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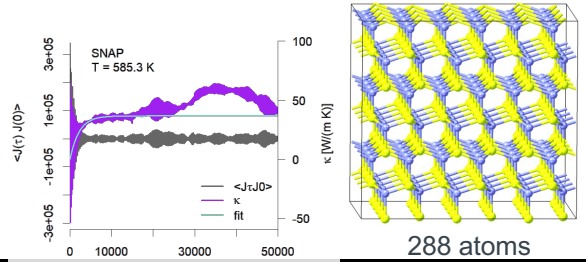


Materials Design 2022 UGM Training Series

Generating and Applying Machine-Learned Potentials with *MedeA*

David Reith

Materials Design

October 13, 2022

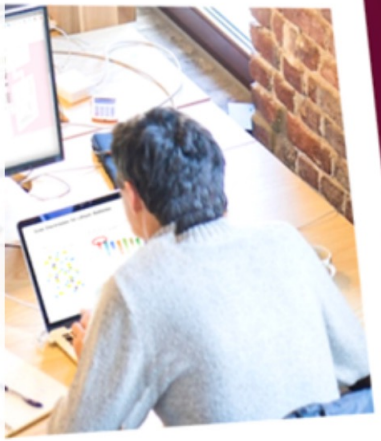
Materials Design UGM

UGM 2022

The Materials Design annual user event will be online for 2022.

Plenary Speakers include:

Prof. Jeffrey Grossman
Prof. Georg Kresse
Dr. Carla Verdi
Prof. Jörg Behler
Dr. Jozef Bicerano



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Materials Design UGM Training Series

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  - Registration details

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- We will be recording this session
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David Reith
Presenter

René Windiks
Moderator

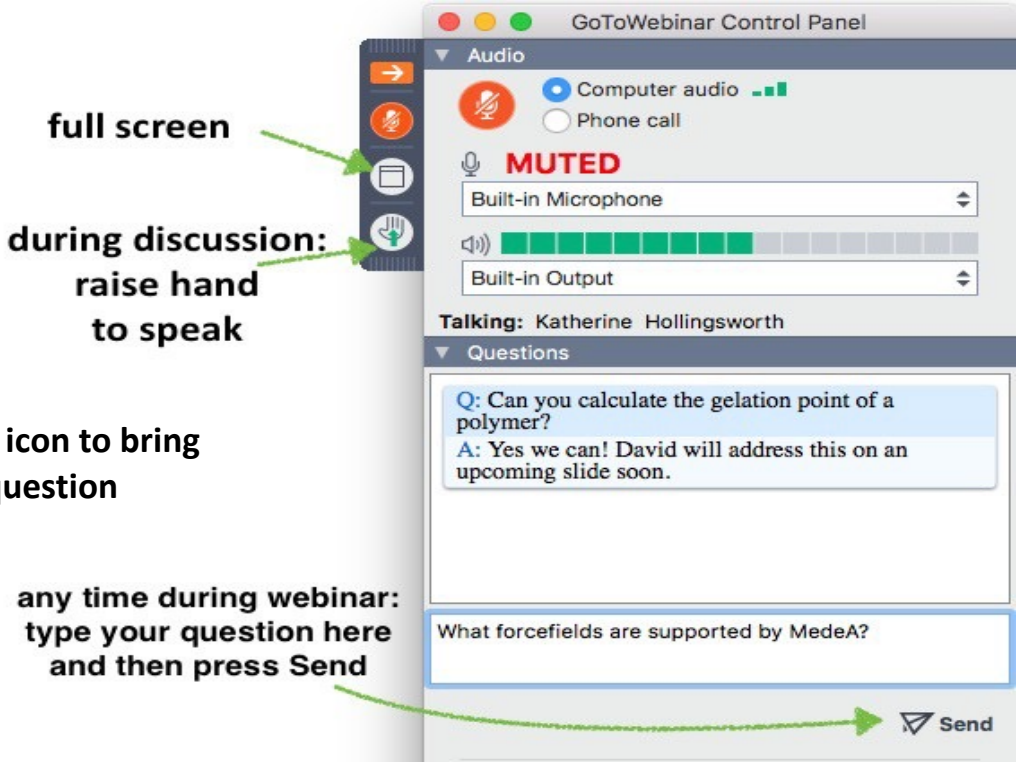
Xiaoli Liu

Shubham Pandey

Garrett Tow

Thomas Nilson

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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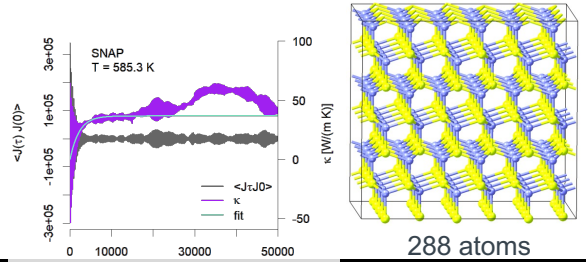


Materials Design 2022 UGM Training Series

Generating and Applying Machine-Learned Potentials with *MedeA*

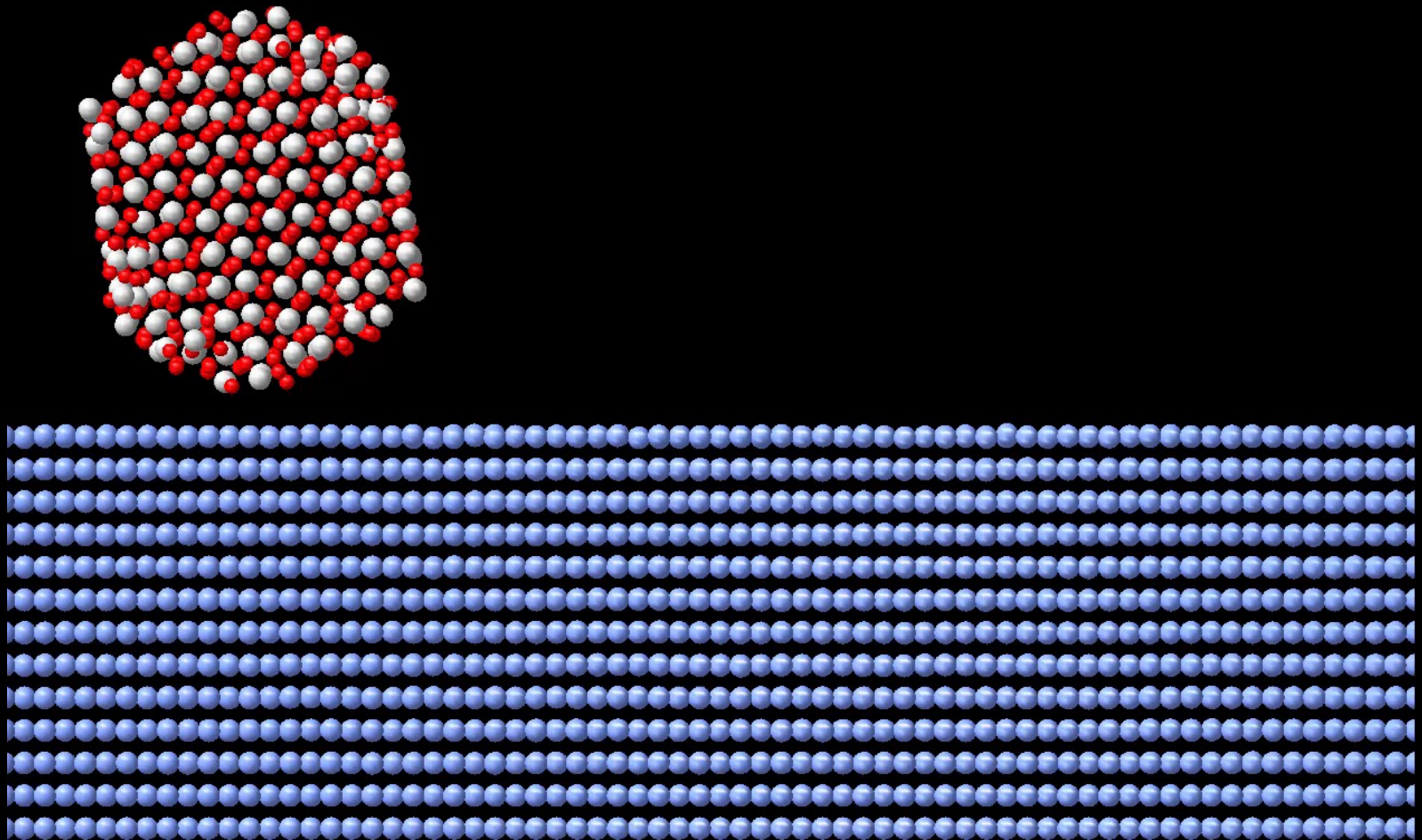
David Reith

Materials Design

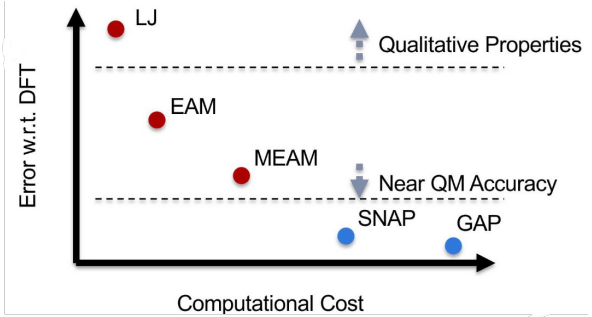
October 13, 2022

Outline

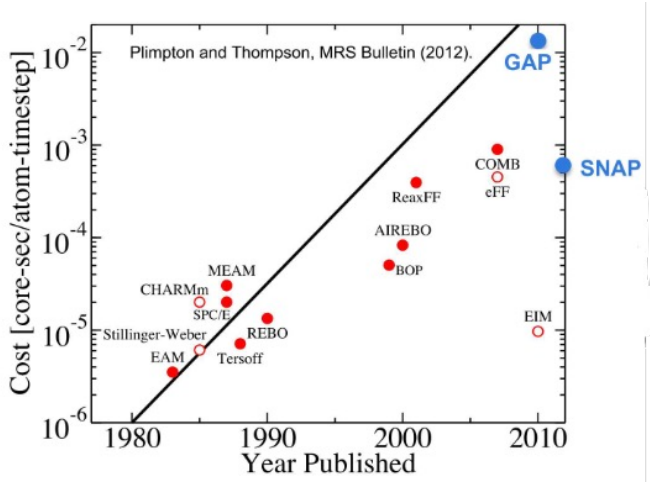
- Overview on machine-learned potentials
- Introduction to machine-learned potential generator
  - Overview on spectral neighbor analysis potentials
  - Hands on
- Neural network potential
  - Neural network potentials theory
  - Behler-Parrinello atomic symmetry functions
  - Hands on
  - Training set convergence
- Δ-learning
  - Basic principle
  - Hands on



Machine-Learned Potentials



- Bridging the time and length scale with VASP accuracy at *LAMMPS* speed
- Build a tailor-made potential
- Reactive by construction
- Generation of an MLP almost automatic from a training set
- Spectral Neighbor Analysis Potentials (SNAP*)
  - Kernel-based method
  - *MedeA* uses FitSNAP to create SNAP potential
- Neural Network Potentials (NNP^{+})
  - Behler-Parrinello neural network method
  - *MedeA* uses n2p2$^{\#}$ to create NNP potentials



* A.P. Thompson *et al.,* J. Comput. Phys. **285**, 316-330 (2015)

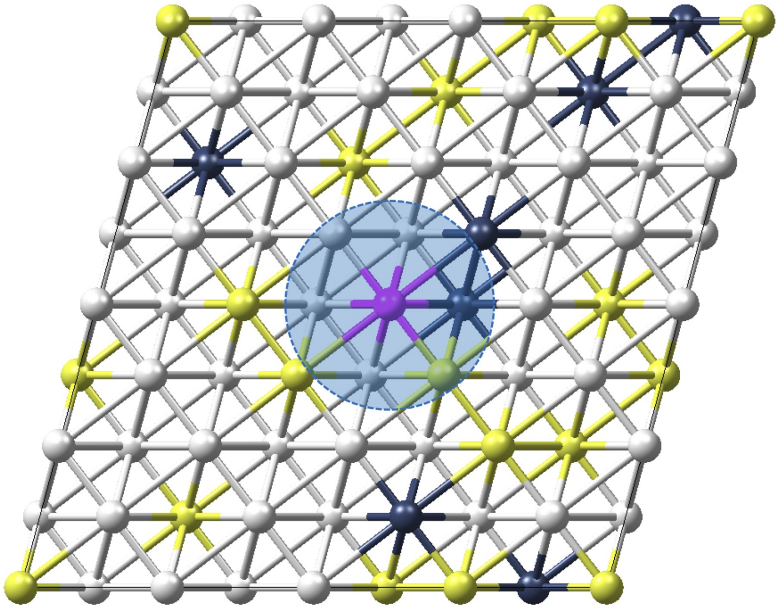
$^{+}$ J. Behler, and M. Parrinello, Phys. Rev. Lett. **98**, 146401 (2007)

$^{\#}$ A. Singraber, T. Morawietz, J. Behler, and C. Dellago, Theory Comput. **15**, 3075-3092 (2019)

Introduction to Machine-Learned Potential Generator

Generate your first SNAP potential with FitSNAP

Spectral Neighbor Analysis Potentials



- Partition structures into local atomic environments
- Local atomic structures described in terms of relative atom positions and types

  $B_i = B_i\,(\{\boldsymbol{r}_{ji}, A_j, j = 1, N_i\}),\ i = 1, N$

  - $\boldsymbol{r}_{ji}$ in local coordinates (r, ϑ, φ), or
  - all pairwise distances r_{jl} in cluster
  - N: all atoms, N_i: all atoms in sphere i

- Local energy centered at atom i

$$E_i = \beta_0 + \Sigma_k \beta_k B_{i,k}$$

- Total energy

$$E = \Sigma_i E_i$$

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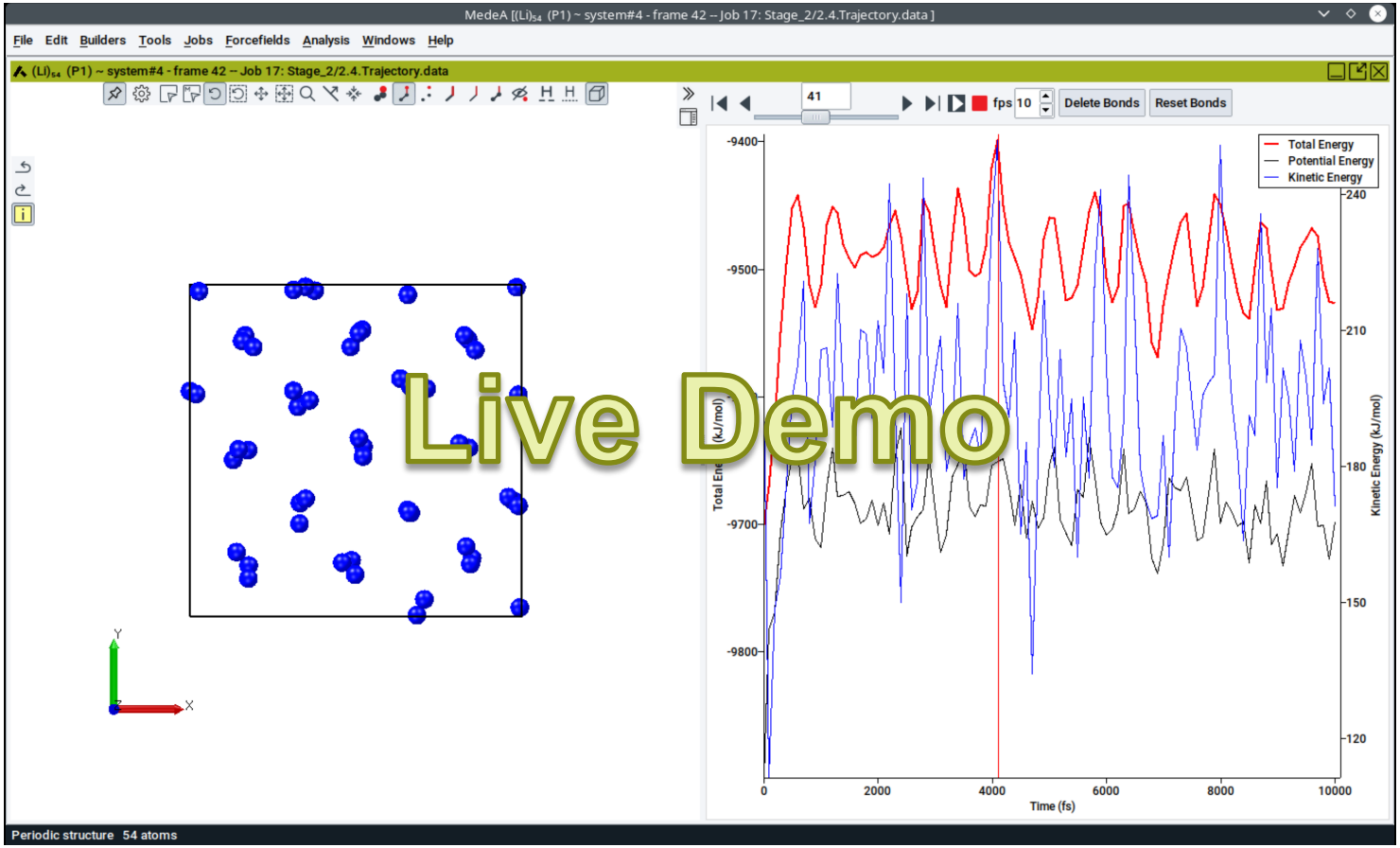

Live Demo

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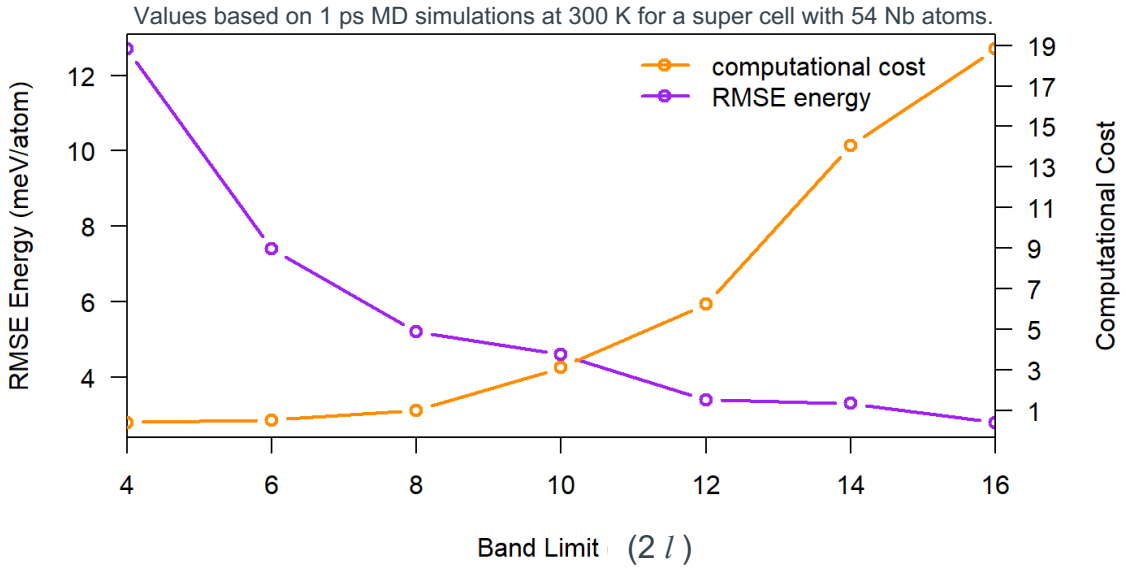
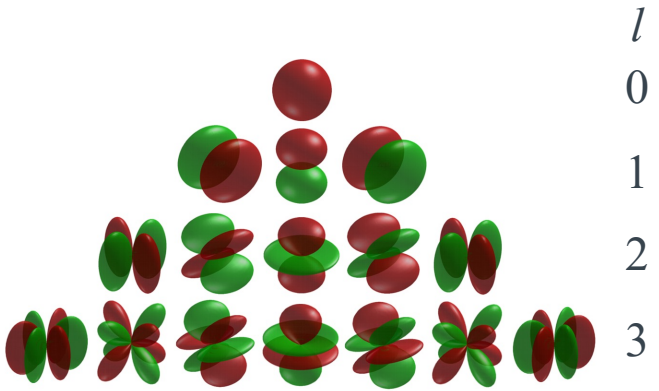
Spectral Neighbor Analysis Potentials:
Band Limit – Cost vs. Accuracy





Spherical harmonics Y_{lm} *(© wikipedia.org)*

- Strong decrease of RMSE with band limit
- Strong increase in computational cost

Band limit

= double maximum angular momentum of spherical harmonics expansion

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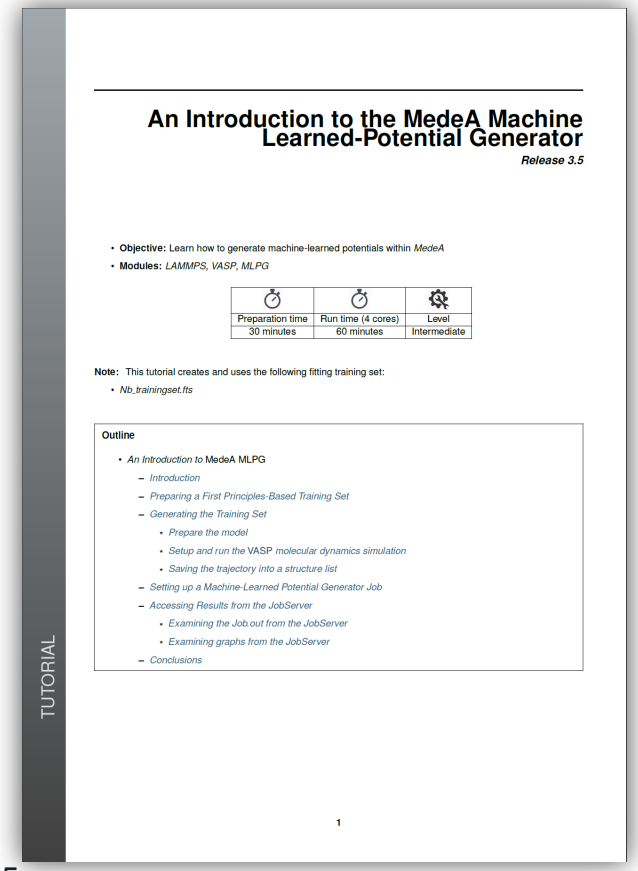
Tutorial: Introduction to *MedeA Machine-Learned Potential Generator*



TUTORIAL

An Introduction to the MedeA Machine Learned-Potential Generator

Release 3.5

- **Objective:** Learn how to generate machine-learned potentials within *MedeA*
- **Modules:** *LAMMPS, VASP, MLPG*

Preparation time	Run time (4 cores)	Level
30 minutes	60 minutes	Intermediate

Note: This tutorial creates and uses the following fitting training set:

- *Nb_trainingset.fts*

Outline

- *An Introduction to* MedeA MLPG
  - *Introduction*
  - *Preparing a First Principles-Based Training Set*
  - *Generating the Training Set*
    - *Prepare the model*
    - *Setup and run the* VASP *molecular dynamics simulation*
    - *Saving the trajectory into a structure list*
  - *Setting up a Machine-Learned Potential Generator Job*
  - *Accessing Results from the JobServer*
    - *Examining the Job.out from the JobServer*
    - *Examining graphs from the JobServer*
  - *Conclusions*

1

- Learn how to generate machine-learned potentials with *MedeA*
  - **Generate** a training set with *MedeA VASP*
  - **Train** a SNAP machine learned potential with *MedeA MLPG*
- Resulting potential can be used like any other classical forcefield by *MedeA LAMMPS*.

- Tutorial available at http://my.materialsdesign.com/tutorials

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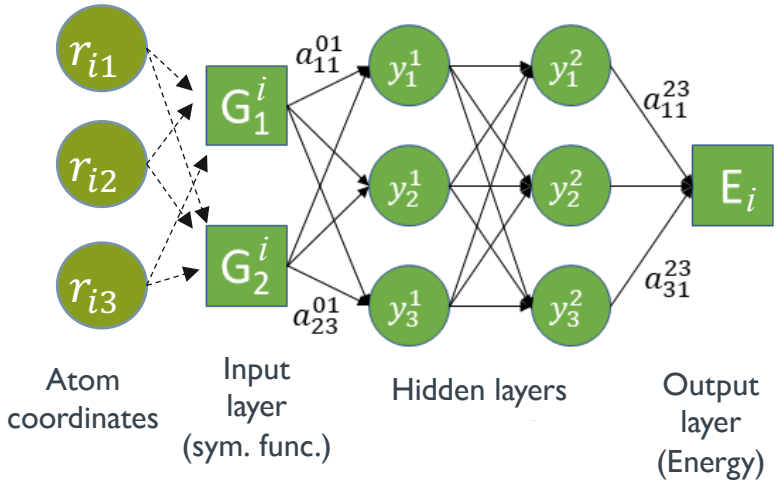
Generating a Neural Network Potential

Create a neural network potential with n2p2

Neural Network Potential



Atomic energy in terms of a hidden layered neural network

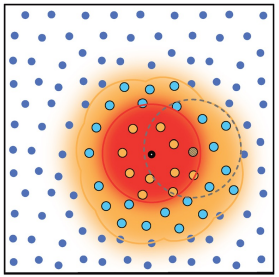
biases weights

$$E_i = f\left(b_1^3 + \sum_{k=1}^{3} a_{k1}^{23} f\left(b_k^2 + \sum_{j=1}^{3} a_{jk}^{12} f\left(b_j^1 + \sum_{i=1}^{2} a_{ij}^{01} G_i\right)\right)\right)$$

activation function: *e.g.*, $\log(1+e^x)$ (softplus function)

$$\rightarrow E = \sum_i E_i$$

- Atomic coordinates not translation/rotation invariant
- Local environment described by "atomic symmetry functions" G
  - Invariant to translation and rotation operations



$$F_{i,\alpha} = \frac{\partial E}{\partial x_{i,\alpha}} = \sum_{m=1}^{N_{loc}} \frac{\partial E_m}{\partial x_{i,\alpha}} = \sum_{m=1}^{N_{loc}} \sum_{k=1}^{N_{SF}} \frac{\partial E_m}{\partial G_k} \frac{\partial G_k}{\partial x_{i,\alpha}}$$

$\alpha = 1,2,3$

J Behler, *Chem. Rev.* 2021, 121, 10037−10072

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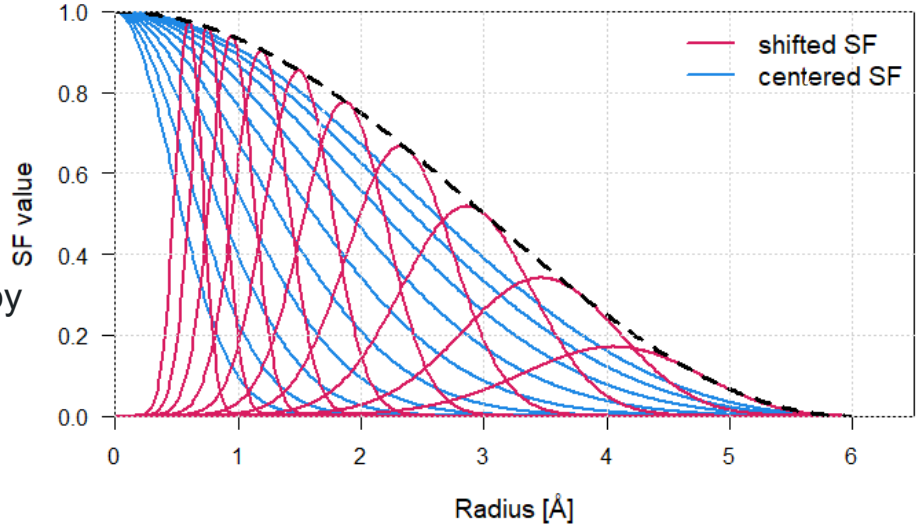
Neural Network Potential
Radial Symmetry Functions

smooth cutoff function

$$G_i^{\mathrm{rad}}(r_C, r_s) = \sum_{j \neq i} f^{\mathrm{rad}}(r_{ij}, r_s)\ f_C(r_{ij}, r_c)$$

Gaussian function $e^{-\eta(r_{ij}-r_s)^2}$



MedeA: symmetry functions on a grid

- For radial symmetry functions use the method by Imbalzano *et al.**
  - $r_s = 0 \ \rightarrow \eta_m = \left(\frac{n_r^{\frac{m}{n_r}}}{r_C}\right)^2$
  - $r_s \neq 0 \ \rightarrow r_{s,m} = \frac{r_C}{n_r^{\left(\frac{m}{n_r}\right)}}$ and $\eta_{s,m} = \frac{1}{\left(r_{s,n_r-m} - r_{s,n_r-m-1}\right)^2}$
  - $m = 0,1,\ldots,n_r$
- Default setting in *MedeA*: shifted radial symmetry functions with $n_r = 10$ and $r_C = 6$ Å

[*] G. Imbalzano *et al.*, J. Chem. Phys. **148**, 241730 (2018)

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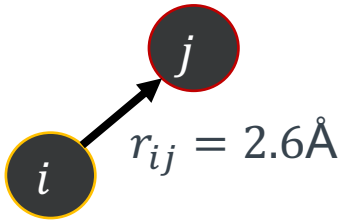
Neural Network Potential
Radial Symmetry Functions

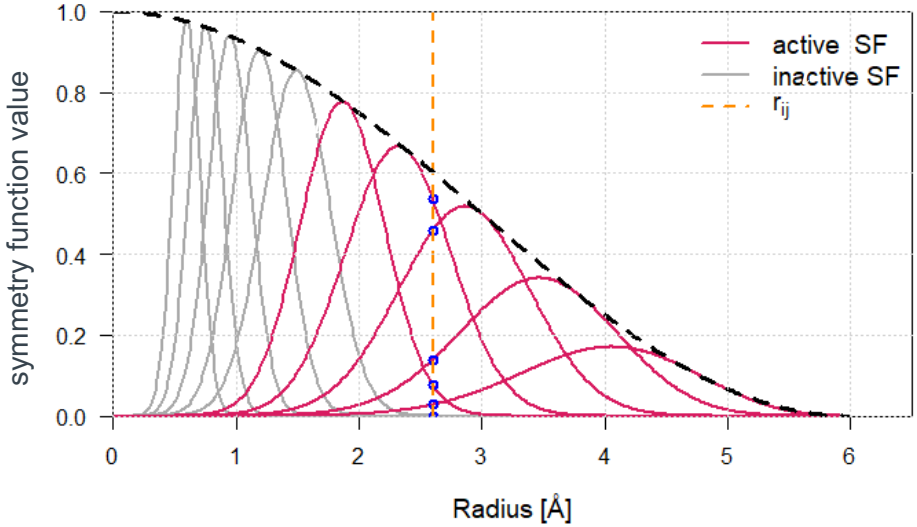
smooth cutoff function

$$G_i^{\mathrm{rad}}(r_C, r_s) = \sum_{j \neq i} f^{\mathrm{rad}}(r_{ij}, r_s)\ f_C(r_{ij}, r_c)$$

Gaussian function $e^{-\eta(r_{ij}-r_s)^2}$

Converting a radial distance between atom i and j into signals





- Not all symmetry functions are activated

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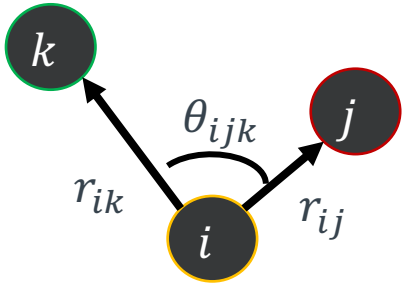
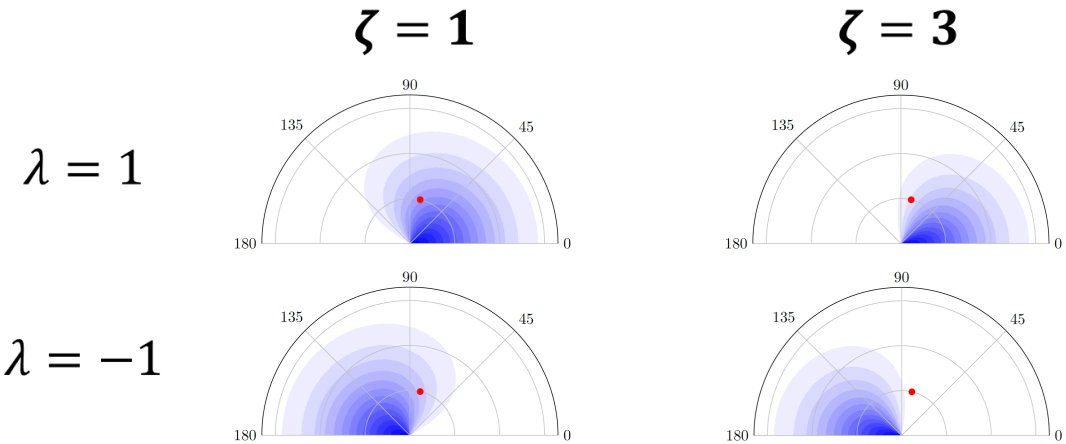
Neural Network Potential
Angular Symmetry Functions

angular function $\left(1+\lambda\cos\theta_{jik}\right)^{\zeta}$

$$G_i^{\mathrm{ang,w}}(r_C,r_s,\lambda,\zeta)=2^{1-\zeta}\sum_{j\neq i,k>j} f^{\mathrm{rad}}(r_{ij},r_s)\, f^{\mathrm{rad}}(r_{ik},r_s) f^{ang}(\theta_{ijk},\lambda,\zeta) f_C(r_{ij},r_c) f_C(r_{ik},r_c)$$





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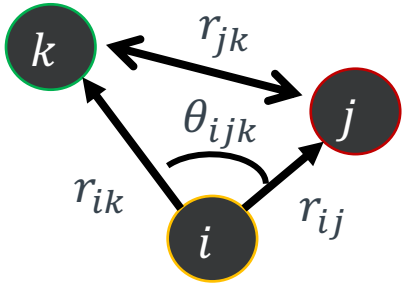
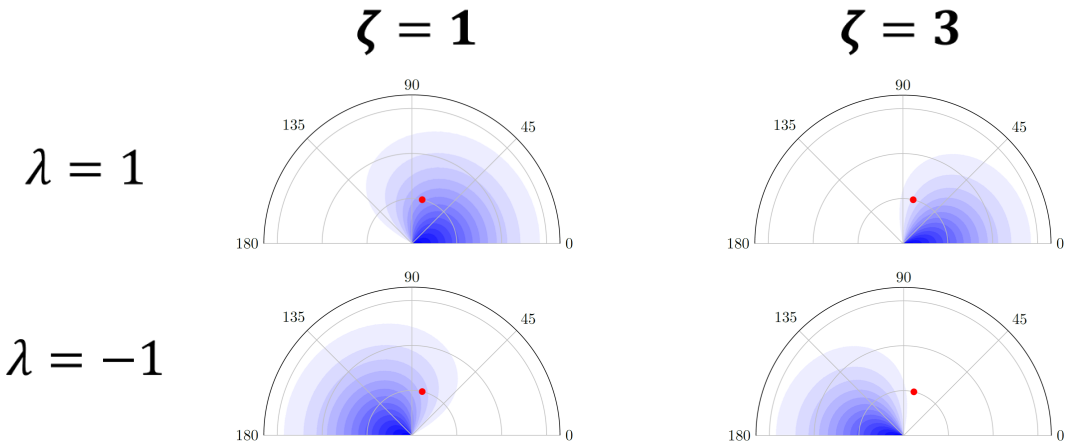
Neural Network Potential
Angular Symmetry Functions

$$G_i^{\mathrm{ang,n}}(r_C, r_s, \lambda, \zeta) = 2^{1-\zeta} \sum_{j \neq i, k>j} f^{\mathrm{rad}}(r_{ij}, r_s)\ f^{\mathrm{rad}}(r_{ik}, r_s) f^{\mathrm{rad}}(r_{jk}, r_s) f^{ang}(\theta_{ijk}, \lambda, \zeta) f_C(r_{ij}, r_c) f_C(r_{ik}, r_c)\ f_C(r_{jk}, r_c)$$

narrow angular symmetry function contain a third radial term for r_{jk}





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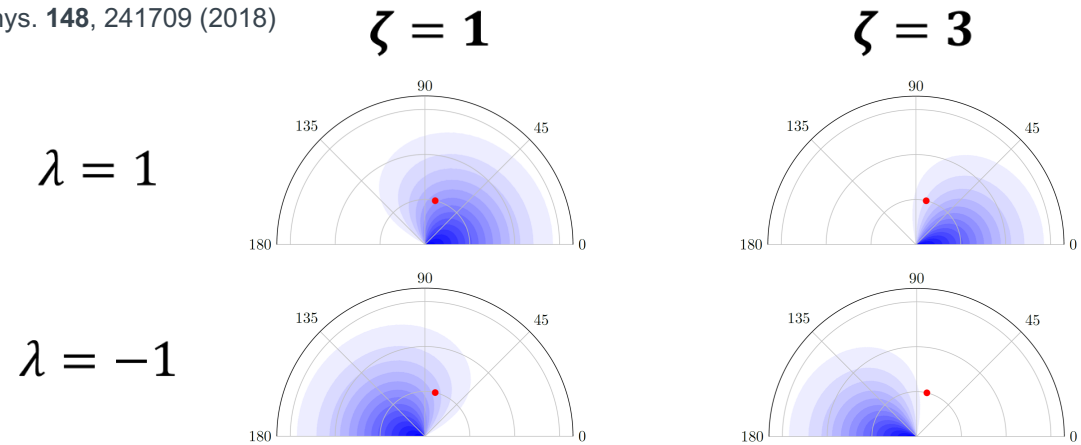
Neural Network Potential Angular Symmetry Functions

- Angular symmetry functions use method by Gastegger *et al.*+
  - $r_s = 0 \rightarrow \eta_i = \frac{1}{2(r_{low}+i\Delta r)^2}$ with $\Delta r = \frac{r_P - r_{low}}{n_a - 1}$ and $r_p = (r_C - 0.5)$
  - $r_s \neq 0 \rightarrow r_{s,i} = r_{low} + i\Delta r$ and $\eta_s = \frac{1}{2\Delta r^2}$ with $r_p = r_C$ (Δr as above)
- Default setting in *MedeA* for narrow angular and wide angular SF: centered, $r_{low} = 1$ Å, $r_C = 6$ Å, $n_a = 4$, $\zeta = 1, 3, 12$, $\lambda = -1, 1$

[+] M. Gastegger *et al.*, J. Chem. Phys. **148**, 241709 (2018)



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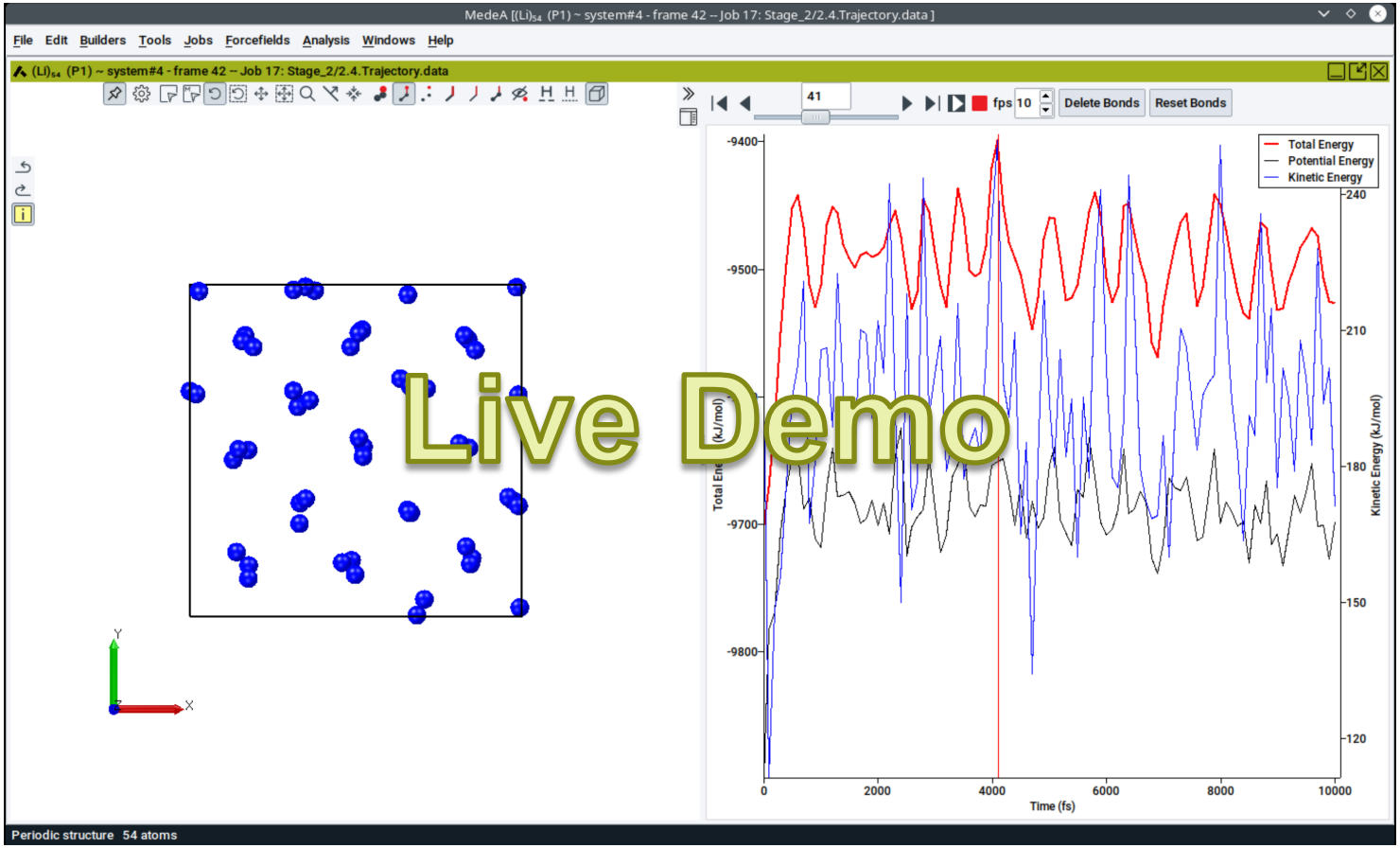

Live Demo

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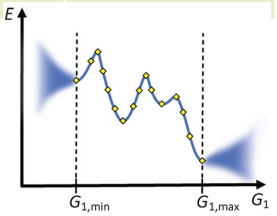
MLPG Training Set Convergence for bcc Li

Warnings: extrapolation warnings (EW) by n2p2 LAMMPS module

Training set composition	# struc.	SNAP RMSE E (meV/at.)	SNAP RMSE F (meV/Å)	NNP RMSE E (meV/at.)	NNP RMSE F (meV/Å)	Warnings per MD steps	
NPT	38	0.1	44	0.0	6	~1000	X
NPT, disNPT	76	0.2	11	0.1	7	0.5 - 10	X
NPT, disNPT, strain	105	0.2	10	0.1	6	0 - 3 / fail	X
NPT, disNPT, strain, NVT	555	0.6	19	0.1	7	0	
NPT, disNPT, strain, NVT, surf NVT	705	1.6	36	0.2	11	0	

NPT: NPT MD flowchart, 8x8x8 Å supercell (2-3 hours)
disNPT: NPT MD flowchart, 6x6x9 Å supercell (2-3 hours)
strain: strain flowchart, isotropic, uniaxial and cell angles, 2x2x2 bcc Li supercell (1 hour)
NVT: VASP MD NVT, 8.0 Å, 7.5 Å, 6.5 Å (3 x 3hours)
surf NVT: VASP MD, (110) Li bcc surface slab



J. Behler, Chem. Rev. **121**, 10037–10072 (2021)

MLPG Training Set Convergence for bcc Li

Warnings: extrapolation warnings (EW) by n2p2 LAMMPS module

Training set composition	# struc.	SNAP		NNP			
		RMSE E (meV/at.)	RMSE F (meV/Å)	RMSE E (meV/at.)	RMSE F (meV/Å)	Warnings per MD steps	
NPT	38	0.1	44	0.0	6	~1000	X
NPT, disNPT	76	0.2	11	0.1	7	0.5 - 10	X
NPT, disNPT, strain	105	0.2	10	0.1	6	0 - 3 / fail	X
NPT, disNPT, strain, NVT	555	0.6	19	0.1	7	0	
NPT, disNPT, strain, NVT, surf NVT	705	1.6	36	0.2	11	0	

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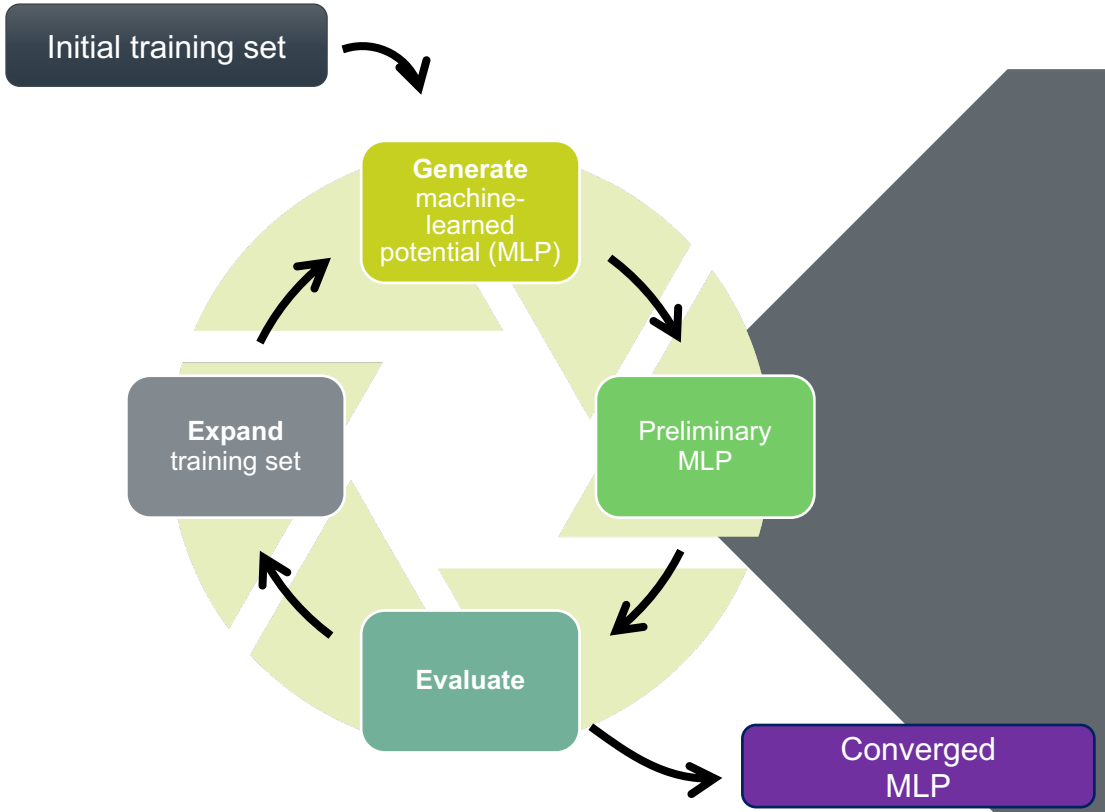
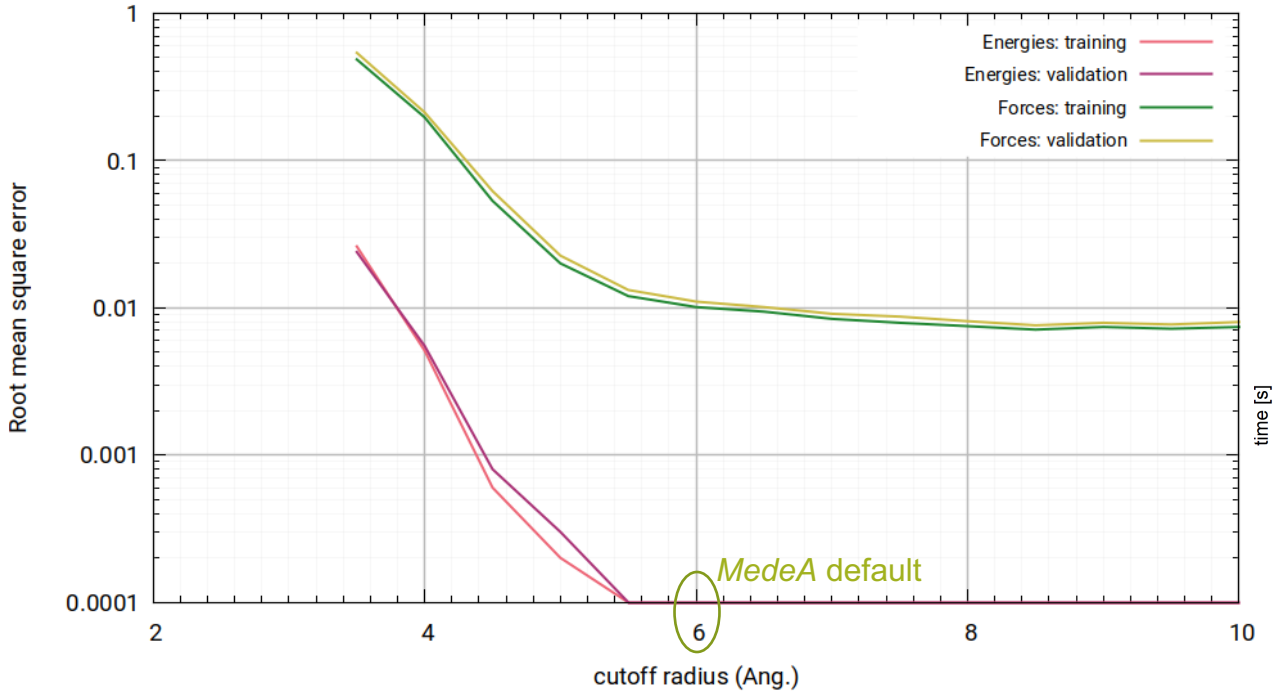
Converging a Machine-Learned Potential



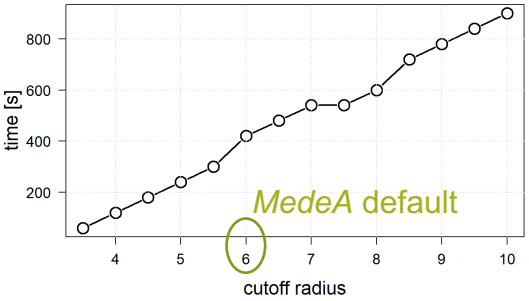
General guidelines

- Always ensure that all entries in the training set have been calculated with consistent VASP settings – *MedeA* will help you (Flowcharts, VASP GUI: “Restore from Job”)
  - PAW potentials
  - Functional
  - K-spacing
  - Planewave cutoff energy
- Training set: include ab initio MD simulations at temperatures higher than the temperature range to which the MLP should be applied
- Create the MLP for the region in phase space that is of interest

Neural Network Potential
Radial Cutoff RMSE Convergence



fitting computational cost



fit performed on full Li training set

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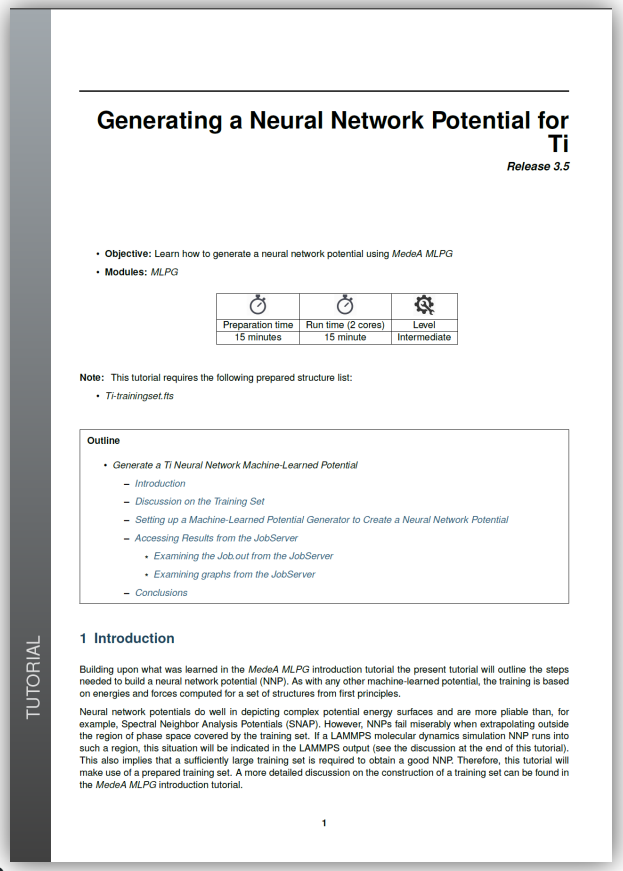
Tutorial: Generating a Neural Network Potential for Ti



TUTORIAL

Generating a Neural Network Potential for Ti

Release 3.5

- **Objective:** Learn how to generate a neural network potential using *MedeA MLPG*
- **Modules:** *MLPG*

Preparation time	Run time (2 cores)	Level
15 minutes	15 minute	Intermediate

Note: This tutorial requires the following prepared structure list:

- *Ti-trainingset.fts*

Outline

- *Generate a Ti Neural Network Machine-Learned Potential*
  - *Introduction*
  - *Discussion on the Training Set*
  - *Setting up a Machine-Learned Potential Generator to Create a Neural Network Potential*
  - *Accessing Results from the JobServer*
    - *Examining the Job.out from the JobServer*
    - *Examining graphs from the JobServer*
  - *Conclusions*

1 Introduction

Building upon what was learned in the *MedeA MLPG* introduction tutorial the present tutorial will outline the steps needed to build a neural network potential (NNP). As with any other machine-learned potential, the training is based on energies and forces computed for a set of structures from first principles.

Neural network potentials do well in depicting complex potential energy surfaces and are more pliable than, for example, Spectral Neighbor Analysis Potentials (SNAP). However, NNPs fail miserably when extrapolating outside the region of phase space covered by the training set. If a LAMMPS molecular dynamics simulation NNP runs into such a region, this situation will be indicated in the LAMMPS output (see the discussion at the end of this tutorial). This also implies that a sufficiently large training set is required to obtain a good NNP. Therefore, this tutorial will make use of a prepared training set. A more detailed discussion on the construction of a training set can be found in the *MedeA MLPG* introduction tutorial.

1

- Learn how to generate neural network potential for Ti with *MedeA MLPG*
  - **Discussion** on the training set
  - **Setup** *MedeA MLPG* to train a Behler-Parrinello neural network potential
- Resulting potential can be used like any other classical forcefield by *MedeA LAMMPS*.

- Tutorial available at http://my.materialsdesign.com/tutorials

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MLP - Δ-Learning

Upgrade your MLP to a higher level

Δ-Learning
Basic Principle

Large training set: at lower QM level (e.g., PBE)

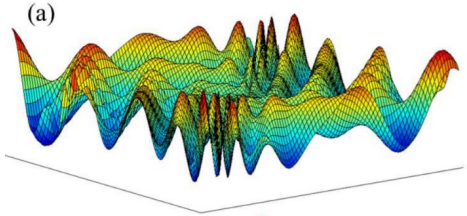
$lT_{QM(L)}$

Small training set: at lower and higher QM levels (e.g., PBE and metaGGA SCAN)

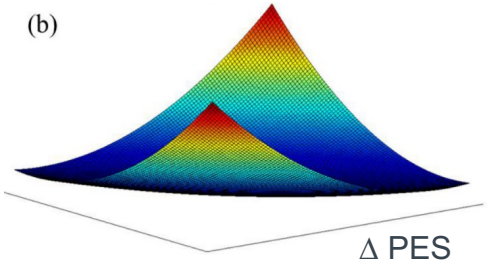
$sT_{QM(L)}$ and $sT_{QM(H)}$

$$\Delta\, MLP = MLP(sT_{QM(H)} - sT_{QM(L)})$$

$$lT_{QM(H)} = \Delta\, MLP\,(\, lT_{QM(L)})$$

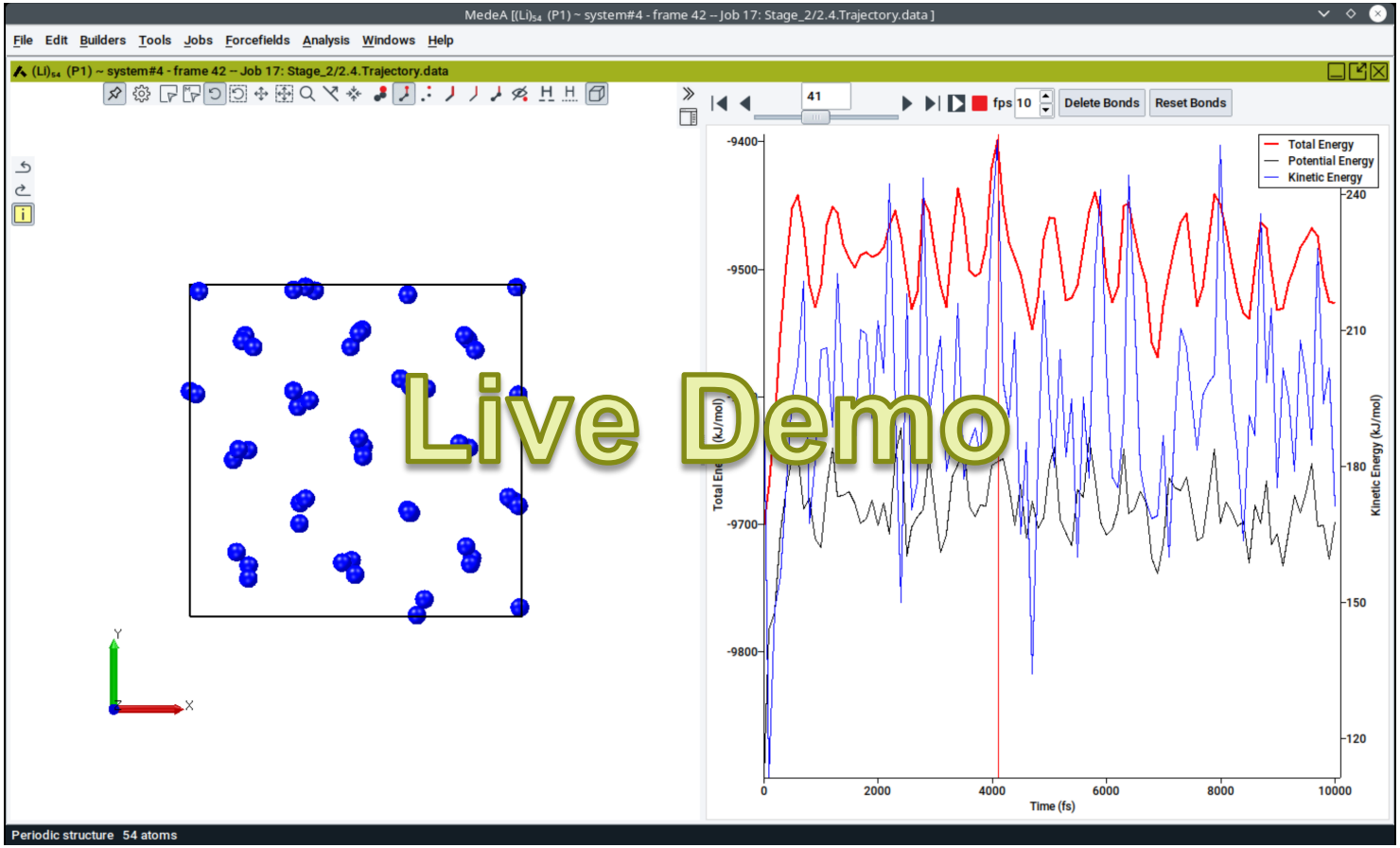


full potential energy surface (PES)



MLFF-Δ(RPA): P. Liu, C. Verdi, F. Karsai, and G. Kresse, PRB 105, L060102 (2022)

Generate final MLP using $lT_{QM(H)}$ as a training set → final MLP based on higher QM level



Live Demo

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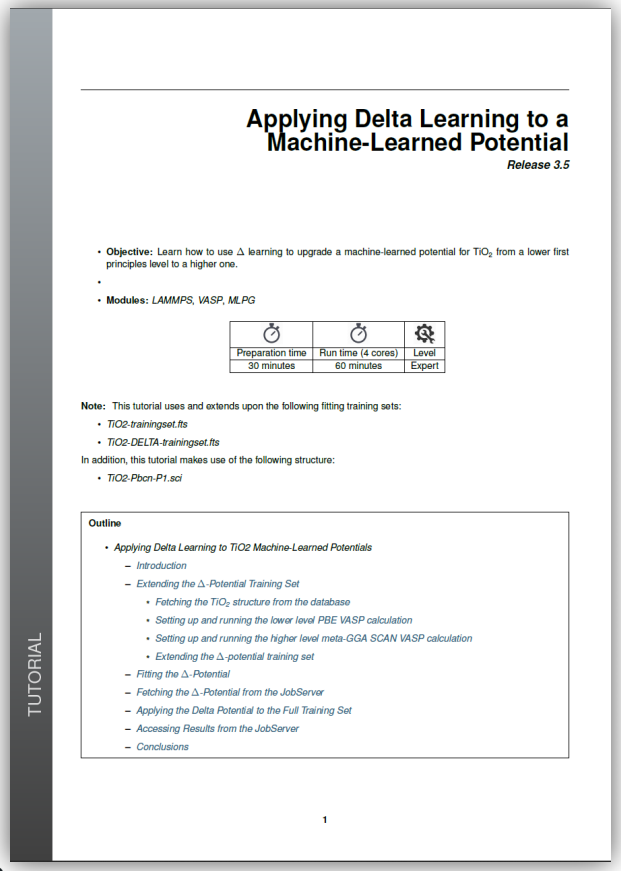
Tutorial: Applying Delta Learning to a Machine-Learned Potential



TUTORIAL

Applying Delta Learning to a Machine-Learned Potential

Release 3.5

- **Objective:** Learn how to use Δ learning to upgrade a machine-learned potential for TiO_2 from a lower first principles level to a higher one.
- **Modules:** *LAMMPS, VASP, MLPG*

Preparation time	Run time (4 cores)	Level
30 minutes	60 minutes	Expert

Note: This tutorial uses and extends upon the following fitting training sets:

- *TiO2-trainingset.fts*
- *TiO2-DELTA-trainingset.fts*

In addition, this tutorial makes use of the following structure:

- *TiO2-Pbcn-P1.sci*

Outline

- *Applying Delta Learning to TiO2 Machine-Learned Potentials*
  - *Introduction*
  - *Extending the Δ-Potential Training Set*
    - *Fetching the TiO_2 structure from the database*
    - *Setting up and running the lower level PBE VASP calculation*
    - *Setting up and running the higher level meta-GGA SCAN VASP calculation*
    - *Extending the Δ-potential training set*
  - *Fitting the Δ-Potential*
  - *Fetching the Δ-Potential from the JobServer*
  - *Applying the Delta Potential to the Full Training Set*
  - *Accessing Results from the JobServer*
  - *Conclusions*

1

- Learn how to use Δ-learning to upgrade a machine-learned potential from a lower first principles level to a higher one
  - **Generate** a Δ-potential training set
  - **Setup** *MedeA MLPG* to train a Δ-potential
  - **Apply** the Δ-potential to a full training set and generate a higher level machine-learned potential

- Tutorial available at http://my.materialsdesign.com/tutorials

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Applications of 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

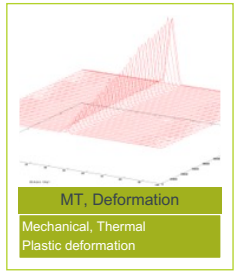
***MedeA* Viscosity:** Calculate viscosity with Green-Kubo or non-equilibrium MD Müller-Plathe

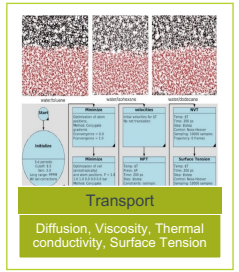
***MedeA* Surface Tension:** Calculate surface tension of fluid slabs

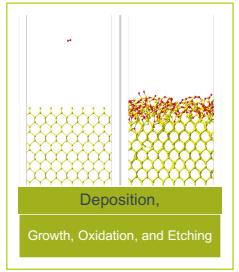
***MedeA* Diffusion:** Automatically calculate diffusivity from mean square displacement

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

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







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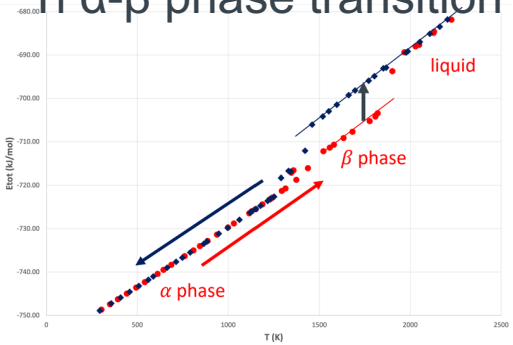
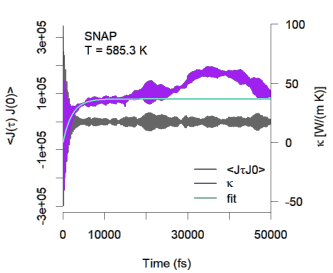
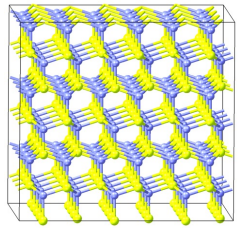
Some Examples

Ti α-β phase transition



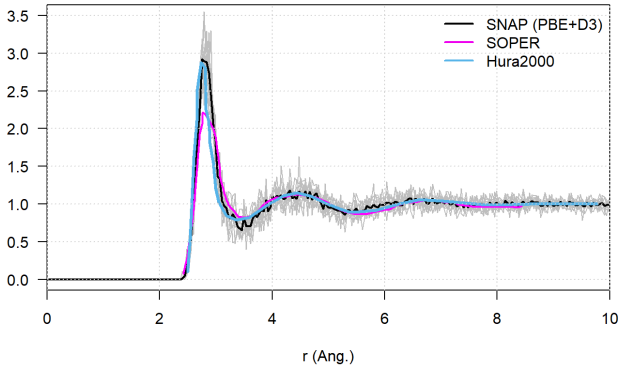
wurtzite Al-N





288 atoms

water



- 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°

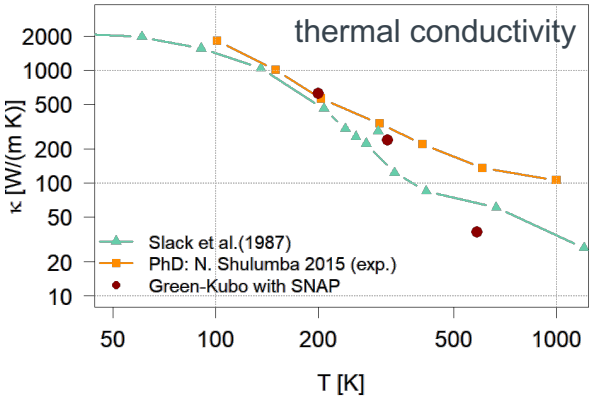
Transition temperatures (exp):
- α-β transition: 1156 K
- melting: 1946 K

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)

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



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Question and Answer Session

Dr. René Windiks

Materials Design

Dr. David Reith

Materials Design

Announcements

ugm.materialsdesign.com

Next MedeA Training

Thursday, October 20th

Next Week's Plenary Session

Tuesday, October 18th

Dr. Shubham Pandey

Materials Design

Dr. Xiaoli Liu

Materials Design

Dr. Carla Verdi

University of Vienna

Professor Georg Kresse

University of Vienna

Questions about Materials Design UGM

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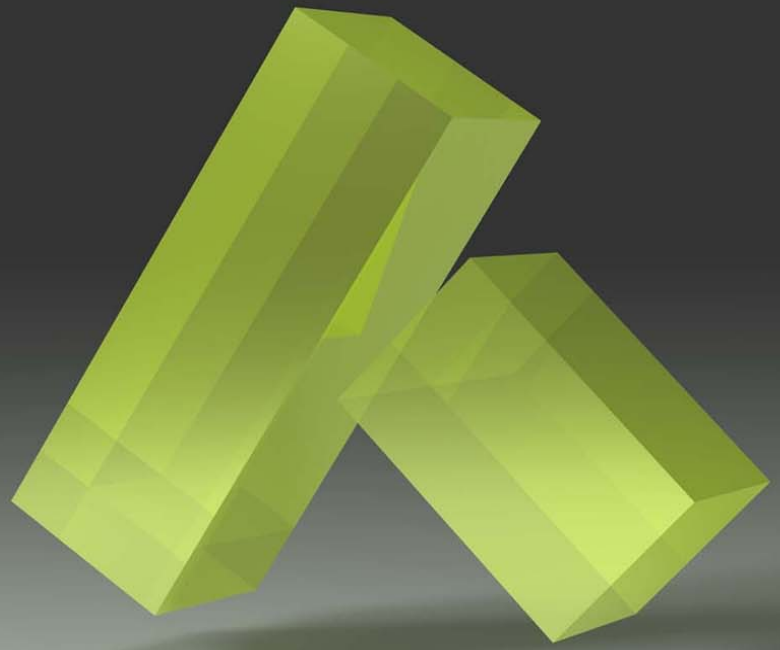

MedeA

Innovation by Simulation