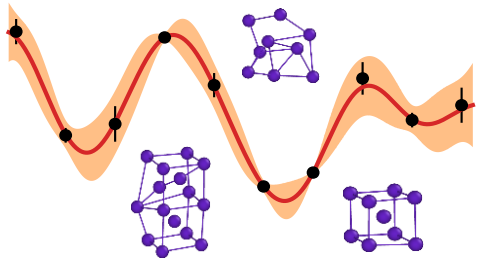
Materials simulations from first principles and Molecular Dynamics with application in the fusion R&D

Jesper Byggmästar, Mikko Koskenniemi, Guanying Wei, Kai Nordlund and Flyura Djurabekova

University of Helsinki



HELSINGIN YLIOPISTO
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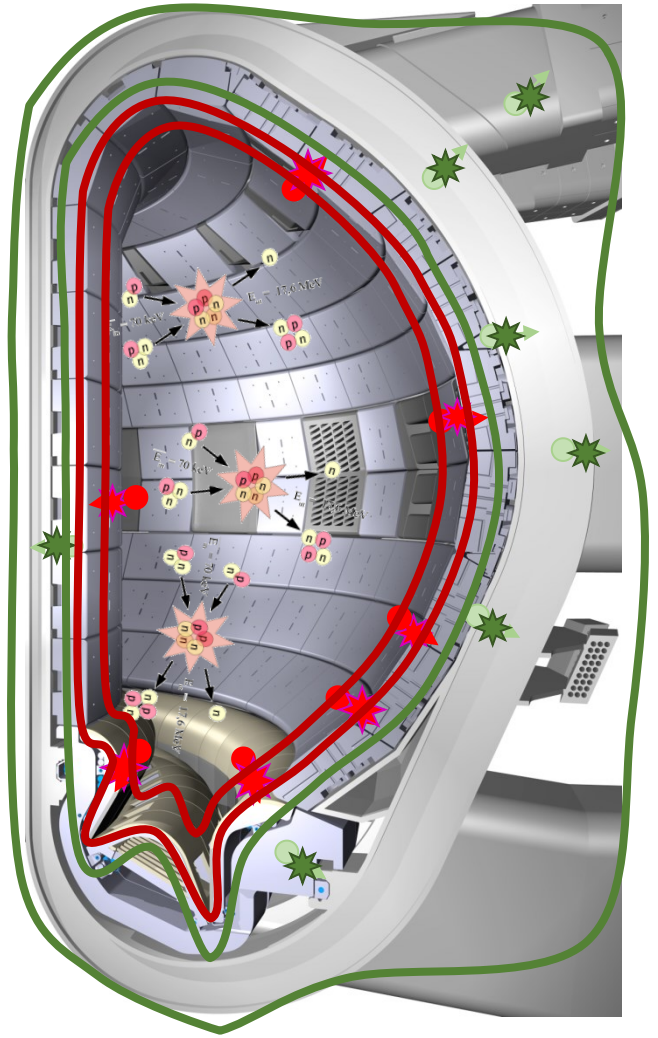
Damage on materials from fusion plasmas

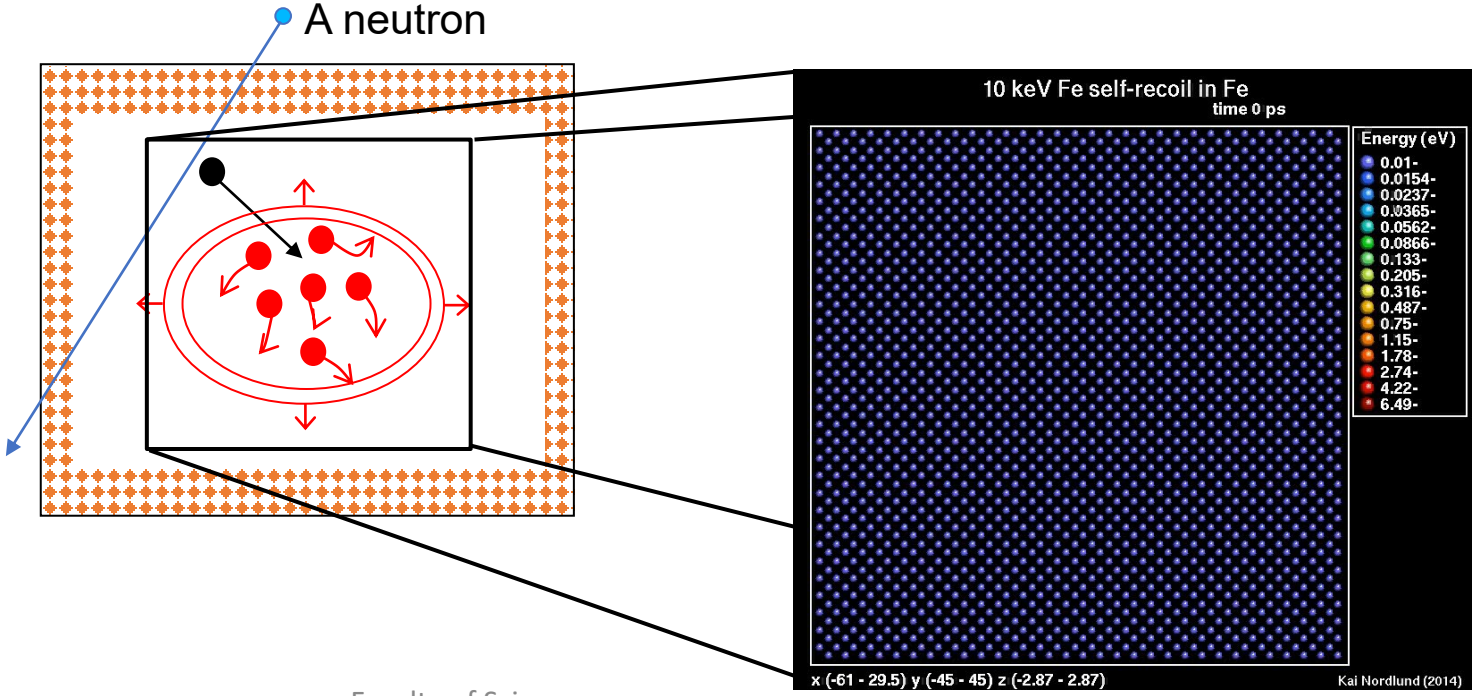
- Physics of ion-surface interactions is able to help to develop durable materials for application in fusion reactors. The two main sources of damage on materials in this condition:
  - ✓ Surface damage from hot nuclei hitting the inner walls (plasma-material interactions)
  - ✓ Damage on materials everywhere from the ~ 14 MeV neutrons produced in the fusion reaction



Nature of neutron damage

- The neutron cross-section is low → travel deep
- Yet they collide with some probability with nuclei and give it a high recoil energy (keV's to 100's of keV's)

Damage event by 10 keV Fe recoil in Fe

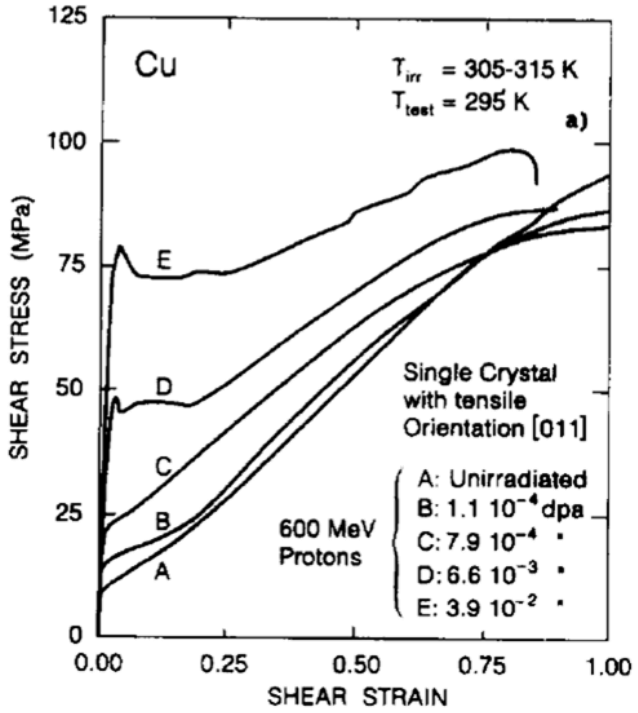


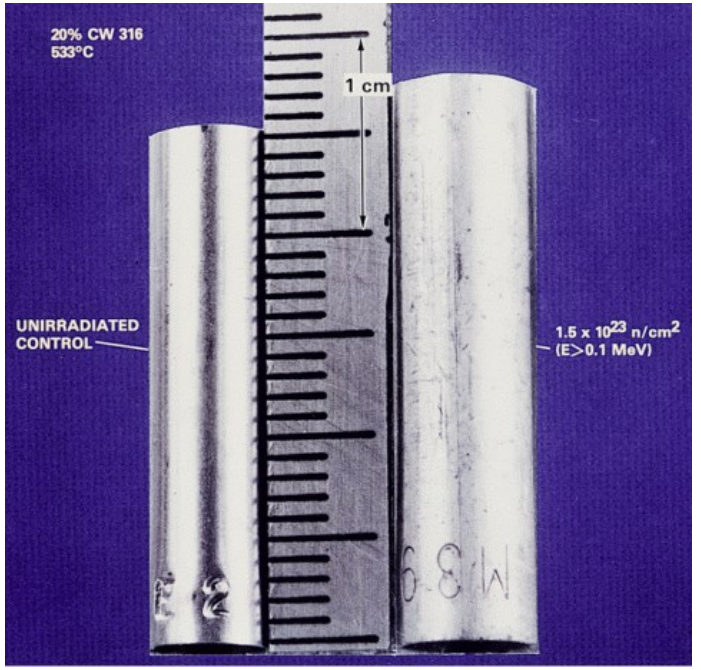
[K. Nordlund et al, Nature comm. **9**, 1084 (2018)]

Radiation damage in nuclear reactors

- Via a complex set of evolutional processes, this nanoscale damage eventually leads to major macroscopic consequences: changes of mechanical properties of materials, swelling, embrittlement, …

Test samples: many years in a **fission** reactor

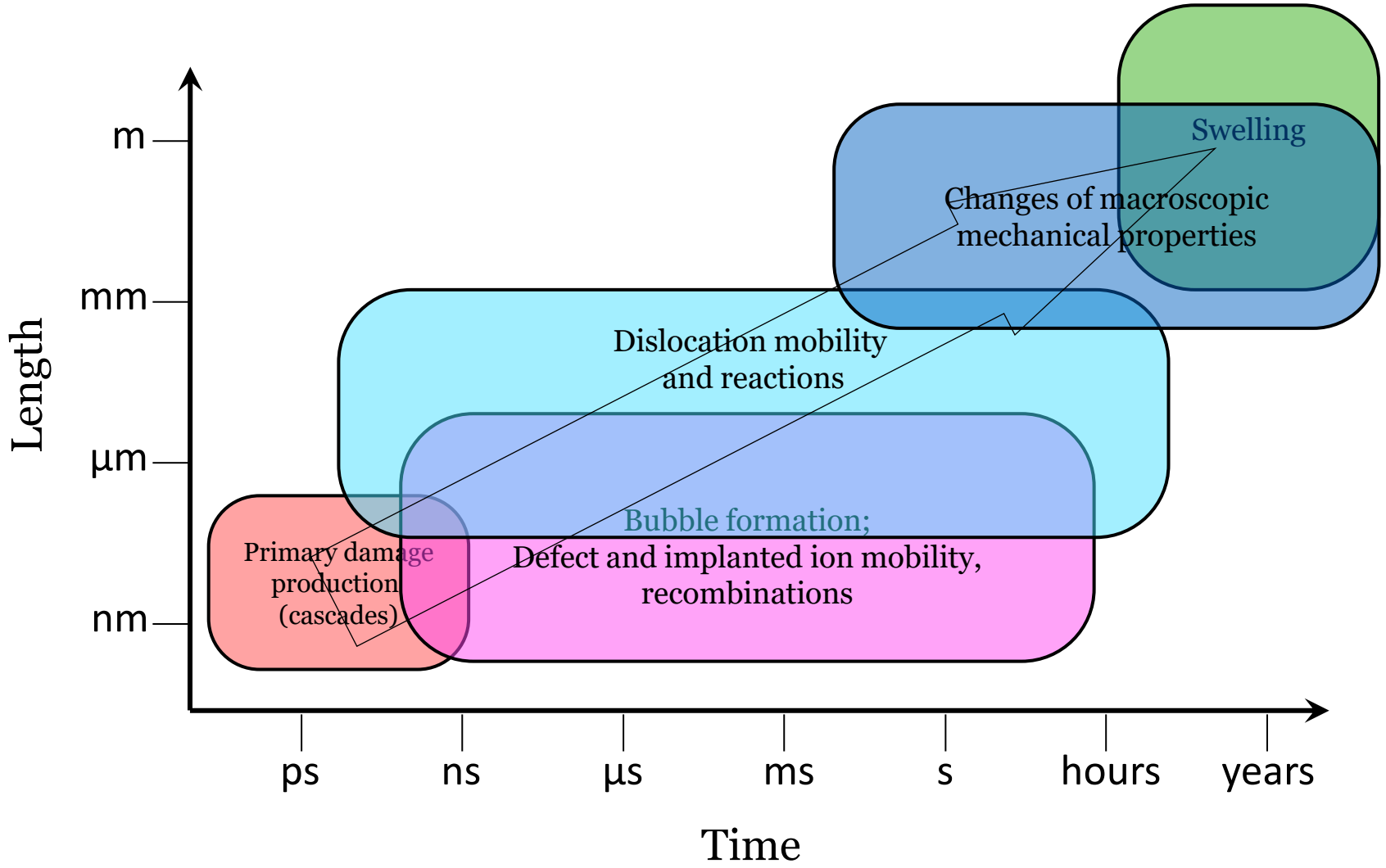




[B.N. Singh, A.J.E. Foreman, H. Trinkaus, Journal of Nuclear Materials **249,** (1997) 103-115]

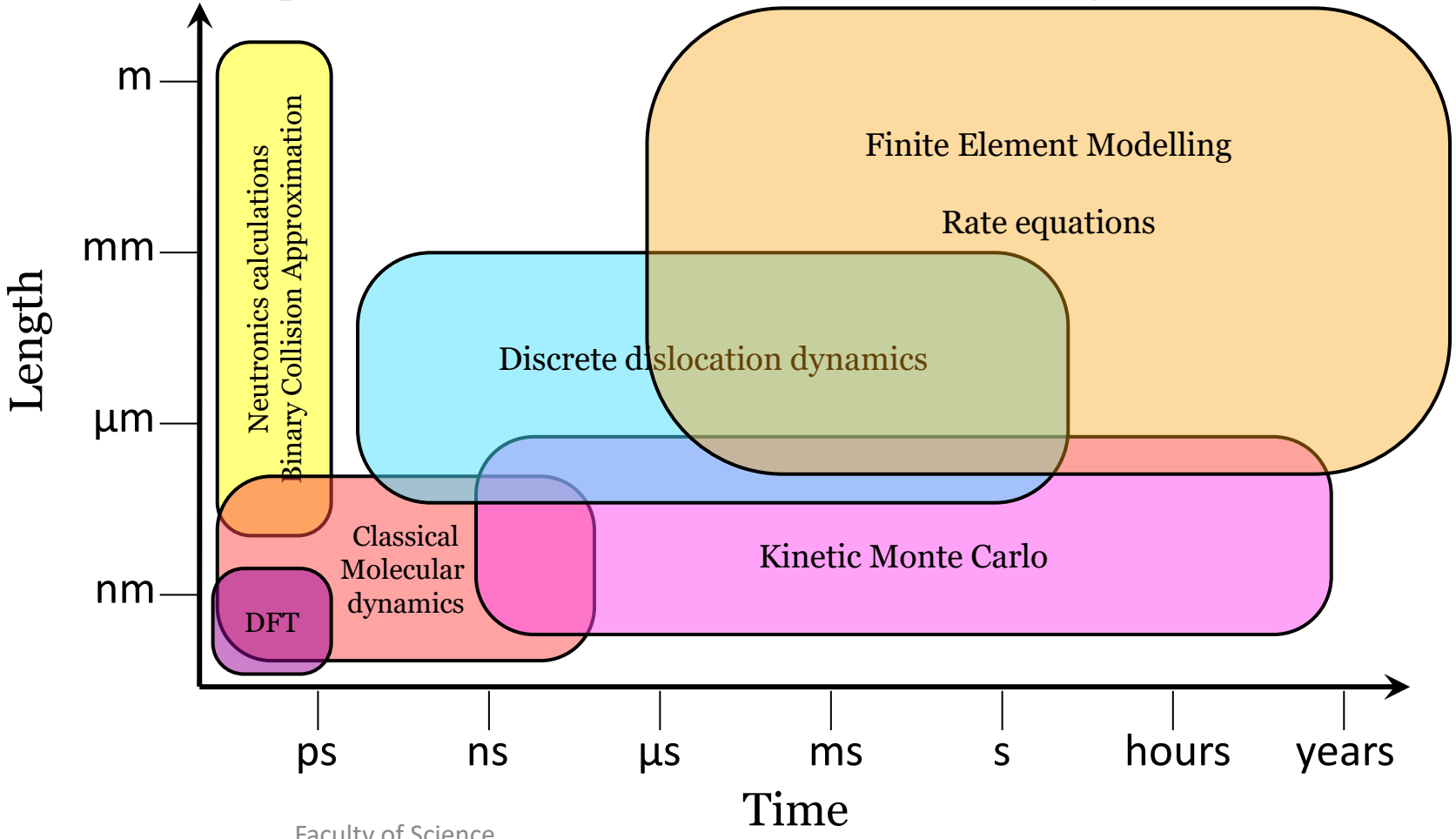
http://cmcsn.phys.washington.edu/book/export/html/467

Multicale physics during the continued neutron irradiation



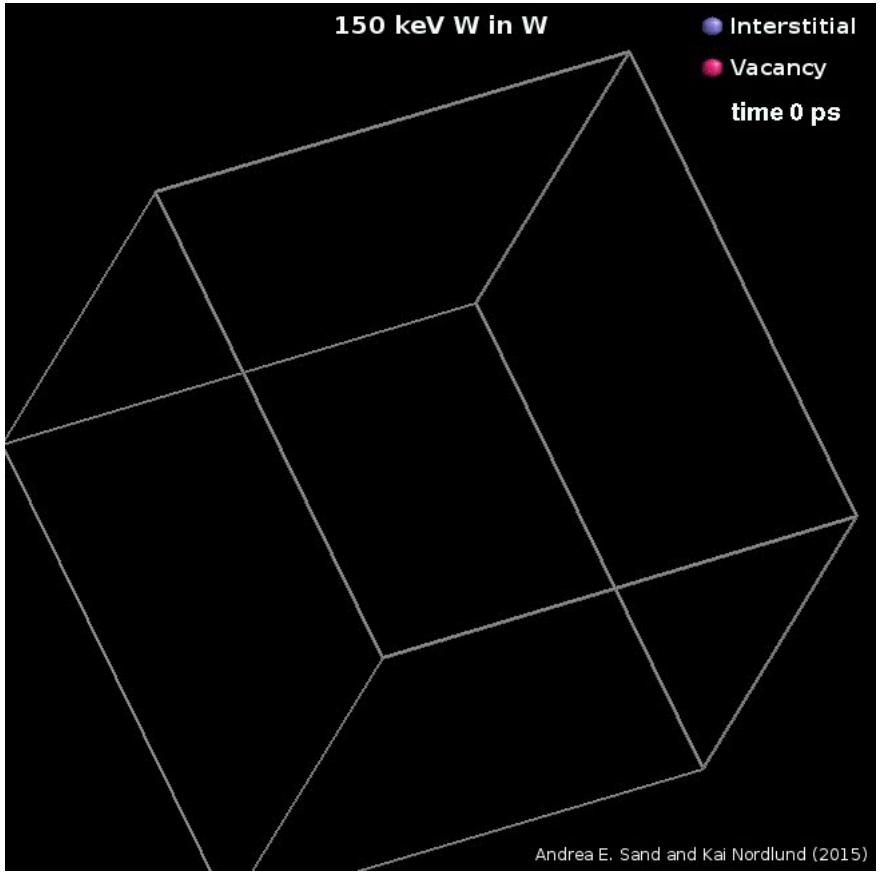
Materials multiscale modelling framework

- Sequential and concurrent multiscale modelling
- In Europe EUROFUSION WPMAT IREMEV has great coordination



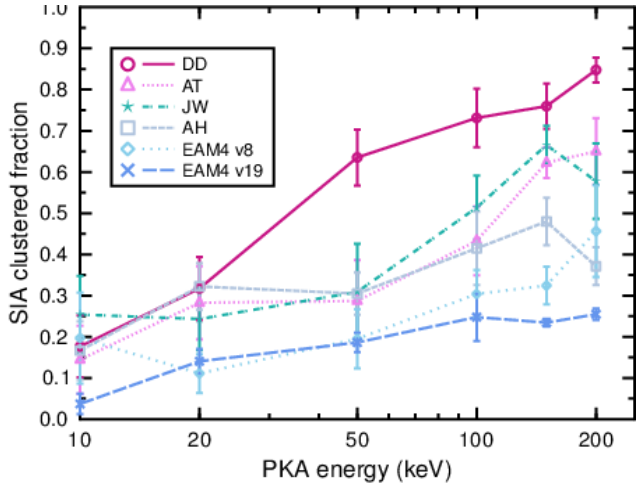
Damage by fusion neutrons

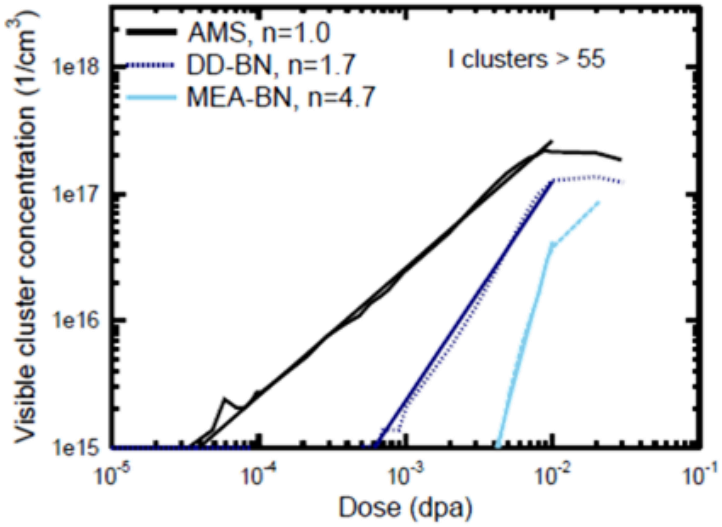
- A typical neutron recoil E in W
  - ✓ in fission is about 10 keV
  - ✓ in fusion is about 150 keV
- The higher energy can produce huge damage clusters immediately => qualitative difference between fission and fusion!



Why does the damage clustering matter?

- There is clear evidence from atomistic simulations that the long-term damage evolution is dominated not by point defects, but by large clusters
- This is (unfortunately) also where the simulation model reliability limit comes in:
  - ✓ different interatomic potentials predict different fractions of damage in large clusters

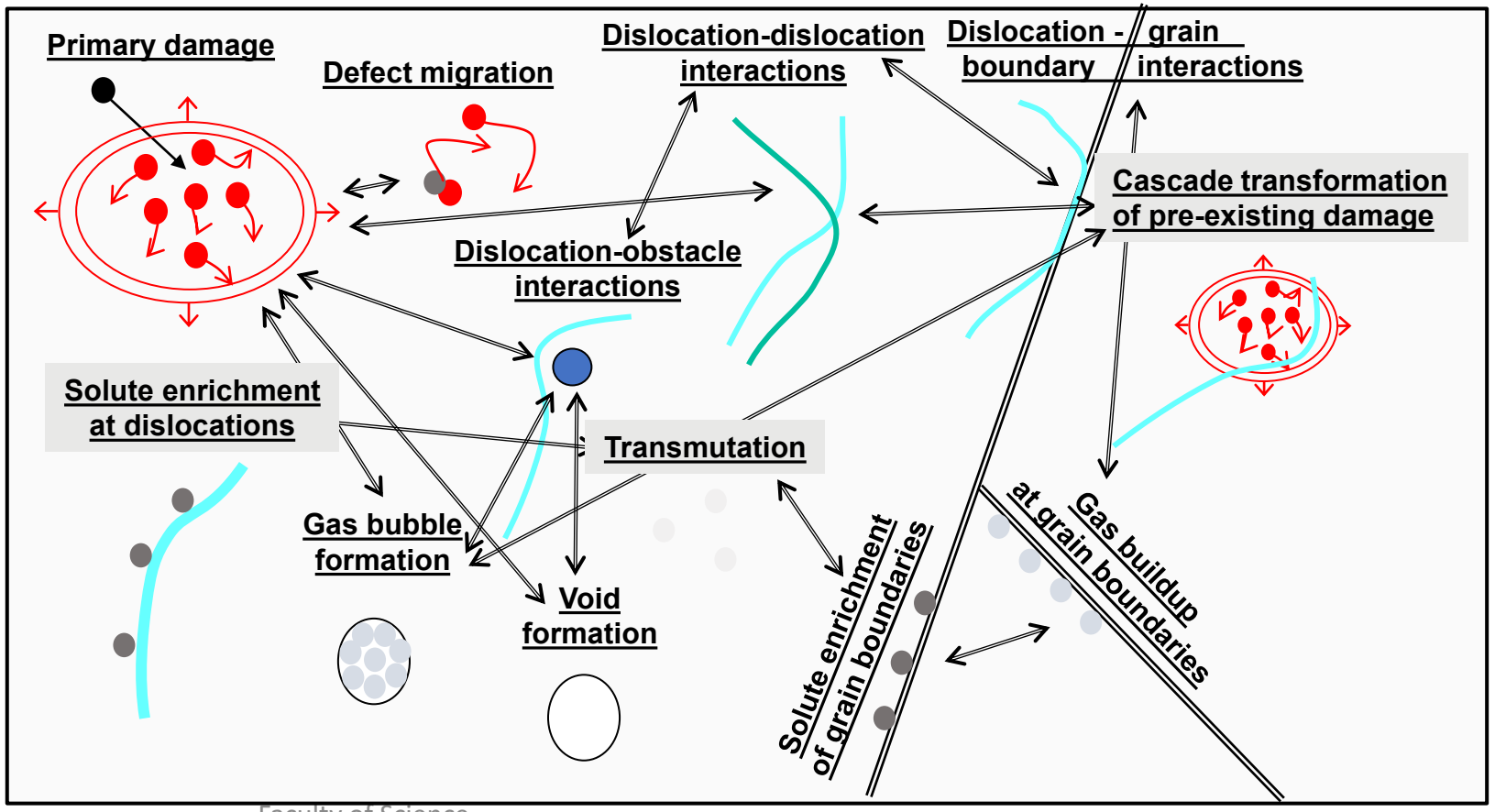




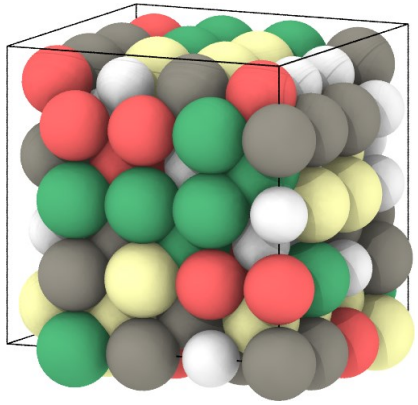
[C. Björkas, K. Nordlund, and M. J. Caturla, Phys. Rev. B **85**, 024105 (2012)]

Complexity of processes

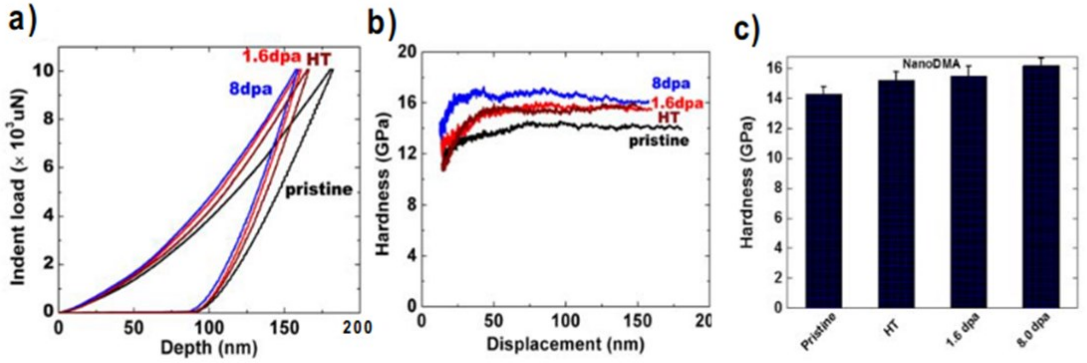
- All of the following are known to happen. However, many of these processes cannot be simulated predictively alone, and the concerted actions even more so



Our latest contribution to modeling fusion-related materials with focus on W-based refractory high-entropy alloys



- Body-centred cubic
  - ✓ Mo-Nb-Ta-V-W, W-Ta-Cr-V
- Promising irradiation resistance:



- ✓ No dislocation loops?
- ✓ Minor hardening

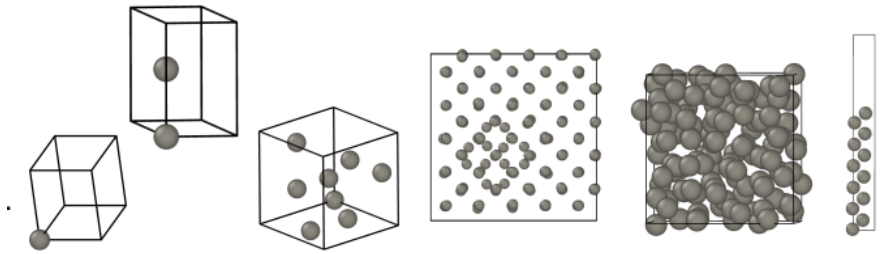
Need for interatomic potentials..!!

Fig. S6. Mechanical response of the HEA. Representative (**a**) load-displacement curves and (**b**) hardness vs displacement curves for the pristine, annealed samples to 1073 K (HT), 1.6 dpa and 8 dpa irradiated samples. Shift of loading curves to the left indicates slight hardening, which is confirmed from the hardness vs displacement curves. (**c**) Nano-hardness (using nanoDMA) values of the pristine, annealed (HT), 1.6 dpa and 8 dpa irradiated HEA samples.

[El-Atwani et al., "Outstanding radiation resistance of tungsten-based HEA." Sci. Adv. 2019; 5]

Machine-learning potential

Energies & forces of structures computed by density functional theory (DFT)



"Descriptor space"

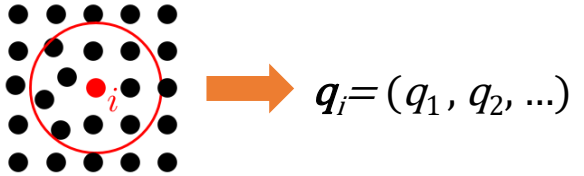
Train ML model

$$E = f_{ML}(\boldsymbol{q}_i)$$

Energy prediction (per atom)
& analytical force computation

Descriptor

- encodes local atomic environment into ”machine-learnable” input (i.e. numbers),
- Should satisfy physical symmetries (permutation, translation, rotation)



Gaussian approximation potentials

$$E = f_{ML}(\boldsymbol{q}_N)$$

AP Bartók, G Csanyi et al. (2010, 2013)

Gaussian process regressor:

$$E_i = \sum_s^M \alpha_s K(\boldsymbol{q}_i, \boldsymbol{q}_s)$$

Loop over M known environments/descriptors

Descriptors of desired environment i and known environment s

Regression coefficients fitted to DFT data

Kernel, measure of similarity between inputs

Gaussian approximation potentials

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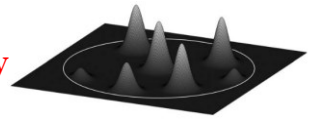
This can combine multiple descriptors, for example:

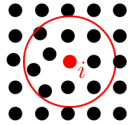
$$E_i = \sum_s^{M_{2b}} \alpha_s K_{2b}(\boldsymbol{q}_i, \boldsymbol{q}_s) + \sum_s^{M_{3b}} \alpha_s K_{3b}(\boldsymbol{q}_i, \boldsymbol{q}_s) + \sum_s^{M_{mb}} \alpha_s K_{mb}(\boldsymbol{q}_i, \boldsymbol{q}_s)$$

Atom pairs

Atom triplets

Many-body





Some descriptors in GAP:

- SOAP [Bartók et al. (2013)]
- 2-body, 3-body
- EAM density (this work): $\rho_i = \sum_j \varphi(r_{ij})$

Gaussian approximation potentials

$$E = f_{ML}(\boldsymbol{q}_N)$$

AP Bartók, G Csanyi et al. (2010, 2013)

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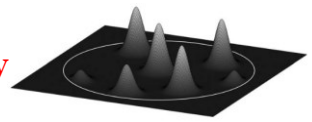
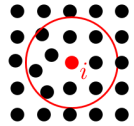
Atom pairs

Atom triplets

Many-body



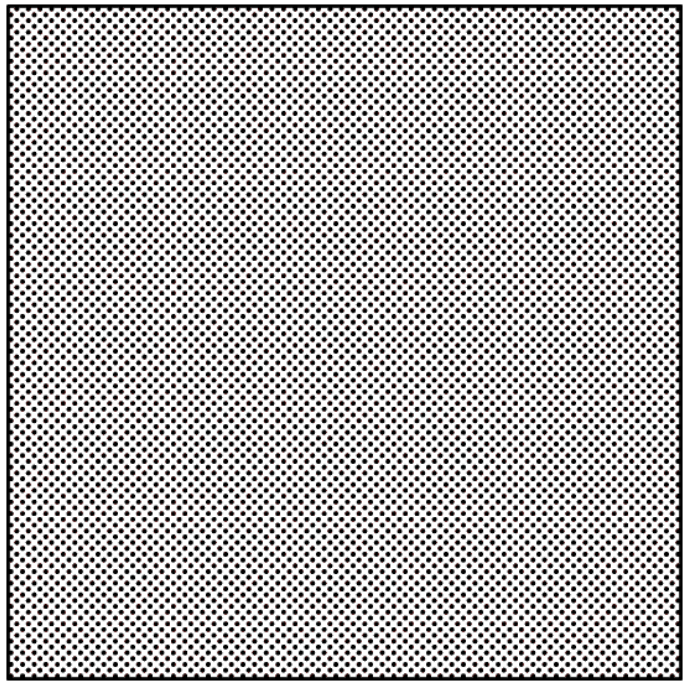


External analytical potential and machine-learnt E_{DFT} can be added: $E_{ext.}$

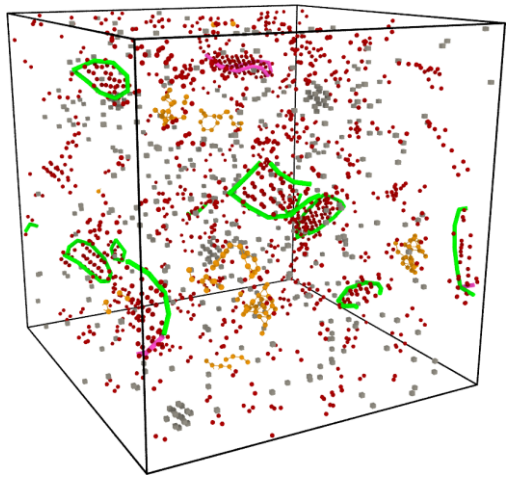
$$E_i = E_{\mathrm{ext.}}(\boldsymbol{r}) + \sum_s^{M_{2b}} \alpha_s K_{2b}(\boldsymbol{q}_i, \boldsymbol{q}_s) + \sum_s^{M_{3b}} \alpha_s K_{3b}(\boldsymbol{q}_i, \boldsymbol{q}_s) + \sum_s^{M_{mb}} \alpha_s K_{mb}(\boldsymbol{q}_i, \boldsymbol{q}_s)$$

Modelling radiation damage

- Collision cascade (10 keV in Fe):



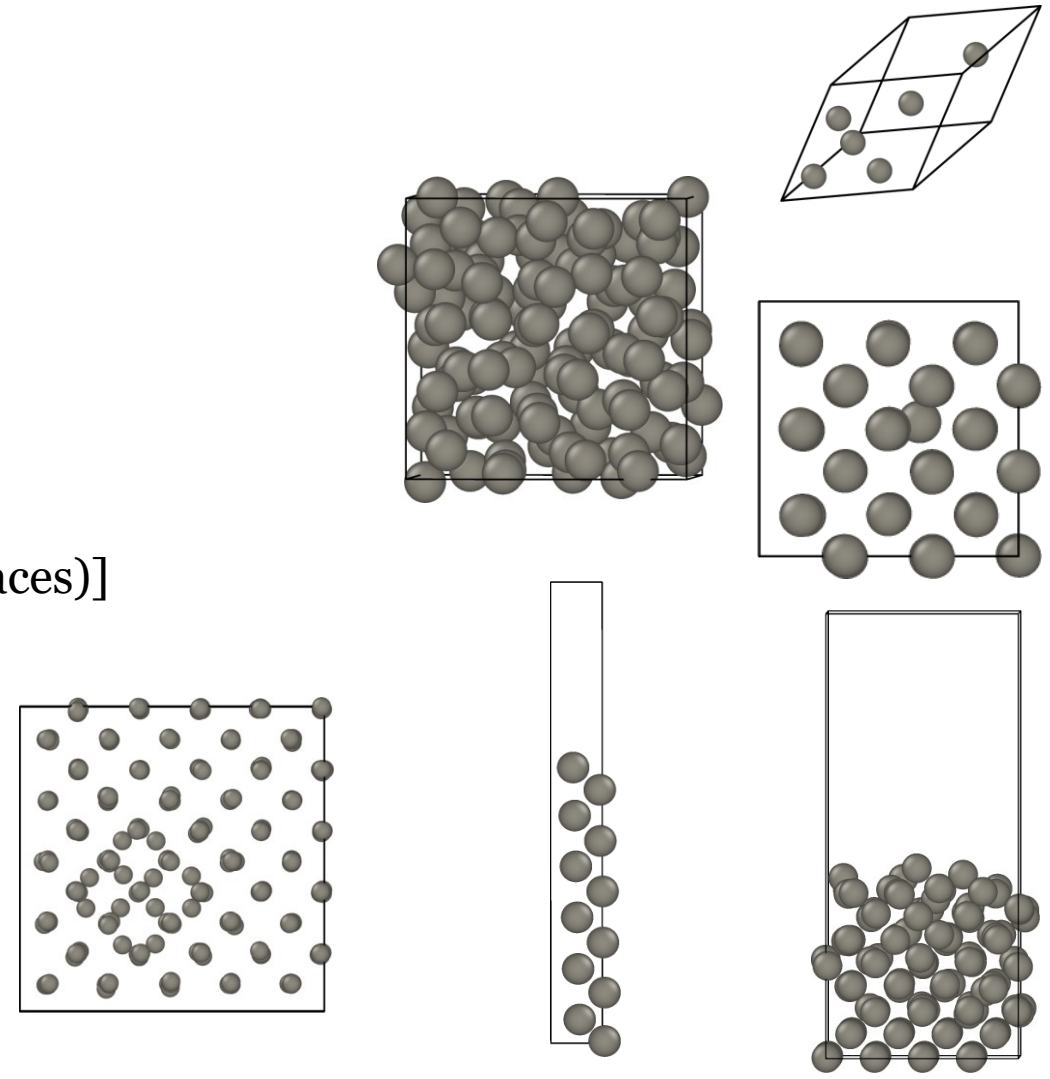
- ✓ Local melting
- ✓ Extreme repulsion
- ✓ Extreme temperature and pressure gradients
- ✓ Simple and complex defect structures
  - → lattice strain fields and swelling



<- high-dose defect structure in Fe

GAP for radiation damage – training structures

- Diverse training database needed!!
  - ✓ Wide range of strain/volumes
  - ✓ Wide range of temperatures
  - ✓ Liquids
  - ✓ Single defects and defect clusters
  - ✓ [Surfaces (including damaged surfaces)]



GAPs for pure BCC metals: W, Mo, Nb, Ta, V

Refitted ZBL-
like

SOAP kernel & descriptor

$E_i =$ ... $, \boldsymbol{q}_s)$

PHYSICAL REVIEW B **100**, 144105 (2019)

Editors' Suggestion

Machine-learning interatomic potential for radiation damage and defects in tungsten

J. Byggmästar,[1,*] A. Hamedani,[1,2] K. Nordlund,[1] and F. Djurabekova[1,3]
[1]*Department of Physics, P.O. Box 43, FI-00014 University of Helsinki, Finland*
[2]*Engineering Department, Shahid Beheshti University, G.C, P.O. Box 1983969411, Tehran, Iran*
[3]*Helsinki Institute of Physics, FI-00014 University of Helsinki, Finland*

PHYSICAL REVIEW MATERIALS **4**, 093802 (2020)

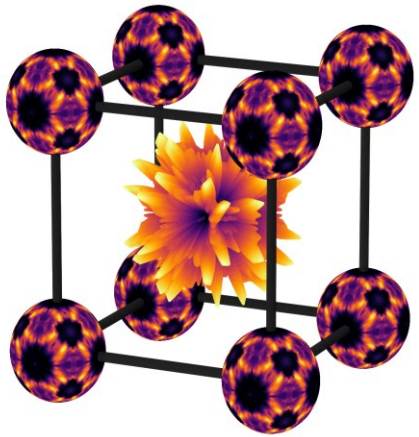
Gaussian approximation potentials for body-centered-cubic transition metals

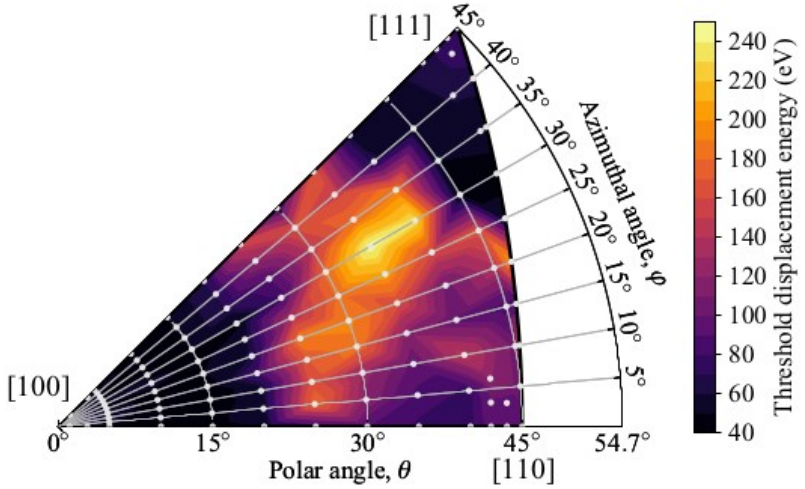
J. Byggmästar,[1,*] K. Nordlund,[1] and F. Djurabekova[1,2]
[1]*Department of Physics, P.O. Box 43, FI-00014 University of Helsinki, Finland*
[2]*Helsinki Institute of Physics, Helsinki, Finland*

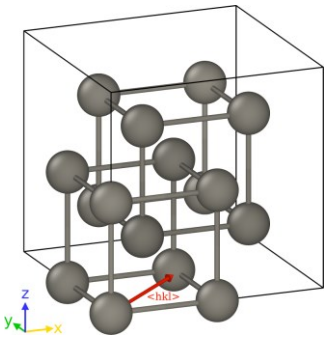
J. Byggmästar, A. Hamedani, K. Nordlund, and F. Djurabekova, Phys. Rev. B, **100**, 144105 (2019)
J. Byggmästar, K. Nordlund, and F. Djurabekova, Phys. Rev. Materials, **4**, 093802 (2020)

Basic properties for pure W, Mo, Nb, Ta, V in GAP

- Threshold displacement energy surface of W:

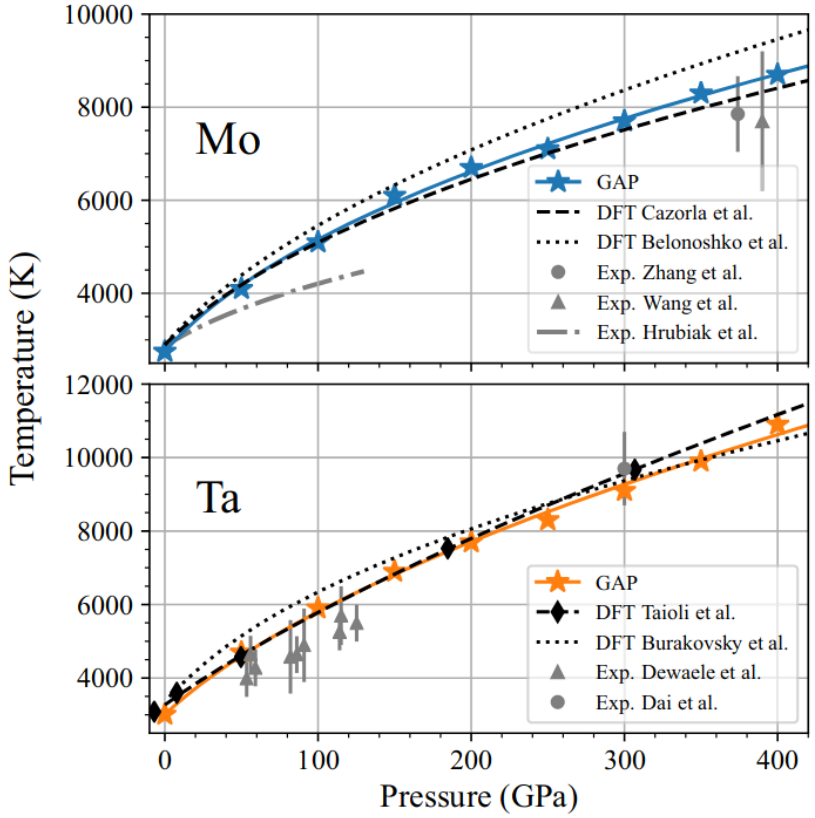




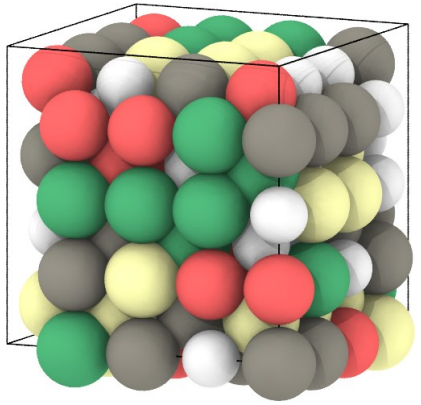


Minimum in <100> is 45.5 eV (expt. 42 eV),

Melting curves



GAP for Mo-Nb-Ta-V-W alloys!?

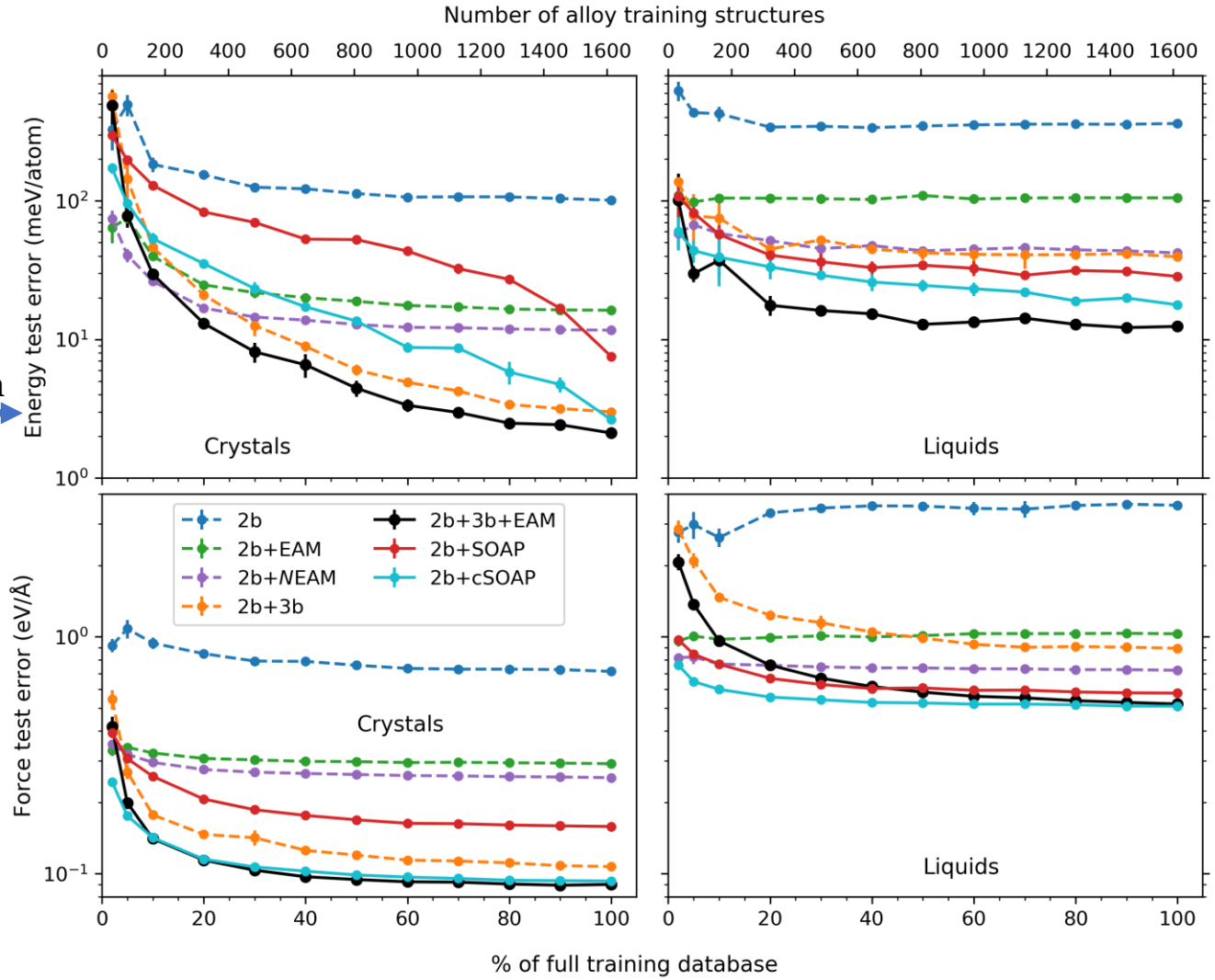


Challenges:

- Huge composition space ➔ huge training data sets!?
- Many-body descriptor vector length scales poorly with number of elements ➔ huge training data sets!?
  - ✓ ~ N^2 for SOAP, ~ N for "compressed-SOAP" [J. Darby et al. (2022)]
- Which descriptors to use??
- Possible solution:
  - ✓ rely on simple and data-efficient *low-dimensional descriptors*!
    - 2-body, 3-body, EAM density

Learning curves for Mo-Nb-Ta-V-W potentials

2 meV/atom



Simple "low-dimensional GAP" 2b+3b+EAM outperforms standard high-dimensional many-body GAPs!

tabGAP: tabulated low-dimensional GAPs

- Tabulate GAP predictions → cubic spline interpolations
- Any combination of low-dimensional descriptors:
  - ✓ Two-body (pairs)
  - ✓ Three-body (triplets)
  - ✓ Many-body: EAM-like density $\rho_i = \sum_j \varphi(r_{ij})$

GAP:

$$E = \sum_{ij}^{N} E_{\text{ext.}}(r_{ij}) + \sum_{ij}^{N}\sum_{s}^{M_{2b}} \alpha_s K_{2b}(\boldsymbol{q}_i, \boldsymbol{q}_s) + \sum_{ijk}^{N}\sum_{s}^{M_{3b}} \alpha_s K_{3b}(\boldsymbol{q}_i, \boldsymbol{q}_s) + \sum_{i}^{N}\sum_{s}^{M_{\text{EAM}}} \alpha_s K_{\text{EAM}}(\boldsymbol{q}_i, \boldsymbol{q}_s)$$

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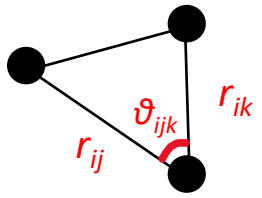
Pair potential: 1D grid

3-body potential: 3D grid

EAM embedding energy: two 1D grids

tabGAP:

$$E = \sum_{ij}^{N} S_{1D}(r_{ij}) + \sum_{ijk}^{N} S_{3D}(r_{ij}, r_{ik}, \cos\theta_{ijk}) + \sum_{i}^{N} S_{1D}(\rho_i), \qquad \rho_i = \sum_{j} s_{1D}(r_{ij})$$

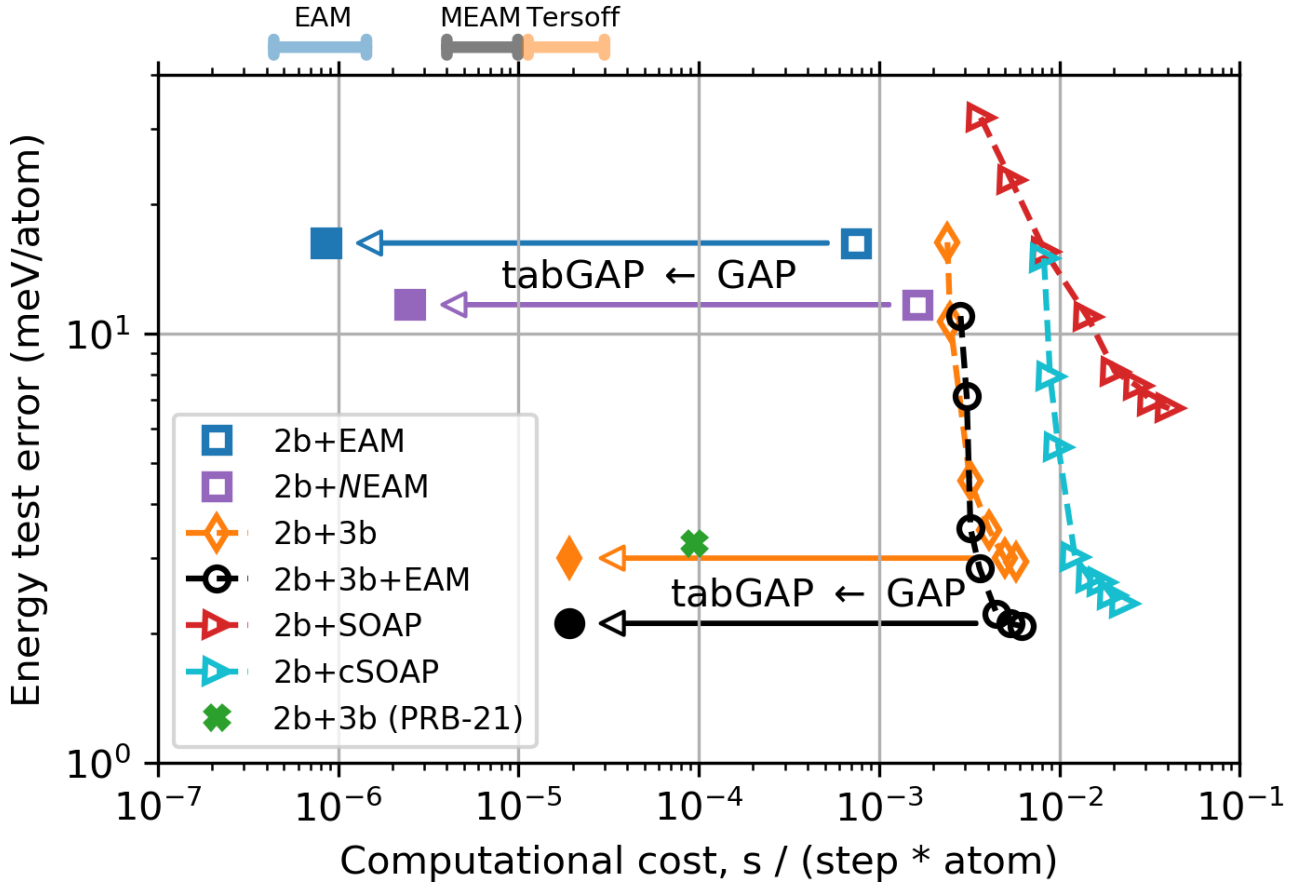


Normal EAM embedding function

Similar work:
- [A. Glielmo, C. Zena, A. De Vita, PRB 97 (2018)]
- FLARE https://github.com/mir-group/flare (J. Vandermause, et al.)

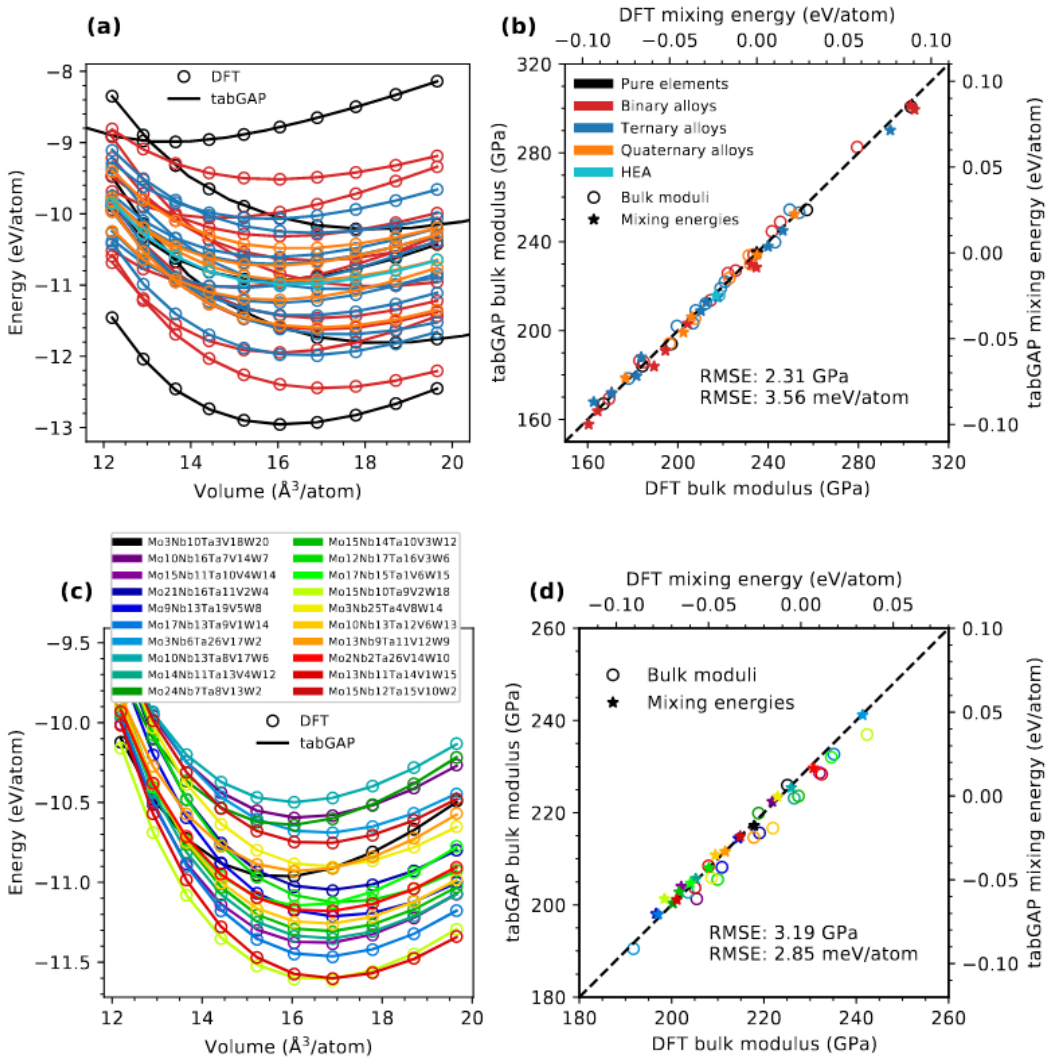
Computational efficiency

Accuracy versus computational cost:



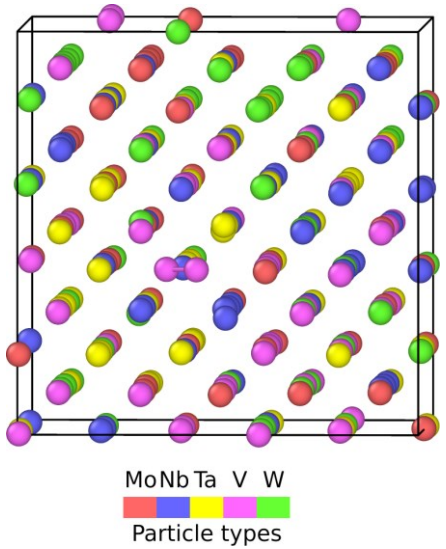
tabGAP for Mo-Nb-Ta-V-W alloys

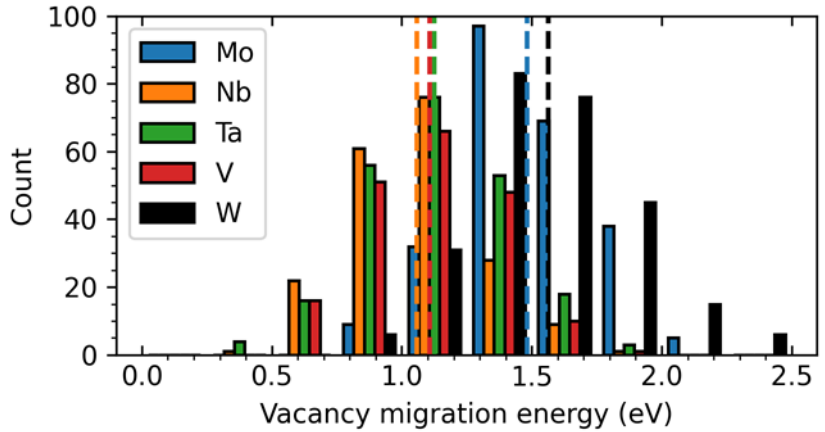
- Comparison of DFT and tabGAP predictions for energy per atom as a function of atomic volume,
  - ✓ a) for all equiatomic compositions for pure, binary, ternary, quaternary and HEA alloys, while in c) the alloy compositions sampled randomly
  - ✓ b) and d) bulk moduli and mixing energies deduced from the curves in a) and c), respectively.



[J. Byggmästar, K. Nordlund, and F. Djurabekova PRB 104, 104101 (2021)]

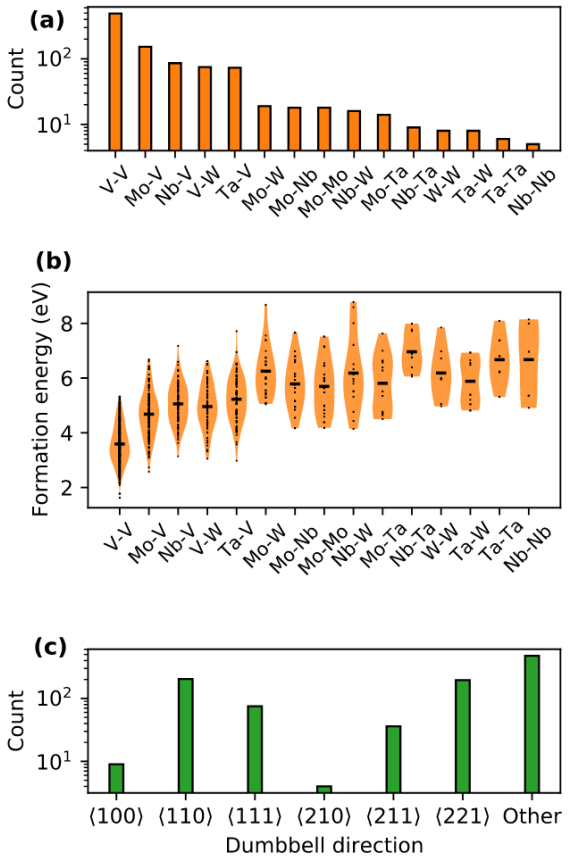
Vacancies and SIAs in MoNbTaVW



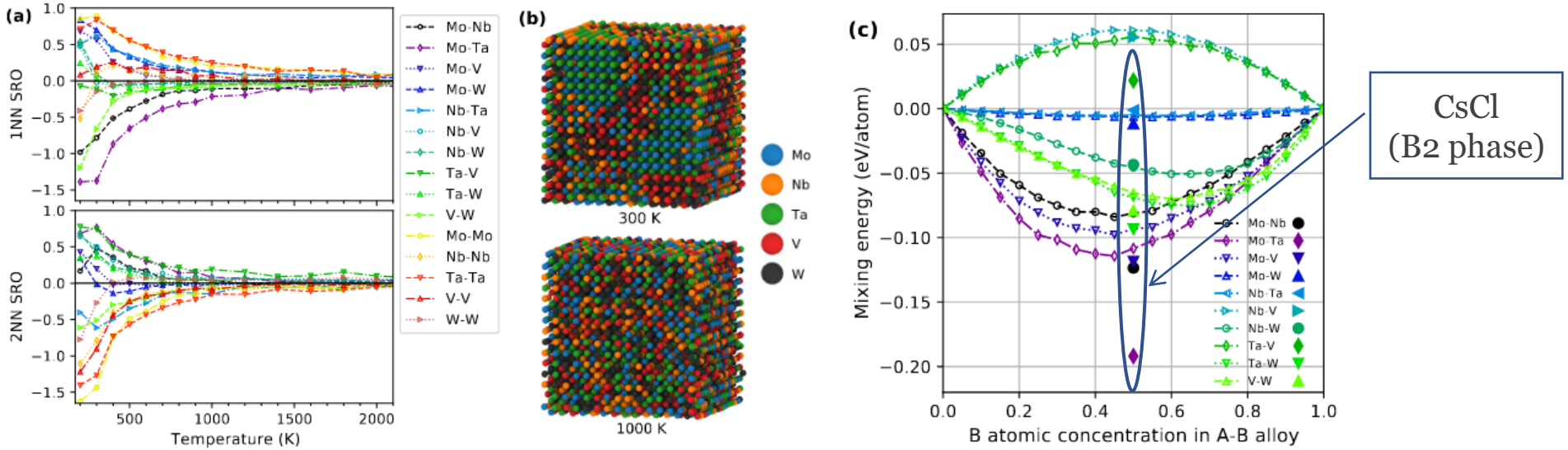


- Strong effects of local chemical neighborhood:
  - ✓ Wide distribution of migration energies!
  - ✓ SIAs not always <111> like in pure metals!
  - ✓ SIAs do not migrate in 1D along <111> with the minimal migration energy!
  - ✓ Avg. 1 eV+ in MoNbTaVW

Self-interstitial atoms:



Short-range ordering in single-crystal MoNbTaVW



- SRO (short-range order) in MoNbTaVW.
  - ✓ SRO parameter as a function of temperature Monte-Carlo +MD simulations for nearest (1nn) and the second nearest (2nn) neighbors.
- Strong energetic preference for (mainly) binary orderings --> explained by binary mixing curves (c)

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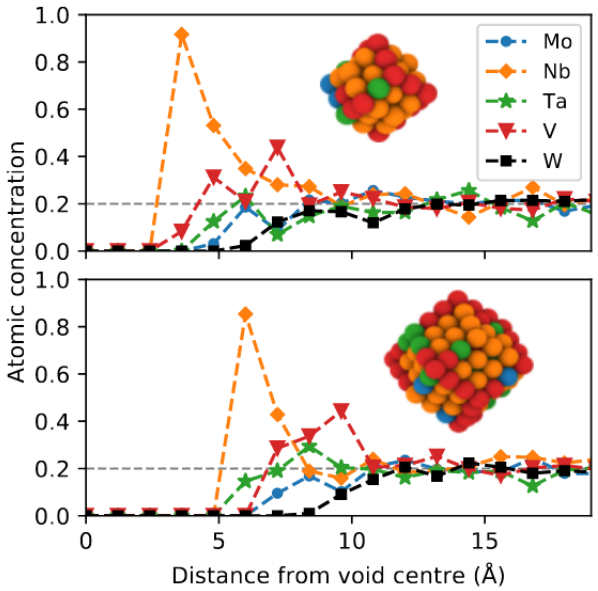
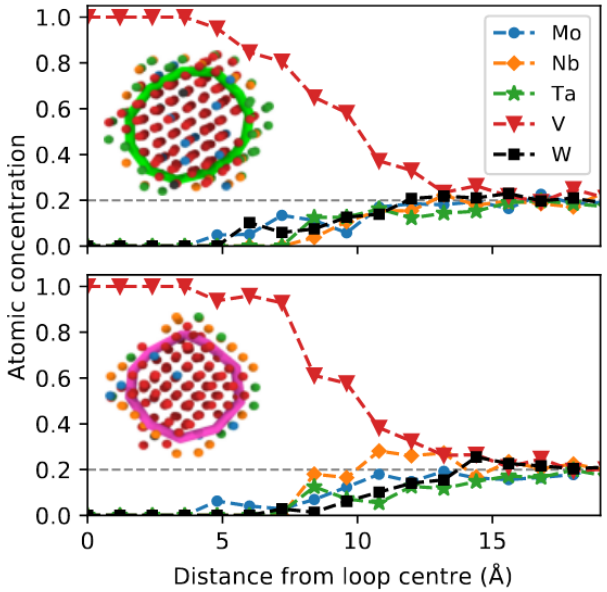
Segregation around defect clusters

Result: Nb → Voids



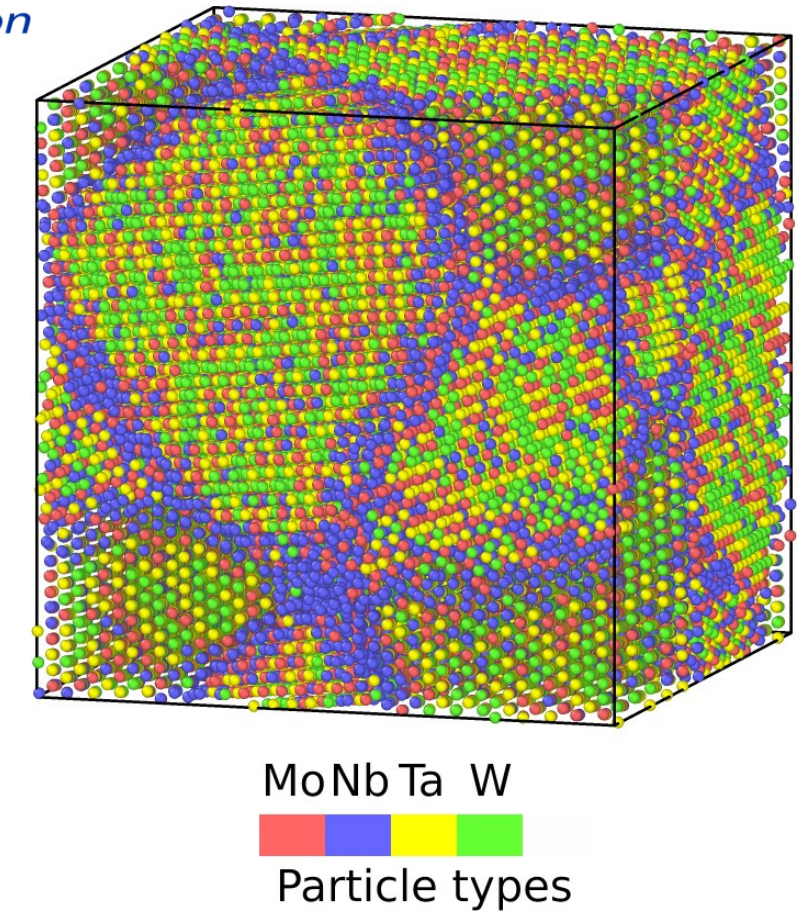
Result: V → interstitial loops

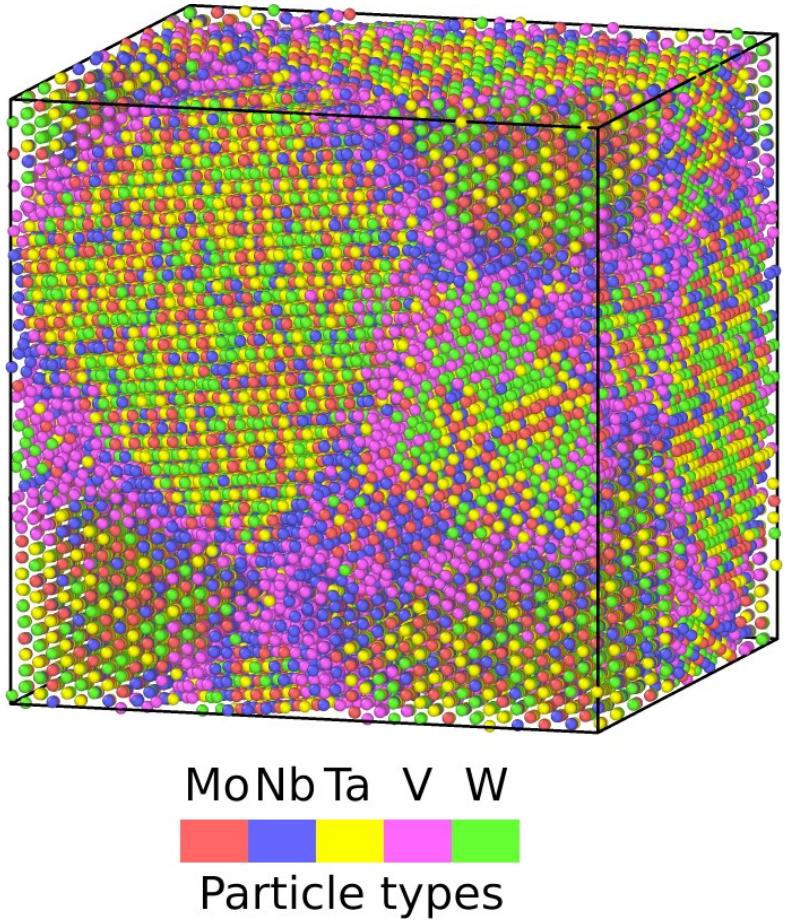


- Segregation around voids: We see that large atoms such as Nb tend to segregate at open surface of voids. Results show two sizes of voids with 15 vacancies (Ø = 0.8 nm, up) and 65 vacancies (Ø = 1.3 nm, down)
- Segregation near interstitial loops: Smaller atoms, V, have tendency to segregate in these regions

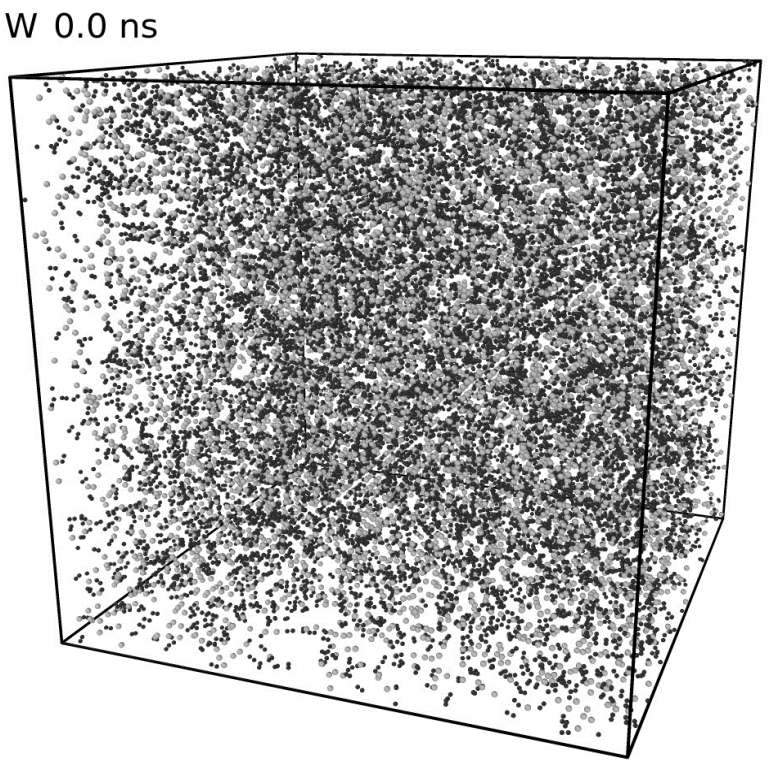
[J. Byggmästar, K. Nordlund, and F. Djurabekova PRB 104, 104101 (2021)]

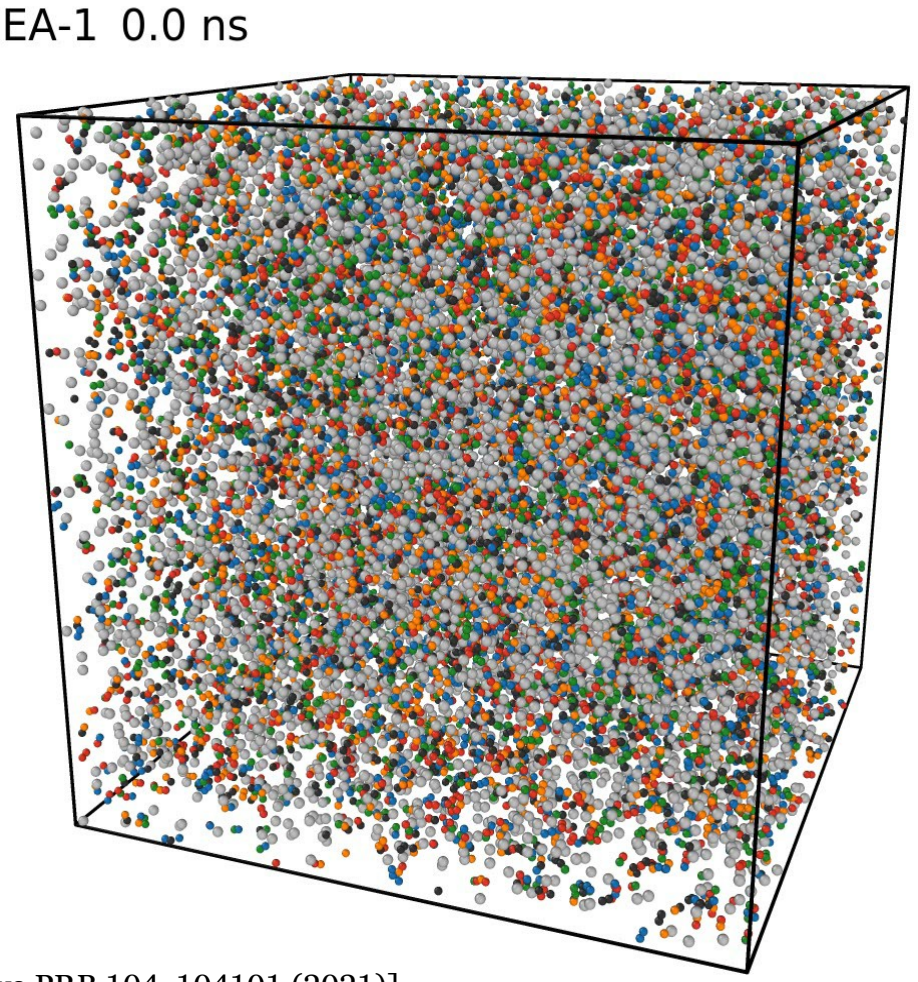
Segregation to grain boundaries





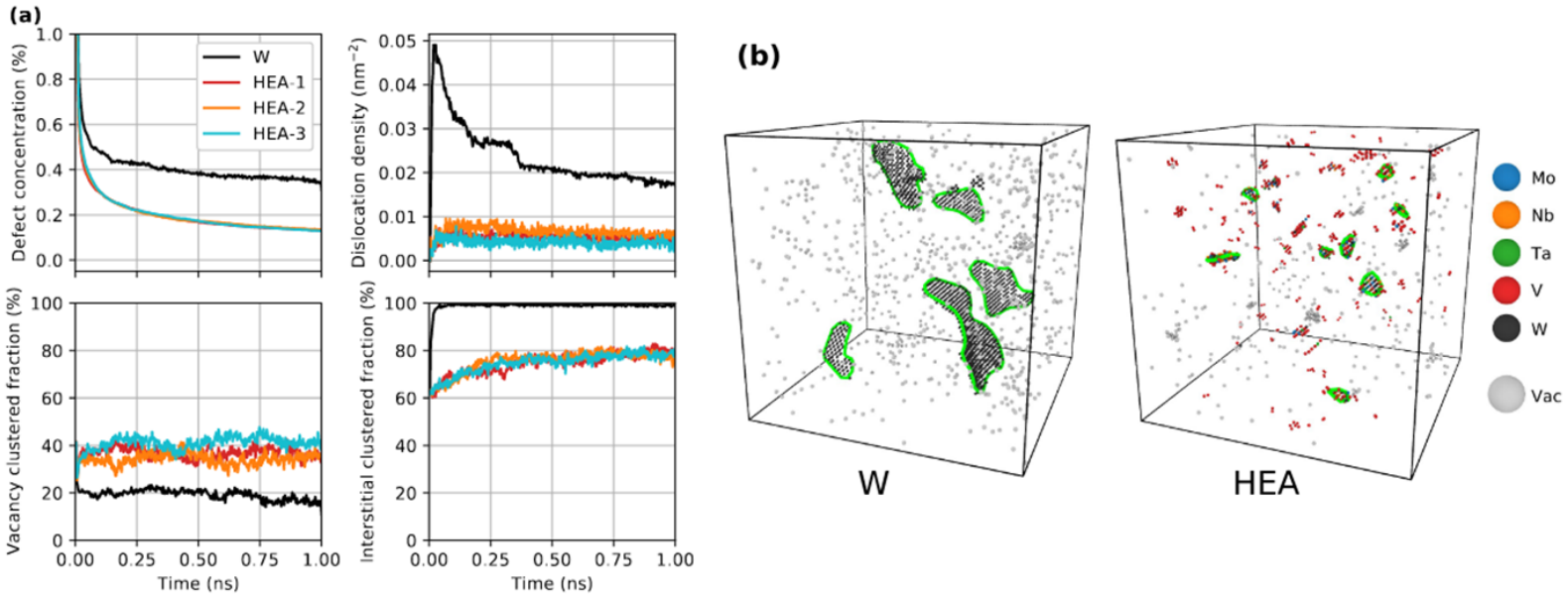
Defect annealing W vs. HEA





[J. Byggmästar, K. Nordlund, and F. Djurabekova PRB 104, 104101 (2021)]

High-dose damage in MoNbTaVW



- Defect evolution during annealing: (a) different defect concentrations as well as the number of interstitials in clusters; (b) snapshots of the final defects in W and MoNbTaVW HEA
- Experiments [1] see no dislocation loops in irradiated bcc HEAs:
  - ✓ They are too small.
- Defects recombine rather than they cluster:
  - ✓ Comparable mobility in 3D for both interstitials and vacancies
  - ✓ dislocation loop immobility

[1] El-Atwani, Osman, et al. "Outstanding radiation resistance of tungsten-based high-entropy alloys." *Science advances* 5.3 (2019):

Conclusions

- "Simple ML potentials for complex alloys"
- tabGAP: tabulated low-dimensional GAP
  - ✓ meV/atom accuracy for Mo-Nb-Ta-V-W alloys
  - ✓ 2 orders of magnitude faster than SOAP-GAP – speed similar to Tersoff/MEAM
- Strong effects of ordering and segregation in MoNbTaVW!
  - ✓ Atom size difference and favorable binary mixing
- Effect of lattice difference in pure-element materials affects the radiation resistance more than the difference in atomic size

Thank you for your attention!

- GAP potential files & training data: https://gitlab.com/acclab/gap-data
- tabGAP: https://gitlab.com/jezper/tabgap
- References/further details:
  - J. Byggmästar, K. Nordlund, and F. Djurabekova, Modeling refractory high-entropy alloys with efficient machine-learned interatomic potentials: defects and segregation, PRB 104, 104101 (2021)
  - J. Byggmästar, K. Nordlund, F. Djurabekova, Simple machine-learned interatomic potentials for complex alloys, https://arxiv.org/abs/2203.08458 (2022)