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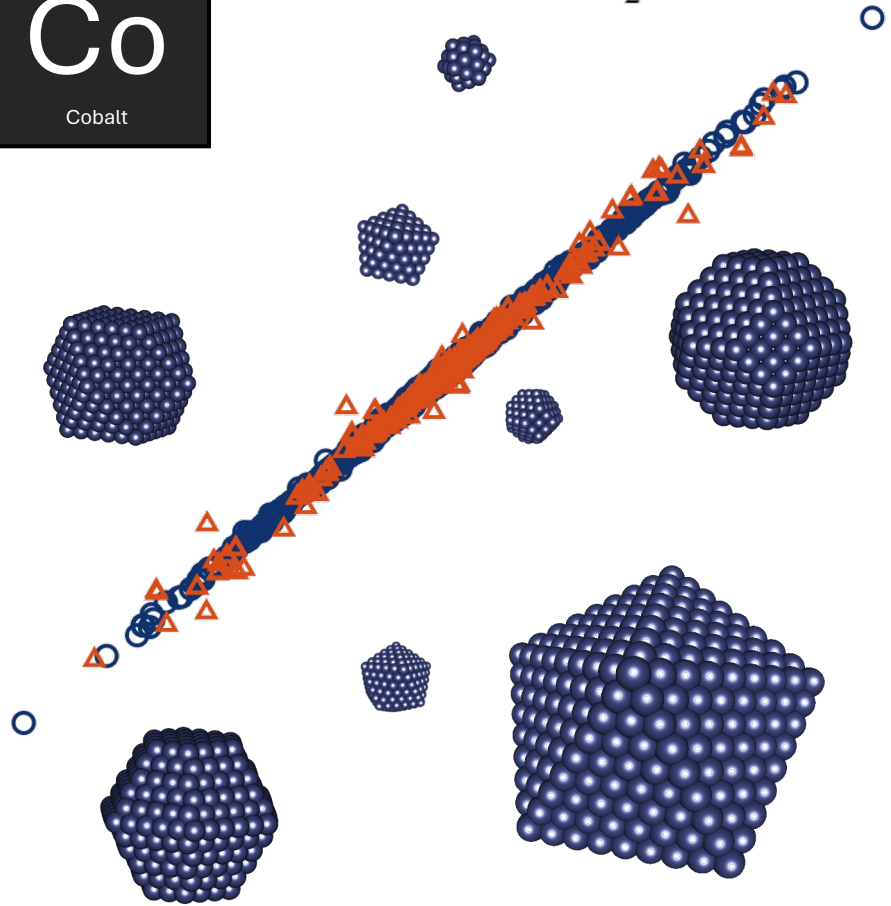
27
Co
Cobalt

$$E^i_{\text{q-SNAP}}(\mathbf{r}^N) = \beta \cdot \mathbf{B}^i + \frac{1}{2}(\mathbf{B}^i)^T \cdot \alpha \cdot \mathbf{B}^i$$

Polyvalent Machine-Learned Potential for Cobalt: from Bulk to Nanoparticles



March 26-28, 2024

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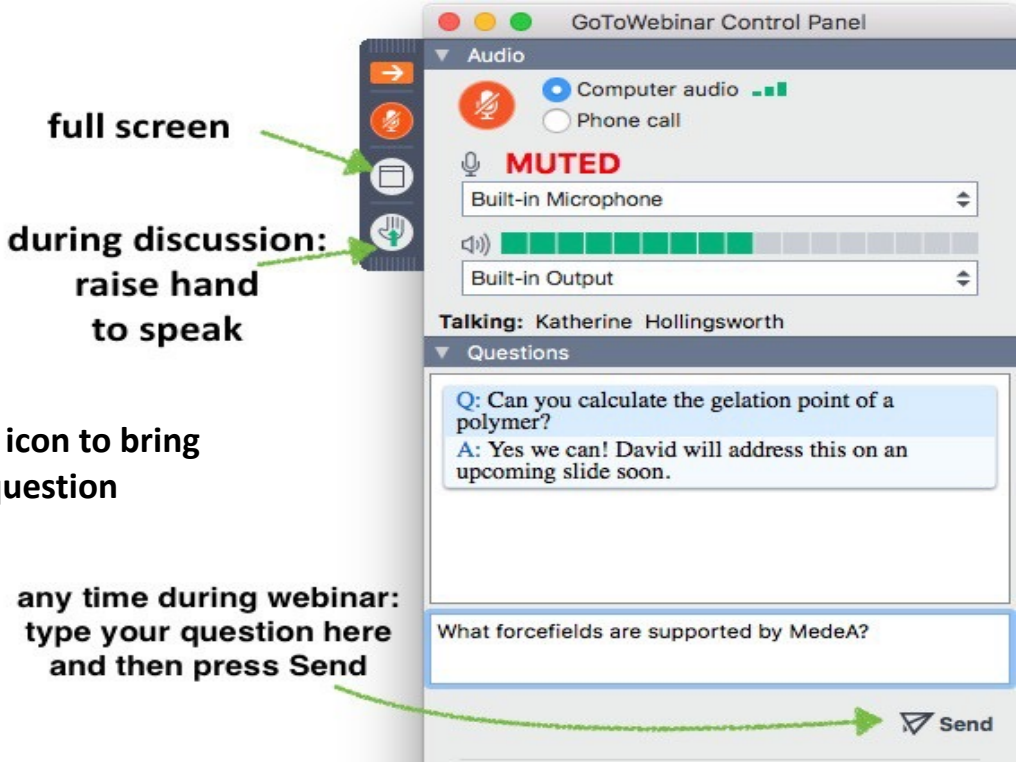

Use the raise hand icon to bring attention to your question

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

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Webinar Speakers

Katherine Hollingsworth

Marthe Bideault

Dr. Erich Wimmer

Agenda

- Why developing Machine-Learned Potentials (MLPs)?
- How can we generate MLPs using *MedeA?*
  - Quick overview about SNAP
  - Training set calculations
  - The *MedeA* Machine-Learned Potential Generator
- Validation: is the MLP able to reproduce cobalt properties?
- Application to cobalt nanoparticles
- Summary and conclusion

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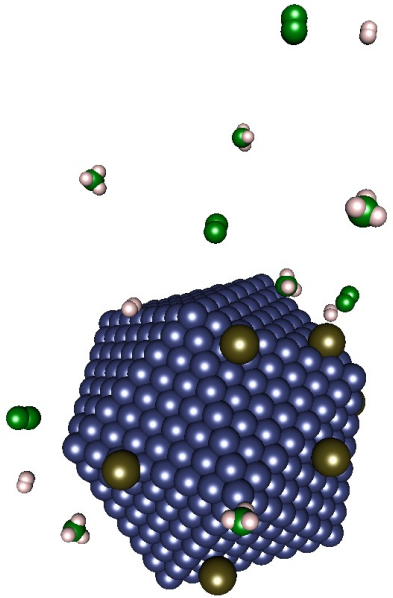
Why developing MLPs?



Complex chemistry
- Bond breaking
- Symmetry breaking
- Surfaces

Density Functional Theory

Limited to hundreds of picoseconds and atoms

Large scales, dynamics
- Several thousand atoms
- Adsorption/Desorption
- Diffusion
- Reaction kinetics
- Pressure and temperature effects

Molecular Mechanics

With traditional FF:
Not well suited for complex chemistry

With MLPs:
- Accuracy close to that of DFT
- Enable large-scales simulations
- Large flexibility

MLPs allow to mix complex chemistry with large scales and dynamics.

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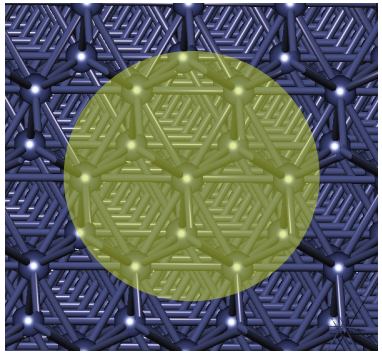
Development of the potential

Quick overview of MLPs

DATABASE

Descriptors of atomic environments labelled by energies, forces and stresses



REGRESSOR

Establishes the relationship between descriptors and their labels. It can be a linear regressor or neural networks.

MLP

$$B_{j_1,j_2,j} = \sum_{m_1,m_1'=-j_1}^{j_1} \sum_{m_2,m_2'=-j_2}^{j_2} \sum_{m,m'=-j}^{j} \left(u^{j}_{m,m'}\right) C^{jm}_{j_1 m_1 j_2 m_2} \times u^{jm'}_{j_1 m_1', j_2 m_2'} u^{j_1}_{m_1',m_1} u^{j_2}_{m_2',m_2}$$

$$E^{i}_{\text{q-SNAP}}(\mathbf{r}^N) = \beta \cdot \mathbf{B}^i + \frac{1}{2}\left(\mathbf{B}^i\right)^T \cdot \alpha \cdot \mathbf{B}^i$$

For more information:
V. Eyert, J. Wormald, W. A. Curtin, and E. Wimmer, **Machine-learned interatomic potentials: Recent developments and prospective applications**, *Journal of Materials Research* 38, 5079 (2023)

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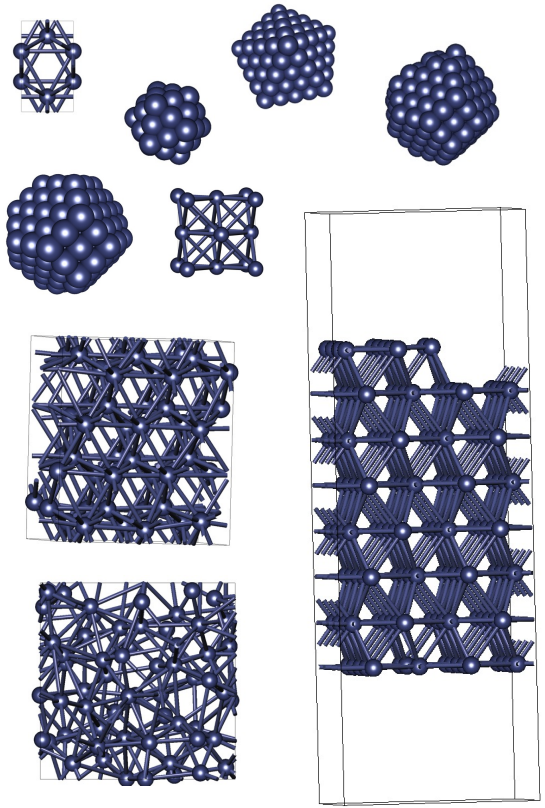
Training set calculations



Training set structures

- Unit cells of hcp, fcc, bcc and ω Co, deformed up to 1% strain
- Structures from MD simulations up to 1200 K:
  - Supercells, some with interstitials and vacancies
  - Surfaces with steps, ad atoms, vacancies
  - Nanoparticles with adatoms and vacancies
- Liquid structures
- Total of 1049 structures

VASP computational parameters

- Spin-polarized
- PBE functional
- 300 eV plane-wave cutoff
- **k**-point spacing 0.2 $Å^{-1}$
- Methfessel Paxton smearing, σ = 0.2 eV
- Additional support grid for the evaluation of augmentation charges
- SCF convergence criterium: 10^{-5} eV

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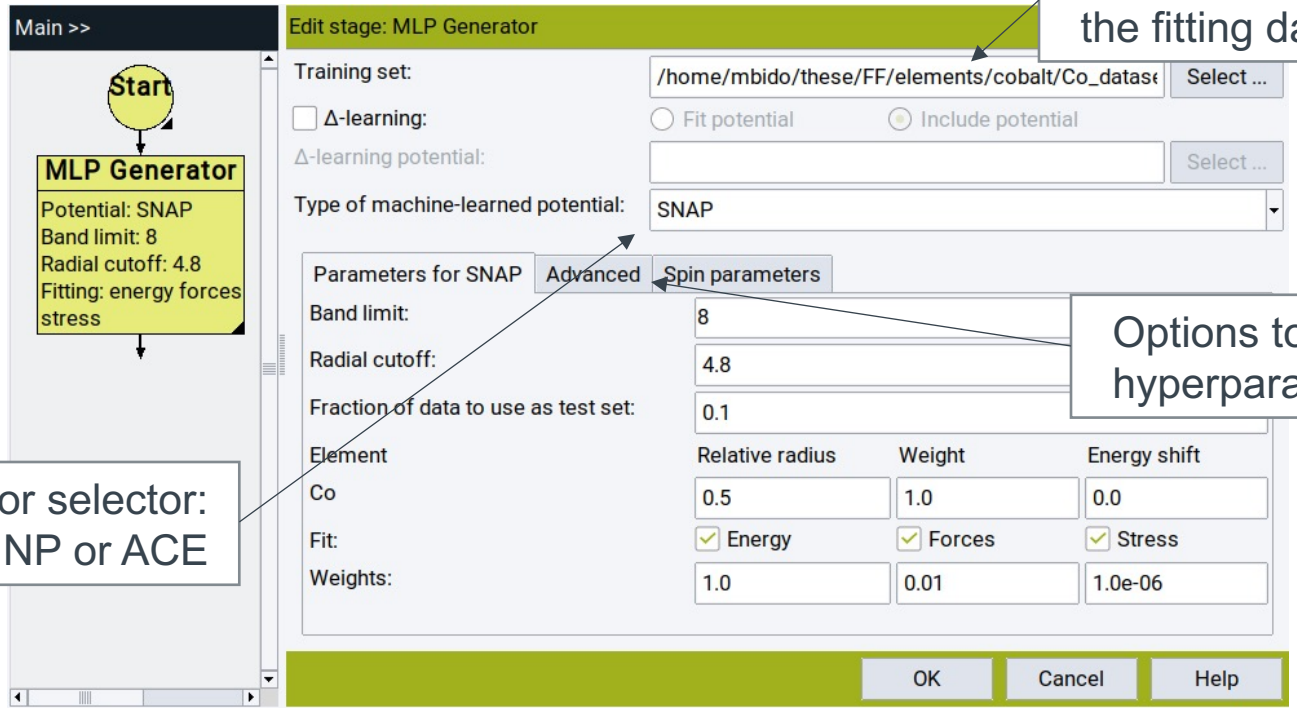
The *MedeA* Machine-Learned Potential Generator



Database created with the fitting data manager

Options to optimize the hyperparameters

Regressor selector: SNAP, NNP or ACE

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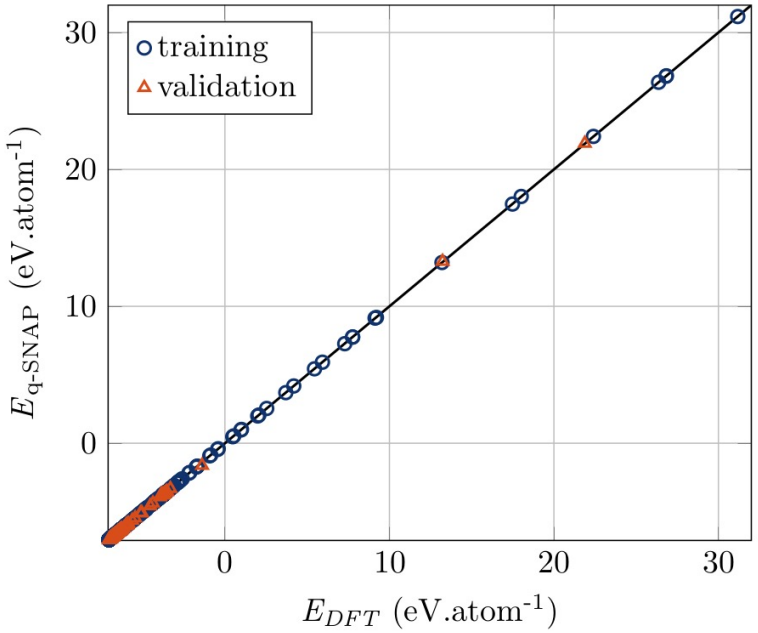
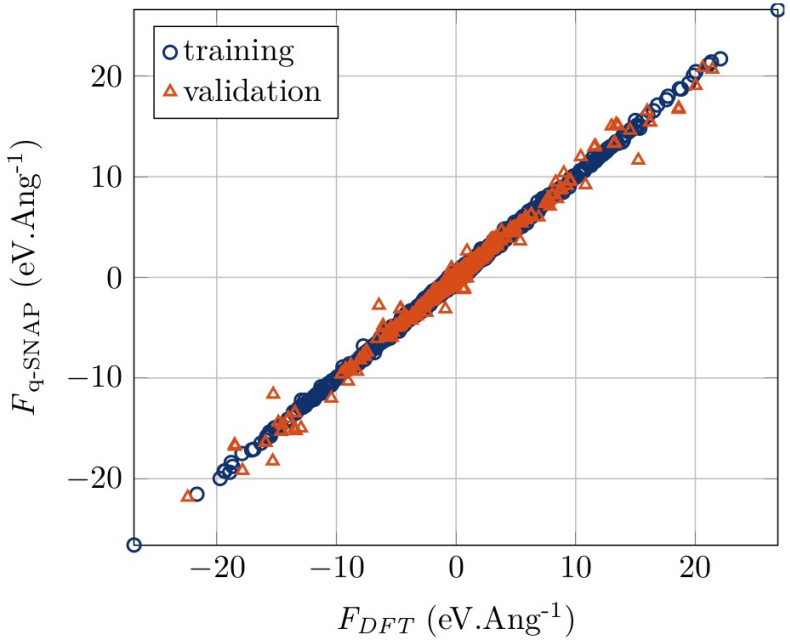
The *MedeA* Machine-Learned Potential Generator





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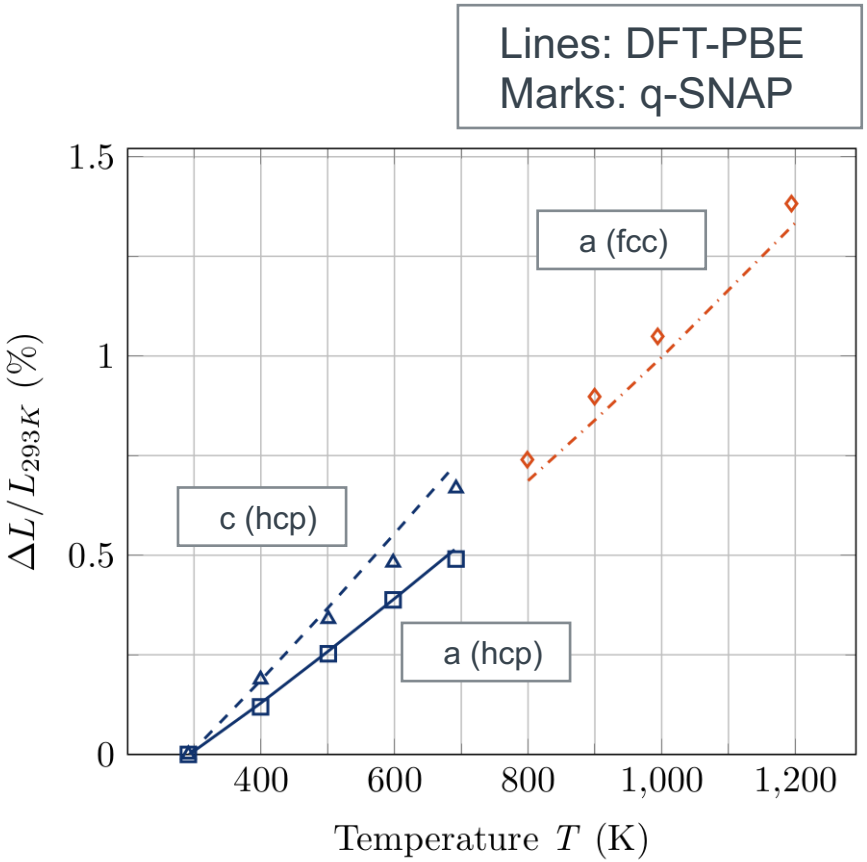
Validation of the potential

Lattice properties

Property	Experiment	DFT-PBE	q-SNAP
a_{hcp} (Å)	2.507[1]	2.4902	2.4899
c_{hcp}/a_{hcp}	1.623[1]	1.6160	1.6149
a_{fcc} (Å)	3.5447[2], 3.568[3]	3.5103	3.5116



[1]W. B. Pearson, **A handbook of lattice spacings and structures of metals and alloys**, 1st ed., Vol. 2 (Pergamon Press, Oxford, 1967).
[2]W. M. Haynes, **CRC Handbook of Chemistry and Physics** (Taylor & F, 2014).
[3]W. Gale and T. Totemeier, **Smithells Metals Reference Book**, 8th ed. (Elsevier, 2004).

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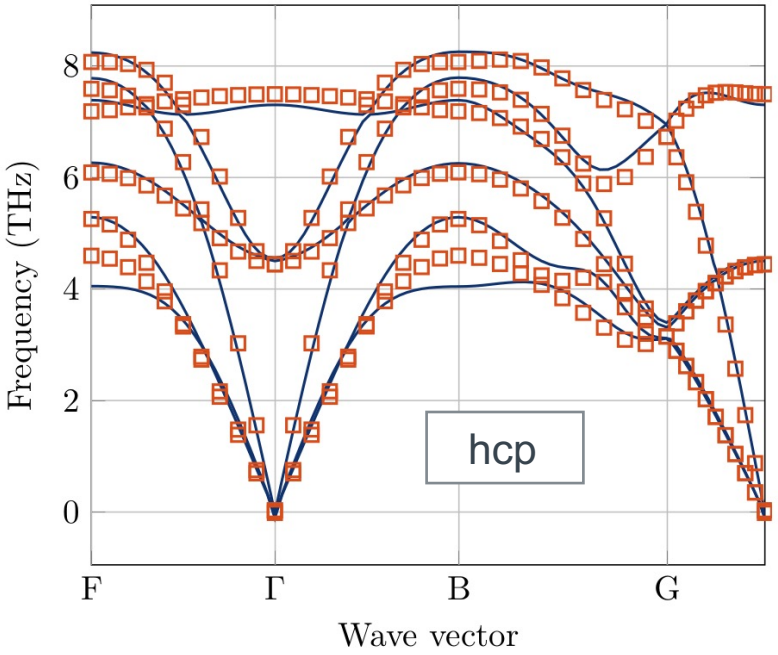
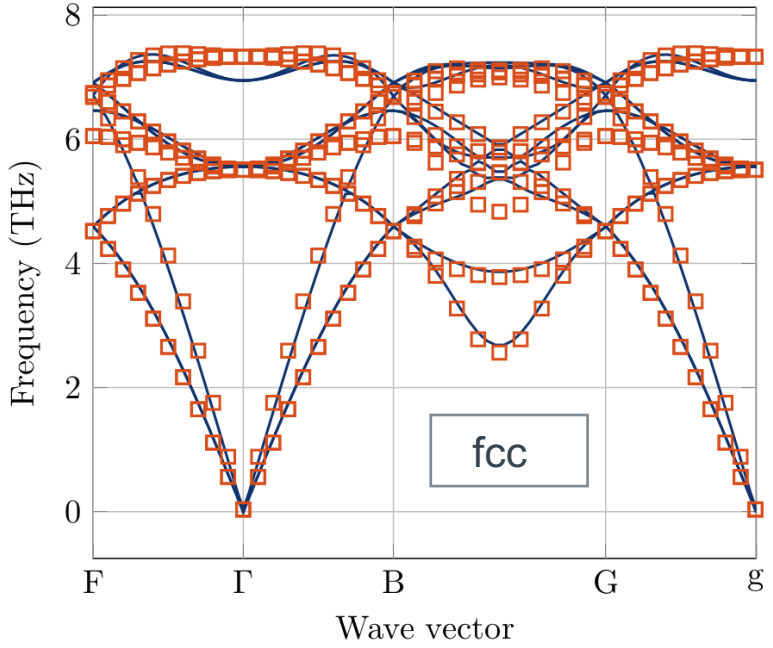
Phonon dispersions

Lines: DFT-PBE
Squares: q-SNAP





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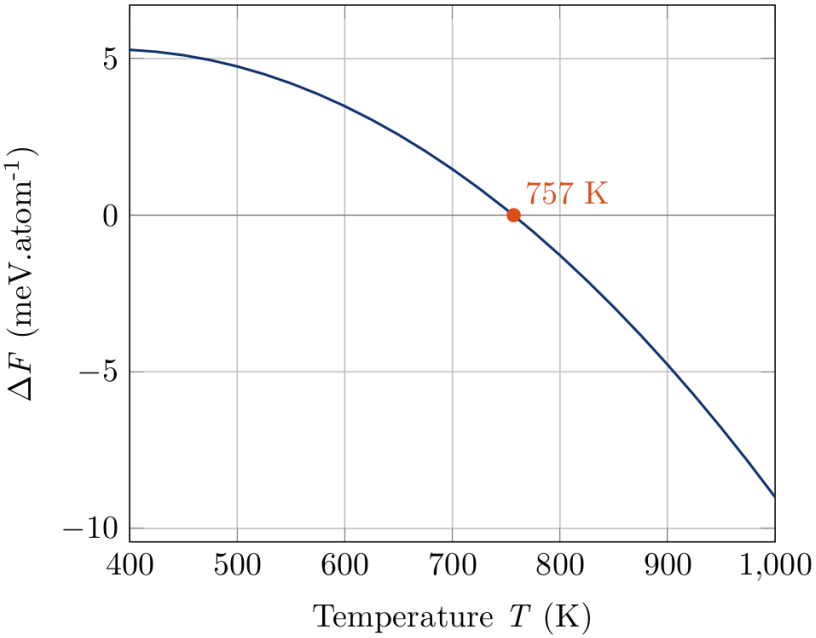
Phase transition temperature

Free energies were computed using *MedeA* MD Phonon.



Experimental value[1]: 693 K

[1]H. Matter, J. Winter, and W. Triftshäuser**, Phase transformations and vacancy formation energies of transition metals by positron annihilation**, *Appl. Phys.* 20, 135 (1979).

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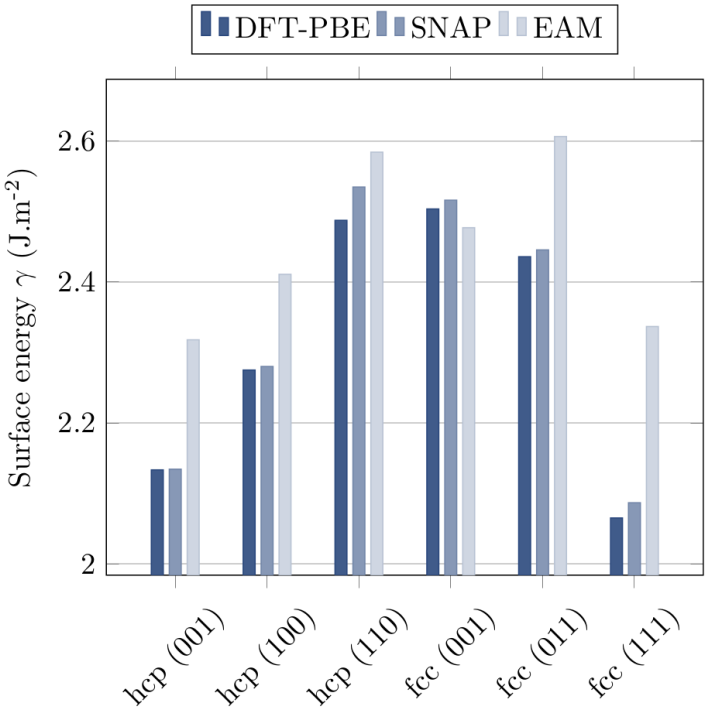
Surface energies



- Getting the surface energies correct is necessary to model correctly nanoparticles.
- The q-SNAP is well better for that task than the EAM[1] which is primarily aimed at accurately reproduce bulk properties.

[1]G. P. P. Pun and Y. Mishin, **Embedded-atom potential for hcp and fcc cobalt**, *Phys. Rev. B* 86, 134116 (2012).

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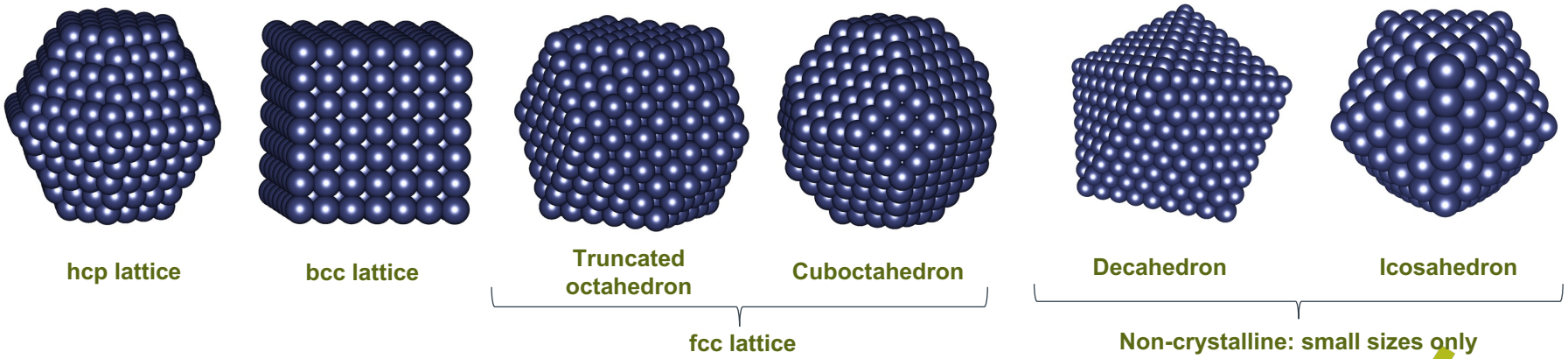
Applications to nanoparticles

Quick overview on nanoparticles

Nanoparticles are clusters of atoms characterized by their high surface-to-volume ratio which depends on the nanoparticle's size. This ratio plays a crucial role in determining the equilibrium shape.

What is the optimal energy balance between:

- Minimizing the number of broken bonds at the surface
- Maintaining a stable crystallographic lattice



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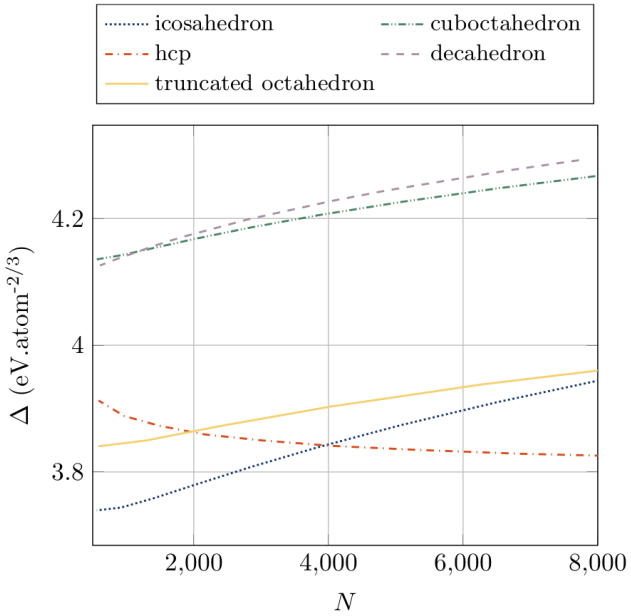
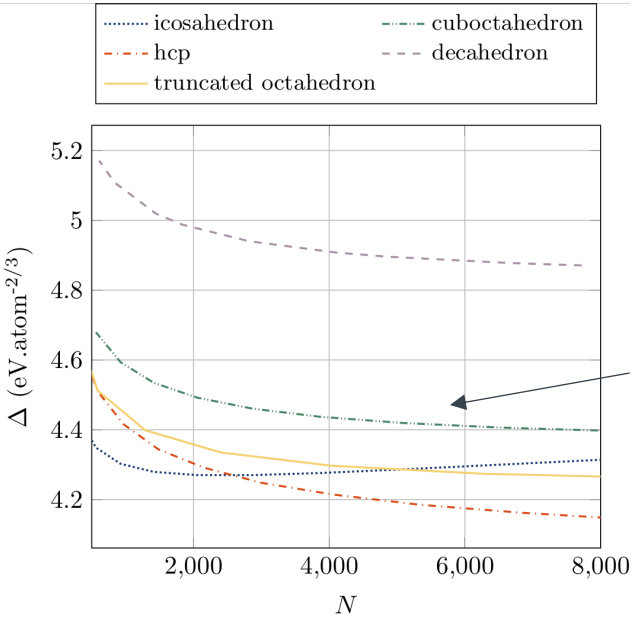
Relative stabilities



q-SNAP:
ico → hcp: ~ 4000 atoms



EAM[1]:
ico → hcp: ~ 2600 atoms

DFT-PBE[2]:
- ico → hcp: ~ 5500 atoms
- cuboctaedron and decahedron highly unstable compared to the three other shapes

The EAM predicts the cuboctahedron to be much more stable than the decahedron.

[1]G. P. P. Pun and Y. Mishin, **Embedded-atom potential for hcp and fcc cobalt**, *Phys. Rev. B* 86, 134116 (2012).
[2]B. Farkaš and N. H. de Leeuw, **Towards a morphology of cobalt nanoparticles: size and strain effects**, *Nanotechnology* 31, 195711 (2020).

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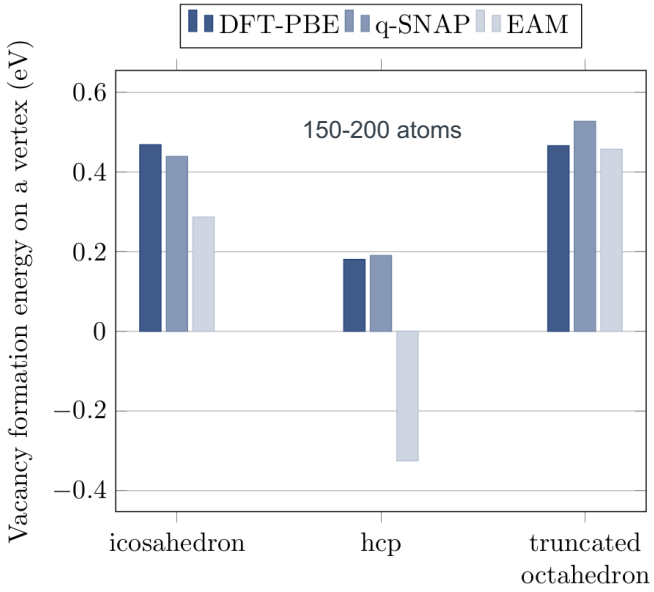
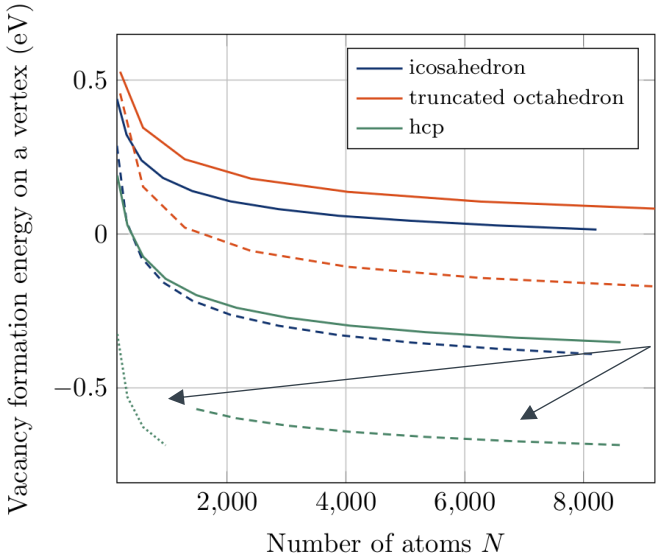
Vacancy formation energy on vertices

- The SNAP is in better agreement with DFT-PBE than the EAM[1] which predicts it is almost always favorable to remove a vertex.





Plain lines: q-SNAP
Dotted lines: EAM

Different minimization paths: for small sizes, the edge is shifted to fill the vacancy

[1]G. P. P. Pun and Y. Mishin, **Embedded-atom potential for hcp and fcc cobalt**, *Phys. Rev. B* 86, 134116 (2012).

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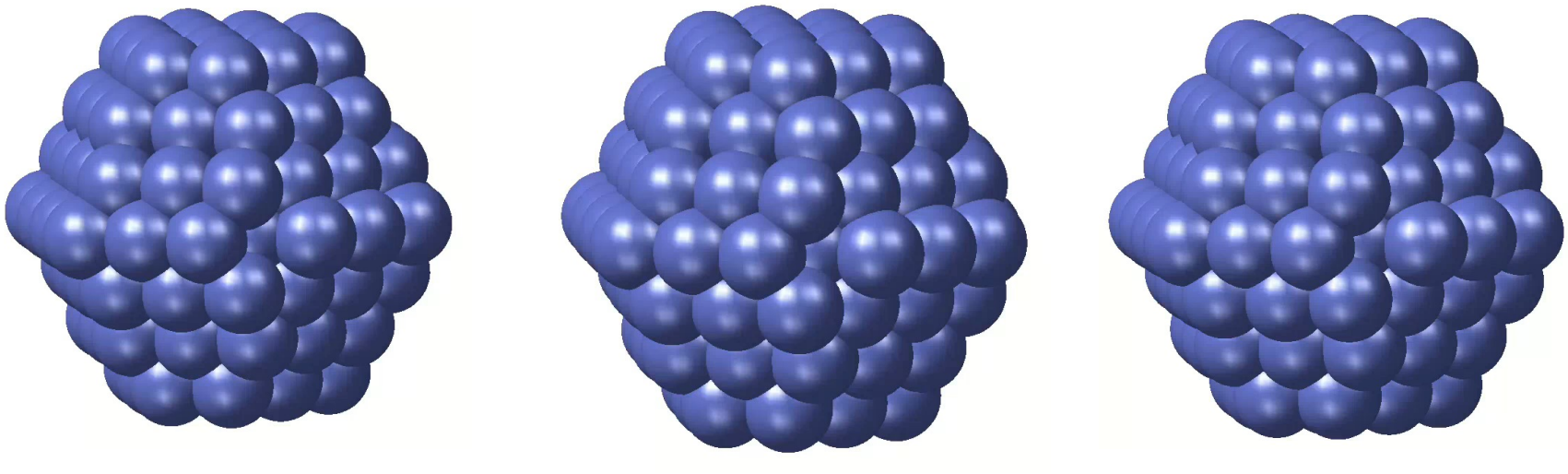
Vacancy formation energy on vertices



DFT-PBE

q-SNAP

EAM[1]

[1]G. P. P. Pun and Y. Mishin**, Embedded-atom potential for hcp and fcc cobalt**, *Phys. Rev. B* 86, 134116 (2012).

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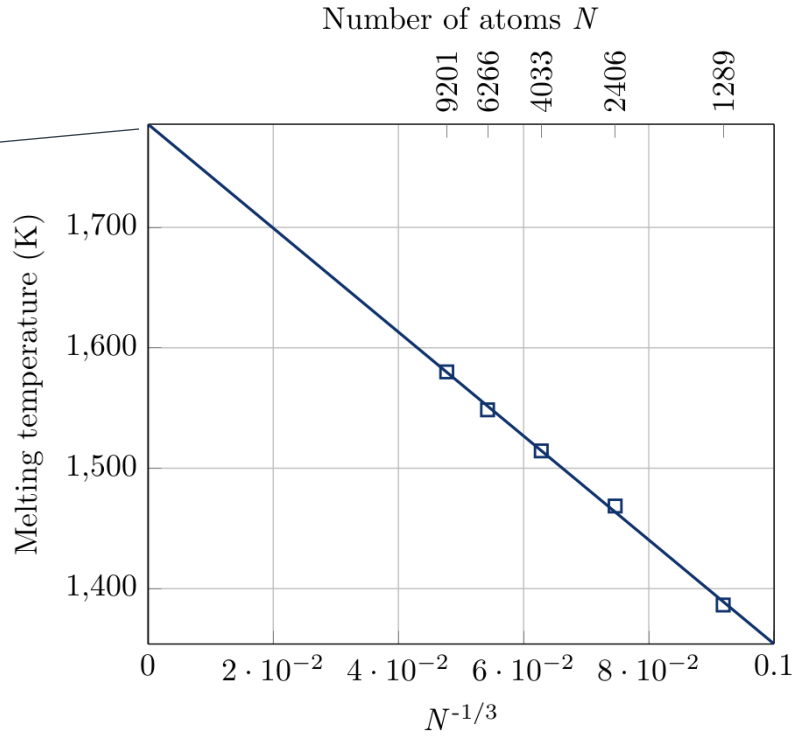
Melting point

Predicted value: 1785 K
Experimental melting point: 1768 K



Melting nanoparticles of increasing size led us to the bulk melting point of cobalt, showing that the q-SNAP is able to correctly reproduce the dynamics of Co nanoparticles.

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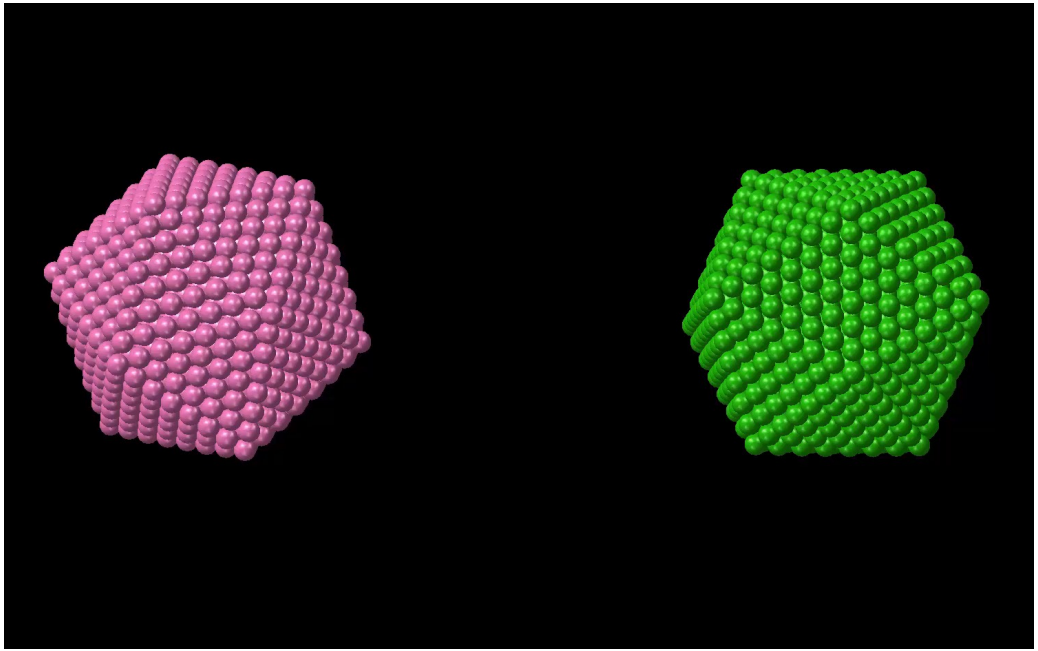
Collision



Initial velocity: 2 Ang/ps

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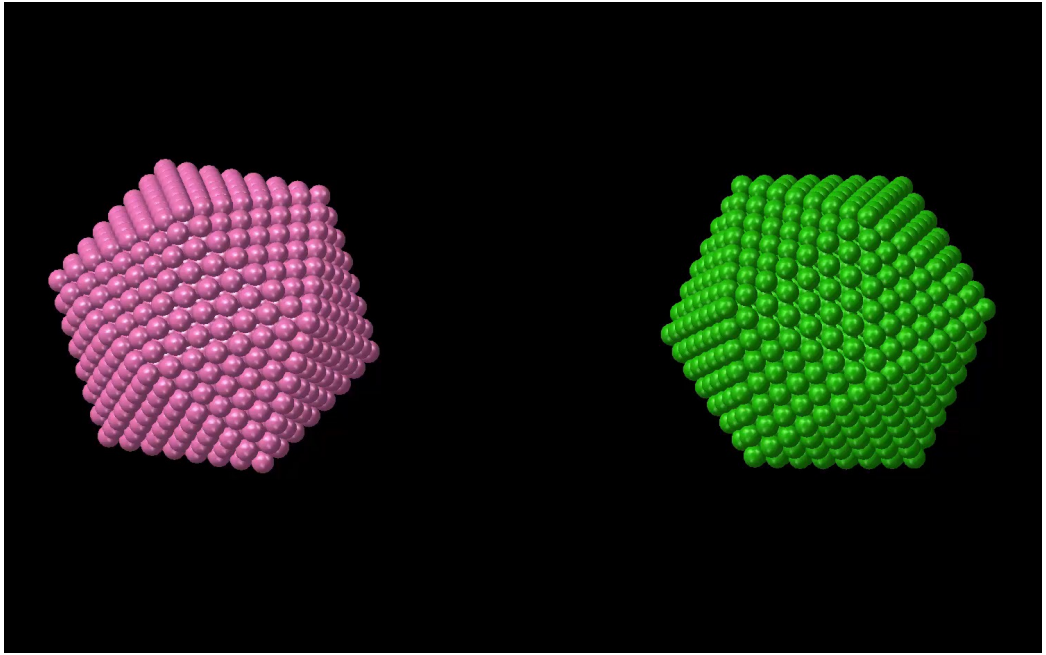
Collision



Initial velocity: 6 Ang/ps

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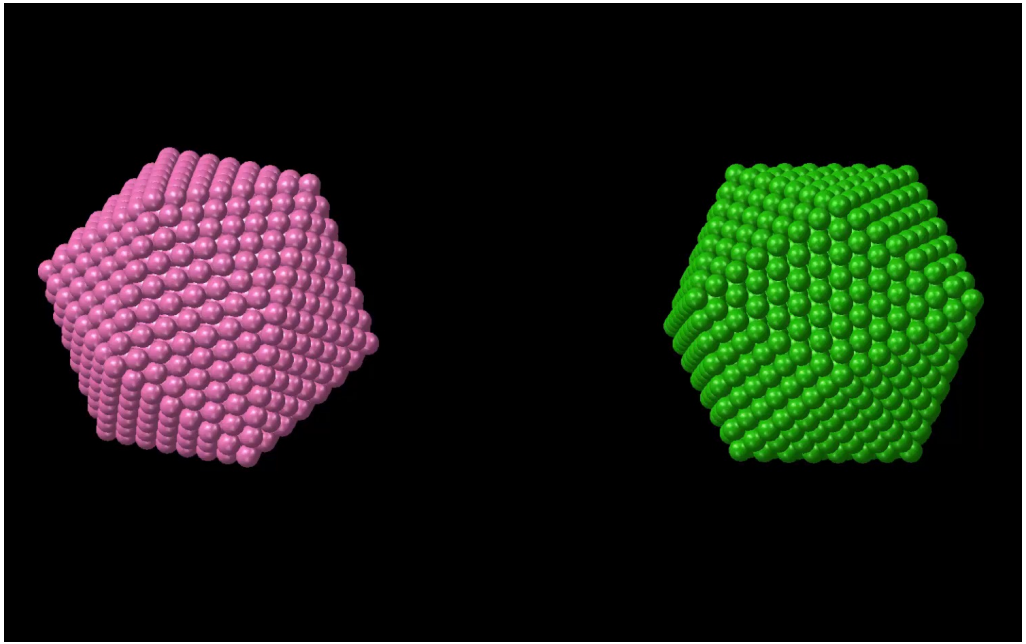
Collision



Initial velocity: 12 Ang/ps

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Summary and conclusion

- This q-SNAP for Co was entirely created using MedeA software.
- It reproduces very well DFT-PBE results but also experimental properties of Co.
- It can be used for nanoparticles modeling.
- In case of catalysis, we can use it to study surface patterns of Co nanoparticles.
- It could be further extended to other elements to model catalysis reactions.

Acknowledgements

Pr. Ryoji Asahi, University of Nagoya, Japan

Pr. Jérôme Creuze, University of Paris-Saclay, France

All colleagues from Materials Design

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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

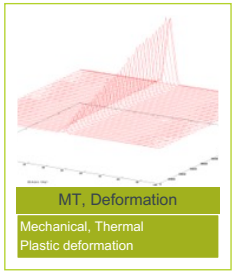
***MedeA* MLPG:** Fully integrated workflow from training-set generation (using *MedeA* HT) and MLP generation to MLP application using *MedeA* LAMMPS

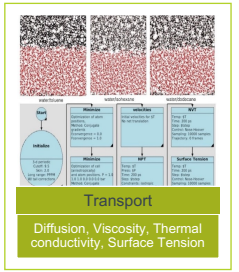
***MedeA* HT:** Generation of large and consistent sets of computed data for input to machine learning procedures

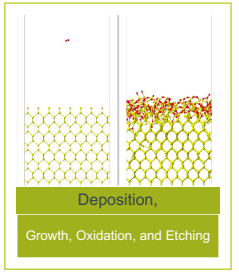
***MedeA* LAMMPS:** Full access to the LAMMPS Code via a graphical user interface based on flowcharts to perform forcefield calculations using MLPs generated by *MedeA* MLPG

MedeA Builders: The *MedeA Environment* includes a rich set of builders for many types of systems and applications. Use *MedeA's* Builders for constructing atomic-scale models of hard, soft or fluid materials.

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







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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*:
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On-the-fly Machine Learning Forcefields with *MedeA* VASP:
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Question and Answer Session

Marthe Bideault

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Dr. Erich Wimmer

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Questions about Materials Design Webinars

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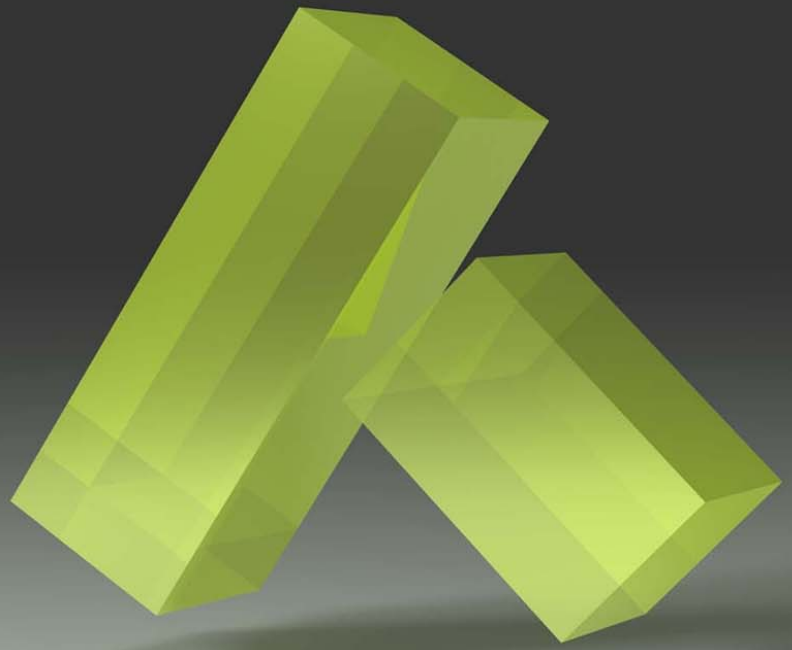

MedeA

Innovation by Simulation