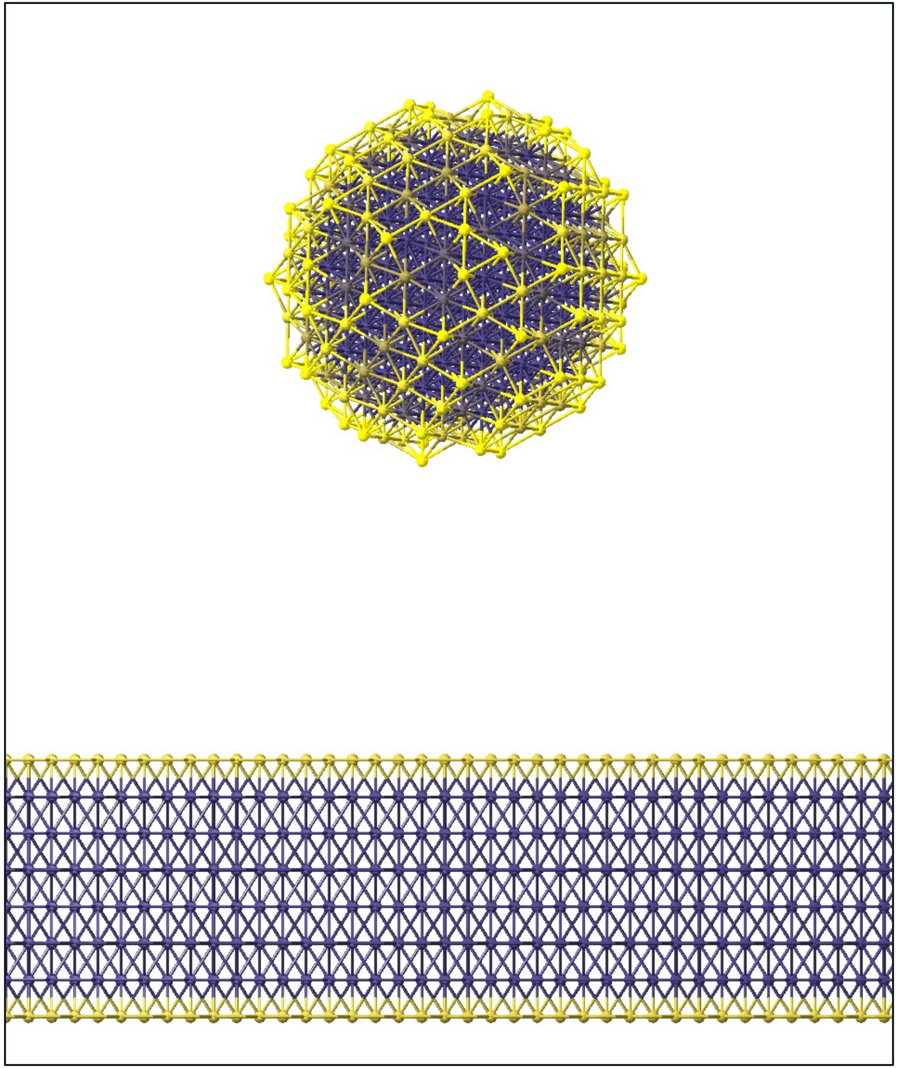
materials design

Machine-Learned Potentials: Surpassing the Limits of the *ab initio* World without leaving it behind



August 1-3, 2023

Materials Design Webinar Series

- Each session runs several times to accommodate schedules
  - Share the webinar series with your colleagues!
  - Registration details http://www.materialsdesign.com/webinars
- We will be recording this webinar
  - Watch any of our earlier webinars anytime
  - We will post upcoming webinars on the webinar page
- Vote for the next webinar topic!
  - Take a 2 minutes brief survey at the end of the webinar!

materials design

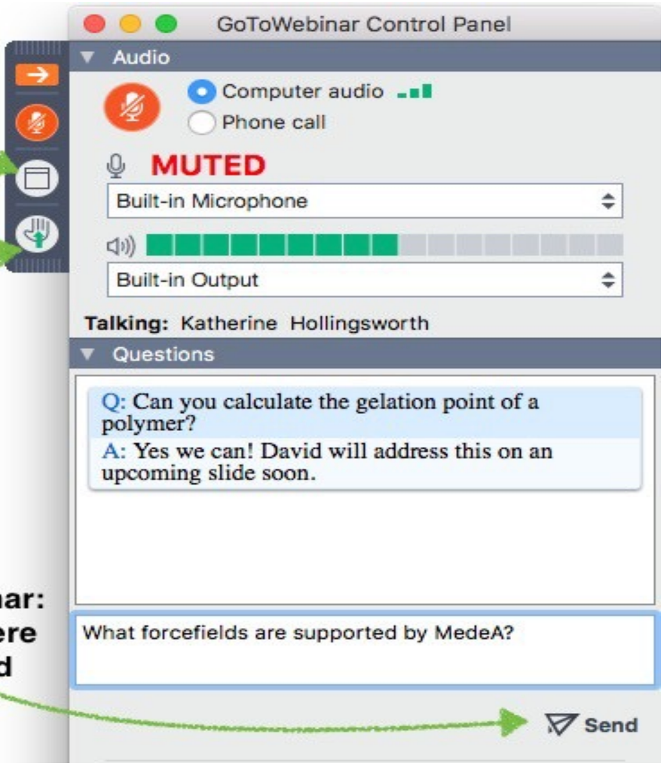
Please Ask Questions!

full screen

during discussion: raise hand to speak

Use the raise hand icon to bring attention to your question

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



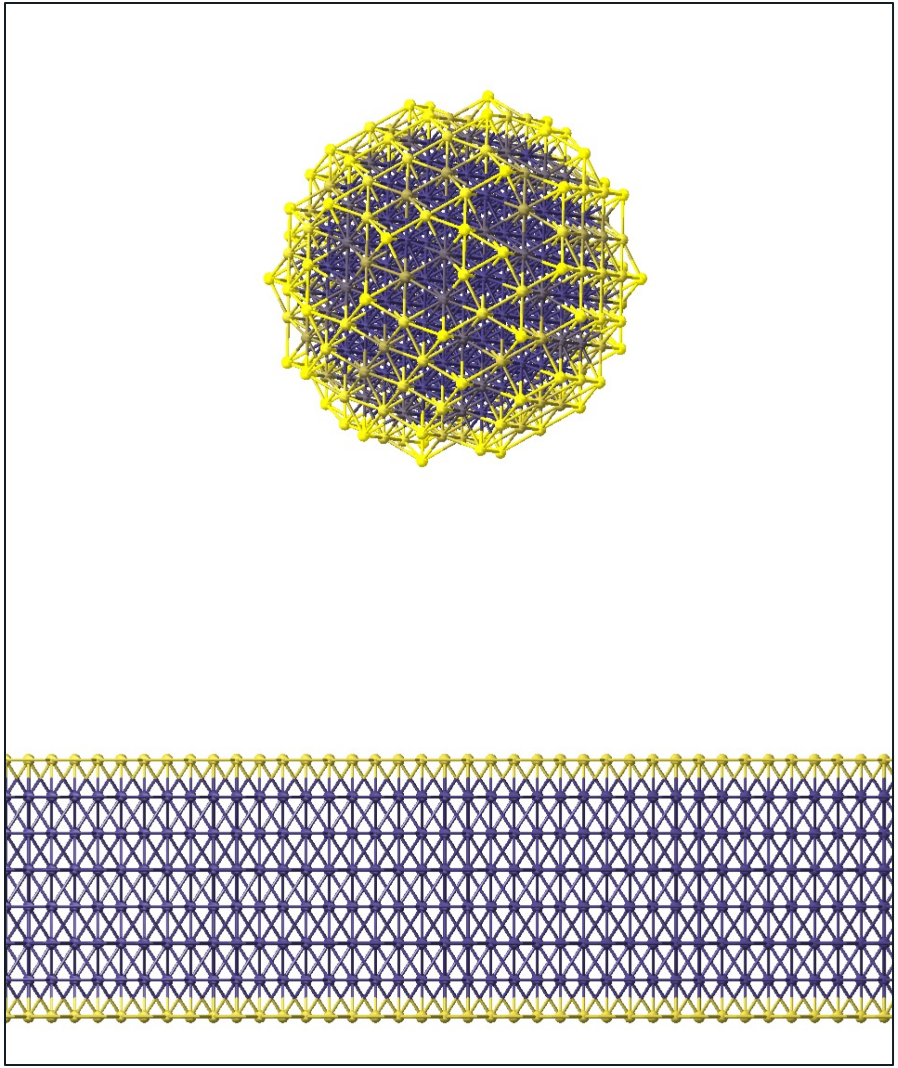
Webinar Speakers

Katherine Hollingsworth *Presenters: Dr. Xiaoli Liu* *Dr. Volker Eyert*

materials design

Machine-Learned Potentials: Surpassing the Limits of the *ab initio* World without leaving it behind

August 1-3, 2023

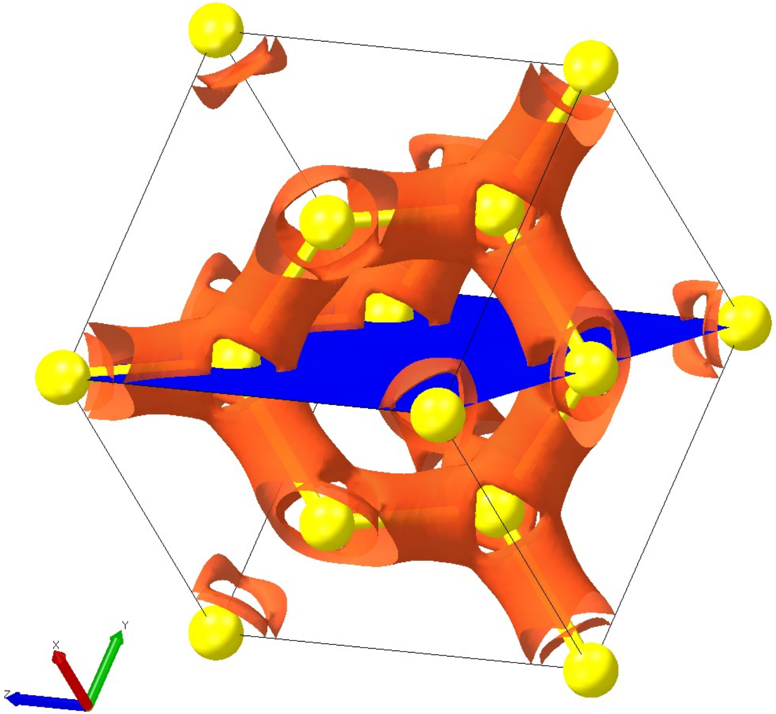


Agenda

- What is the value of Machine-Learned Potentials (MLPs)?
- How can we generate MLPs using *MedeA?*
  - The *MedeA* Machine-Learned Potential Generator
- How do MLPs work? A quick view behind the scenes
- What kind of problems can be solved with MLPs?
  - α-β Phase Transition of Ti
  - Impact of Ti Nanoclusters on Ti Surfaces
  - Metal-Insulator Transition of VO_2
  - Green Electronics: Diffusion of Ni into Si

materials design

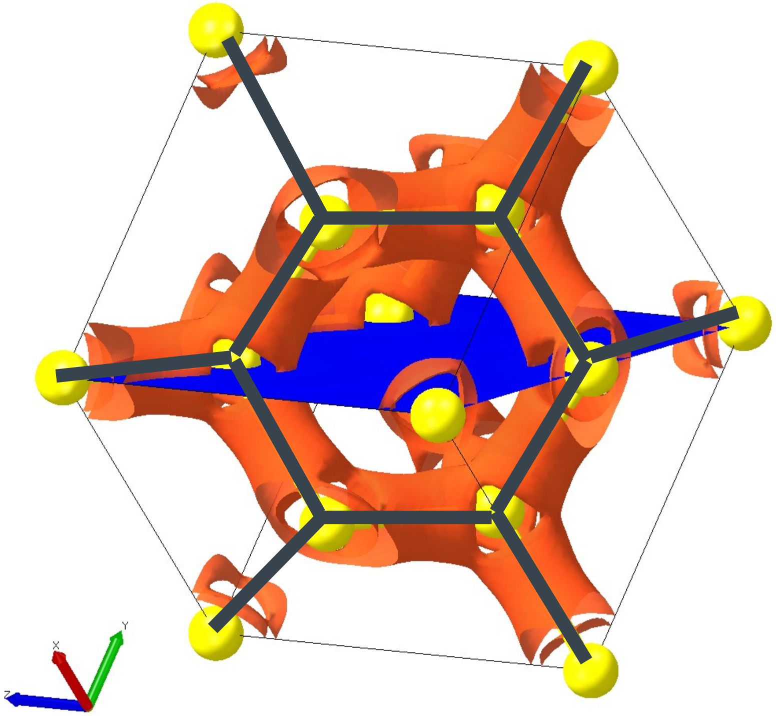
From Nuclei and Electrons to Interatomic Potentials



Si: total valence density

- The behavior of materials is determined by the motion and Coulomb interaction of nuclei and electrons.
- DFT provides a most efficient description of matter in terms of the electron density.

From Nuclei and Electrons to Interatomic Potentials



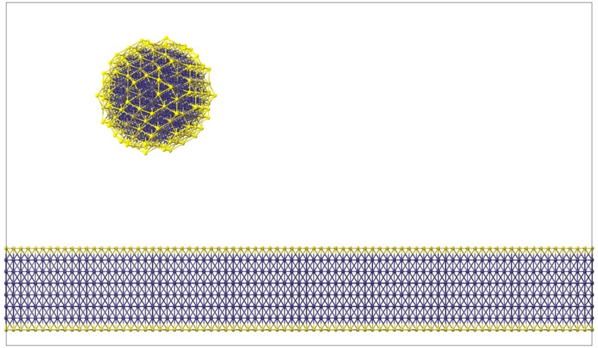
Si: total valence density

“The Taming of the Glue”

- The behavior of materials is determined by the motion and Coulomb interaction of nuclei and electrons.
- DFT provides a most efficient description of matter in terms of the electron density.
- Simulation of millions of atoms and millions of configurations requires replacing the glue of the electron distribution by effective interatomic interactions.
- Machine learning offers the systematic generation of highly efficient yet accurate interatomic potentials from DFT training sets.

Titanium

Phase Stability and Surface-Nanoparticle Interactions



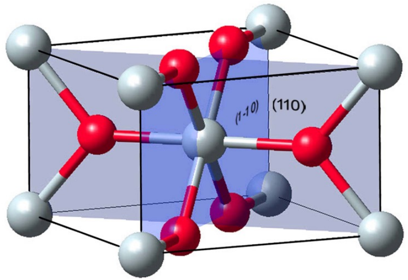
Why Titanium?

- Lightweight, high corrosion resistance, high strength-to-density ratio, biocompatible
- Strong lightweight alloys with Al, V, Fe, and Mo for applications in
  - aerospace and marine
  - automotive
  - orthopedic implants (compatible with MRI)
  - dental instruments and dental implants
  - sporting goods, jewelry, mobile phones
- Oxides: TiO_2 (anatase, brookite, rutile)
  - pigments, additives, coatings
- TiN: hardness, electrical/thermal conductivity
- TiC: hardness

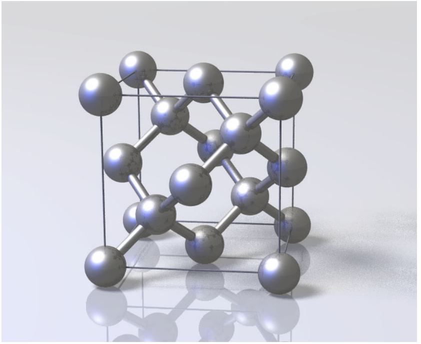
https://www.visualcapitalist.com/sp/titanium-the-metal-of-the-future/

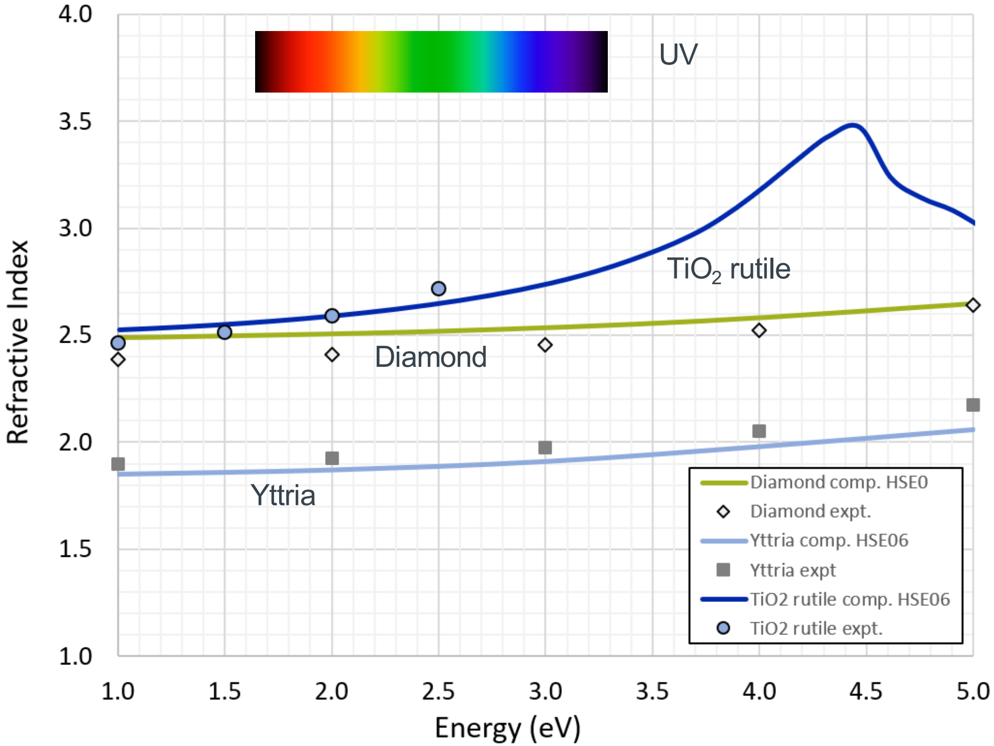
materials design

High UV Refractive Index of Rutile TiO_2



TiO_2 rutile





See: The Color of Materials (Webinar 2019)

materials design

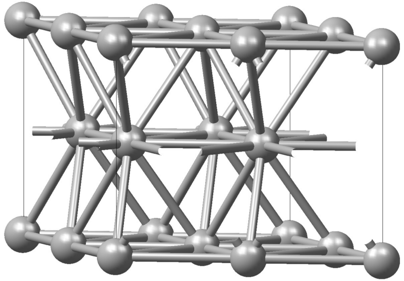
Training Set Calculations

Training set structures

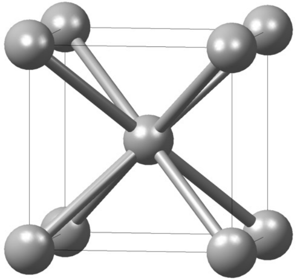
- Initial structures, supercells
- Vacancies, self-interstitial atoms, surfaces, stacking faults
- Isotropic and uniaxial strain up to ±6%
- Angular deformations up to ±2°
- NPT/NVT MD simulations at 300-1700 K
- Total of 1005 structures

VASP computational parameters

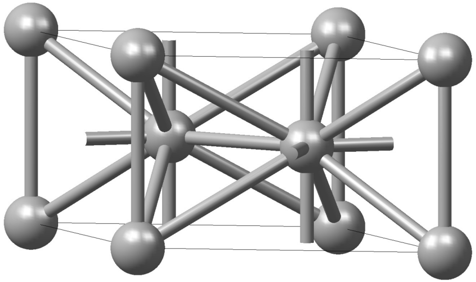
- PBE functional
- 520 eV plane-wave cutoff
- **k**-point spacing 0.2 Å^{-1}
- Gaussian smearing, σ = 0.05 eV



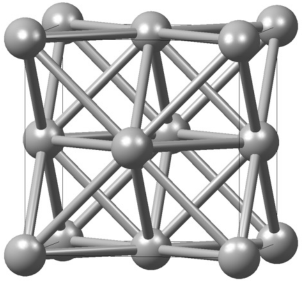
α-phase, hcp
3×3×2 supercell



β-phase, bcc
3×3×3 supercell



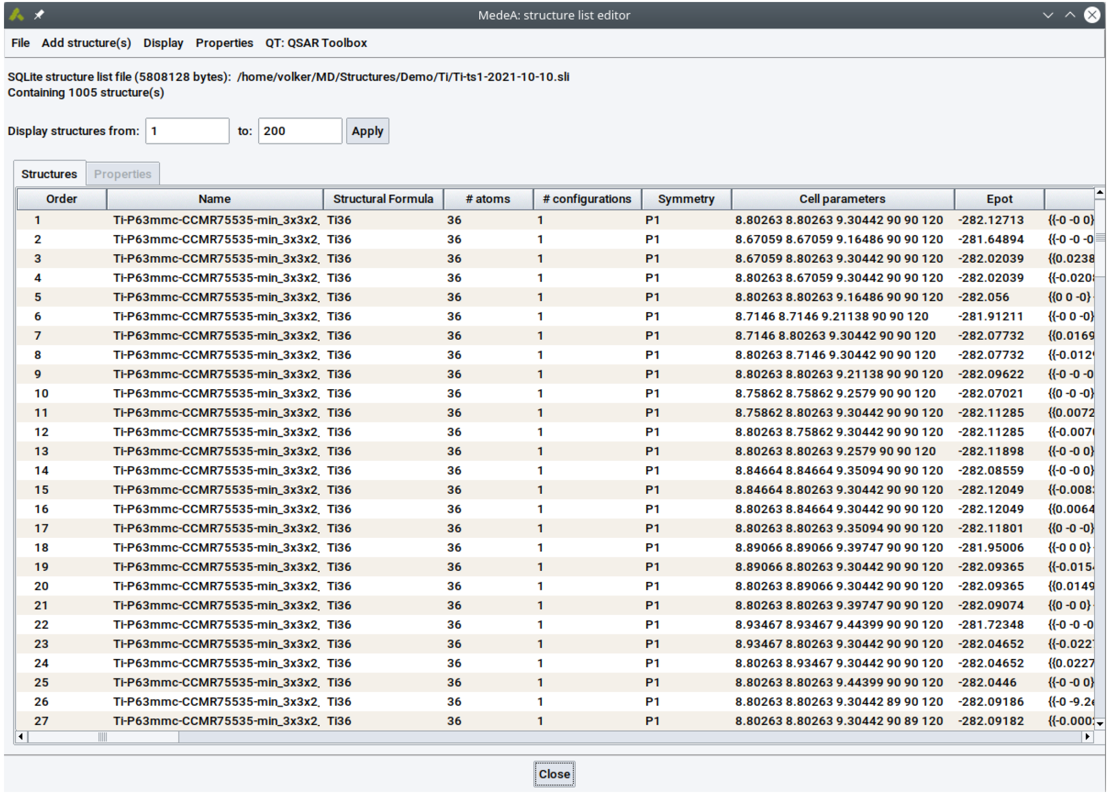
ω-phase, hex
2×2×3 supercell



fcc
2×2×2 supercell

materials design

MedeA MLP Generator: Prepare



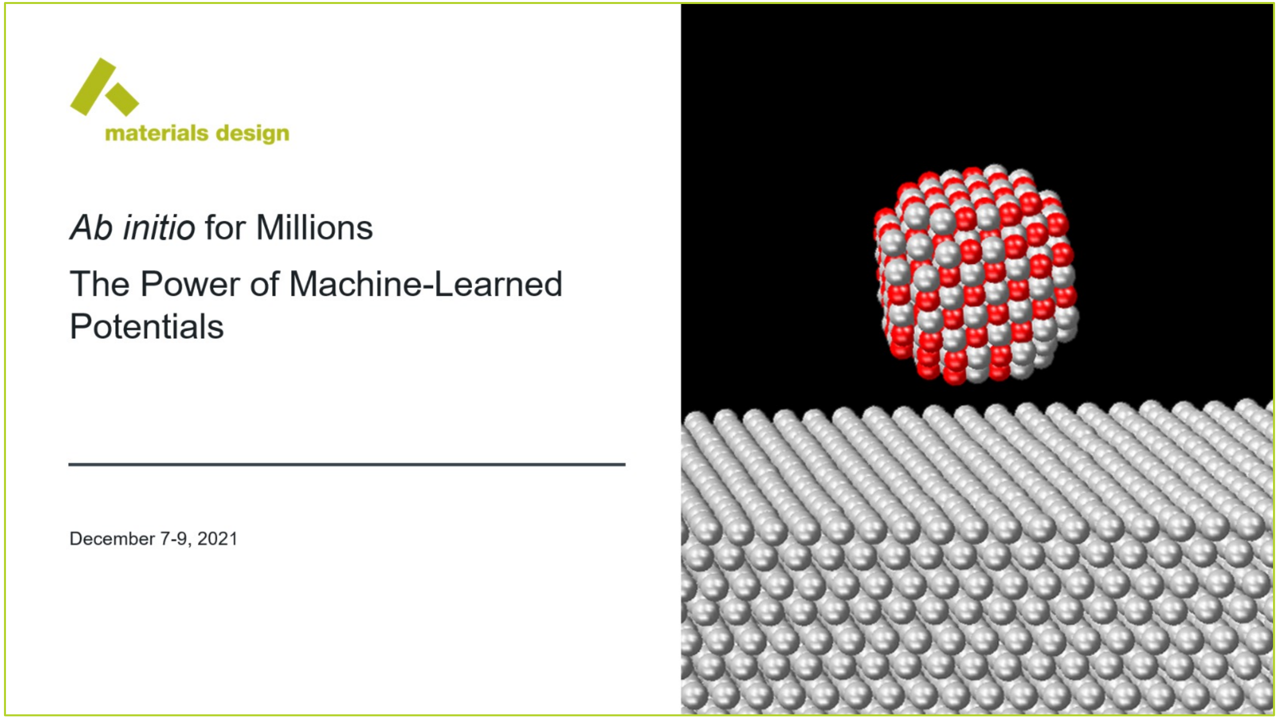
Collect information about training set in structure list

- Structures
- Energies
- Forces
- Stresses

materials design

MedeA Machine-Learned Potential Generator (MLPG)

For details see:



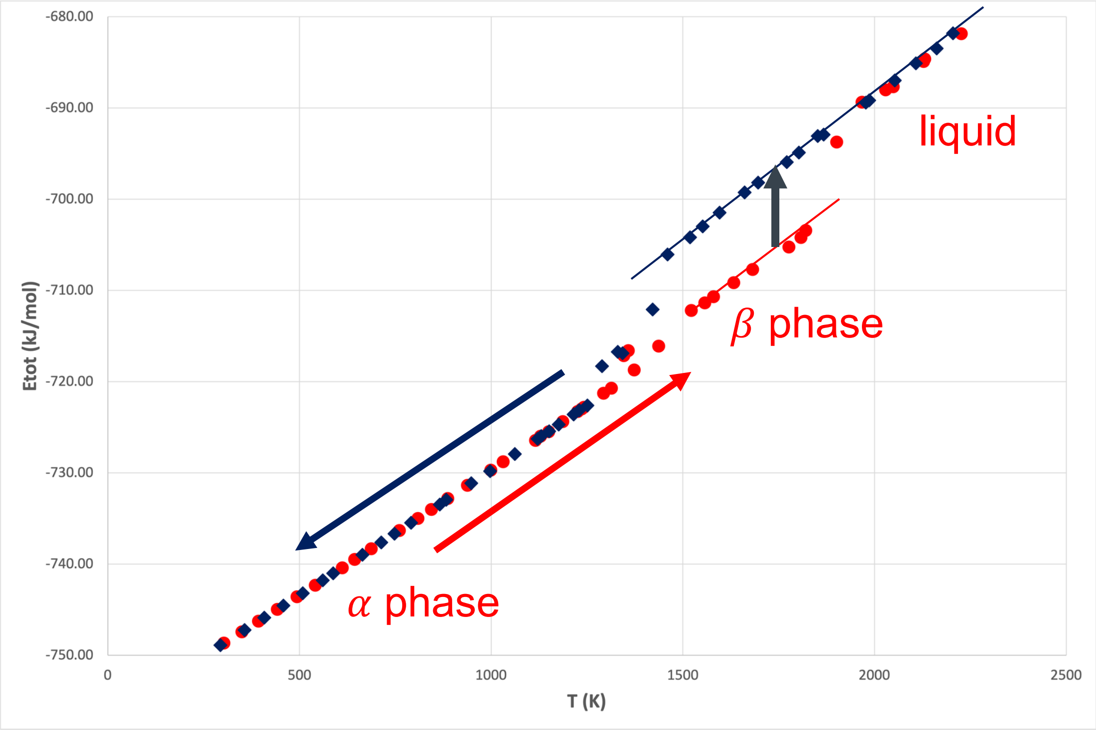
α-Ti: Defect Properties

	E_{vac} (eV)	E_{SIA} (eV)	E_{SF} (meV/Å^2)	$E_{bcc\text{-}hcp}$ (eV)	$E_{fcc\text{-}hcp}$ (eV)
Exp.			21.0		
DFT	2.05	2.55	20.7	0.108	0.056
MLP	2.02	2.17	20.4	0.129	0.060

DFT: *MedeA* VASP, Exp.: B. Zhao *et al.*, Nature Sci. Rep. **10**, 3086 (2020)

- Lattice parameters, densities, defect formation energies, phase stabilities, and elastic constants and moduli calculated using MLPs are in very good agreement with the respective results obtained from VASP.

Ti: α-β Phase Transition



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

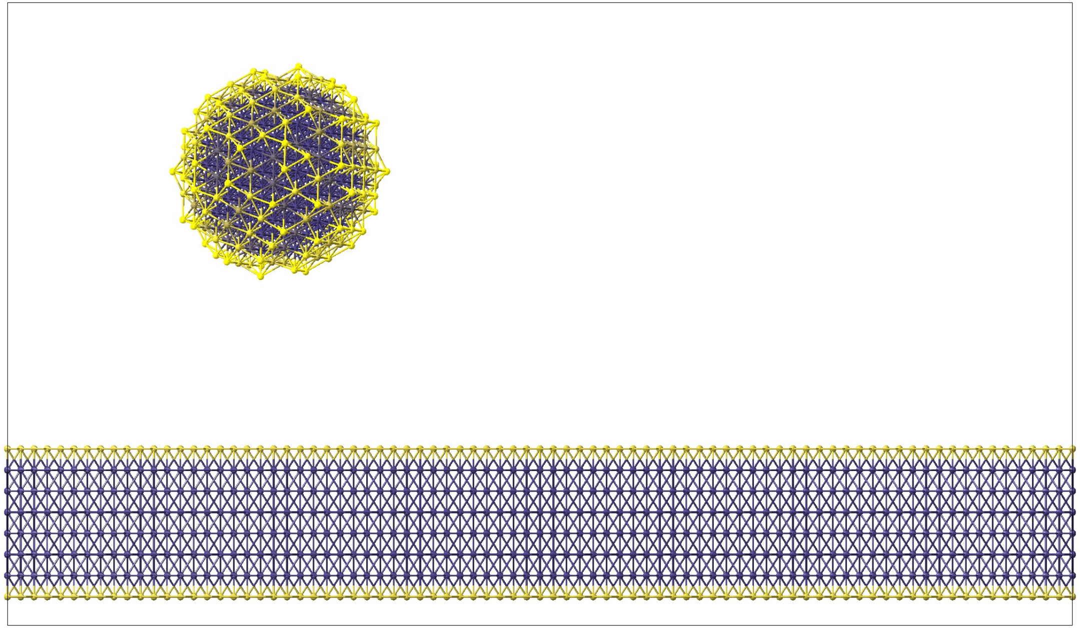
Specific heat:

- 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)

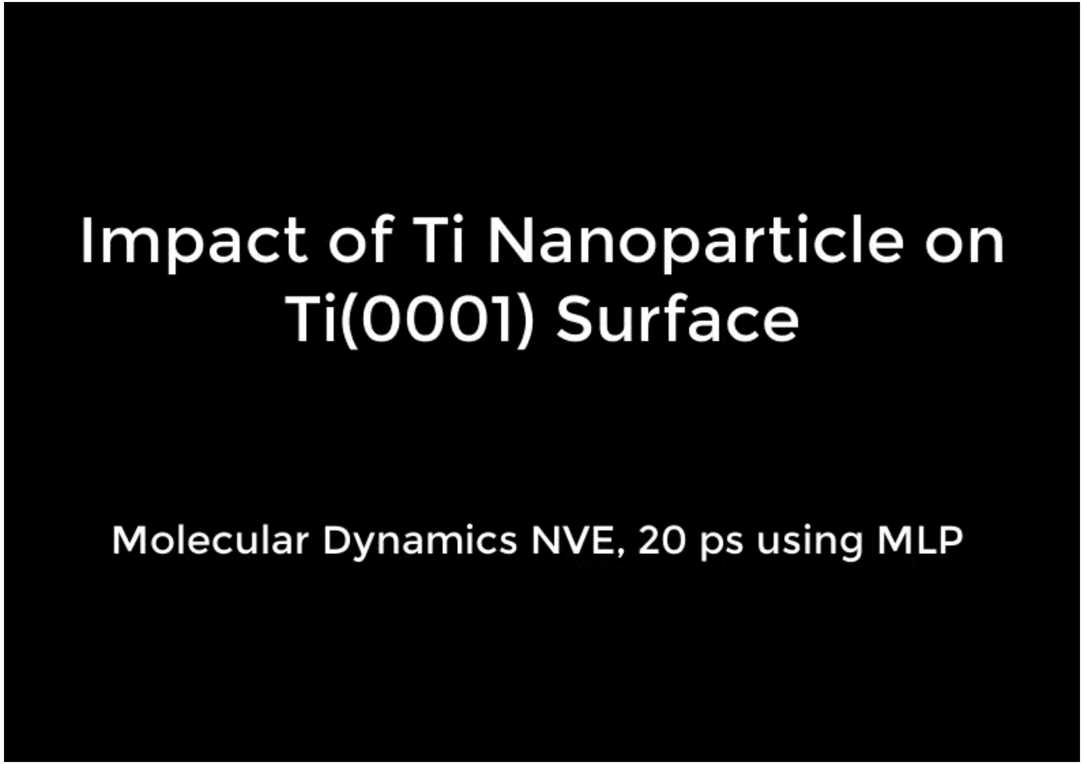
materials design

Impact of Ti Nanoparticle on α-Ti Surface



- The simulation uses a SNAP MLP generated from a training set containing over 1000 structures.
- The model contains 13214 atoms and was constructed with *MedeA*.
- This simulation was run on a GPU with *MedeA LAMMPS*.
- 20,000 steps were computed in 24 minutes.
- The rendering was done with screen capture from *MedeA.*

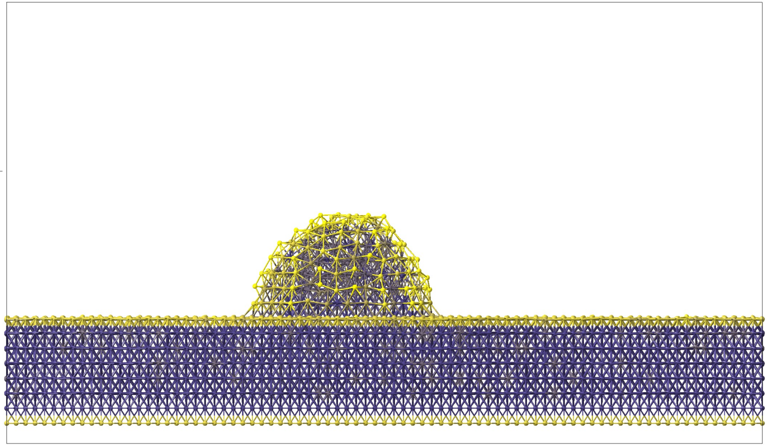
Impact of Ti Nanoparticle on α-Ti Surface



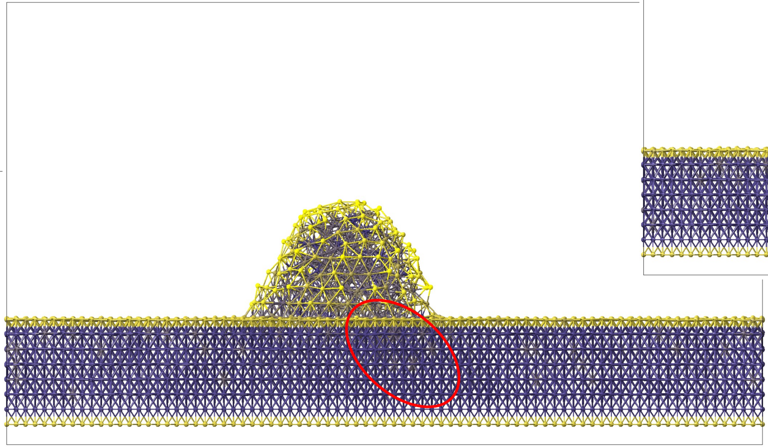
- The simulation uses a SNAP MLP generated from a training set containing over 1000 structures.
- The model contains 13214 atoms and was constructed with *MedeA*.
- This simulation was run on a GPU with *MedeA LAMMPS*.
- 20,000 steps were computed in 24 minutes.
- The rendering was done with screen capture from *MedeA.*

Impact of Ti Nanoparticle on α-Ti Surface

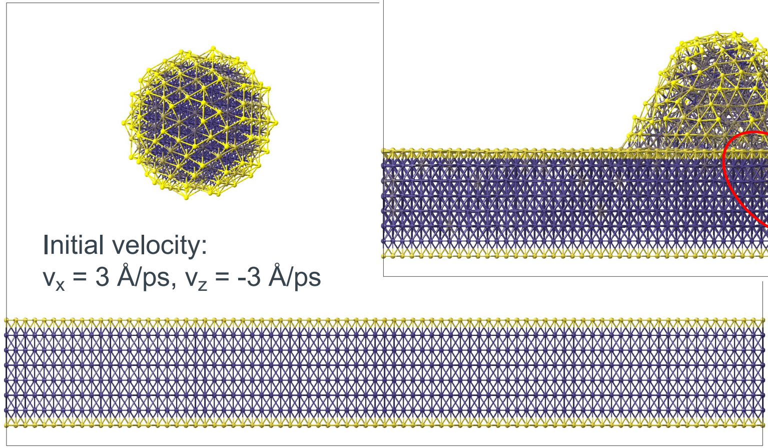
- Initially, only surface atoms are found at elevated energies as indicated by the yellow color
- Nanoparticle starts to recrystallize immediately after hitting the surface



Final configuration



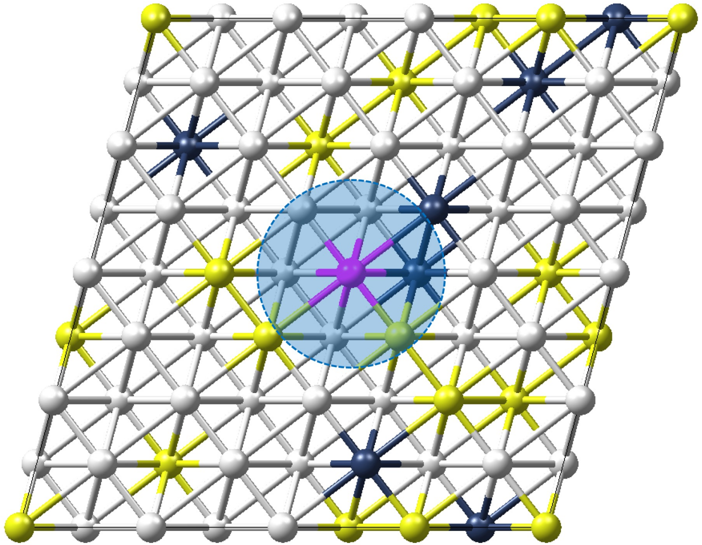
After 9 ps



Initial configuration

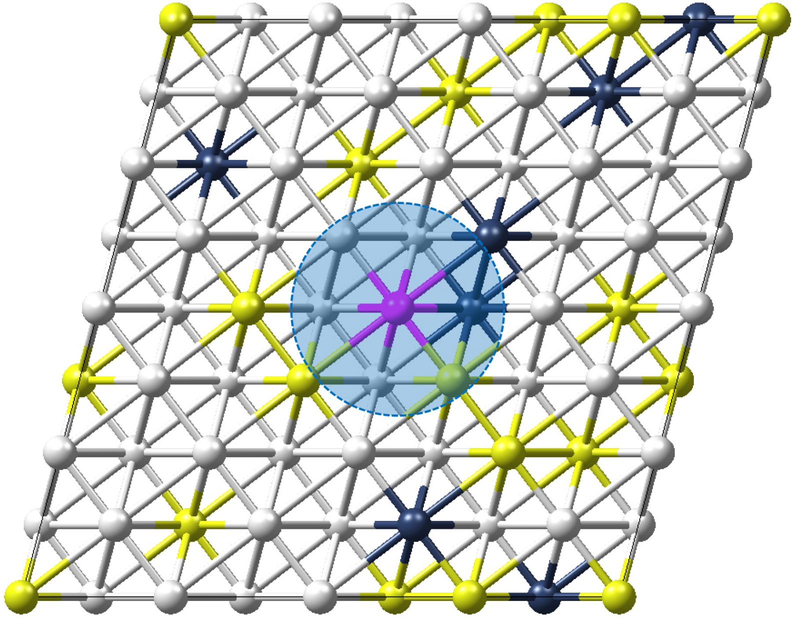
- After the impact, energies of bulk atoms increase, especially inside the nanoparticle and in the compression zone
- Finally, slab and nanoparticle start to relax

Machine-Learned Potentials



Descriptors and Regressors

Descriptor: describes atomic structures



- Divide structure into local atomic environments/clusters
- Describe local atomic structures in terms of atom positions and types

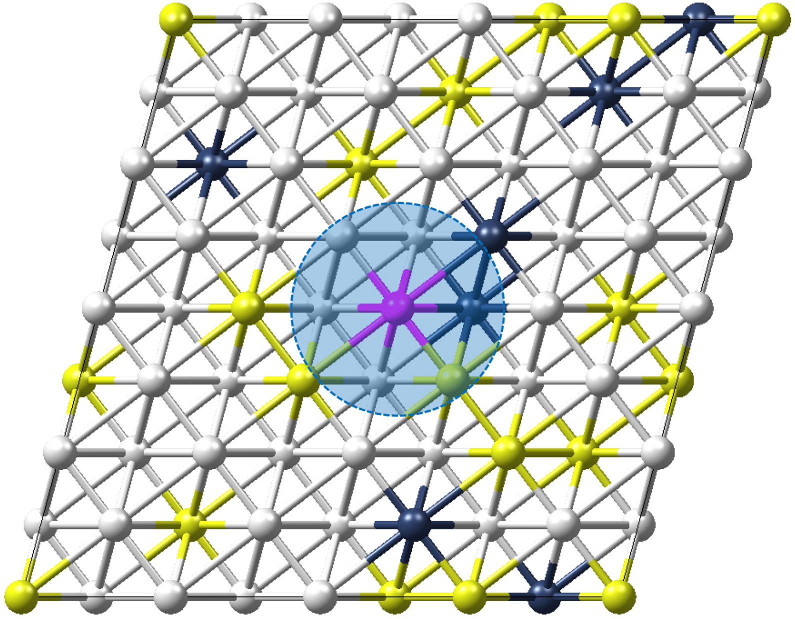
$$B_i = B_i\,(\{\mathbf{r}_{ji}, A_j, j = 1, N_i\}),\ i = 1, N$$

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

materials design

Descriptors and Regressors

Regressor: maps structures to energies/forces/stresses



- Divide energies/forces/stresses into contributions from all clusters

$$E = \Sigma_i E_i$$

- Express energies/forces/stresses in terms of descriptors B_i

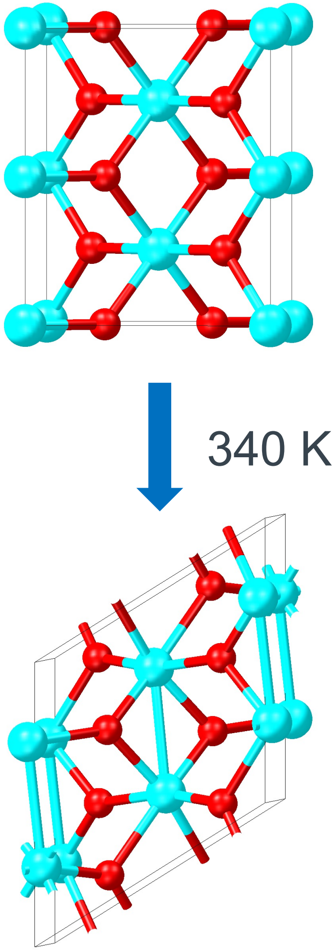
$$E_i = \beta_0 + \Sigma_k \beta_k B_{i,k}$$

$$\boldsymbol{F}_{i,j} = -\nabla_j E_i = -\sum_k \beta_k \frac{\partial B_{i,k}}{\boldsymbol{r}_j}$$

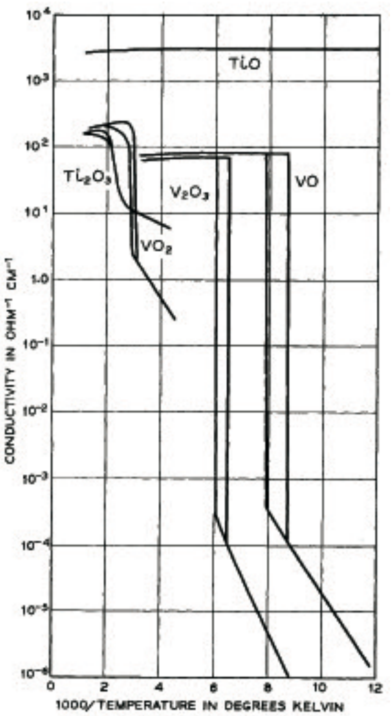
- Determine (train) coefficients β_k using *ab initio* calculations

Vanadium Dioxide

The Metal-Insulator Transition



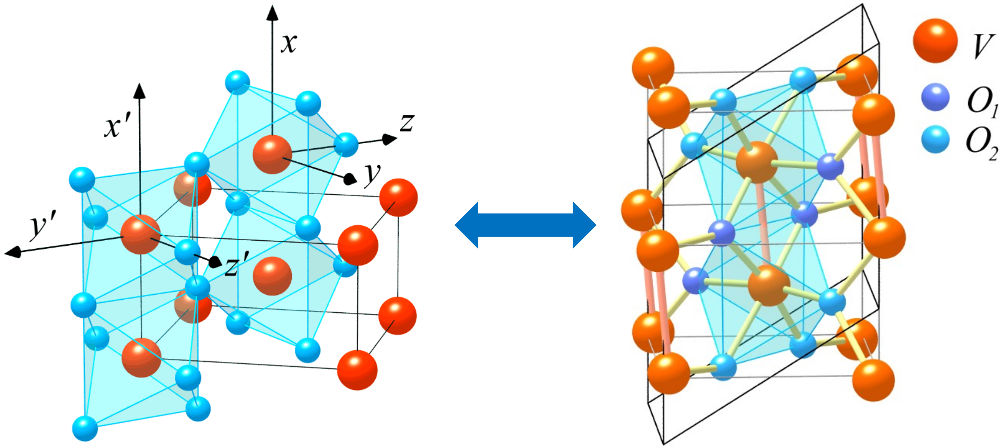
Metal-Insulator Transition of VO_2



F. J. Morin, PRL **3**, 34 (1959)

- MIT at 340 K
- $\Delta\sigma > 10^2\ (\Omega cm)^{-1}$

- V-V dimerization || c
  - V 3d – V 3d interaction
  - splitting of $d_{||}$ band
- V displacement ⊥ c
  - V 3d – O 2p interaction
  - upshift of π* band

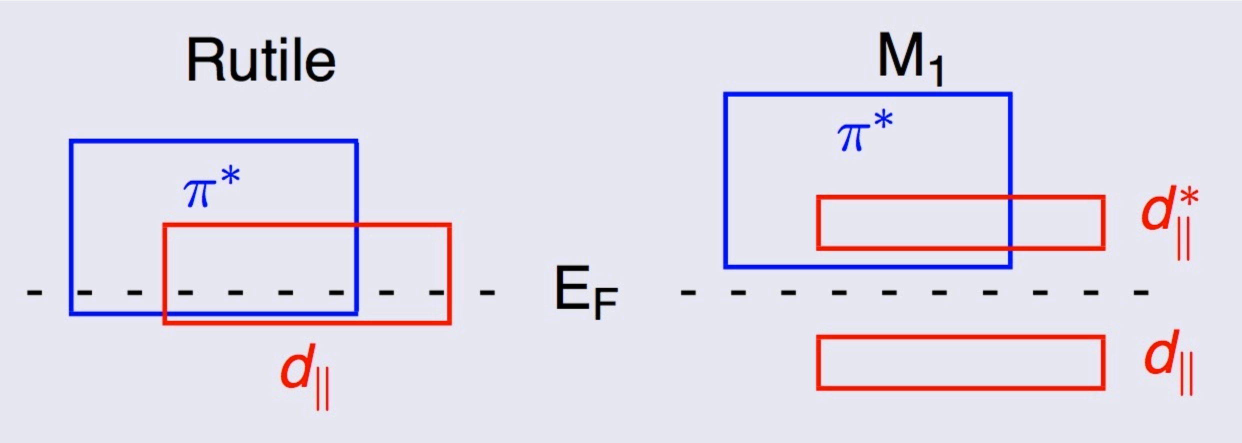


T > 340 K: Rutile

T < 340 K: M_1

materials design

Origin of MIT: Hen (Phonons) or Egg (Electrons)?



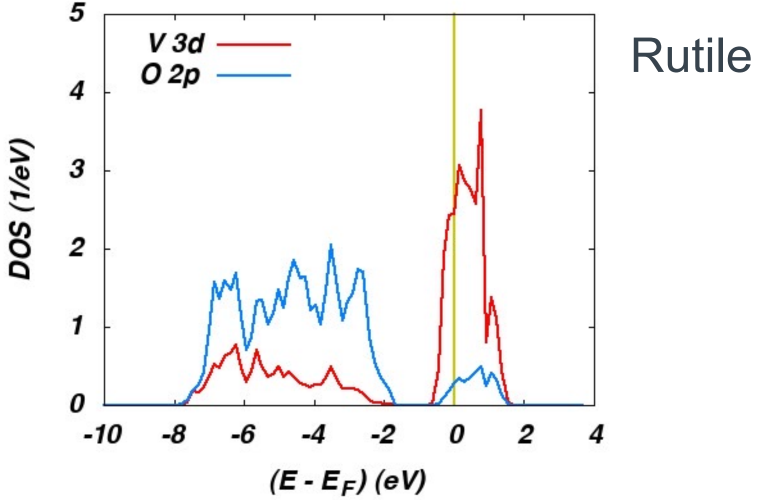
J. B. Goodenough, PR **117**, 1442 (1960)

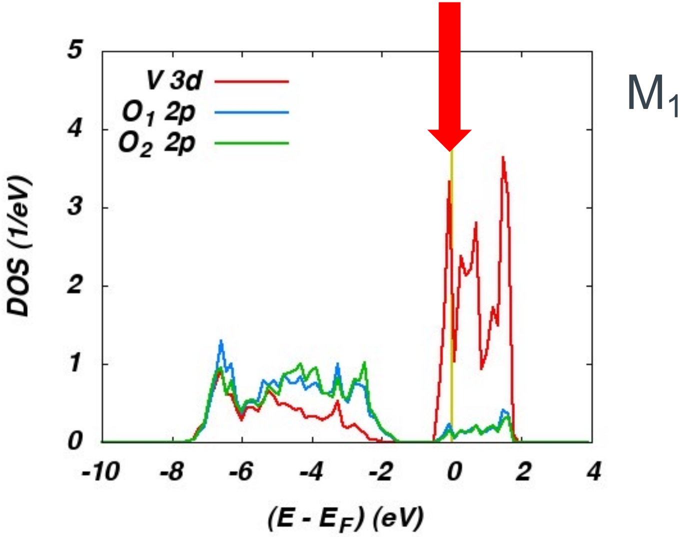
J. B. Goodenough
Nobel Prize 2019

- Goodenough: MIT due to structural distortions
- Zylberstejn and Mott: (Structural distortions) and MIT due to electronic correlations

Sir N. F. Mott
Nobel Prize 1977

Ab initio Calculations using GGA





DFT calculations using a GGA functional fail to obtain band gap of the M_1 phase!

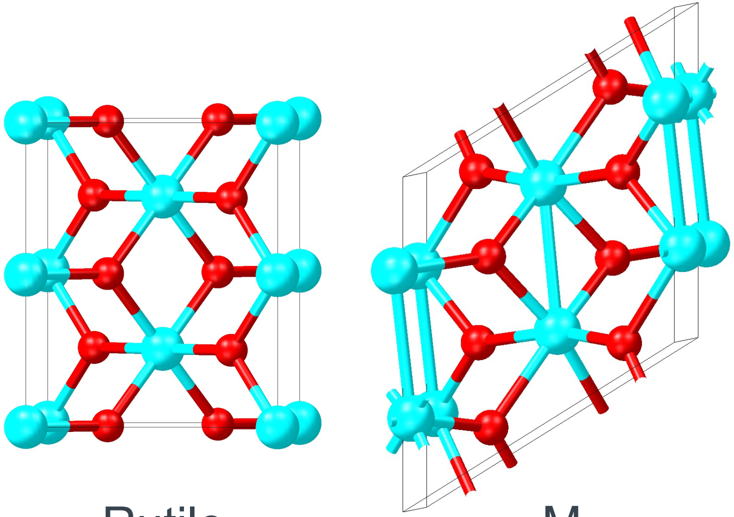
DFT calculations using the r2SCAN metaGGA functional show metallic behavior for rutile VO_2 and a band gap of 0.3 eV for M_1 VO_2

V. Eyert, Ann. Phys. **11**, 650 (2002); PRL **107**, 016401 (2011)

materials design

Training Set Calculations

Training set structures
- Initial structures (R, M1)
- Isotropic and uniaxial strain up to ±6%
- Angular deformations up to ±2°
- NPT MD simulations at 300-3000 K
- Total of 314 structures

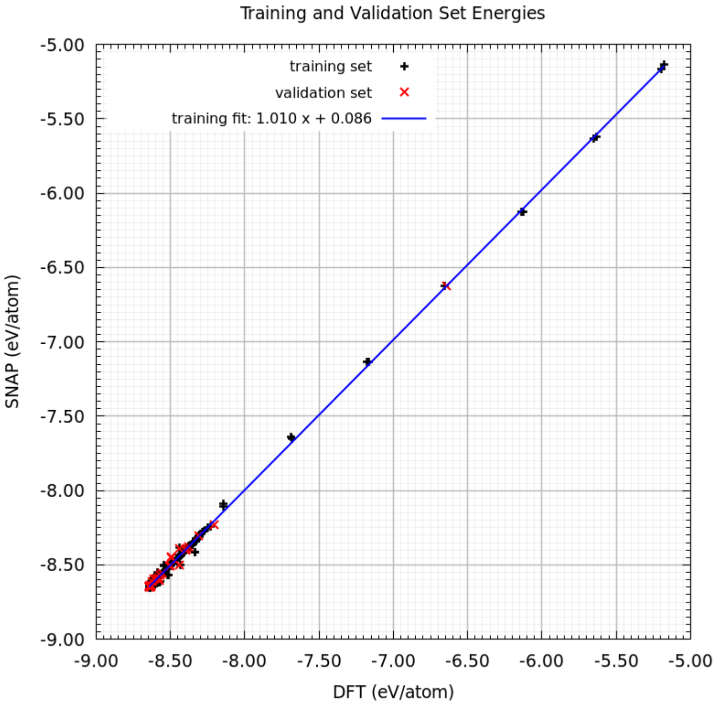


Rutile

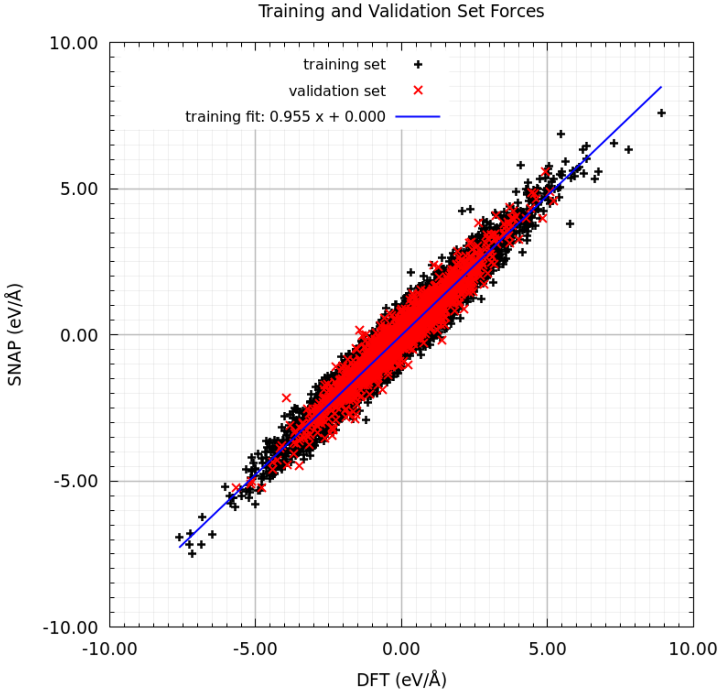
M_1

VASP computational parameters
- PBE functional
- 400 eV plane-wave cutoff
- **k**-point spacing 0.2 Å^{-1}
- Gaussian smearing, σ = 0.05 eV

MedeA MLP Generator: Assess



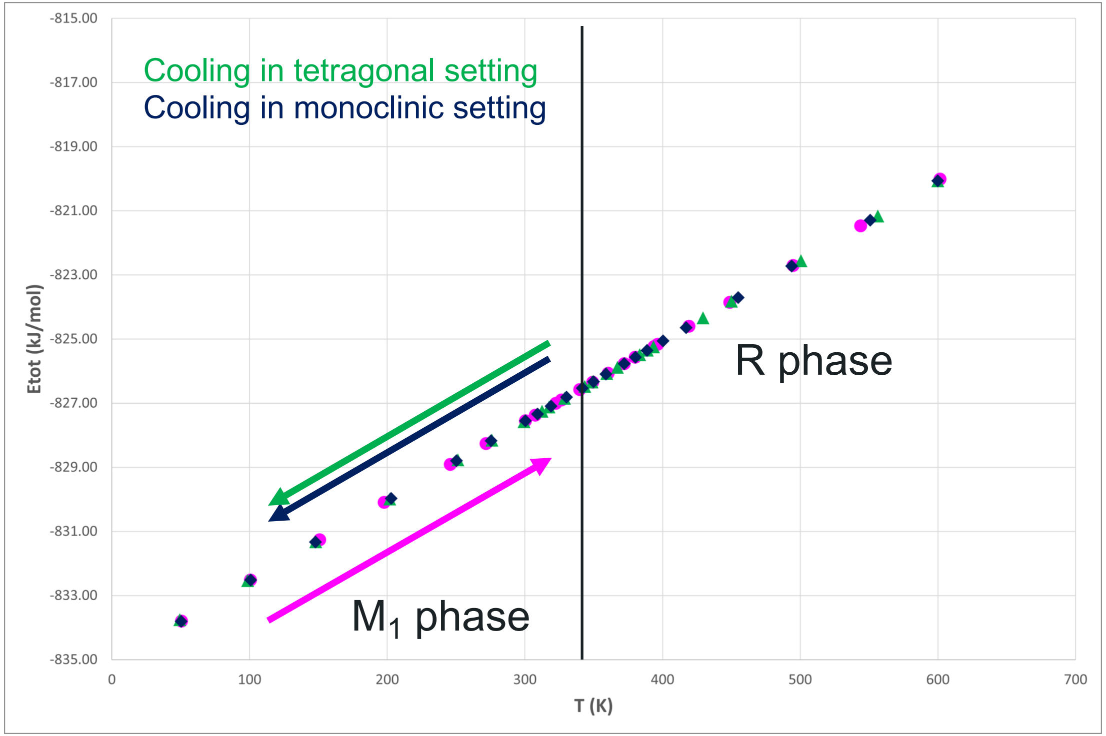
Energies



Forces

materials design

VO_2: Heating and Cooling



Energy/atom

Transition temperatures (exp):

- M_1-R transition: 340 K

Specific heat:

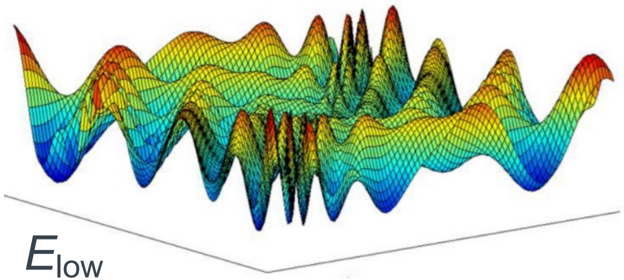
- Exp.: c_p = 63-67 J/(mol·K) (400-500 K)
- MLP: c_p = 75 J/(mol·K)

No indication of the transition from NPT MD simulations using an MLP based on GGA calculations!

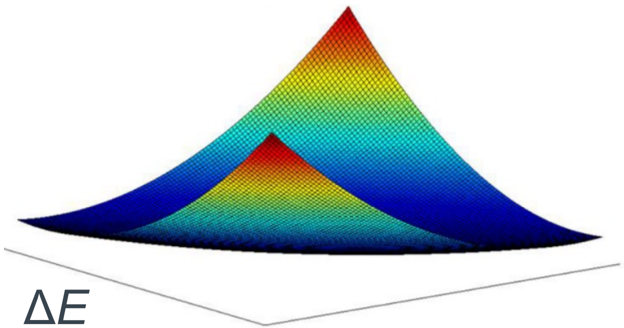
Berglund and Guggenheim, Phys. Rev. **185**, 1022 (1969); Pintchovski et al., J. Phys. Chem. Solids **39**, 941 (1978)

materials design

Δ–Learning: Idea



E_{low}



ΔE

Courtesy of G. Kresse;
P. Liu, C. Verdi, F. Karsai, and G. Kresse, *Phase transitions of zirconia: Machine-learned force fields beyond density functional,* PRB **105**, L060102 (2022);
see also: VASP, Machine Learning, and Multi-Scale Physics, Webinar (2022)

Large Training Set

↓

Calculate E_{low}, $\boldsymbol{F}_{low}$ from low-level DFT, e.g., GGA

↓

Generate MLP_{low} from E_{low}, $\boldsymbol{F}_{low}$

↓

$E_{high} = E_{low} + \Delta MLP(E)$,
$\boldsymbol{F}_{high} = \boldsymbol{F}_{low} + \Delta MLP(\boldsymbol{F})$

↓

Generate MLP_{high} from E_{high}, $\boldsymbol{F}_{high}$

Small Training Set

↓

Calculate E_{low}, $\boldsymbol{F}_{low}$ from low-level DFT, e.g., GGA

↓

Calculate E_{high}, $\boldsymbol{F}_{high}$ from high-level DFT, e.g., metaGGA, RPA

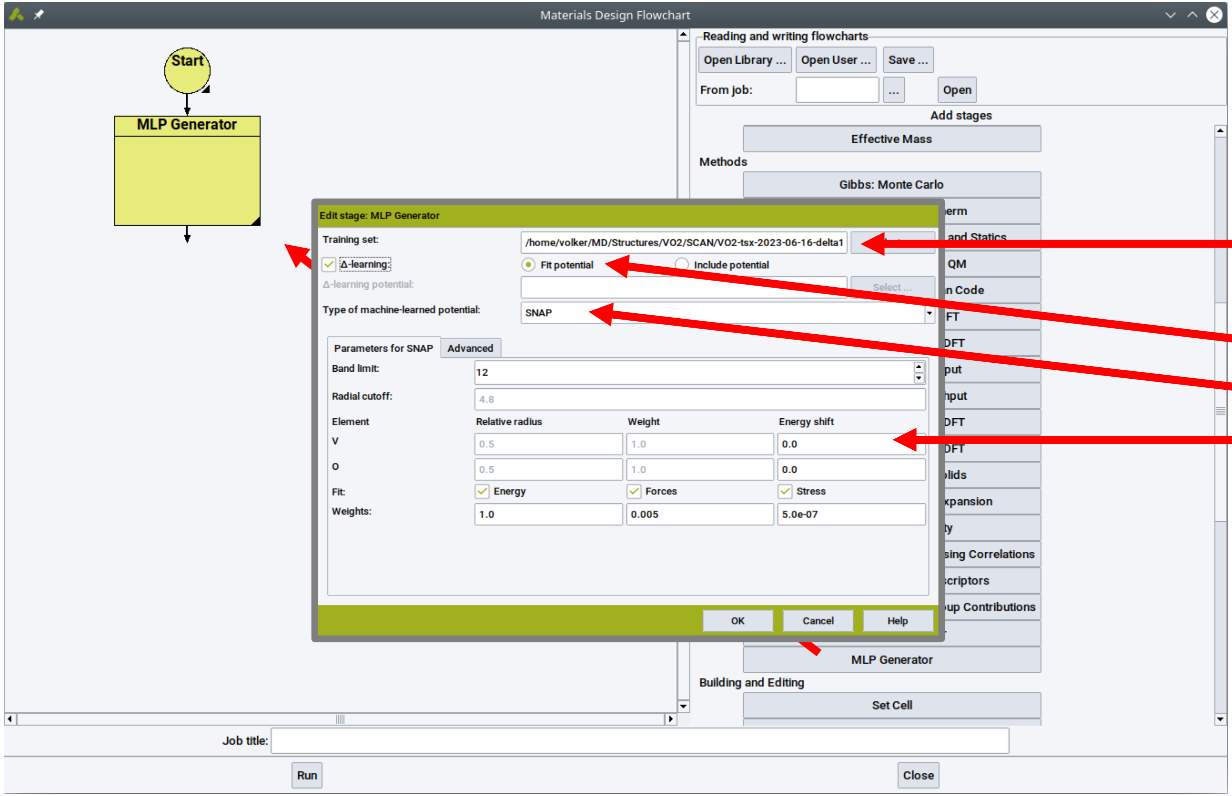
↓

$\Delta E = E_{high} - E_{low}$,
$\Delta \boldsymbol{F} = \boldsymbol{F}_{high} - \boldsymbol{F}_{low}$

↓

Generate ΔMLP from ΔE, $\Delta \boldsymbol{F}$ (→ into $E_{high} = E_{low} + \Delta MLP(E)$ box)

Δ–Learning fully integrated in *MedeA* MLP Generator



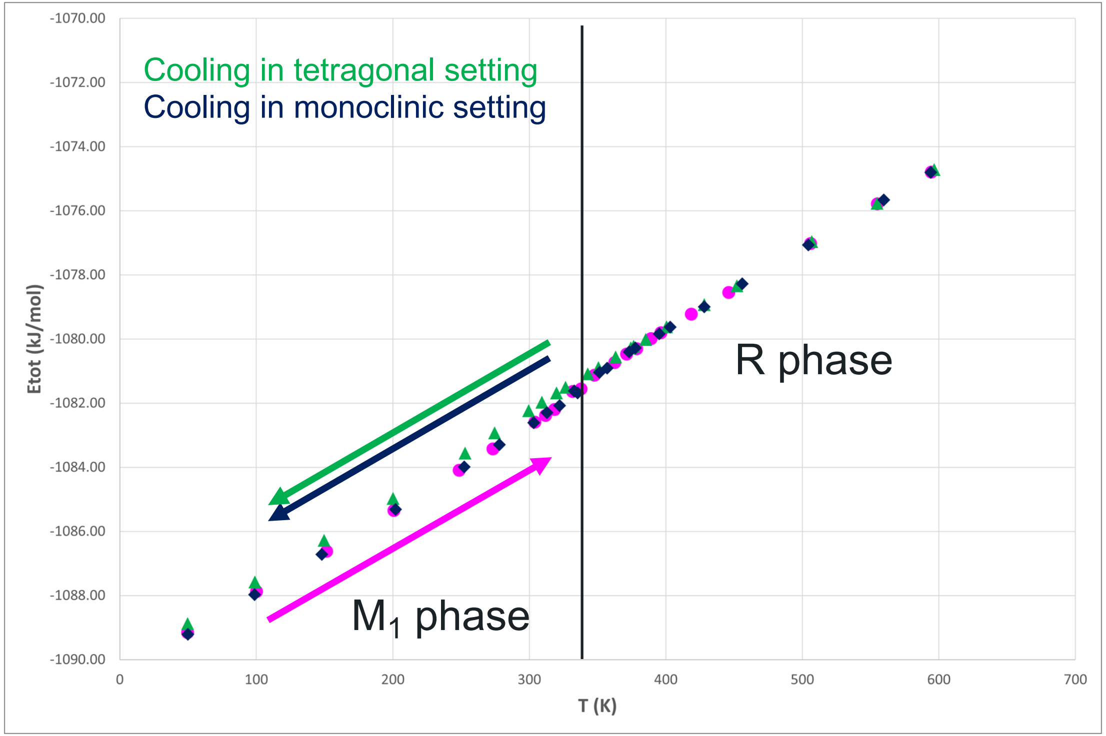
Call MLP Generator in Flowchart

Specify

- Training set, i.e, structure list
- Specify Δ–learning
- Type of potential
- Basic parameters

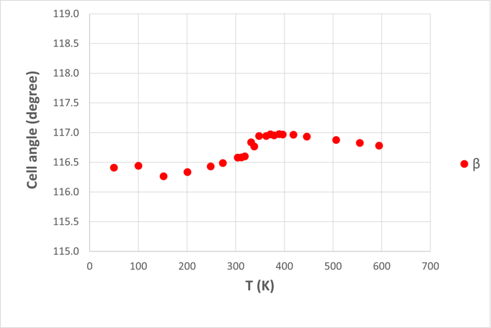
materials design

VO_2: M_1-R Phase Transition after Δ–Learning



Energy/atom

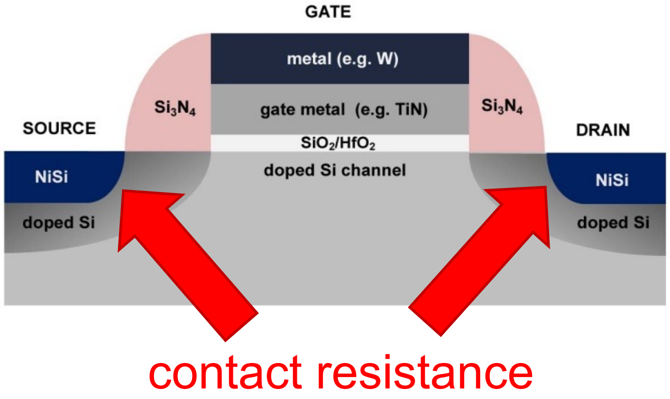
The phase transition of VO_2 is identified from NPT MD simulations using an MLP based on GGA calculations with results from metaGGA calculations included via Δ–learning!



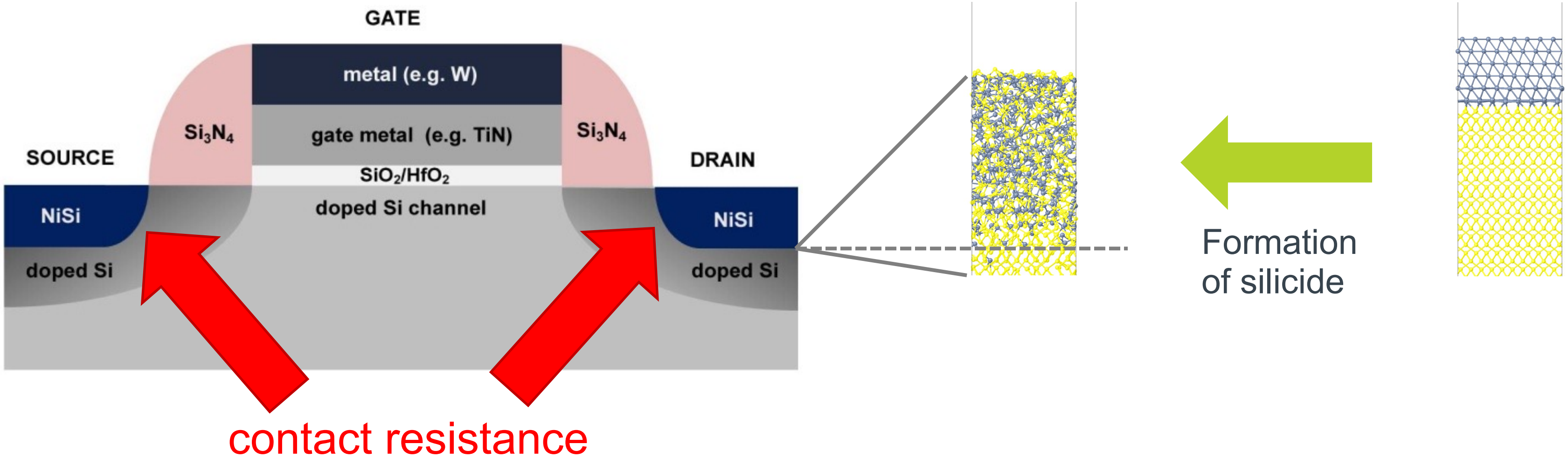
Kink of the monoclinic angle at the phase transition

Nickel Silicide

Green Electronics



Ni-Si as Metallic Contact in CMOS Devices



Contact resistance at the source and drain in CMOS devices

- is controlled by the Schottky barrier (SB) at the interface between the metallic (NiSi) and semiconducting (doped Si) regions,
- is not reduced by scaling to smaller device sizes, i.e., is a critical bottleneck.

materials design

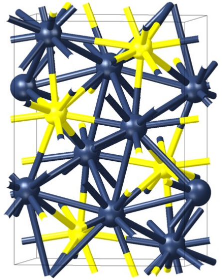
Training Set Calculations

Training set structures

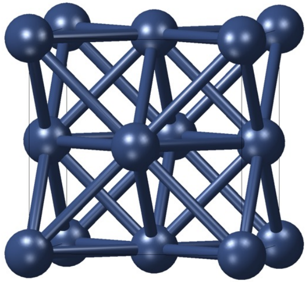
- Initial structures, supercells
- Surfaces, interfaces
- Isotropic and uniaxial strain up to ±6%
- Angular deformations up to ±2°
- NPT/NVT MD simulations at 100-2000 K
- Total of ≈ 2700 structures

VASP computational parameters

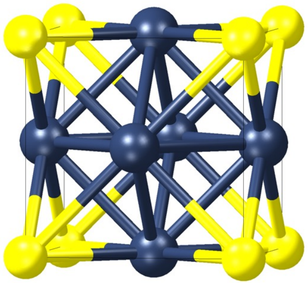
- PBE functional
- 300 eV plane-wave cutoff
- **k**-point spacing 0.2 $Å^{-1}$
- Gaussian smearing, σ = 0.05 eV



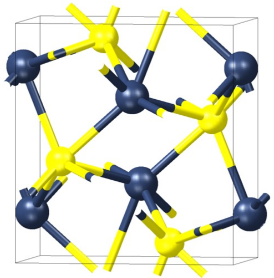
Ni_2Si, Pnma



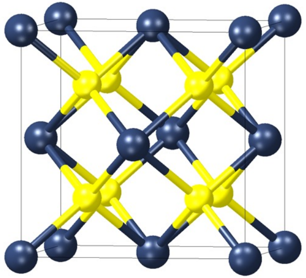
Ni, Fm-3m



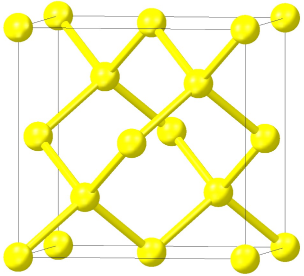
Ni_3Si, Pm-3m



NiSi, Pnma



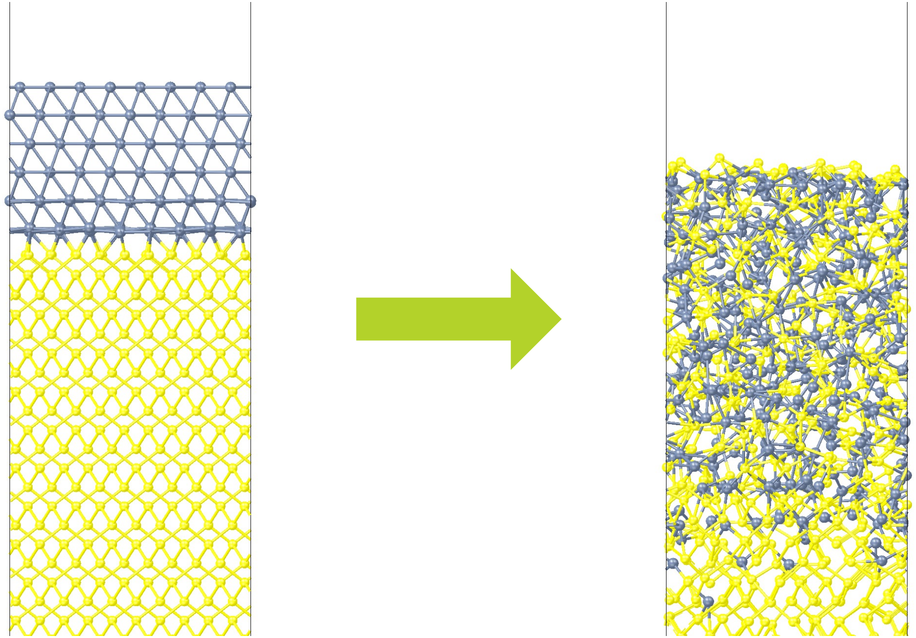
$NiSi_2$, Fm-3m



Si, Fd-3m

materials design

Silicidation from MD Simulation at 850 K



- The simulation uses a SNAP MLP generated from a training set including ≈ 2700 structures.
- The model contains ≈ 1300 atoms and was constructed with *MedeA*.
- This simulation was run on a GPU with *MedeA LAMMPS*.
- 400,000 steps were computed in 2½ hours.
- The rendering was done with screen capture from *MedeA.*

materials design

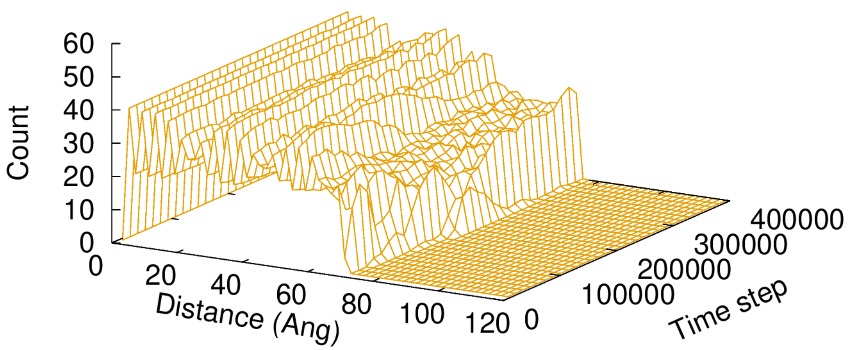
Silicidation from MD Simulation at 850 K

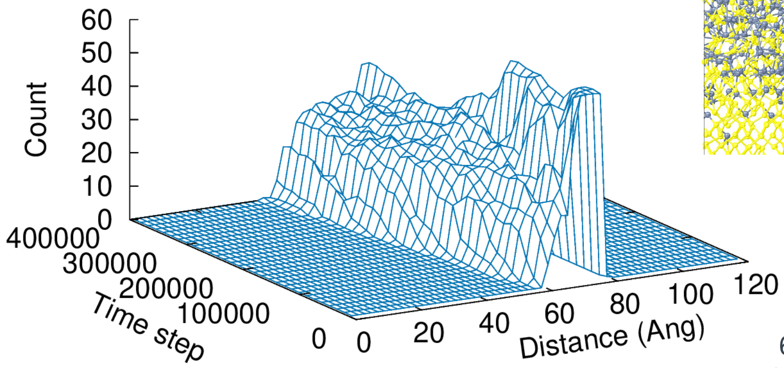
Formation of a Si/Silicide Interface by Ni Diffusion into Si Substrate

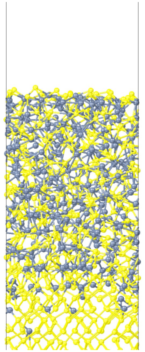
Molecular Dynamics Simulation at 850 K using *MedeA* LAMMPS

- The simulation uses a SNAP MLP generated from a training set including ≈ 2700 structures.
- The model contains ≈ 1300 atoms and was constructed with *MedeA*.
- This simulation was run on a GPU with *MedeA LAMMPS*.
- 400,000 steps were computed in 2½ hours.
- The rendering was done with screen capture from *MedeA.*

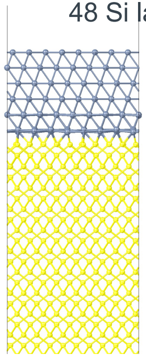
Silicidation from MD Simulation at 850 K







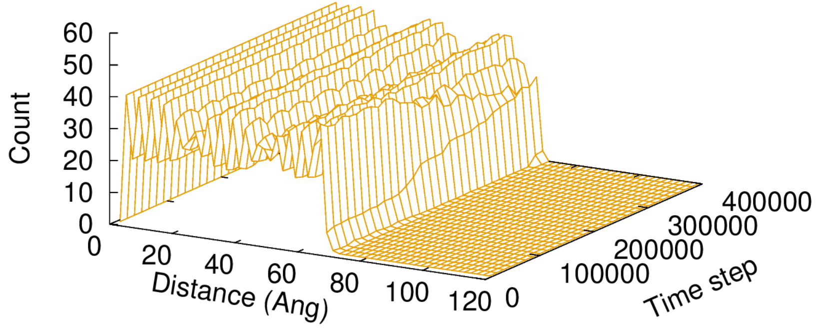
6 Ni layers
48 Si layers

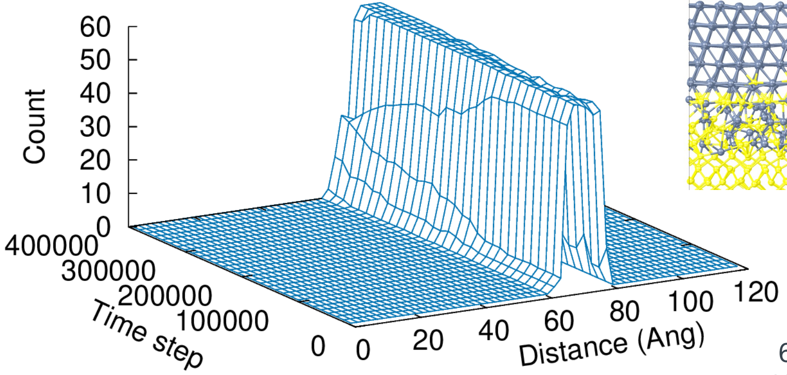


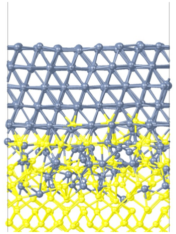
- Ni atoms diffuse from the top ≈ 10 Å layer into the substrate.
- Ni diffusion comes to an end at ≈ 40 Å below the surface.
- The top Ni layer is preserved for about 180 ps.
- Si atoms diffuse into the Ni layer.
- The top ≈ 40 Å region shows a combination of Ni_2Si and NiSi.

materials design

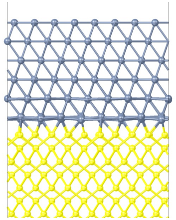
Silicidation from MD Simulation at 600 K







6 Ni layers
48 Si layers



- Ni atoms diffuse from the top ≈ 10 Å layer into the top ≈ 10 Å of the substrate.
- Only few Si atoms diffuse into the Ni layer.
- After about 180 ps a thin Ni_2Si layer is observed, which finally turns into a NiSi layer.

materials design

Concluding Remarks

Summary

MLPs in *MedeA*: VASP accuracy with LAMMPS speed

- Machine-Learned-Potentials (MLPs) enable highly efficient atomistic simulations at extended length and time scales with unprecedented levels of accuracy.
- *MedeA* offers a fully integrated MLP workflow from creation of training-sets (*MedeA* HT), generation of MLPs (*MedeA* MLPG), and their application (*MedeA* LAMMPS).
- Applications demonstrated in this webinar:
  - Phase transitions in metals and oxides
  - Impact of nanoparticles on surfaces
  - Diffusion at metal-semiconductor interfaces

Acknowledgements

All customers of Materials Design

All science and technology partners

All colleagues at Materials Design

The Advanced Materials Simulation Engineering Tool (AMSET) project, sponsored by the US Naval Nuclear Laboratory (NNL)

materials design

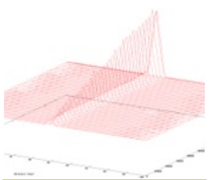
Highlighted *MedeA* Modules

***MedeA* VASP:** Comprehensive access to the VASP Code via a graphical user interface (GUI) to set up, run and analyze multi-step VASP calculations

***MedeA* MLPG:** Fully integrated workflow from training-set generation (using *MedeA* HT) and MLP generation to MLP application using *MedeA* LAMMPS

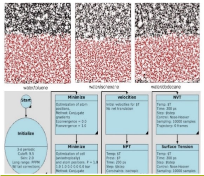
***MedeA* HT:** Generation of large and consistent sets of computed data for input to machine learning procedures

***MedeA* LAMMPS:** Full access to the LAMMPS Code via a graphical user interface based on flowcharts to perform forcefield calculations using MLPs generated by *MedeA* MLPG



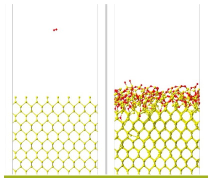
MT, Deformation

Mechanical, Thermal
Plastic deformation



Transport

Diffusion, Viscosity, Thermal conductivity, Surface Tension



Deposition,

Growth, Oxidation, and Etching

materials design

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

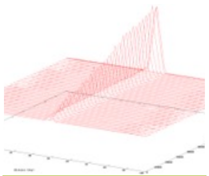
***MedeA* Viscosity:** Calculate viscosity with Green-Kubo or non-equilibrium MD Müller-Plathe

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

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

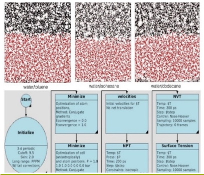
***MedeA* Deposition:** Atomistic scale simulation to study deposition, growth, oxidation and etching

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



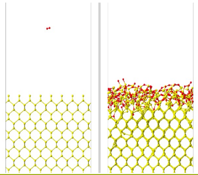
MT, Deformation

Mechanical, Thermal
Plastic deformation



Transport

Diffusion, Viscosity, Thermal conductivity, Surface Tension



Deposition,

Growth, Oxidation, and Etching

materials design

Related *MedeA* Webinars

Ab Initio for Millions - the Power of Machine-learned Potentials:
https://www.materialsdesign.com/webinars/recorded/Ab-Initio-for-MLPG

***MedeA* Training: *MedeA* Machine Learning Potential Generator (MLPG):**
https://www.materialsdesign.com/webinars/recorded/UGM-2021-Training-MLPG

VASP, Machine Learning, and Multi-Scale Physics: Defining the State of the Art in Materials Modeling:
https://www.materialsdesign.com/webinars/recorded/MedeA-VASP-Machine-Learning

Training: Generating and Applying Machine-Learned Potentials with *MedeA*:
https://www.materialsdesign.com/webinars/recorded/UGMtraining-Generating-and-Applying-Machine-Learned-Potentials-with-MedeA

On-the-fly Machine Learning Forcefields with *MedeA* VASP:
https://www.materialsdesign.com/webinars/recorded/MedeA-Training-On-the-Fly-Machine-Learning-Forcefields-with-MedeA-VASP

Related *MedeA* Application Notes

Accurate Band Gaps of Correlated Transition-Metal Oxides from Hybrid-Functional Calculations:

https://www.materialsdesign.com/all-application-notes/VO2_Band_Gaps

Prediction of Schottky Barrier in Electronic Devices:

https://www.materialsdesign.com/all-application-notes/Schottky_Barriers

The α-β-phase-transition in Ti investigated using a machine-learned interatomic potential:

https://www.materialsdesign.com/all-application-notes/Titransition

Related *MedeA* Tutorials

An Introduction to *MedeA* MLP:

Learn how to run LAMMPS simulations with Machine Learning Potential

An Introduction to *MedeA* MLPG:

Learn how to generate machine-learned potentials within *MedeA*

Generate a Ti Neural Network Machine-Learned Potential:

Learn how to generate a neural network potential using MedeA MLPG

Applying Delta-Learning to TiO_2 Machine-Learned Potentials:

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

materials design

Announcement

Upcoming

- Webinar: The Basis of Success - A Conversation with the President of Gaussian Inc., Dr. Mike Frisch

- 2023 Materials Design UGM

  Vienna, Austria

  October 9-11

  Registrations open this week

  https://www.ugm.materialsdesign.com

TAKE A PRODUCT TOUR

materials design

Question and Answer Session

Dr. Xiaoli Liu

Materials Design

Dr. Volker Eyert

Materials Design

Questions about Materials Design Webinars

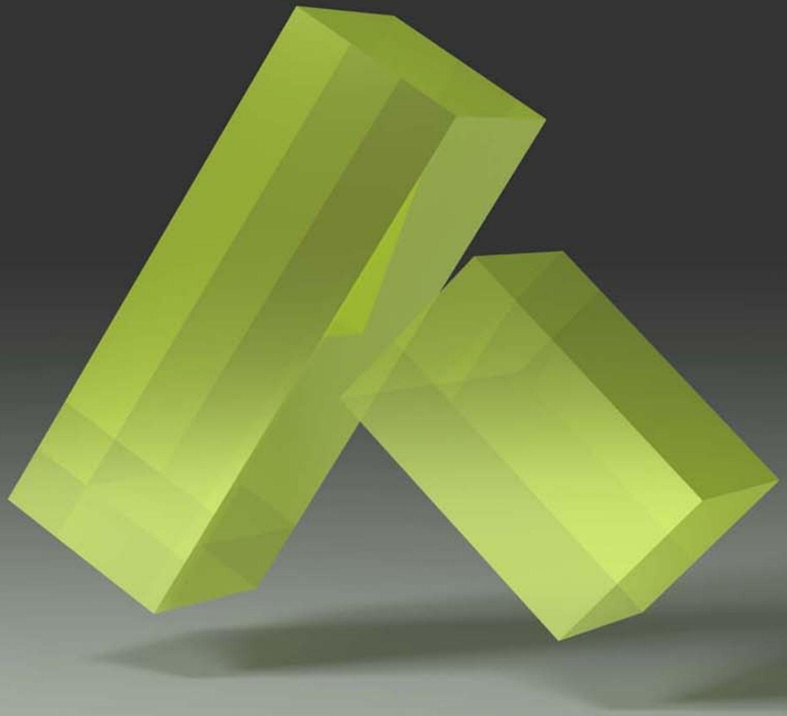
Katherine Hollingsworth

khollingsworth@materialsdesign.com

materials design

info@materialsdesign.com

www.materialsdesign.com



MedeA

Innovation by Simulation