CHALLENGES IN SIMULATING INTERFACE STRUCTURES & PROPERTIES

Srikanth Patala

Department of Materials Science and Engineering

North Carolina State University

ACKNOWLEDGMENTS

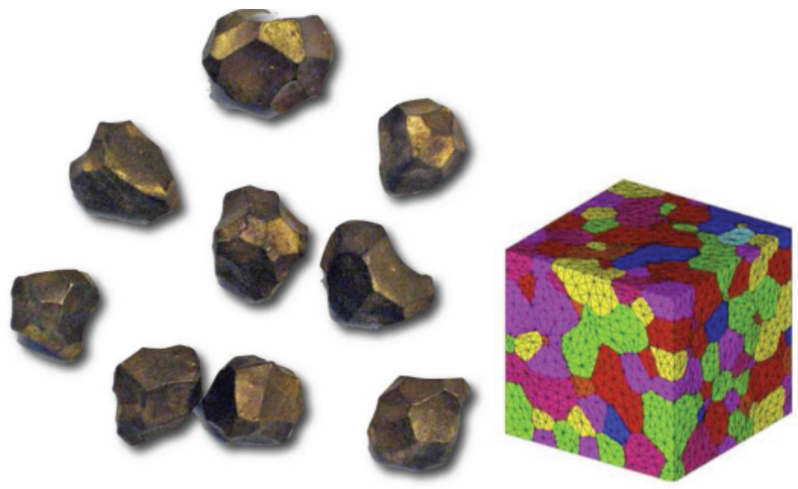
Aerospace Materials for Extreme Environments
Contract # FA9550-17-1-0145

Arash Banadaki
NCSU

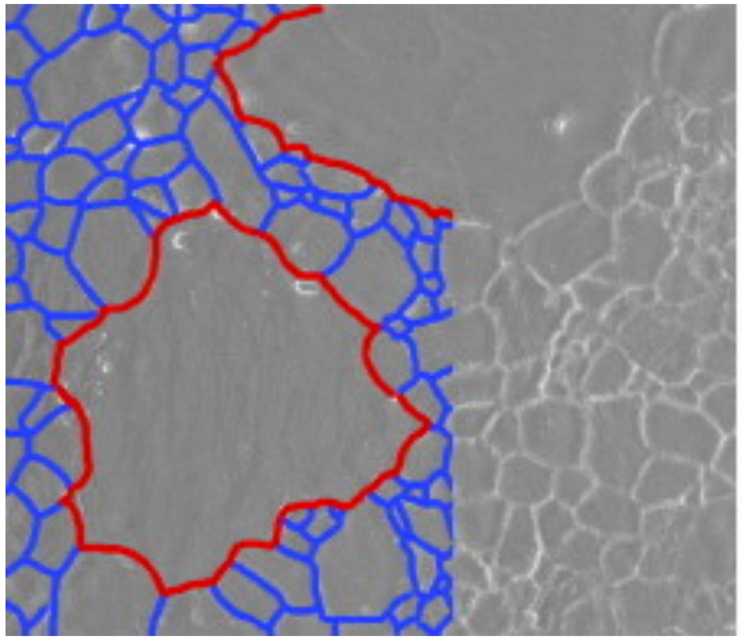
Mark A. Tschopp
ARL

GRAIN BOUNDARIES

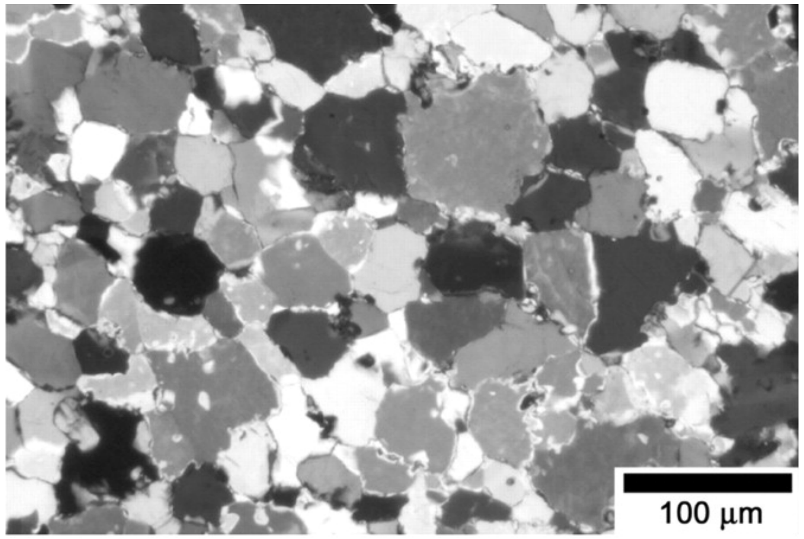
- Interfaces in polycrystalline materials (metals/ ceramics/ minerals)



Beta brass grains and visualization of a meshed polycrystal [1]



Microstructure of alumina (Al_2O_3) [2]



Micrograph of a natural dunite rock specimen from New Zealand [3]

1. Sharp, R. (2010). Mathematics and Polycrystalline Materials: An Entropic Approach to Texture Development. SIAM News, 43(8).
2. Dillon, S. J., Tang, M., Carter, W. C., & Harmer, M. P. (2007). Complexion: A new concept for kinetic engineering in materials science. Acta Materialia, 55(18), 6208-6218.
3. Aizawa, Y., Barnhoorn, A., Faul, U. H., Gerald, J. D. F., Jackson, I., & Kovács, I. (2008). Seismic properties of Anita Bay dunite: an exploratory study of the influence of water. Journal of Petrology, 49(4), 841-855.

GRAIN BOUNDARIES

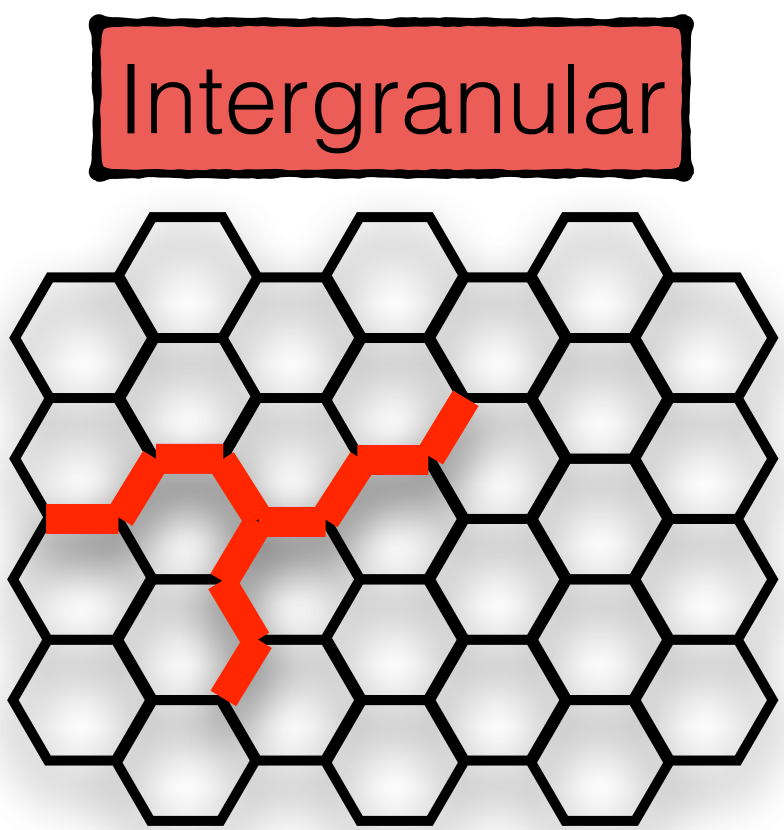
- Influence on properties:

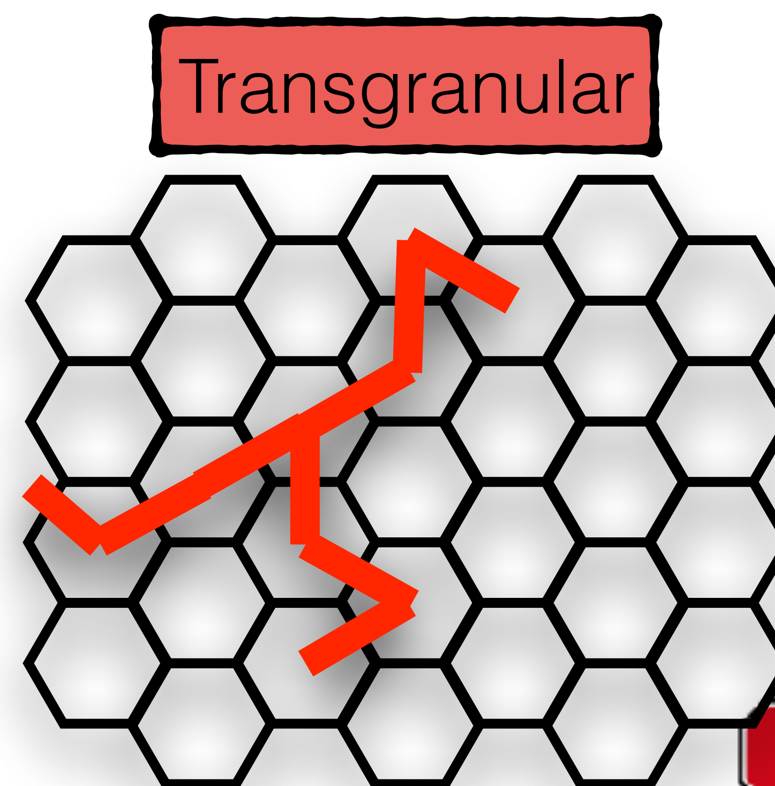
Structural
Cavitation
Intergranular Cracking
Radiation Resistance

Chemical
Corrosion
Stress-corrosion cracking

Cleavage cracking

Electrical/Electronic
Magneto-transport
Superconductivity





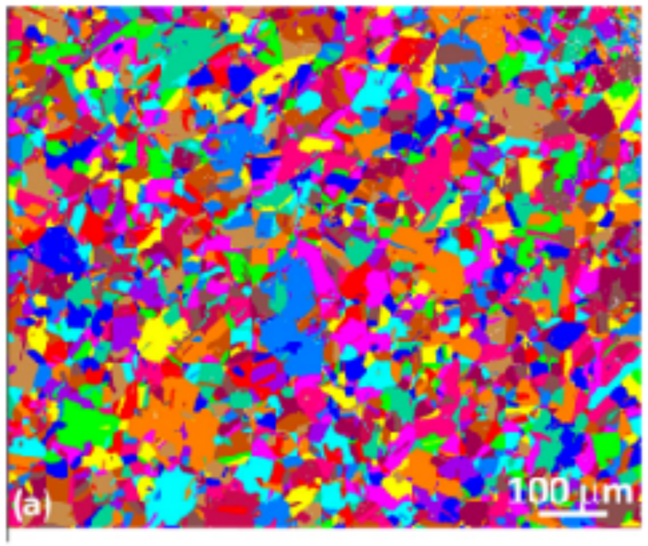
GRAIN BOUNDARY ENGINEERING

- GBE: Manipulate connectivity and distribution of interfaces

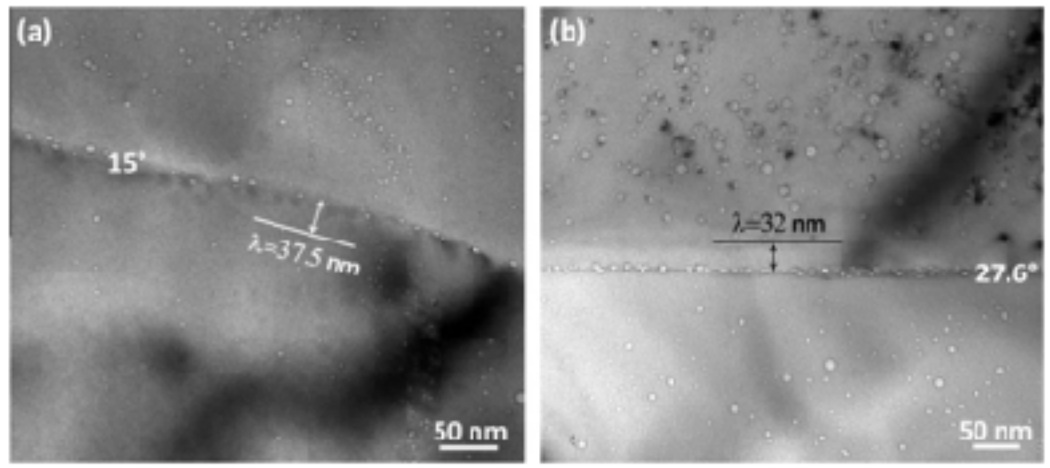
Property	Materials	GBE effect on property [1]
Stress-Corrosion Cracking	Ni-based, Stainless Steel	>2X
Fatigue	Various Ni-based	2-3X
Intergranular Corrosion	Lead, Nickel, Copper alloys	>2X
Ductility (Room Temp)	Brass, Stainless	>2X
Creep Rate	Ni-based, various	>20X
Hot Ductility (Weldability)	Ni-based Inconel	~40X
Superconducting Current	Tetragonal Oxides	>10X
Electromigration Resistance	Aluminum	>10X

[1] Palumbo, G., Lehockey, E. M., & Lin, P. (1998). Applications for grain boundary engineered materials. JOM, 50(2), 40-43.

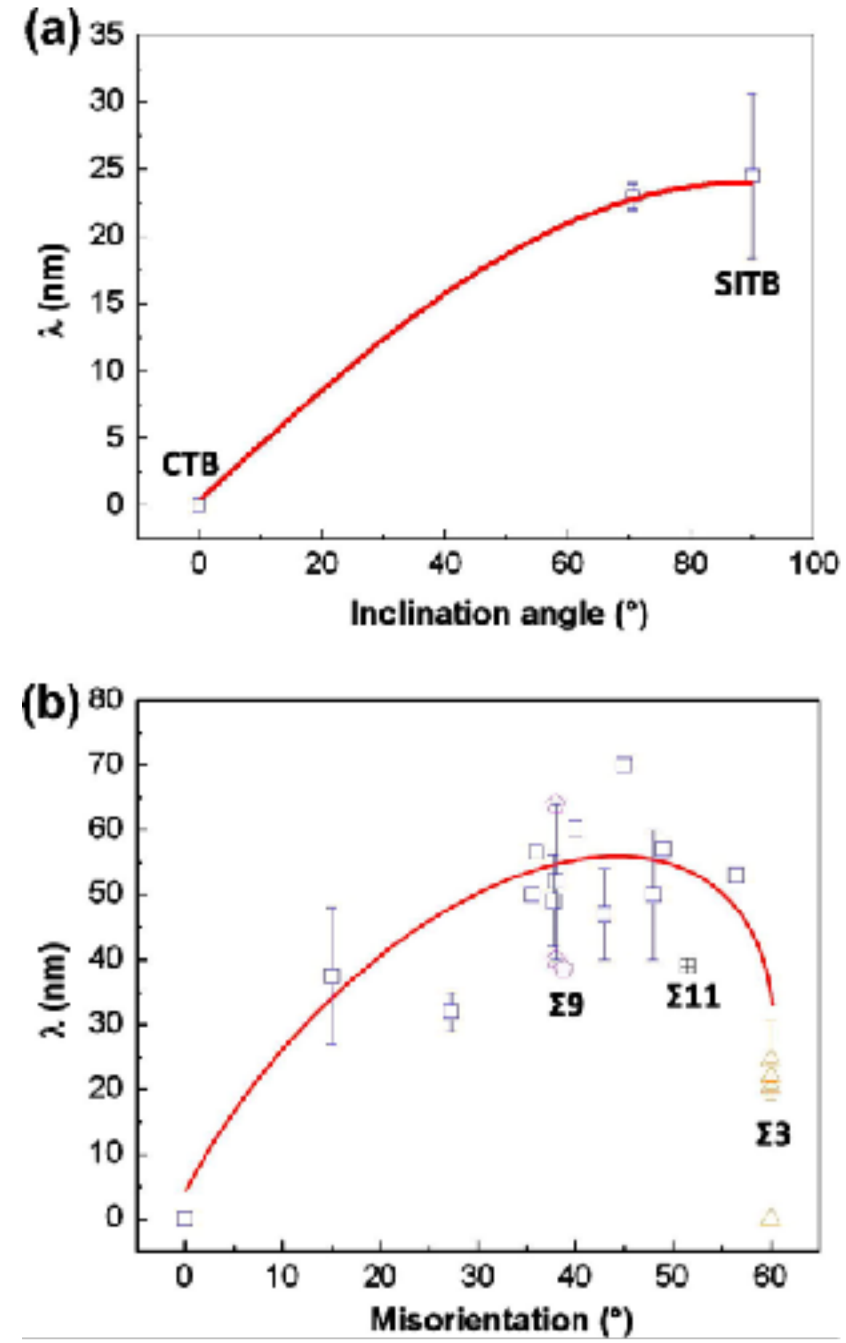
ANISOTROPY OF GRAIN BOUNDARY PROPERTIES

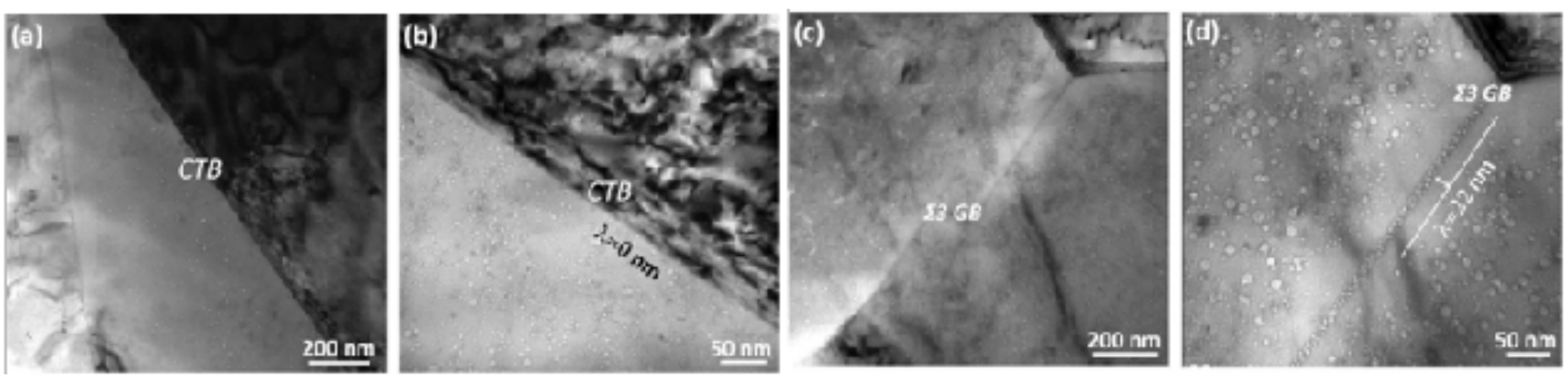


EBSD image of high-purity Cu after annealing at 500 °C for 10 min (unique grain color code) [1].



Radiation-induced damage features around GBs with misorientation angle of (a) 15° and (b) 27.6° at 450 °C by 200 keV He ions with fluence of 2 × 1017 ions cm^{-2} [1].



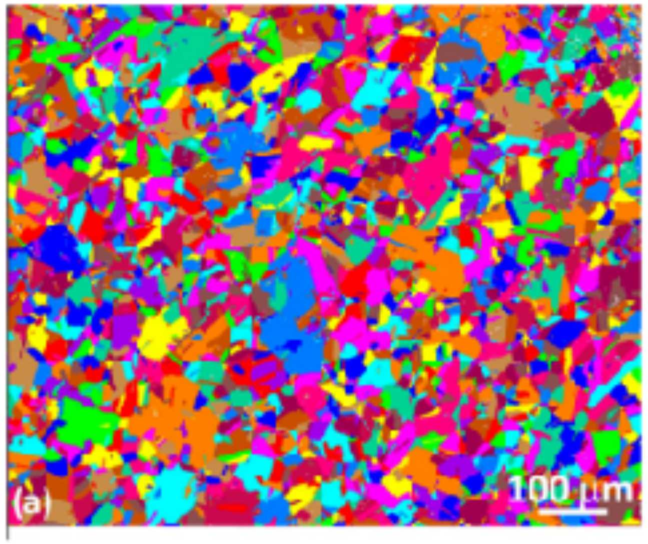


Σ3 [110] tilt GBs irradiated at 450°C by 200 keV He ions with a fluence of 2 × 10^{17} ions cm^{-2}: (a) and (b) show radiation-induced voids but no VDZ near a CTB; (c) and (d) show a VDZ near an asymmetric Σ3 [110] tilt GB [1].

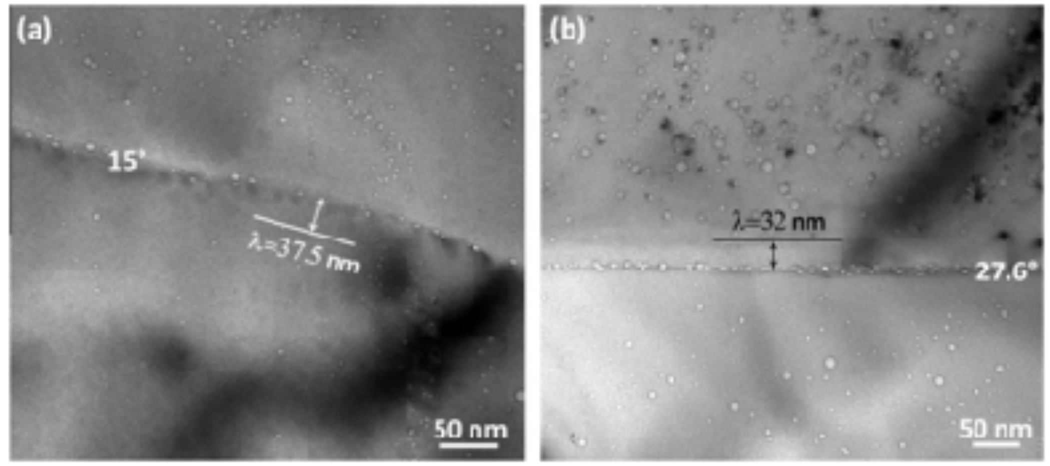
(a) Plot of VDZ width as a function of inclination angle for Σ3 GBs (solid line is the model prediction); (b) VDZ width as a function of misoriention for non-Σ3 and Σ3 GBs. The solid line in (b) is added for legibility [1].

[1] Han, W. Z., Demkowicz, M. J., Fu, E. G., Wang, Y. Q., & Misra, A. (2012). Effect of grain boundary character on sink efficiency. Acta materialia, 60(18), 6341-6351.

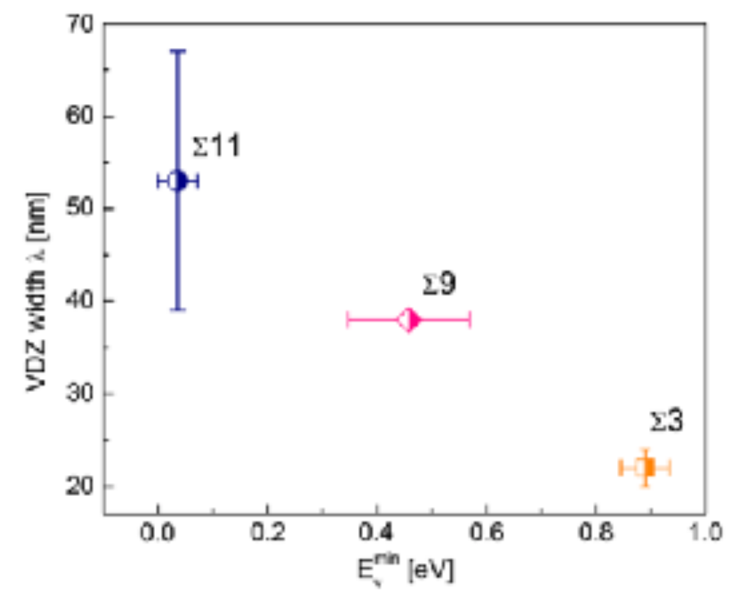
ANISOTROPY OF GRAIN BOUNDARY PROPERTIES



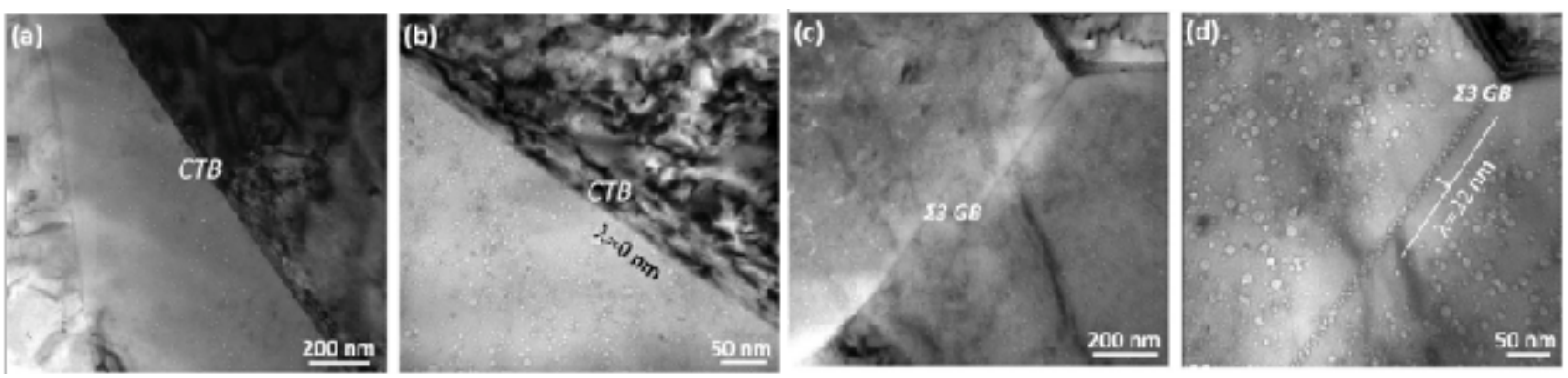
EBSD image of high-purity Cu after annealing at 500 °C for 10 min (unique grain color code) [1].



Radiation-induced damage features around GBs with misorientation angle of (a) 15° and (b) 27.6° at 450 °C by 200 keV He ions with fluence of 2 × 1017 ions cm^{-2} [1].



Void denuded zone widths from [1] plotted against minimum vacancy formation energies in the <u>lowest energy states for GBs</u> [2].



Σ3 [110] tilt GBs irradiated at 450°C by 200 keV He ions with a fluence of 2 × 10^{17} ions cm^{-2}: (a) and (b) show radiation-induced voids but no VDZ near a CTB; (c) and (d) show a VDZ near an asymmetric Σ3 [110] tilt GB [1].

[1] Han, W. Z., Demkowicz, M. J., Fu, E. G., Wang, Y. Q., & Misra, A. (2012). Effect of grain boundary character on sink efficiency. Acta materialia, 60(18), 6341-6351.
[2] Yu, W. S., & Demkowicz, M. J. (2015). Non-coherent Cu grain boundaries driven by continuous vacancy loading. Journal of Materials Science, 50(11), 4047-4065.

VISION

High-Throughput Grain Boundary Property Measurements

Atomistic/DFT Simulations

- GB Energy [1]
- Temperature-Dependent Mobility [3]
- GB Yield, Shear Strength

Experiment GB Property Measurements

- GB Energy [2]
- Radiation Resistance (VDW)
- Susceptibility to Hydrogen Embrittlement

Statistical Analysis Framework

GB Knowledge Systems

- Crystallography and Metrics
- Atomic Descriptor
- Two-Point Correlation Function
- Calibrated Localization Linkages

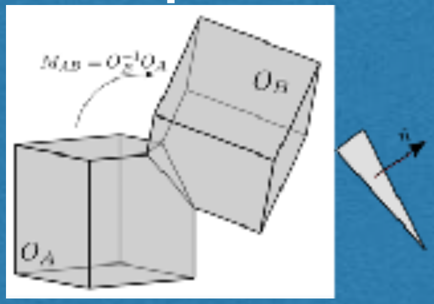
Basis Functions on the Five-Parameter Phase-Space

Crystallography:

$\mathrm{GB} = (M;\, \hat{n})$

Topology:

$\left[S^3 \times S^3\right]/S^1$

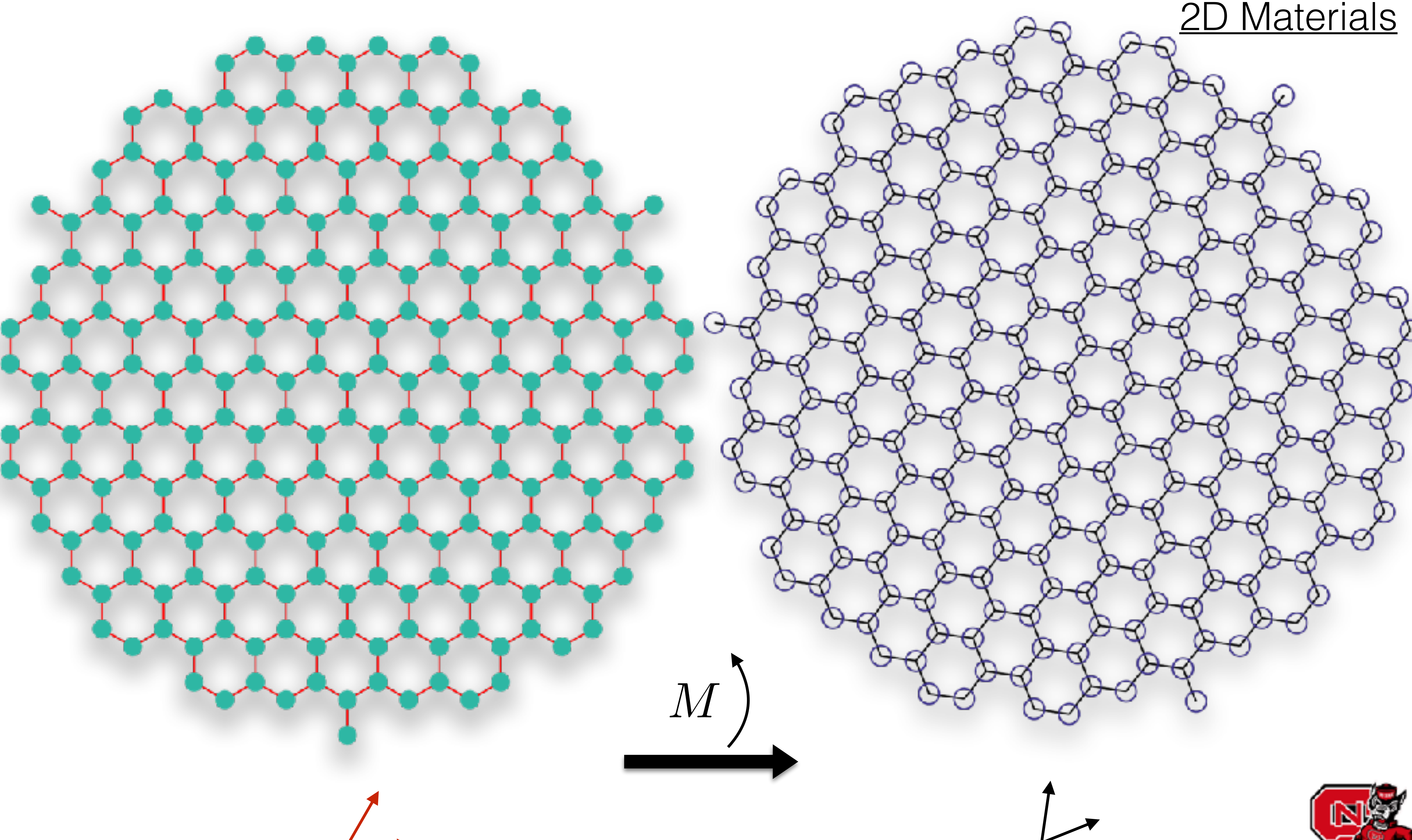


GB Structure-Property Relationships → **GB Crystallography-Structure Relationships** → **GB Crystallography-Property Relationships**

[1] AD Banadaki, MA Tschopp, S. Patala. A Monte-Carlo Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)
[2] Morawiec, A. (2000). Method to calculate the grain boundary energy distribution over the space of macroscopic boundary parameters from the geometry of triple junctions. *Acta materialia*, *48*(13), 3525-3532.
[3] E. R. Homer, E. A. Holm, S. M. Foiles, and D. L. Olmsted. Trends in grain boundary mobility: Survey of motion mechanisms. *JOM*, 66, 114–120, 2014.

CRYSTALLOGRAPHY OF GBs

2D Materials



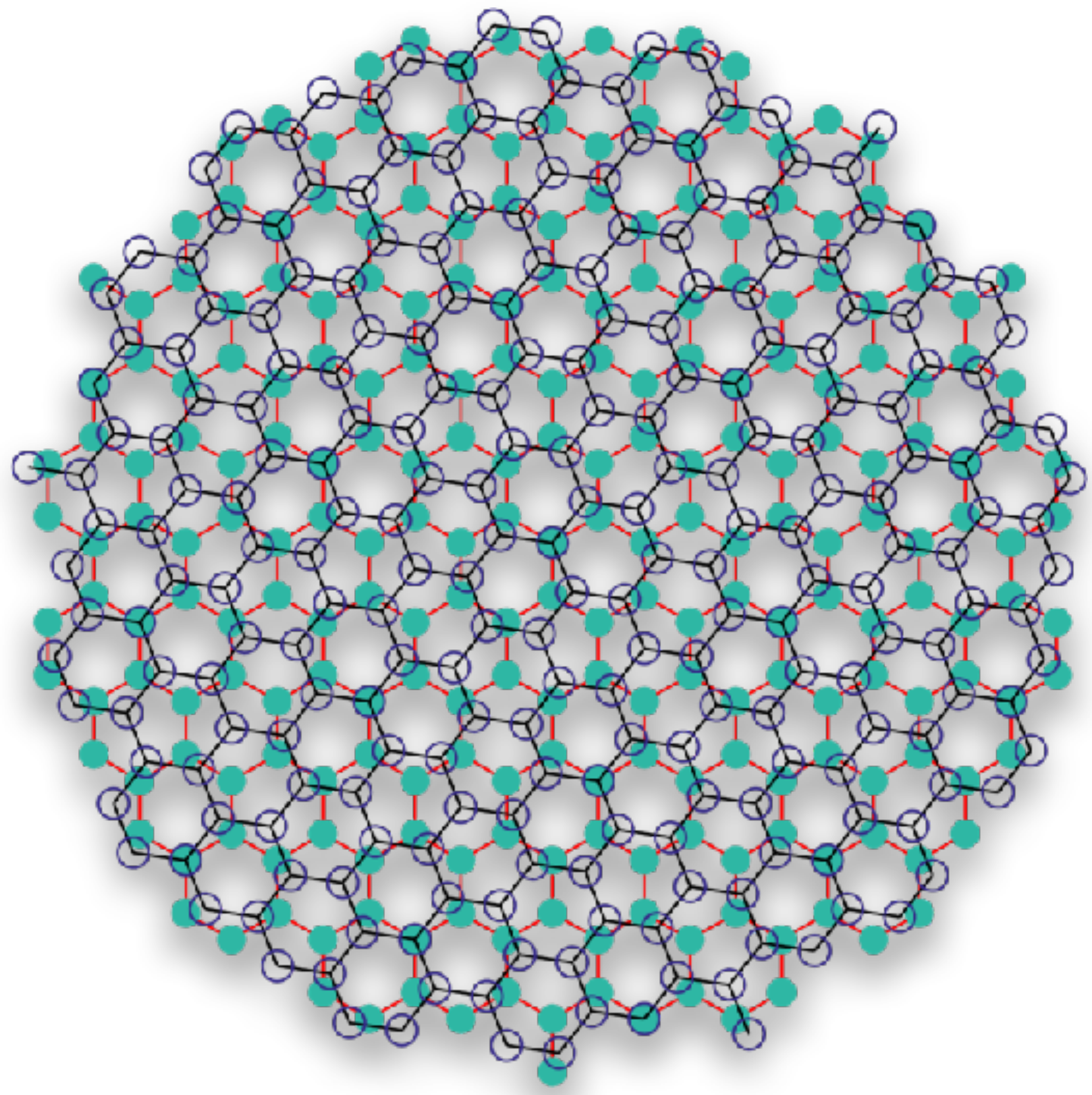
CRYSTALLOGRAPHY OF GBs IN 2D MATERIALS

3D Materials

Misorientation

$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters

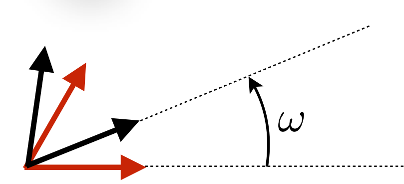


2D Materials

Misorientation

$$M = (\omega, \hat{z}) = \omega$$

One-Parameter



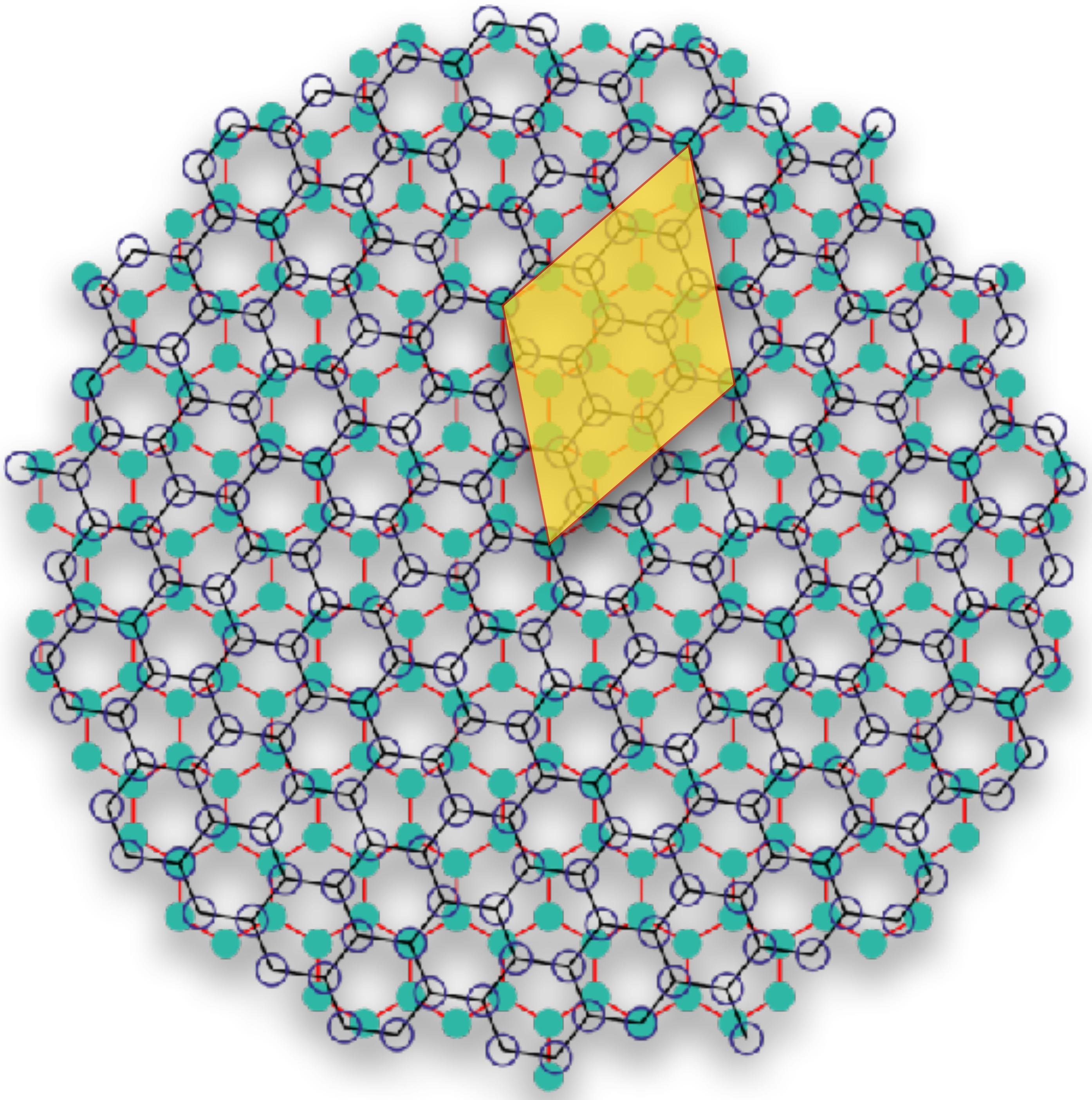
CRYSTALLOGRAPHY OF GBs IN 2D MATERIALS

<u>3D Materials</u>

Misorientation

$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters



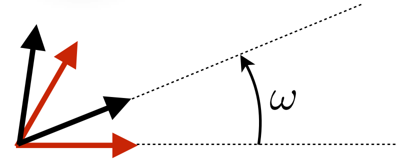
<u>2D Materials</u>

Misorientation

$$M = (\omega, \hat{z}) = \omega$$

Σn

One-Parameter



CRYSTALLOGRAPHY OF GBs IN 2D MATERIALS

3D Materials

Misorientation

$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters

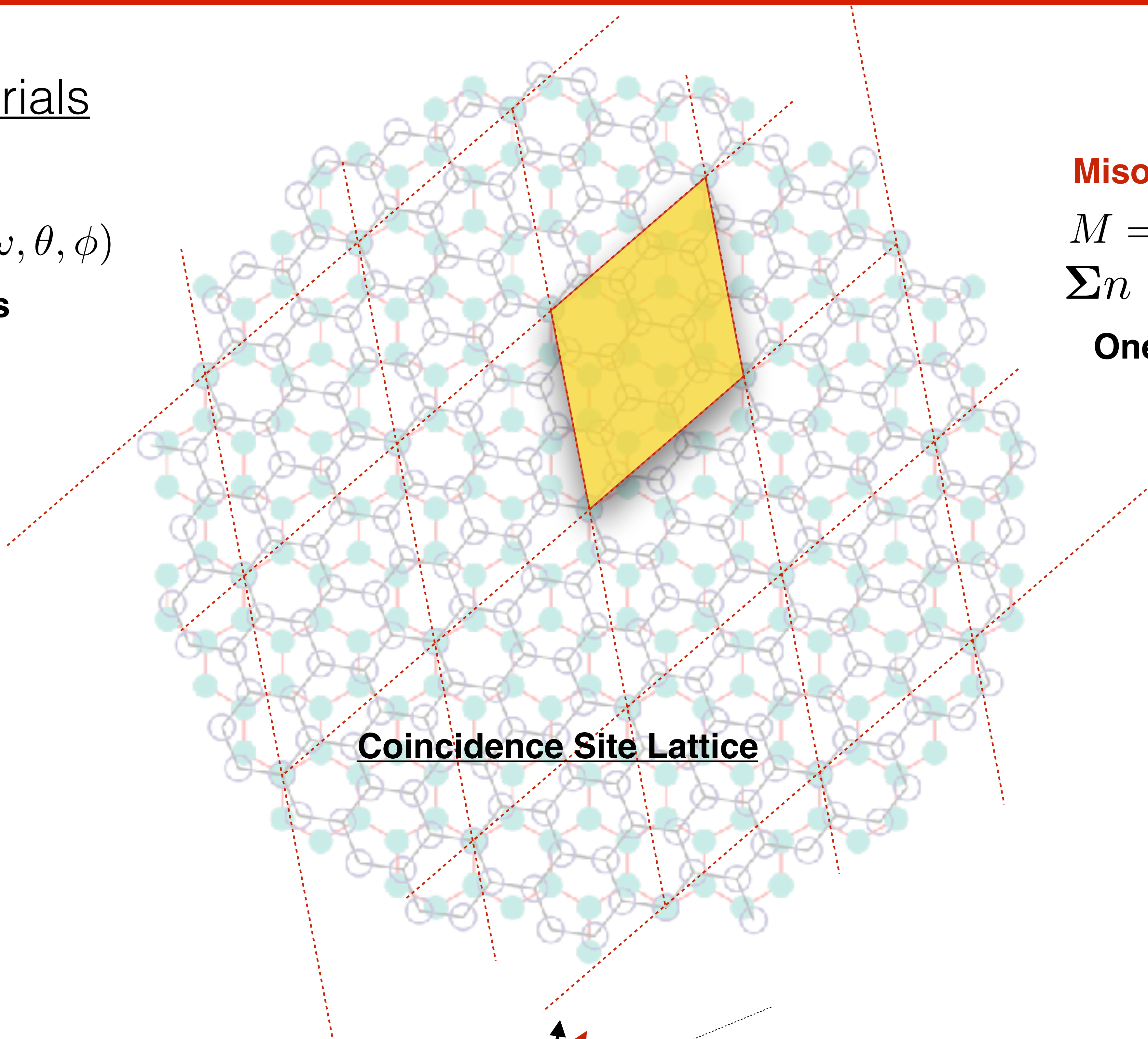
2D Materials

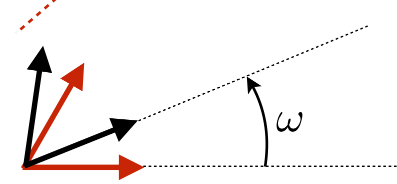
Misorientation

$$M = (\omega, \hat{z}) = \omega$$

$$\Sigma n$$

One-Parameter





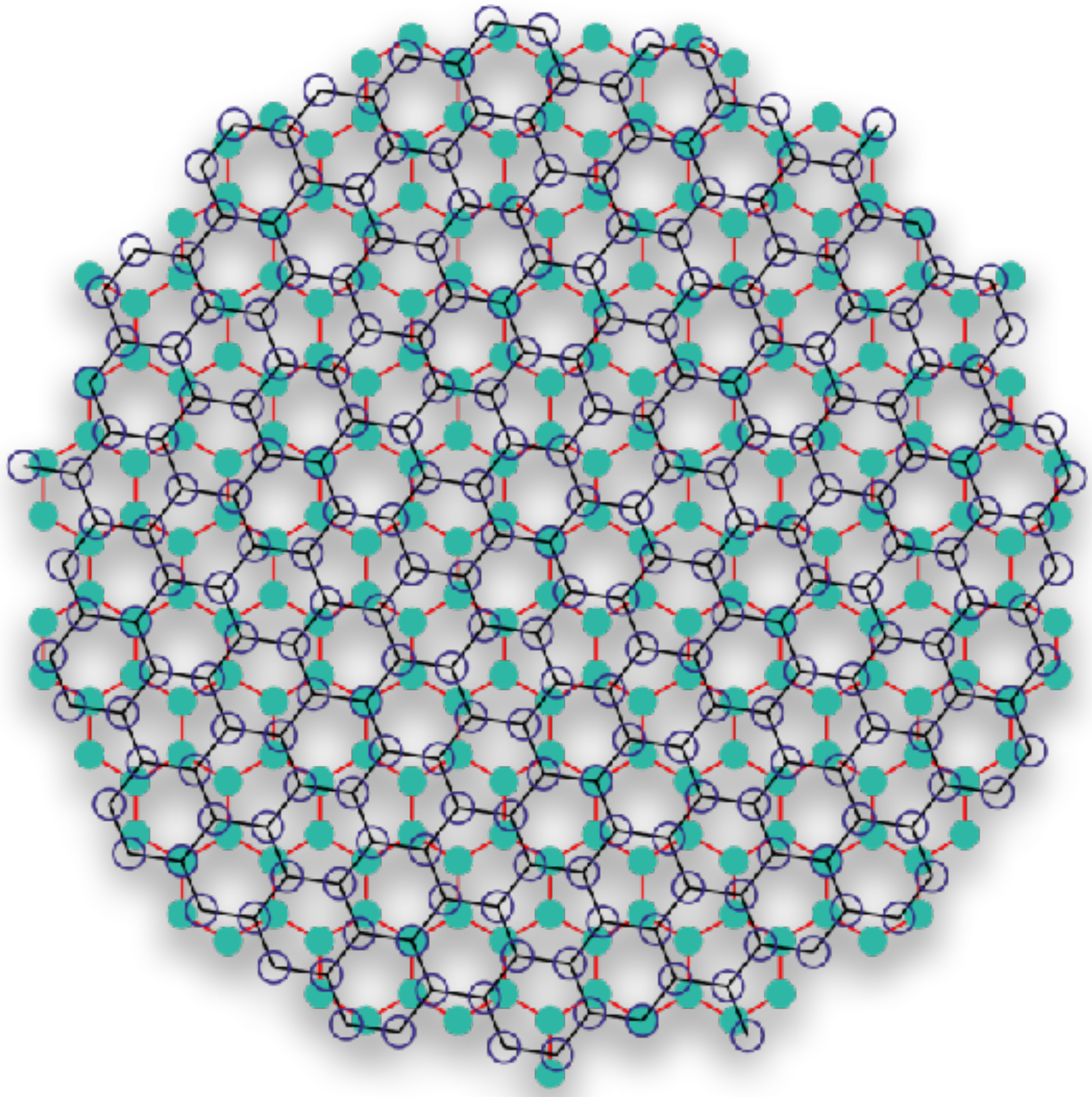
CRYSTALLOGRAPHY OF GBs IN 2D MATERIALS

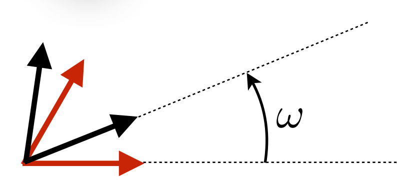
3D Materials

Misorientation

$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters





2D Materials

Misorientation

$$M = (\omega, \hat{z}) = \omega$$

$$\Sigma n$$

One-Parameter

CRYSTALLOGRAPHY OF GBs IN 2D MATERIALS

3D Materials

Misorientation

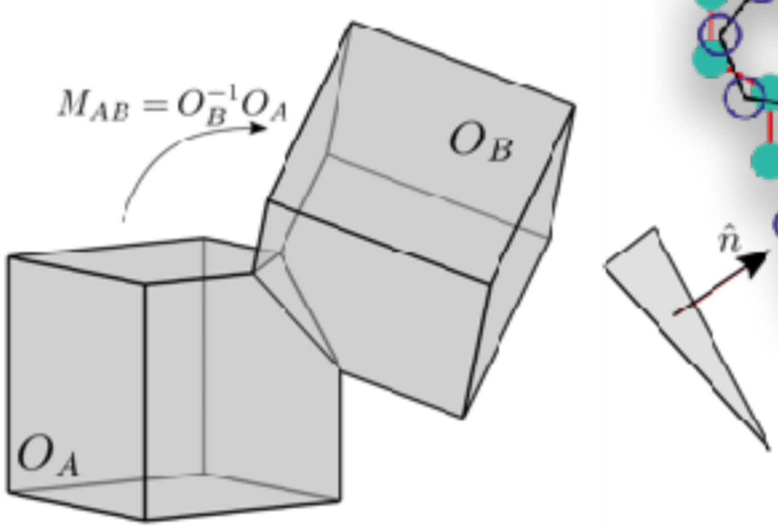
$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

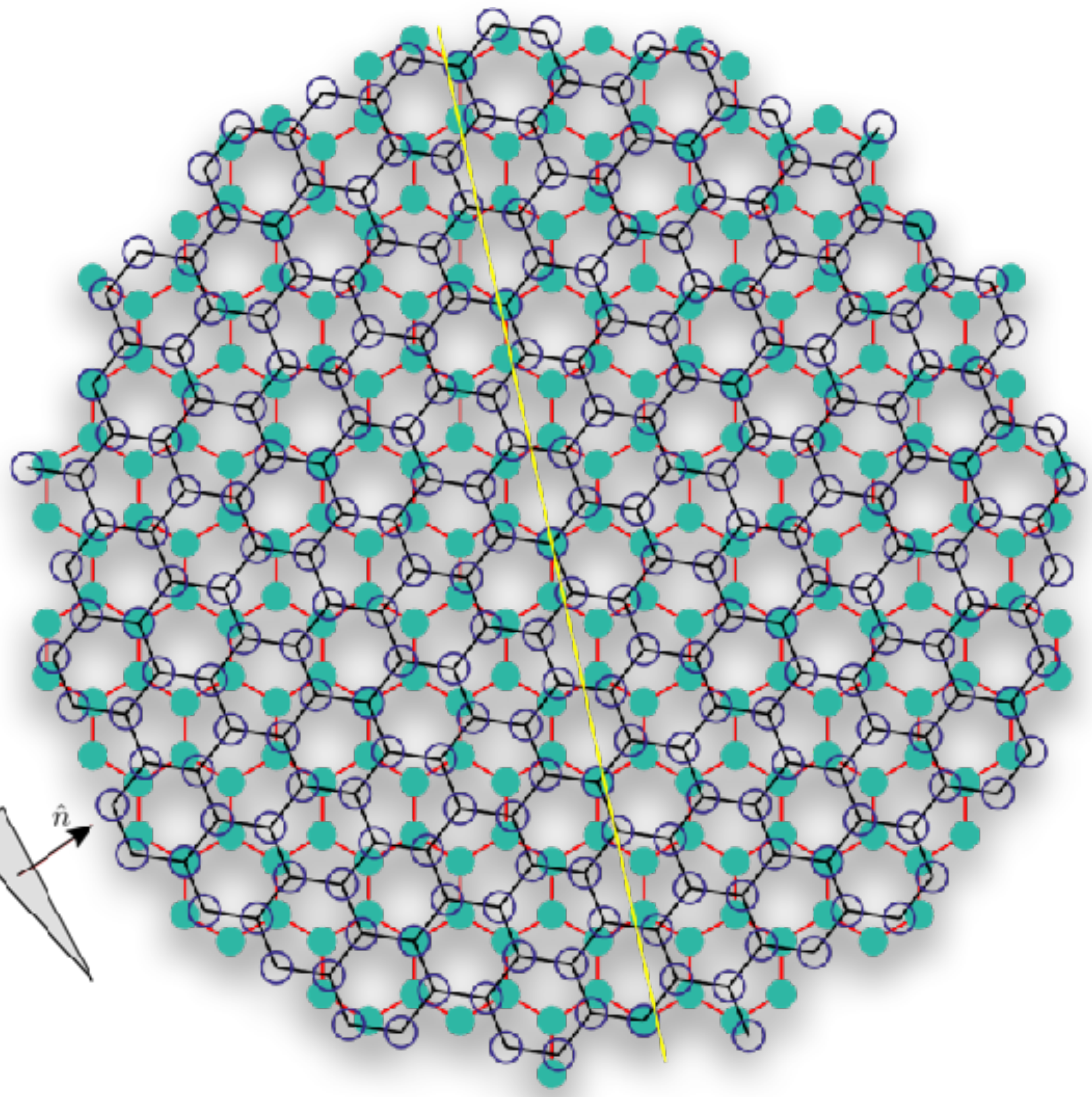
Three-Parameters

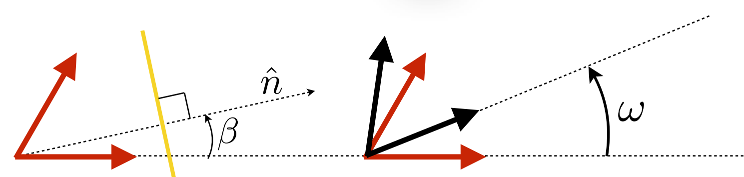
Boundary-Plane Orientation:

$$\hat{n} = (\alpha, \beta)$$

Two-Parameters







2D Materials

Misorientation

$$M = (\omega, \hat{z}) = \omega$$

$$\Sigma n$$

One-Parameter

Boundary-Plane Orientation:

$$\hat{n} = \beta$$

One-Parameter

CRYSTALLOGRAPHY OF GBs IN 2D MATERIALS

3D Materials

Misorientation

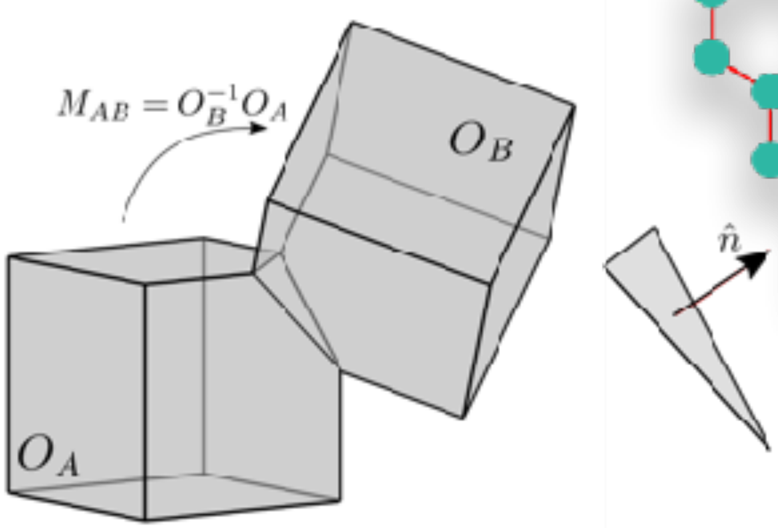
$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters

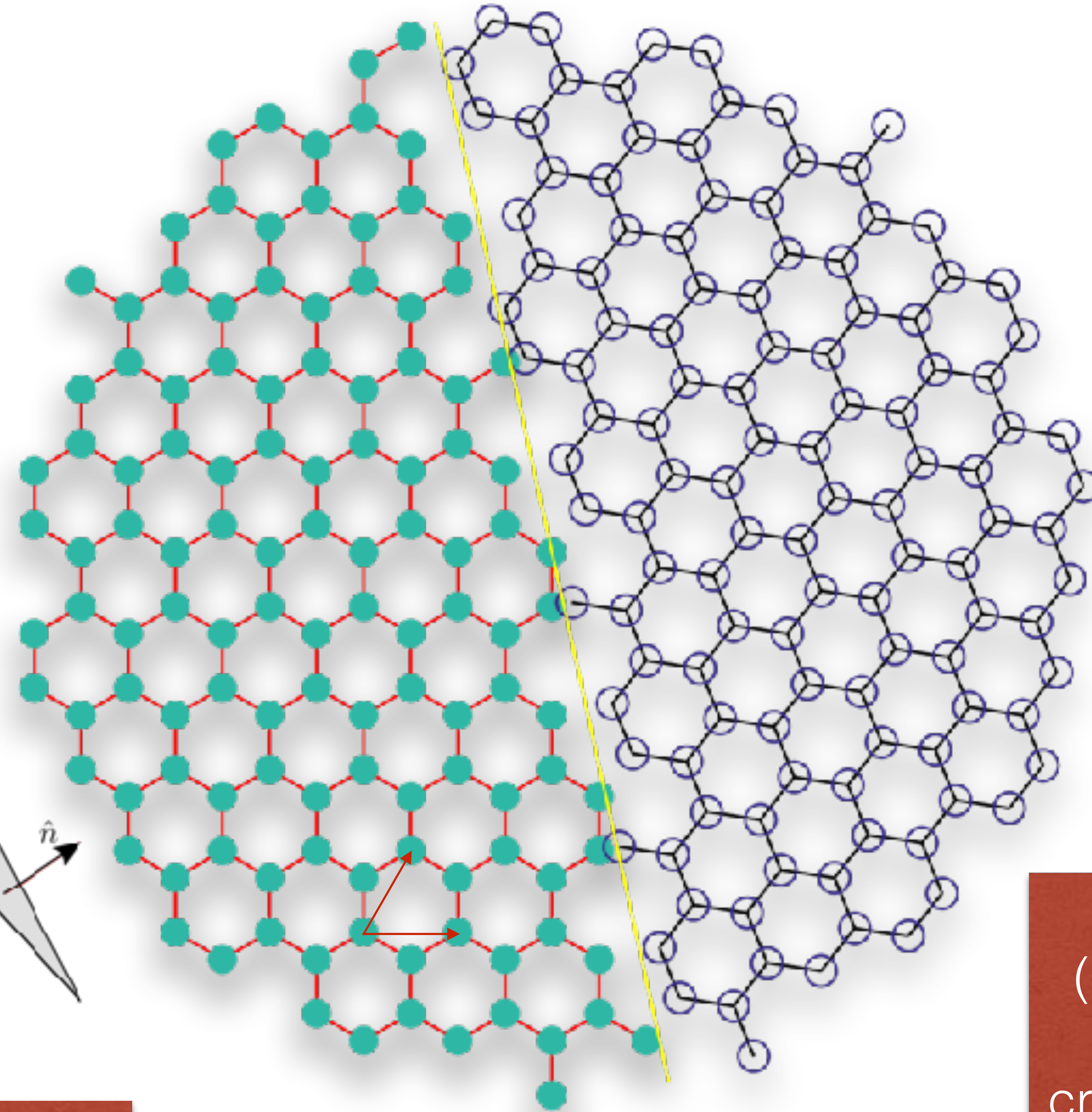
Boundary-Plane Orientation:

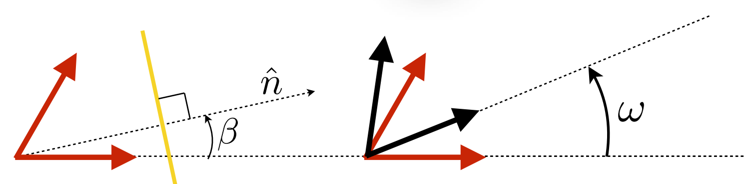
$$\hat{n} = (\alpha, \beta)$$

Two-Parameters



Five parameters for uniquely specifying the crystallography of GBs in 3D bulk materials.





2D Materials

Misorientation

$$M = (\omega, \hat{z}) = \omega$$

$$\Sigma n$$

One-Parameter

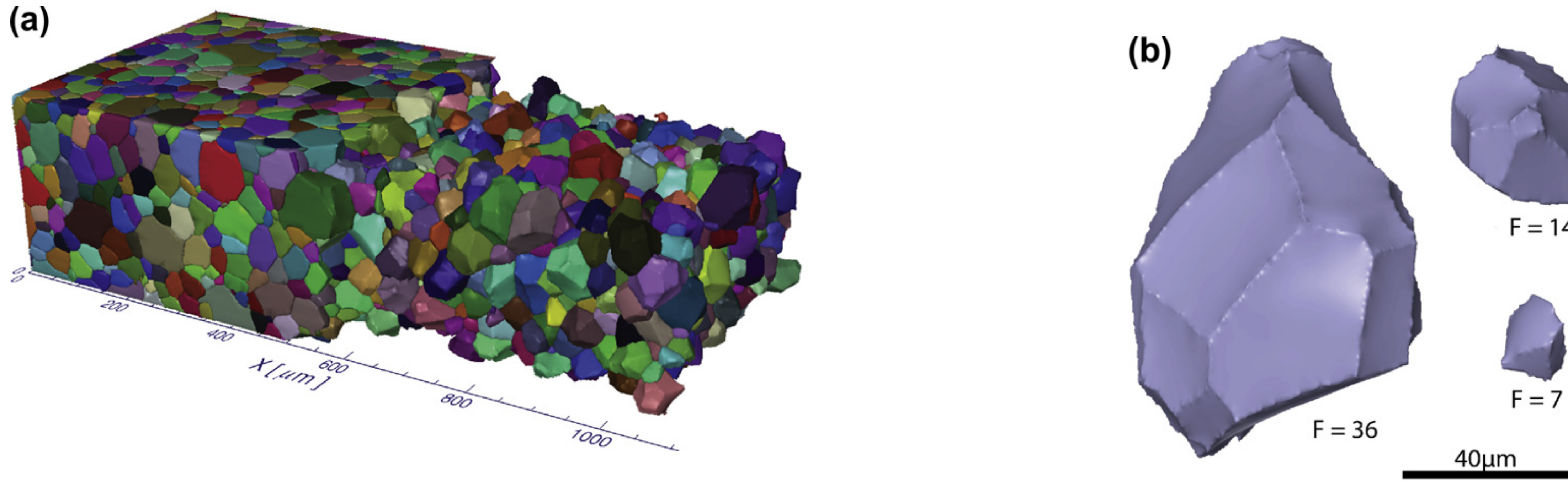
Boundary-Plane Orientation:

$$\hat{n} = \beta$$

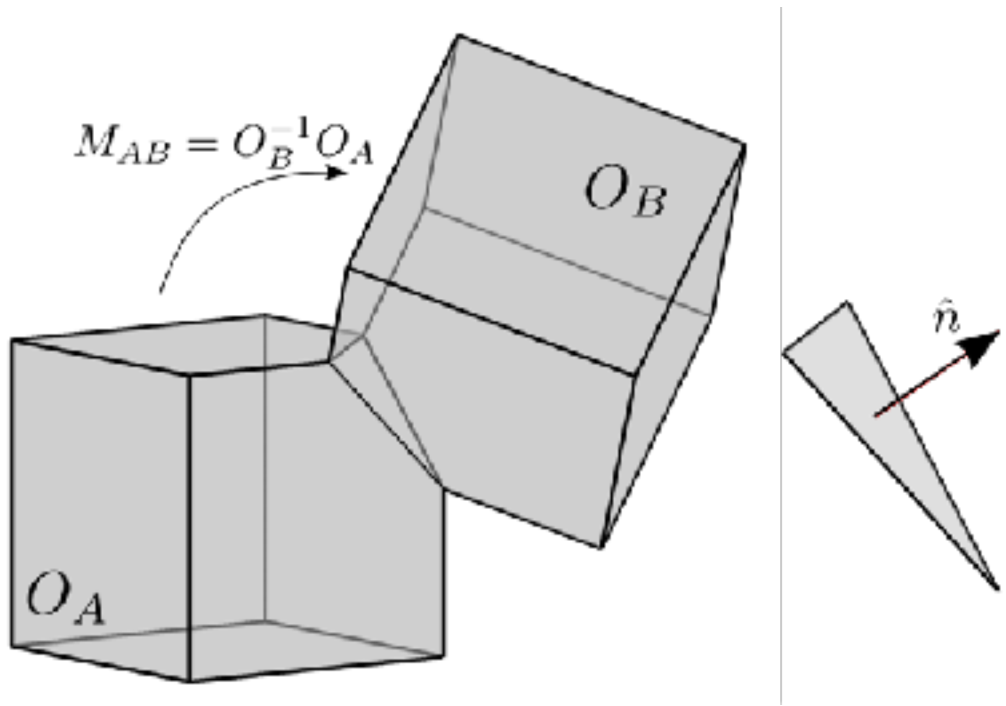
One-Parameter

Two parameters (angles) for uniquely specifying the crystallography of GBs in 2D materials.

3D MICROSTRUCTURES



- Spatial mapping of local crystal orientations enables direct measurement of grain boundary parameters [1].
- Three dimensional Orientation Imaging (3D OIM™ [2])



Misorientation

$$M = (\omega, \hat{a})$$

$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters

Boundary-Plane Orientation:

$$\hat{n} = (\alpha, \beta)$$

Two-Parameters

[1] **D. J. Rowenhorst, A. C. Lewis, G. Spanos,** Acta Materialia, Vol 58, (2010) 5511-5519.
[2] **S. Zaefferer, S.I. Wright, D. Raabe,** Metallurgical and Materials Transactions A, No. 2, (2008) 374-389

STATISTICAL FRAMEWORK

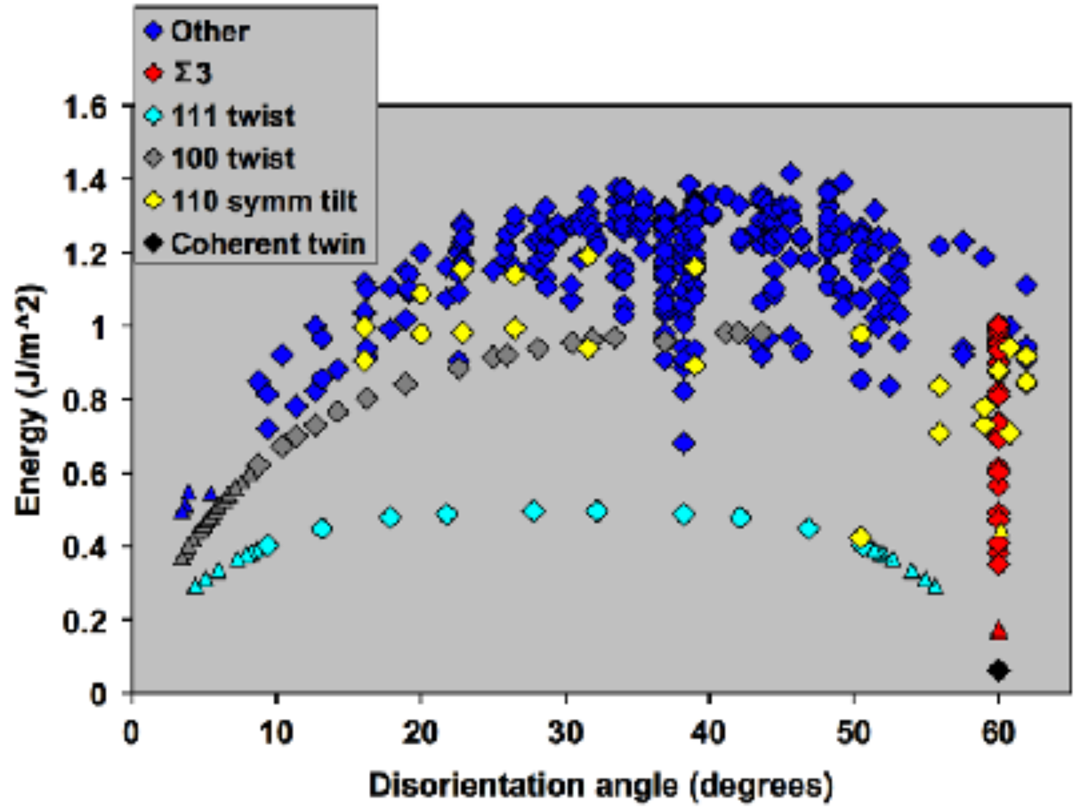
High-Throughput Grain Boundary Property Measurements

Atomistic/DFT Simulations

- GB Energy [1]
- Temperature-Dependent Mobility
- GB Yield, Shear Strength

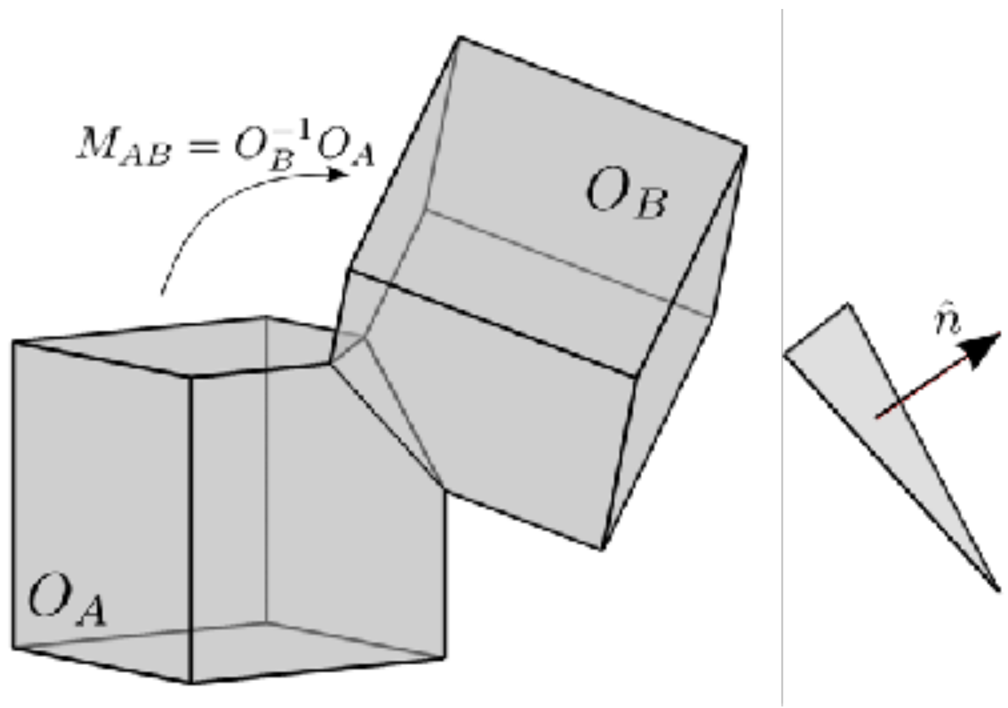
Experiment GB Property Measurements

- GB Energy[2]
- Corrosion Resistance
- Susceptibility to Hydrogen Embrittlement



[1] AD Banadaki, MA Tschopp, S. Patala. A Monte-Carlo Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)
[2] Morawiec, A. (2000). Method to calculate the grain boundary energy distribution over the space of macroscopic boundary parameters from the geometry of triple junctions. *Acta materialia*, *48*(13), 3525-3532.

CRYSTALLOGRAPHY OF GBs



Macroscopic DOF

Misorientation

$$M = (\omega, \hat{a})$$

$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters

Boundary-Plane Orientation:

$$\hat{n} = (\alpha, \beta)$$

Two-Parameters

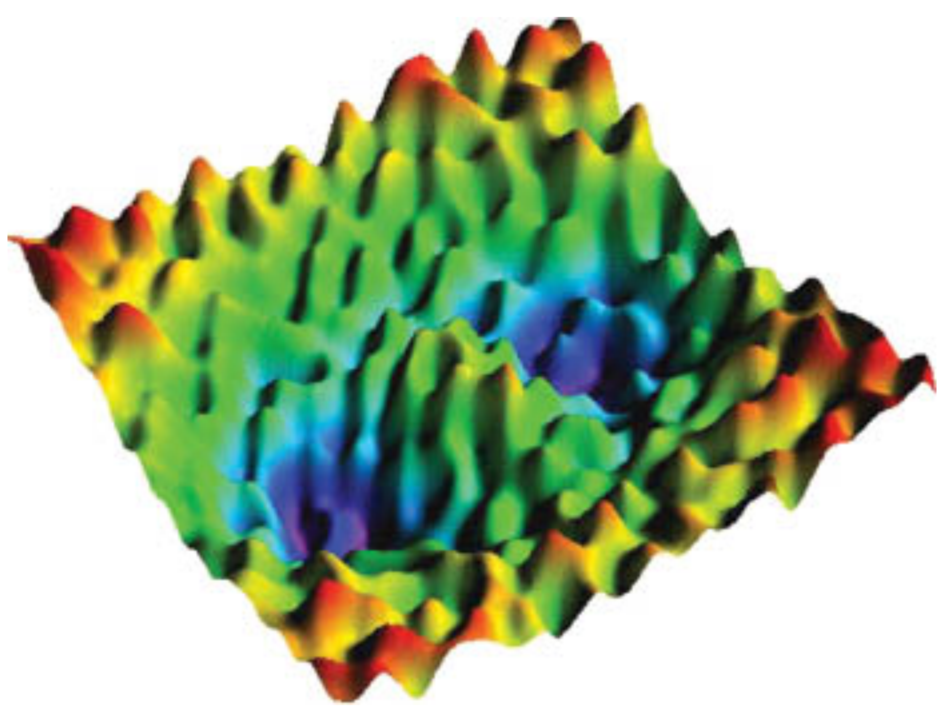
Microscopic DOF

Relative Lattice Translation: $\vec{d}$

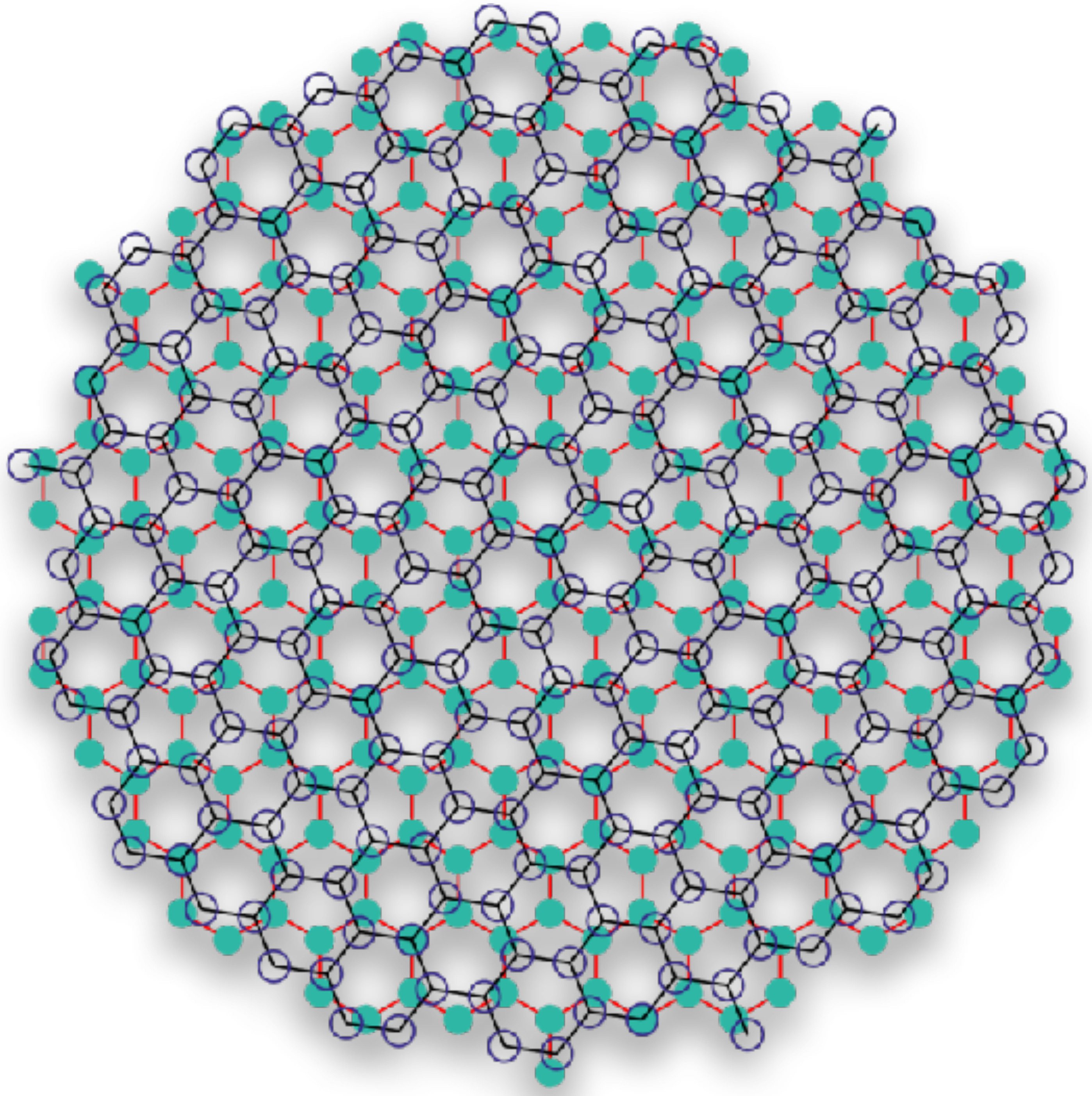
Boundary-Plane Translation: t

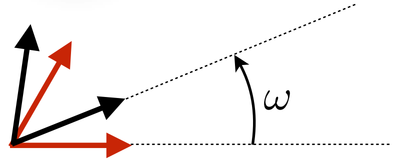
Grain Boundary Density: λ

μ-energy-landscape



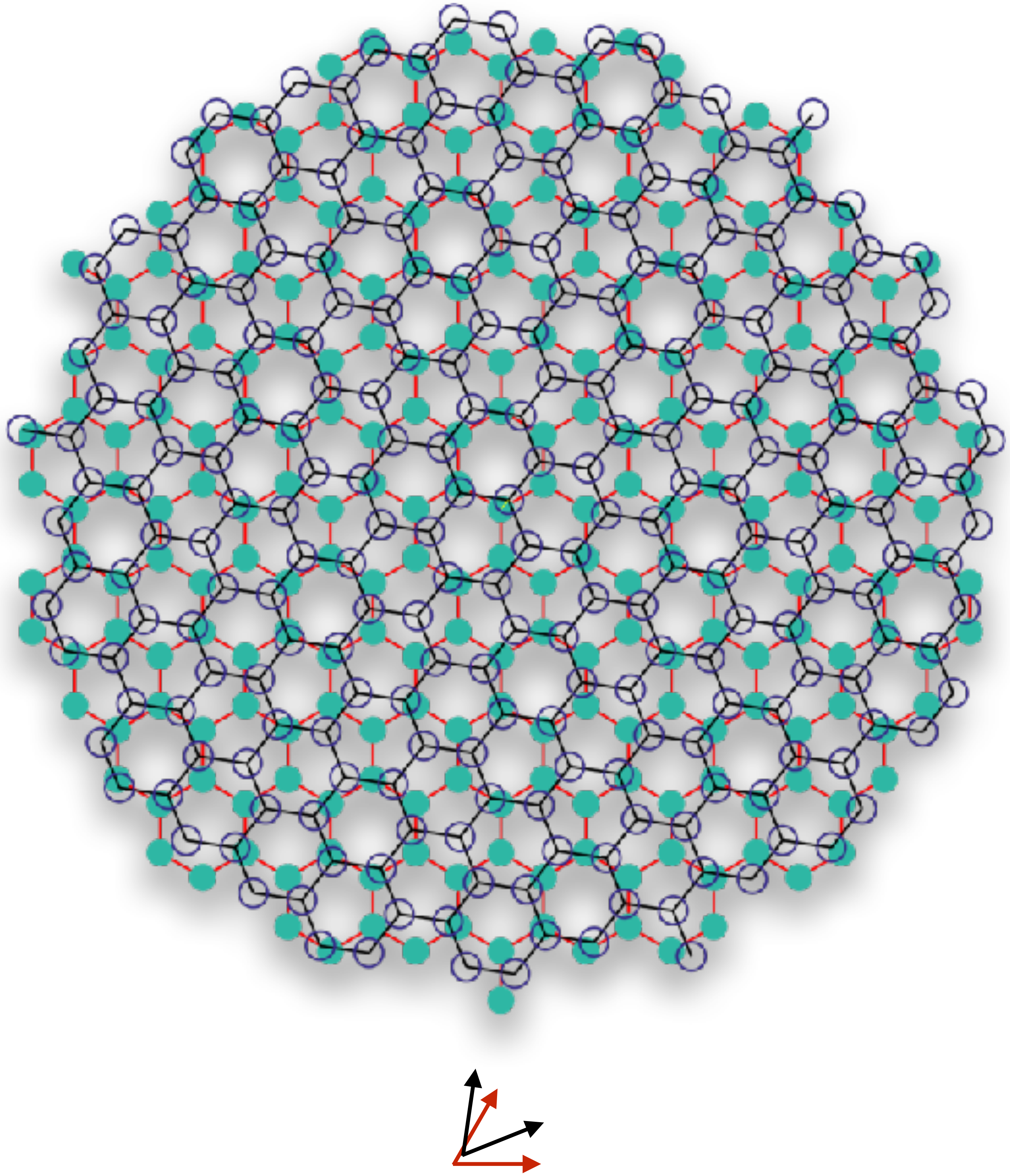
MICROSCOPIC DEGREES OF FREEDOM





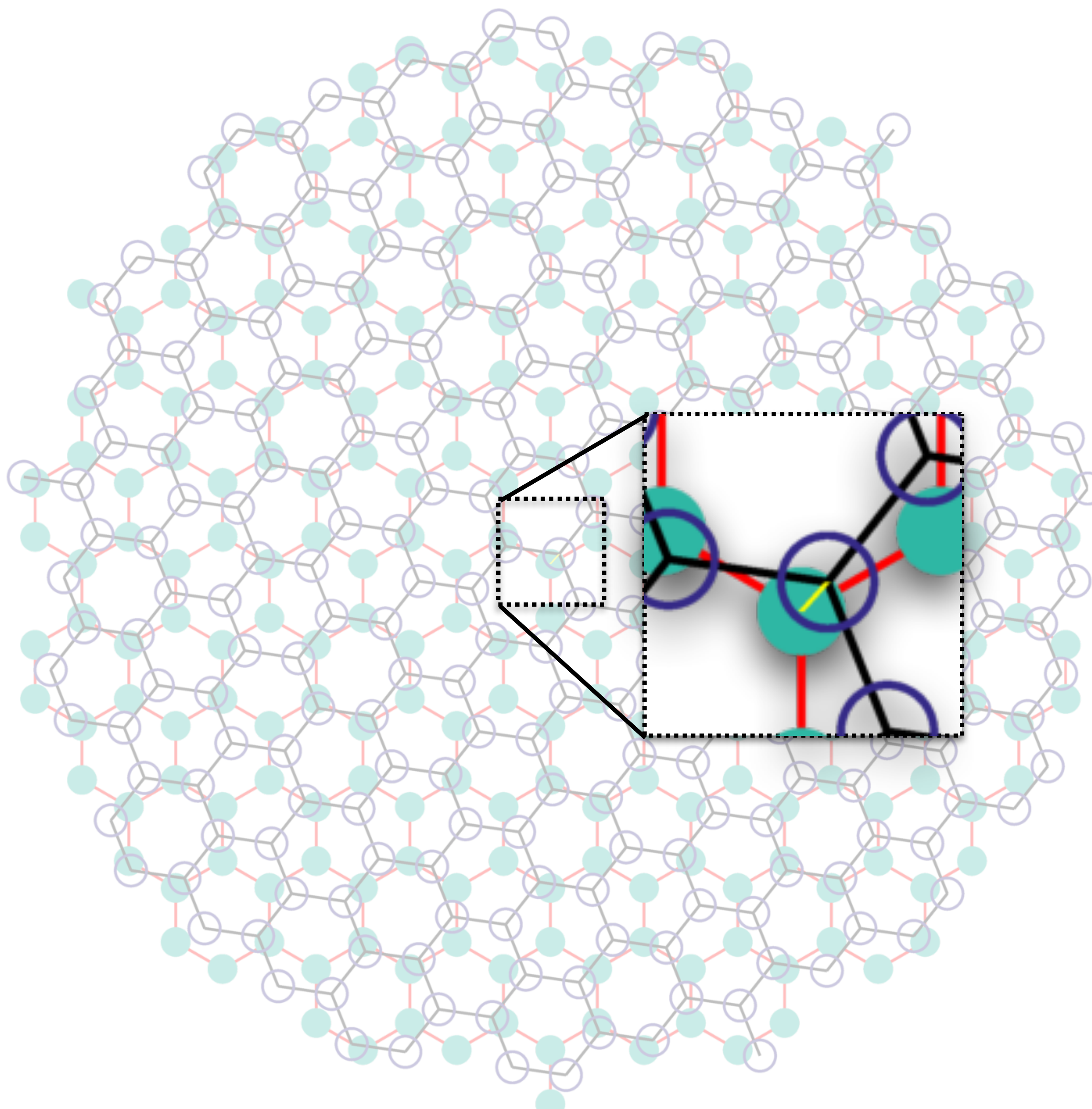
MICROSCOPIC DEGREES OF FREEDOM

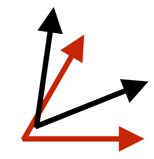
- Relative Lattice Translation: $\vec{d}$



MICROSCOPIC DEGREES OF FREEDOM

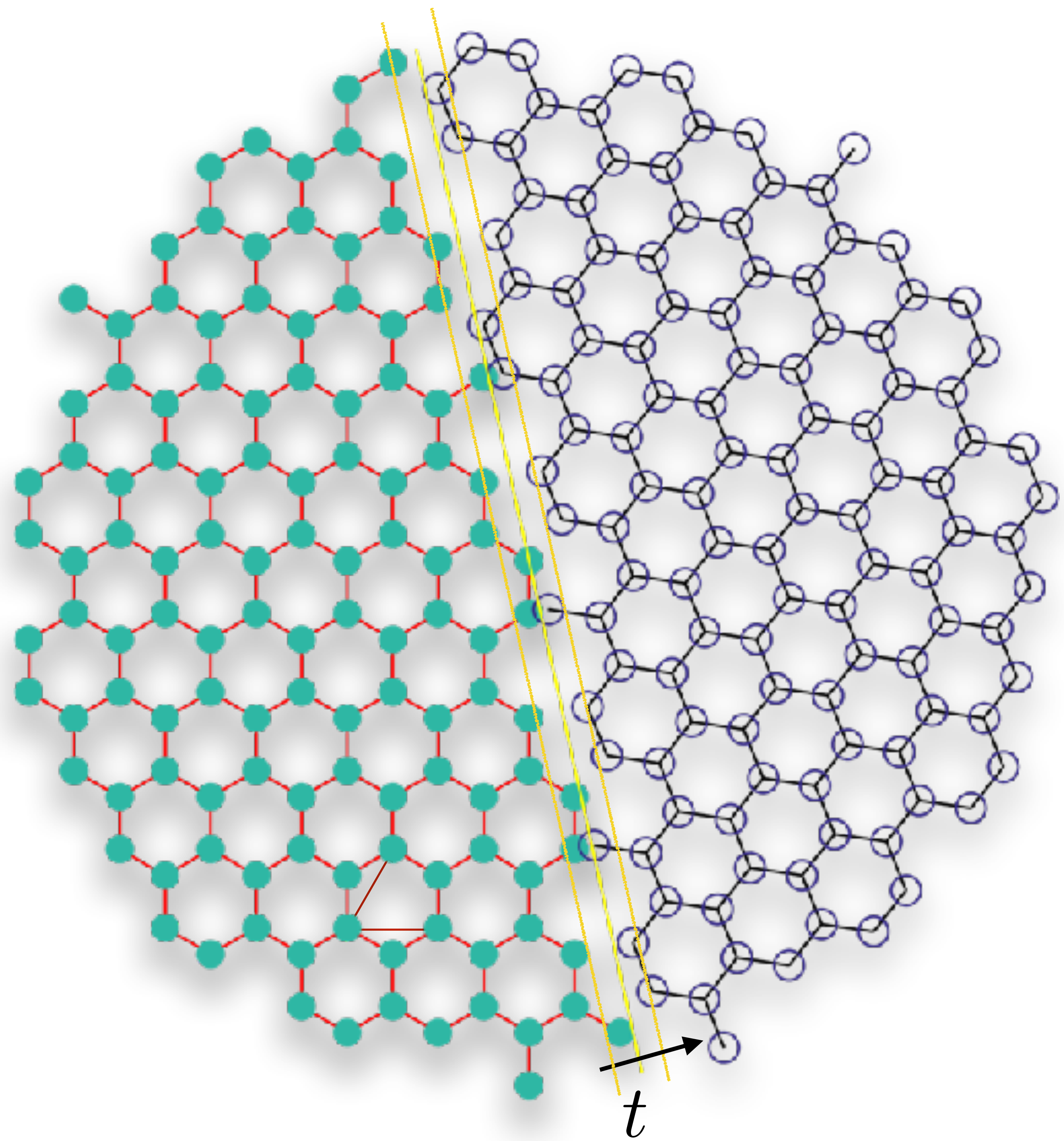
- Relative Lattice Translation: $\vec{d}$





MICROSCOPIC DEGREES OF FREEDOM

- Relative Lattice Translation: $\vec{d}$
- Boundary-plane translation: t



BOUNDS ON MICROSCOPIC DEGREES OF FREEDOM

Relative Lattice Translation: $\vec{d}$

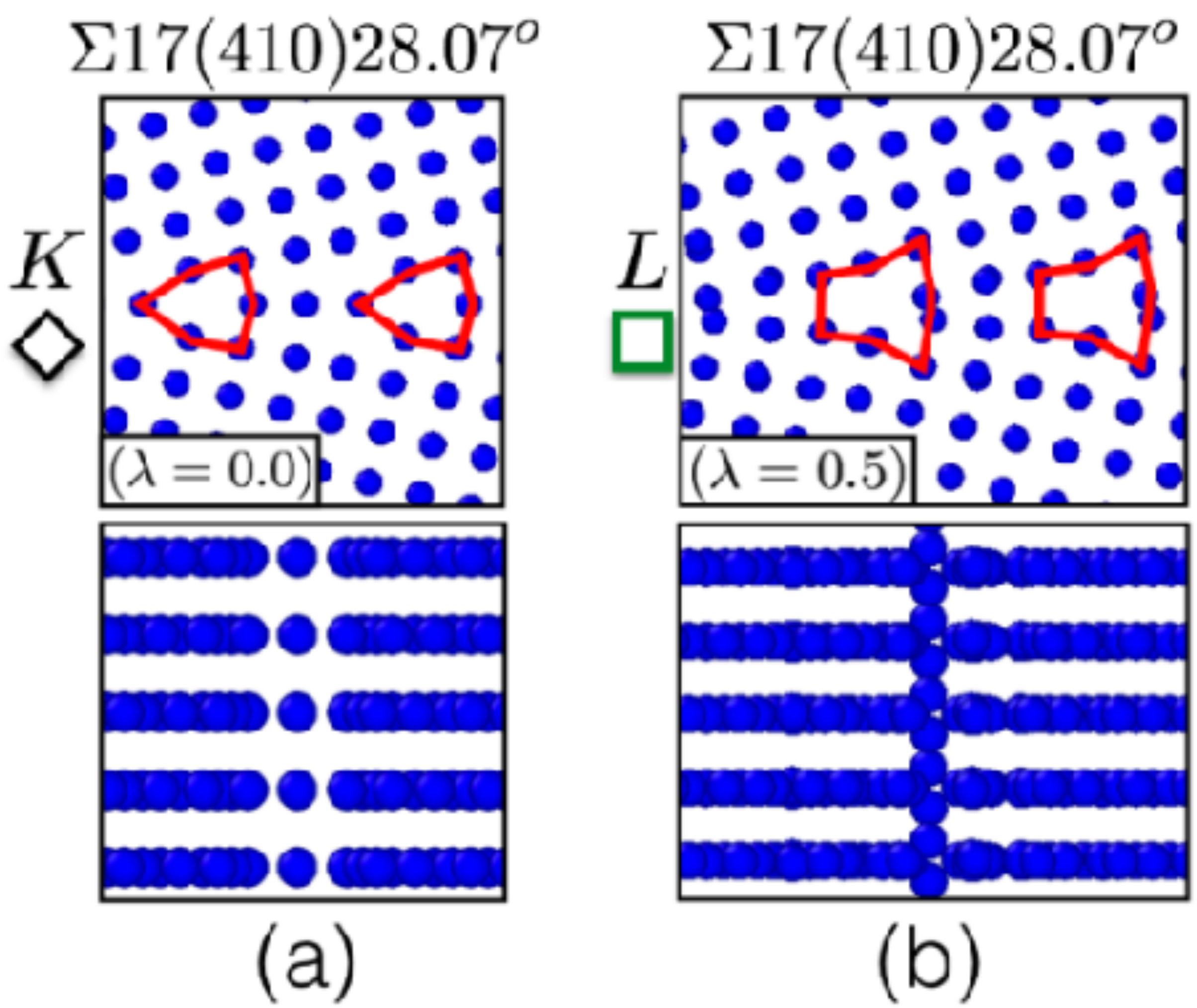
DSC Lattice

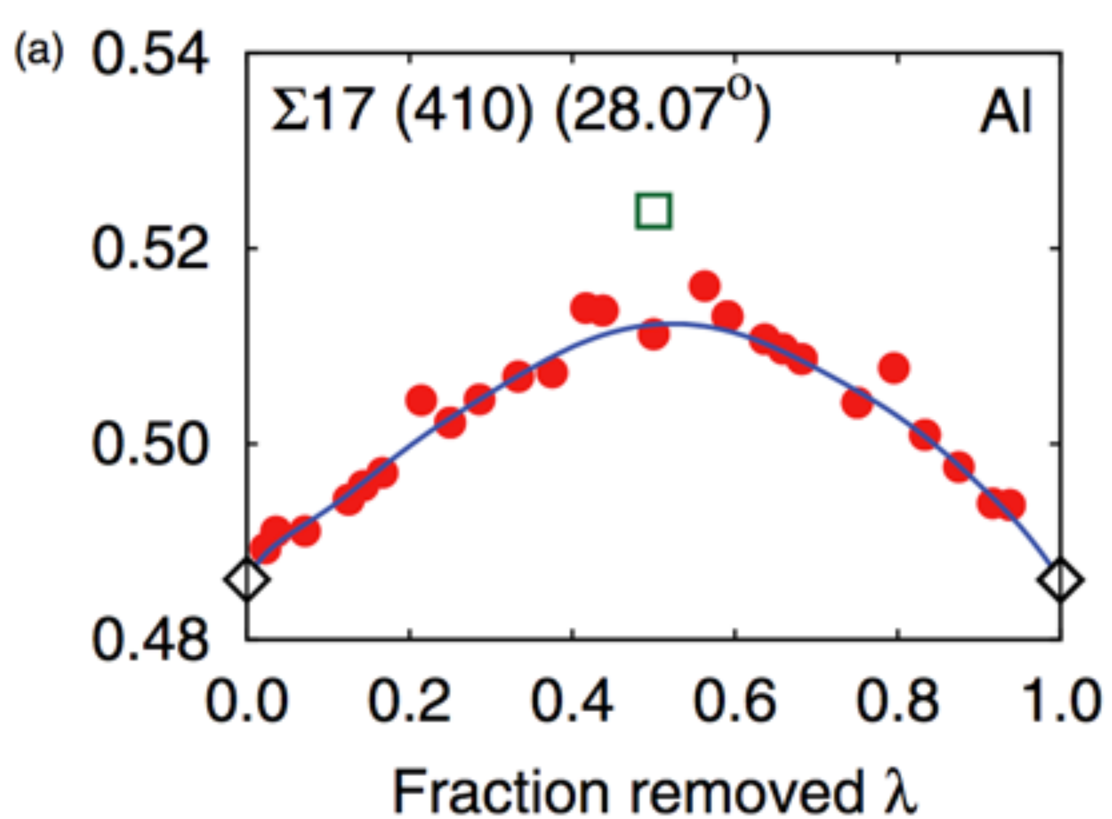
Boundary-Plane Translation: t

Inter-planar spacing (in CSL)

Grain Boundary Density [1]: λ

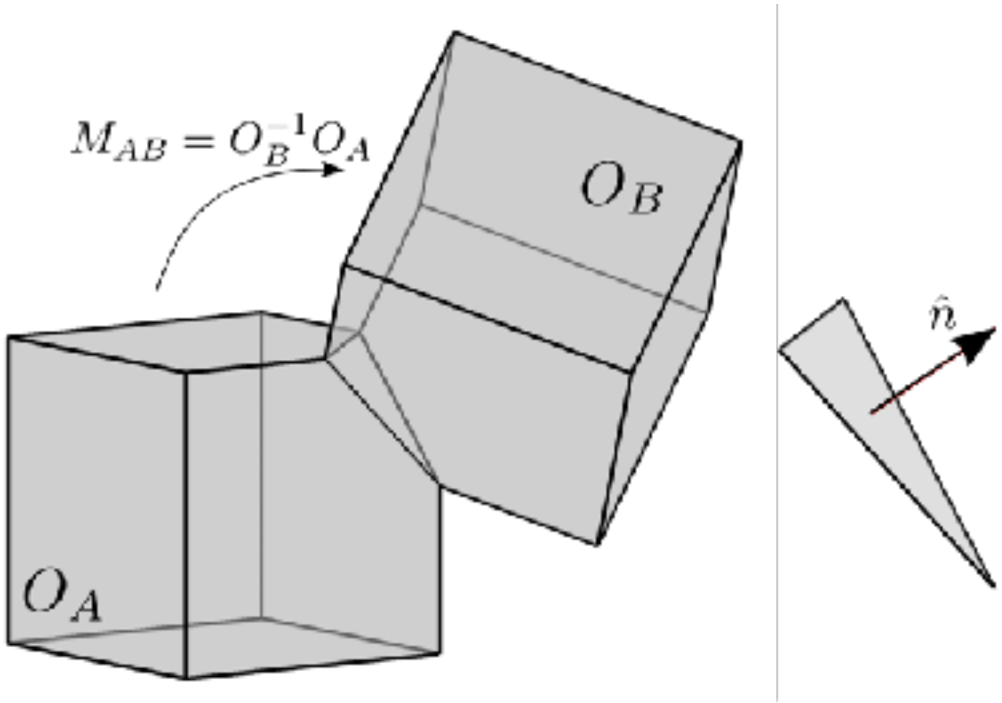
Extent of overlap





[1] Hickman, J., & Mishin, Y. (2017). Extra variable in grain boundary description. *Physical Review Materials*, *1*(1), 010601.

CRYSTALLOGRAPHY OF GBs



Macroscopic DOF

Misorientation

$$M = (\omega, \hat{a})$$

$$M = (\omega, \hat{a}) = (\omega, \theta, \phi)$$

Three-Parameters

Boundary-Plane Orientation:

$$\hat{n} = (\alpha, \beta)$$

Two-Parameters

Microscopic DOF

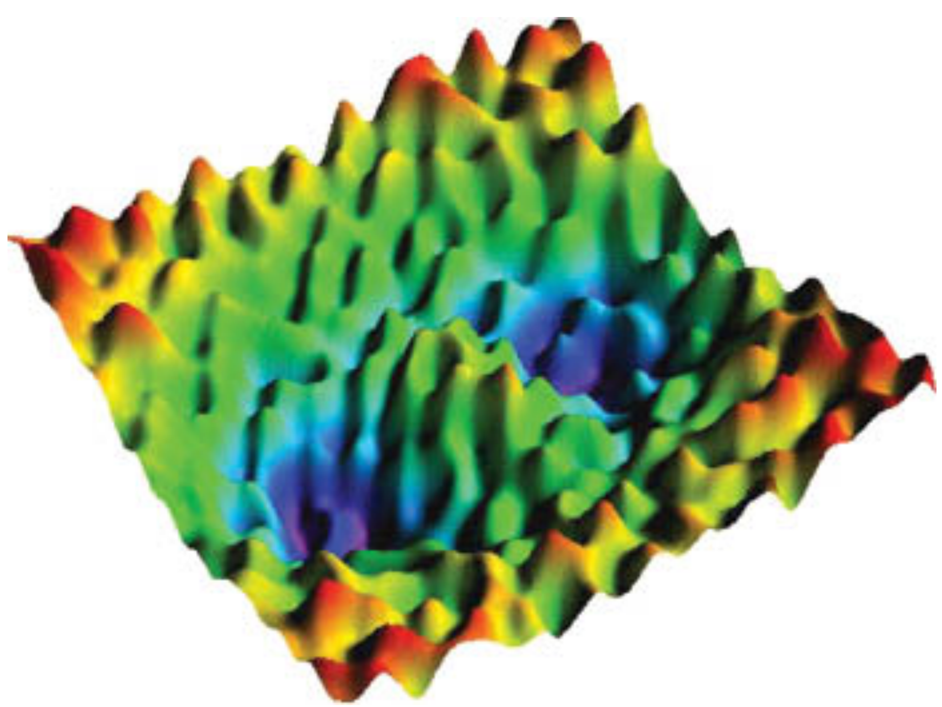
Relative Lattice Translation: $\vec{d}$

Boundary-Plane Translation: t

Grain Boundary Density: λ

Five Parameters

μ-energy-landscape



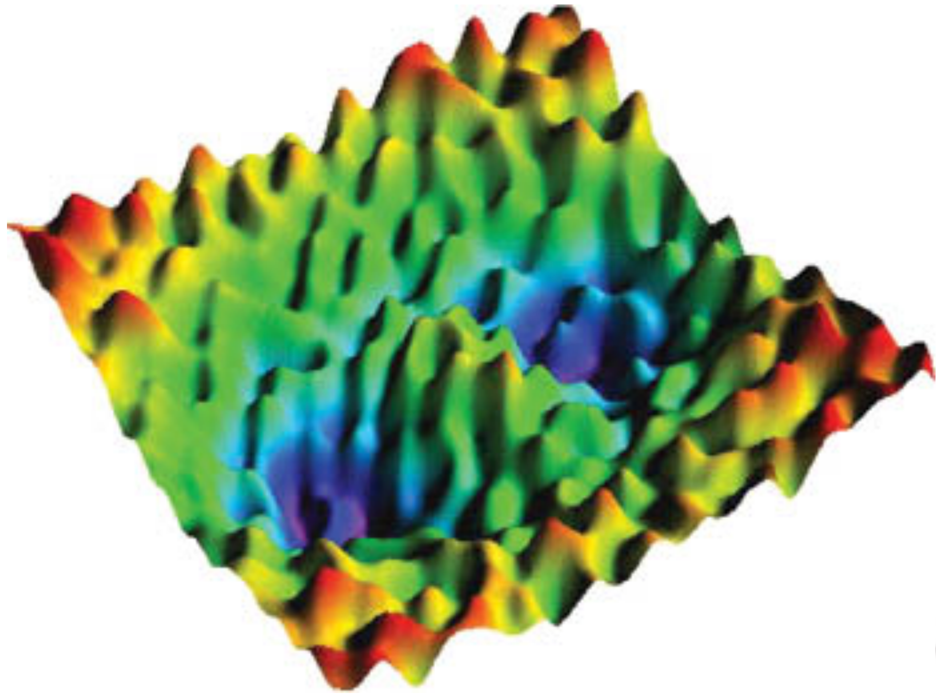
GB SIMULATIONS - BRUTE FORCE ALGORITHM

- **Minimum Energy Structures**
  - Discretize all possible micro-DOF
  - Usually 10,000-150,000 unique GB configurations have to be minimized.
- **Limitations**
  - Uncertainty in Convergence
    - Due to Discrete sampling of a Continuum space!
  - Computational Cost
    - A vast microscopic configurational search space!

Microscopic DOF

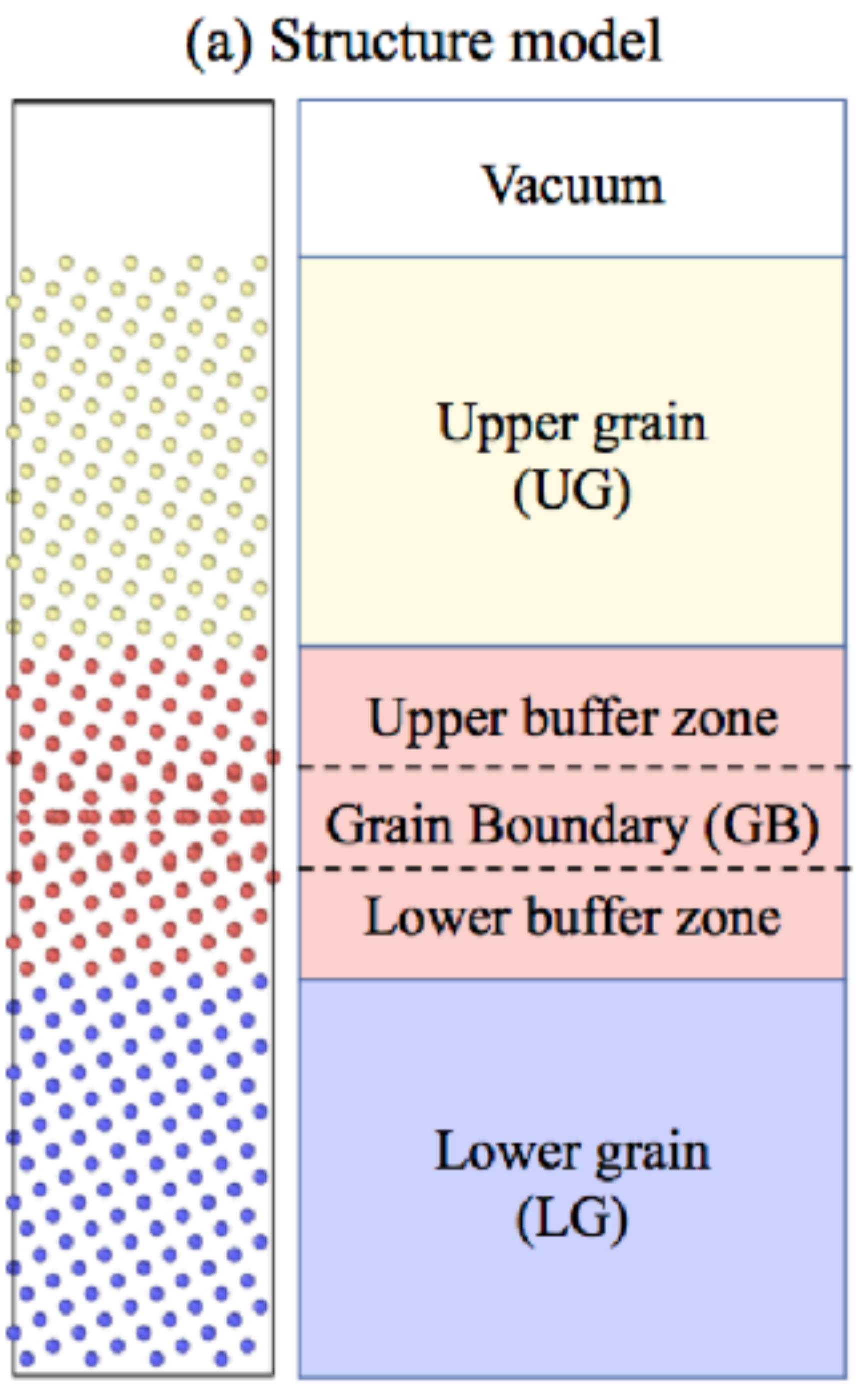
Relative Lattice Translation:	$\vec{d}$
Boundary-Plane Translation:	t
Grain Boundary Density:	λ

μ-energy-landscape



EVOLUTIONARY SEARCH

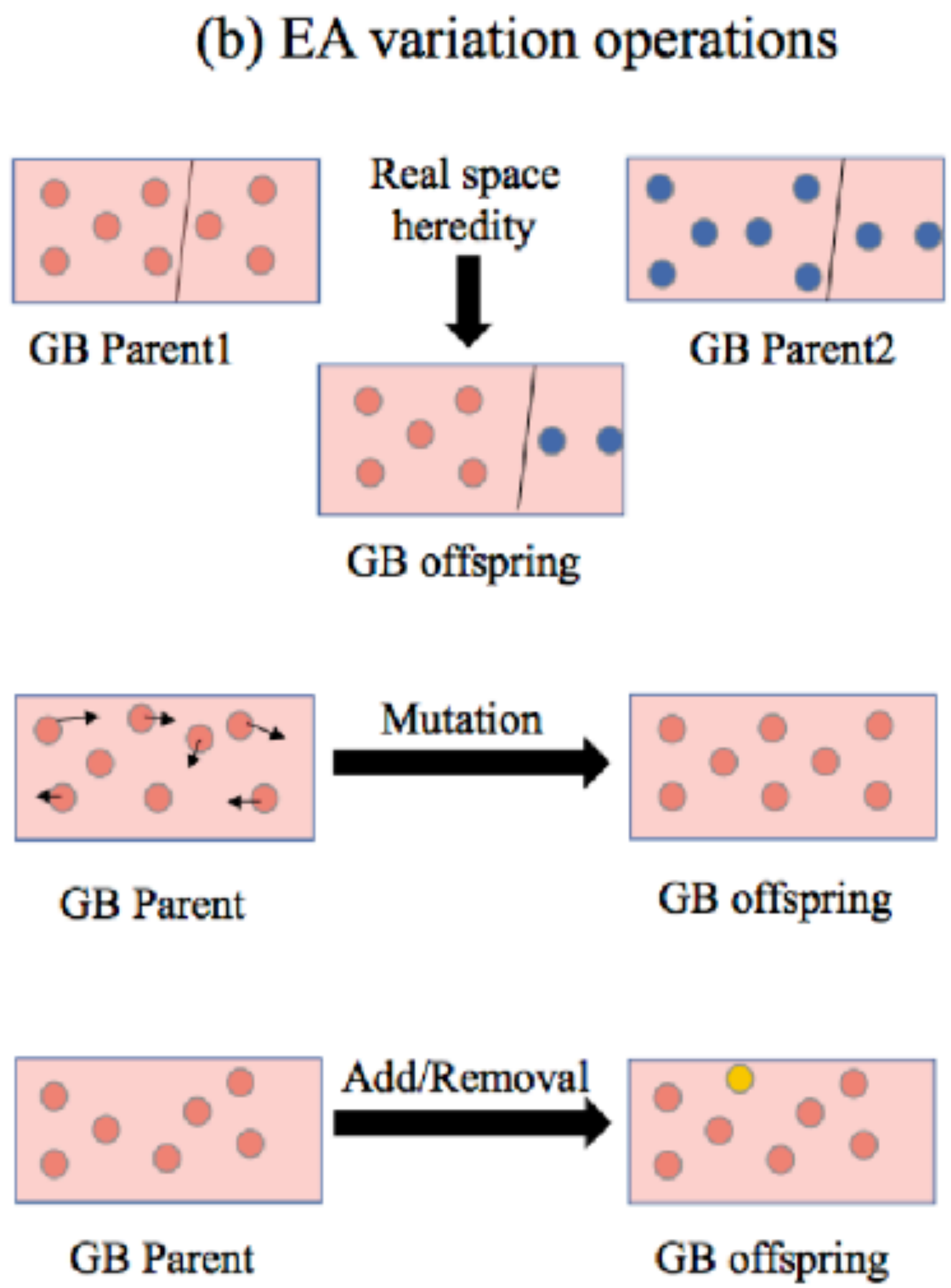
- The first generation of GB structures are created (randomly with certain symmetry constraints).
- The energy minimization is followed by fitness evaluation, namely, the excess GB energy calculation.
- After that the parents are chosen from the top 60% of structures according to the tournament selection, which ensures a higher probability for GBs with higher fitness.



Zhu, Q., Samanta, A., Li, B., Rudd, R. E., & Frolov, T. (2018). Predicting phase behavior of grain boundaries with evolutionary search and machine learning. Nature communications, 9(1), 467.

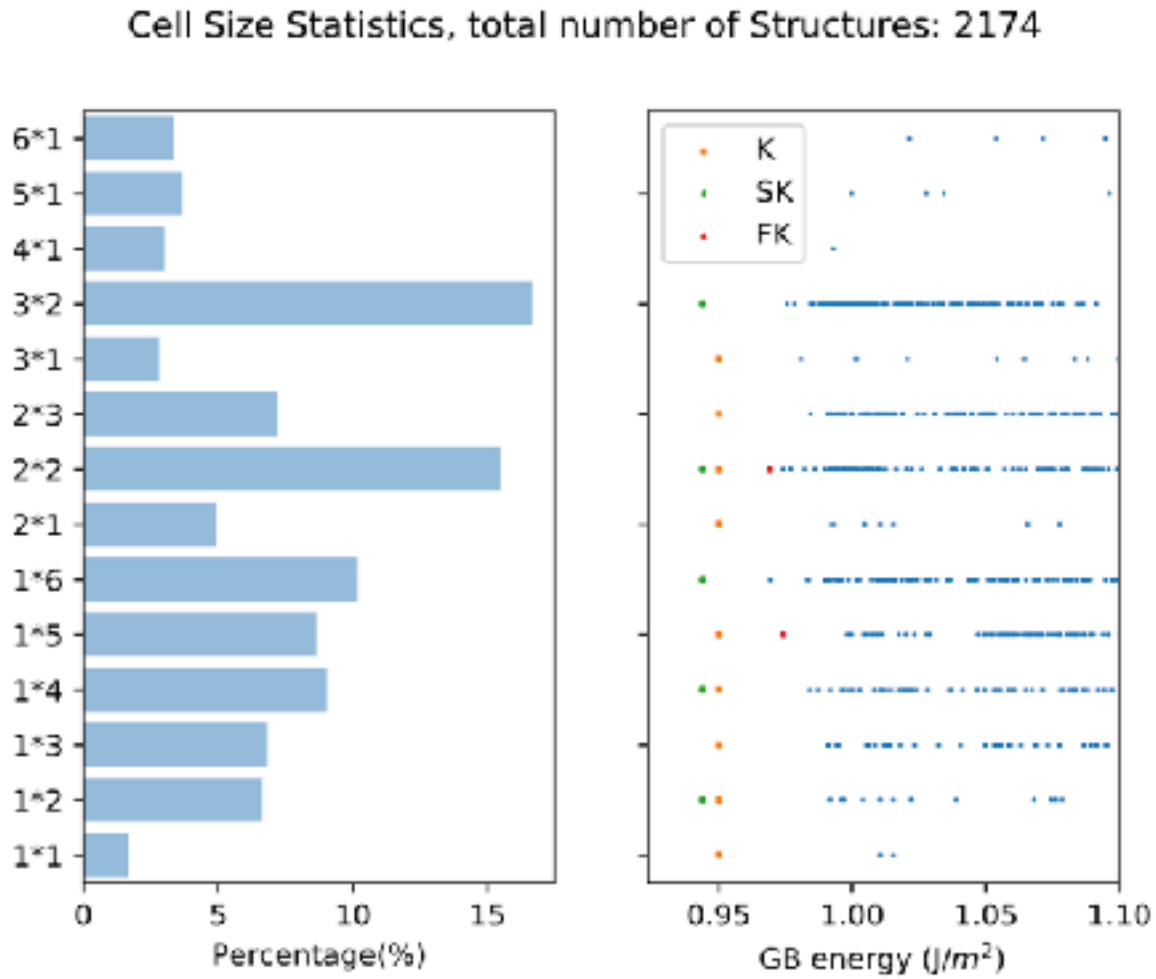
EVOLUTIONARY SEARCH

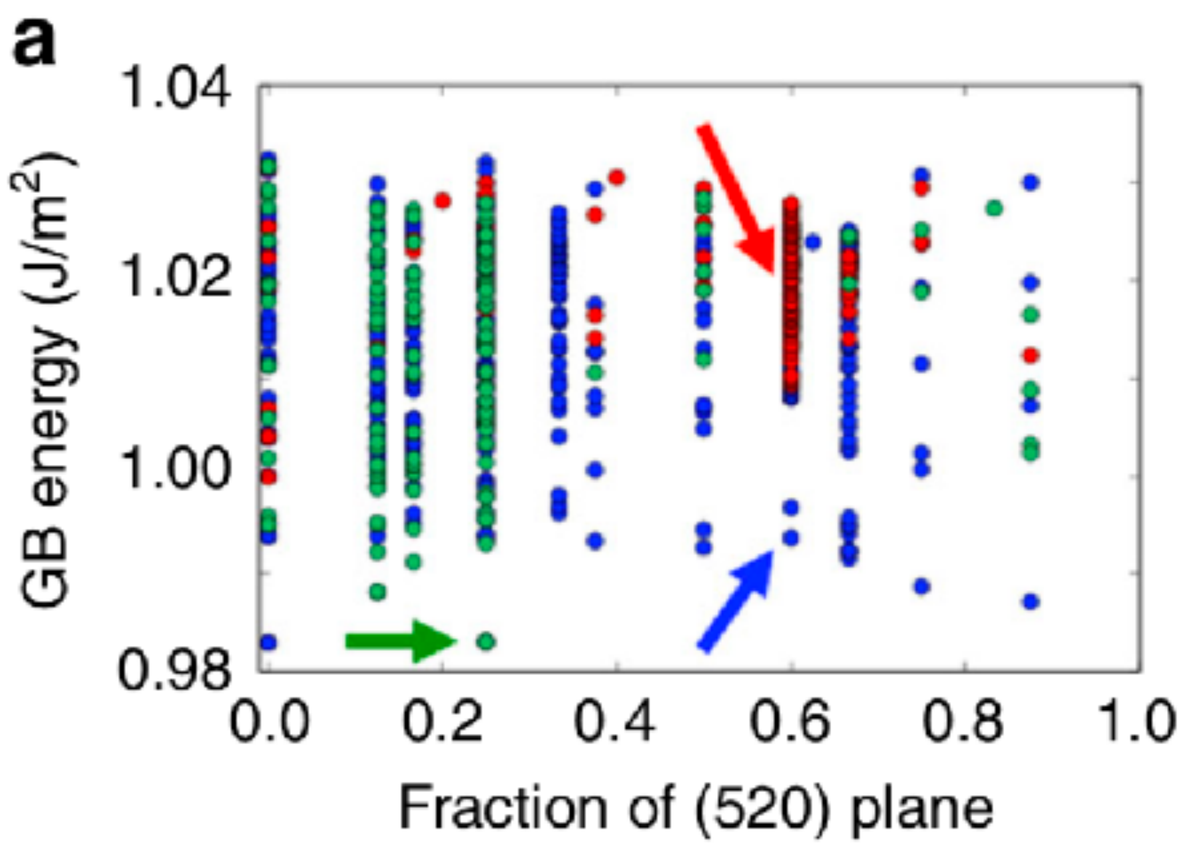
- The child structures for the next generation are produced in the following way:
  - **Heredity**, which choses two GB structures and randomly slices them at the same position in the GB unit cell and then combines the pieces to generate the offspring;
  - **Mutation**
    - **Displace** GB atoms according to the stochastically picked soft vibrational modes based a bond-hardness model;
    - **GB dimensions** are allowed to change automatically during the search
    - **Insertion/removal** of atoms, which changes the number of atoms in the GB slab.



Zhu, Q., Samanta, A., Li, B., Rudd, R. E., & Frolov, T. (2018). Predicting phase behavior of grain boundaries with evolutionary search and machine learning. Nature communications, 9(1), 467.

EVOLUTIONARY SEARCH





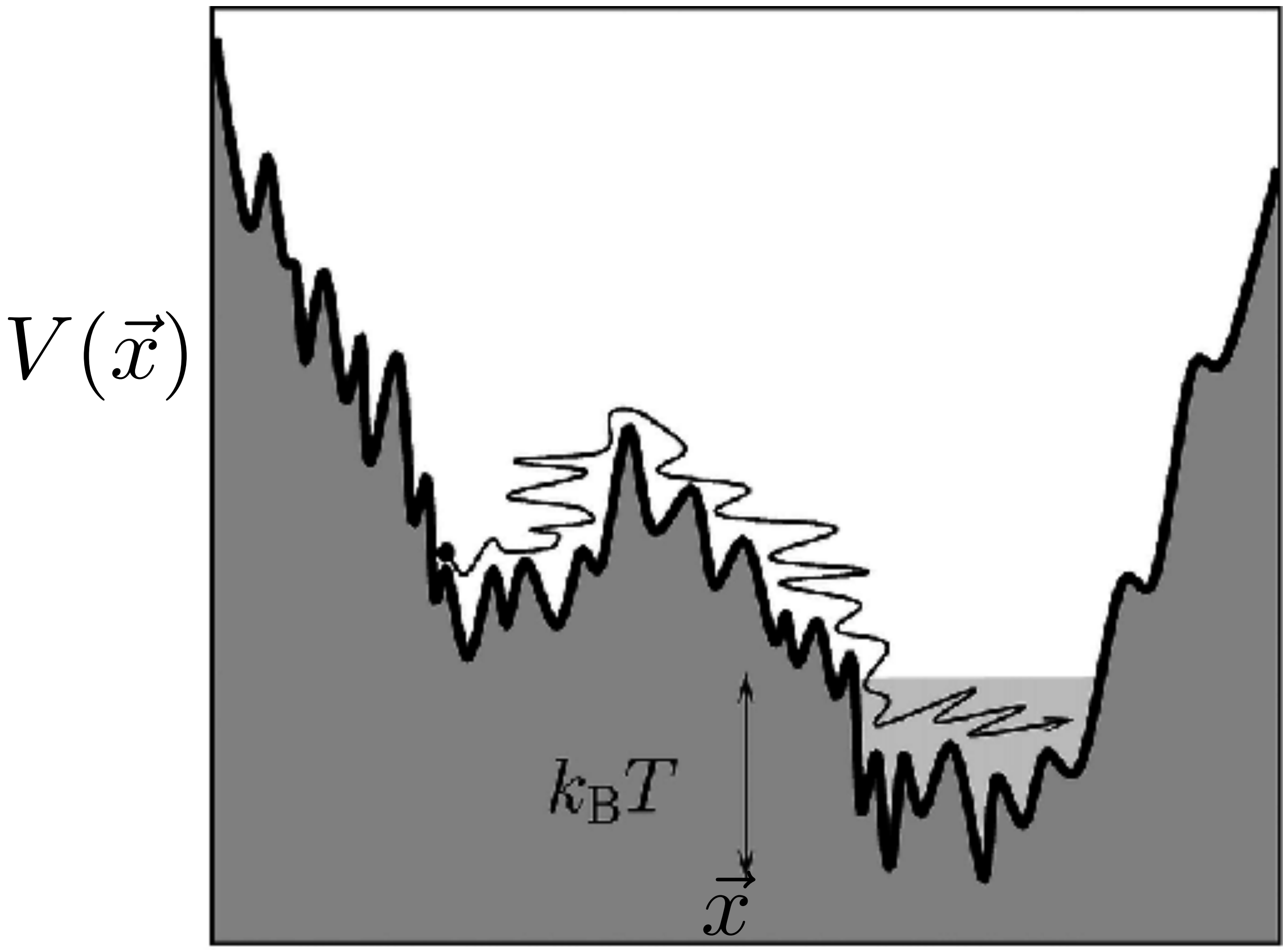
- The GB cross-section statistics for a typical structure search for the Σ5(210)[001] GB in Cu.

- Evolutionary search for the Σ29(520) [001] predict three grain boundary phases

Zhu, Q., Samanta, A., Li, B., Rudd, R. E., & Frolov, T. (2018). Predicting phase behavior of grain boundaries with evolutionary search and machine learning. Nature communications, 9(1), 467.

EFFICIENT GB STRUCTURE GENERATION

- A hybrid Monte-Carlo/Molecular-Dynamics (MC/MD) Scheme for sampling the GB energy landscape.



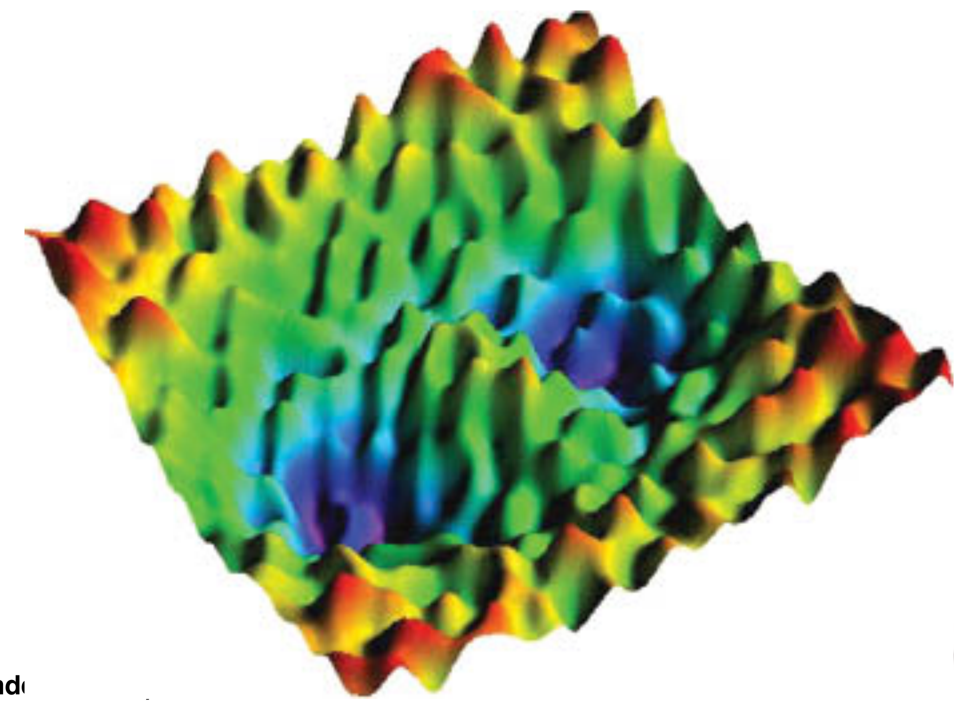
Microscopic DOF

Relative Lattice Translation: $\vec{d}$

Boundary-Plane Translation: t

Grain Boundary Density: λ

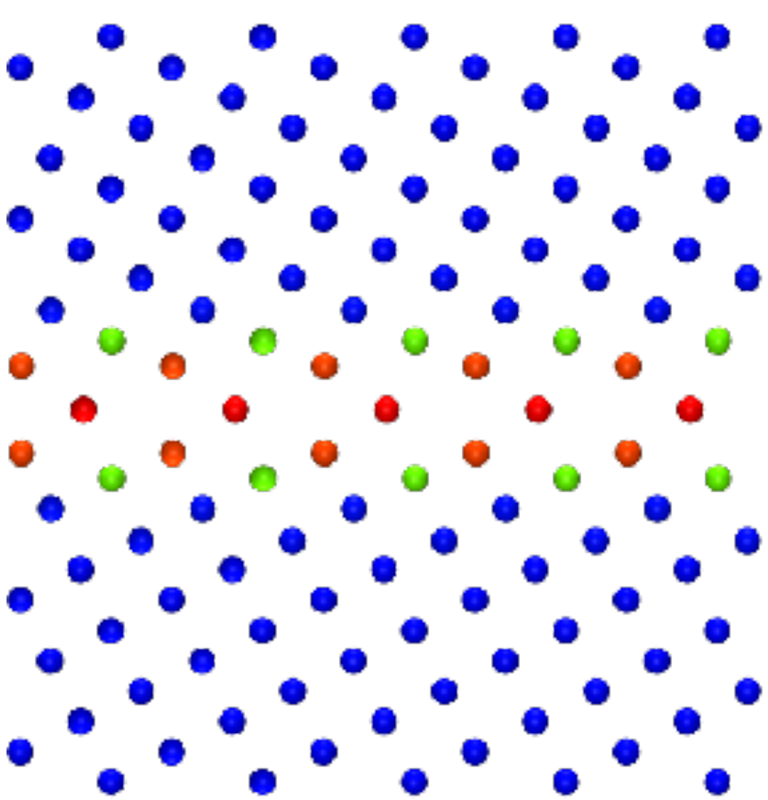
μ-energy-landscape

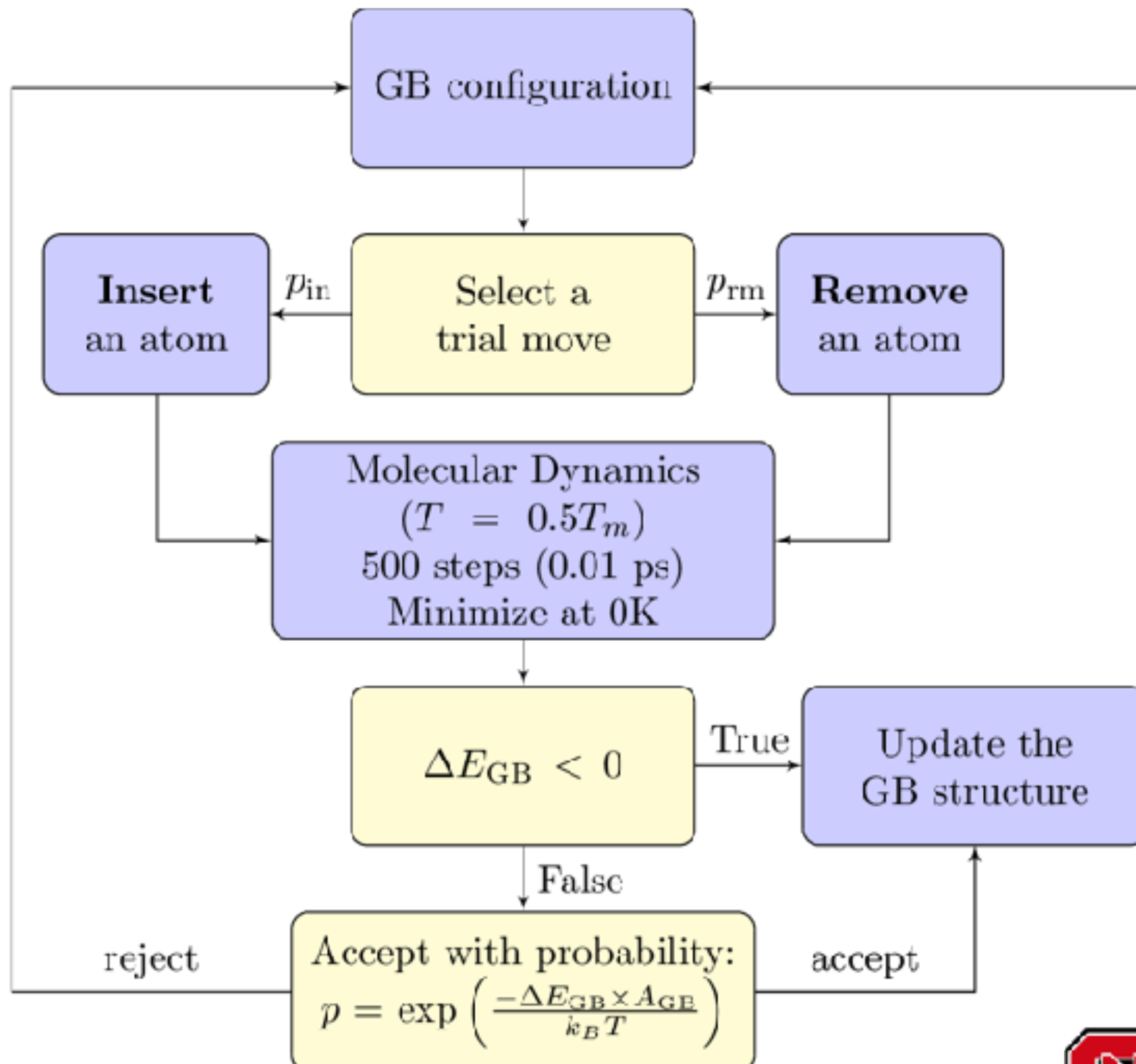


[1] AD Banadaki, MA Tschopp, S. Patala. A Monte-Carlo Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Und

TRIAL MOVES - FUNDAMENTAL PERTURBATIONS

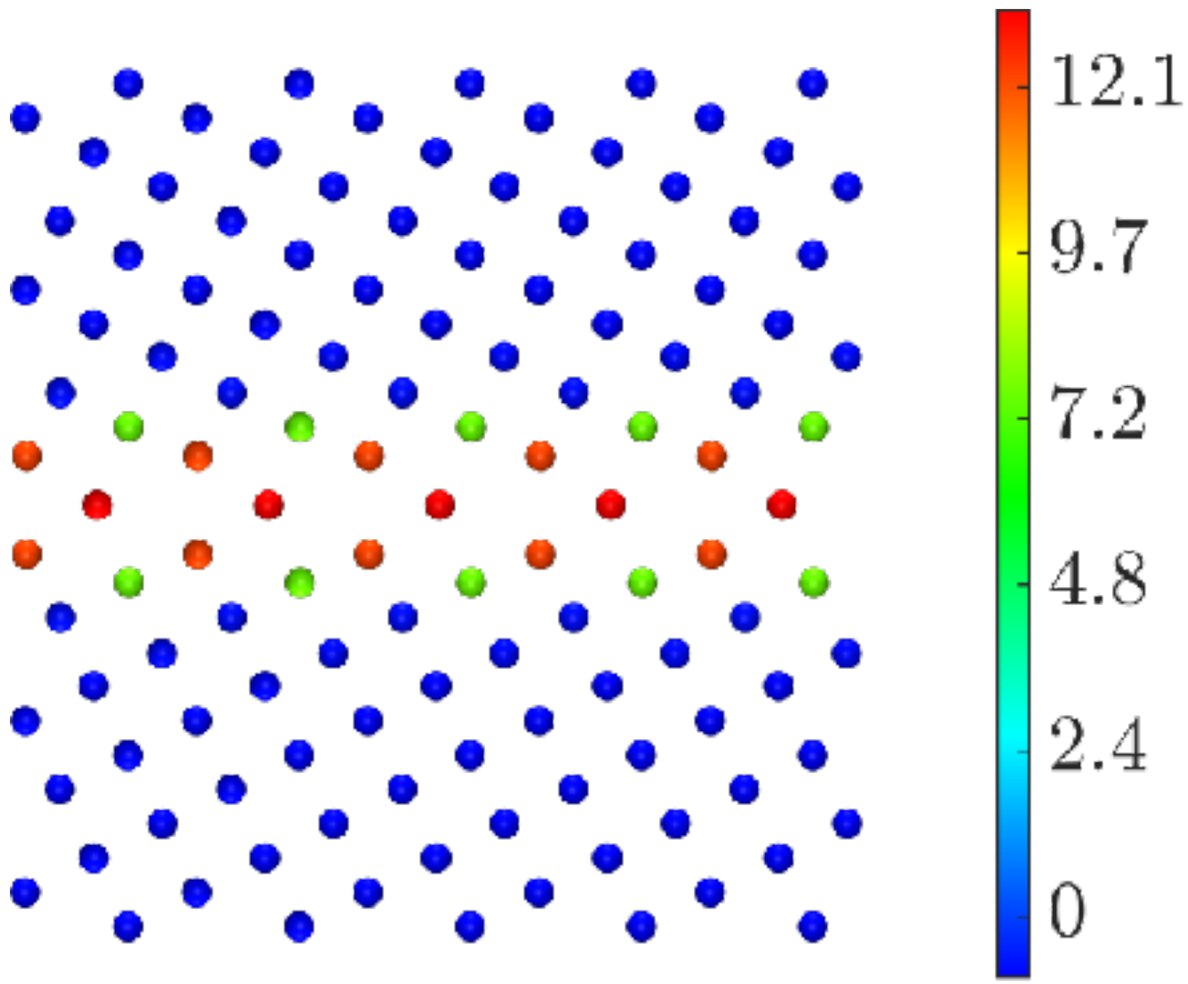
- Hybrid MC/MD Scheme
  - Monte Carlo
    - **Atom Removal**
    - **Atom Insertion**
  - Molecular Dynamics
    - Thermal vibrations



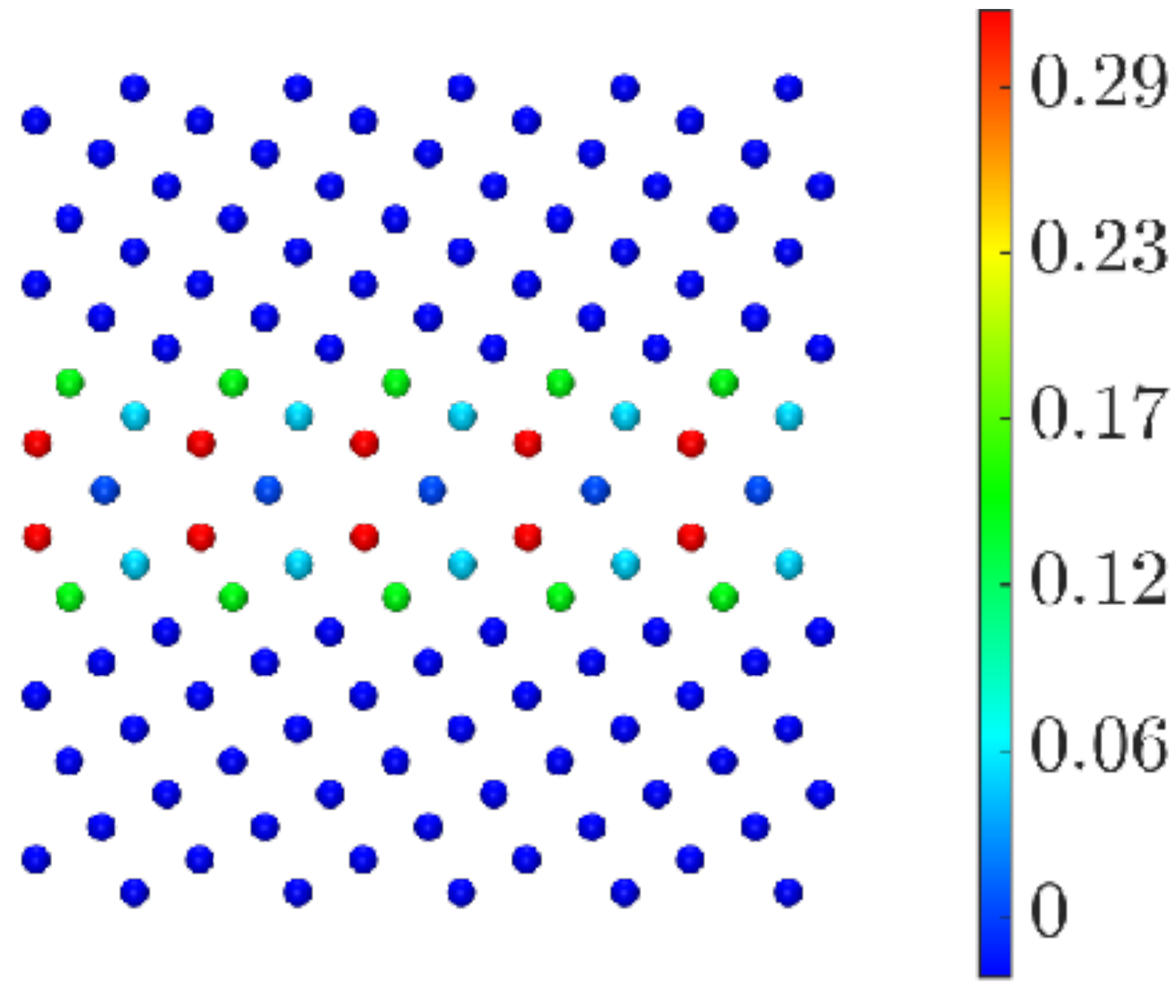


[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

TRIAL MOVES - ATOM REMOVAL



Atom Energy (eV/atom)

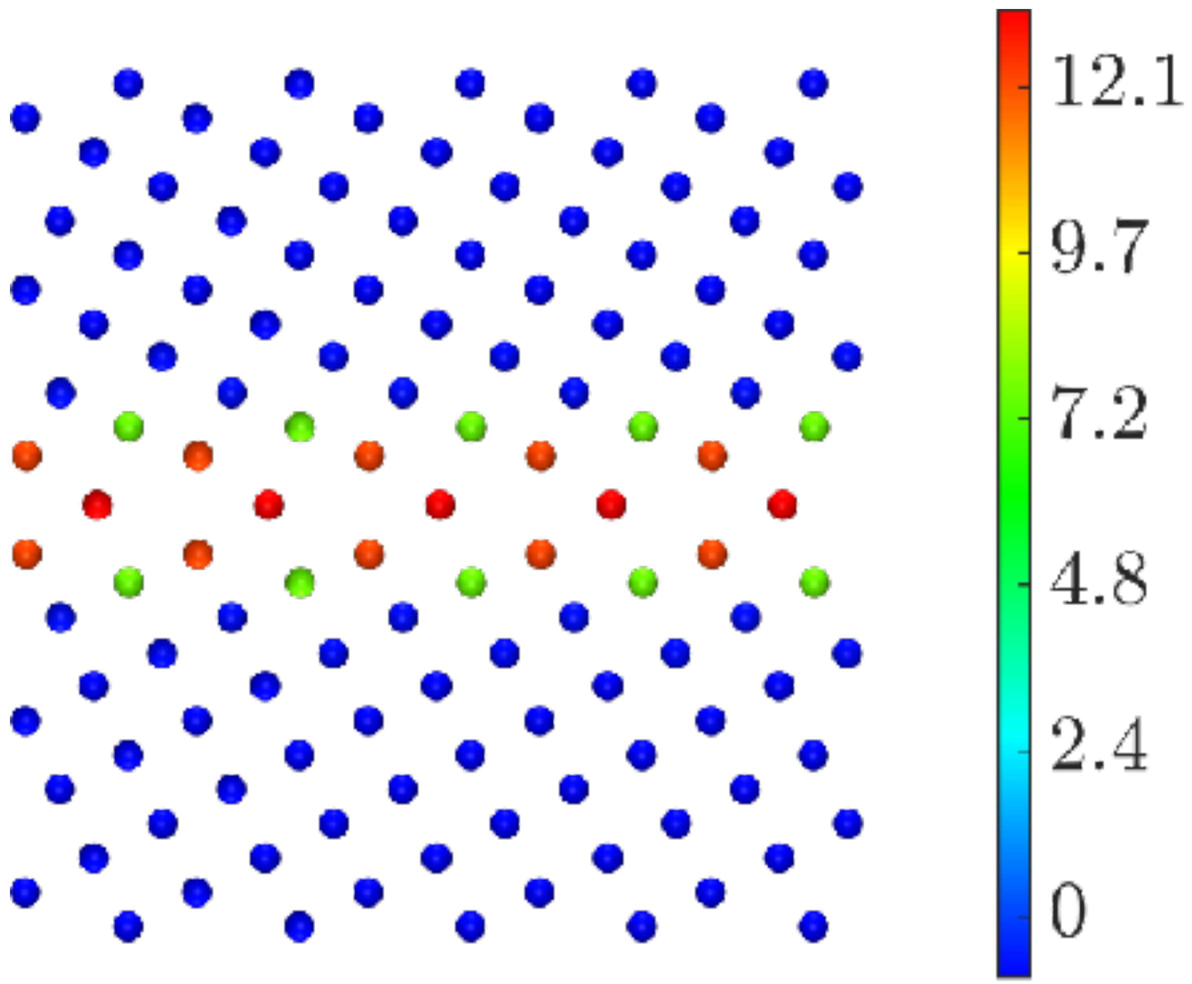


Removal Probability

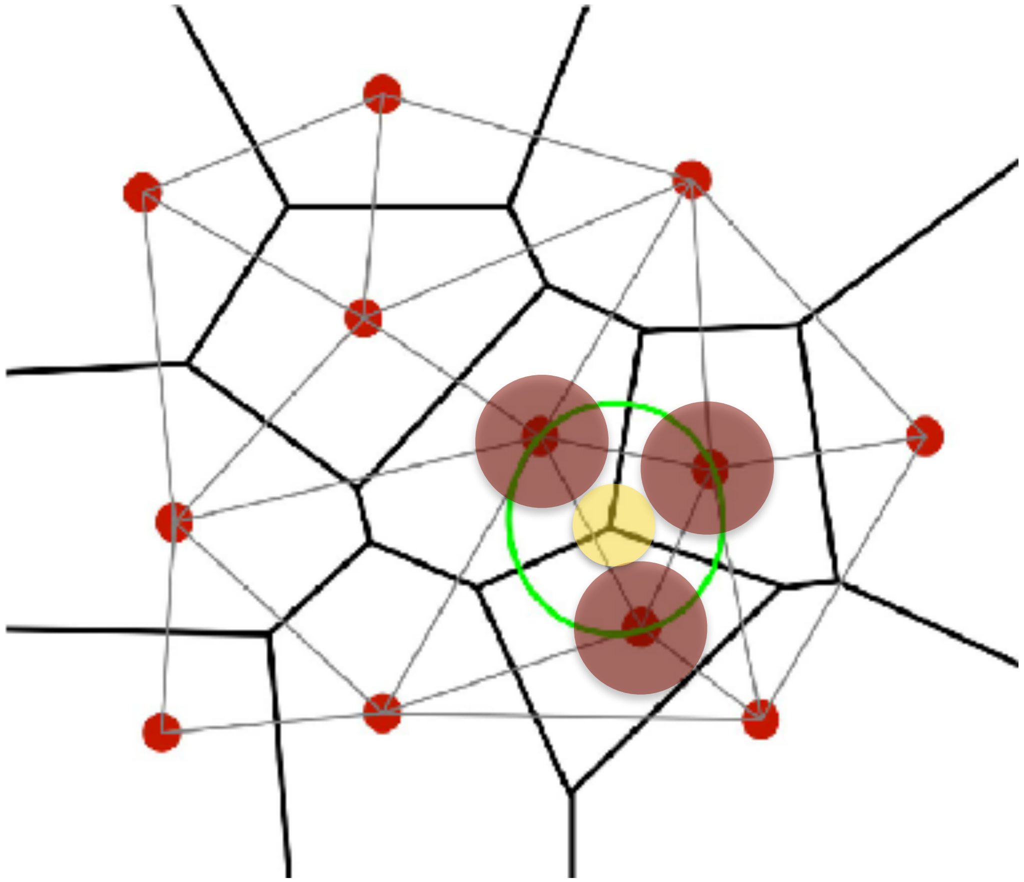
$$p_{\mathrm{rm},i} = \begin{cases} (E_i - E_0) \,/\, \left(\sum_{j=1}^{N_{\mathrm{GB}}} (E_j - E_0) \right), & \text{if } E_i \geq E_0 \\ 0, & \text{otherwise} \end{cases}$$

[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

TRIAL MOVES - ATOM INSERTION

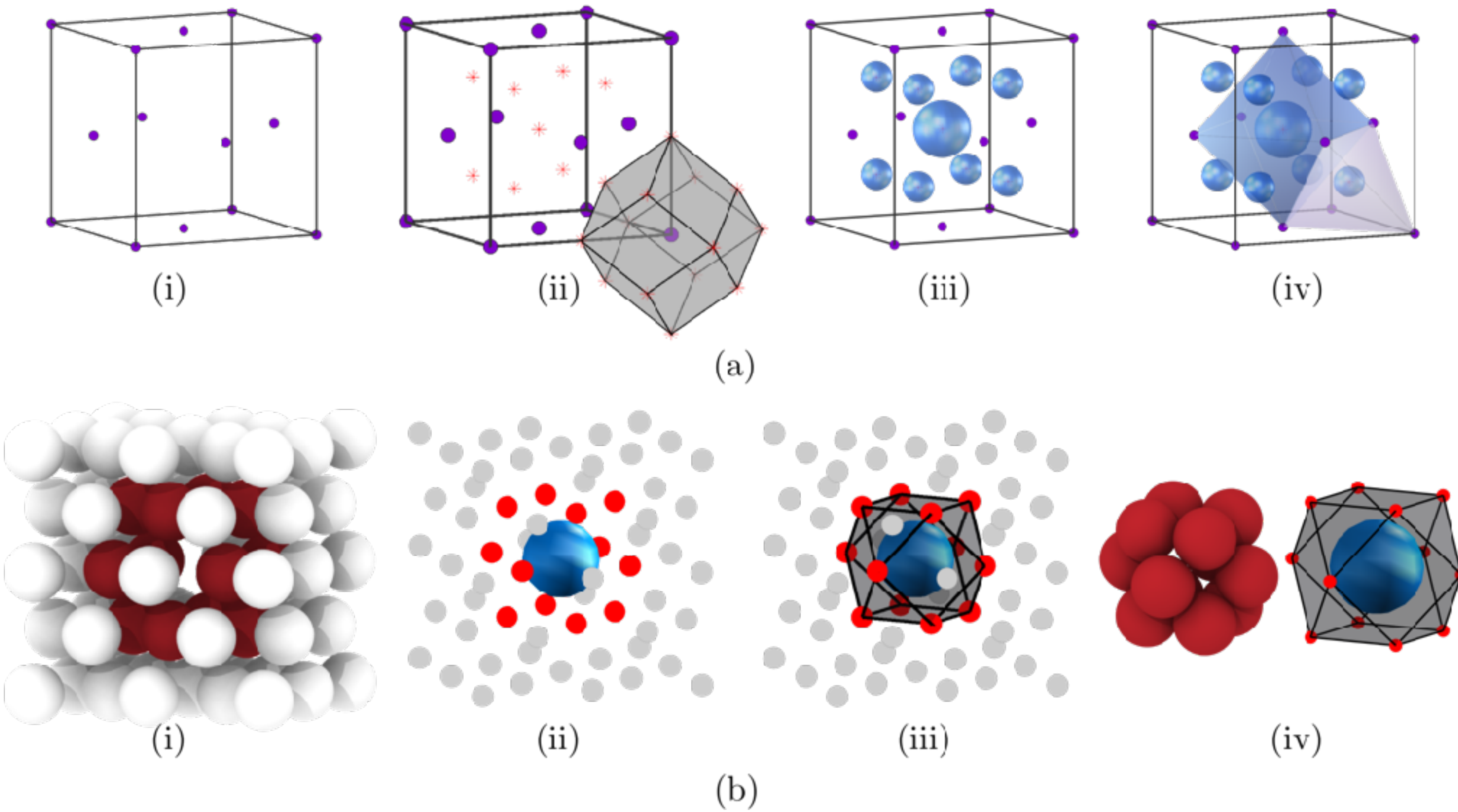


Atom Energy (eV/atom)



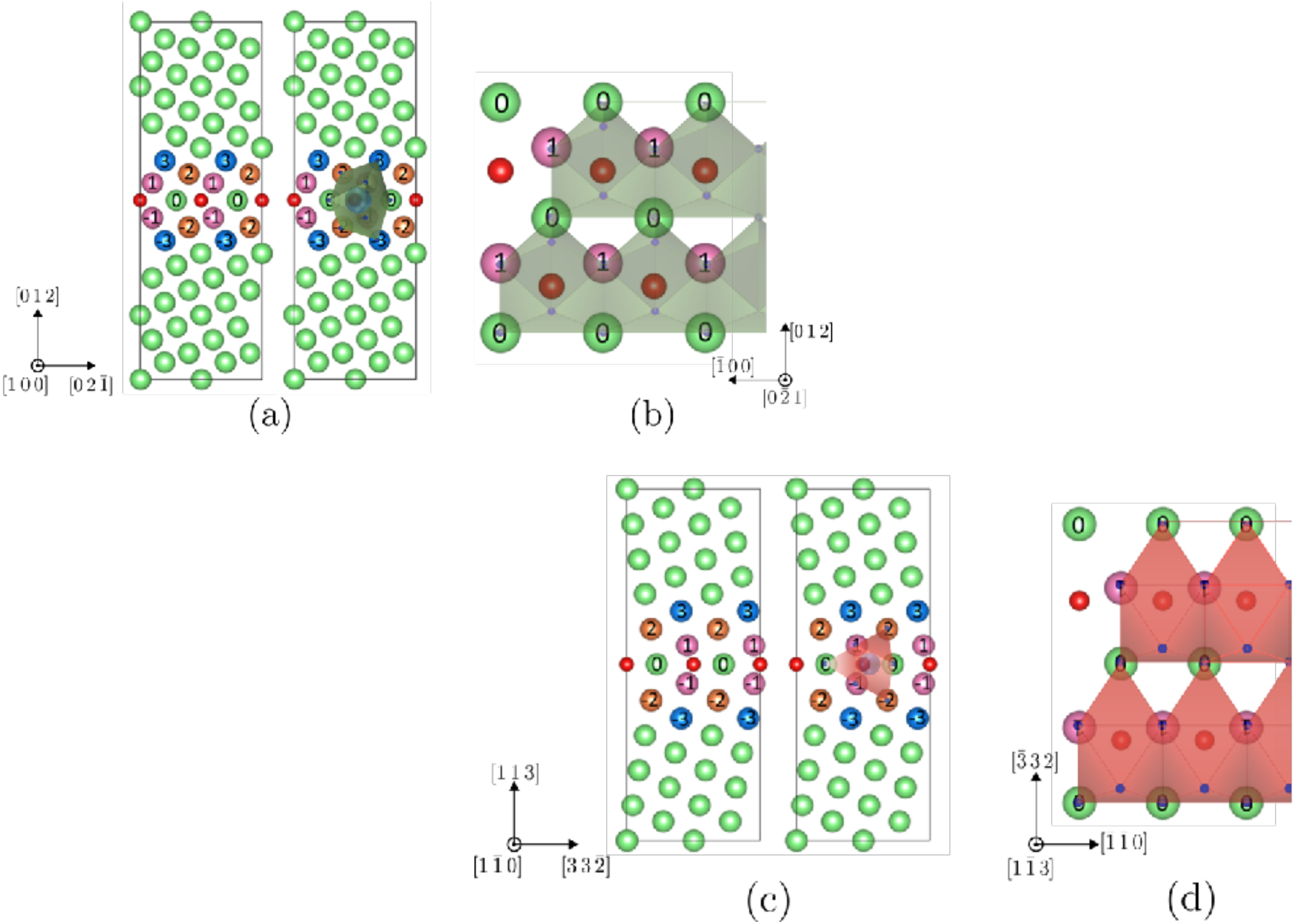
[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

IDENTIFYING VOIDS IN CONDENSED MATTER SYSTEMS



Banadaki, A. D., & Patala, S. (2017). A three-dimensional polyhedral unit model for grain boundary structure in fcc metals. *npj Computational Materials*, *3*(1), 13.

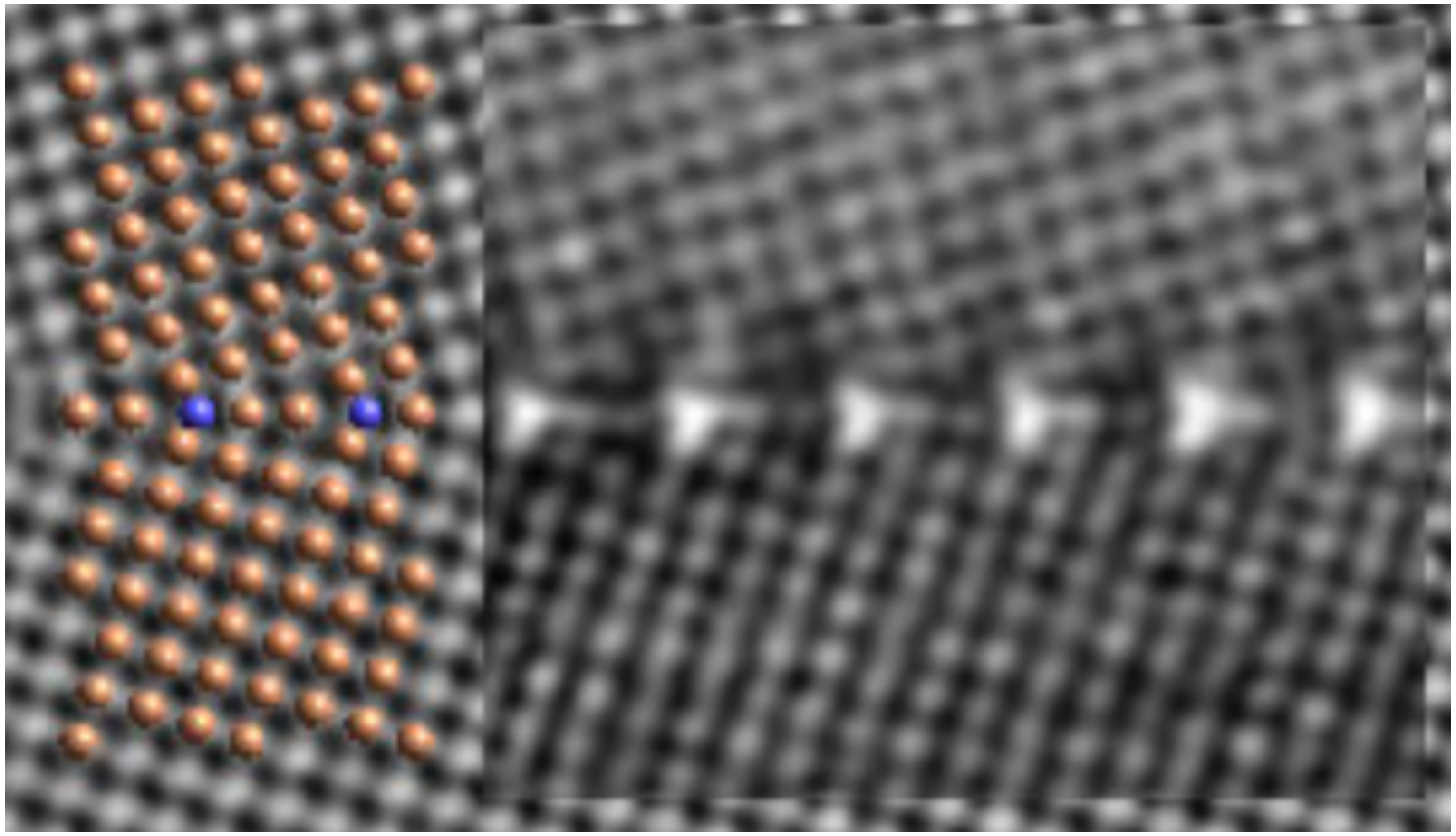
SEGREGATION - HYDROGEN IN NICKEL GBs



Banadaki, A. D., & Patala, S. (2017). A three-dimensional polyhedral unit model for grain boundary structure in fcc metals. *npj Computational Materials*, *3*(1), 13.

SEGREGATION - COPPER IN ALUMINUM GBs

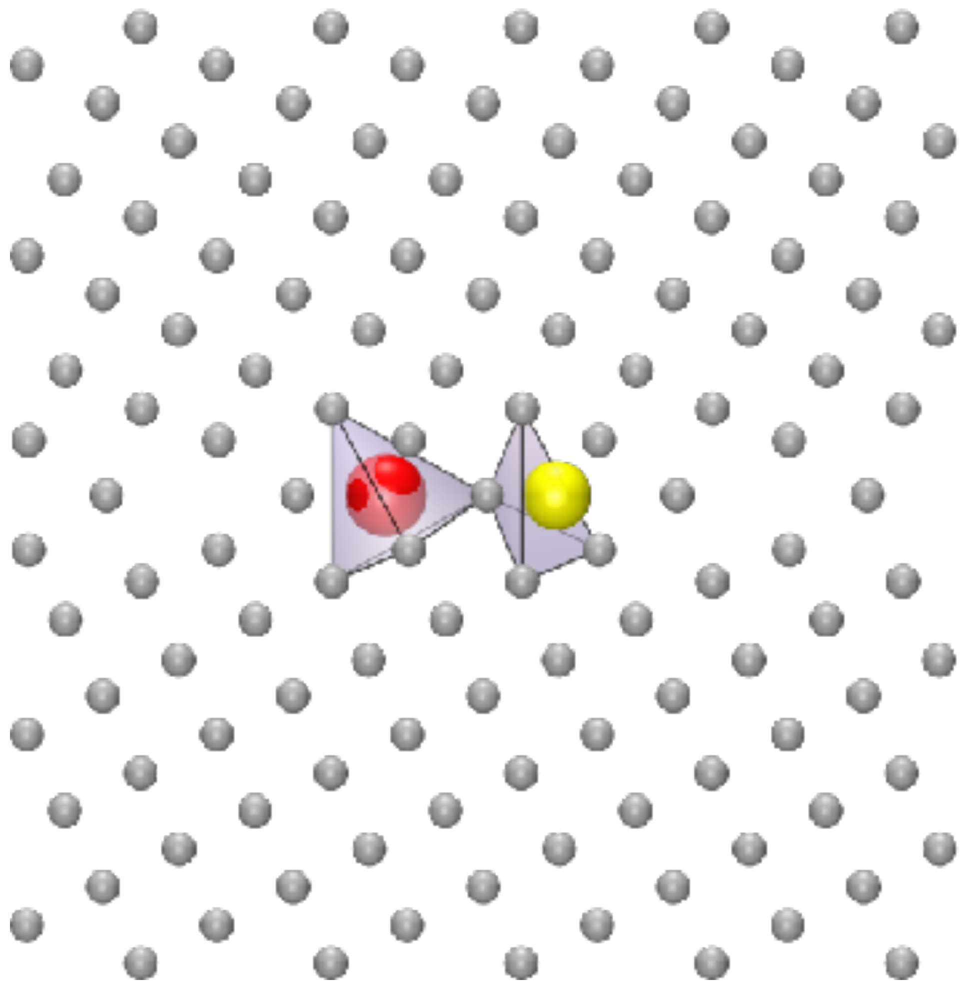
- Grain Boundaries in Multi-component systems

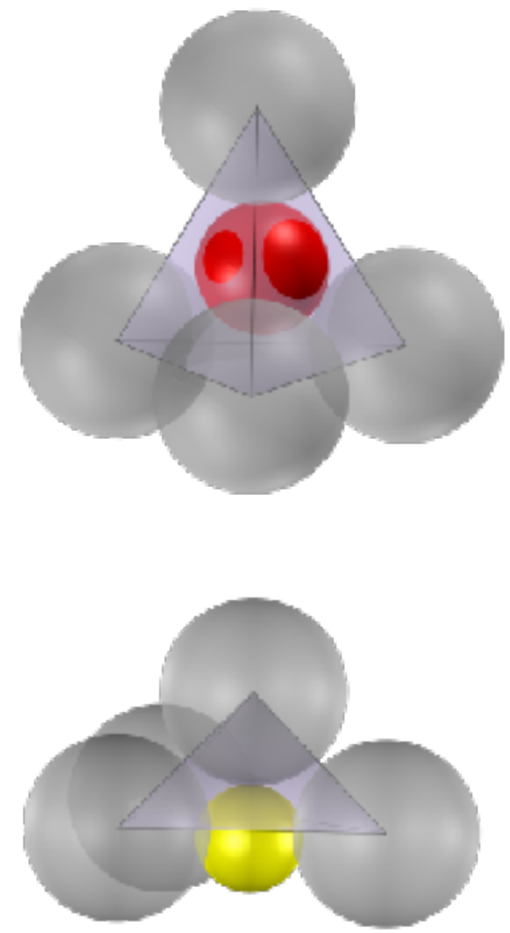


A composite image combining the data acquired in the TEM and the atomistic simulations. The alignment of the z-contrast image with the atomic model clearly show the signal arising from the interstitial site in the GB [1].

[1] Campbell, G. H., Plitzko, J. M., King, W. E., Foiles, S. M., Kisielowski, C., & Duscher, G. J. (2004). Copper segregation to the Σ5 (310)/[001] symmetric tilt grain boundary in aluminum. *Interface Science*, *12*(2-3), 165-174.

TRIAL MOVES ATOM INSERTION



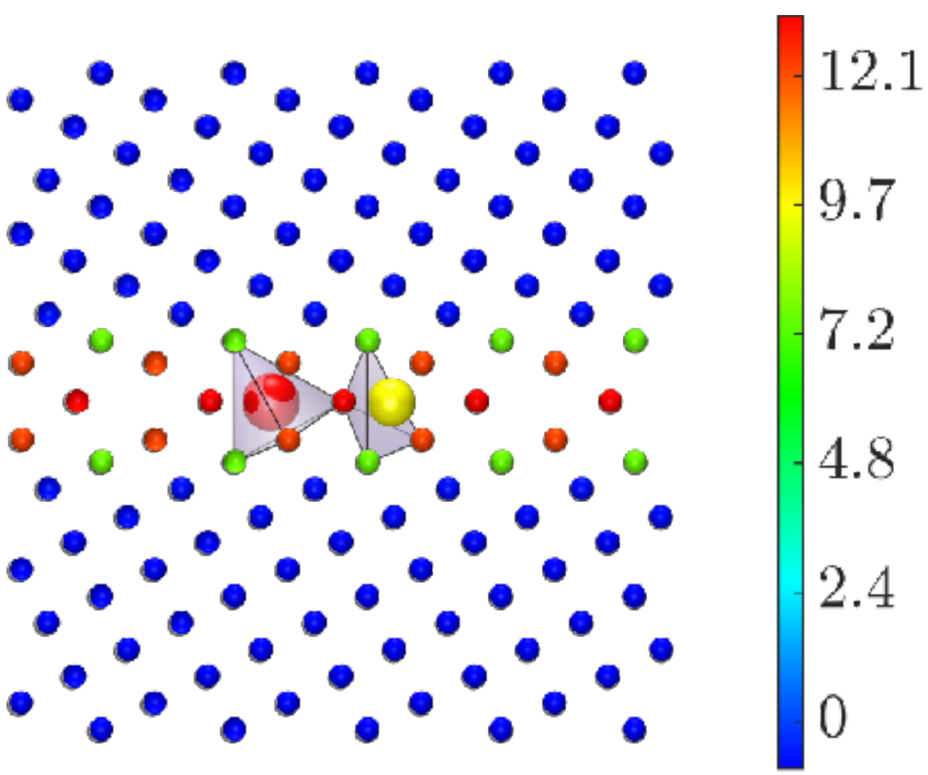


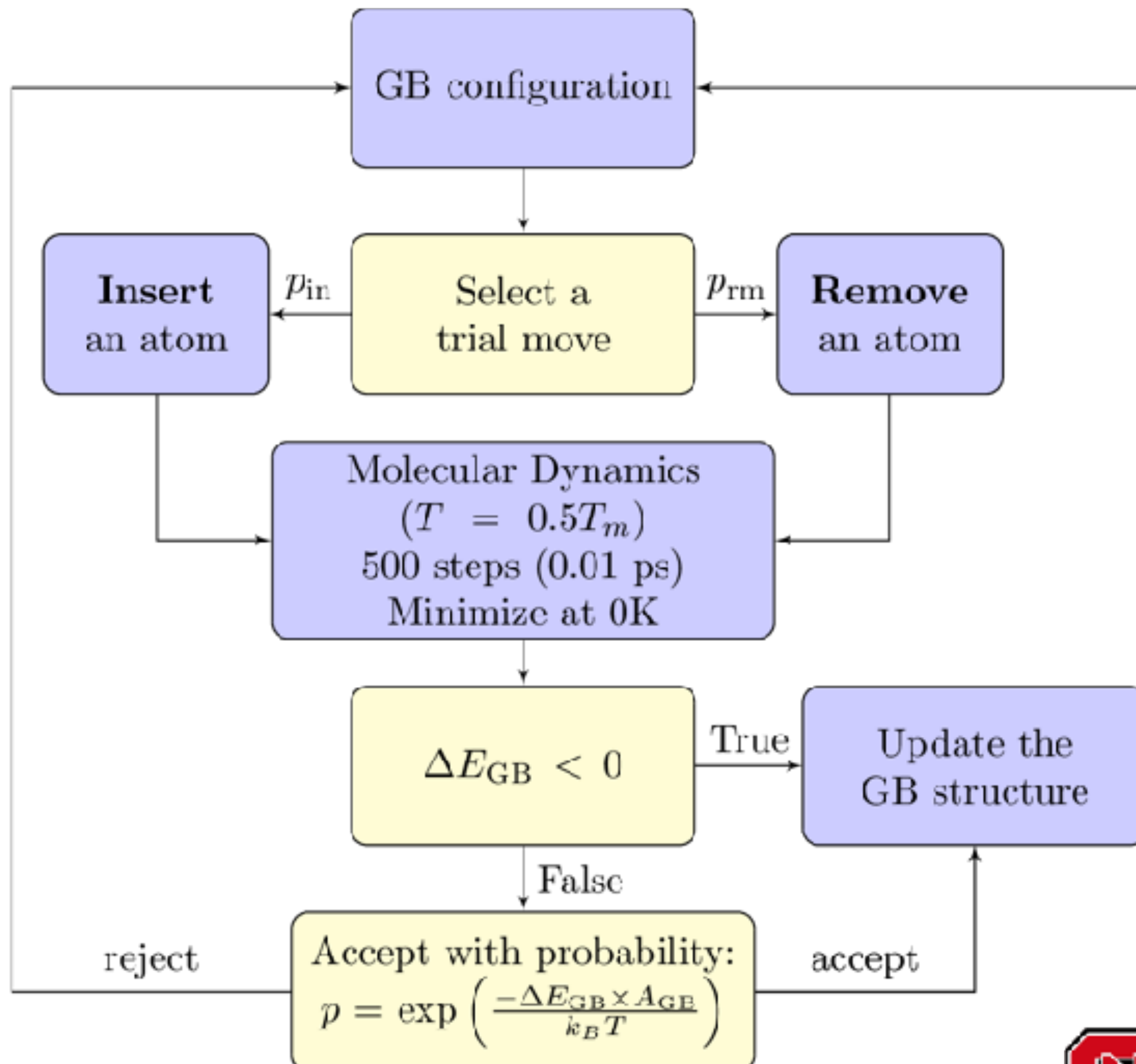
$$p_{\text{in},i} = \frac{r_{v,i}}{\sum_{j=1}^{N_v}(r_{v,j})}$$

[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

TRIAL MOVES - FUNDAMENTAL PERTURBATIONS

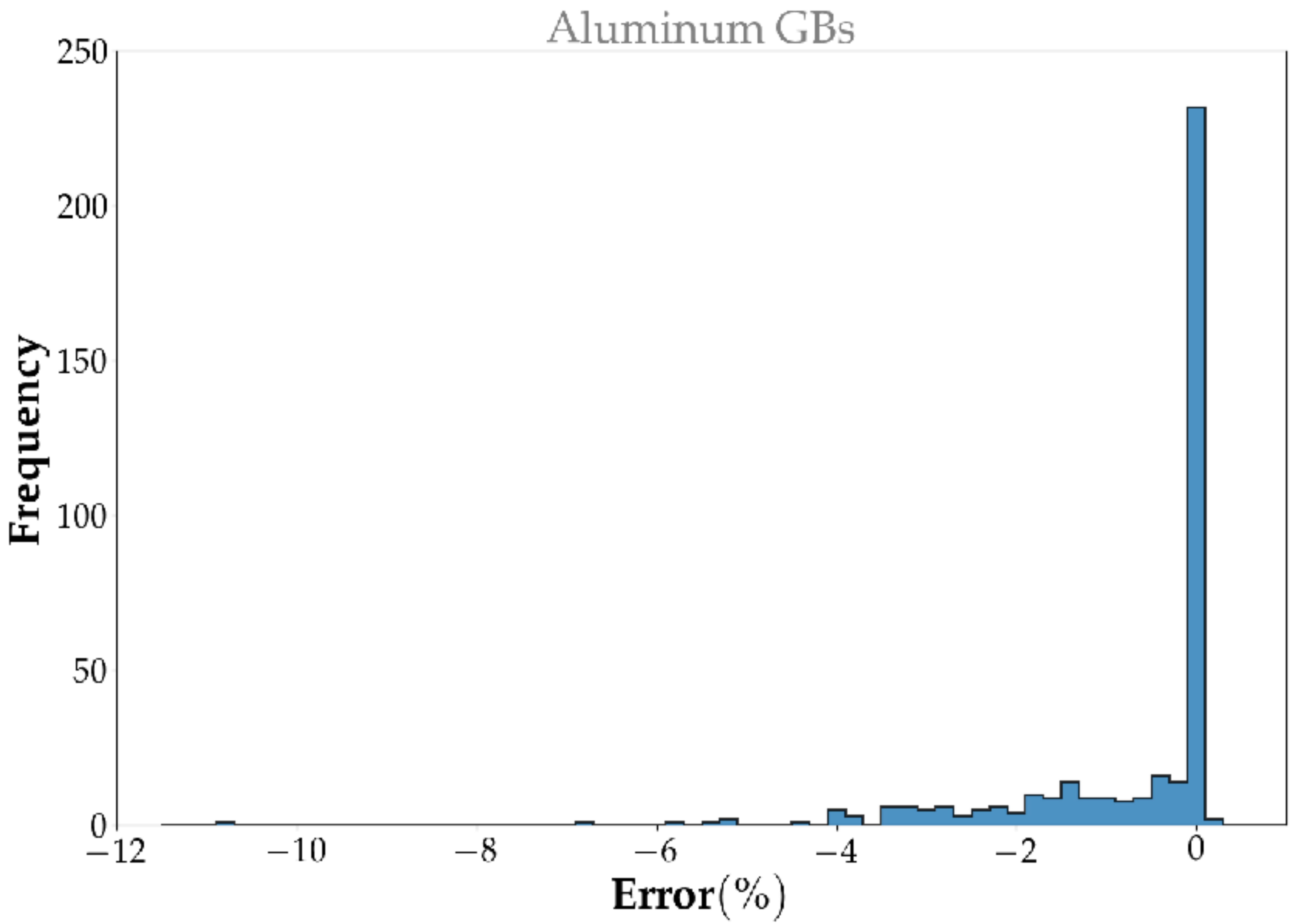
- Hybrid MC/MD Scheme
  - Monte Carlo
    - **Atom Removal**
    - **Atom Insertion**
  - Molecular Dynamics
    - Thermal vibrations





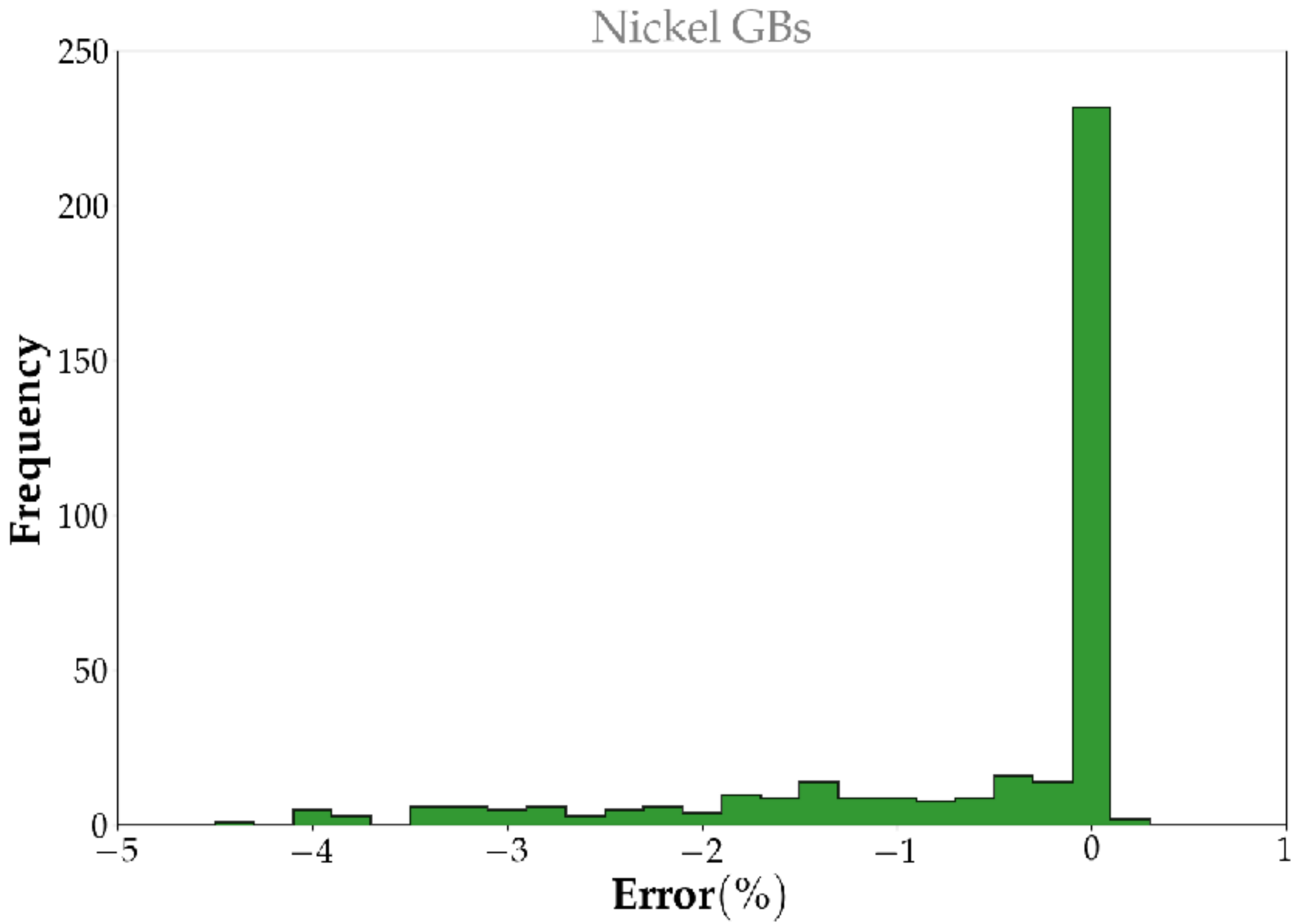
[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

HYBRID MC/MD ALGORITHM vs. BRUTE FORCE



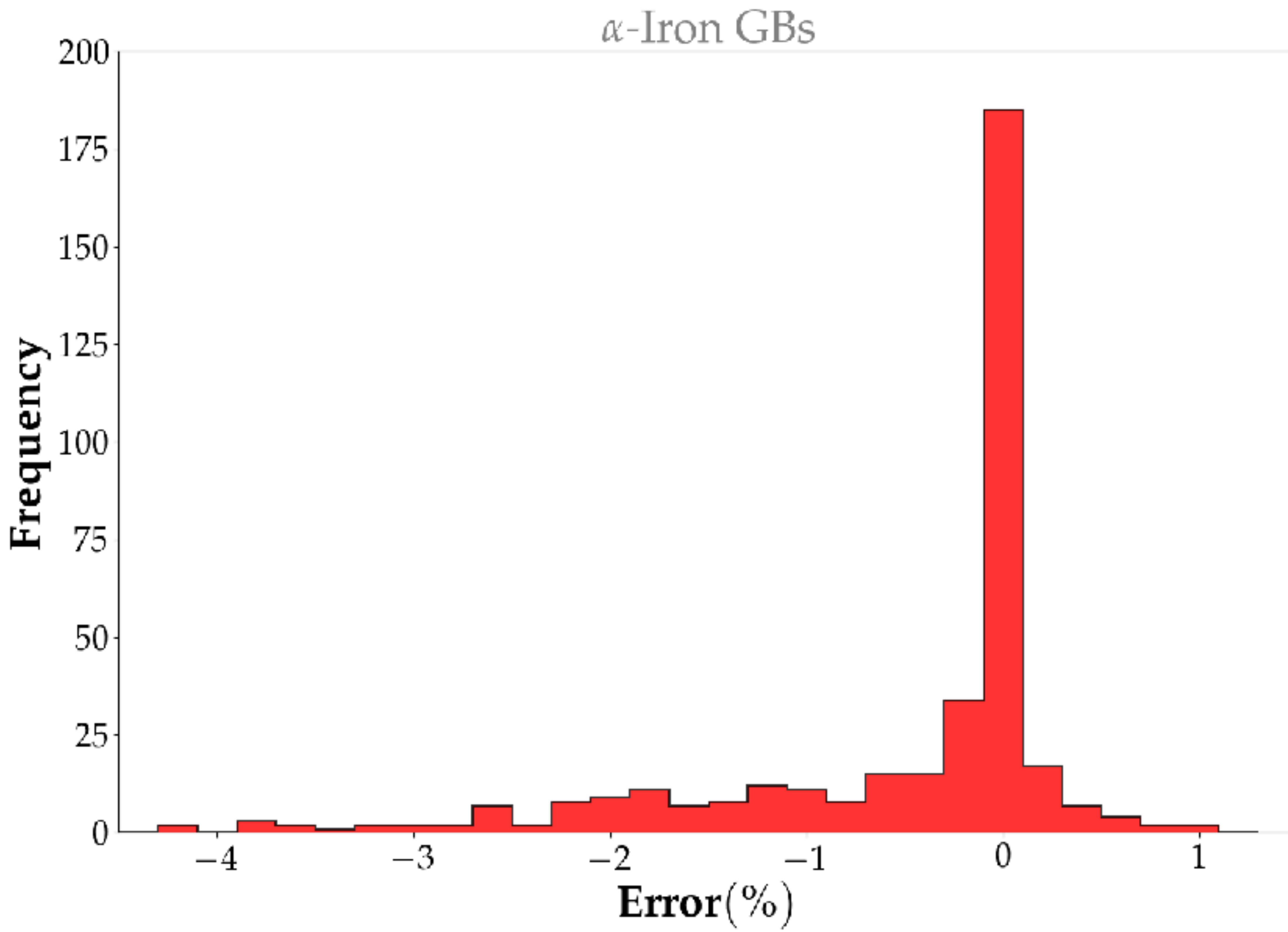
[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

HYBRID MC/MD ALGORITHM vs. BRUTE FORCE



[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

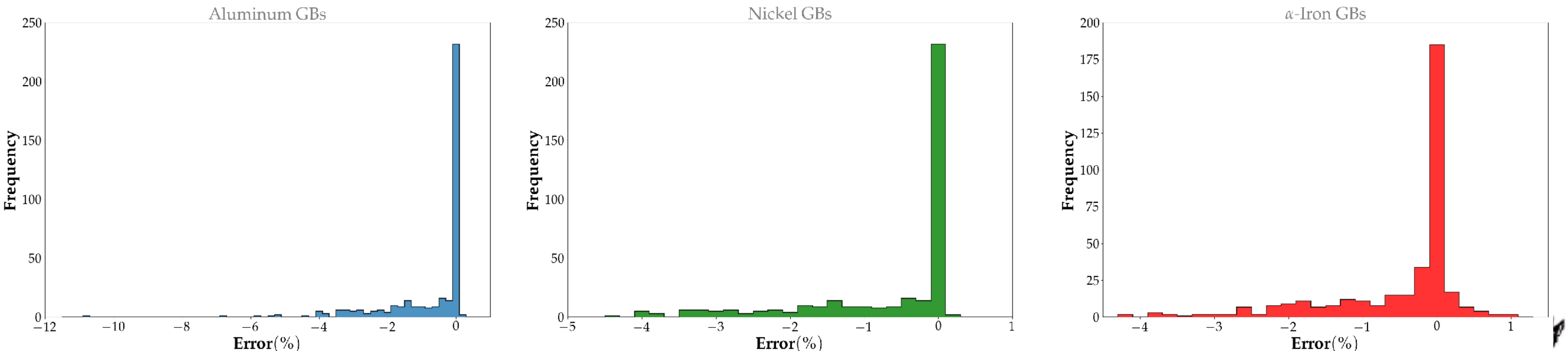
HYBRID MC/MD ALGORITHM vs. BRUTE FORCE



[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

HYBRID MC/MD GB GENERATOR

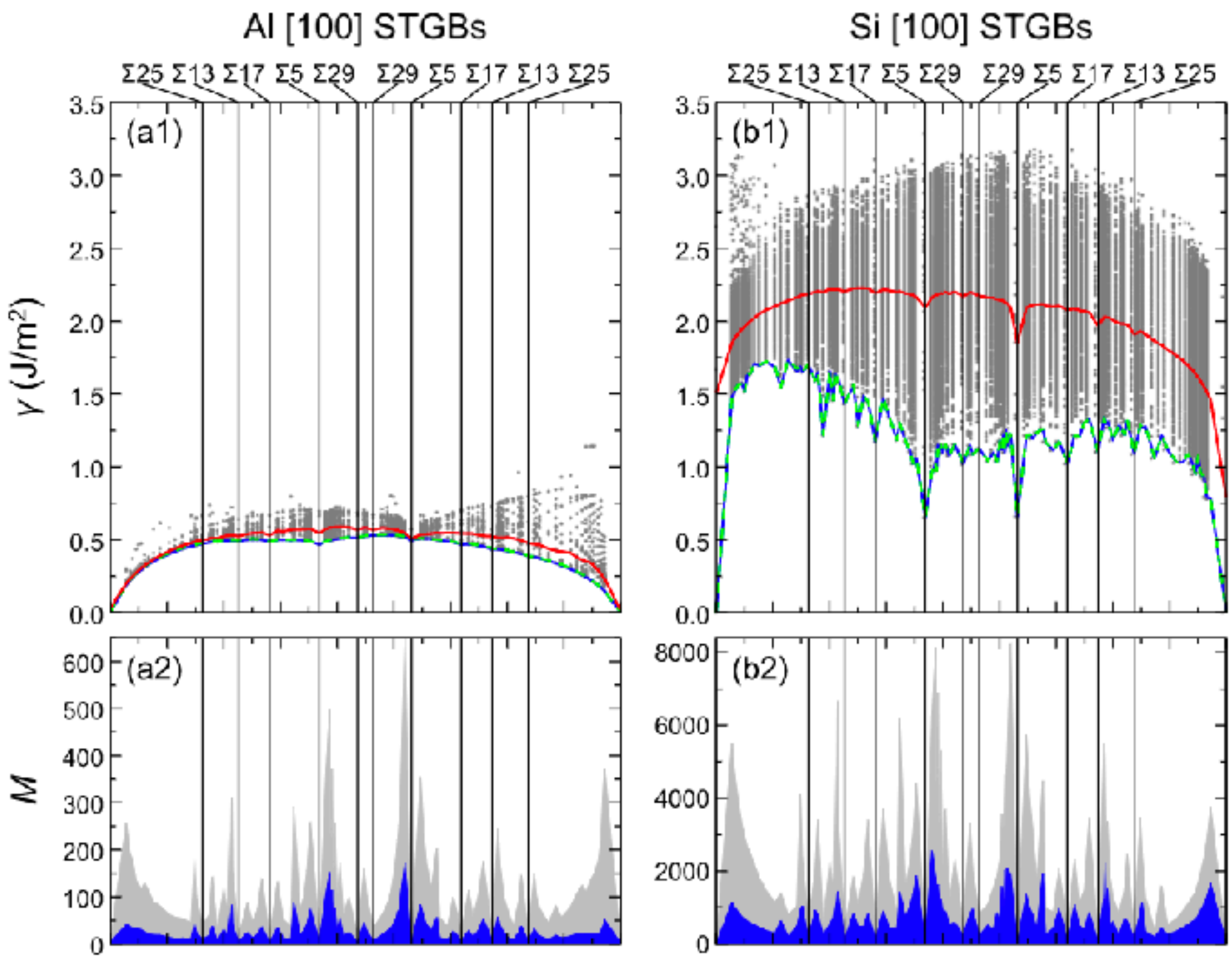
- "Convergence" to brute-force energies in less than 5000 energy minimizations.
- **1184** GBs of Fe, Ni and Aluminum
  - **5000 energy minimizations in MC scheme**
    - 500 MD steps between each MC step.
  - ~96% of the GBs have error less than 0.1%.
  - The maximum error is ~1% (BCC **α**-Iron).



[1] AD Banadaki, MA Tschopp, S. Patala. An Efficient Algorithm for Determining the Minimum Energy Structures of Grain Boundaries (Under Review)

UNIQUE CHALLENGES FOR GB STRUCTURES

- Metastability and GB Free energies



The results of conservative sampling for (a) Al [100] STGBs, and Si [100] STGBs.

The top panels show the γ-bands, i.e. the GB-energy spectra of all the GB states (black dots) for each misorientation.

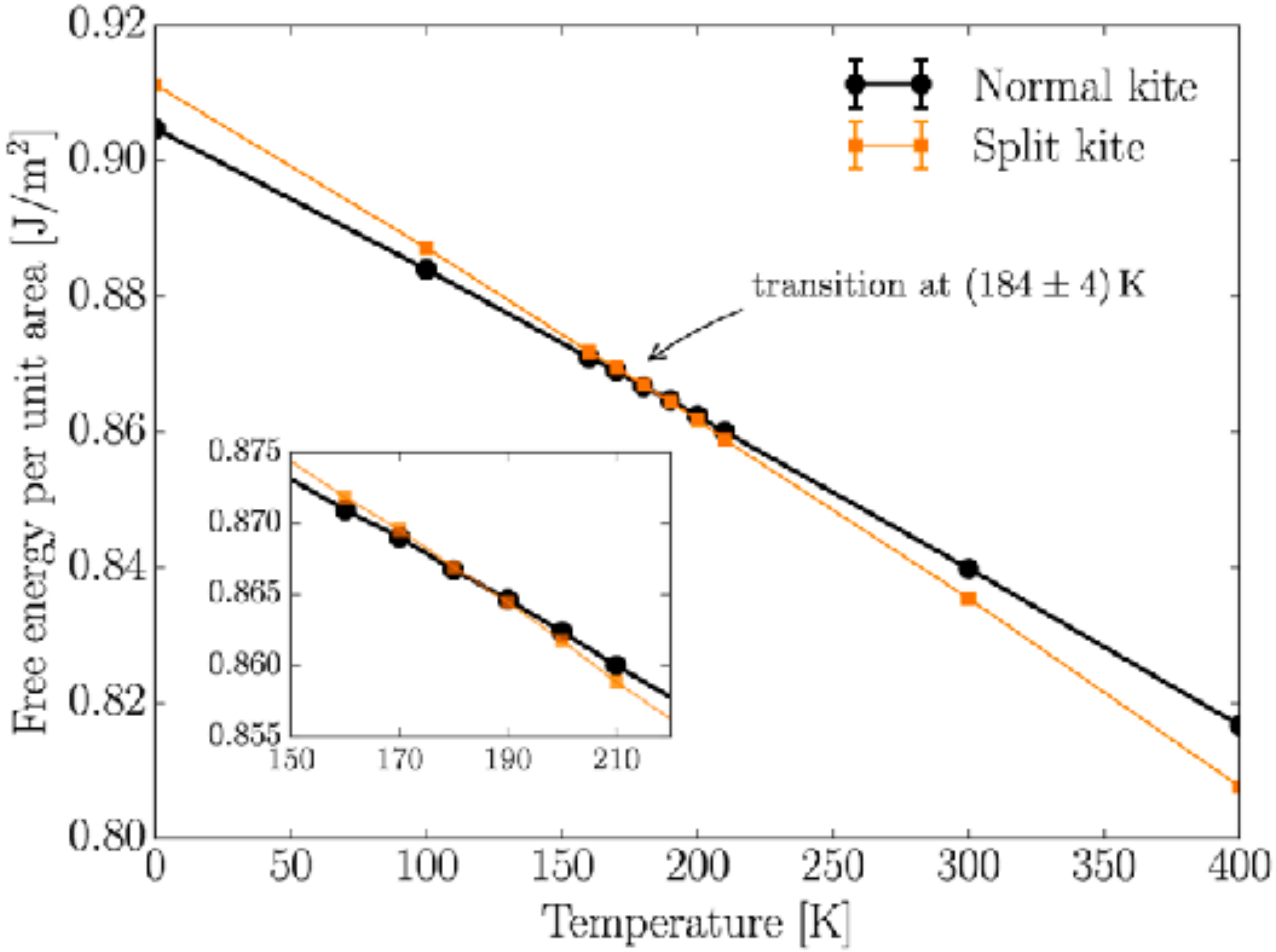
The blue solid, green dashed, and red solid lines represent the minimum GB energy γ_{min}, the equilibrium ensemble-averaged GB energy $\langle\gamma\rangle_{eq}$ (at half bulk melting point), and the nonequilibrium ensemble-averaged GB energy $\langle\gamma\rangle_{sq}$, respectively.

The panels of the second row show the number of GB states M that are shaded in gray, along with the number of states with distinct energies M_e that are shaded in blue.

Han, J., Vitek, V., & Srolovitz, D. J. (2016). Grain-boundary metastability and its statistical properties. *Acta Materialia*, *104*, 259-273.

UNIQUE CHALLENGES FOR GB STRUCTURES

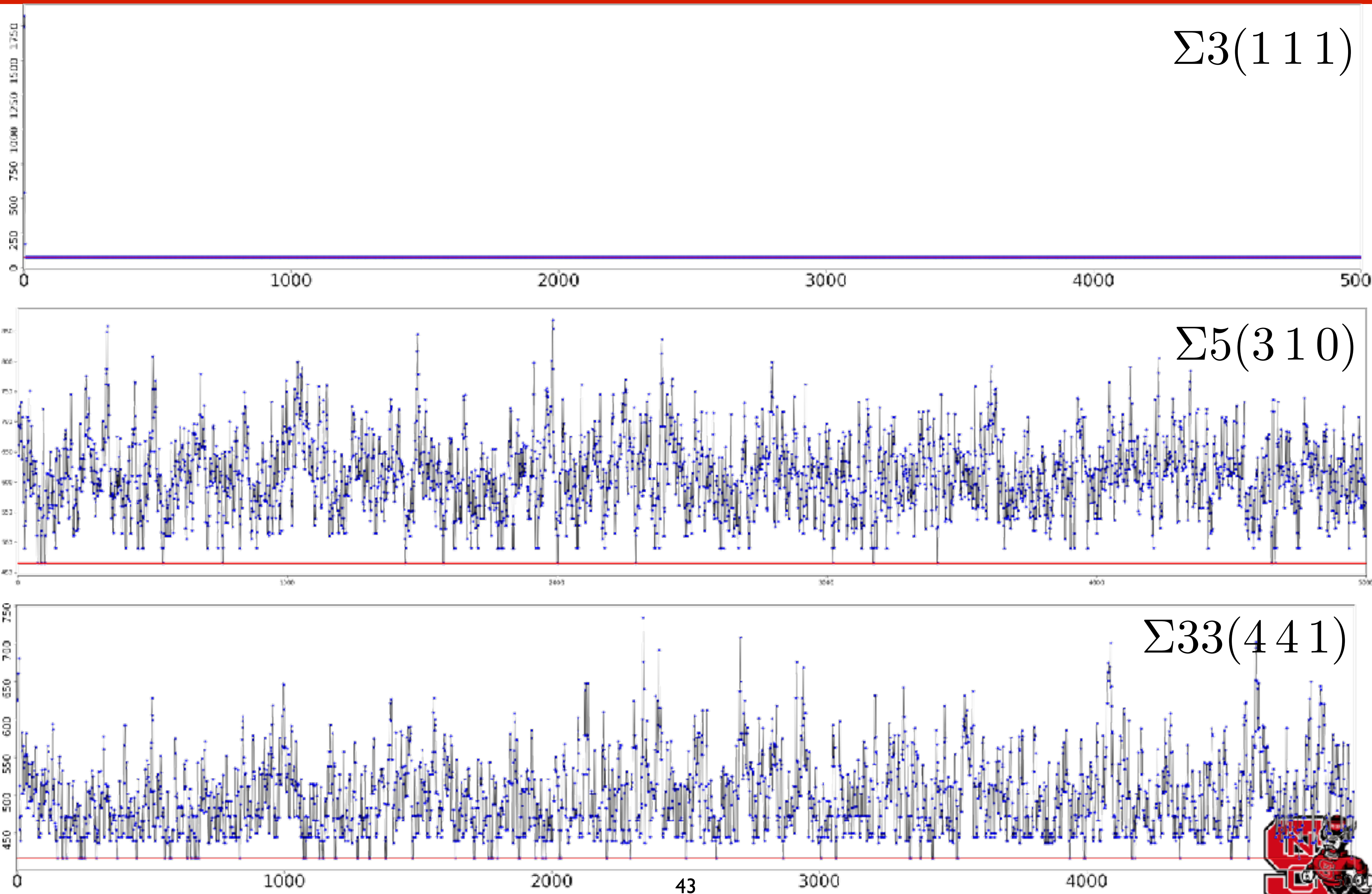
- Metastability and GB Free energies



Temperature dependence of the free energy of NK and SK phases as computed with the non-equilibrium Frenkel-Ladd method using the subsystem approach for Σ5(310) Copper GB [1].

Freitas, R., Rudd, R. E., Asta, M., & Frolov, T. (2018). Free energy of grain-boundary phases: Atomistic calculations for $\Sigma $5 (310)[001] grain boundary in Cu. arXiv preprint arXiv:1807.03274.

UNIQUE CHALLENGES FOR GB STRUCTURES



UNIQUE CHALLENGES FOR GB STRUCTURES

- Grand-Canonical Monte-Carlo Simulations
  - Different Ensembles (μVT, μPT)
    - How to apply appropriate boundary conditions?
    - How to satisfy detailed-balance equations?
  - Free-Energies
    - Phase-volumes of different metastable structures (Partition function and probabilities).
    - Are GBs more like single-crystals or glasses?