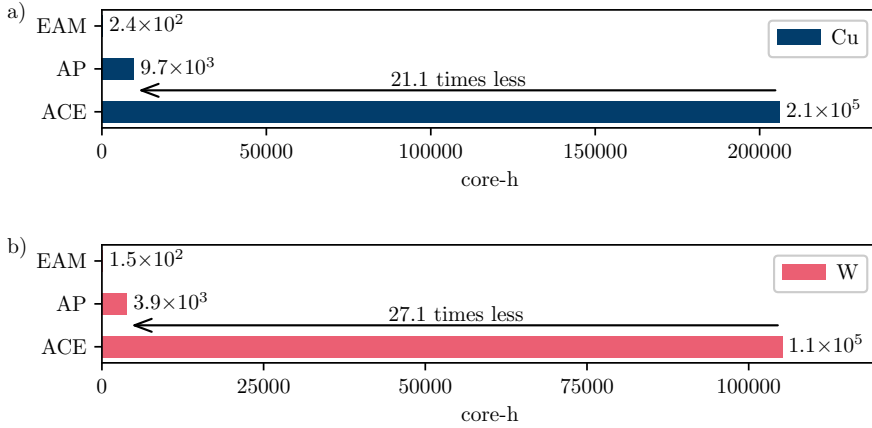


Figure 5.12: a) Total computation time of a nanoindentation with 4 million Cu atoms simulated for 100 ps with the AP potential compared to ACE and EAM simulations. EAM, AP and ACE simulations are calculated on 128, 128 and 2048 cores of JURECA-DC[166], respectively. b) Total computation time of a nanoindentation with 1.27 million W atoms simulated for 125 ps using the AP potential compared to ACE and EAM simulations. EAM, AP and ACE simulations are calculated on 128, 128 and 640 cores of JURECA-DC[166], respectively.

the ACE simulation is 27.1, as shown in Fig. 5.12b.

The computational time required for the simulation of the nanoindentations are reported in Fig. 5.12 for the whole nanoindentation. The automatic precision adjustment during the AP simulations affects, as discussed in Section 4.2.4, also the computational time, which is why an analysis of the computational time dependent on the progress of the simulation provides further insights. LAMMPS measures the number of simulated timesteps per wall-time second. The computational time τ_{atom} per atom and timestep is given as

$$\tau_{\text{atom}} = \frac{\# \text{ processors}}{\# \text{ timesteps per wall-time second} \times \# \text{ particles}}, \quad (5.10)$$

which is shown in Fig. 5.13(a) dependent on the simulated time. τ_{atom} is approximately constant for EAM and ACE for the whole simulation, while τ_{atom} starts to increase during the AP simulations. The number of precisely calculated atoms increases during the simulation, as the dislocation density ρ^{dxa} (Eq. (5.6)) increases (cf. Figs. 5.5 and 5.11). As the number of precisely calculated atoms and τ_{atom} are positively correlated (cf. Fig. 5.13(c)), the increase of τ_{atom} over time is expected.

The imbalance I is calculated by Eq. (4.31). As discussed in Section 4.2.4, $I = 1$ applies for a perfectly balanced system, while $I > 1$ indicates load imbalances. The mean imbalance in the adaptive-precision simulations of W and Cu is 1.09 and 1.29, where the visualisation dependent on the simulated time in Fig. 5.13(b) shows for Cu a decrease of the imbalance when τ_{atom} increases. Hence, the increase of τ_{atom} in the AP simulations is independent of the imbalance for W and weakened by the imbalance for Cu. Furthermore, the fixing of the switching parameter in the zones of interest, as described in Section 5.1.2.3, results in a decrease of the imbalance of 0.12 in the AP Cu simulation compared to the AP Cu simulation of the previous chapter (see Section 4.4.2.1). Thermal fluctuations of atoms affect the centro-symmetry parameter, which is used to calculate the switching parameter. As discussed in Section 4.4.2.1, an atom, which is only due to thermal fluctuations detected for a precise calculation, causes due to the spatial transition zone the computation of ACE also for neighbouring atoms and, thereby, increases the imbalance. The existence of such atoms at room temperature is less likely for W than for Cu due to the higher melting point of W. Therefore, the imbalance I is smaller for W than for Cu in the AP nanoindentations (cf. Fig. 5.13(b)). As the number of fast calculated atoms decreases over time, in particular through the gliding of prismatic dislocation loops (cf. Fig. 5.4), the number of atoms, that are susceptible to incorrect precise treatment, and, thus, the imbalance decrease at the end of the AP Cu simulation.

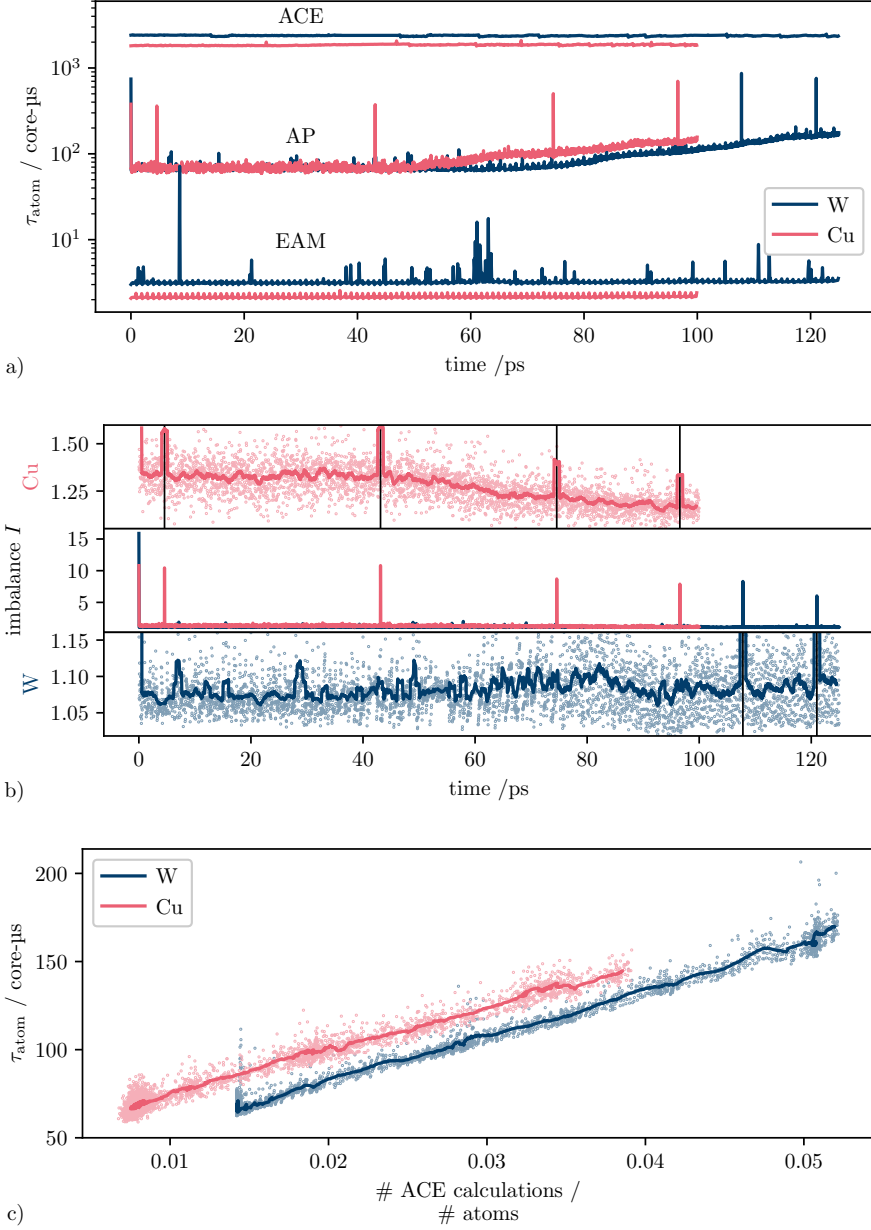


Figure 5.13: a) Computing time τ_{atom} per atom and time step according to Eq. (5.10) dependent on the simulated time for all nanoindentations. b) Imbalance I according to Eq. (4.31) dependent on the simulated time for the AP simulations. The central plot shows that the system is at the beginning and at restarts of the simulation unbalanced. The part of the imbalance-axis corresponding to a balanced system, i.e. $I \approx 1$, is enlarged at the top for Cu and at the bottom for W. The time of restart is marked with vertical lines in the enlarged plots. c) Correlation of τ_{atom} and the number of precisely calculated atoms. All measured values in c) and in the enlarged plots in b) are shown as points, while the a moving average of 1 ps-intervals is shown as line.

5.3 Conclusion

In this chapter, we systematically compared results of nanoindentation simulations for the prototypical fcc metal copper and bcc metal tungsten performed with interatomic potentials of different accuracies and computational costs. We employed computationally efficient embedded atom method (EAM) potentials, accurate but less efficient machine learning atomic cluster expansion (ACE) potentials, and an AP combination of both [146]. The selection of atoms treated by the AP potential was extended, compared to the previous chapter, to ensure an accurate description of atoms whose environment differs significantly from that of bulk crystal.

The results show that for Cu, all potentials yield similar dislocation morphologies under the indenter with only small quantitative differences. This confirms that the EAM potential can describe well the metallic bonding in free-electron metal Cu. Therefore, despite the achieved speedup of 21.1 of the AP simulation compared to the ACE simulation, one does not gain additional insights from the additional accuracy offered by the ACE potential in this case.

In contrast, markedly different plasticity mechanisms were observed for W in simulations performed with the central-force EAM potential compared to results obtained using the ACE potential, which is able to accurately describe the angular character of bonding in W due to its half-filled d-band. The EAM potential (also with optimised parameters as shown in Appendix C) predicts dislocation-mediated plasticity and sustained presence of the $\langle 100 \rangle$ dislocations below the indenter. In the ACE simulations, we instead observed a transition from initial twin-mediated plasticity to dislocation-mediated plasticity and predominance of the common $1/2\langle 111 \rangle$ dislocations. All ACE-specific mechanisms were reproduced in the AP nanoindentation simulations, however, with a significant speedup of almost 30 times compared to the ACE-only simulations. Hence, the AP potential overcomes the performance gap between the precise ACE and the fast EAM potential by combining the advantages of both potentials and is beneficial for materials where simple central-force models are not appropriate, such as for bcc transition metals.

Chapter 6

Conservative adaptive-precision potentials

The present chapter is based on my paper *Conservative adaptive-precision interatomic potentials*[209].

Computer simulations of atomistic systems are a cornerstone of research in Physics, Chemistry, Biology and Materials Science. Today atomistic simulations occupy a large part of the worlds supercomputers and high-performance computing clusters[7–11]. Making atomistic simulations more efficient is therefore an integral part of resource responsible computing.

Often the outcome of simulations is determined by only a small number of atoms. For example, in simulations of crack propagation, bond-breaking at the crack tip is decisive[107]. In simulations of plastic deformation, the generation and propagation of dislocations are key[167]. The same holds for defect nucleation sites or reaction centres in biology[91]. For many years researchers have developed frameworks to exploit hierarchical structures in simulations, where some atoms are more important or decisive than others[6, 90–102, 106, 107, 146, 167]. A common strategy is to model the decisive atoms with a method that enables accurate atomic forces and energies at a high computational cost and to model atoms that are less important with a simpler, less accurate description of forces and energies at a smaller numerical cost. The combination of precise and fast simulation models within one simulation leads to a speed-up compared to a simulation where all atoms are described by the precise model by a factor

$$S = \frac{t^{(p)}}{x_{\text{fpm}} t^{(p)} + (1 - x_{\text{fpm}}) t^{(f)} + t^{(d)}}, \quad (6.1)$$

where $t^{(p)}$ and $t^{(f)}$ are the execution times per atom of the precise and fast models, respectively, and $t^{(d)}$ is an overhead that will be specified later. In many simulations the fraction of atoms x_{fpm} that need to be described by the precise model is of the order of 1% or 1‰[91, 107, 167]. Often one can assume that the evaluation time for the fast model¹ as well as the overhead[146] is orders of magnitude smaller than the computational cost of the precise model, $t^{(f)} \approx t^{(d)} \ll t^{(p)}$, such that a potential speedup of orders of magnitude can be expected[146, 167]. The coupling of fast and precise models therefore is very promising to allow for simulations of either much larger system sizes or diminished resource requirements at constant simulation size.

The coupling of two different models in one simulation presents a formidable scientific challenge that has a long history in concurrent multi-scale modelling. Traditionally regions for precise and fast models are identified before the start of a simulation and the challenge is a sound description of the interface between the two regions[90–98].

However, this requires knowledge of expected key atomistic events before the start of the simulation, while for many simulations it is desirable that regions of fast or precise description appear or vanish dynamically as dictated by the trajectories of the atoms. Variable switching between fast and precise models can be achieved with the help of descriptors that quantify the atomic environment of the atoms. For each atom i the descriptor $\mathbf{d}_i$ is evaluated. The fast model is trained as a surrogate model to reproduce the precise model in the vicinity of reference descriptor values $\mathbf{d}^{(0)}$. For example, the reference descriptor $\mathbf{d}^{(0)}$ corresponds to a perfect lattice at equilibrium volume. Then for small deviations of $\mathbf{d}$ from $\mathbf{d}^{(0)}$, the fast potential provides atomic forces and

¹The recently developed ML potentials[37–40] provide – unlike traditional empirical potentials[20–22] – an accuracy comparable to electronic structure calculations[4, 41–43], but remain several orders of magnitude slower than the fast empirical potentials[4, 62, 66].

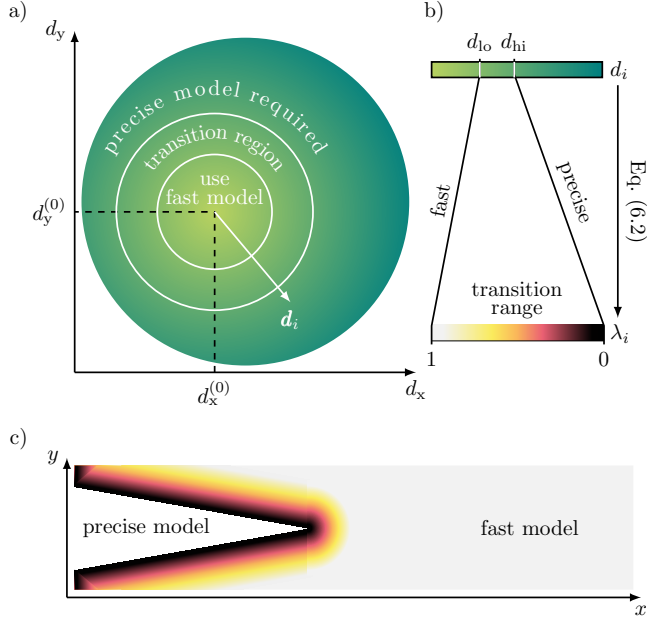


Figure 6.1: The in the a) descriptor space measured deviation $\mathbf{d}_i$ is used to calculate the c) switching parameter λ_i between fast and precise model for all atoms i as visualised in b).

energies that closely match the precise model. At larger deviations of $\mathbf{d}$ from $\mathbf{d}^{(0)}$, the fast surrogate model is insufficient and the precise model is used as illustrated in Fig. 6.1a.

The transition between fast and precise model is achieved by a smooth switching function $\lambda(\mathbf{d})$ (cf. Fig. 6.1b), i.e.,

$$\lambda_i = f^{(\text{trans})}(d_i, d_{lo}, d_{hi}), \quad (6.2)$$

where $f^{(\text{trans})}$ (cf. Eq. (3.37)) is 1 for $d_i \leq d_{lo}$, decays differentiable to 0 until $d_i = d_{hi}$ and stays zero for $d_i > d_{hi}$.

There are different ways to combine fast and precise models. An intuitive approach consists in superimposing the atomic forces of two potentials weighted by a switching parameter λ_i evaluated for atom i as [92, 99, 107]

$$\mathbf{F}_i^{\text{FM}} = \lambda_i \mathbf{F}_i^{(\text{f})} + (1 - \lambda_i) \mathbf{F}_i^{(\text{p})}. \quad (6.3)$$

However, the energy that corresponds to the force is not known and, consequently, force mixing simulations do not conserve energy and require a thermostat to prevent spontaneous heating during a simulation; besides, Eq. (6.3) violates Newton's third law, i.e., it does not conserve the momentum either [106]. A conservative potential may be obtained by mixing the atomic energies directly, [146]

$$E_i^{\text{CAP}} = \lambda_i E_i^{(\text{f})} + (1 - \lambda_i) E_i^{(\text{p})}. \quad (6.4)$$

Thereby, the Hamiltonian is in the absence of external fields given as $H = \sum_i (E_i + T_i)$, where T_i denotes the kinetic energy of an atom i .

The difficulty – caused by superimposing the energies – is that the forces on the atoms, [146]

$$\mathbf{F}_i^{\text{CAP}} = -\nabla_i \sum_k E_k^{\text{CAP}} = \sum_k (-\lambda_k \nabla_i E_k^{(\text{f})} - (1 - \lambda_k) \nabla_i E_k^{(\text{p})} + (\nabla_i \lambda_k) (E_k^{(\text{p})} - E_k^{(\text{f})})), \quad (6.5)$$

may deviate significantly from the forces of the precise reference potential: If the energy of the fast and precise potential differ in the switching region, the gradient of the switching parameter contributes to the force, and a steep gradient of the switching parameter can induce large forces neither present in the precise reference nor in the fast potential (cf. Section D.2.5). Furthermore, $\mathbf{F}_i^{\text{CAP}}$ (Eq. (6.5)) depends on the switching parameters λ_k of neighbouring atoms k . Therefore, a smooth transition is required between both the reference and the fast potential to minimise artefacts in forces from the coupling region.

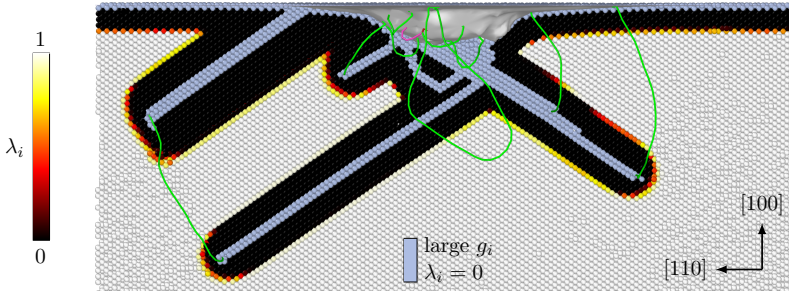


Figure 6.2: Switching parameters λ_i – calculated from g_i via Eqs. (6.2) and (6.6) – and dislocations lines. Not shown are the quickly calculated atoms at the bottom of the simulation box.

A simple and common way to achieve such a transition between precise and fast model is to interpolate the switching parameter dependent on an atom’s position from 1 to 0 between spatial reference positions[92–95], which implies that the force (cf. Eq. (6.5)) caused by the differentiation of the switching function also depends on an atom’s position and, therefore, does not conserve the momentum²[93]. This example illustrates the challenge of the switching-parameter calculation for energy-based approaches, that is addressed in this Chapter.

Therefore, the descriptor $\mathbf{d}$, which we use to calculate the switching parameter (cf. Eq. (6.2)), cannot depend on spatial constants, but only on atomic distances $\mathbf{r}^d$ and non-spatial constants β . In general atomic quantities $\mathbf{g}_k(\{\mathbf{r}^d\}, \beta)$ can be used. $\mathbf{d}_i$ could be calculated from the strain energy to study cracking[107], depend on the chemical species[91], or depend on multiple quantities as visualised in Fig. 6.1a. Note that, d_i – and thus also $\mathbf{g}_k$ – needs to be differentiable as the switching parameter is differentiated for the force calculation (cf. Eq. (6.5)).

The most direct way to calculate the descriptor $\mathbf{d}$ would be $\mathbf{d}_i(\mathbf{g}_i)$, which would result in accurate energies E_i^{AP} (cf. Eq. (6.4)). However, the usage of $\mathbf{d}_i(\mathbf{g}_i)$ neglects the previously discussed dependency of $\mathbf{F}_i^{\text{CAP}}$ on λ_k of neighbouring atoms k that might have a small $\mathbf{d}_k(\mathbf{g}_k)$, which could cause an inaccurate $\mathbf{F}_i^{\text{CAP}}$. To prevent this issue, one can instead use a local average of $\mathbf{g}_k$, i.e.,

$$\mathbf{d}_i = \sum_k \mathbf{g}_k(\{\mathbf{r}^d\}, \beta) \frac{w^d(r_{ik})}{\sum_j w_i^d(r_{ij})} - \mathbf{d}^{(0)}, \quad (6.6)$$

where $w^d(r)$ is a weighting function, that is zero beyond the cutoff distance r_w^{cut} and $\sum_j w^d(r_{ij})$ is a normalisation factor that ensures the independence of d_i from the number of neighbouring atoms within the cutoff distance.

r_w^{cut} should be in the order of magnitude of the cutoff radius of the precise potential, so that atoms with a large $\mathbf{g}_k$ increase $\mathbf{d}_i$ for all atoms k in their force cutoff. Thereby, the neighbouring switching parameters $\lambda_k(d_k) = 0$ can ensure a precise force $\mathbf{F}_i^{\text{CAP}}$ (cf. Eq. (6.5)). Significantly larger averaging radii offer no further advantage as more and more atoms would require the expensive calculation of the precise potential.

The local summation in Eq. (6.6) averages out some fluctuations of $\mathbf{g}_i$, that may be caused by the thermal fluctuation of atoms, like the non-differentiable time average does in Ref. [146]. Note, that one can modify this approach and include fast/precise atoms to speedup a simulation further analogously to Ref. [167] by using a constant time independent switching parameter ($\lambda = 0$ or 1) for a subset of atoms without loosing the conservativity (cf. Section D.2.4).

The partially filled d-band of the transition metal tungsten (W) causes directional, i.e., angular-dependent, bonding[52], which cannot be modelled with central-force potentials[55, 57, 59]. Thus, W benefits from an adaptive-precision description[167] and serves as test material in the following. We construct a conservative adaptive precision potential from the adaptive-precision EAM-ACE W potential of Ref. [167], for which we use a differentiable centro-symmetry parameter (CSP) as g_k (cf. Appendix D). The achievable accuracy of such a conservative EAM-ACE potential is investigated for a nanoindentation snapshot of W taken from a simulation of Ref. [167]. The analysed nanoindentation snapshot is visualised with OVITO[143] in Fig. 6.2 for $r_w^{\text{cut}} = 16 \text{ \AA}$. The visualisation shows that two layers of atoms i have a significant g_i at the surface, whereas nine

²With $\lambda_i(\mathbf{r}_i, \{\mathbf{r}^{\text{ref}}\})$ follows for the force (Eq. (6.5)) $\mathbf{F}_i = (E_i^{(p)} - E_i^{(f)})\nabla_i \lambda_i(\mathbf{r}_i, \{\mathbf{r}^{\text{ref}}\}) + \sum_k (-\lambda_k \nabla_i E_k^{(f)} - (1 - \lambda_k)\nabla_i E_k^{(p)})$, where $(E_i^{(p)} - E_i^{(f)})\nabla_i \lambda_i(\mathbf{r}_i, \{\mathbf{r}^{\text{ref}}\})$ changes the momentum of atom i , but no other atom feels the same force in the opposite direction.

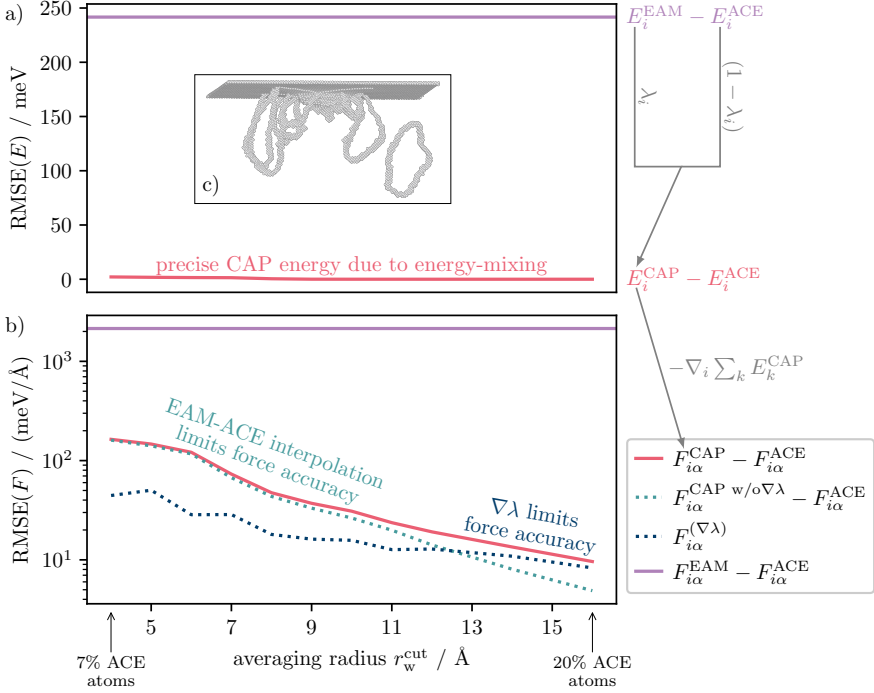


Figure 6.3: RMSE of the a) potential energies E_i and b) force components $F_{i\alpha}$ of an conservative AP potential compared with the precise ACE potential dependent on the radius r_w^{cut} of the averaging region of the descriptor for the in c) shown atoms. The RMSE of EAM is given for comparison.

layers have $\lambda_i = 0$ and the tenth layer transfers with $\lambda_i \in (0, 1)$ to the quickly calculated atoms with $\lambda_i = 1$ in the eleventh layer. This comparison between g_i and λ_i shows that the calculation of the local average according to Eq. (6.6) changes the switching parameter in the force cutoff of atoms with large g_i towards a precise calculation, which is essential for the force accuracy (cf. Section D.2.5). The same effect occurs near dislocation lines, that are identified using the dislocation extraction algorithm[142] of OVITO[143]. Furthermore, slip traces are visible as the gliding dislocations change the neighbourhood of atoms and the CSP-opposite neighbour pairs are not updated (as discussed in Section D.1).

Therefore, to assess the precision of the conservative EAM-ACE potential, we neglect these slip traces by analysing only the by the non-differentiable CSP identified atoms ($\text{CSP}_i > 1.5 \text{ \AA}^2$ [167]). Furthermore, we neglect the fixed atoms of the bottom boundary, as they are calculated efficiently as described in Section D.2.4. Hence, the in Fig. 6.3c shown group of atoms is analysed, i.e., atoms near the top surface or near dislocations – 1.5% of the atoms – which are not well described by the fast EAM potential.

The root mean square error (RMSE) of conservative AP forces and energies compared with the precise ACE potential is shown in Fig. 6.3. The precise ACE-energies are used (cf. Fig. 6.3a) due to the energy interpolation with the switching parameter (cf. Eq. (6.4)). The AP forces are conservatively calculated and improve with the averaging radius r_w^{cut} (cf. Fig. 6.3b) up to a RMSE of 9.6 meV/Å at the largest analysed r_w^{cut} of 16 Å. There are two factors, which decrease the force-precision of the conservative AP potential: 1. force contributions of the fast model and 2. the gradient of the switching parameter (cf. Section D.2.5). We investigate the second factor by calculating the force $F_i^{(\nabla \lambda)}$ caused by the differentiation of the switching parameter. $F_{i\alpha}^{\text{CAP w/o } \nabla \lambda} = F_{i\alpha}^{\text{CAP}} - F_{i\alpha}^{(\nabla \lambda)}$ is a non-conservative AP force independent of the gradient of the switching function and, thus, allows to assess the influence of the fast model on the conservatively calculated AP force. The force contributions of the fast model dominate for small averaging radii (< 13 Å) but become negligible for larger averaging radii (> 13 Å), for which the gradient of the switching function limits the force-accuracy as shown in Fig. 6.3b. As the influence of these

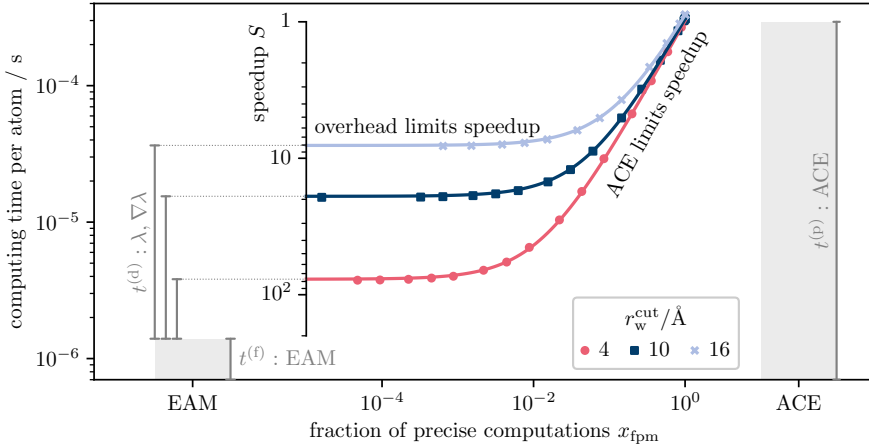


Figure 6.4: Achievable speedup factor S (cf. Eq. (6.1)) and computing time³ per atom dependent on the fraction x_{fpm} of ACE atoms and the averaging radius r_w^{cut} of the AP potential.

two mechanisms decays with the averaging radius r_w^{cut} , one can adjust the force accuracy at the cost of ACE computations dependent on the numerical requirements.

The possible speedup S of a conservative AP potential compared to a complete precise simulation depends on the fraction x_{fpm} of precise computations (cf. Eq. (6.1)). We measure this dependency of S on x_{fpm} for the conservative EAM-ACE W potential by generating more and more vacancy-interstitial pairs – which require the precise potential – in a system of 2×10^6 atoms at room temperature with periodic boundary conditions. Figure 6.4 shows that for large x_{fpm} (near to 1) the speedup S is limited by the required ACE computations. However, a speedup of one or two orders of magnitude is possible when only a small fraction of ACE computations is required (x_{fpm} near to 0). In the latter case, the speedup is limited by the overhead caused mainly by the computation of descriptor (Eq. (6.6)) and switching parameter (Eq. (6.2)). Therefore, a fast descriptor is desirable as the speedup depends on both the descriptor and the averaging radius.

The time integration, e.g., with the velocity-Verlet integrator[131], of an atomic system without external forces described by a conservative interatomic potential results in a *NVE* ensemble, i.e., number of particles, volume and total energy are conserved. We assess this conservativity at room temperature for a system of 20100 W atoms with two surfaces, that is shown in Appendix E. For non-conservative potentials (like LOTF potentials⁴) and force-mixing (cf. Eq. (6.3); here calculated with ML-MIX⁵[106]), the total energy is not conserved, which may lead to numerical instability or the system heating up like shown in Fig. 6.5d and Refs. [100, 215, 216]. Therefore, one should not neglect the gradient of the switching function, as the more precise forces (cf. Fig. 6.3b) come at the cost of the conservativity of the potential. Force-mixing has the further disadvantage that it violates momentum conservation as shown in Fig. 6.5a. In contrast, the total momentum change is numerically zero for the conservative AP W potential, i.e., Newton’s third law applies within numerical precision (cf. Fig. 6.5b). Furthermore, the AP potential conserves the energy within the limit of the finite timestep of the time integrator (cf. Fig. E.1).

We have shown that one can construct a conservative adaptive-precision potential with accurate forces based on the interpolation of potential energies with a switching parameter λ_i that is calculated from a locally averaged descriptor $\mathbf{d}_i$. This new approach requires the usage of a differentiable descriptor $\mathbf{d}_i(\{\mathbf{r}^{\text{d}}\}, \beta)$, that only depends on atomic distances $\mathbf{r}^{\text{d}}$ and non-spatial constants β like a nuclear charge number⁶ as the usage of spatial reference positions changes the

⁴Existing *learn-on-the-fly* (LOTF)[96, 210, 211] potentials are not energy-conserving when the Hamiltonian (if existent at all[210]) is changed during the simulation[212, 213].

⁵ $r_{\text{core}} = 6 \text{ \AA}$ and $r_{\text{blend}} = 4 \text{ \AA}$ are used as parameters for the force mixing like in the Fe-dumbbell example of ML-MIX[214] and all atoms i whose 0.1 ps-time average of CSP_i is larger than $1.5 \text{ \AA}^2$ served as seed atoms for the force mixing. As EAM was incompatible with ML-MIX[214], the communication within the EAM computation was removed like in the `eam/fs/apip` pair style of LAMMPS[1].

⁶ $g_i = \sum_j f^{(\text{trans})}(r_{ij}, r_{\text{low}}, r_{\text{high}})(Z_j - Z_{\text{ref}})^2$ can be used to treat atoms i precisely, when there are neighbouring atoms j , whose nuclear charge number Z_j differs from the one Z_{ref} of the reference environment. $f^{(\text{trans})}$ is given

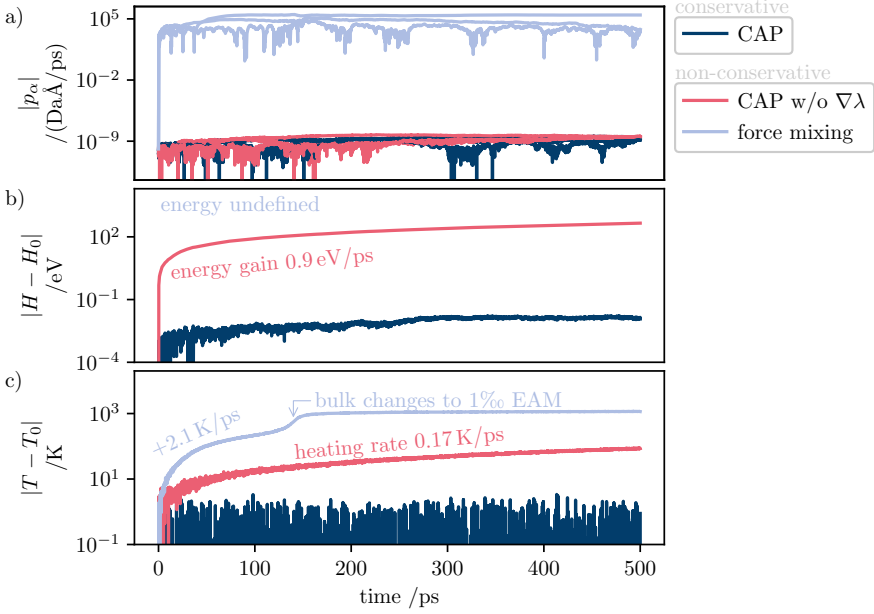


Figure 6.5: a) Momentum, b) energy and c) temperature change in a *NVE* simulation dependent on the combination method of EAM and ACE potential.

total momentum. The differentiable CSP works as descriptor for solids, but one can introduce adaptive-precision also in other materials like fluids[91, 108]. In general, the descriptor needs to be small (cf. Fig. 6.1a) for all atoms, which are well described by the fast surrogate model, so that the expensive precise model can be avoided. Thereby, one can – like demonstrated – use exact potential energies and accurate forces for the atoms of interest, whereby the force accuracy improves when the averaging radius of d_i is increased. Therefore, force-based approaches, in which λ_i interpolates directly the forces, are not required any more in order to obtain accurate forces on specific atoms.

This approach can speed up simulations of microcanonical ensembles, in which the highest precision is required only for a subset of atoms, by a factor of up to 100. Thus, the usage of conservative AP potentials can save a significant amount of energy and pave the way to more resource-responsible computing.

Chapter 7

APIP package of LAMMPS

The Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS)[1] is a modularly designed open-source MD code, that is widely used[14, 68, 74, 77, 109] and whose capabilities have been extended over the years by many community contributions[4, 110–112]. LAMMPS is used for many simulations, which have the potential to benefit from the usage of adaptive-precision, like catalytic reactions[113, 114], surface-growth processes[118, 119], nanoindentations[2, 117] and dynamical crack propagation[115, 116], to name a few. Therefore, all adaptive-precision approaches, which were developed in the present work, have been implemented for LAMMPS.

The code for the computation of the energy- and momentum conserving AP potentials according to Chapter 4, has been combined to a LAMMPS package¹ – the adaptive-precision interatomic potential (APIP) package. The capabilities of this code have been demonstrated by the AP nanoindentation simulations in Chapter 5. This APIP package has been already accepted as part of the LAMMPS distribution by the LAMMPS developers². The APIP package was extended due to the development of conservative adaptive-precision potentials (cf. Chapter 6). This recent development of the APIP package is available at Ref. [217], but not yet a part of the LAMMPS distribution.

The present chapter gives an overview about the APIP package. How to create a new adaptive-precision potential is described in Section 7.1. Section 7.2 concludes with a comparison of the two AP potential approaches, that are implemented in the APIP package, i.e., the approaches of Chapters 4 and 6. Implementation details, a usage example and instructions how to add other detection mechanisms, fast or precise potentials are given in Appendix F.

7.1 Defining a new adaptive-precision potential

Just like their constant precision analogues, both the adaptive-precision EAM and the adaptive-precision ACE commands read the parametrisation of a potential from a file. Thus, one can couple any EAM potential to any ACE potential. In principle, any two potentials can be coupled – however, other potentials must be implemented beforehand as discussed in Section F.2.

So far, optimised fast potentials have been used in the present work to make the fast potential as similar as possible to the precise potential. However, such an optimisation like in Sections B.3.2.1 and 4.3.1 with `atomicrex` might be difficult, e.g., for piecewise defined functions (like in Ref. [147]), that need to remain continuous and differentiable. Therefore, the application of not perfectly optimised fast potentials is discussed in the following.

When a non-optimised fast potential is used, there will be a systematic non-zero energy difference between the fast and the precise potential as both potentials, i.g., were fitted against different reference data. During the course of a simulation some atoms will change their switching parameter, e.g., through a near dislocation in a nanoindentation. With an energy offset between fast and precise potential, these atoms will systematically gain or loose energy due to the changed switching parameter. Dependent on the used potential, this systematic energy change is corrected by the local thermostat (cf. Eq. (4.21)) or causes large forces due to the differentiation of the switching parameter (cf. Eq. (6.5)). Therefore, correcting systematic energy differences between fast and

¹The staggered-grid domain decomposition for LAMMPS, which was used in Chapters 4 and 5, will be published separately in the future, but the APIP package does not depend on the geometry of the spatial domain decomposition[161] and can be used also with the load-balancing via recursive coordinate bisectioning[184], that is implemented in LAMMPS[1].

²LAMMPS is available on <https://github.com/lammps/lammps>.

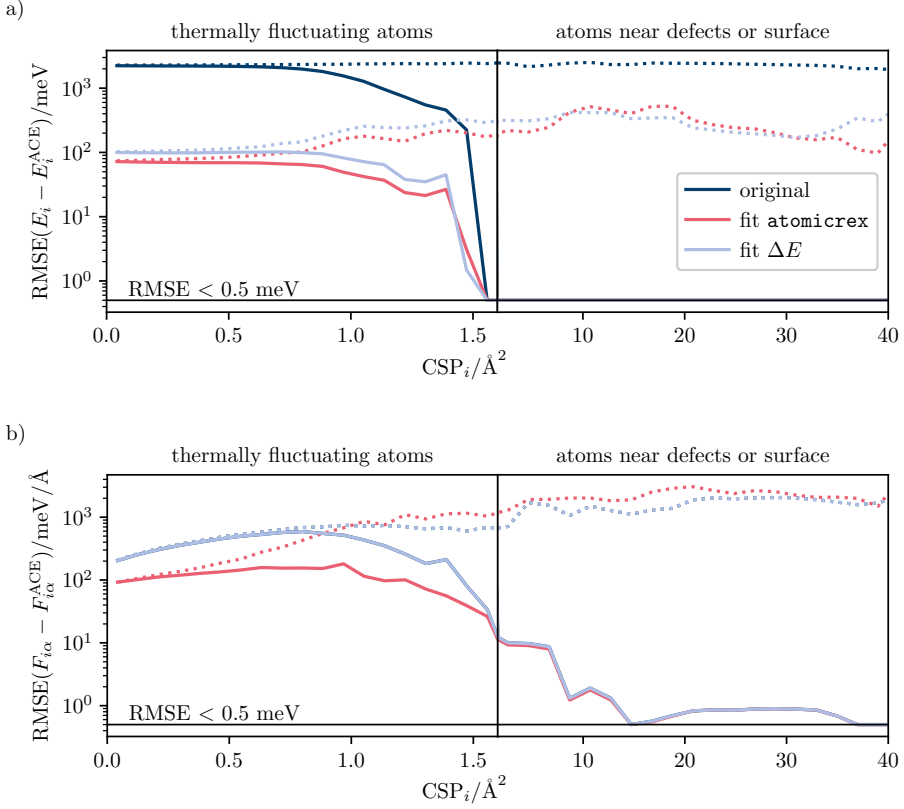


Figure 7.1: The RMSE of a) energies and b) forces of adaptive-precision EAM-ACE W potentials compared with the precise ACE W potential (see Section B.2.2) calculated for the final timestep of the ACE W nanoindentation, that is visualised in Fig. 5.10c. Forces and energies are computed with the input script given in Listing F.1. For comparison, the RMSE of the EAM potentials is shown with dashed lines. The atoms are grouped depended on the CSP and the RMSE is calculated for all groups. The original EAM potential is described in Section B.1.2. Original EAM and 'fit ΔE ' differ only in a constant introduced in the embedding function to correct energy differences between EAM and ACE (cf. Eq. (B.10)). As a constant in the embedding function does not change the computed force, the RMSE of the original EAM potential is identical to the RMSE of the 'fit ΔE ' EAM potential and, thus, not visible in b). 'fit *atomicrex*' is the in Section B.3.2.1 optimised EAM W potential, whose parameters are optimised with *atomicrex*. The atoms at the bottom of the simulation box, whose the CSP was ignored (see line 22 in Listing F.1), are also ignored in this analysis, as they are treated with the fast potential and would be static in a real simulation anyway. This analysis does not contain rescaling effects of a local thermostat, as the example LAMMPS script only calculates the initial energies and forces, but does not include time integration (see line 39 in Listing F.1).

precise potential significantly decreases the influence of the local thermostat or the derivative of the switching parameter.

A potential file, that defines an EAM potential for LAMMPS, contains the embedding function dependent on the local electron density only in a tabulated form³[218, page 2223]. This tabulated form of the embedding function allows to correct systematic energy differences between an EAM and a precise potential.

The energy offset can be measured by computing and comparing the total potential energy of few (< 10) small (< 10000 atoms) reference structures with both the fast and the precise potential. These reference structures should be similar to the real simulation, for which the fast potential is optimised. However, these reference structures should not contain atomistic environments, which should be calculated by the precise potential; e.g., for a nanoindentation, the reference structures should have the same temperature and lattice type like the targeted AP simulation, but the reference structures should neither contain a surface nor defects. The small reference calculation allows to directly calculate the average potential energy difference per atom and adjust the EAM file: One can correct the systematic energy offset between EAM and a precise potential by increasing or decreasing all tabulated values of the embedding function in the EAM file.

This simple optimisation of a fast EAM potential is independent of the functional form of the potential, which may even be unknown, and does not require an additional tool like `atomicrex`. The effect of this optimisations is shown in Fig. 7.1a for a tungsten potential: the RMSE of the potential energy of the adaptive-precision EAM-ACE potential compared to the precise ACE potential for thermally fluctuating atoms, i.e., atoms in the reference environment, is decreased from about 2 eV to less than 102 meV. Hence, using non-optimised fast EAM potentials should be avoided.

The optimisation of the EAM potential with `atomicrex` as described in Section B.3.2.1 further decreases the RMSE of the potential energy of thermally fluctuating atoms to less than 72 meV and additionally decreases the RMSE of the forces from less than 590 meV/Å to less than 181 meV/Å (cf. Fig. 7.1). Thus, a complete optimisation of the fast potential with `atomicrex` is recommended if possible.

The optimised fast potential and the precise potential are combined to an adaptive-precision description via a switching function – either to an energy- and momentum conserving potential according to Chapter 4 or to a conservative AP potential according to Chapter 6. For the former case, the parametrisation of the switching function can be done following the strategy described in Section 4.3.2; for the latter case the strategy is described in Chapter 6 and Section D.1.

7.2 Conclusion

The APIP package of LAMMPS allows to compute energy- and momentum conserving AP potentials according to Chapter 4 and conservative AP potentials according to Chapter 6. The demonstrated capabilities of the APIP package (cf. Chapter 5) can be easily expanded – as demonstrated with the conservative switching function – due to the modular approach used in its implementation: one can simply replace the CSP calculation by another detection mechanism, replace the whole switching function or use another fast or precise potential.

Both approaches – the energy- and momentum conserving one and the conservative approach – have their own advantages. The non-conservative method has the advantage, that it can be better optimised for performance: The calculation of the switching parameter can be done within the force calculation routine, which allows to include the switching parameter calculation in the load-balancer (cf. Section 4.2.4). Thereby, the AP energies and forces are computed with the switching parameters, which were computed in the previous timestep. Using the switching parameters of the previous timestep is possible since the differentiation of this switching parameters is – due to the usage of a local thermostat – not required. However, as a differentiation of the switching parameter λ_i is required for a conservative AP potential, one has to compute λ_i for all atoms i prior to the force-calculation and – in a parallel simulation – communicate these computed λ_i to neighbouring processors before the force calculation. Obviously, this is no disadvantage for a serial simulation. In a parallel simulation, there might be idle time on some processors after the computation of λ_i as the communication of the switching parameters synchronises all processors. However, the energy- and force computation of conservative AP potentials can be balanced by a load balancer in a parallel simulation. Furthermore, most importantly, a conservative AP potential is straightforward to apply as it naturally conserves both momentum and energy – like any constant-precision conservative potential such as EAM or ACE.

³The description of the EAM potential file is also available under https://docs.lammps.org/pair_eam.html.

Currently ACE and EAM are available as fast potentials. ACE is available as precise potential, which already allows the usage of APIP for many materials due to the variety of existing ACE potentials[4, 61–65] and the possibility to create new ones with **pacemaker**[134]. This range of already for APIP available materials could be further extended by implementing more fast and precise potentials, which is possible due to the modular approach of APIP. With all these customisation options, it is straightforward to apply an adaptive-precision interatomic potential (APIP) for other situations, in which the highest precision is required only locally but not globally, like interfaces, cracks or grain boundaries.

Chapter 8

Summary and conclusions

The present work was focussed on the development of adaptive-precision (AP) approaches for the atomistic simulation of microcanonical ensembles. Ideally, the interatomic potential is fast and precise. However, the most precise potentials are machine learning (ML) potentials, that are orders of magnitude slower than less precise traditional empirical potentials. Therefore, with computational resources being limited, there is a conflict of interest between the accuracy of the potential on the one hand and time and length scale of the simulation on the other hand. For simulations, in which the highest accuracy is not required globally, one can solve this conflict of interest with an adaptive-precision approach: the subsystem, that does not need the highest precision, can be simulated with a fast potential, while a precise but expensive potential is only used, where the highest precision is required. However, there were no approaches that conservatively coupled two interatomic potentials since either energy, momentum or both were not conserved (cf. Chapter 2). Hence, such an energy- and momentum conserving approach was developed in the present work to speedup large-scale atomistic simulations. As simulations are dynamic and evolve with time, the distinction between subsystems works dynamically and adaptively in space and time using a switching parameter per atom.

One can, in principle, reconstruct a Hamiltonian from an adaptive-precision force-coupling approach, in particular for pair potentials (cf. Section 2.4). However, as one needs to solve integrals, such a reconstruction of a Hamiltonian is not straightforward and may not be possible at all, in particular for precise modern (machine learning) many-body potentials and an adaptive switching parameter. In contrast, one can conserve the energy with an energy-based coupling approach, which, consequently, was used in the present work. To conserve the momentum and to avoid violating Newton’s third law, one has to be cautious when defining the switching parameter, i.e., one cannot use spatial reference positions other than particles, as this would result in a force on an atom that only depends on the distance to the non-particle reference position, while no other atom feels the same force in the opposite direction. In other words, an interatomic potential, that is not invariant under translation of all atoms, may violate momentum conservation.

The fast potential should be as similar as possible to the precise potential for all cases, in which the fast potential is used. To get a fast potential for the coupling, one can speedup the computation of a ML potential itself by using fewer descriptors, as demonstrated with the creation of fast atomic cluster expansion(ACE[40]) potentials (cf. Chapter 3), or use a fast empirical potential like the embedded atom model(EAM[53]). To make the fast potential as similar as possible to the precise potential in the vicinity of a reference environment, one can use molecular dynamics reference data calculated with the precise potential (for example at room temperature in an fcc lattice like for Cu) to optimise the parameters of the fast potential through the minimisation of a loss function (in this case with `pacemaker`[134] for ACE and `atomicrex`[165] for EAM). The fast potential is tailored for a specific simulation environment. Therefore, one cannot create one single AP potential for a broad range of applications. Instead, one has to optimise a fast potential again when, e.g. a second AP simulation at a (significantly) different temperature is required.

Apart from a fast and a precise potential, one also needs a switching function, that decides which potential is evaluated depending on the atomic environment. In a first coupling attempt, a conservative ACE-ACE coupling was investigated, in which the cluster basis functions of the fast ACE potential were used to measure the deviation of an environment from the reference environment and calculate the switching parameter. Tests with nanoindentation snapshots revealed that this conservative ACE-ACE coupling only prevents unphysical extrapolations, when enough cluster basis functions are used as detection mechanism, which, however also increases the computation cost. The analysis of the force-precision of the adaptive-precision ACE-ACE potential showed the

necessity of a spatial transition region between detected atoms and the remaining atoms, in which the switching parameter slowly changes from 0 to 1 to smoothly interpolate between the forces of the fast and the precise potential.

With the learnings from the ACE-ACE coupling, a more general coupling approach was introduced, that does not depend on any specific potential or detection mechanism (cf. Chapter 4). The atoms of special interest are automatically detected by a customisable detection mechanism, e.g., the CSP for nanoindentations, and simulated with the precise potential while the fast potential is used for the remaining atoms. The energy-based approach ensures precise energies for the detected atoms while the switching function is used to get precise forces through the introduction of the spatial transition zone. This general approach includes the estimation of the load with a calculation specific per-atom load per processor to prevent the load-balancing issues arising from the combination of two potentials of different cost. Moreover, the gradient of the switching parameter can be neglected through the application of the energy- and momentum conserving local thermostat, which distributes the precision decreasing force due to this gradient over more atoms and prevents large deviations of the adaptive-precision force from the force of the precise potential for atoms within the transition zone. The local thermostat also allows the usage of non-differentiable switching functions and, thereby, simplifies the choice of the detection mechanism. Furthermore, one can use the switching parameters of the last timestep, which allows to include the switching parameter calculation into the load-balancing of the force-computation (cf. Section 4.2.4) and prevents idle times in parallel simulations. The latter is not possible for conservative AP potentials, which require the computation of the new switching parameter after the position-calculation prior to the force-calculation.

The amount of computational time that can be saved heavily depends on the number of not required precise calculations and, therefore, on the simulation setup and the detection mechanism for atoms of interest. Hence, the expected speedup cannot be generalised, but can be estimated on-the-fly. Thus, one can detect (and prevent) cases, in which the overhead introduced by the computation of detection mechanism and switching parameter dominates.

The developments were tested with simulations of nanoindentations (cf. Chapter 5) since nanoindentations primarily require the precise treatment of dislocation nucleation and dislocations itself rather than a precise treatment of the whole system. The general adaptive-precision interatomic potential approach was used to obtain adaptive-precision interatomic potentials for copper and tungsten that are tailored for a nanoindentation at room temperature: a fast empirical EAM and a precise ACE potential were coupled with the centro-symmetry parameter (CSP) as detection mechanism for atoms that require the precise potential. The results showed that for Cu, all potentials yield similar dislocation morphologies under the indenter with only small quantitative differences, which confirmed that the EAM potential can describe the metallic bonding in free-electron metal Cu accurately enough. In contrast, markedly different plasticity mechanisms were observed for W in simulations performed with the central-force EAM potential compared to results obtained using the ACE potential, which is able to describe accurately the angular character of bonding in W caused by the half-filled d-band. All ACE-specific mechanisms were reproduced in the adaptive-precision nanoindentation simulations, however, with a significant speedup of almost 30 times compared to the ACE-only simulations. Hence, the adaptive-precision potential overcomes the performance gap between the precise ACE and the fast EAM potential by combining the advantages of both potentials which is beneficial for materials where simple central-force models are not applicable.

The switching function of the EAM-ACE potentials, that was used for the nanoindentation simulations, contains the time averaging of a detection mechanism, i.e., the CSP. Such a time averaging prevents the calculation of the gradient of the switching parameter and, thus, a conservative adaptive-precision potential. Therefore, the descriptor d_i (cf. Chapter 6) uses spatial averaging instead to average out fluctuations. Thereby, the novel energy and momentum conserving and dynamically self adapting coupling approach is further improved to a conservative adaptive-precision potential. As the force precision improves with the averaging radius, force-based approaches, in which an atomic switching parameter λ_i interpolates the forces directly, are not required any more in order to obtain accurate forces on specific atoms. Hence, conservative adaptive-precision potentials can be used regularly in the future – without the currently existing problems caused by the violation of conservativity. Such conservative adaptive-precision potentials can speed up simulations of microcanonical ensembles, in which the highest precision is required only for a subset of atoms, by a factor of up to 100. Thus, the usage of conservative AP potentials can save a significant amount of energy and pave the way to more resource-responsible computing.

The developed APIP package is available for the community as part of LAMMPS¹. Furthermore,

¹The LAMMPS source code is available on <https://github.com/lammps/lammps> and the LAMMPS documen-

the demonstrated capabilities of the APIP package can be easily expanded – as demonstrated with the conservative switching function – due to the modular approach used in its implementation: one can simply replace the CSP calculation by another detection mechanism, replace the whole switching function or use another fast or precise potential. Currently ACE and EAM are available as fast potentials. ACE is available as precise potential, which already allows the usage of APIP for many materials due to the variety of existing ACE potentials[4, 61–65] and the possibility to create new ones with **pacemaker**. This range of available materials for APIP could be further extended by implementing more fast and precise potentials, which is possible due to the modular approach of APIP. With all these customisation options, it is straightforward to apply an adaptive-precision interatomic potential (APIP) for other situations, in which the highest precision is required only locally but not globally, like interfaces, cracks and grain boundaries.

The advantages of APIP have been proven in the nanoindentations and APIP is available for the community. Next steps, that were out of scope for the present work, include a tool for an automated optimisation of the fast potential, e.g., for the energy-offset correction (cf. Section 7.1), or the usage of GPUs to maximise the speedup currently achieved by CPUs. An open question is the choice of the detection mechanisms for fluids or amorphous materials as the CSP can be only used for solids.

One can obtain – independent of the state of matter – conservative adaptive-precision potentials for systems, in which the atoms, that require the highest precision, are known at the start of the simulation. Each atom k could have a constant property g_k , that is zero for atoms, that can be calculated using a fast potential, and positive for the remaining atoms. Such a division of atoms into two groups is known from QM/MM simulations. One can construct a conservative AP potential based on these g_k according to Eqs. (6.2) and (6.6) with $d_{\text{lo}} = 0$ (cf. Chapter 6). However, the precise simulation of a phenomenon in a gas or fluid, that is not restricted to previously known atoms, requires the development of an atomic property g_k that can detect this phenomenon. Thereby, one could simulate other states of matter than solids based on the conservative AP potential developed in the present work.

tation is available at <https://docs.lammps.org/latest/>. The conservative AP potentials have been added to the APIP package and are available at https://github.com/d-immel/lammps_ap/tree/apip_local_average, but have not yet been merged into the official LAMMPS repository. The staggered-grid domain decomposition for LAMMPS, which was used in Chapters 4 and 5, will be published separately in the future, but the APIP package does not depend on the geometry of the spatial domain decomposition[161] and can be used also with the load-balancing via recursive coordinate bisectioning[184], that is implemented in LAMMPS[1].

Appendix A

Dependency on weights of the APA detection mechanism

Adaptive-precision ACE (APA) potentials are presented in Chapter 3. The detection mechanism, that is used to calculate the switching parameter per atom i , is based on the cluster basis functions $B_{i\mathbf{n}^i\mathbf{l}^i}^{(K^f)}$ of the fast ACE potential (cf. Section 3.2.2). The deviation Δ_i^B of a reference environment, to which the fast ACE potential is fitted, is measured per atom and cluster basis function, i.e., per $\left(B_{i\mathbf{n}^i\mathbf{l}^i}^{(K^f)} - B_{i\mathbf{n}^i\mathbf{l}^i}^{(K^f, \text{ref})}\right)^2$, where this squared difference is weighted by the factor $w_{\mathbf{n}^i\mathbf{l}^i}^{(K^f, B)}$ (cf. Eq. (3.33)). This weighting factor can be calculated according to

$$\tilde{w}_{\mathbf{n}^i\mathbf{l}^i}^{(K^f, B)} = \frac{w^B}{(\text{std}(B_{i\mathbf{n}^i\mathbf{l}^i}^{(K^f)}))^2 \Big|_{T=T^{\text{sim}}}}, \quad (\text{A.1})$$

where the variance (squared standard deviation) measures the expected fluctuations of a cluster basis function in a short simulation at the temperature T^{ref} of a system, that has no defective-atoms, and the scaling factor w^B ensures, that $\Delta_i^B = 1$ applies on average in this short reference simulation (cf. Eq. (3.35)).

The standard deviation has the same unit like the data, from which it is calculated. Thus, using the squared standard deviation in Eq. (A.1) is intuitive, since the scaling factor w^B is a real number in this case. In contrast, the standard deviation was used instead of the squared standard deviation in Chapter 3. This appendix compares both approaches.

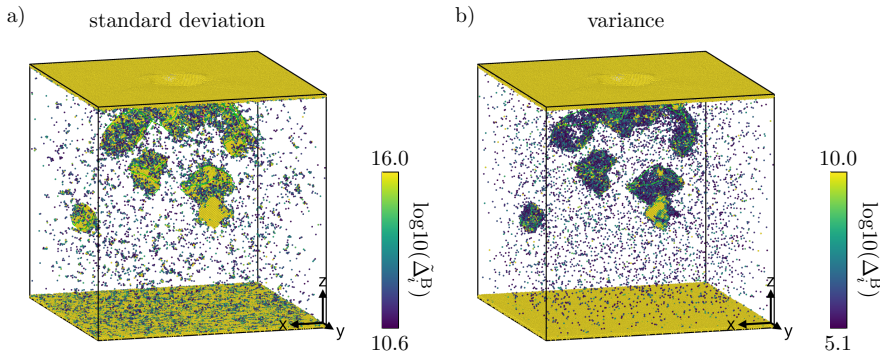


Figure A.1: Visualisations of an at room temperature simulated nanoindentation of a (100) copper surface for an indentation depth of 20 Å. The coloured lines are dislocation lines which are identified by the dislocation extraction algorithm (DXA)[142] of OVITO[143]. Only the 4% of the atoms with the largest deviation $\Delta_i^{B, \text{ACE } 50}$ from the reference environment according to Eq. (3.33) are shown, where the weights used to calculate this deviation are calculated according to a) Eq. (3.34) with the standard deviation and b) according to Eq. (A.1) with the variance. The atoms are coloured according to $\Delta_i^{B, \text{ACE } 50}$.

As test, the deviation from the reference environment is calculated for the two approaches using the ACE 50 potential, which is fitted in Section 3.3.1 for copper. The copper nanoindentation, which is visualised in Fig. 3.6, is used as test system. The deviation $\bar{\Delta}_i^{\text{B,ACE } 50}$ is calculated with the squared standard deviation according to Eq. (A.1), while $\Delta_i^{\text{B,ACE } 50}$ is calculated with the standard deviation according to Eq. (3.34).

Like visualised in Fig. A.1a, one can treat the majority of the thermally fluctuating atoms, which are far away from a dislocation line or the surface, with the fast potential when the standard deviation is used to calculate the weights. In contrast, in Fig. A.1b, where the weights are calculated with the squared standard deviation according to Eq. (A.1), the atoms near dislocations have a deviation from the reference environment that also occurs for many atoms far away from dislocation lines and the surface. Therefore, one cannot distinguish atoms near dislocations and thermally fluctuation atoms, which are far away from any dislocation or surface, with the latter weights definition. Thus, the intuitive approach with the squared standard deviation results in a worse detection of atoms, which are not in a environment similar to the reference environment. Hence, the unintuitive approach with the non-squared standard deviation was used in Chapter 3.

Appendix B

Parametrisations of interatomic potentials

This chapter provides details regarding the interatomic potentials, that were used to simulate nanoindentations of copper and tungsten in Chapter 5. The EAM potentials are described in Section B.1, ACE in Section B.2 and the adaptive-precision EAM-ACE potentials in Section B.3.

B.1 EAM potentials

An embedded atom model (EAM; cf. Section 4.1) potential is given according to Eq. (4.1) by an embedding function ξ , an electron density ζ and a pair potential Φ .

B.1.1 Copper

The EAM copper potential from Ref. [145] is given by the pair potential¹

$$\begin{aligned} \Phi^{\text{Cu}}(x) = & \left(E_1^{\text{Cu}} M^{\text{Cu}}(x, r_0^{(\text{Cu},1)}, \alpha_1^{\text{Cu}}) + E_2^{\text{Cu}} M^{\text{Cu}}(x, r_0^{(\text{Cu},2)}, \alpha_2^{\text{Cu}}) + \delta^{\text{Cu}} \right) \\ & \times \psi^{\text{Cu}} \left(\frac{x - r_{\text{cut}}^{\text{EAM,Cu}}}{h^{\text{Cu}}} \right) - \sum_{n=1}^3 \Theta(r_s^{(\text{Cu},n)} - x) S_n^{\text{Cu}} (r_s^{(\text{Cu},n)} - x)^4, \end{aligned} \quad (\text{B.1})$$

where

$$\psi^{\text{Cu}}(x) = \frac{x^4}{1 + x^4} \Theta(-x) \quad (\text{B.2})$$

and

$$M^{\text{Cu}}(x, r_0, \alpha) = \exp(-2\alpha(x - r_0)) - 2\exp(-\alpha(x - r_0)) \quad (\text{B.3})$$

with the Heaviside step function $\Theta(x) = 1$ for $x \geq 0$, 0 for $x < 0$. The embedding function is

$$\xi^{\text{Cu}}(x) = \begin{cases} \xi^{(\text{Cu},0)} + \frac{1}{2}\xi^{(\text{Cu},2)}(x-1)^2 + \sum_{n=1}^4 q_n^{\text{Cu}}(x-1)^{n+2} & \text{if } x \leq 1 \\ \frac{\xi^{(\text{Cu},0)} + \frac{1}{2}\xi^{(\text{Cu},2)}(x-1)^2 + q_1^{\text{Cu}}(x-1)^3 + Q_1^{\text{Cu}}(x-1)^4}{1 + Q_2^{\text{Cu}}(x-1)^3} & \text{otherwise.} \end{cases} \quad (\text{B.4})$$

The electron density is

$$\begin{aligned} \zeta^{\text{Cu}}(x) = & \left(a^{\text{Cu}} \exp \left(-\beta_1^{\text{Cu}}(x - r_0^{(\text{Cu},3)})^2 \right) + \exp \left(-\beta_2^{\text{Cu}}(x - r_0^{(\text{Cu},4)}) \right) \right) \\ & \psi^{\text{Cu}} \left((x - r_{\text{cut}}^{\text{EAM,Cu}})/h^{\text{Cu}} \right). \end{aligned} \quad (\text{B.5})$$

B.1.2 Tungsten

The EAM potential for W developed in Ref. [148] is given by the embedding function

$$\xi^{\text{W}}(x) = a_1^{\xi} \sqrt{x} + a_2^{\xi} x^2, \quad (\text{B.6})$$

¹The functions, which are used to define potentials in this chapter, just appear in this appendix. Thus they are not listed in the nomenclature.

the pair potential

$$\Phi^W(x) = \sum_{i=1}^{n^\Phi} a_i^\Phi (\delta_i^\Phi - x)^3 \Theta(\delta_i^\Phi - x), \quad (\text{B.7})$$

and the electron charge density

$$\zeta_0^W(x) = \sum_{i=1}^{n^\zeta} a_i^\zeta (\delta_i^\zeta - x)^3 \Theta(\delta_i^\zeta - x), \quad (\text{B.8})$$

where $\Theta(x) = 1$ for $x \geq 0$, 0 for $x < 0$. The electron charge density $\zeta_0^W(r)$ becomes negative for small r and has a maximum at r_{cut}^ζ . Thus, a constant value of the electron charge density is used in the form of

$$\zeta^W(x) = \begin{cases} \zeta_0^W(r_{\text{cut}}^\zeta) & \text{for } r \leq r_{\text{cut}}^\zeta, \\ \zeta_0^W(r) & \text{for } r > r_{\text{cut}}^\zeta \end{cases} \quad (\text{B.9})$$

to ensure a continuous derivative of the electron charge density. Furthermore, the potential is extended to short range using the universal potential of Ref. [219].

B.2 ACE potentials

The atomic cluster expansion (ACE)[40] is described in detail in Section 3.1.

B.2.1 Copper

The ACE copper potential from Ref. [4] was trained against DFT reference data calculated using the PBE functional [220] in the FHI-aims code. The potential has a cutoff of 7.4 Å and uses a basis of 742 many-body basis functions.

B.2.2 Tungsten

The ACE parametrisation for W was trained on a large dataset of DFT data obtained using the FHI-aims all-electron code [221]. The training structures included bulk as well as defective configurations. The potential has a cutoff of 7.9 Å and uses a basis of 929 many-body basis functions. This parametrisation has been extensively tested but remains a preliminary version. The final ACE model for W will be presented in a separate publication.

B.3 Adaptive-precision EAM-ACE potentials

How to couple an EAM potential to an ACE potential is described in Chapter 4 and my paper [146]. The potential energy of an atom i according to an adaptive-precision potential is given by Eq. (4.5), in which a quickly calculated potential energy, i.e., the energy of an EAM potential, is coupled to a precisely calculated potential energy, i.e., to the energy of an ACE potential, by a switching parameter λ_i . The EAM potential should be as similar as possible to the ACE potential in the reference environment, in which the EAM potential is used. Thus, an EAM potential is optimised for the usage in an adaptive-precision potential as described in Section 4.3.1.

B.3.1 Copper

The adaptive-precision copper potential is based on the precise ACE potential, that is described in Section B.2.1. The EAM potential described in Section B.1.1 is used as starting point for the fast potential. The EAM potential is optimised for the usage at room temperature and in combination with the ACE potential in Section B.3.1.1 and coupled to the ACE potential via the switching function given in Section B.3.1.2. This adaptive-precision potential was developed for my paper [146].

B.3.1.1 Optimisation of the EAM potential

The EAM copper potential, that is described in Section B.1.1, was optimised for the usage in an adaptive-precision potential. This optimisation with `atomicrex` is described in Section 4.3.1. The target values and tolerances for the minimised loss function (4.34) are shown in Table 4.2. The fitted parameters are shown in Table B.1.

Table B.1: Optimised parameters of the EAM copper potential compared with the original values of EAM2 of Ref. [145].

parameter	value original	value fit 300K	parameter	value original	value fit 300K
$E_1^{\text{Cu}}/\text{eV}$	2.01458e+02	2.01486e+02	$\delta^{\text{Cu}}/\text{\AA}$	8.62250e-03	2.18016e-02
$E_2^{\text{Cu}}/\text{eV}$	6.59288e-03	6.44057e-03	$F_0^{\text{Cu,Fit}}/\text{eV}$	0.00000e+00	-1.50036e-01
$\xi^{(\text{Cu},0)}/\text{eV}$	-2.30000e+00	-2.30089e+00	$h^{\text{Cu}}/\text{\AA}$	5.00370e-01	5.00370e-01
$\xi^{(\text{Cu},2)}/\text{eV}$	1.40000e+00	1.39912e+00	$q_1^{\text{Cu}}/\text{eV}$	-1.30000e+00	-1.29894e+00
Q_1^{Cu}	4.00000e-01	3.99178e-01	$q_2^{\text{Cu}}/\text{eV}$	-9.00000e-01	-8.99672e-01
Q_2^{Cu}	3.00000e-01	3.00455e-01	$q_3^{\text{Cu}}/\text{eV}$	1.80000e+00	1.79940e+00
$S_1^{\text{Cu}}/\text{eV}\text{\AA}^{-4}$	4.00000e+00	4.00000e+00	$q_4^{\text{Cu}}/\text{eV}$	3.00000e+00	2.99944e+00
$S_2^{\text{Cu}}/\text{eV}\text{\AA}^{-4}$	4.00000e+01	4.00000e+01	$r_0^{(\text{Cu},1)}/\text{\AA}$	8.35910e-01	8.36347e-01
$S_3^{\text{Cu}}/\text{eV}\text{\AA}^{-4}$	1.15000e+03	1.15000e+03	$r_0^{(\text{Cu},2)}/\text{\AA}$	4.46867e+00	4.46690e+00
a^{Cu}	3.80362e+00	3.80541e+00	$r_0^{(\text{Cu},3)}/\text{\AA}$	-2.19885e+00	-2.19834e+00
$\alpha_1^{\text{Cu}}/\text{\AA}^{-1}$	2.97758e+00	2.96972e+00	$r_0^{(\text{Cu},4)}/\text{\AA}$	-2.61984e+02	-2.61984e+02
$\alpha_2^{\text{Cu}}/\text{\AA}^{-1}$	1.54927e+00	1.55510e+00	$r_s^{(\text{Cu},1)}/\text{\AA}$	2.24000e+00	2.24000e+00
$\beta_1^{\text{Cu}}/\text{\AA}^{-2}$	1.73940e-01	1.66589e-01	$r_s^{(\text{Cu},2)}/\text{\AA}$	1.80000e+00	1.80000e+00
$\beta_2^{\text{Cu}}/\text{\AA}^{-1}$	5.35661e+02	5.35661e+02	$r_s^{(\text{Cu},3)}/\text{\AA}$	1.20000e+00	1.20000e+00
$r_{\text{cut}}^{\text{EAM,Cu}}/\text{\AA}$	5.50679e+00	5.50679e+00			

Table B.2: Parameters of the adaptive-precision model and values of the parameter set Hyb1 for copper at room temperature.

parameter	value	parameter	value	parameter	value
N_{buffer}	0 atoms	CSP_{hi}	$3.0 \text{\AA}^2$	$N_{\lambda,\text{avg}}$	110
$N_{\text{CSP,avg}}$	110	$r_{\lambda,\text{lo}}$	$4.0 \text{\AA}$	$\Delta\lambda_{\text{min}}$	$1.0/N_{\lambda,\text{avg}}$
CSP_{lo}	$2.5 \text{\AA}^2$	$r_{\lambda,\text{hi}}$	$12.0 \text{\AA}$	$ \Omega_i $	800 atoms

B.3.1.2 Switching function

The switching parameter λ_i of an atom i is calculated according to Eqs. (4.13) to (4.15) from $\lambda_{0,j}$ of neighbouring atoms j . $\lambda_{0,j}$ is calculated based on the centro-symmetry parameter according to Eq. (4.11) for most of the atoms. To enforce or prevent the evaluation of the precise ACE potential, one can set $\lambda_{0,j}$ to 0 or 1 as discussed in Section 5.1.2.3. $\lambda_{0,j} = 1$ is used for the static atoms at the bottom of the simulation box. Note that a precise hemisphere ($\lambda_{0,j} = 0$) below the indenter is used together with an otherwise fast surface ($\lambda_{0,j} = 1$) only in Chapter 5 but not in Chapter 4. The parameters of the switching function are given in Table B.2.

B.3.2 Tungsten

The adaptive-precision tungsten potential is based on the precise ACE potential, that is described in Section B.2.2. The EAM potential described in Section B.1.2 is used as starting point for the fast potential. The EAM potential is optimised for the usage at 300 K and in combination with the ACE potential in Section B.3.2.1 and coupled to the ACE potential via the switching function given in Section B.3.2.2. This adaptive-precision potential was developed for my paper [167].

B.3.2.1 Optimised EAM potential

The EAM tungsten potential, that was described in Section B.1.2, is optimised following the strategy described in Section 4.3.1. First, we introduce an energy offset $\Delta\xi$ since the equilibrium energies of atoms differ between the EAM and the ACE description. Thus, the embedding energy is given as

$$\xi^{\text{W,Fit}}(\zeta) = \xi^{\text{W}}(\zeta) + \Delta\xi. \quad (\text{B.10})$$

The EAM potential described by Eqs. (B.7), (B.8) and (B.10) is optimised using `atomicrex` [165]. The minimised loss function is given in Eq. (4.34). The target values calculated with ACE and the used tolerances are given in Table B.3. The optimised parameters are shown in Table B.4.

Table B.3: Target values with tolerance used in the loss function Eq. (4.34) for the optimisation of the EAM tungsten potential.

property	target value	tolerance δ^{tol}
scalar properties A_o		
lattice parameter a_0^{bcc}	3.1840 Å	0.001 Å
cohesive energy E_{coh}	-11.1694 eV/atom	0.1 eV/atom
bulk modulus B	279.0494 GPa	1 GPa
elastic constant C_{11}	506.4702 GPa	1 GPa
elastic constant C_{12}	165.3389 GPa	1 GPa
elastic constant C_{44}	135.9924 GPa	1 GPa
properties per structure s		
force F	MD simulation	0.01 eV Å ⁻¹
potential energy E	MD simulation	0.01 eV

Table B.4: Parameters of the optimised EAM tungsten potential for usage at 300 K.

parameter	value	parameter	value	parameter	value
$r_{\text{lo}}^{\text{int}}/\text{Å}$	1.10002200044	a_{14}^{Φ}/eV	-1.40395	a_2^{ρ}/eV	0.486763
$r_{\text{hi}}^{\text{int}}/\text{Å}$	2.10004200084	a_{15}^{Φ}/eV	1.13035	a_3^{ρ}/eV	-0.0425619
Z	74.0	$\delta_1^{\Phi}/\text{Å}$	2.74456	a_4^{ρ}/eV	0.0330214
$\epsilon_0/e^2\text{eV}^{-1}\text{Å}^{-1}$	0.005526349406	$\delta_2^{\Phi}/\text{Å}$	2.74451	$\delta_1^{\rho}/\text{Å}$	2.5
$a_{\text{ZBL}}/\text{Å}$	0.087024572489	$\delta_3^{\Phi}/\text{Å}$	2.28653	$\delta_2^{\rho}/\text{Å}$	3.1
a_1^{Φ}/eV	3663.54	$\delta_4^{\Phi}/\text{Å}$	2.91147	$\delta_3^{\rho}/\text{Å}$	3.5
a_2^{Φ}/eV	-3663.85	$\delta_5^{\Phi}/\text{Å}$	2.96512	$\delta_4^{\rho}/\text{Å}$	4.9
a_3^{Φ}/eV	128.443	$\delta_6^{\Phi}/\text{Å}$	3.07694	a_5^{ξ}/eV	-6.24657
a_4^{Φ}/eV	2.10845	$\delta_7^{\Phi}/\text{Å}$	3.53116	a_6^{ξ}/eV	-0.0836583
a_5^{Φ}/eV	5.10311	$\delta_8^{\Phi}/\text{Å}$	3.85904	$\Delta\xi/\text{eV}$	-2.14333
a_6^{Φ}/eV	-4.06894	$\delta_9^{\Phi}/\text{Å}$	4.10323	b_0^{ρ}/eV	15390.7852469191
a_7^{Φ}/eV	1.25081	$\delta_{10}^{\Phi}/\text{Å}$	4.73354	$b_1^{\rho}/\text{eV Å}^{-1}$	-41436.4792651818
a_8^{Φ}/eV	1.64931	$\delta_{11}^{\Phi}/\text{Å}$	4.8959	$b_2^{\rho}/\text{eV Å}^{-2}$	45168.4307517119
a_9^{Φ}/eV	-1.4248	$\delta_{12}^{\Phi}/\text{Å}$	5.09081	$b_3^{\rho}/\text{eV Å}^{-3}$	-24693.6420961014
a_{10}^{Φ}/eV	-0.761194	$\delta_{13}^{\Phi}/\text{Å}$	5.27739	$b_4^{\rho}/\text{eV Å}^{-4}$	6736.28696027297
a_{11}^{Φ}/eV	1.93524	$\delta_{14}^{\Phi}/\text{Å}$	5.40309	$b_5^{\rho}/\text{eV Å}^{-5}$	-731.611626209140
a_{12}^{Φ}/eV	-0.70151	$\delta_{15}^{\Phi}/\text{Å}$	5.45078	$r_{\text{cut}}^{\rho}/\text{Å}$	2.46396652011306
a_{13}^{Φ}/eV	0.0935973	a_1^{ρ}/eV	-267.394		

Just like for the EAM original potential form Ref. [148], the short-range interaction is given by the Coulomb energy screened by the ZBL screening function ϕ^{ZBL} , namely

$$\Phi^{\text{ZBL}}(r) = \frac{1}{4\pi\epsilon_0} \frac{Z_1 Z_2 e^2}{r} \phi^{\text{ZBL}}(r), \quad (\text{B.11})$$

with the vacuum permittivity $\epsilon_0 \approx 55.26 \times 10^{-4} \text{ e}^2/\text{eV Å}$, the nuclear charge number $Z_{1,2}$ of the respective atom. The ZBL screening function ϕ^{ZBL} is [222]

$$\phi^{\text{ZBL}} = 0.1818e^{-3.2x^{\text{ZBL}}} + 0.5099e^{-0.9423x^{\text{ZBL}}} + 0.2802e^{-0.4029x^{\text{ZBL}}} + 0.02817e^{-0.2016x^{\text{ZBL}}} \quad (\text{B.12})$$

with the reduced distance $x^{\text{ZBL}} = r/a^{\text{ZBL}}$, where

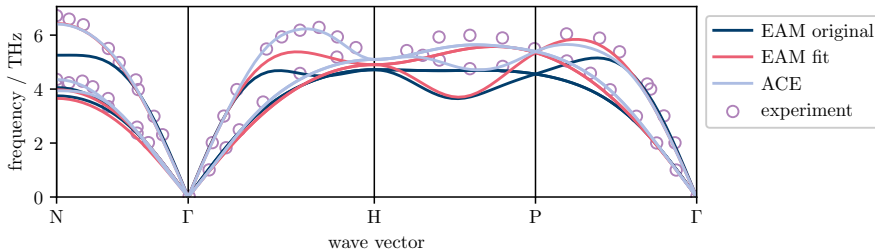
$$a^{\text{ZBL}} = \frac{0.8854a_0}{Z_1^{0.23} + Z_2^{0.23}}, \quad (\text{B.13})$$

where $a_0 = 0.529 \text{ Å}$ is the Bohr radius. For W-W interaction with $Z_1 = Z_2 = 74$ follows $a_{\text{ZBL}} \approx 0.0870 \text{ Å}$. The ZBL potential Φ^{ZBL} and the pair potential Φ are interpolated according to Ref. [223] between $r_{\text{lo}}^{\text{int}}$ and $r_{\text{hi}}^{\text{int}}$ with the polynomial

$$\Phi^{\text{int}}(x) = \sum_{m=0}^5 b_m^{\Phi} x^m \quad (\text{B.14})$$

Table B.5: Properties calculated with tungsten potentials.

	ACE	EAM	
		fit	original
interstitial formation energy / eV			
100-dumbbell / eV	12.47	8.77	12.94
tetrahedral / eV	11.03	8.66	10.43
elastic constant C11 / GPa	506	508	523
elastic constant C12 / GPa	165	162	203
elastic constant C44 / GPa	136	140	160
bulk modulus / GPa	279	277	310
shear modulus 1 / GPa	136	140	160
shear modulus 2 / GPa	171	173	160
poisson ratio	0.25	0.24	0.28
lattice constant / Å	3.1840	3.1837	3.1400
cohesive energy / eV	-11.17	-11.17	-8.90
vacancy formation energy (bcc) / eV	3.36	4.01	3.49
surface energy 111 / J/m ²	3.62	3.55	2.96
surface energy 100 / J/m ²	4.11	3.10	2.72
surface energy 110 / J/m ²	3.51	2.95	2.31

**Figure B.1:** Phonon spectra of tungsten calculated with ASE[136] of the fitted EAM potential, original EAM potential, the ACE potential and experimental values[224] measured by inelastic neutron scattering at room temperature.

so the pair potential and its first two derivatives are continuous at $r_{\text{lo}}^{\text{int}}$ and $r_{\text{hi}}^{\text{int}}$. Thereby we get the pair potential

$$\Phi^{\text{W,Fit}}(r) = \begin{cases} \Phi^{\text{ZBL}}(r) & \text{for } r < r_{\text{lo}}^{\text{int}}, \\ \Phi^{\text{int}}(r) & \text{for } r_{\text{lo}}^{\text{int}} \leq r \leq r_{\text{hi}}^{\text{int}}, \\ \Phi^{\text{W}}(r) & \text{for } r_{\text{hi}}^{\text{int}} < r. \end{cases} \quad (\text{B.15})$$

Hence, we use the optimised EAM potential described by Eqs. (B.9), (B.10) and (B.15) with the parameters given in Table B.4. Basic properties calculated with the optimised EAM potential compared with the original EAM potential and the ACE potential are shown in Table B.5. The elastic constants and related bulk properties correspond to the ACE reference values. Properties not included in the **atomicrex** optimisation, such as surface energies or formation energies of interstitials, are not reproduced. The phonon spectra of the optimised EAM potential and the ACE potential compared in Fig. B.1 are in good agreement.

B.3.2.2 Switching function

The parameters of the switching function (cf. Section 4.2.2.2) are set according to the strategy used in Ref. [146]. The used values are shown in Table B.6. The centro-symmetry parameter uses neighbouring atoms which are on opposite positions of the central-atom and is usually calculated for the atoms of the nearest neighbour shell[76]. The distance in a bcc lattice between the first and second neighbour shell is with $(1 - \sqrt{3}/2) \approx 0.13$ lattice constants relatively small, where the distance between the second and third neighbour shell is $(\sqrt{2} - 1) \approx 0.41$ lattice constants. Therefore, one can easily separate the third from the first two neighbour shells. Thus, we calculate

Table B.6: Parameters of the adaptive-precision model and values of the parameters for tungsten at 300 K.

parameter	value	parameter	value	parameter	value
N_{buffer}	0 atoms	CSP_{hi}	$1.6 \text{ \AA}^2$	$N_{\lambda, \text{avg}}$	110
$N_{\text{CSP, avg}}$	110	$r_{\lambda, \text{lo}}$	$4.0 \text{ \AA}$	$\Delta\lambda_{\text{min}}$	0.01
CSP_{lo}	$1.5 \text{ \AA}^2$	$r_{\lambda, \text{hi}}$	$12.0 \text{ \AA}$	$ \Omega_i $	800 atoms

the CSP with 14 nearest neighbouring atoms which corresponds to the number of neighbours in the first two neighbour shells.

Note that the switching function is not completely defined by the parameters in Table B.6 as one can enforce or prevent the evaluation of the precise ACE potential by setting $\lambda_{0,j}$ to 0 or 1 for a subset of atoms as discussed in Section 5.1.2.3.

Appendix C

Tungsten nanoindentation with optimised EAM potential

The AP and ACE nanoindentations of tungsten are similar, as discussed in Section 5.2.2, while the EAM nanoindentation differs markedly from the other two nanoindentations. To validate whether the nanoindentation simulations with the optimised EAM potential would give results similar to the reference ACE potential, we performed additional simulation runs. Dislocation lines, defects and the shear strain from these simulations are visualised in Fig. C.1 for the same indentation depths as in Figs. 5.9 and 5.10.

For the normalised indentation depth $h/a_c \approx 0.41$ (left column), there are primarily non-screw dislocations detected by the BDA algorithm for the original EAM potential (cf. Fig. 5.9(g)) as well as for the optimised EAM potential (cf. Fig. C.1(e)). Unlike in the AP/ACE simulations, no twins are detected.

At the maximum indentation depth (right column), we observe less pile-up when the optimised EAM potential is used than when the original EAM potential is used (cf. Fig. C.1(d) and Fig. 5.10(a)). Furthermore, twins are detected by BDA in the optimised-EAM nanoindentation (cf. Fig. C.1(d)) while there are none in the original-EAM nanoindentation. However, there are still numerous $\langle 100 \rangle$ dislocation lines below the indenter (cf. Fig. C.1(b)) and the shear strain is higher only below the indenter for both EAM potentials (cf. Fig. C.1(h) and Fig. 5.10(j)), in contrast to the observed $1/2\langle 111 \rangle$ loops observed in the AP/ACE nanoindentation.

In conclusion, the optimised EAM potential predicts a less pronounced pile-up at the indent's rim and the existence of twins, unlike the original EAM potential, which somewhat resembles the AP/ACE results. However, the dislocation mechanisms remain the same as for the original EAM potential. Therefore, the mechanisms observed in the AP/ACE nanoindentation simulations cannot be simply achieved via optimisation of the EAM potential.

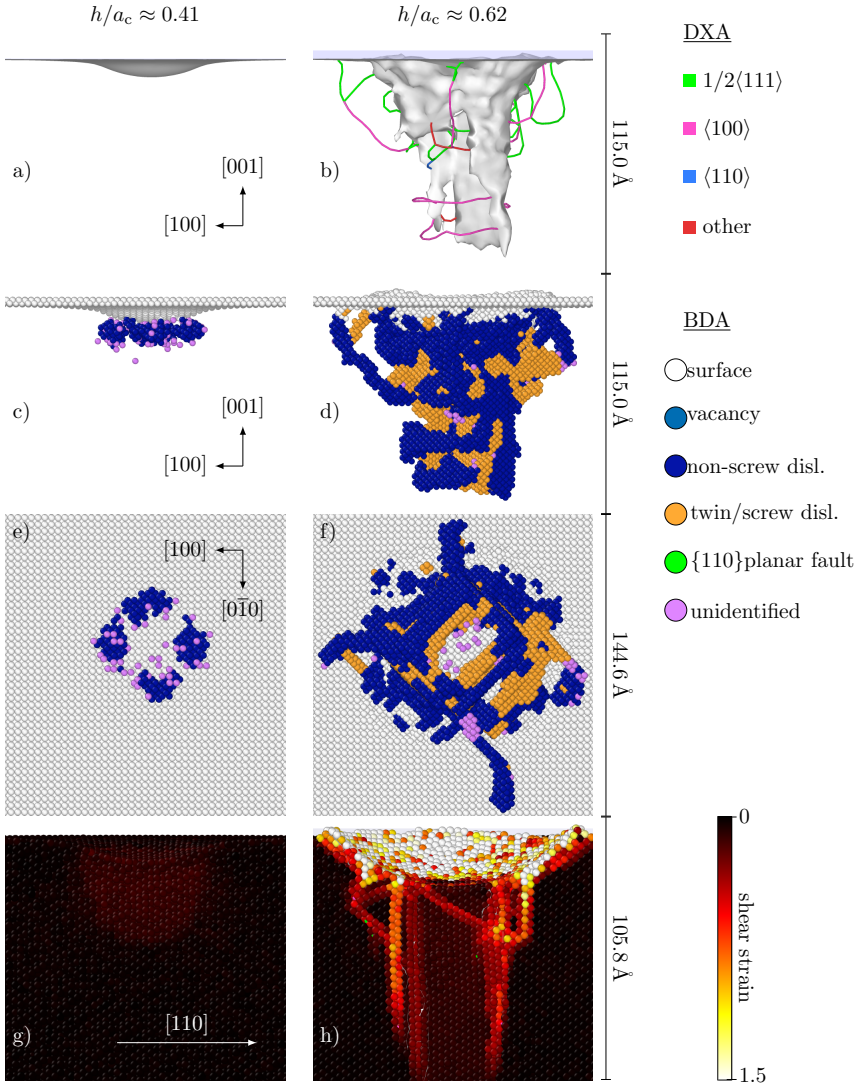


Figure C.1: Tungsten nanoindentation simulated with the optimised EAM potential (Table B.4) visualised with a-b) the dislocation extraction algorithm (DXA) and c-f) the bcc defect analysis (BDA). g-h) A cut in the $(1\bar{1}0)$ plane through the contact point is used to visualise the by OVITO calculated shear strain. Scale bars are given on the right side of each row.

Appendix D

Conservative EAM-ACE adaptive-precision potential

This chapter describes the conservative EAM-ACE potential for tungsten that was developed according to the in Chapter 6 described approach. This potential is based on the non-conservative EAM-ACE W potential that was developed according to Chapter 4 for the simulation of nanoin indentations in Chapter 5. Therefore, the EAM and ACE parametrisations of the potential in Section B.3.2 are reused. However, as the centro-symmetry parameter CSP[76], that was used in Chapter 4 for the switching-parameter calculation, is not differentiable[225], a differentiable CSP is developed in Section D.1. This potential is completely defined using this differentiable CSP as g_i for the descriptor calculation (cf. Eq. (6.6)). Thus, the forces of this conservative AP W potential are calculated in Section D.2 and combined in Section D.2.5.

D.1 Differentiable centro-symmetry parameter

The centro-symmetry parameter (CSP)[76] is used within a non-differentiable switching parameter in Ref. [167]. The CSP uses pairs of neighbours in exact opposite directions of atom i to detect whether i is near defects or surfaces. In LAMMPS[1], these neighbour pairs are recalculated every timestep for the calculation of CSP_i [225]. Therefore, CSP_i is not differentiable[225] and needs to be modified to be used for the calculation of a differentiable descriptor. Thus, we detect these neighbour pairs initially and do not update them afterwards for the calculation of $\text{CSP}_i^{\text{list}}$, i.e.,

$$\text{CSP}_i^{\text{list}} = \sum_j \frac{\Theta_{ij}^{\text{in}}}{2} \left| \mathbf{r}_{i,j} + \mathbf{r}_{i,\Theta_{ij}^{\text{ngh}}} \right|^2, \quad (\text{D.1})$$

where Θ_{ij}^{in} is 1 if j is an atom in the pair-list of i and 0 otherwise and Θ_{ij}^{ngh} denotes the atom, that was initially on the opposite side of i than j . Thereby, $\text{CSP}_i^{\text{list}}$ is differentiable and used as g_i in the descriptor calculation (cf. Eq. (6.6)). The CSP fluctuates around zero for atoms at a finite temperature in a the defect-free lattice, for which the fast EAM potential was optimised[167]. Therefore, the reference descriptor $d^{(0)}$ (cf. Eq. (6.6)) is zero for this potential. For simplicity, we use $w^{\text{d}}(r) = f^{\text{trans}}(r, r_{\text{w}}^{\text{cut}} - 1 \text{ \AA}, r_{\text{w}}^{\text{cut}})$ as weighting function for the averaging within the descriptor (cf. Eq. (6.6)). Thereby, thermal fluctuations of atoms near the averaging cutoff $r_{\text{w}}^{\text{cut}}$ change the averaged descriptor smoothly rather than abruptly.

Finally, the switching parameter λ_i is calculated according to Eq. (6.2) from d_i . $\text{CSP}_i \leq 1.5 \text{ \AA}^2$ is expected in body-centred cubic tungsten at room temperature due to thermal fluctuations of atoms[167]. By using $d_{\text{hi}} = 1.5 \text{ \AA}^2$ (cf. Eq. (6.2)) we ensure that only atoms with a higher than for thermal fluctuations expected CSP cause the evaluation of the precise ACE potential. d_{lo} (cf. Eq. (6.2)) is set just high enough so that all atoms in defect-free bulk have $\lambda_i = 1$, i.e., require only the fast EAM computation.

Atoms need a unique identifier due to the pair-list (cf. Eq. (D.1)). Thus, this exemplary potential is not invariant under permutation of atoms any more, i.e., the differentiable CSP of increases for some atoms when two atoms swap their position in a lattice. Such permutations occur, when a dislocation glides through a system (cf. Section 5.2.2). Therefore, slip traces are visible in Fig. D.1. Note, that the general averaging approach (Eqs. (6.2) and (6.6)) is invariant under permutation of atoms if $\mathbf{g}_i$ is invariant under permutation of atom indices.

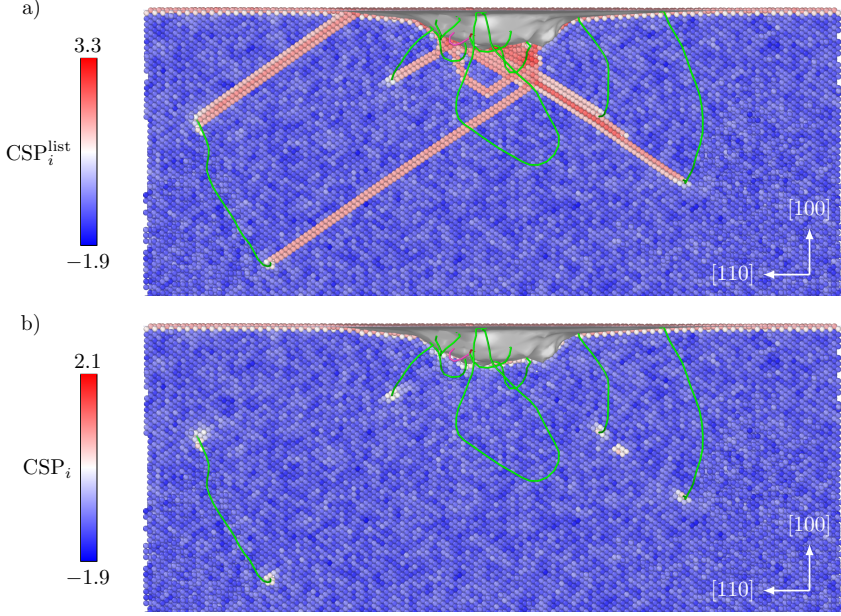


Figure D.1: Comparison between a) differentiable and b) non-differentiable centro-symmetry parameter for a tungsten nanoindentation snapshot. Furthermore, dislocation lines identified by OVITO are shown. Not shown are the quickly calculated atoms at the bottom of the simulation box.

D.2 Force calculation

The force of a conservative adaptive-precision potential is calculated according to Eq. (6.5). This force calculation requires the calculation of the gradient of three components: 1. of the fast potential energy, 2. of the precise potential energy and 3. of the switching parameter λ . These gradients are calculated in the following subsections.

D.2.1 Embedded atom model

The energy E_i^{EAM} of an atom i described with EAM is given according to Eq. (4.1) by an embedding function ξ , a pair potential Φ and an electron charge density ζ . The contributions to Eq. (6.5) caused by the differentiation of a fast EAM potential are

$$\mathbf{F}_i^{\text{CAP,EAM}} = \sum_k (-\lambda_k \nabla_i E_k^{\text{EAM}}) = \sum_k \left(-\lambda_k \left(\frac{\partial \xi}{\partial \zeta} \Big|_{\zeta=\zeta_k} \sum_{j \neq k} \frac{\partial \zeta}{\partial r} \Big|_{r=r_{kj}} \hat{\mathbf{r}}_{kj} (\delta_{ki} - \delta_{ji}) + \frac{1}{2} \sum_{j \neq k} \frac{\partial \Phi}{\partial r} \Big|_{r=r_{kj}} \hat{\mathbf{r}}_{kj} (\delta_{ki} - \delta_{ji}) \right) \right), \quad (\text{D.2})$$

where $\hat{\mathbf{r}} = \mathbf{r}/r$ is a unit vector. Resolving the sums over the Kronecker deltas and rearranging the terms gives

$$\mathbf{F}_i^{\text{CAP,EAM}} = \sum_k \left(\lambda_i \left(\frac{\partial \xi}{\partial \zeta} \Big|_{\zeta=\zeta_i} \frac{\partial \zeta}{\partial r} \Big|_{r=r_{ki}} + \frac{1}{2} \frac{\partial \Phi}{\partial r} \Big|_{r=r_{ki}} \right) \hat{\mathbf{r}}_{ki} - \lambda_k \left(\frac{\partial \xi}{\partial \zeta} \Big|_{\zeta=\zeta_k} \frac{\partial \zeta}{\partial r} \Big|_{r=r_{ik}} + \frac{1}{2} \frac{\partial \Phi}{\partial r} \Big|_{r=r_{ik}} \right) \hat{\mathbf{r}}_{ik} \right), \quad (\text{D.3})$$

where the identification of the EAM pair force contribution $\mathbf{f}_{ki}^{\text{EAM}}$ (cf. Eq. (4.4)) of the constant precision EAM potential allows to write the force as

$$\mathbf{F}_i^{\text{CAP,EAM}} = \sum_k (\lambda_i \mathbf{f}_{ki}^{\text{EAM}} - \lambda_k \mathbf{f}_{ik}^{\text{EAM}}). \quad (\text{D.4})$$

The force on an atom i according to a constant precision EAM potential can be directly obtained by assuming $\lambda = 1$ for all atoms in Eqs. (D.2) and (D.4), i.e.,

$$\mathbf{F}_i^{\text{EAM}} = -\nabla_i \sum_k E_k^{\text{EAM}} = \mathbf{F}_i^{\text{CAP,EAM}} \Big|_{\lambda=1} = \sum_k (\mathbf{f}_{ki}^{\text{EAM}} - \mathbf{f}_{ik}^{\text{EAM}}). \quad (\text{D.5})$$

The EAM pair force contributions $\mathbf{f}_{ki}^{\text{EAM}}$ remain valid in the adaptive-precision case (cf. Eq. (D.4)), as they are independent of the switching parameter.

D.2.2 Atomic cluster expansion

The force contributions $\mathbf{f}_{ki}^{\text{ACE}}$ of an ACE[40] potential are given in Eq. (42) of Ref. [132] and Eq. (16) of Ref. [4]. Comparing Eqs. (D.4) and (D.5) shows that one only has to weight the constant-precision force contributions by a switching parameter to get an adaptive-precision force. Thus, the contributions to the adaptive-precision force (cf. Eq. (6.5)) by the differentiation of a precise ACE potential are

$$\mathbf{F}_i^{\text{CAP,ACE}} = -\sum_k (1 - \lambda_k) \nabla_i E_k^{\text{ACE}} = \sum_k ((1 - \lambda_i) \mathbf{f}_{ki}^{\text{ACE}} - (1 - \lambda_k) \mathbf{f}_{ik}^{\text{ACE}}). \quad (\text{D.6})$$

D.2.3 Switching parameter

In general, the gradient of the switching parameter contributes to the adaptive-precision force (Eq. (6.5)) in form of

$$\mathbf{F}_i^{(\nabla\lambda)} = \sum_k (\nabla_i \lambda_k) (E_k^{(\text{p})} - E_k^{(\text{f})}). \quad (\text{D.7})$$

For the definition of the switching parameter with a locally averaged detection mechanism (cf. Eqs. (6.2) and (6.6)) follows

$$\mathbf{F}_i^{(\nabla\lambda)} = \sum_k (E_k^{(\text{p})} - E_k^{(\text{f})}) \frac{\partial \lambda_k}{\partial d_k} \nabla_i d_k = \sum_k \tilde{E}_k \left(\sum_j (\nabla_i g_j) w^{\text{d}}(r_{kj}) + \sum_j (g_j - d_k) (\nabla_i w^{\text{d}}(r_{kj})) \right), \quad (\text{D.8})$$

where the abbreviation

$$\tilde{E}_k = \frac{E_k^{(\text{p})} - E_k^{(\text{f})}}{\sum_j w^{\text{d}}(r_{kj})} \frac{\partial \lambda_k}{\partial d_k} \quad (\text{D.9})$$

is used. The force $\mathbf{F}_i^{(\nabla\lambda)}$ requires the calculation of $\nabla_i g_j$ and $\nabla_i w^{\text{d}}(r_{kj})$ (cf. Eq. (D.8)). As the former differentiation depends on the atomic quantities g_j , we start with the latter differentiation by calculating

$$\nabla_i w^{\text{d}}(r_{kj}) = \frac{\partial w^{\text{d}}}{\partial r} \Big|_{r=r_{kj}} \hat{\mathbf{r}}_{kj} (\delta_{ik} - \delta_{ij}) \quad (\text{D.10})$$

and resolving the sums over the Kronecker deltas to

$$\begin{aligned} \mathbf{F}_i^{(\nabla\lambda, \text{w})} = \sum_k \tilde{E}_k \sum_j (g_j - d_k) \nabla_i w^{\text{d}}(r_{kj}) &= \sum_j \left(\tilde{E}_i (g_j - d_i) \frac{\partial w^{\text{d}}}{\partial r} \Big|_{r=r_{ij}} \hat{\mathbf{r}}_{ij} \right. \\ &\quad \left. - \tilde{E}_j (g_i - d_j) \frac{\partial w^{\text{d}}}{\partial r} \Big|_{r=r_{ji}} \hat{\mathbf{r}}_{ji} \right). \end{aligned} \quad (\text{D.11})$$

With the force contribution

$$\mathbf{f}_{ij}^{(\nabla\lambda, \text{w})} = \frac{E_i^{(\text{f})} - E_i^{(\text{p})}}{\sum_j w^{\text{d}}(r_{ij})} \frac{\partial \lambda_i}{\partial d_i} (g_j - d_i) \frac{\partial w^{\text{d}}}{\partial r} \Big|_{r=r_{ij}} \hat{\mathbf{r}}_{ij} \quad (\text{D.12})$$

follows

$$\mathbf{F}_i^{(\nabla\lambda, \text{w})} = \sum_j (\mathbf{f}_{ji}^{(\nabla\lambda, \text{w})} - \mathbf{f}_{ij}^{(\nabla\lambda, \text{w})}). \quad (\text{D.13})$$

D.2.4 Centro-symmetry parameter

A modification of the centro-symmetry parameter[76] is used as atomic quantity g_i for the calculation of the descriptor in Chapter 6. This differentiable CSP is given by Eq. (D.1). For the $\nabla_i g_j$ term in Eq. (D.8) follows with $\text{CSP}_j^{\text{list}}$ (Eq. (D.1)) as atomic quantity g_j

$$\nabla_i \text{CSP}_j^{\text{list}} = \sum_l \frac{\Theta_{jl}^{\text{in}}}{2} \nabla_i \sum_{\alpha=0}^2 \left(2r_{j\alpha} - r_{l\alpha} - r_{\Theta_{jl}^{\text{ngb}} \alpha} \right)^2 = \sum_l \Theta_{jl}^{\text{in}} \left(\mathbf{r}_{j,l} + \mathbf{r}_{j,\Theta_{jl}^{\text{ngb}}} \right) \left(2\delta_{ij} - \delta_{il} - \delta_{i,\Theta_{jl}^{\text{ngb}}} \right), \quad (\text{D.14})$$

where the contribution of the last two Kronecker deltas are identical as $\Theta_{jl}^{\text{in}} = \Theta_{j\Theta_{jl}^{\text{ngb}}}^{\text{in}}$ applies by definition of Θ_{jl}^{ngb} and the sum over all atoms l is calculated. Thus, the derivative of the differentiable CSP is given by

$$\nabla_i \text{CSP}_j^{\text{list}} = 2 \sum_l \Theta_{jl}^{\text{in}} \left(\mathbf{r}_{j,l} + \mathbf{r}_{j,\Theta_{jl}^{\text{ngb}}} \right) (\delta_{ij} - \delta_{il}). \quad (\text{D.15})$$

Therefore, one can calculate the force caused by the differentiation of the CSP in Eq. (D.8) by resolving the sums over the Kronecker deltas, i.e.,

$$\begin{aligned} \mathbf{F}_i^{(\nabla\lambda, \text{CSP})} &= \sum_k \tilde{E}_k \sum_j \left(\nabla_i \text{CSP}_j^{\text{list}} \right) w^{\text{d}}(r_{kj}) = \sum_j 2 \left(\sum_k \tilde{E}_k w^{\text{d}}(r_{ki}) \right) \Theta_{ij}^{\text{in}} \left(\mathbf{r}_{i,j} + \mathbf{r}_{i,\Theta_{ij}^{\text{ngb}}} \right) \\ &\quad - \sum_j 2 \left(\sum_k \tilde{E}_k w^{\text{d}}(r_{kj}) \right) \Theta_{ji}^{\text{in}} \left(\mathbf{r}_{j,i} + \mathbf{r}_{j,\Theta_{ji}^{\text{ngb}}} \right), \end{aligned} \quad (\text{D.16})$$

and identifying the force contribution

$$\mathbf{f}_{ij}^{(\nabla\lambda, \text{CSP})} = \Theta_{ij}^{\text{in}} \left(\sum_k \frac{E_k^{(\text{f})} - E_k^{(\text{p})}}{\sum_j w^{\text{d}}(r_{kj})} \frac{\partial \lambda_k}{\partial d_k} w^{\text{d}}(r_{ki}) \right) 2 \left(\mathbf{r}_{i,j} + \mathbf{r}_{i,\Theta_{ij}^{\text{ngb}}} \right). \quad (\text{D.17})$$

Thereby, the force can be simplified to

$$\mathbf{F}_i^{(\nabla\lambda, \text{CSP})} = \sum_j \left(\mathbf{f}_{ji}^{(\nabla\lambda, \text{CSP})} - \mathbf{f}_{ij}^{(\nabla\lambda, \text{CSP})} \right). \quad (\text{D.18})$$

The force $\mathbf{F}_i^{(\nabla\lambda)}$ (cf. Eq. (D.7)) caused by the differentiation of the switching parameter requires the differentiation of the weighting function w^{d} (cf. Eqs. (D.10) to (D.13)) and the atomic quantity g_i (cf. Eqs. (D.14) to (D.18)). Thus, $\mathbf{F}_i^{(\nabla\lambda)} = \mathbf{F}_i^{(\nabla\lambda, w)} + \mathbf{F}_i^{(\nabla\lambda, \text{CSP})}$ is calculated from Eqs. (D.13) and (D.18) to

$$\mathbf{F}_i^{(\nabla\lambda)} = \sum_j \left(\mathbf{f}_{ji}^{(\nabla\lambda)} - \mathbf{f}_{ij}^{(\nabla\lambda)} \right), \quad (\text{D.19})$$

where the force contribution $\mathbf{f}_{ij}^{(\nabla\lambda)} = \mathbf{f}_{ji}^{(\nabla\lambda, \text{CSP})} + \mathbf{f}_{ji}^{(\nabla\lambda, w)}$ follows from Eqs. (D.12) and (D.17), i.e.,

$$\mathbf{f}_{ij}^{(\nabla\lambda)} = \Theta_{ij}^{\text{in}} \left(\sum_k \frac{E_k^{(\text{f})} - E_k^{(\text{p})}}{\sum_j w^{\text{d}}(r_{kj})} \frac{\partial \lambda_k}{\partial d_k} w^{\text{d}}(r_{ki}) \right) 2 \left(\mathbf{r}_{i,j} + \mathbf{r}_{i,\Theta_{ij}^{\text{ngb}}} \right) + \frac{E_i^{(\text{f})} - E_i^{(\text{p})}}{\sum_j w^{\text{d}}(r_{ij})} \frac{\partial \lambda_i}{\partial d_i} (g_j - d_i) \frac{\partial w^{\text{d}}}{\partial r} \bigg|_{r=r_{ij}} \hat{\mathbf{r}}_{ij}. \quad (\text{D.20})$$

One can introduce atoms i with a constant switching parameter as described in Ref. [167]. Thereby, one can enforce the fast or the precise calculation for a subset of atoms for a whole simulation. Equation (D.20) remains valid in this case, where $\frac{\partial \lambda_i}{\partial d_i} = 0$ applies for the corresponding atoms as λ_i is constant and not calculated from d_i (cf. Eq. (6.2)).

D.2.5 Total force

The force on an atom described by a conservative EAM-ACE potential according to Chapter 6 with the differentiable CSP (cf. Section D.1) as g_i in the detection mechanism of the potential (cf. Eq. (6.6)) is given by Eq. (6.5). This force $\mathbf{F}_i^{\text{CAP}} = \mathbf{F}_i^{\text{CAP, EAM}} + \mathbf{F}_i^{\text{CAP, ACE}} + \mathbf{F}_i^{(\nabla\lambda)}$ was calculated in three parts in the previous sections (cf. Eqs. (D.4), (D.6) and (D.19)). Combining these three parts gives

$$\mathbf{F}_i^{\text{CAP}} = \sum_k \left(\mathbf{f}_{ki}^{\text{CAP}} - \mathbf{f}_{ik}^{\text{CAP}} \right), \quad (\text{D.21})$$

where the pair force contributions $\mathbf{f}_{ik}^{\text{CAP}}$ of the conservative adaptive-precision potential,

$$\mathbf{f}_{ki}^{\text{CAP}} = \lambda_i \mathbf{f}_{ki}^{(\text{f})} + (1 - \lambda_i) \mathbf{f}_{ki}^{(\text{p})} + \mathbf{f}_{ki}^{(\nabla\lambda)}, \quad (\text{D.22})$$

are given by Eqs. (3.19), (4.4) and (D.20) and correspond to the differentiation of the fast potential, the precise potential and the switching function, respectively. Note, that $\mathbf{f}_{ki}^{(\text{f})}$ and $\mathbf{f}_{ki}^{(\text{p})}$ can be calculated only from the fast and precise potential, respectively.

Equations (D.22) and (6.4), i.e., the equations for the calculation of the adaptive-precision energy and force, have the same form, apart from the additional force contribution $\mathbf{f}_{ki}^{(\nabla\lambda)}$ caused by the differentiation of the switching parameters. Furthermore, Eqs. (D.21) and (D.22) show the dependence of $\mathbf{f}_i^{\text{CAP}}$ on the switching parameters λ_k of all neighbouring atoms k within the cutoff. Thus, although $\lambda_i = 0$ implies a precise potential energy E_i^{CAP} (cf. Eq. (6.4)), it does not imply an accurate force $\mathbf{F}_i^{\text{CAP}}$. Furthermore, it is important to remember that although two models compute similar forces $\mathbf{F}_i$, their force contributions $\mathbf{f}_{ki}$ may not be correlated any more as shown in Fig. 4.1. Thus, a slow spatial change of the switching parameter from 1 to 0 over few neighbouring atoms is desirable to smoothly transfer the fast force model into the precise one.

Appendix E

Energy conservation of conservative potentials

The conservation of energy and momentum in a *NVE* simulation was investigated in Chapter 6 for a conservative adaptive-precision W potential. The energy change is – in contrast to the momentum change – not numerically zero. Thus, the observed energy change is further investigated in this Appendix.

Figure E.1 shows the energy changes measured in reference *NVE* simulations of the same system using constant precision potentials. The total energy H of the conservative AP potential fluctuates systematically with a small amplitude on a short timescale and drifts randomly during the whole simulation to a negligible extend (cf. Fig. E.1b). Both, fluctuation and drift, are expected due to the finite timestep (1 fs) of the time integrator [226] (see also Refs. [216, 227]). The random energy fluctuation in the conservative AP simulation is in the same order as found in the reference simulation with the EAM potential, that is used as fast potential in the AP potential (cf. Fig. E.1b).

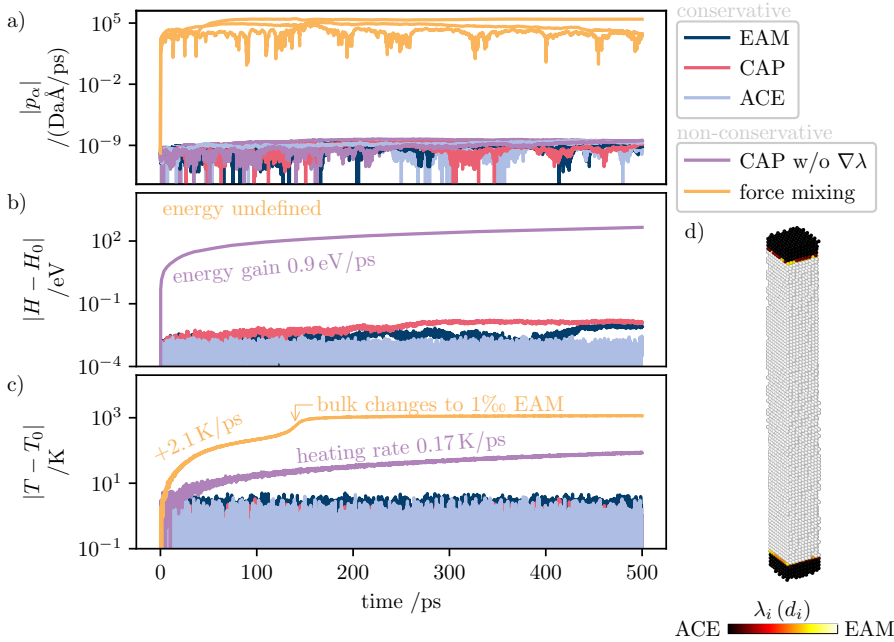


Figure E.1: a) Total momentum, b) energy change and c) temperature change in a *NVE* simulation of the in d) shown system with conservative potentials compared to non-conservative approaches (cf. Chapter 6). d) System of 20100 W atoms at room temperature with two surfaces visualised with OVITO [143]. The atoms are coloured according to their switching parameter λ_i of the conservative adaptive-precision potential (cf. Eq. (6.2)).

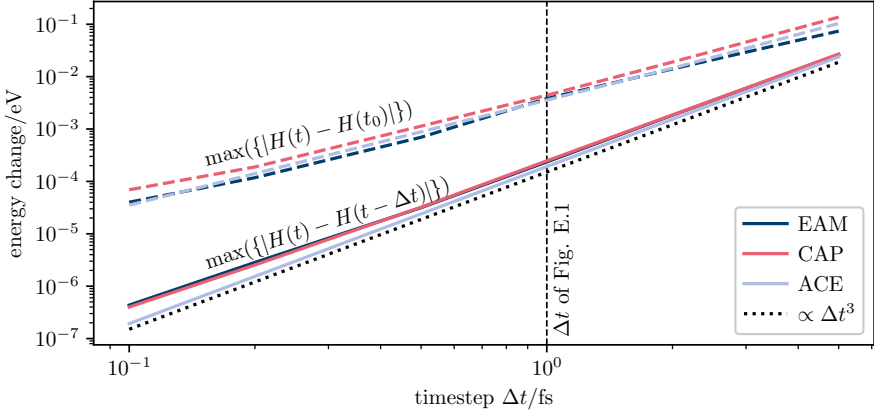


Figure E.2: Timestep dependence of the energy change between two consecutive timesteps and total energy change analysed in a 10 ps simulation starting after the simulations shown in Fig. E.1. An energy change proportional to Δt^3 is shown for comparison.

To investigate the energy change further, we continued the simulation with the conservative potentials with different timesteps Δt of the velocity-Verlet integrator. Figure E.2 shows a dependence of the energy change between two consecutive timesteps on Δt^3 , i.e., the energy change vanishes with the timestep – like expected from Ref. [228]. Furthermore, the energy change between consecutive timesteps is similar for EAM and AP, while the energy change of ACE is smaller. As the majority of the system uses the quickly computed EAM potential energy (cf. Fig. E.1d), it is expected that the energy change of EAM dominates in AP compared to the ACE energy change. Thus, the AP potential conserves the energy within the limits set through the finite timestep.

Appendix F

APIP package of LAMMPS

The adaptive-precision interatomic potential (APIP) package for LAMMPS is presented in Chapter 7. The present appendix provides some implementation details. Note that the full package documentation¹ is only contained in the LAMMPS documentation[156].

The APIP package is – like LAMMPS – designed modularly. The components of the APIP package and a usage example are given in Section F.1. The modularity allows to add other detection mechanisms, fast or precise potentials as explained in Section F.2.

F.1 New LAMMPS commands

LAMMPS is a modularly designed software. The user provides a LAMMPS input file, which contains commands and is read line by line by LAMMPS. First, one has to initialise the atoms and the simulation box. LAMMPS provides a time-stepping algorithm for a MD simulation. This time-stepping algorithm provides multiple points during a timestep, at which modules are called: a `pair_style` provides an interatomic potential, such as `pace` for an ACE potential or `eam/fs` for an EAM potential. A `fix` changes properties of the system, such as `nve` for the update of positions and velocities based on the stored atomic forces. Users can define multiple fixes to tailor a simulation to their own requirements. The reader is referred to the LAMMPS documentation[156] for further details.

The APIP package provides new pair styles and fixes for the usage of AP potentials. Currently, the APIP package contains the following commands:

- `fix atom_weight/apip` computes the computational weight per particle as input for the load balancer.
- `fix lambda_thermostat/apip` applies the local thermostat according to Section 4.2.3.2.
- `fix lambda/apip` computes the switching parameter together with the pair styles `lambda/zone/apip` and `lambda/input/apip`.
- `fix lambda/la/csp/apip` computes the switching parameter of a conservative AP potential based on the CSP according to Chapter 6.
- `pair_style lambda/zone/apip` computes the spatial transition zone according to Eq. (4.13).
- `pair_style lambda/input/apip` computes the input for the calculation of Eq. (4.10), e.g., the CSP.
- `pair_style eam/apip` computes EAM contributions to energies and forces.
- `pair_style pace/apip` computes ACE contributions to energies and forces.
- `pair_style la/csp/apip` computes the forces caused by the differentiation of a conservative AP potential based on the CSP according to Chapter 6.

¹The recent development of the conservative AP potentials is included in APIP at Ref. [217], but not yet part of the LAMMPS distribution.

As one needs multiple commands to define an AP potential, the APIP commands print a warning to the log or abort the simulation at the start of a simulation if a relevant command is missing. Furthermore, there are plausibility checks of the provided parameters, e.g., a negative radius cannot be used.

The integration of APIP commands into the time integration is done as discussed in Section 4.2.5. An input script for LAMMPS, that uses most of these commands to apply an adaptive-precision EAM-ACE potential, is shown in Listing F.1. The reader is referred to the howto discussion[218, pages. 345-347] of the APIP package in the LAMMPS manual for further details². Furthermore, the LAMMPS documentation contains descriptions of all APIP commands, that also explain arguments in detail.

Listing F.1: LAMMPS input script, that reads the initial atom configuration from data.nanoindentation and applies an adaptive-precision EAM-ACE potential with the EAM potential from W.eam.fs and the ACE potential from W.yace. Potential energy, forces and the centro symmetry parameter of the atoms are computed and written to dump.nanoindentation. The arguments of the used commands are explained in the documentation of LAMMPS[156].

```

1  # setup atoms and simulation box
2  units metal
3  atom_style apip # own atom style to store the switching parameter
4  comm_style tiled # for efficient load-balancing
5  timestep 0.001 # in ps due to metal units
6  boundary p p s
7  read_data data.nanoindentation
8  region bottom_region block INF INF INF INF 2.28 units box
9  group bottom_group region bottom_region
10
11 # adaptive-precision EAM-ACE potential with CSP detection mechanism
12 pair_style hybrid/overlay &
13     eam/fs/apip &
14     pace/precise/apip &
15     lambda/input/csp/apip 14 cutoff 6.0 &
16     lambda/zone/apip 12.0
17 pair_coeff * * eam/fs/apip W.eam.fs W
18 pair_coeff * * pace/precise/apip W.yace W
19 pair_coeff * * lambda/input/csp/apip
20 pair_coeff * * lambda/zone/apip
21 fix lambda all lambda/apip 1.5 1.6 time_averaged_zone 4.0 12.0 &
22     110 110 group_ignore_lambda_input bottom_group
23
24 # time integration
25 fix nve all nve
26 fix thermostat all lambda_thermostat/apip
27
28 # dynamic load balancing
29 fix aw all atom_weight/apip 50 eam ace lambda/input lambda/zone all
30 variable myweight atom f_aw
31 fix balance all balance 50 1.1 rcb weight var myweight
32
33 # output: dump potential CSP, energy and force of all atoms
34 compute csp all property/atom apip_lambda_input
35 compute pe all pe/atom
36 dump 1 all custom 1 dump.nanoindentation id fx fy fz c_csp c_pe
37
38 # compute initial timestep
39 run 0

```

²The howto discussion is also available under https://docs.lammps.org/latest/Howto_apip.html.

F.2 Implementing new features

One can provide a new switching function by writing an own fix that computes the switching parameter λ_i for all atoms – like `fix lambda/la/csp/apip` for the conservative switching parameter of Chapter 6. An implementation of a new switching function for a conservative potential requires the implementation of the derivative $\sum_k (\nabla_i \lambda_k) (E_k^{(p)} - E_k^{(f)})$ as pair style for the force calculation – like `pair_style la/csp/apip` for the conservative switching parameter of Chapter 6.

Alternatively, one can solely replace the CSP with another detection mechanism: the class, that provides the code for the command `lambda/input/apip`, contains a virtual function for the computation of the input itself, so that a child class can only provide the detection mechanism in form of this virtual function. The class of the pair style `lambda/input/csp/apip`, that contains only the code required for the CSP calculation, can serve as example. Thereby, one can create custom switching functions by only providing another child class of the pair style `lambda/input/apip`.

Currently, ACE or EAM can serve as fast potential and ACE can serve as precise potential. The potentials are combined via the pair style `hybrid/overlay`. Due to this modularity, a fast potential computes with known switching parameters λ_i the potential energy $\lambda_i E_i^{(f)}$ for all atoms i while the precise potential computes $(1 - \lambda_i) E_i^{(p)}$ (cf. Eq. (4.5)). Therefore, it is straightforward to add new fast or precise potentials to APIP based on an existing constant-precision implementation. First, one has to calculate $F_i = -\sum_k \lambda_k \nabla_i E_k^{(f)}$ or $F_i = -\sum_k (1 - \lambda_k) \nabla_i E_k^{(p)}$. Based on this calculation, one can add λ_k where needed in the energy and force computation. Afterwards, one needs to prevent the computation of energy and force contributions, which are not required, i.e., from products that include the factor 0 in form of λ_k or $(1 - \lambda_k)$. Furthermore, one should prevent communication within the force communication routine to allow effective load-balancing as discussed in Section 4.2.4. As LAMMPS provides well designed classes and functions for the force-calculation, one does not need to use MPI[229] directly. The pair style `eam/apip` can serve as example for such a communication prevention. For the whole implementation, the pair styles `eam/apip` and `pace/apip` can serve as reference.

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