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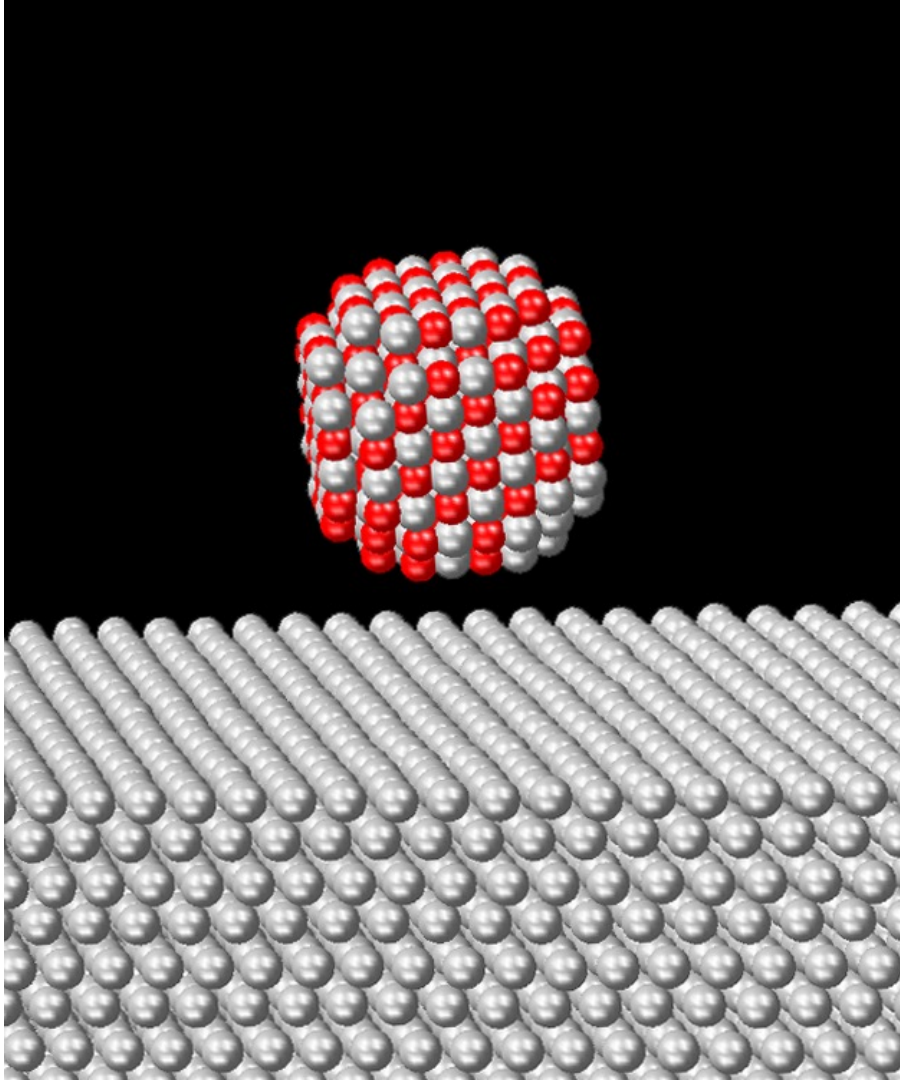
Ab initio for Millions

The Power of Machine-Learned Potentials

December 7-9, 2021



Materials Design Webinar Series

- Each session runs several times to accommodate schedules
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- We will be recording this webinar
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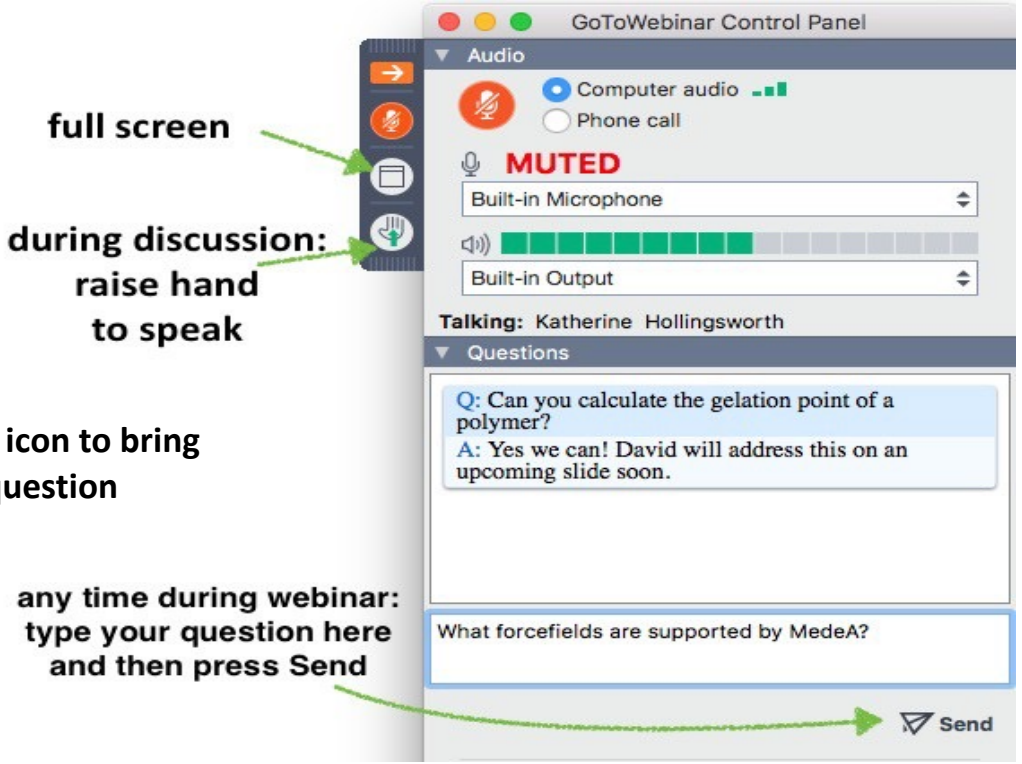
Please Ask Questions!



Use the raise hand icon to bring attention to your question

any time during webinar: type your question here and then press Send

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Webinar Speakers

Katherine Hollingsworth

Volker Eyert

Erich Wimmer

David Reith

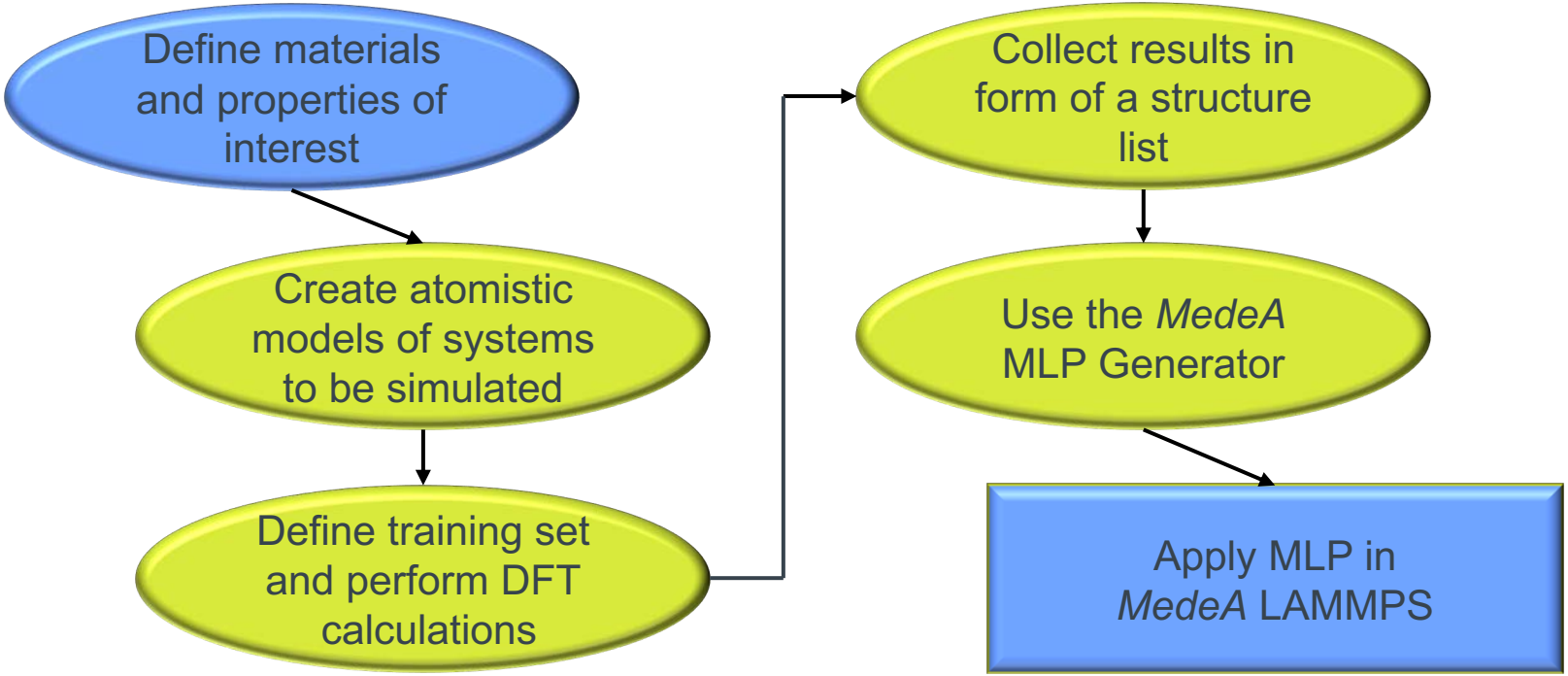
Agenda

- Introduction
- Applications
  - Phase Transitions of Titanium
  - AlN – Thermal Conductivity
  - Water – The Accuracy of MLPs
  - SiO_2 Polymorphs – One for All
  - Impact of Iodine on Zirconium
- Concluding Remarks
- Q&A

Impact of MgO Nanoparticle on Mg Slab

Molecular Dynamics Simulation using a Machine-Learned Potential generated with MedeA MLPG

Generating MLPs in *MedeA*: Workflow



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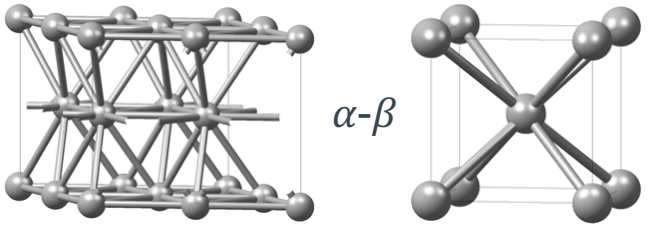
Titanium

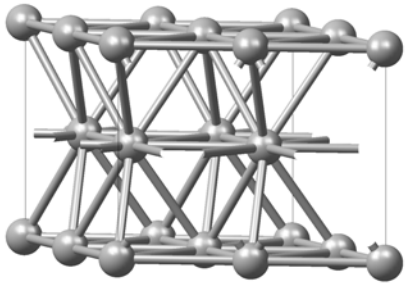
Explore α-β Phase Transition of Ti with SNAP



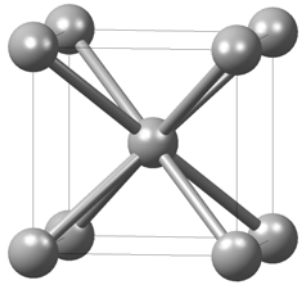
Training Set Calculations

Training set structures
- Initial structures, supercells
- Vacancies, self-interstitial atoms, surfaces, stacking faults
- Isotropic and uniaxial strain by ±0.5%, ±1%, ±1.5%, ±2%, ±4%, and ±6%
- Angular deformations by 0.5°, 1°, and 2°
- NPT/NVT MD simulations at 300-1700 K
- Total of 1005 structures

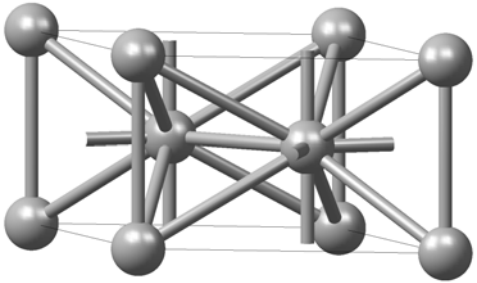
VASP computational parameters
- PBE functional
- 520 eV plane-wave cutoff
- **k**-point spacing 0.2 $Å^{-1}$



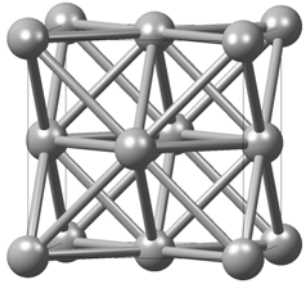
α-phase, hcp
3×3×2 supercell



β-phase, bcc
3×3×3 supercell



ω-phase, hex
2×2×3 supercell



fcc
2×2×2 supercell

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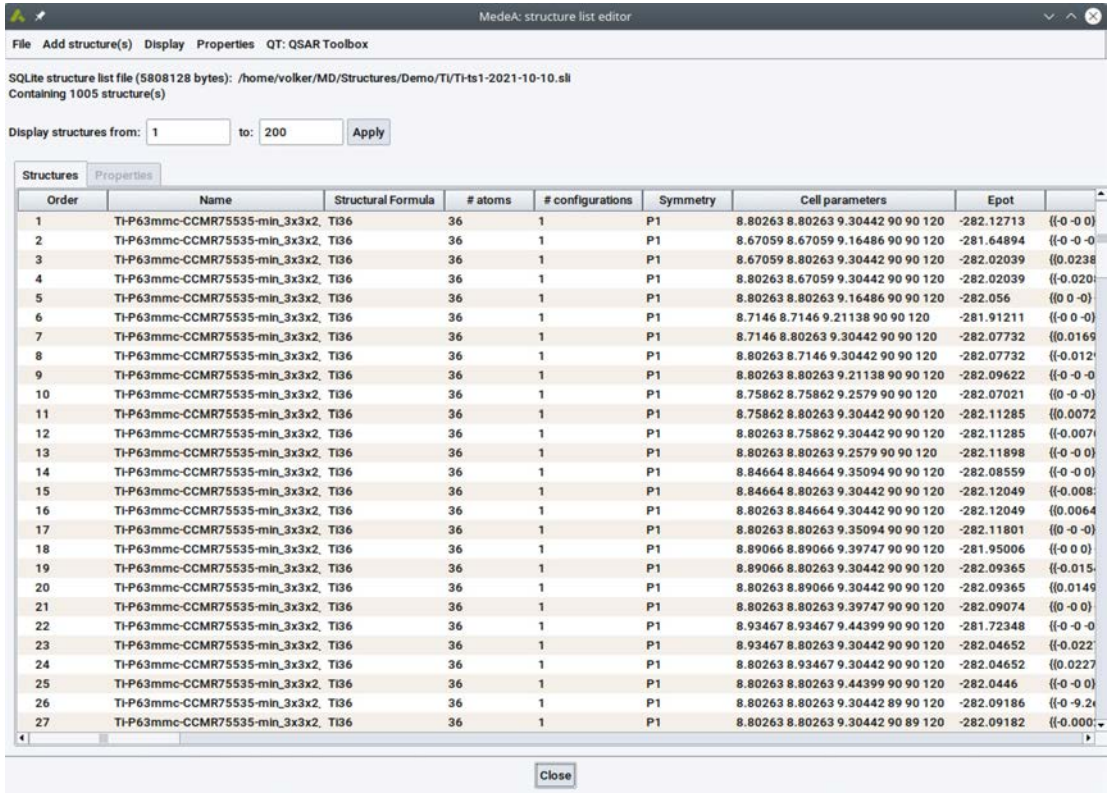
MedeA MLP Generator: Prepare



Collect information about training set in structure list

- Structures
- Energies
- Forces
- Stresses

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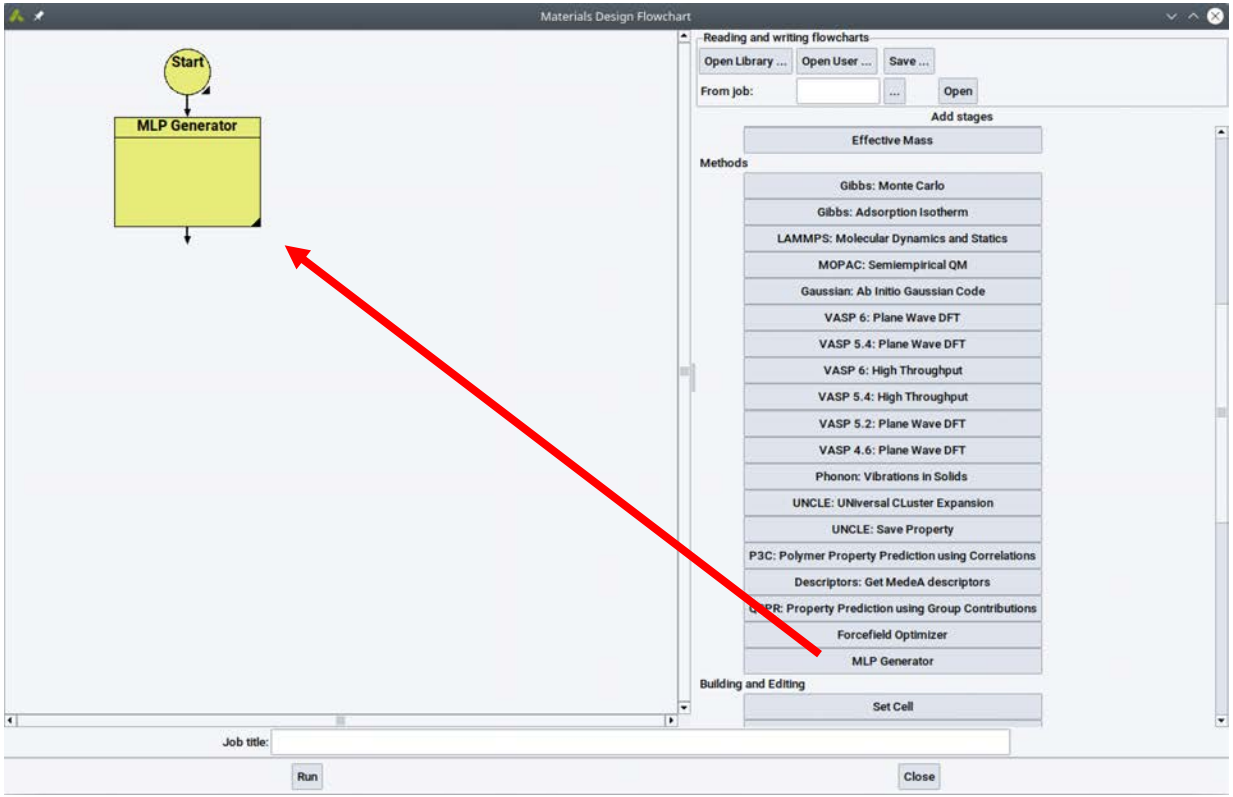
MedeA MLP Generator: Start



Call MLP Generator in Flowchart

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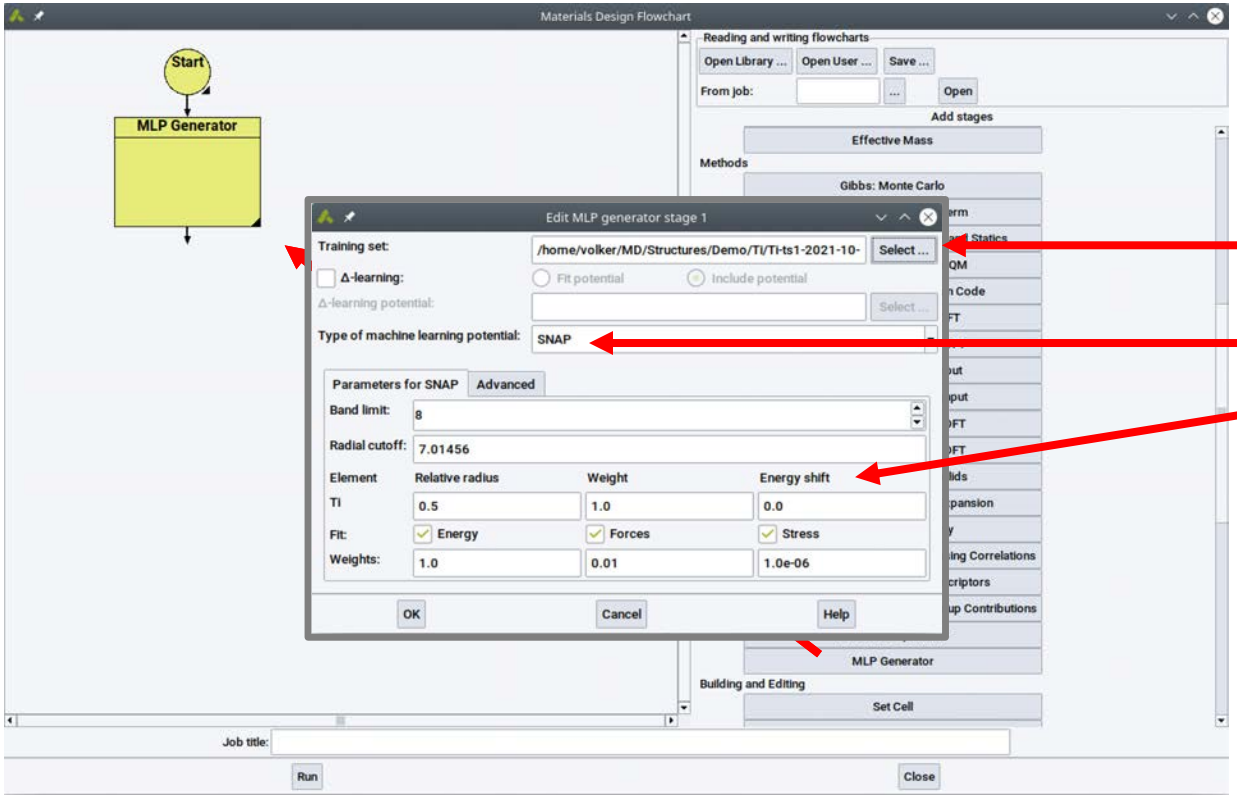
MedeA MLP Generator: Specify



Call MLP Generator in Flowchart

Specify

- Training set, i.e, structure list
- Type of potential
- Basic parameters

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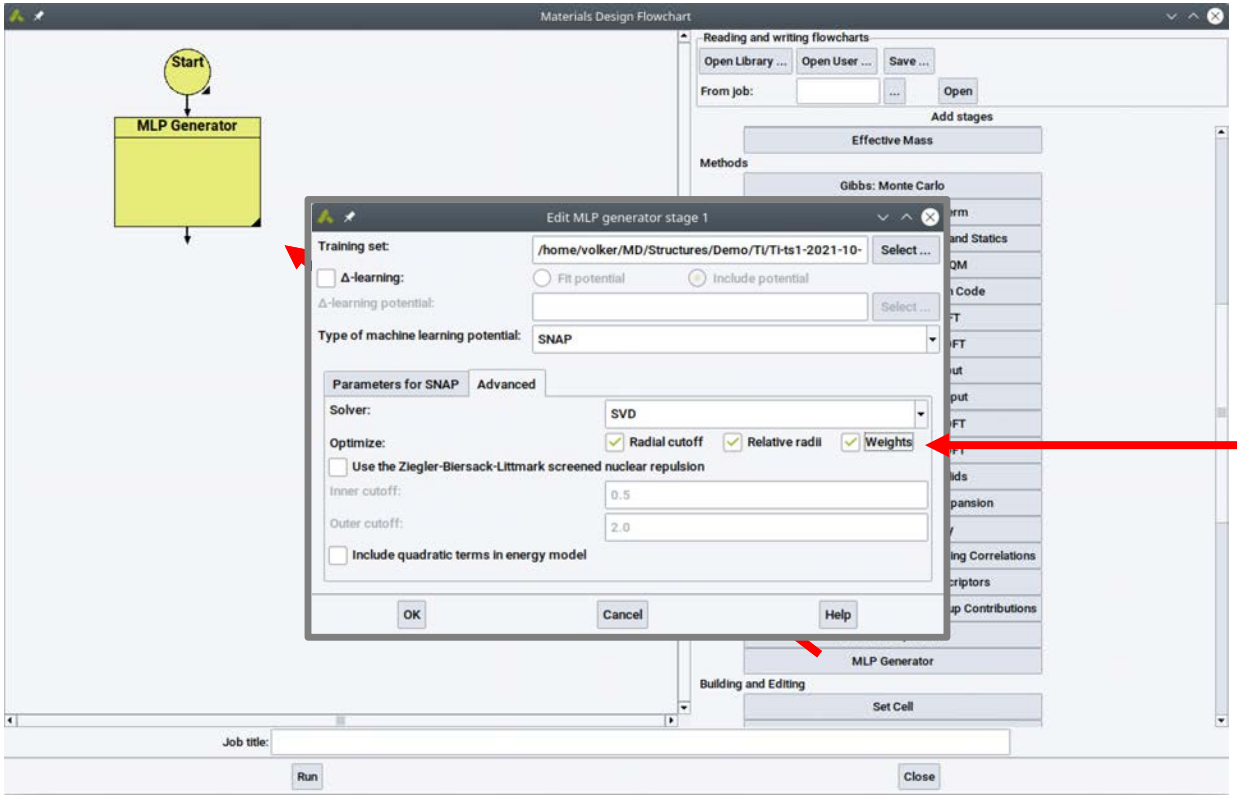
MedeA MLP Generator: Finetune



Call MLP Generator in Flowchart

Specify

- Training set, i.e, structure list
- Type of potential
- Basic parameters
- Advanced settings

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MedeA MLP Generator: Analyze

```
Stage 1: MLP Generator

        Input for FitSNAP is in "Ti-ts1-2021-10-10.in":
          Band limit:    10
          Radial cutoff: 7.01456 Ang
          Elements:
          +-----------------------------------------+
          |     Rel. radius  Weight   Energy shift |
          +-----------------------------------------+
          | Ti     1.0000    0.5000     0.000000   |
          +-----------------------------------------+

          Properties used in the fit: energy forces stress
          with weights:               1.0 0.05 1.0e-05

          The Ziegler-Biersack-Littmark screened nuclear repulsion with inner/outer radius of 0.5/2.0 Ang is being used.

          The following parameters are optimized during the MLP generator run:
          Parameter               Initial    Optimized
          Radial cutoff           7.01456     6.02630
          Relative radius for Ti  0.50000     0.59345
          Weight for Ti           1.00000     1.00917

        Fitting statistics
        +-------------------------------------------------------------------------------------------------------------+
        | Group  Weighting  Subsystem    Count   Mean abso-  Root mean ab-    Root mean     Relative root mean     R    |
        |                                        lute error  solute error  squared error      squared error    squared |
        +-------------------------------------------------------------------------------------------------------------+
        |   all Unweighted  Combined    127401      59.9335        0.0739       487.8104             0.0586   0.9966 |
        |   all Unweighted  Energy        1005       0.0133        0.1452         0.0178             0.1341   0.9820 |
        |   all Unweighted  Forces      120366       0.0583        0.3595         0.1091             0.2428   0.9411 |
        |   all Unweighted  Stress        6030    1265.1000        0.0738      2242.2232             0.0586   0.9966 |
        |   all   Weighted  Combined    127401       0.0035        0.0453         0.0074             0.0108   0.9999 |
        |   all   Weighted  Energy        1005       0.0133        0.1452         0.0178             0.1341   0.9820 |
        |   all   Weighted  Forces      120366       0.0029        0.3595         0.0055             0.2428   0.9411 |
        |   all   Weighted  Stress        6030       0.0127        0.0738         0.0224             0.0586   0.9966 |
        +-------------------------------------------------------------------------------------------------------------+
        Units: energy: eV/atom, force: eV/Angstrom, pressure: bar

        Generated SNAP has been converted to "/data/medea/ContractJobs/dir97000/97338/Stage_1/Ti-ts1-2021-10-10.frc".
```

Information in Job.out

- Type of potential (SNAP, NNP)
- (Optimized) parameters
- Fitting statistics
- Pointer to generated MLP

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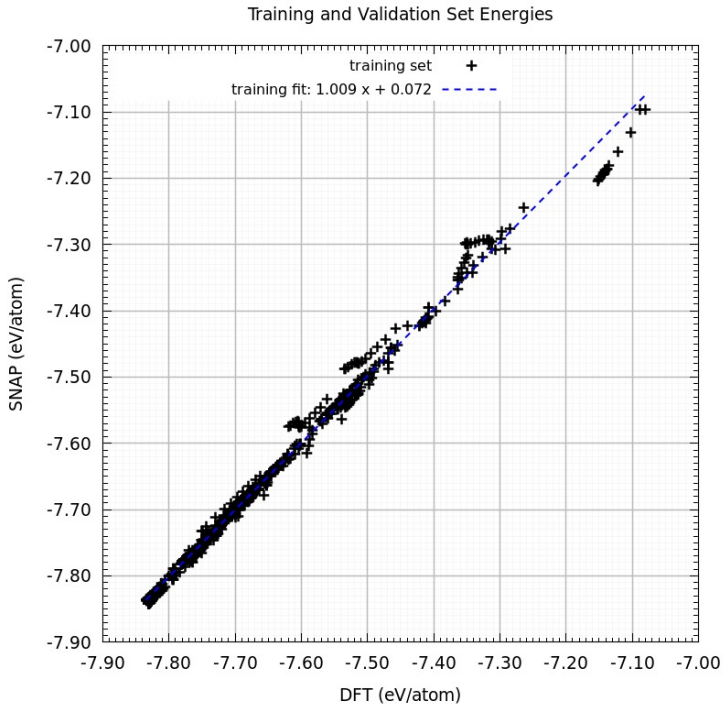
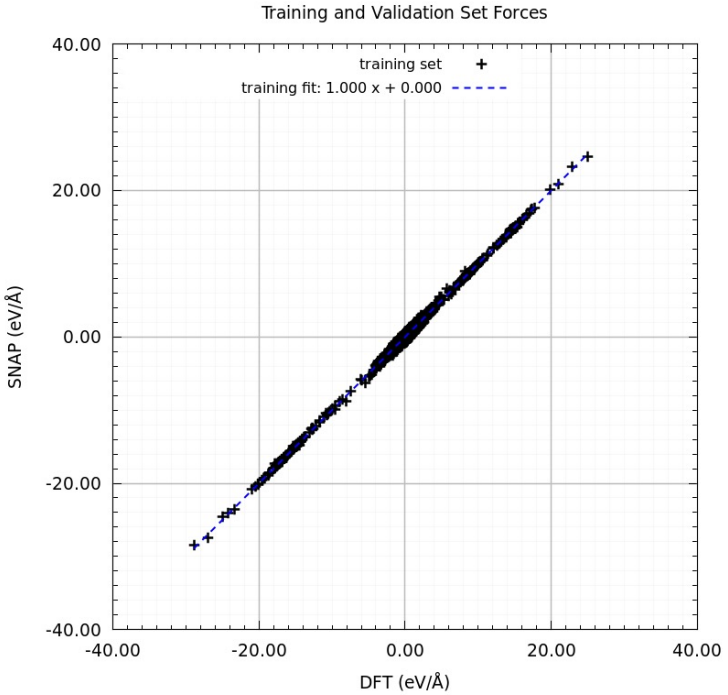
MedeA MLP Generator: Assess



Energies



Forces

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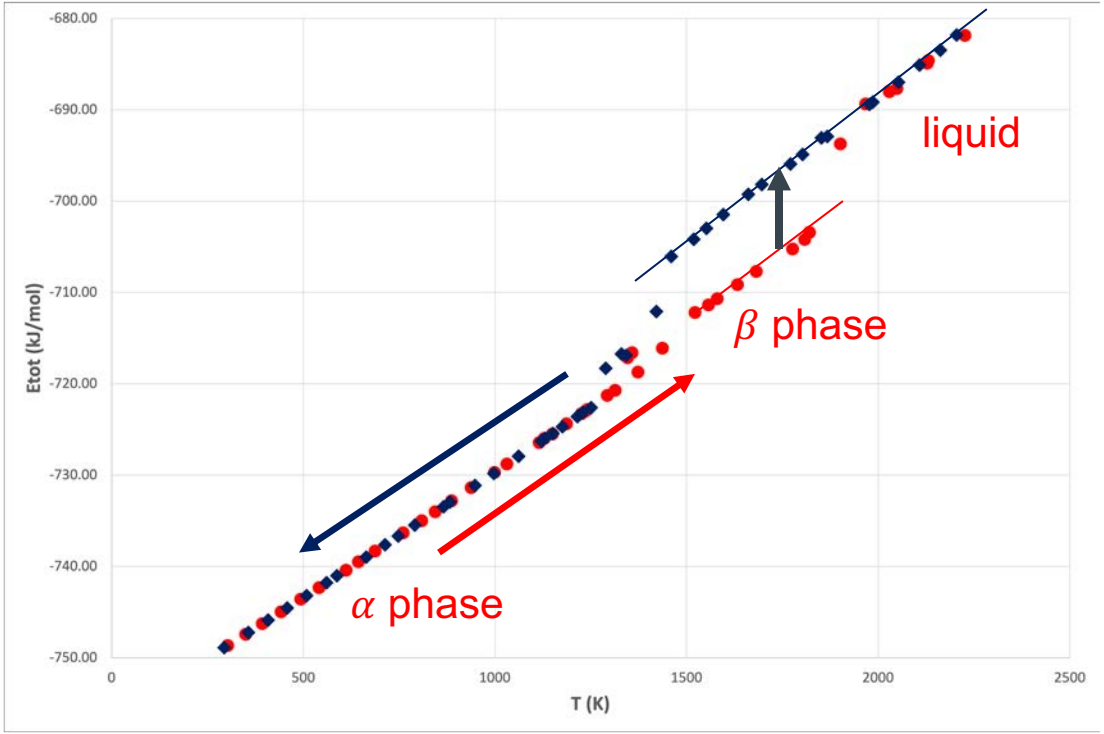
Ti: α-β Phase Transition



CCMR 109654, 109657

Transition temperatures (exp):
- α-β transition: 1156 K
- melting: 1946 K

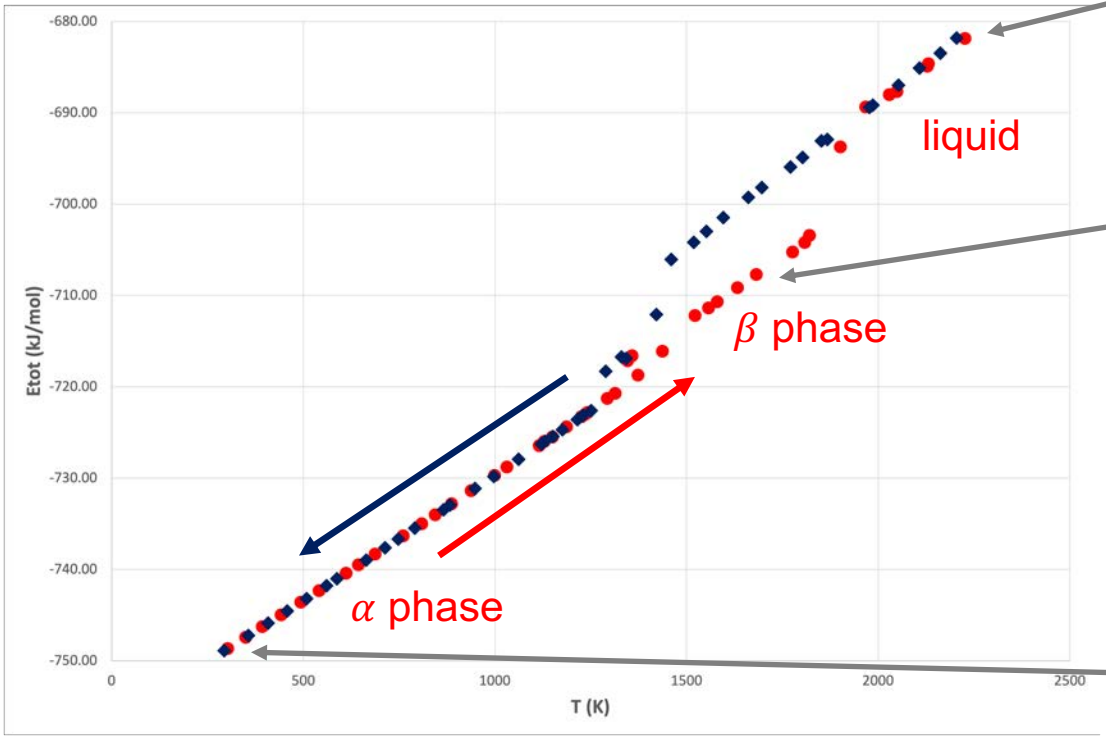
Latent heat of α-β transition:
- MLP: ΔH = 2.7 kJ/mol
- Exp.: ΔH = 4.3 kJ/mol

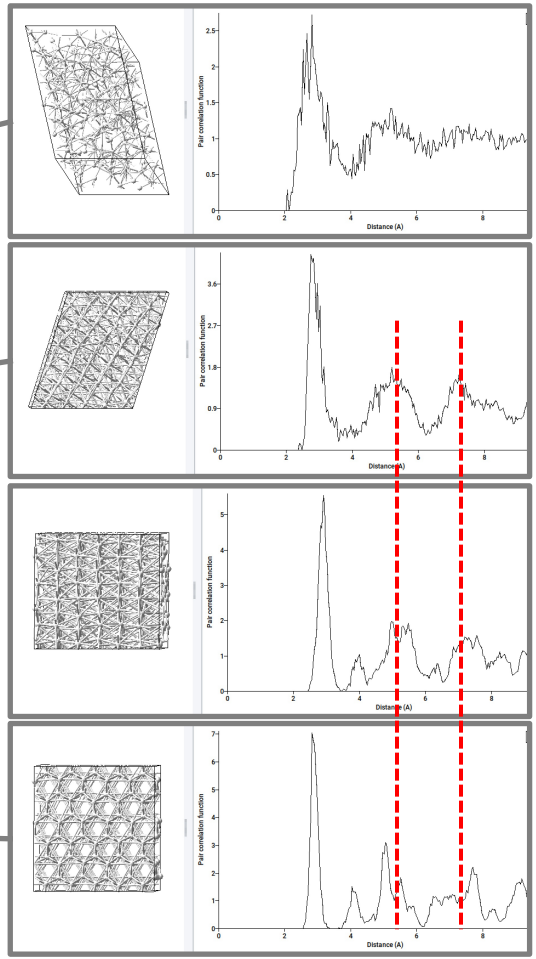
Heat of fusion:
- MLP: ΔH = 10.4 kJ/mol
- Exp.: ΔH = 13 kJ/mol

Specific heat:
- MLP: c_p = 28-30 J/(mol·K)
- Exp.: c_p = 25-37 J/(mol·K)

McClure et al., Int. J. Thermophys. **13**, 75 (1992), Kaschnitz et al., J. Therm. Anal. Calorim. **64**, 351 (2001), Int. J. Thermophys. **23**, 1339 (2002)

Ti: α-β Phase Transition





β phase at 300 K

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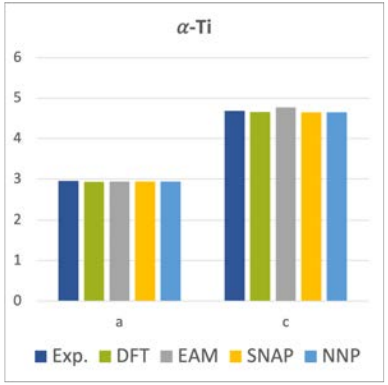
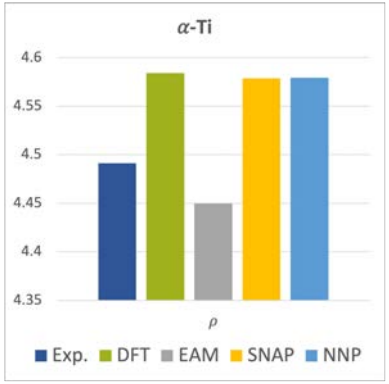
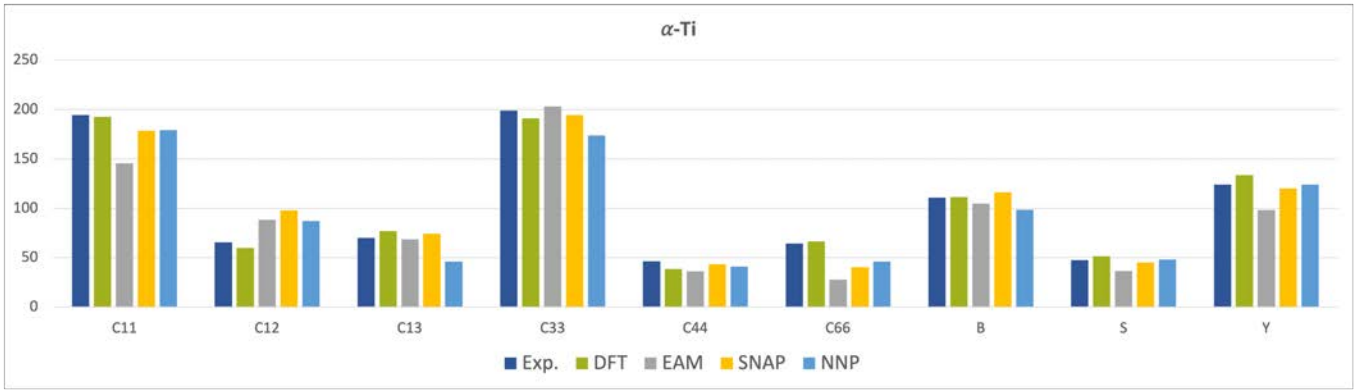
α-Ti: Equilibrium and Elastic Properties





DFT: *MedeA* VASP, EAM: Zhou et al., Francis et al., Exp.: P. D. Hao et al., J. Mater. Res. Technol. **9**, 3488 (2020)



a,c in Å
ρ in g/cm^3
C_{ij} in GPa

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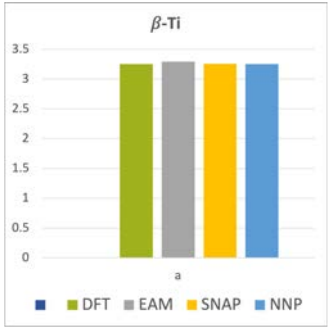
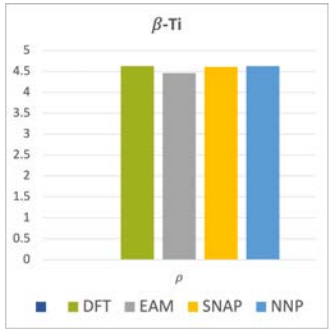
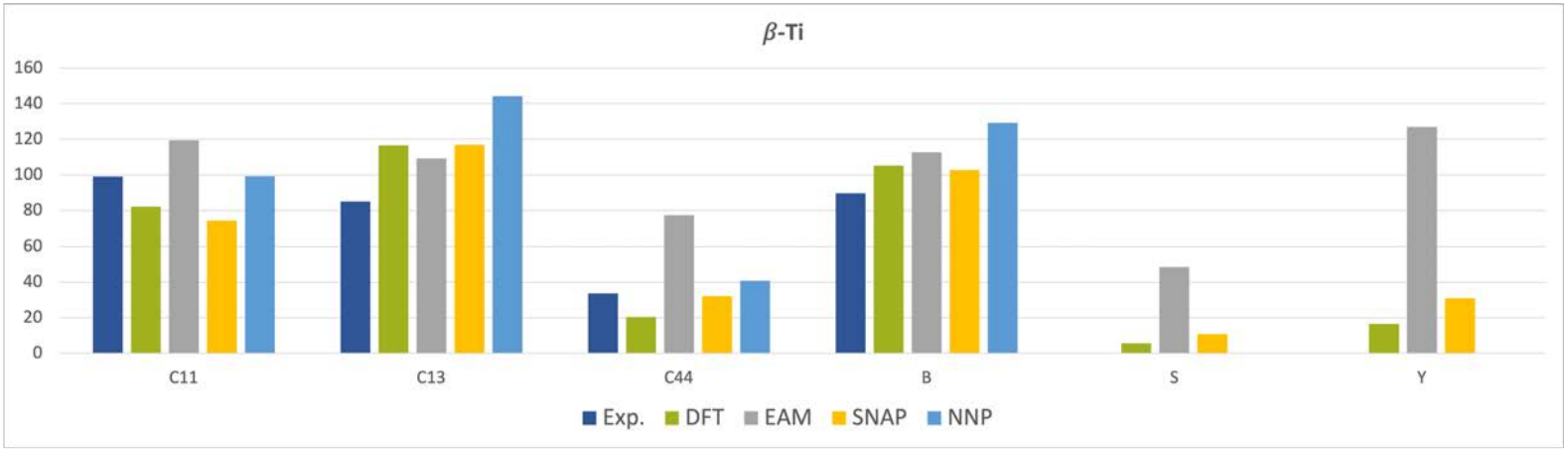
β-Ti: Equilibrium and Elastic Properties





DFT: *MedeA* VASP, EAM: Zhou et al., Francis et al., Exp.[a]: H. Ledbetter et al., J. Appl. Phys. **95**, 4642 (2004), Exp.[b]: E. Fisher and D. Dever, *Science, Technology, and Application of Titanium* (Pergamon, NY 1970), p. 373



a,c in Å
ρ in g/cm^3
C_{ij} in GPa

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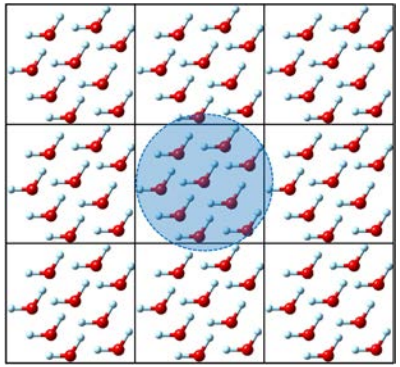
Approaches to MLPs

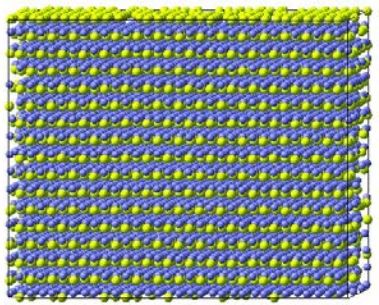


- Divide structure into local atomic environments/clusters described in terms of atom positions and types
- Express energies/forces/stresses in terms of local descriptors of all clusters
- Determine (train) parameters relating energies/forces stresses and local descriptors using *ab initio* calculations

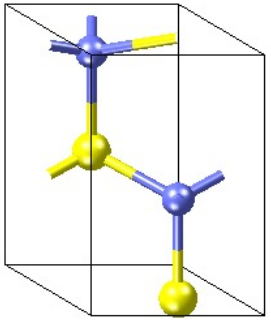
- Spectral Neighbor Analysis Potential (SNAP)
  - A. Thompson and coworkers
  - FitSNAP code by A. Thompson *et al.*, Sandia
- Neural Network Potential (NNP)
  - J. Behler and M. Parrinello
  - n2p2 code by A. Singraber, Univ. Vienna

Thermal Conductivity

AlN in Wurtzite Structure

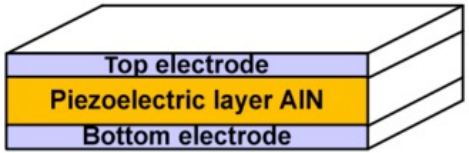


Industrial Application of Wurtzite Aluminum Nitride



- High thermal conductivity
  - Remove heat with an insulating material
- Good piezoelectric and acoustic properties
  - Thin film AlN on Si wafers used as surface acoustic wave sensors
  - Used as ultrasound transducers: emit and receive ultrasound
  - Acoustic resonator (resonance frequency $= \frac{v_{acoustic}}{2d}$) can be used as a radio frequency filter or a pressure sensor

Piezoelectric plate to convert audio signal to sound waves (wikipedia.org)



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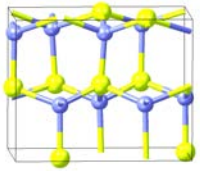
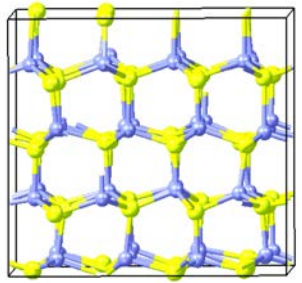
Training Set

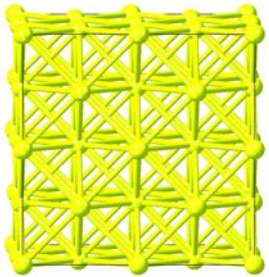
- NPT MD trajectory for 16 atoms and 96 atoms wurtzite AlN supercell
  - Temperatures: 150 K, 300 K, 600 K, 1500 K
- Strained Al-N structures: wurtzite, rocksalt, zinc blende
- NVT MD trajectories of (001) and (110) wurtzite AlN surface slabs
- NPT MD trajectory for a 32 atoms fcc Al supercell
  - Temperature: 300 K to 1000 K
- Total number of structures in training set: 431



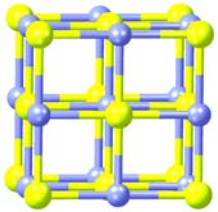
16 atoms wurtzite AlN



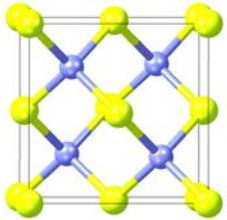
96 atoms wurtzite AlN



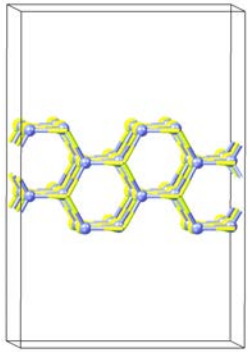
32 atoms fcc Al



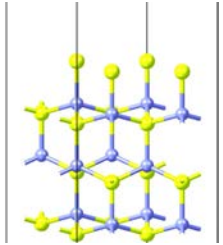
AlN rocksalt



AlN zinc blende



AlN (110) surface slab



AlN (001) surface slab

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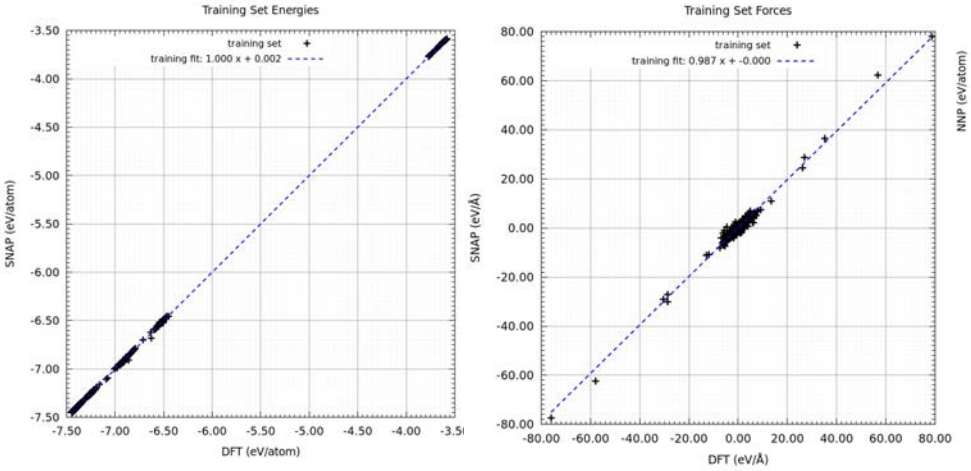
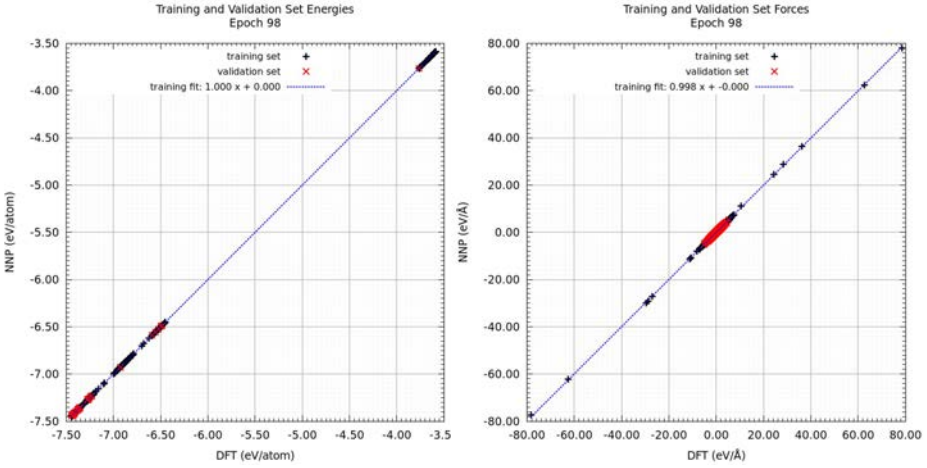
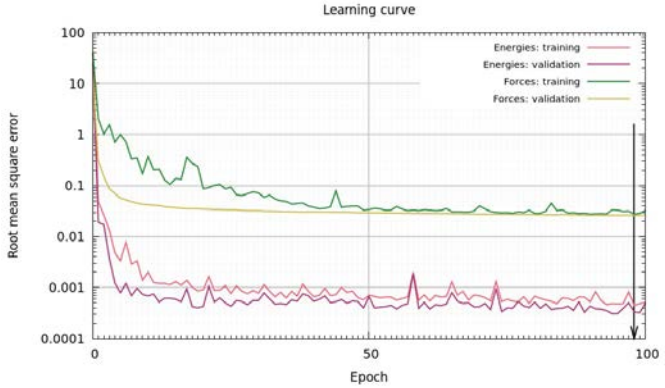
Machine Learned Potential

SNAP



NNP





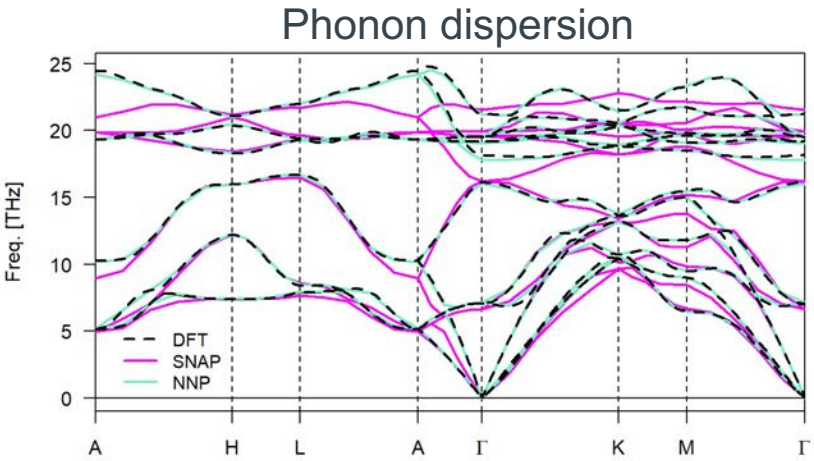
Elastic and Vibrational Properties

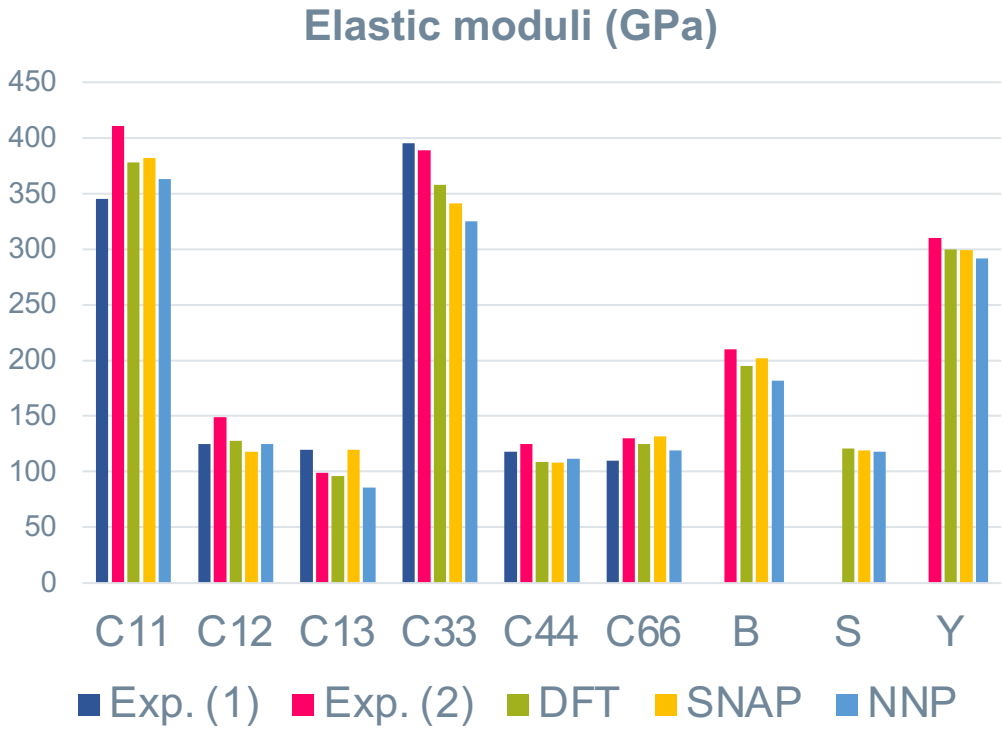
NNP

- Excellent phonon dispersion
- Good elastic properties

SNAP

- Good phonon dispersion
- Good elastic properties





[1] C.M. Lin et al., Appl. Phys. **109**, 033514 (2011) [2] McNeil et al., J. Am. Ceram. Soc. **76**, 1132 (1993)

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Thermal Conductivity with Green-Kubo

- Green-Kubo expression relating lattice thermal conductivity to the integral of the time autocorrelation function of the heat-flux density

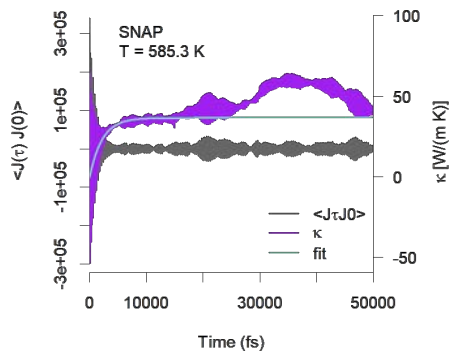
$$\kappa\,(t_m) = \frac{1}{\Omega k_B\, T^2} \int_0^{t_m} < J(t) \cdot J(0) > dt$$

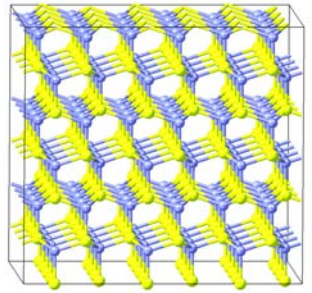
Ω volume, k_B Boltzmann constant, T temperature, J thermal current

- Thermal current: $J(t) = \frac{d}{dt} \sum_i r_i(t) \epsilon_i(t)$

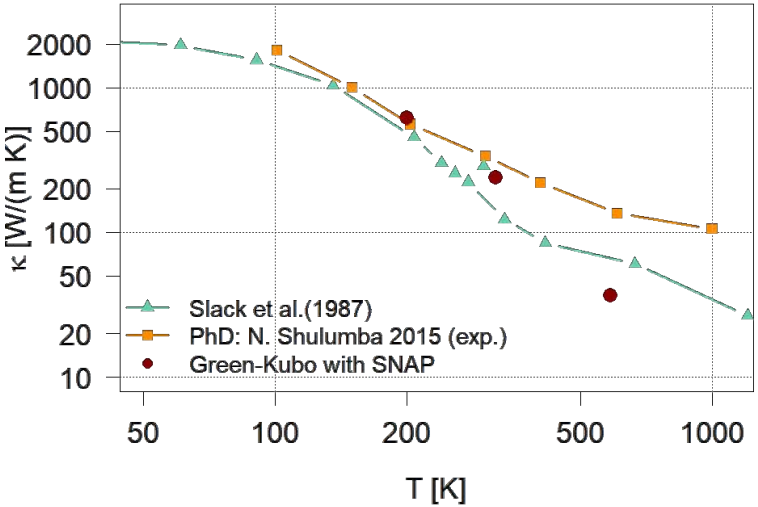
$r_i(t)$ time-dependent coordinate of atom i, $\boldsymbol{\varepsilon}_i(t)$ site energy

Temperature [K]	exp. κ [W/(mK)]	calc. κ [W/(mK)]
300	285[1], 320[2]	238 (@320 K)





288 atoms

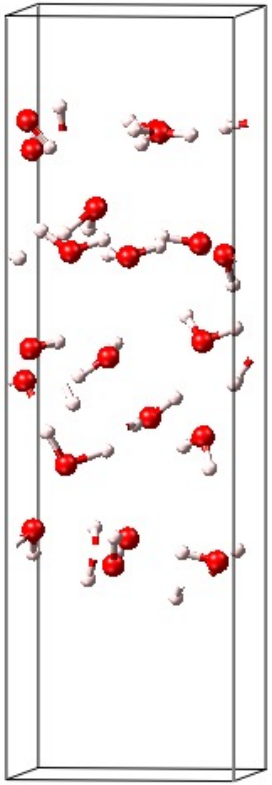


[1] G. A. Slack, R. Tanzilli, R. Pohl, and J. Vandersande, J. Phys. Chem. Solids **48**, 641-647 (1987)
[2] PhD: N. Shulumba, Linköping University (2015)

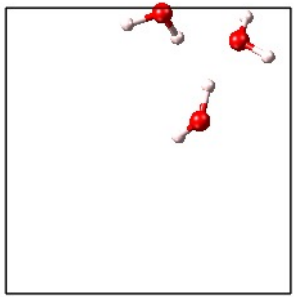
Water

The Accuracy of MLPs

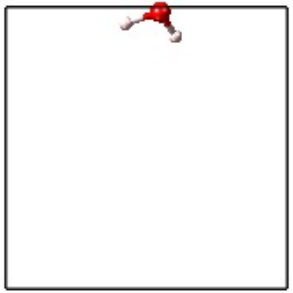
Training Set



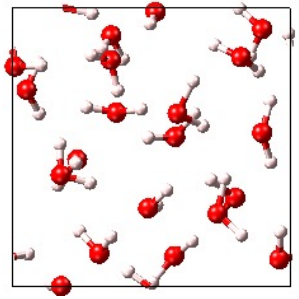
water slab



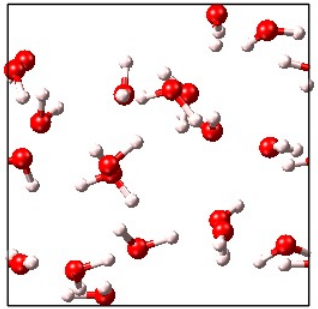
3 water molecules



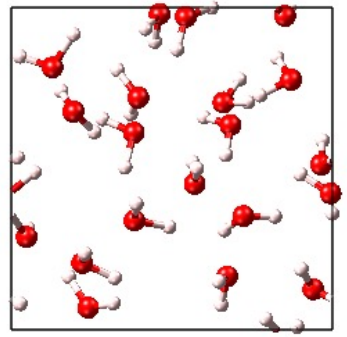
1 water molecules



ρ = 1100 kg/m^3



ρ = 1000 kg/m^3

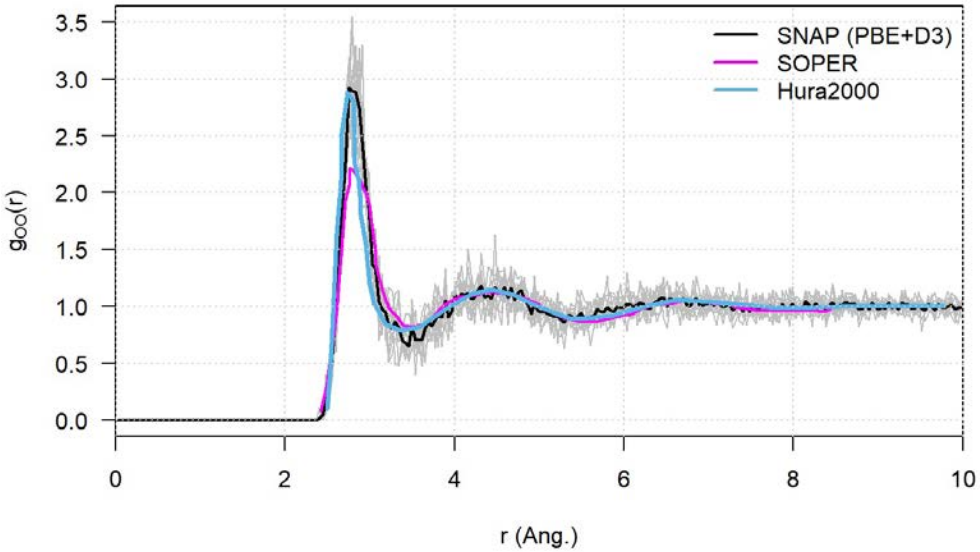


ρ = 900 kg/m^3

- Training set composed of VASP MD simulations
- VASP: PBE with van der Waals D3-zero-damping corrections

materials design

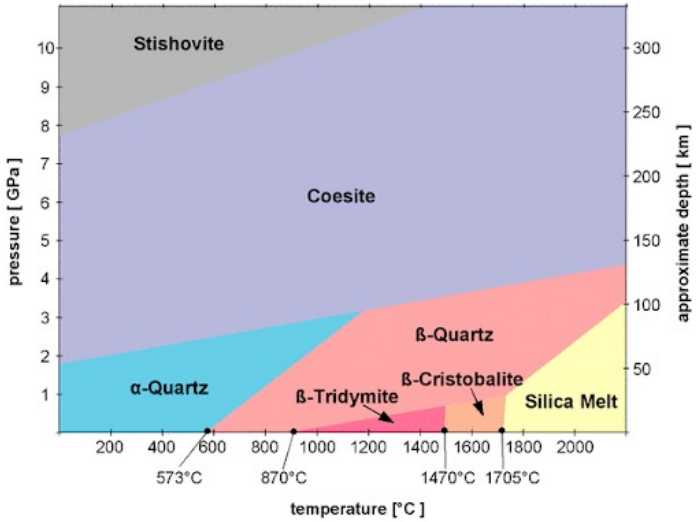
Water Structure



- Excellent $g_{OO}(r)$ agreement with exp.
- Density (298.2K, 1atm): 991.6 kg/m^3
  exp.: 997 kg/m^3
- H-O-H angle: 103.4° (4.3°)
  exp.: 104.45°

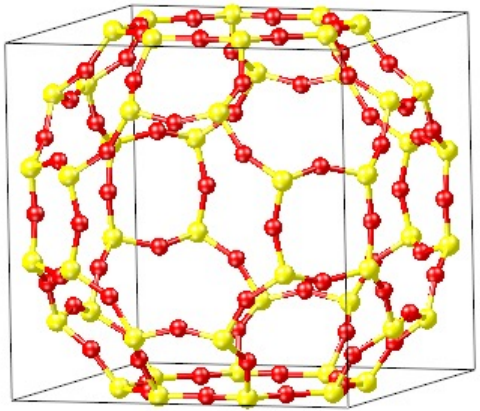
SiO_2 Polymorphs

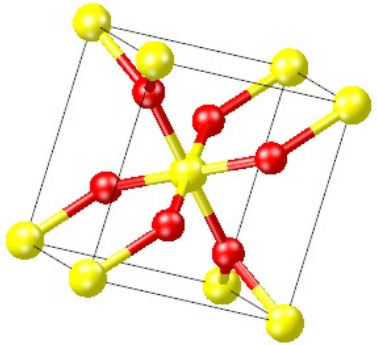
A Single MLP for a Multitude of Structures



Structures and Data for Fit

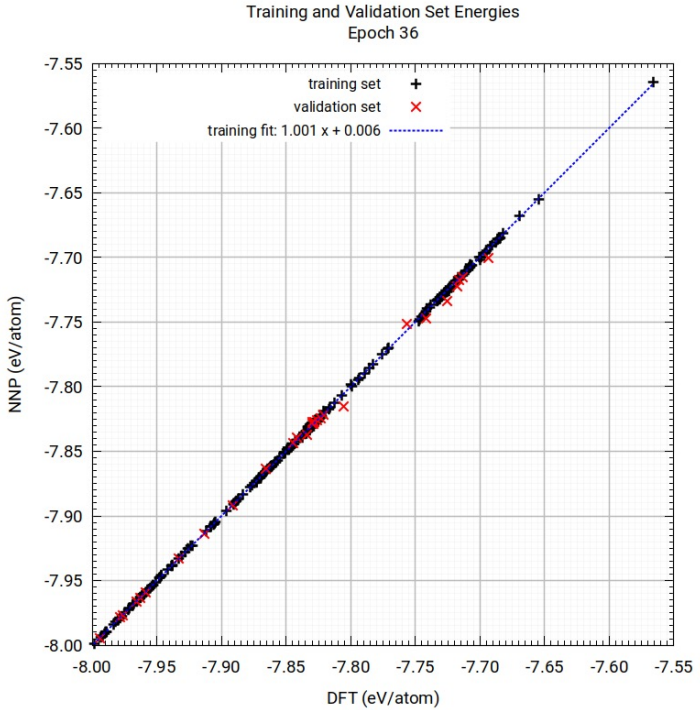
- Structures used to fit MLPs
  - α-quartz (2x2x2), zeolite Linde Type A, stishovite (2x2x2)
  - Cells distorted by -6%, -4%, -2%, 2%, 4%, 6% along a, b and c (simultaneous and separate), -4%, -2%, 2% and 4% for α, β and γ
  - NPT MD simulations at 298, 1100 and 1900 K for quartz and stishovite
  - Total of 298 structures
- VASP parameters
  - GGA-PBE with Grimme D3 correction
  - Plane wave cutoff 510 eV
  - k spacing 0.178
- SNAP and NNP fitted



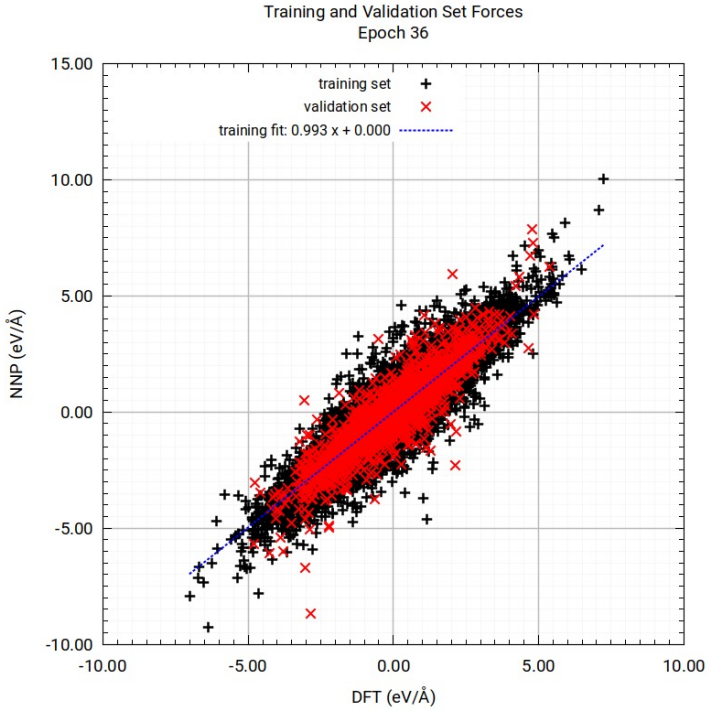


materials design

Quality of Fit



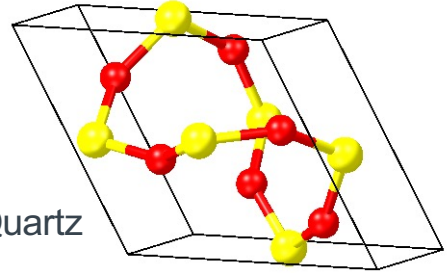
Energies



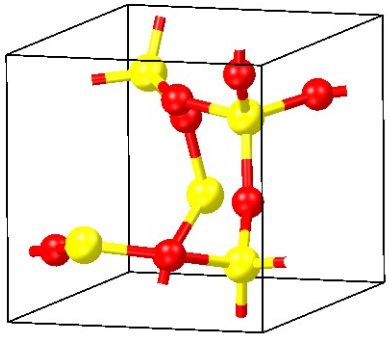
Forces

materials design

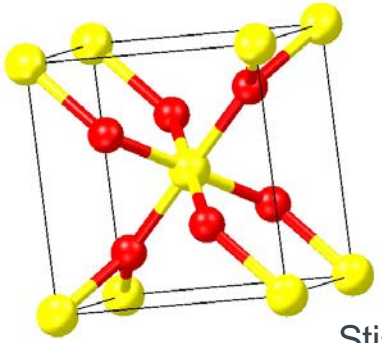
Validation of MLPs



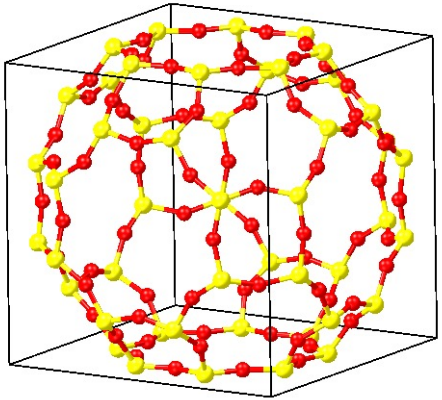
α-Quartz



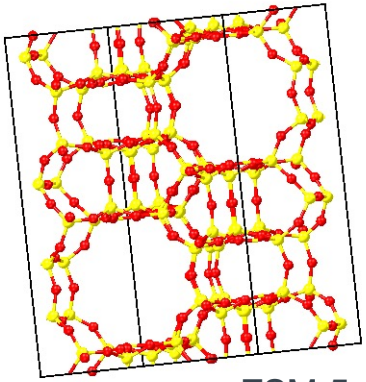
α-Cristobalite



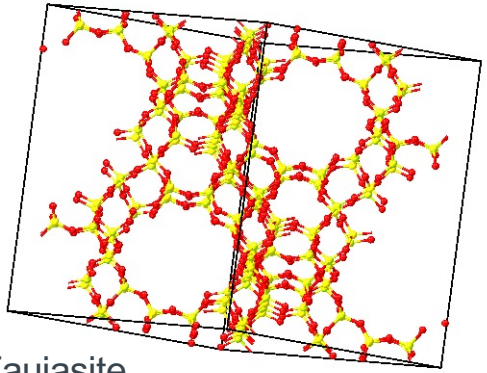
Stishovite



Linde type A



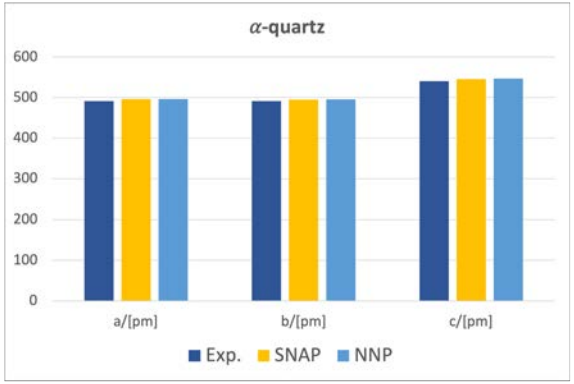
ZSM-5

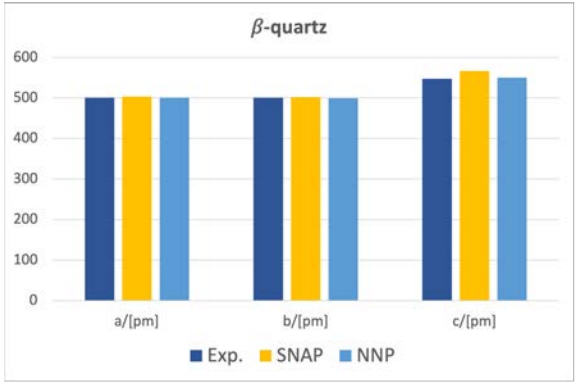


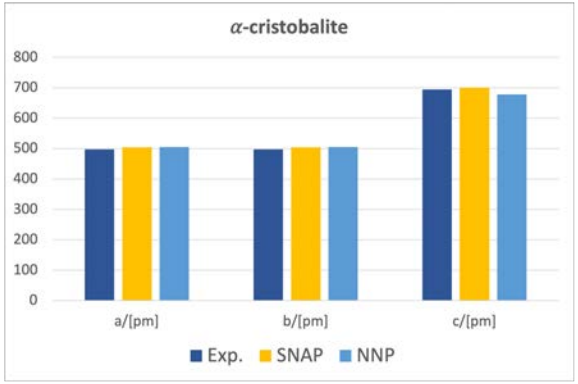
Faujasite

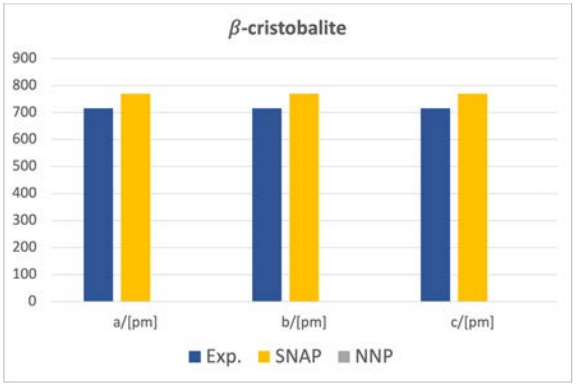
materials design

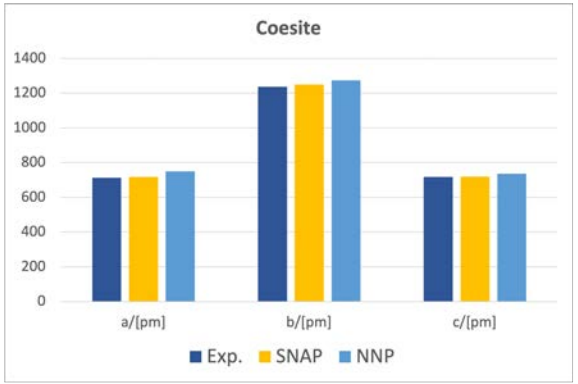
Results

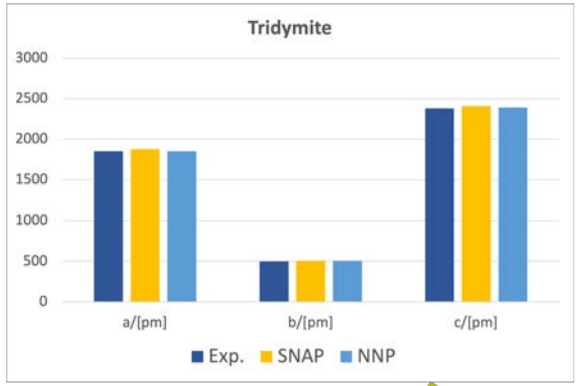






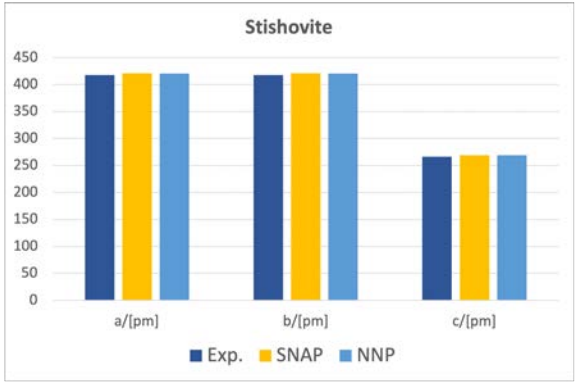


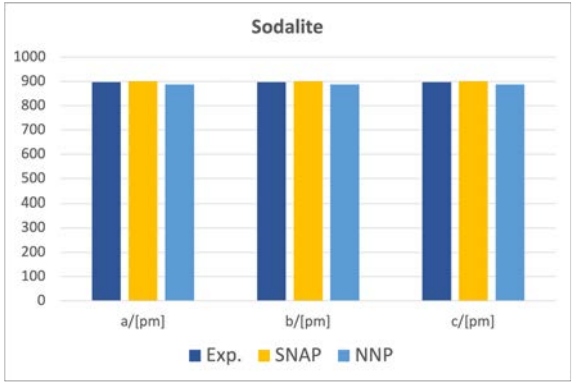


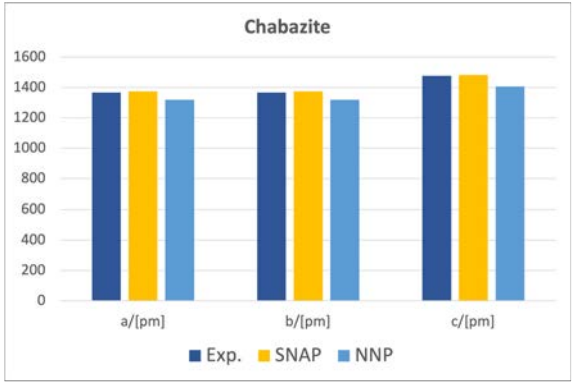


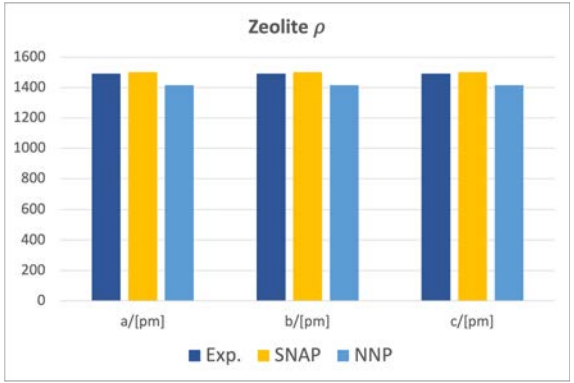
materials design

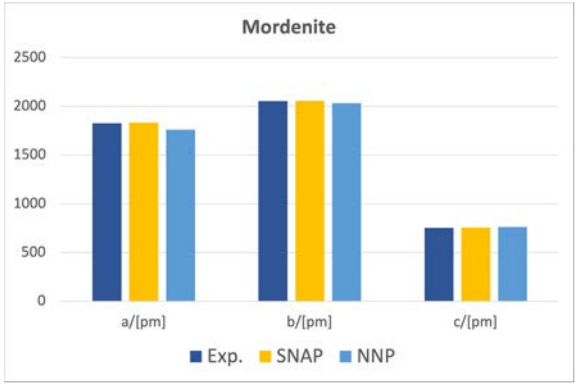
Results

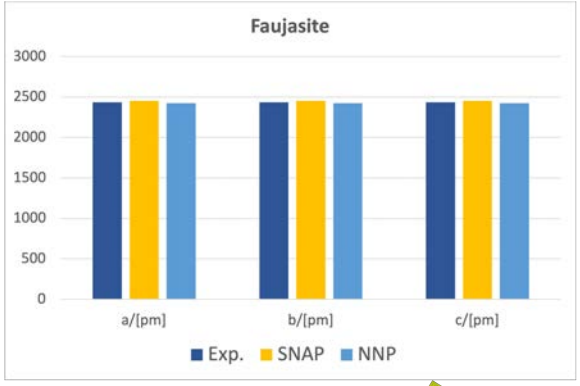






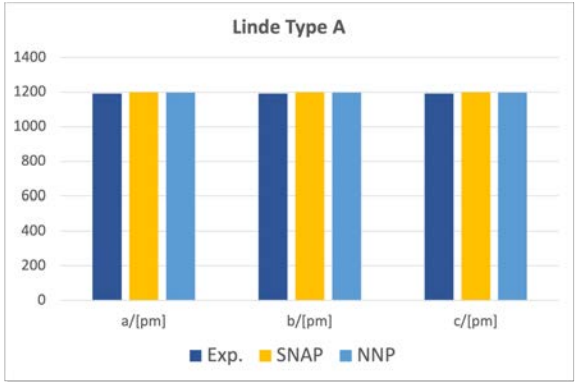






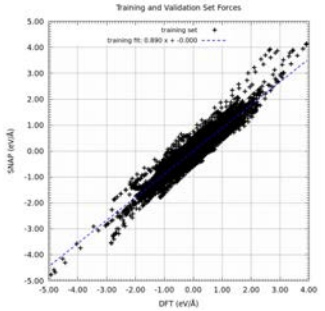
materials design

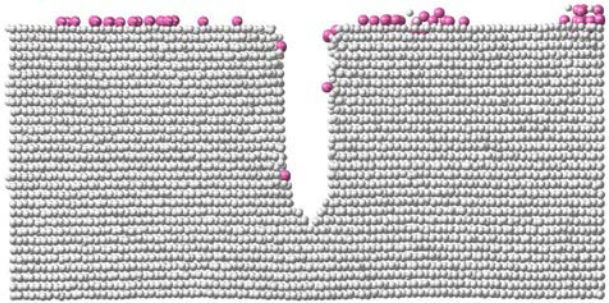
Results



Maximum deviation: 7.4 % (β-cristobalite), 5.1 % (coesite)

Iodine on Zr


Training and Validation Set Forces
training set
training fit: 0.890 x + -0.000
SNAP (eV/Å)
DFT (eV/Å)
5.00
4.00
3.00
2.00
1.00
0.00
-1.00
-2.00
-3.00
-4.00
-5.00



Impact of Iodine Molecules on a Zirconium Surface with Nano-Groove

Generation of MLP: Training Set

MedeA: structure list editor

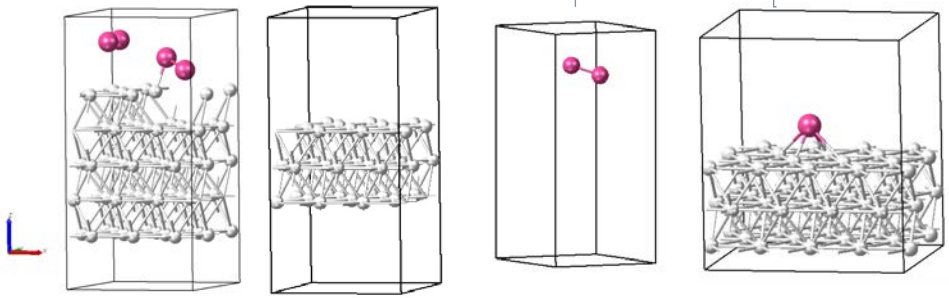
File Add structure(s) Display Properties QT: QSAR Toolbox

SQLite structure list file (1011712 bytes): C:/Active/MLPG demo/Iodine on Zr/Zr-iodine-1.sli
Containing 4 structure(s)

Display structures from: 1 to: 4 Apply

Structures | Properties

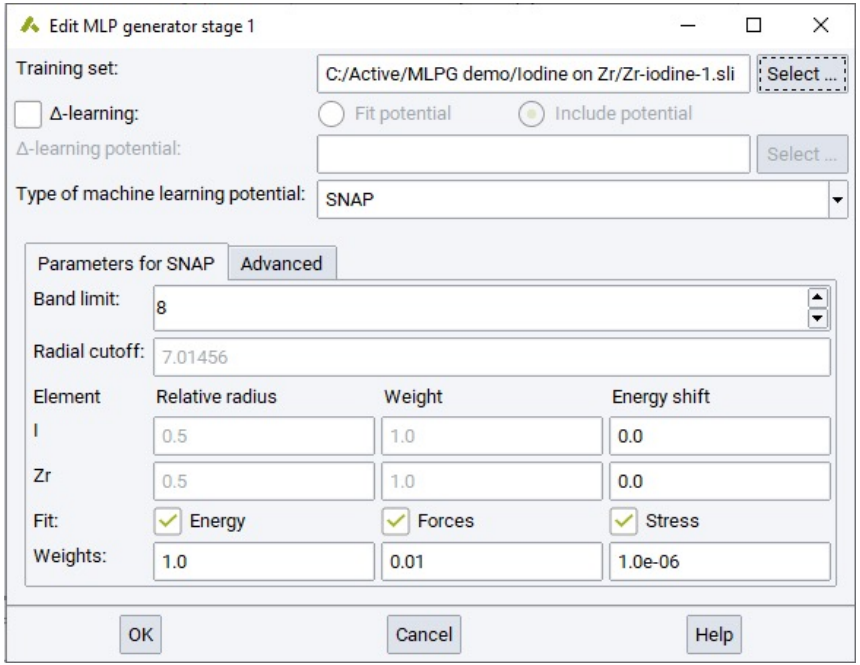
Order	Name	Structural Formula	# atoms	# configurations	Symmetry	Cell parameters
1	Iodine on Zr surface	Zr56I4	60	100	P1	9.70277 11.2038 20 90 90 90
2	Zr 3-layer slab	Zr36	36	100	P1	9.70277 11.2038 20 90 90 90
3	I2 molecule	I2	2	10	P1	9.70277 11.2038 20 90 90 90
4	Zr(0001) slab with I atom heating to 1500 K	Zr48I	49	100	P1	12.9989 11.2574 15 90 90 90

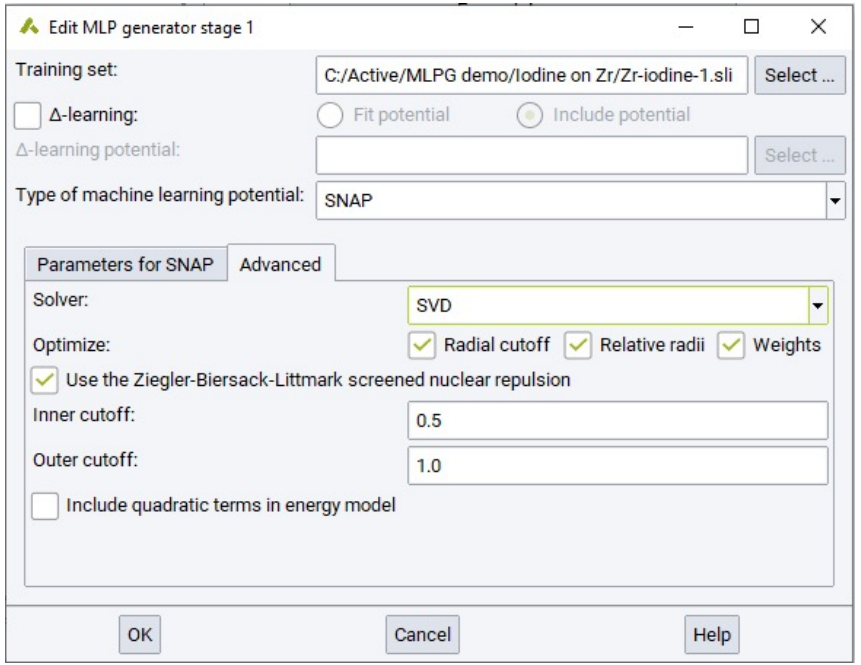


For each of these small structures, short *ab initio* molecular dynamics simulations were carried out using *MedeA* VASP, generating a total of 310 configurations. The energies, forces, and stress tensors are stored in a structure list.

materials design

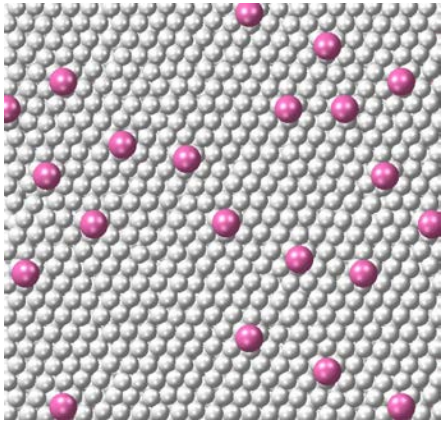
Generation of SNAP with MLPG

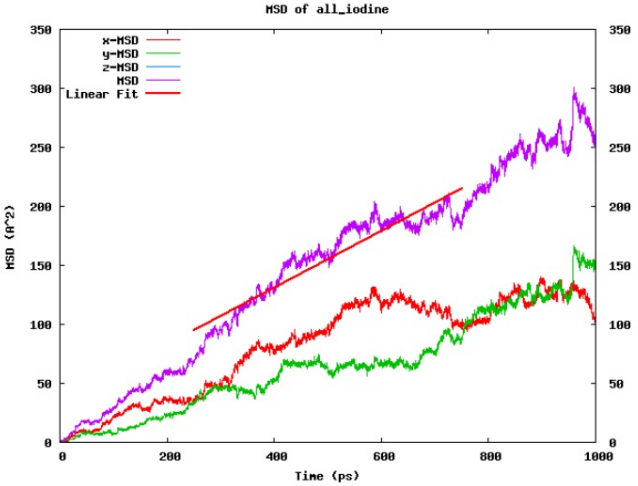




materials design

Diffusion of Iodine on Zr Surface





$$D = \frac{\langle R^2 \rangle}{\alpha t}$$

Mean-square displacement of per iodine atom vs. time during 1 ns with a time step of 1 fs (1 million configurations) using SNAP MLP on GPU. The model contains 1800 atoms.

Compute time: 3h 45 min (job T379)

T=1000 K

Quantity	Value	+/- Uncertainty	Units
MSD D:	4.022e-06	1.9e-08	cm^2/s
MSD-X D:	7.762e-06	5.9e-08	cm^2/s
MSD-Y D:	4.302e-06	3.1e-08	cm^2/s
MSD-Z D:	1.13e-09	1e-10	cm^2/s

Concluding Remarks

MedeA MLP/MLPG

VASP accuracy with LAMMPS speed at your fingertips all within *MedeA*

- Machine-Learned-Potentials (MLPs) offer a combination of extended length and time scales with unprecedented ease in generation and high fidelity with respect to DFT to describe so far inaccessible physical phenomena.
- The *MedeA* MLP Generator (MLPG) offers a fully integrated workflow from training-set generation (using *MedeA* HT) and MLP generation to MLP application using *MedeA* LAMMPS.

MLPs with *MedeA LAMMPS*

***MedeA* MT:** Elastic, mechanical and thermodynamic properties (also at finite temperature)

***MedeA* Deformation:** Perform deformation beyond the elastic regime

***MedeA* Thermal Conductivity:** Calculate lattice thermal conductivity with Green-Kubo or non-equilibrium MD Müller-Plathe

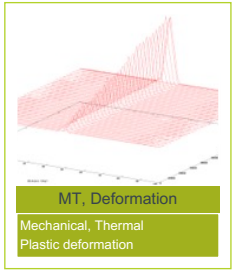
***MedeA* Viscosity:** Calculate viscosity with Green-Kubo or non-equilibrium MD Müller-Plathe

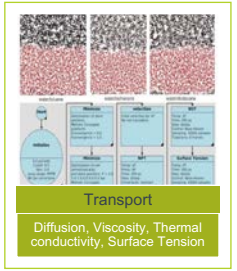
***MedeA* Surface Tension:** Calculate surface tension of fluid slabs

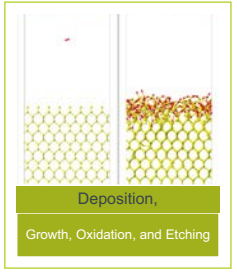
***MedeA* Diffusion:** Automatically calculate diffusivity from mean square displacement

***MedeA* Deposition:** Atomistic scale simulation to study deposition, growth, oxidation and etching

***MedeA* Phonon:** Phonon spectra and thermodynamic functions (vibrational free energy, heat capacities)







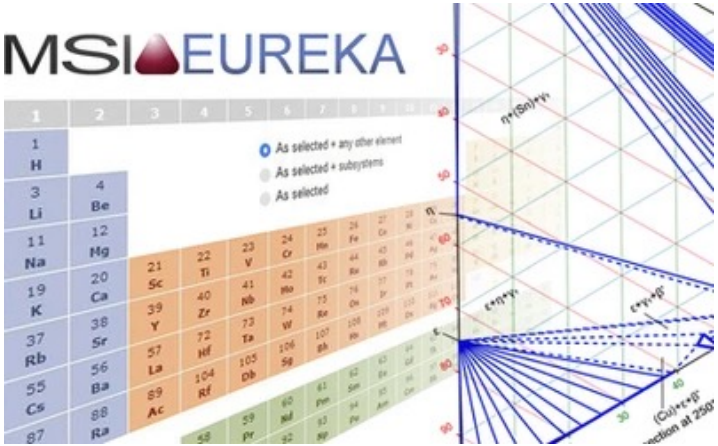
materials design

Announcement

Upcoming

- Webinar Next Week: Materials Constitution Data in MSI Eureka – Fundamentals for Efficient R&D with Live Q&A

- The *MedeA* 3.4 Release



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Question and Answer Session

Dr. Erich Wimmer

Materials Design

Dr. Volker Eyert

Materials Design

Dr. David Reith

Materials Design

Questions about Materials Design Webinars

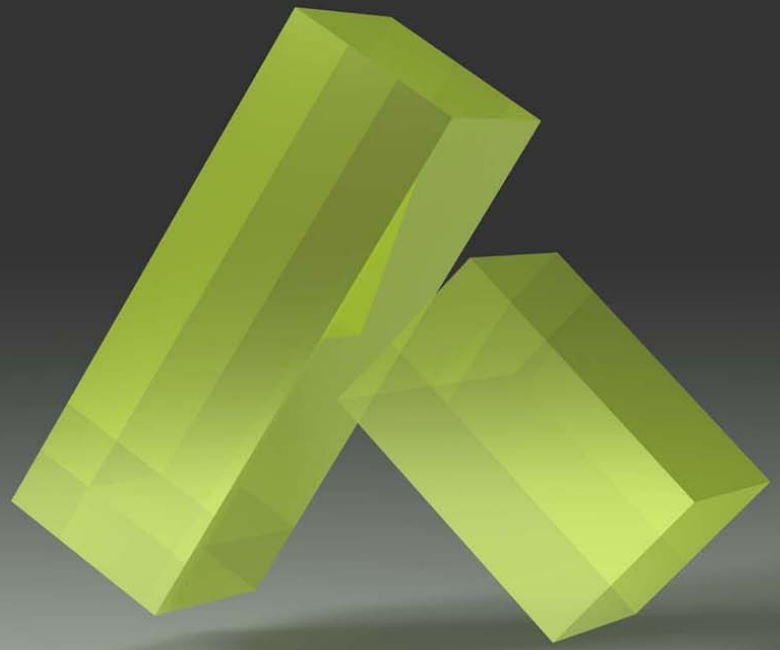
Katherine Hollingsworth

khollingsworth@materialsdesign.com

materials design

info@materialsdesign.com

www.materialsdesign.com



MedeA

Innovation by Simulation