High Throughput Computational Framework of Materials Properties for Extreme Environments

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Team members

- **PI:** Zi-Kui Liu
  - In charge of the project
- **Co-PI**: Allison Beese
  - In charge of simulations based on finite element method (FEM)
- **Co-PI**: Shun-Li Shang
  - In charge of first-principles calculations based on density functional theory (DFT) and thermodynamic modeling based on the CALPHAD method
- **Senior Personnel**: Yi Wang
  - Development of DFTTK, calculations of thermodynamic properties
- **Graduate students**: Shipin Qin and John Shimanek
  - Perform both simulations and experiments

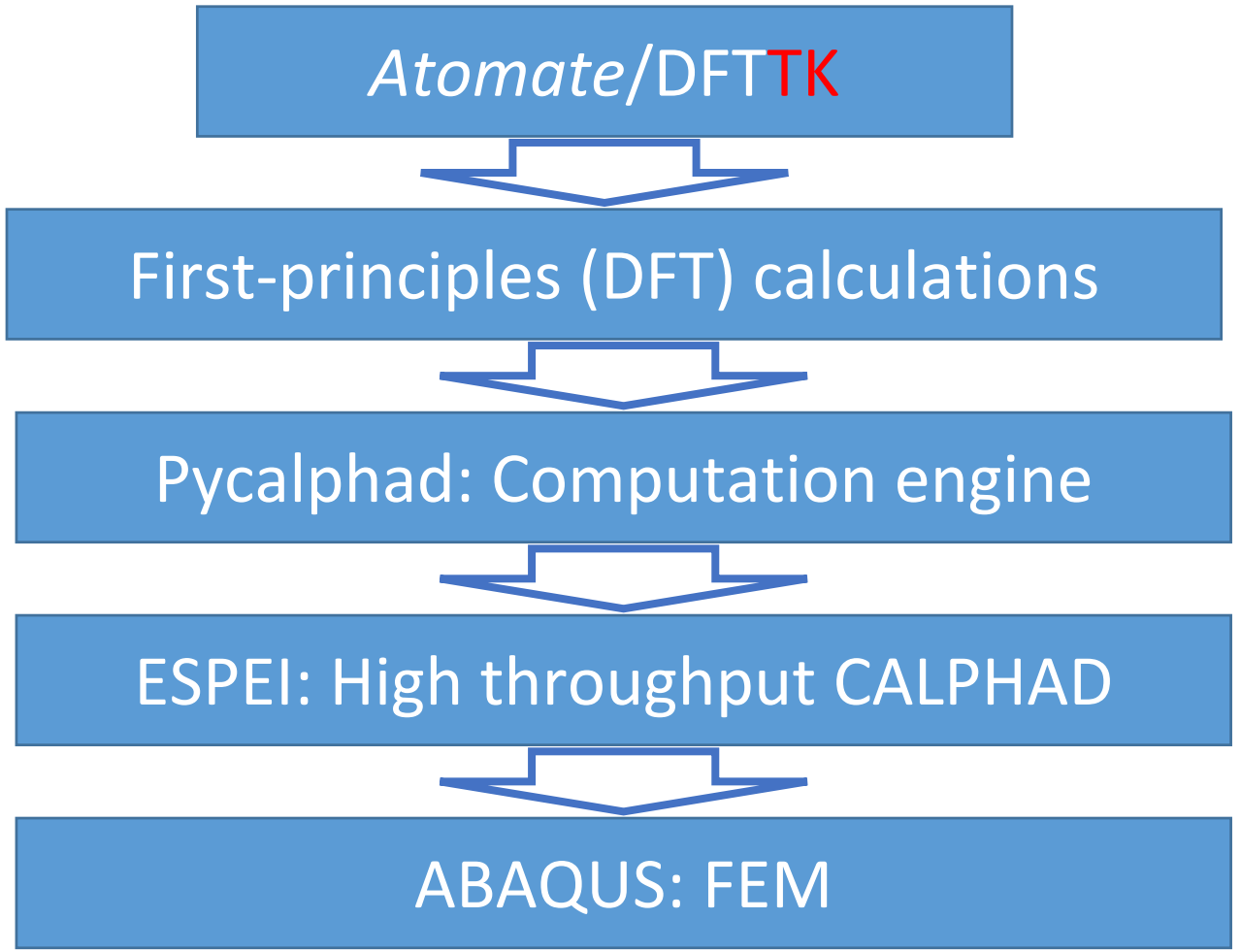
Goal ⇒ To establish a Python based open-source infrastructure with the following capabilities

- High throughput first-principles calculations for properties of interest, including such as phonon, thermodynamic, elastic, diffusion, vacancy formation, stacking and twin fault properties;
- High throughput CALPHAD modeling of the above properties using our unique capability based on *ESPEI* and *pycalphad;*
- New capabilities to predict the stress-strain behavior; and
- Phase-based new models for tensile strength prediction in common finite element method (FEM) analysis software.

Outline

- Overview of the proposed tasks
- Computational approaches
  - First-principles calculations based on density functional theory (DFT)
  - Thermodynamic modeling using the CALPHAD technique
  - Simulations of strain-stress curve using finite element method (FEM)
- Results and discussion
  - DFT results of Ni and Ni_3Al from pure shear deformation
  - FEM results of strain-stress curve for pure Ni single crystal
- Summary

Overview of high throughput computational framework for materials properties under extreme environments

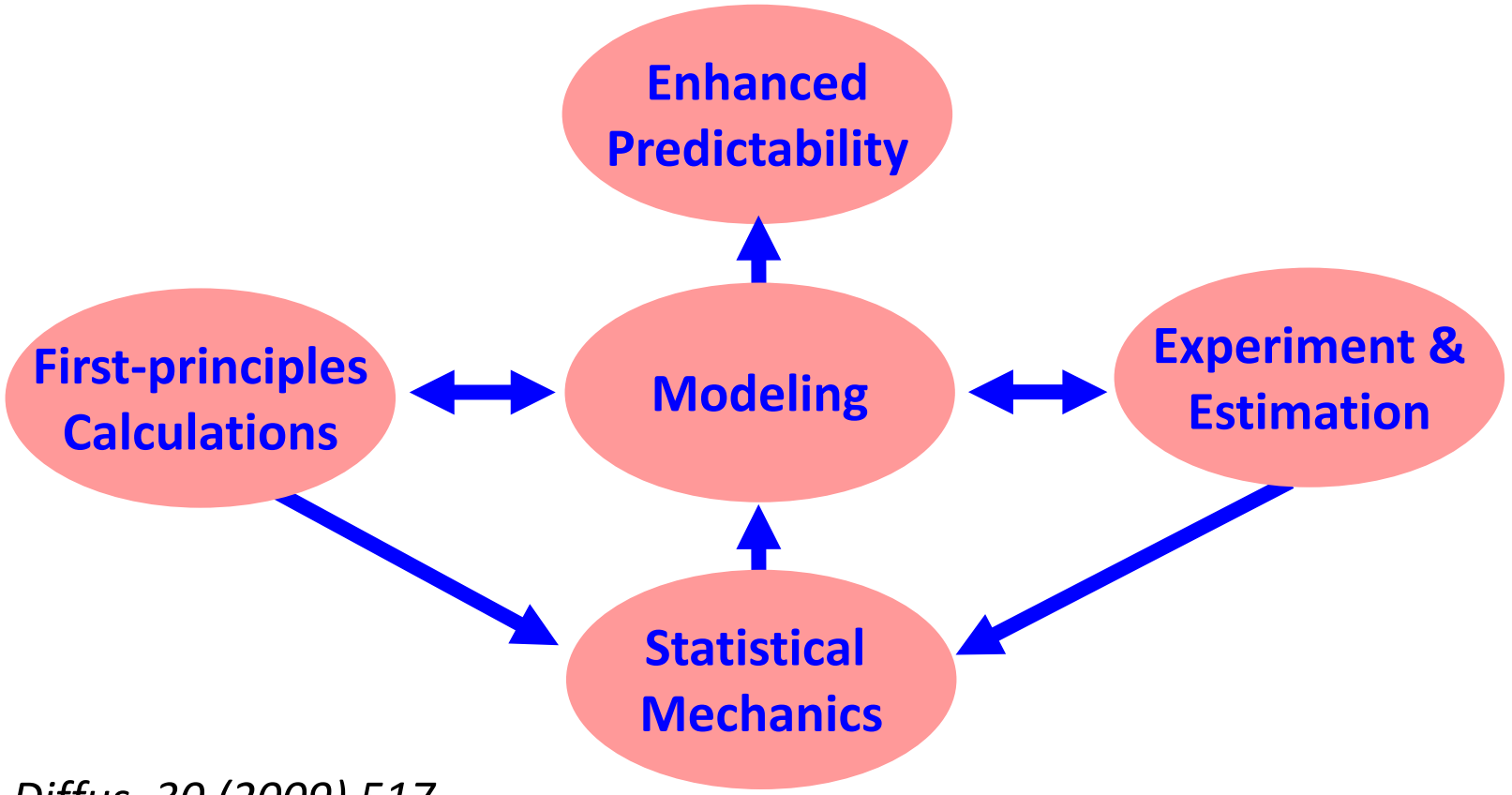


- ✓ Predict structure and thermodynamic properties using DFT and CALPHAD
- ✓ Use machine learning to reduce the amount of DFT calculations
- ✓ Apply FEM to simulate tensile strength
- ✓ Validate results and improve models

DFT, thermodynamic modeling, FEM and experiments

First-Principles Calculations and CALPHAD Modeling of Thermodynamics

Zi-Kui Liu



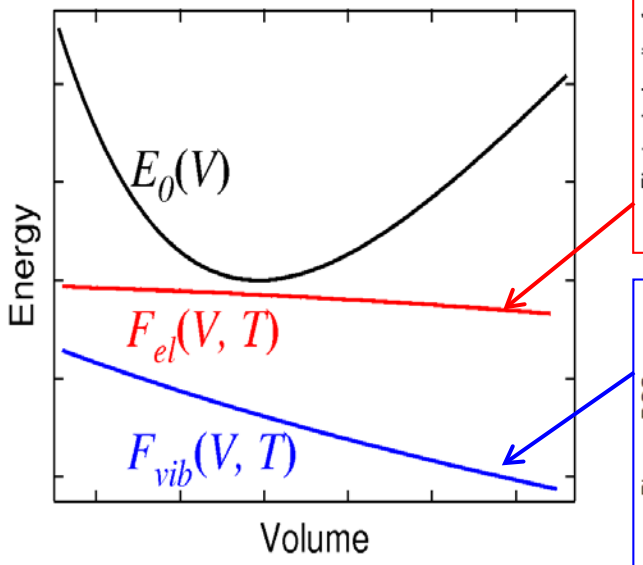
Liu, J. Phase Equilib. Diffus. 30 (2009) 517

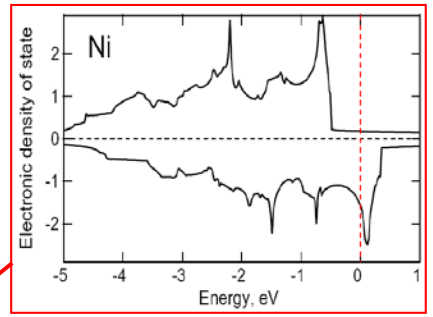
DFT calculations of thermodynamic properties

- **Quasi-Harmonic approach**

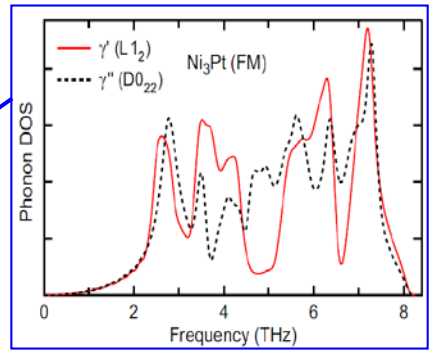
$$F(V,T) = E_0(V) + F_{vib}(V,T) + F_{el}(V,T)$$

- $E_0(V)$ Static energy at 0 K and volume V, i.e., EOS (by VASP)
- $F_{vib}(V,T)$ Vibrational contribution at V & T (Phonon or Debye model)
- $F_{el}(V,T)$ Thermal electronic contribution at V & T (by VASP)

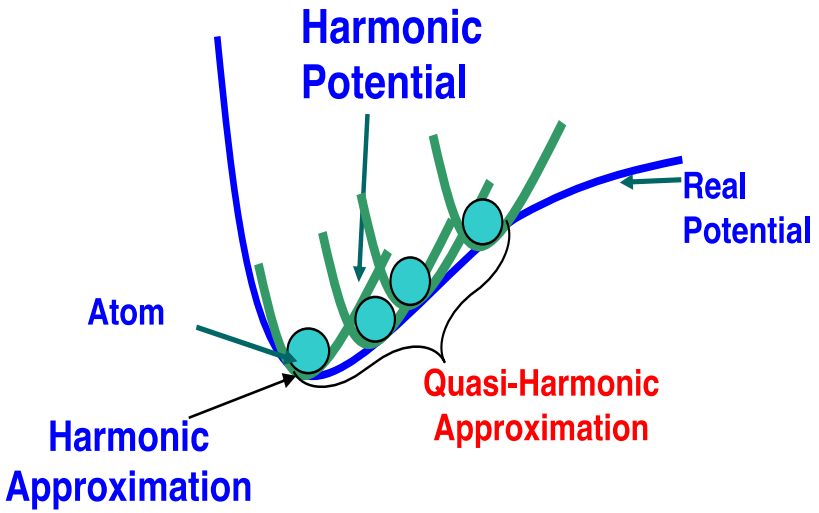




E-DOS



P-DOS



Acta Mater. 52 (2004), 2665, Comput. Mater. Sci. 47 (2010) 1040

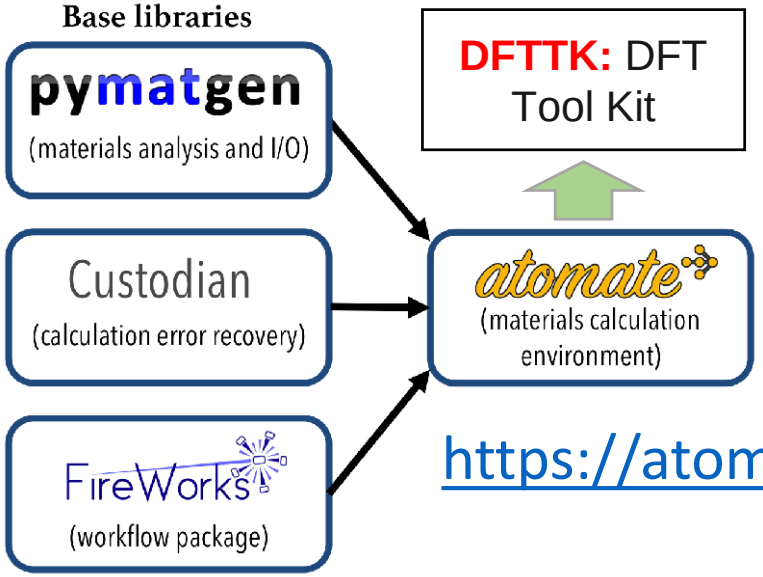
Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Atomate: A high-level interface to generate, execute, and analyze computational materials science workflows

- High throughput DFT calculations
  - Allows large scale calculations in an efficient way
  - Allows greater control and automation of DFT (via e.g. the VASP code)
  - Allows for the introduction of machine learning
- DFTTK: DFT calculations at high temperatures based on *atomate,* developed at Penn State

https://github.com/PhasesResearchLab/dfttk



https://atomate.org

CALPHAD modeling

Derivatives of Gibbs energy →
Thermochemical data: heat capacity, enthalpy, entropy, activity

$D_j d\xi_j = 0$ **→ Phase equilibrium data: phase stability, phase composition, phase boundary**

Gibbs Energy of Individual Phases
$G^{\phi}(T, P, N_i, \xi_j)$

Materials Design: Equilibrium, driving force, physical/chemical properties (1st, 2nd derivatives)

Pure elements → Binary → Ternary → Multicomponent

Kaufman & Bernstein: *Computer Calculation of Phase Diagram. 1970*

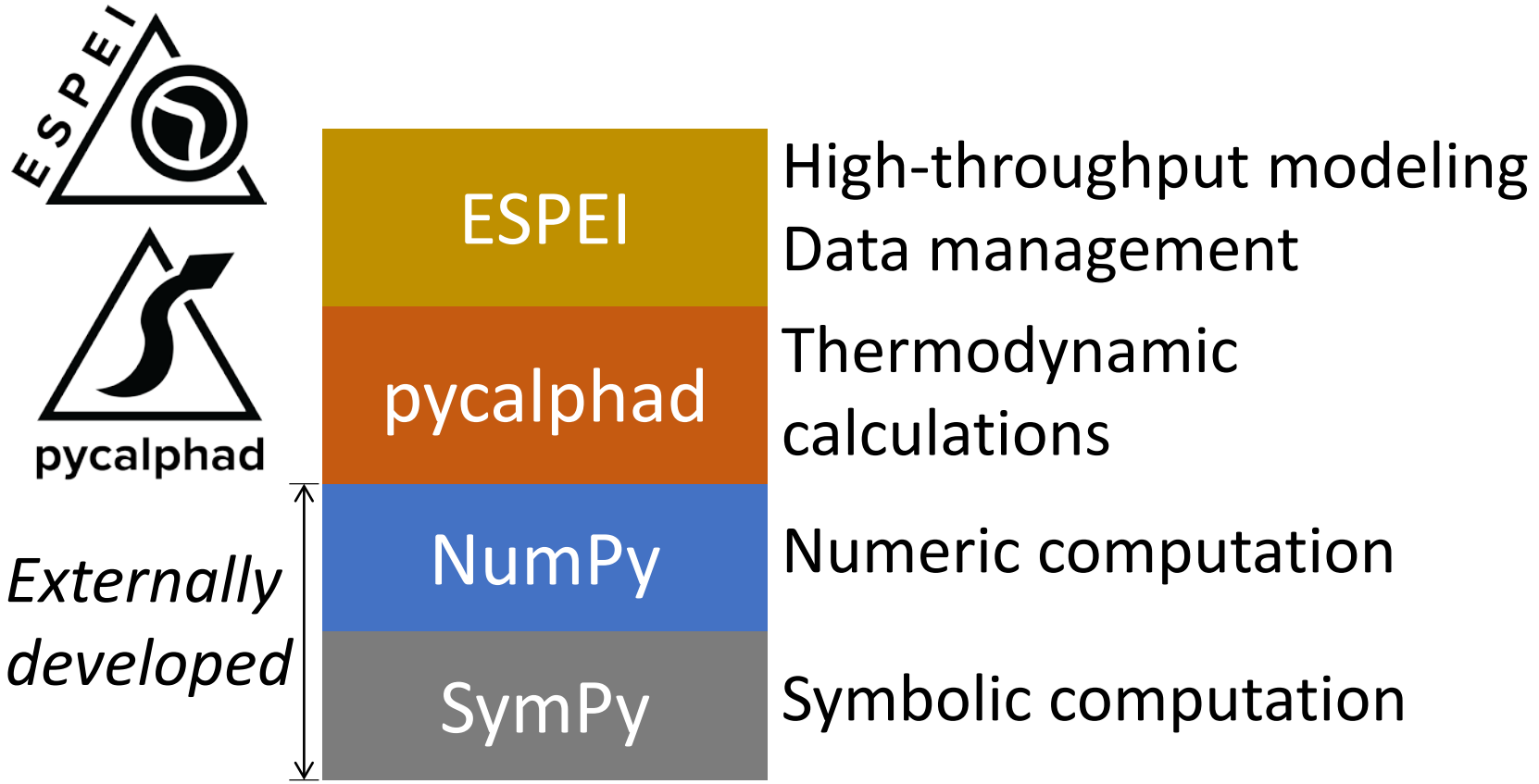
CALPHAD Community

- Commercial Tools: ThermoCalc, Factsage, ComputTherm, JMatPro
  - Special issues in CALPHAD journal: volume 26 (2002) and 33 (2009)

- Open Source Codes: OpenCALPHAD, pycalphad/ESPEI (our Python-based codes)

- CALPHAD Organization, http://www.calphad.org
  - CALPHAD Journal: published by Elsevier since 1975
  - CALPHAD, Inc.: Private foundation for scholarships and awards
  - Annual conference: Gordon conference style

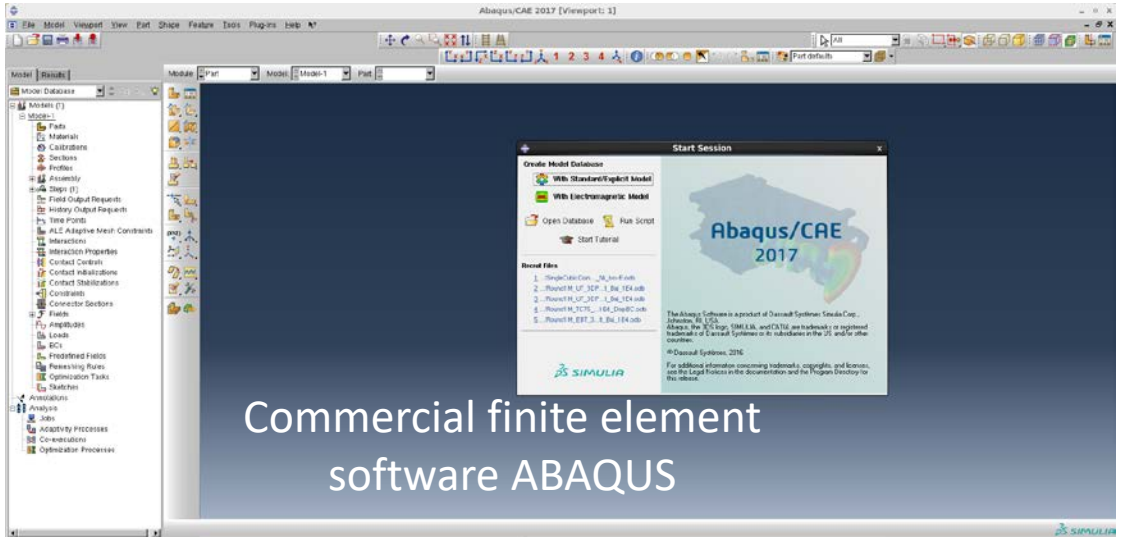
ESPEI.org/pycalphad.org Software Stack



- **Model flexibility**
- **Model parameter evaluation**
- **Uncertainty quantification**

JOM **69** (2017) 886–892

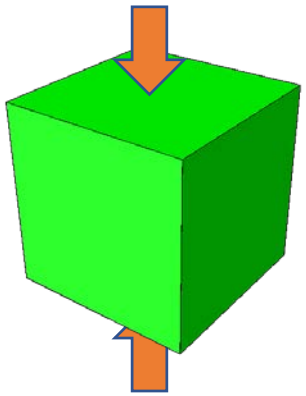
Finite Element Method (FEM)



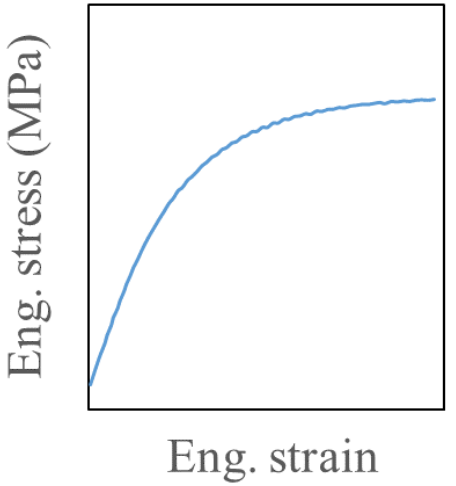
Commercial finite element software ABAQUS

```
      SUBROUTINE UMAT(stress,statev,ddsdde,sse,spd,scd,
     1 rpl, ddsddt, drplde, drpldt,
     2 stran,dstran,time,dtime,temp,dtemp,predef,dpred,cmname,
     3 ndi,nshr,ntens,nstatv,props,nprops,coords,drot,pnewdt,
     4 celent,dfgrd0,dfgrd1,noel,npt,layer,kspt,kstep,kinc)
c  WRITE (6,*) '
c  NOTE:  MODIFICATIONS TO *UMAT FOR ABAQUS VERSION 5.3 (14 APR '94)
c
c  (1)  The list of variables above defining the *UMAT subroutine,
c  and the first (standard) block of variables dimensioned below,
c  have variable names added compared to earlier ABAQUS versions.
```

Material model: a crystal plasticity model. Implemented through ABAQUS subroutine UMAT (User defined MATerial)



Single element finite element model for macroscopic material behavior prediction



Engineering stress strain curves in macroscopic mechanical tests

Huang, Yonggang. *Harvard Univ.*, 1991.

Crystal plasticity FEM (CPFEM) framework: **Phenomenological models and Physics/Dislocation-based models**

Phenomenological models

- Incorporate the concept of critical resolved shear stress (CRSS)
- Phenomenologically describe the evolution of CRSS with strain hardening and calibrate with 3 – 8 parameters

Physics/Dislocation-based models

- Explicitly describe the evolution of dislocation densities, based on flow stress.
- Contain ~ 20 parameters or more, challenge for calibrations

Roters, Franz et al. *Acta Materialia,* 58, no. 4 (2010): 1152-1211.

Crystal plasticity FEM (CPFEM) framework

Slipping rate in slip system α under a constant resolved shear stress τ^α

Change of slipping resistance in slip system α due to the slipping in all slip systems with n being the number of slip systems (12 for FCC)

$$\dot{\gamma}^\alpha = \dot{\gamma}_0 \left|\frac{\tau^\alpha}{\tau_c^\alpha}\right|^{\frac{1}{m}} sgn(\tau^\alpha)$$

$$\dot{\tau}_c^\alpha = \sum_{\beta=1}^{n} h_{\alpha\beta} \left|\dot{\gamma}^\beta\right|$$

$$h_{\alpha\beta} = q_{\alpha\beta} h_0 \operatorname{sech}^2 \left|\frac{h_0 \gamma}{\tau_s - \tau_0}\right|$$

$$q_{\alpha\beta} = \begin{cases} 1, & \alpha = \beta \\ 1.4, & \alpha \neq \beta \end{cases}$$

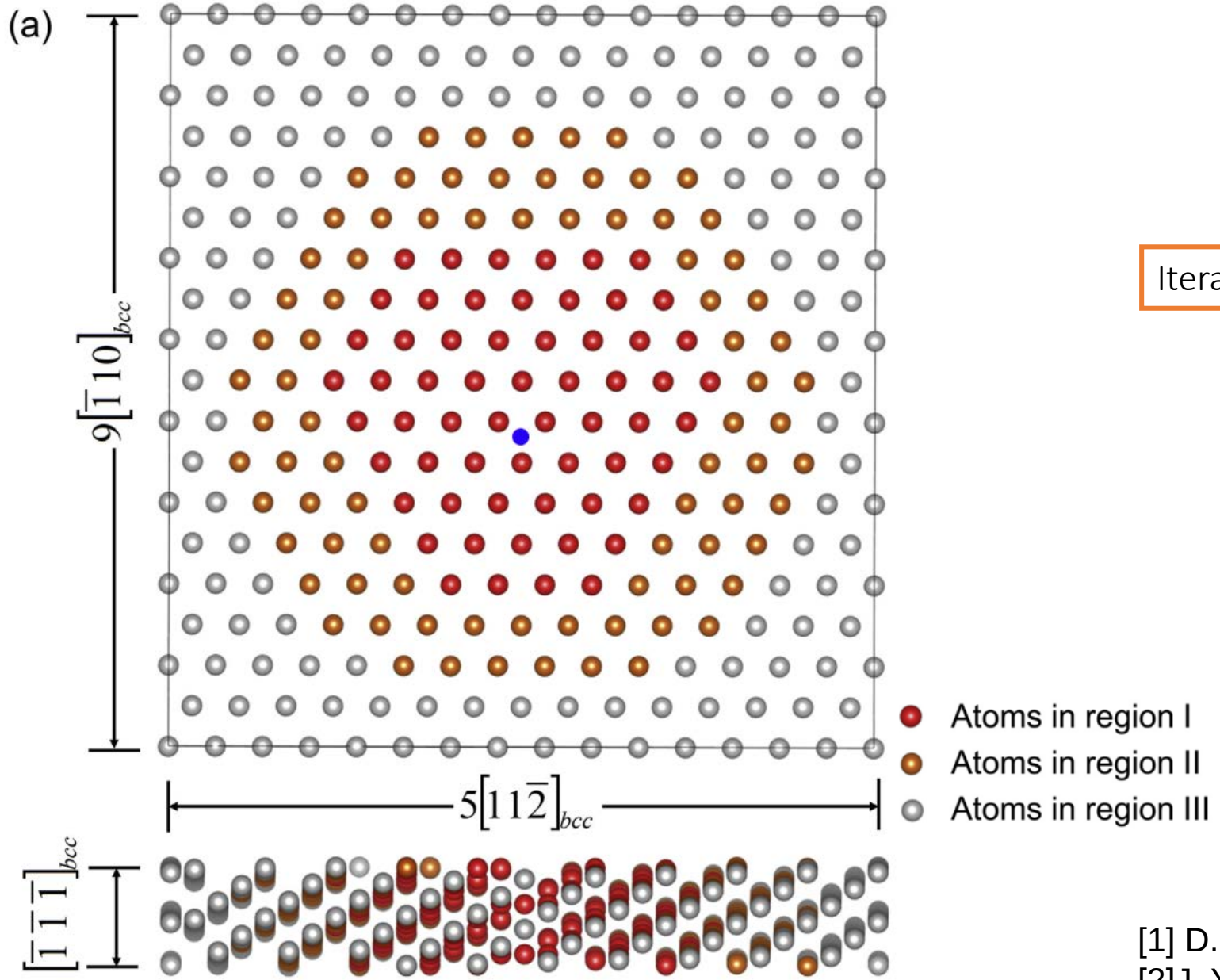
The shear stress (τ) and shear strain (γ) relationship on slip systems

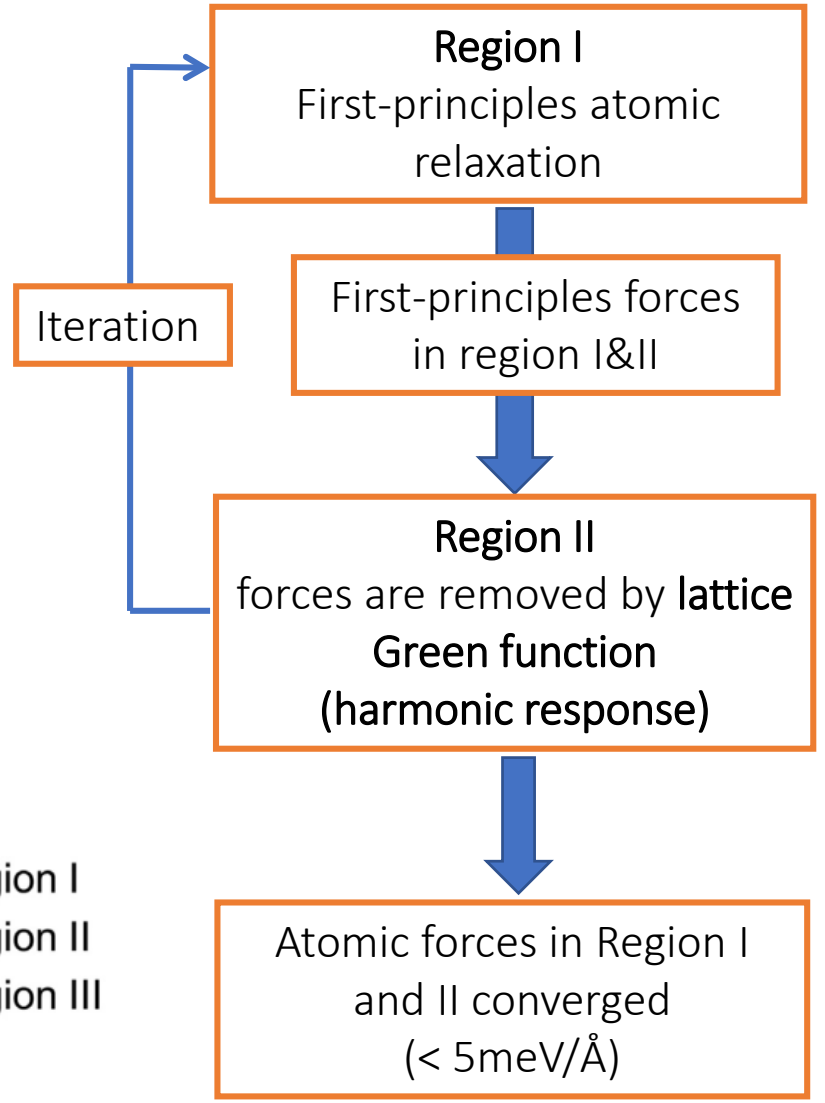
Determined from DFT calculations in the present study

$\dot{\gamma}_0$ – Reference slipping rate (usually $0.001\ s^{-1}$)
τ^α – Applied resolved shear stress on slip system α
τ_c^α – Current strength of slip system α
m – hardening exponent (usually 0.02 ~ 0.05)
h_0, τ_s – hardening parameters
τ_0 – CRSS

Roters, Franz et al. *Acta Materialia,* 58, no. 4 (2010): 1152-1211.
Huang, Yonggang. *Harvard Univ.*, 1991.

Flexible boundary condition method



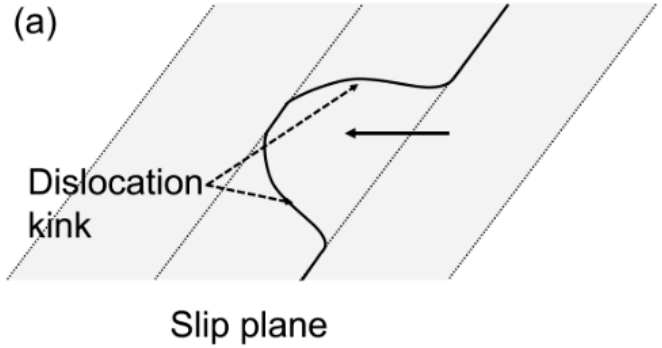


[1] D.R. Trinkle, *Phys. Rev. B*, (2008)
[2]J. Yasi, et al., *Phys. Rev. E*, (2012)

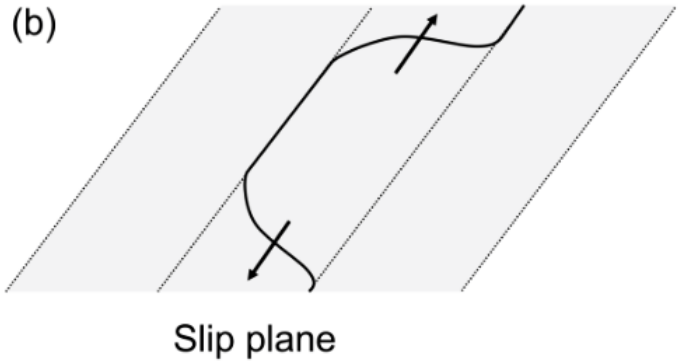
Solid-solution plasticity model: Acta Mat. 141 (2017) 304

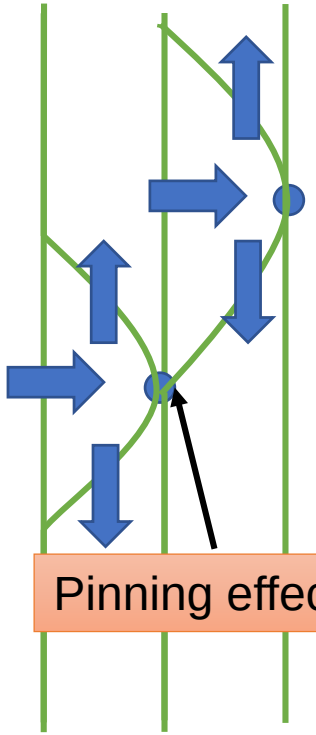
Motion of ½<111>{110} screw dislocation

Step I: Double-kink nucleation



Step II: Kink migration





Pinning effects of solute

Double-kink nucleation

Kink migration

Thermally activated dislocation motion

Nucleation Rate	Migration Rate
$\nu_{nucl.} \approx \nu_{dk} \exp\left(-\frac{\Delta H_{nucl.}(\sigma)}{k_B T}\right)$	$\nu_{mig.} \approx \nu_{km} \exp\left(-\frac{\Delta H_{mig.}(\sigma)}{k_B T}\right)$

$$\Delta H(\sigma) = \Delta H^0 [1 - (\frac{\sigma}{\tau^0})^p]^q$$

Solute-dislocation interaction

E_{int}
Solute-dislocation
Interaction energy

$\tau' = \tau^{-1} d\tau / dc$
Peierls misfit

A. S. Argon, *Strengthening Mechanisms in Crystal Plasticity* (2006)

D. Trinkle, et al., *Science*, 2005

Peierls-Nabarro (P-N) model & Peierls stress (σ_p)

- P-N model: to describe dislocation
  - A hybrid atomistic-continuum approach, to bridge atomistic and continuum description of dislocations
  - For example of Peierls stress σ_p (wide dislocation):

σ_p or CRSS at 0 K $\quad \sigma_p = \frac{Kb}{a'}\exp(-2\pi\zeta/a')$

b: Burgers vector; *a'*: atomic planes spacing

Half width of dislocation $\quad \zeta = \frac{Kb}{4\pi\tau_{max}}$

τ_{max}: ideal shear strength

Energy coefficient for isotropic polycrystal $\quad K = \mu\left(\frac{\sin^2\theta}{1-\nu} + \cos^2\theta\right)$

μ: shear modulus; ν: Poisson's ratio
θ: angle; 0° for screw & 90° edge dislocations

Energy coefficient for anisotropic single crystal, for example: $\quad K_{e_x} = (\bar{c}'_{11} + c'_{12})\left[\frac{c'_{66}(\bar{c}'_{11} - c'_{12})}{(\bar{c}'_{11} + c'_{12} + 2c'_{66})c'_{22}}\right]^{1/2}$

c_{ij}': Translated elastic constants with "ex" of edge dislocation along x-direction

Joos and Duesbery *PRL* 78 (1997) 266
Hirth and Lothe, Theory of dislocation, 1982.

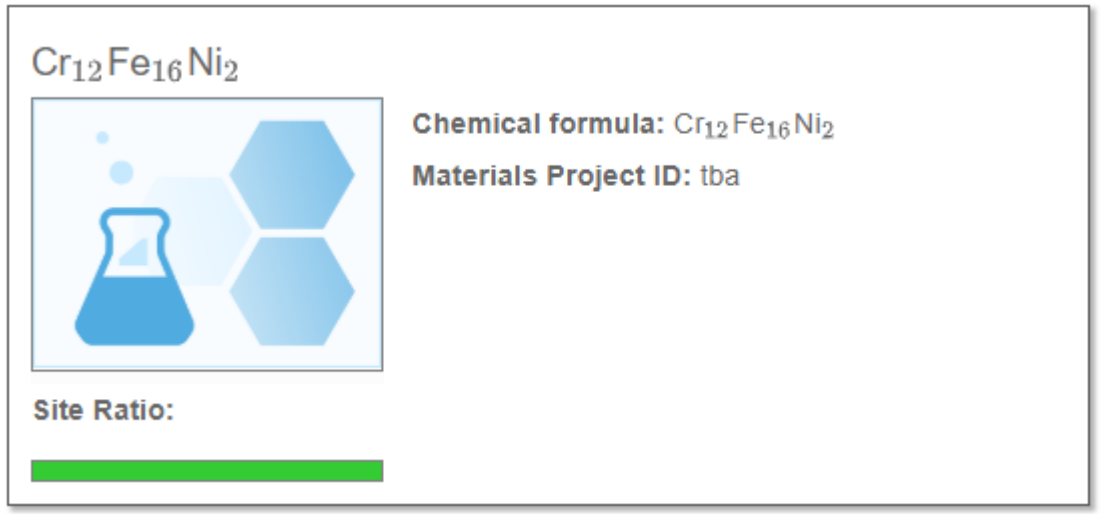
Two key input parameters:
Elastic properties and ideal shear strength

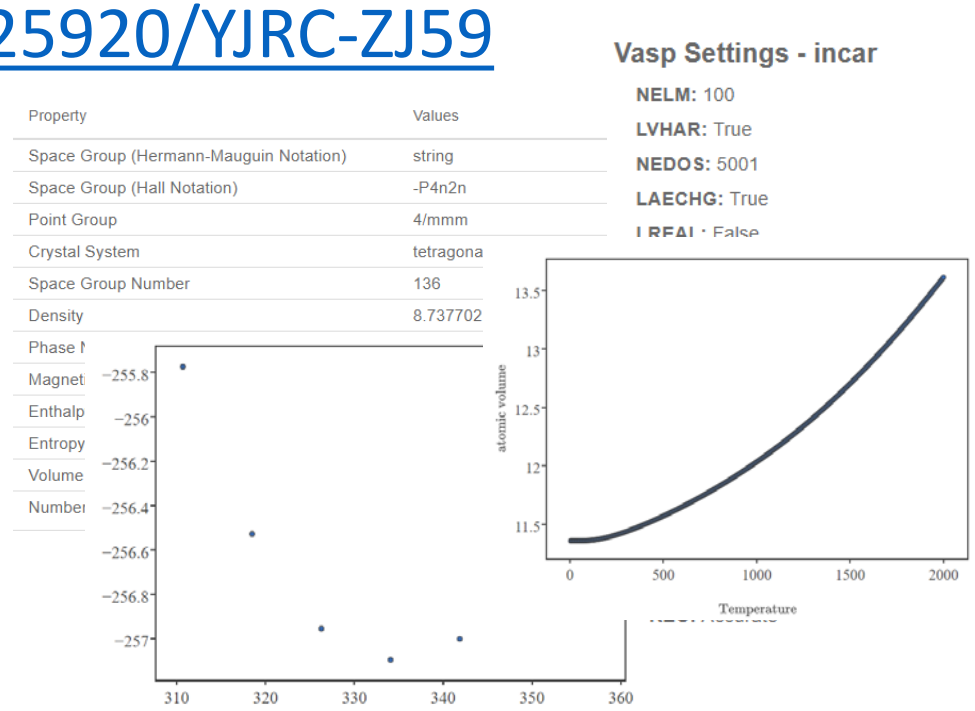
Data Repositories

- DFT-based first-principles calculations
  - Materials Project, http://materialsproject.org/
  - OQMD: An Open Quantum Materials Database, http://oqmd.org
  - AFLOW: Automatic Flow for Materials Discovery, http://www.aflowlib.org
  - In the process of establishing a data repository of free energy
- NIST
  - Materials Data Repository, https://mgi.nist.gov/materials-data-repository
  - Phase-Based Data Repository, https://phasedata.nist.gov/
  - Thermodynamic Research Center, http://www.trc.nist.gov
- Citrine Informatics, https://citrine.io

Citrine Dataset Example: Cr-Fe Sigma Phase

- 192 endmembers with the remaining ~50 calculations to be uploaded soon
- Contains phase information and finite temperature properties
- Citrine Informatics. https://doi.org/10.25920/YJRC-ZJ59

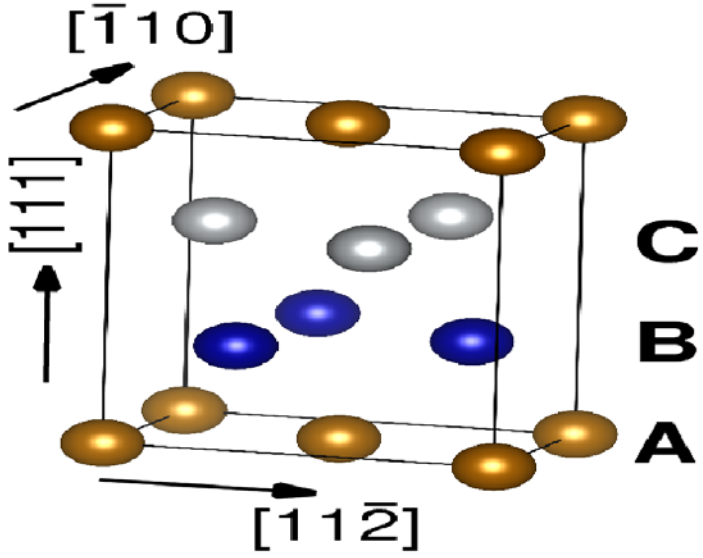




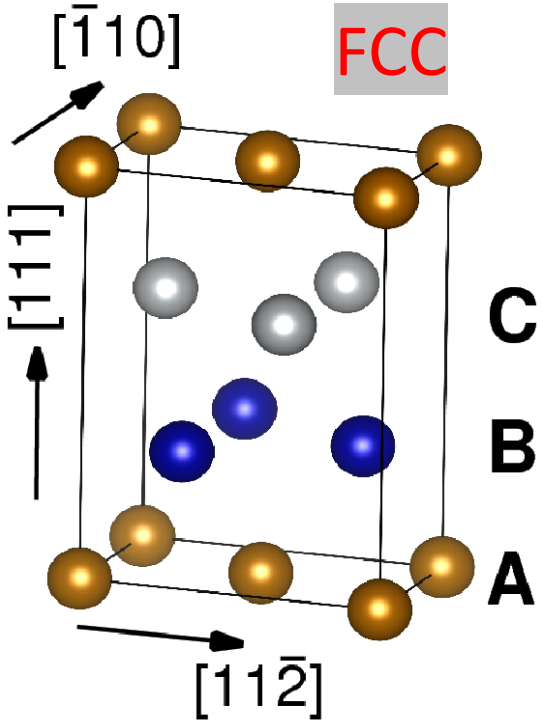
Case study of Ni and Ni_3Al: From DFT, FEM to mechanical properties

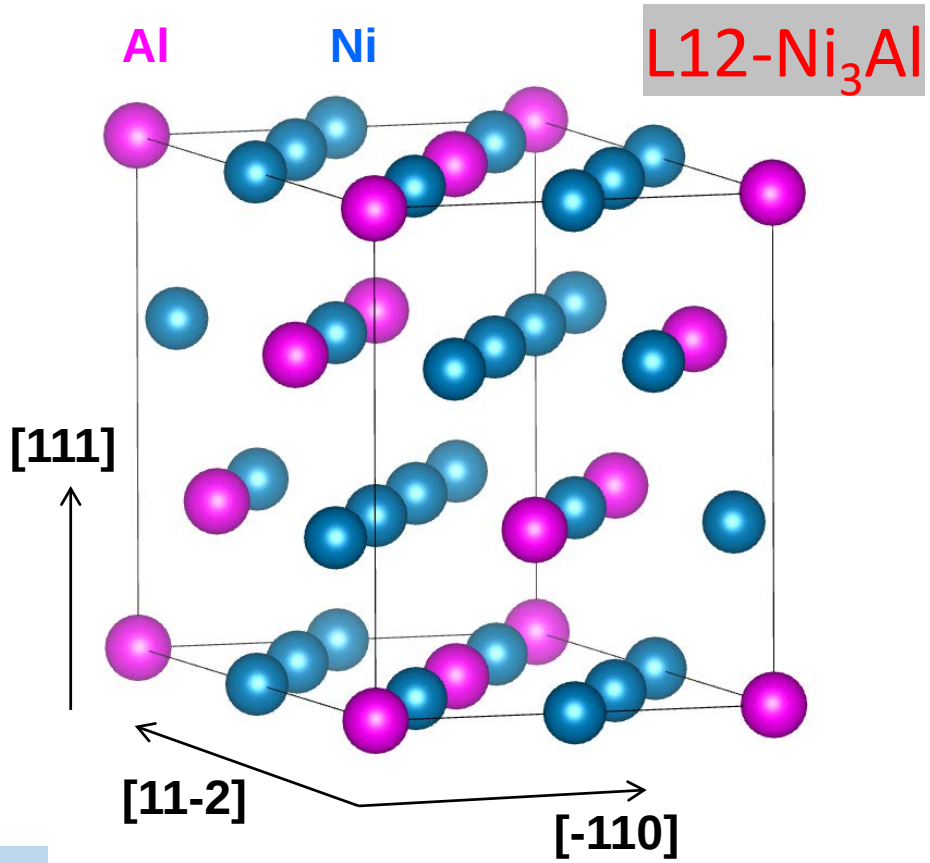
DFT calculations of mechanical properties by shear deformation

- **Calculations of ideal shear stress τ_{IS}, stacking fault energies (γ_{SF} , γ_{USF}), CRSS using the Peierls-Nabarro model**



Shear deformation





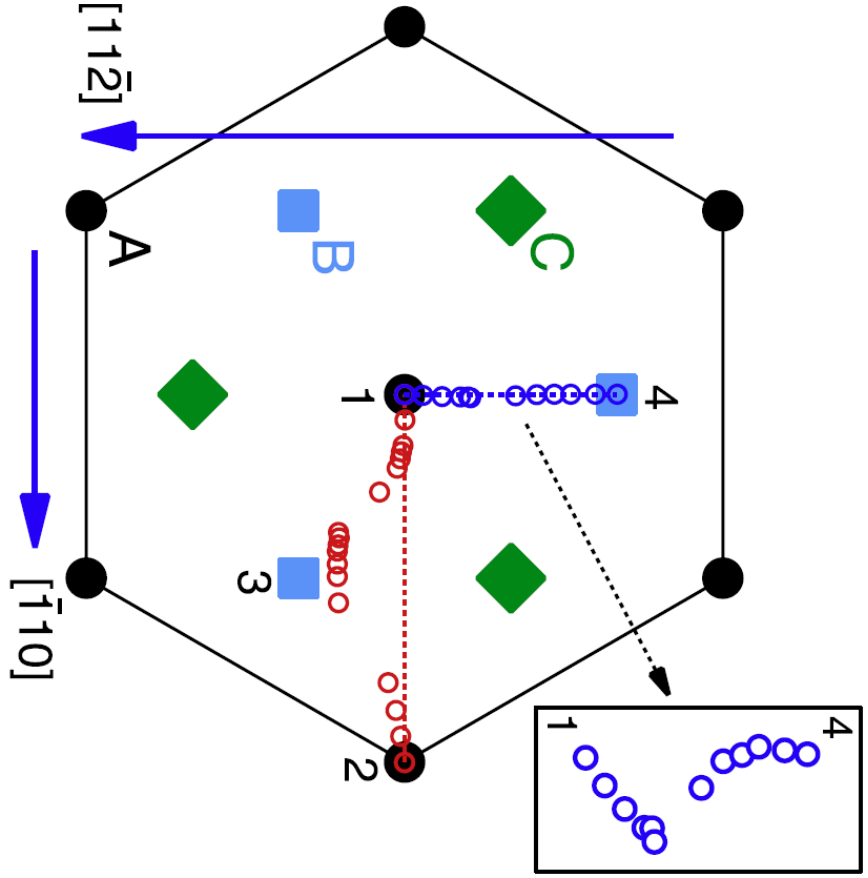
n=1: Alias shear deformation (close to real case), we use this one
n=∞: Affine shear deformation (far from real case)

J. Phys.: Condens. Matter 24 (2012) 155402

Dislocation decomposition from pure shear deformation

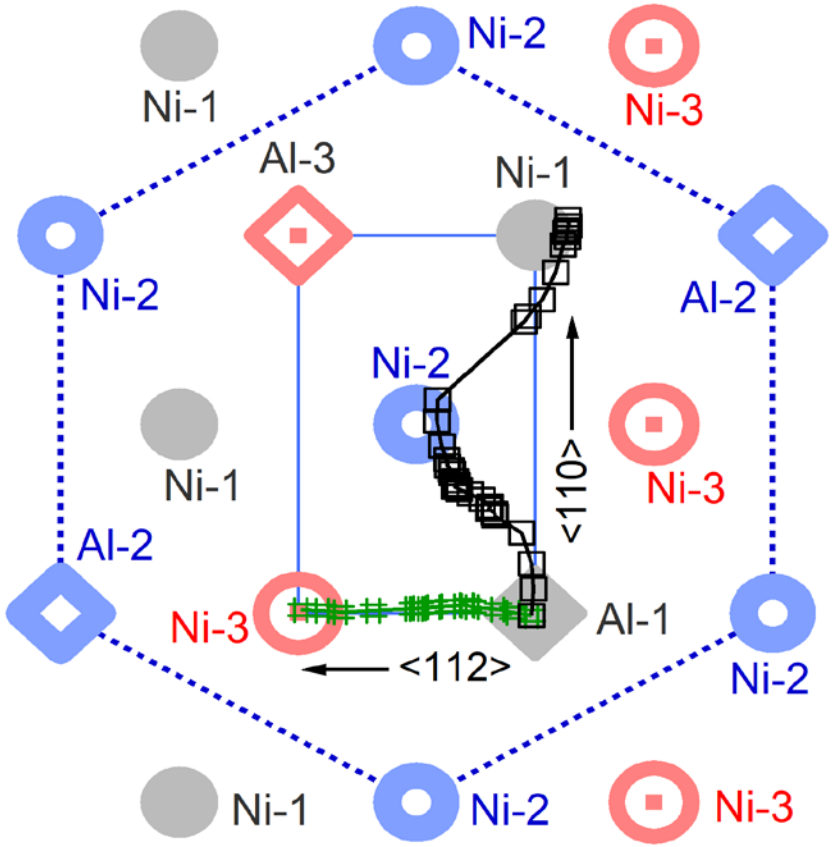
{111}<110> shear of Ni and Ni_3Al along the <112> directions

fcc Ni



<110>/2 → <121>/6 + SF + <211>/6

$L1_2$ Ni_3Al

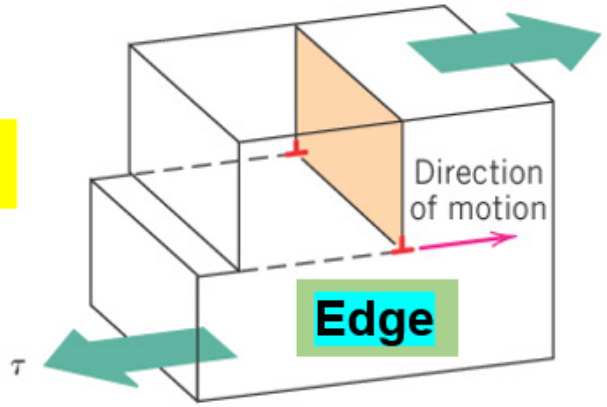


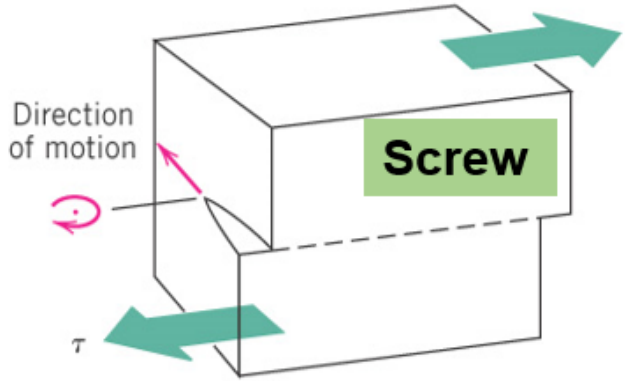
<110>/2 → <121>/6 + CSF + <211>/6

Dislocation decomposition from pure shear deformation

{111}<110> shear of Ni and Ni_3Al along the <112> directions

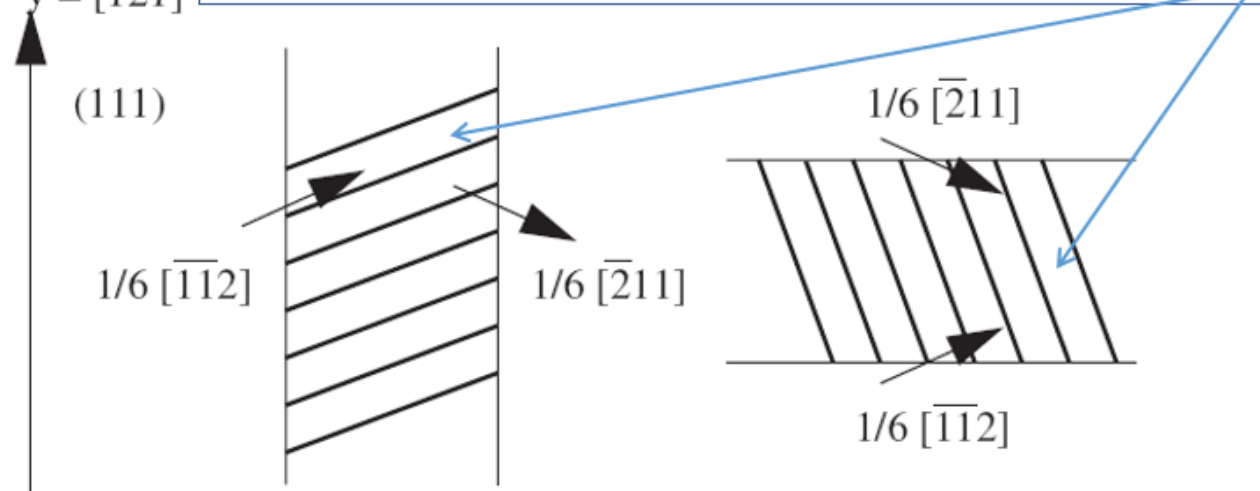
Summary





Edge

$$\frac{1}{2}[\bar{1}01] \rightarrow \frac{1}{6}[\bar{1}\bar{1}2] + \frac{1}{6}[\bar{2}11] +$$ **(complex/) stacking fault (C/SF)**

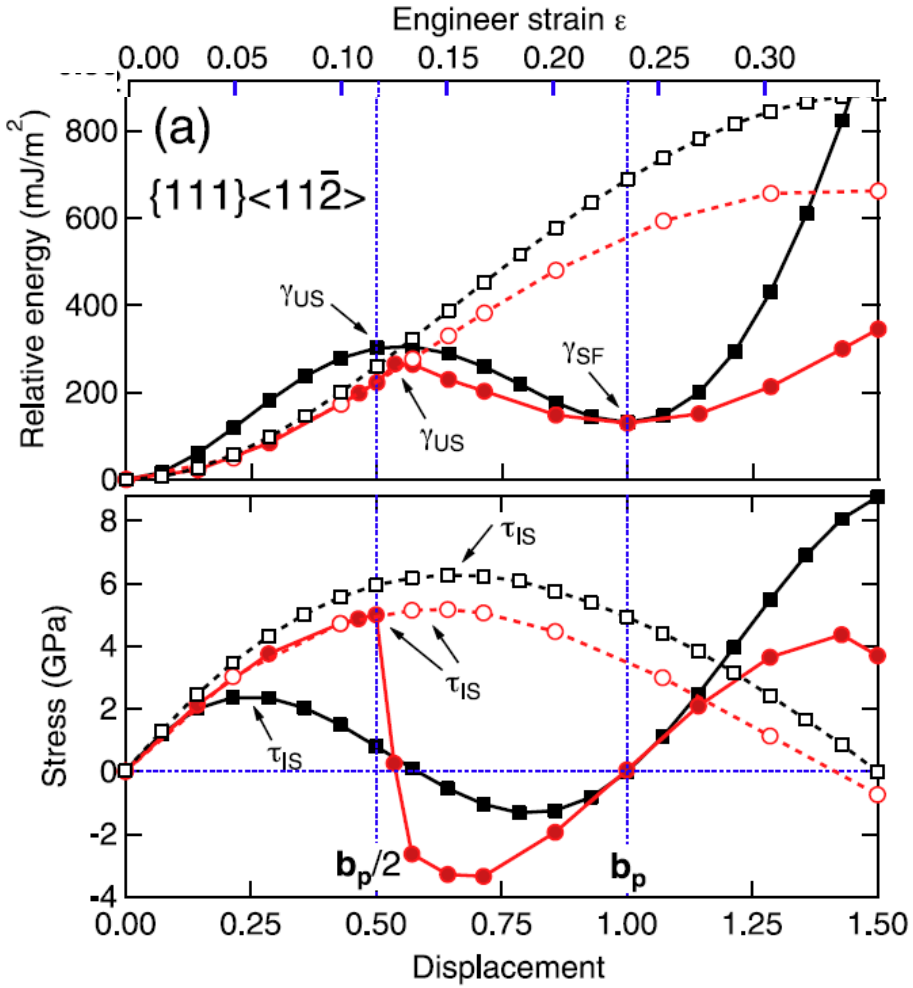


MSMSE 11, 2003, 883

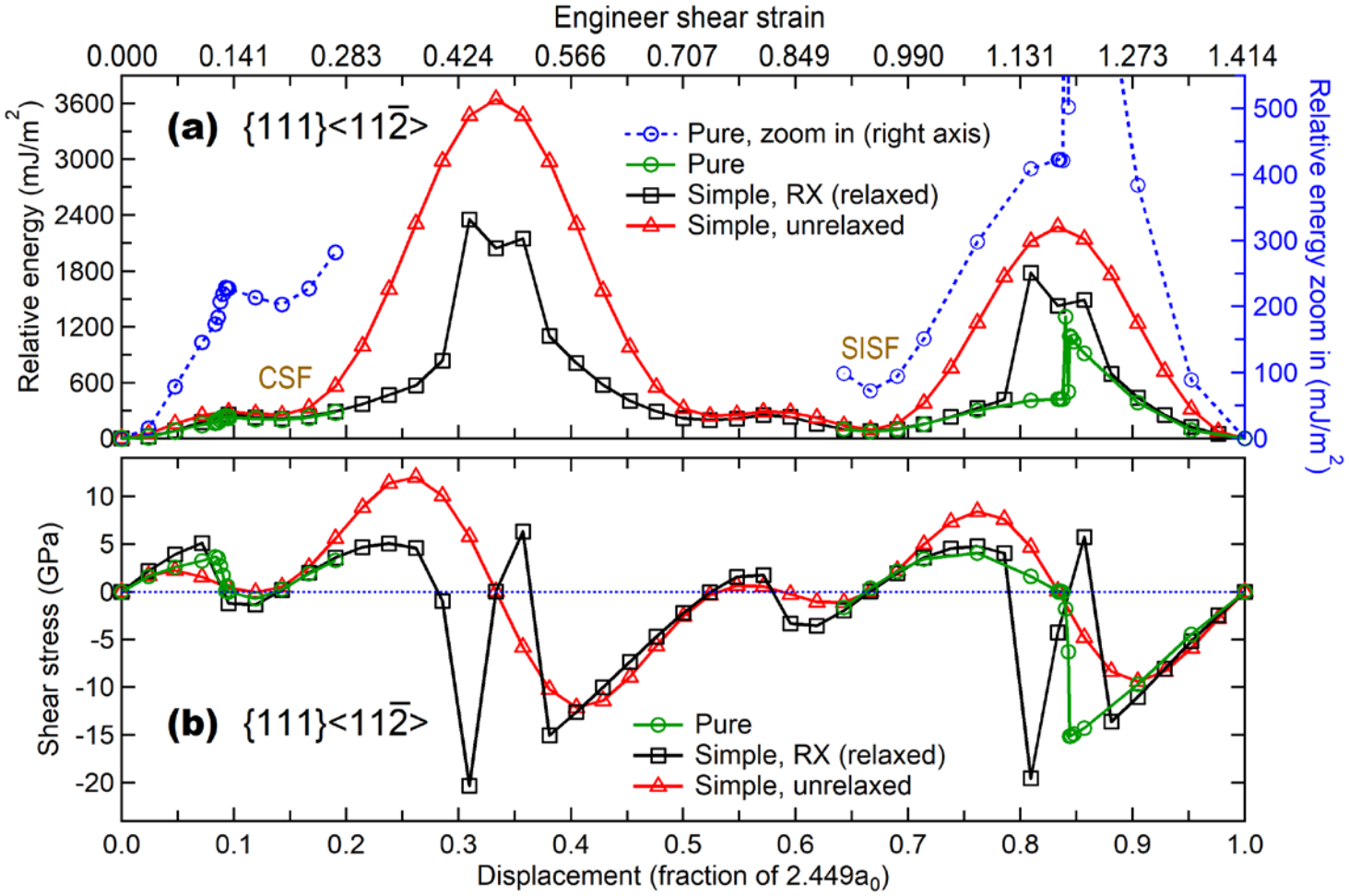
Screw

Shear deformation along {111}<112>: fcc Ni, $L1_2$ Ni_3Al

fcc Ni



$L1_2$ Ni_3Al



Shang et al., JPCM 24 (2012) 155402 for Ni and unpublished work for Ni_3Al

Stacking fault energy and ideal shear stress of Ni and Ni_3Al (pure shear deformation based on the above slide)

Material	Slip system	Name	Stacking fault (mJ/m^2)		Ideal shear stress τ_{max} (GPa)
			Stable	Unstable	
Ni	{111}<112>	SF	127	263	5.1
Ni_3Al	{111}<112>	CSF	203	229	3.7
Ni_3Al	{111}<112>	SISF	**72**	1308	15.2
Ni_3Al	{111}<110>	APB_{111}	205	230	4.2

fcc Ni: SF: intrinsic stacking faults energy
Ni_3Al: CSF: complex stacking fault
Ni_3Al: SISF: Superlattice intrinsic stacking fault
Ni_3Al: APB: antiphase boundary on {111}<110>

NOTE: SF and CSF are comparable

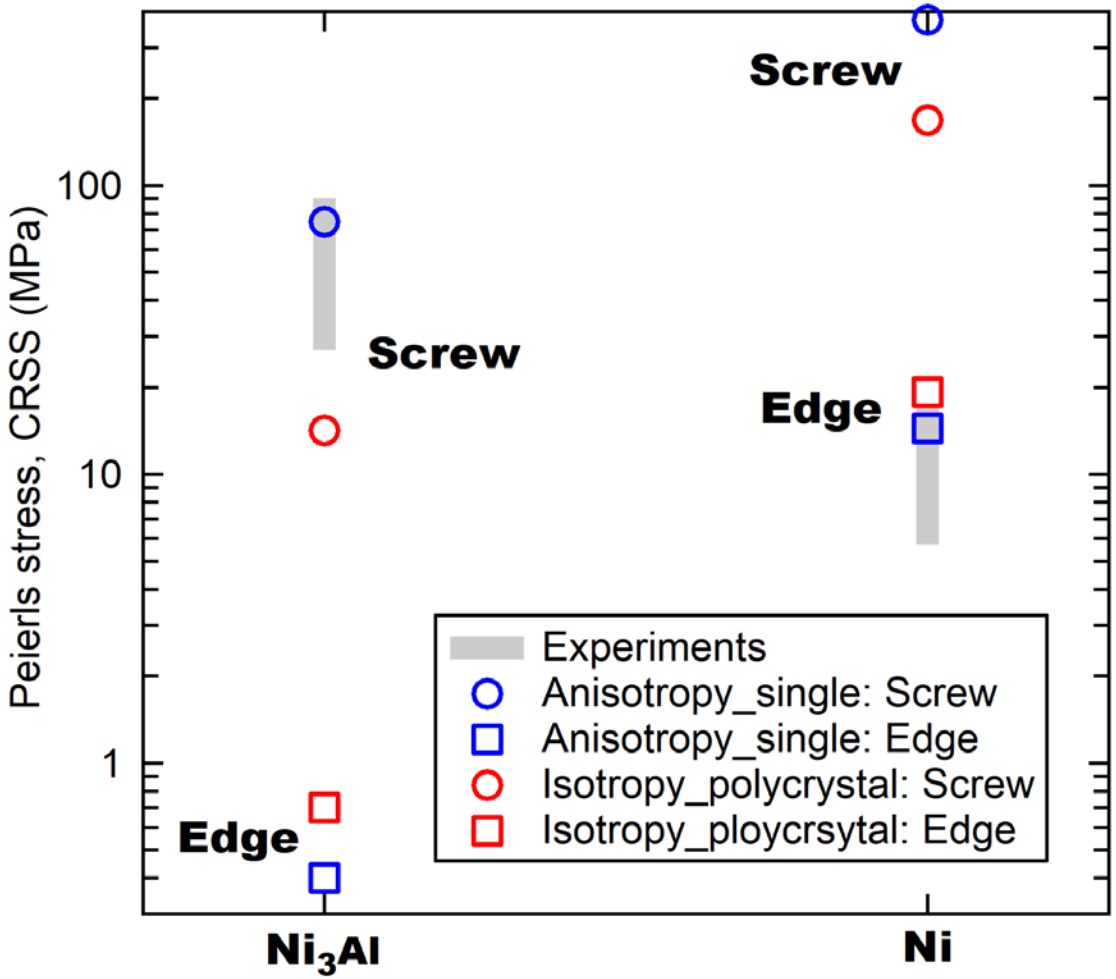
Common belief:

τ_{max} values of Ni and Ni_3Al are similar (~ 8 GPa)

This work:

τ_{max} value of Ni_3Al (3.7) is ~ 30% lower w.r.t. Ni (5.1)
Why? (see the next slide)

Predicted Peierls stress (CRSS at 0 K) of Ni and Ni_3Al



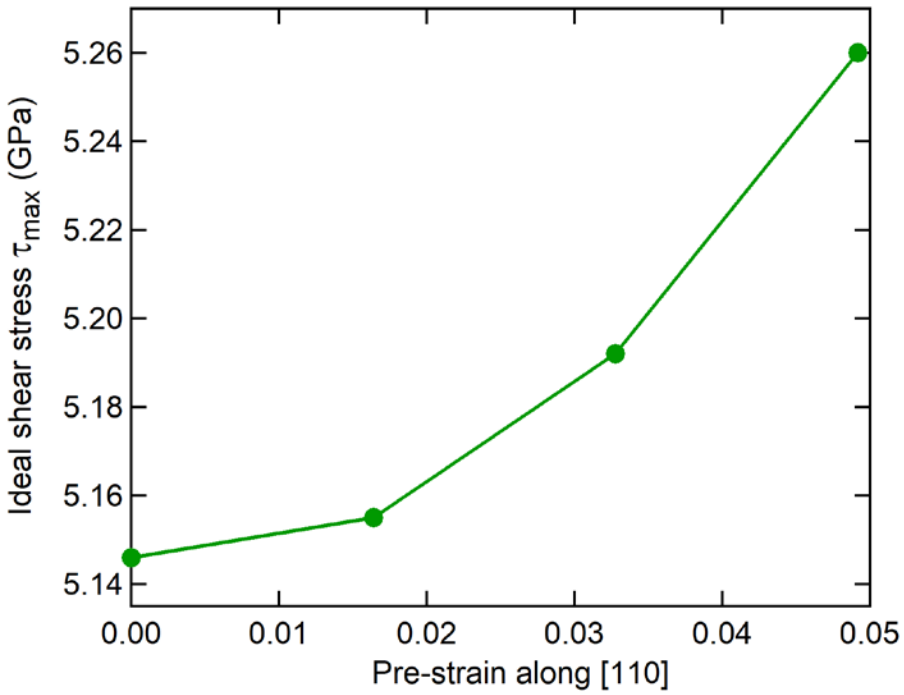
- **fcc γ-Ni: more slip systems**
  - This work: edge dislocations are dominant
  - Expt:[a] Edge dislocation dipoles are dominant in the first stage of fcc single crystal deformation

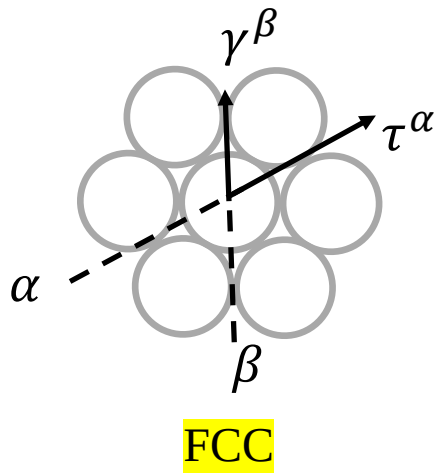
- **$L1_2$ γ'-Ni_3Al: few slip systems**
  - This work: screw dislocations are dominant
  - Expt:[b] Mainly screw dislocations, edge dislocations mainly act as links between screw dislocations. Cross-slip of screw dislocations, adopting the Kear-Wilsdorf (K-W) locking configurations, will result in yield strength anomaly (YSA).

[a]Nöhring and Curtin, Acta Mater 128 (2017) 135-148
[b]Wang-Koh, Mater Sci Technol 33 (2017) 934-943.

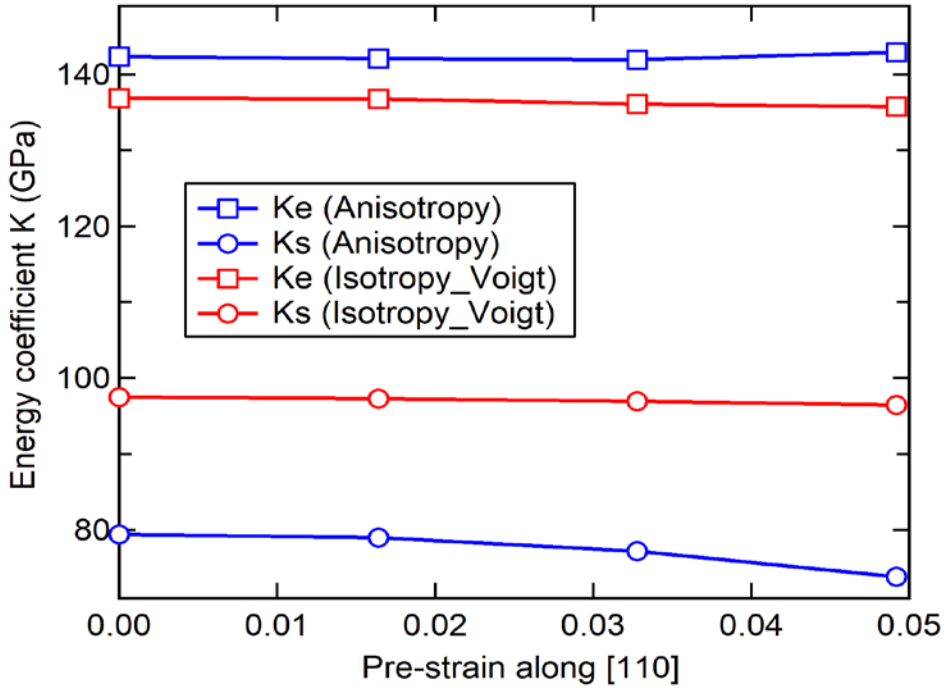
{111}<112> pure shear deformation of fcc Ni

Ideal shear stress τ_{max} and energy coefficient K along α-direction as a function of pre-strain along β-direction:





FCC



τ_{max} slightly increases, hence, CRSS will slightly increases

Ke (edge) almost constant

Ks (screw) constant or slightly decreasing

Crystal plasticity FEM (CPFEM) predictions

CPFEM

$$\dot{\gamma}^{\alpha} = \dot{\gamma}_0 \left|\frac{\tau^{\alpha}}{\tau_c^{\alpha}}\right|^{\frac{1}{m}} sgn(\tau^{\alpha})$$

$$\dot{\tau}_c^{\alpha} = \sum_{\beta=1}^{n} h_{\alpha\beta}\left|\dot{\gamma}^{\beta}\right|$$

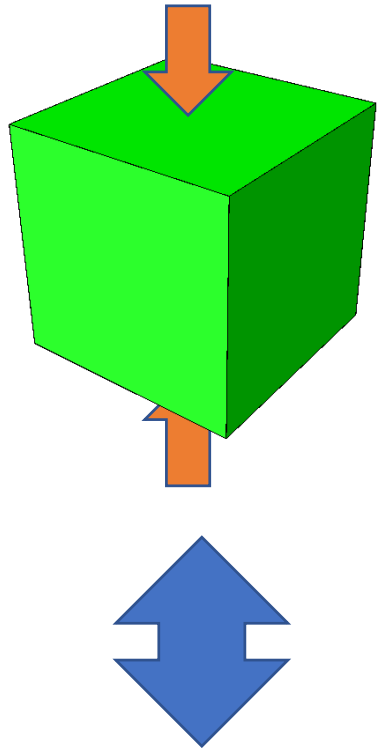
$$h_{\alpha\beta} = q_{\alpha\beta} h_0 \operatorname{sech}^2 \left|\frac{h_0 \gamma}{\tau_s - \tau_0}\right|$$

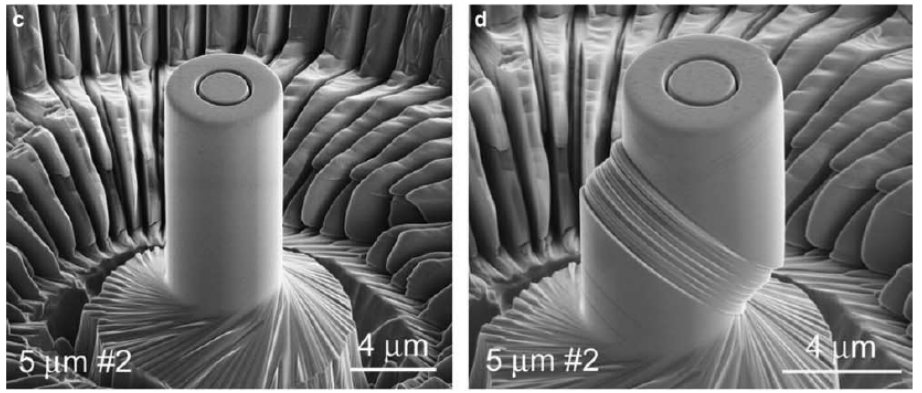
$$q_{\alpha\beta} = \begin{cases} 1, & \alpha = \beta \\ 1.4, & \alpha \neq \beta \end{cases}$$

$$\tau_c = \sigma_P$$

DFT

$$\sigma_p = \frac{Kb}{a'} \exp(-2\pi\zeta / a')$$

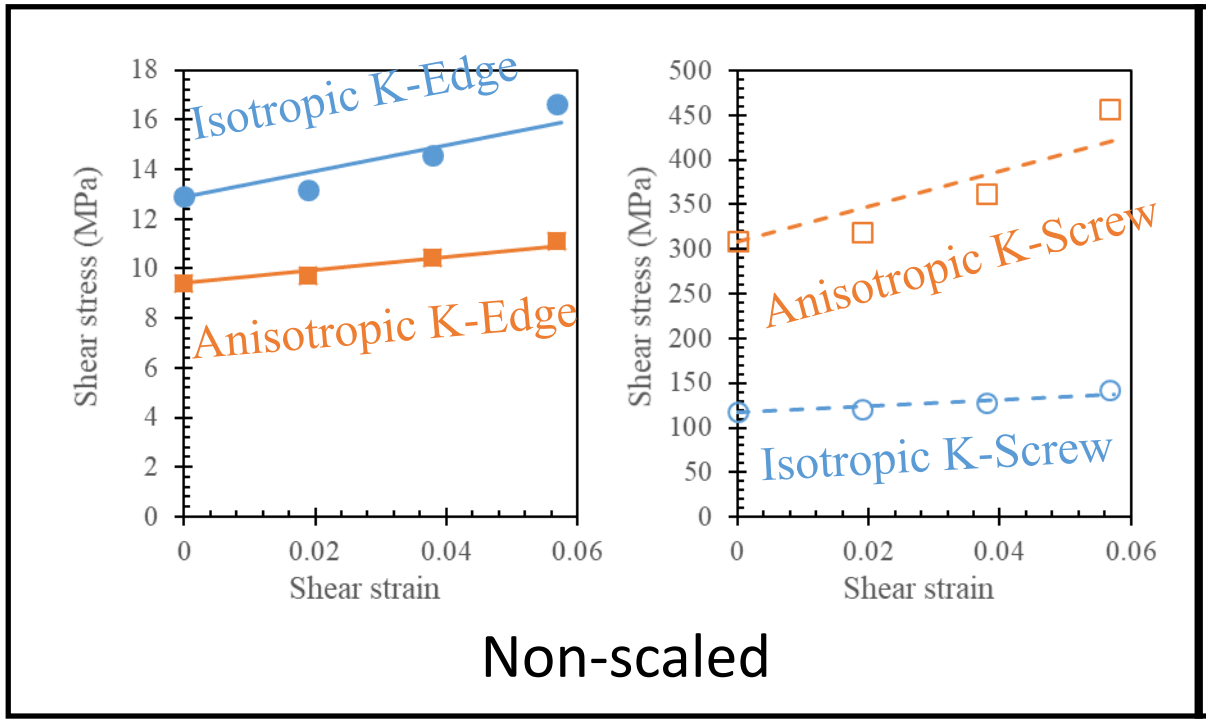




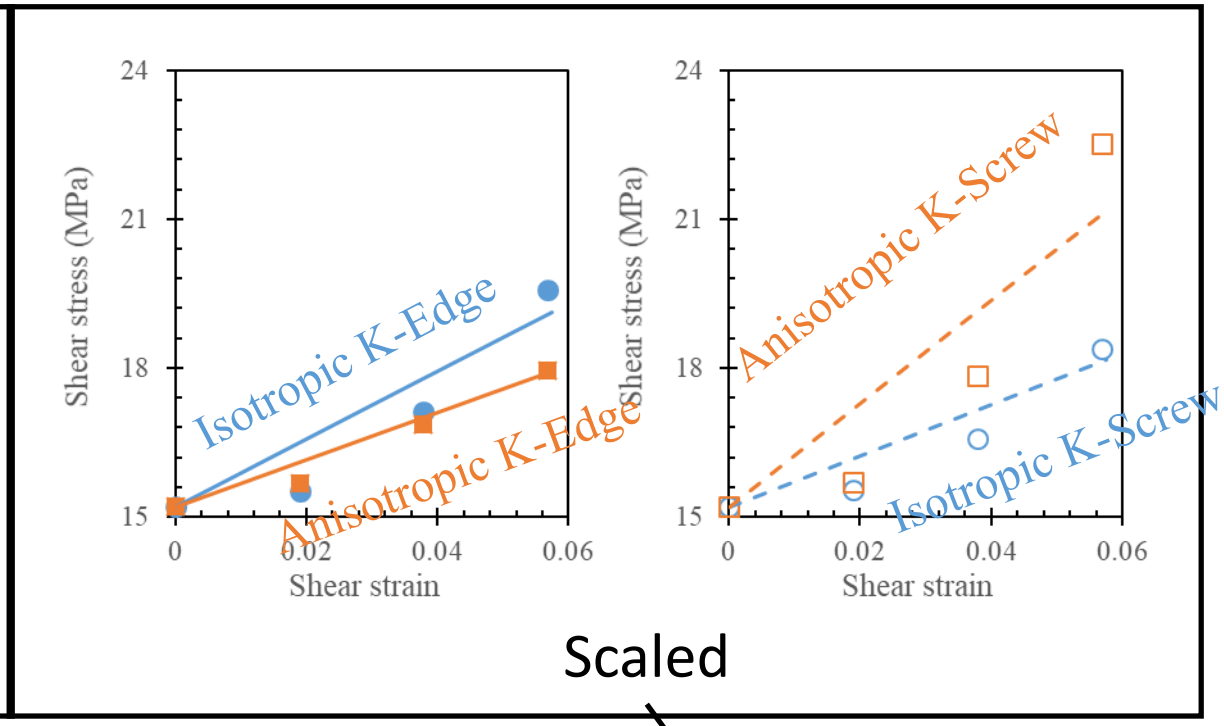
Dimiduk et al., Acta Mater 53(2005) 4065.

Crystal plasticity model parameter calibrations

Shear stress strain behavior on slip systems

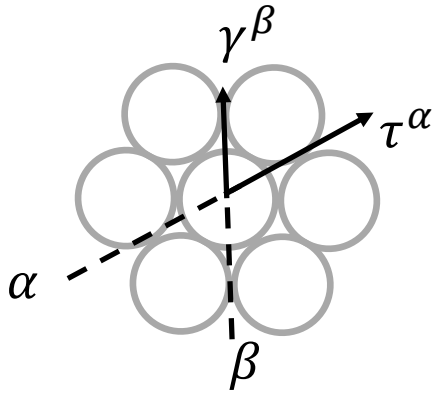


Non-scaled



Scaled

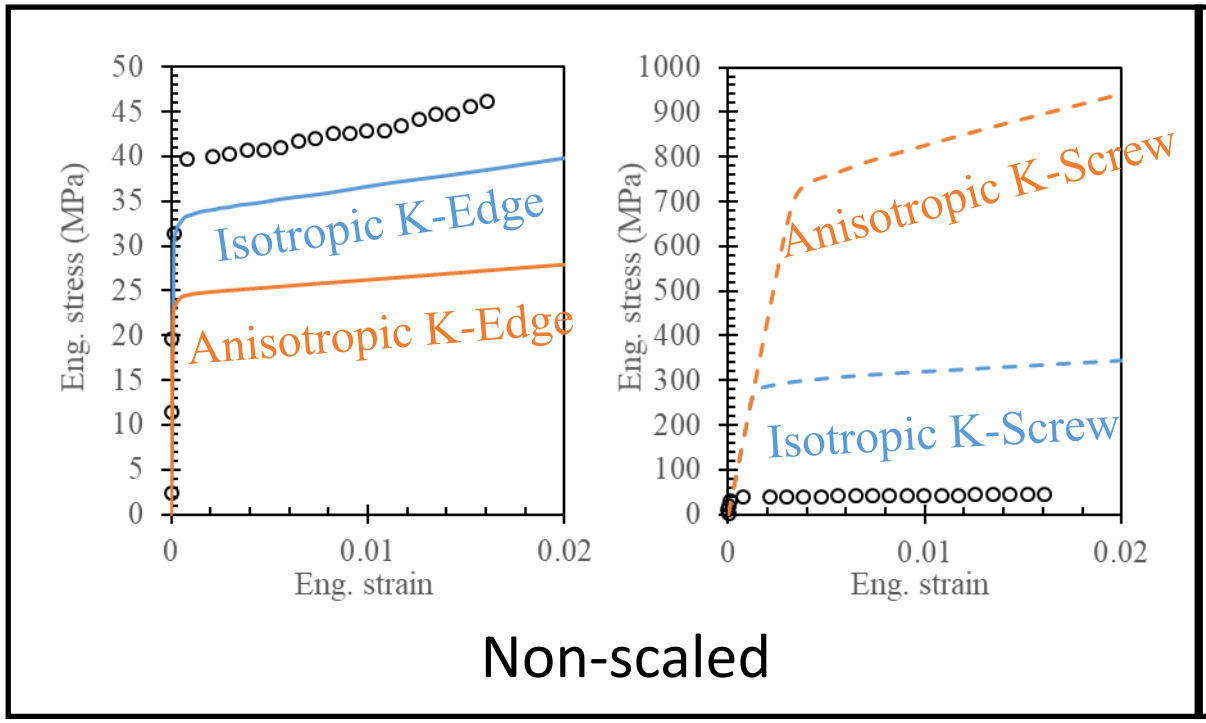
Symbols: DFT calculations
Lines: CPFEM calibration



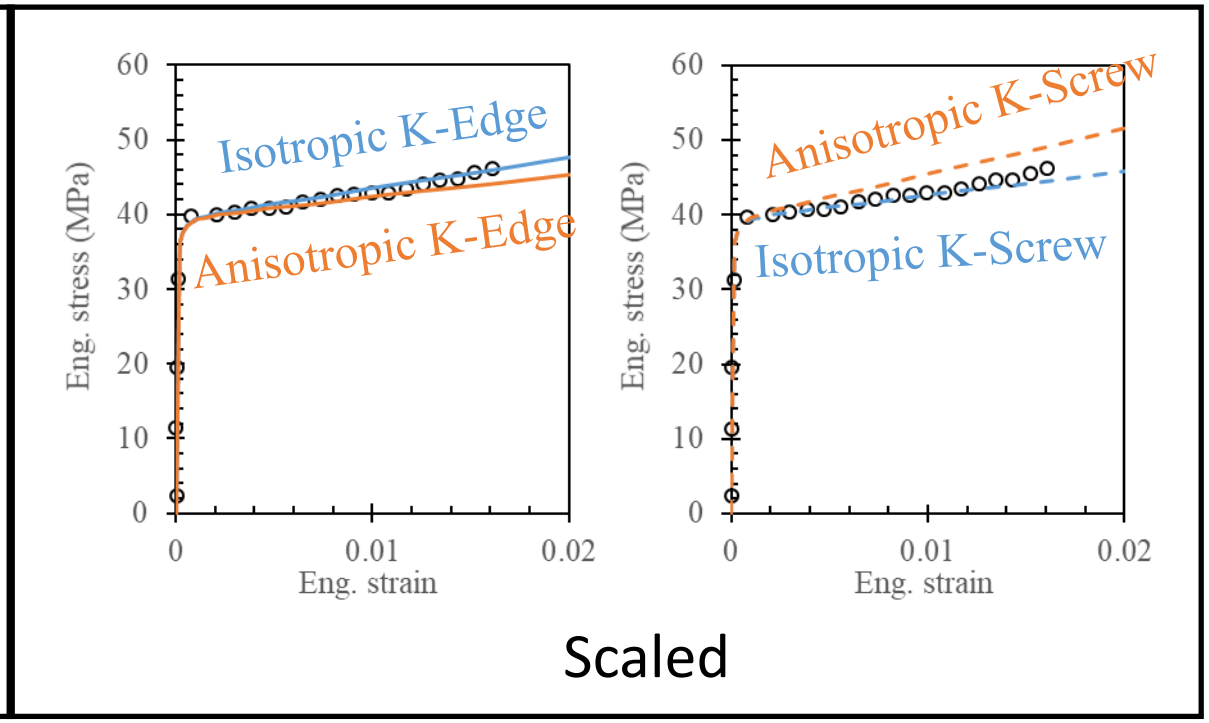
Based on experimental initial CRSS value (15.2 MPa)

Results of CPFEM predictions for single crystal of fcc Ni

Engineering stress strain behavior of a bulk single crystal under uniaxial compression



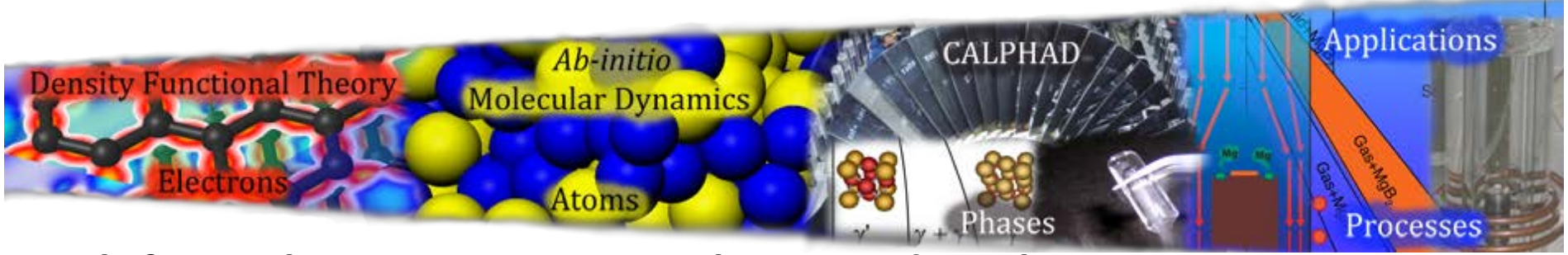
Non-scaled



Scaled

Symbols: Experiments (Dimiduk et al. Acta Mater 2005)
Lines: CPFEM results

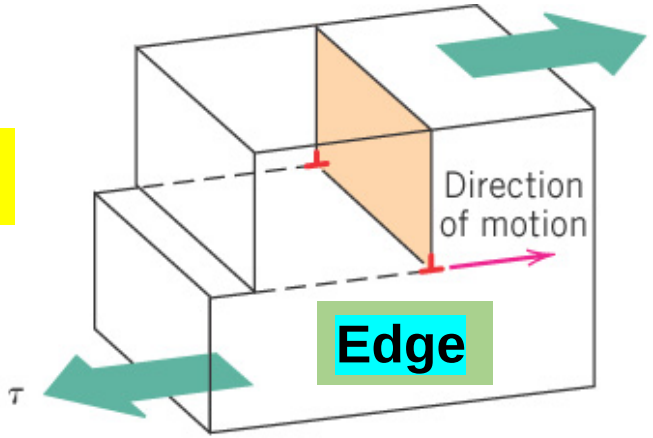
Summary

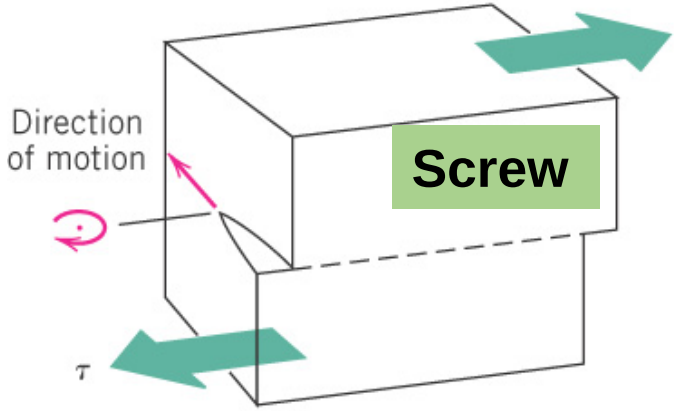


Multiscale approach from electron, atoms, to phase, and applications

- High-throughput calculations for fast data-gain
  - DFT-based first-principles calculations of thermodynamic/mechanical properties
  - CALPHAD modeling to develop the databases of interest
- Web-accessible codes and databases for broader impact
  - Python-based open source **codes:** DFTTK, *ESPEI*, *Pycalphad*
  - **Databases:** via Citrine.io
- Understanding and prediction of materials properties for extreme environments
  - DFT calculations of shear deformation
  - Phase-based FEM simulations of mechanical properties
  - Case study of Ni and Ni_3Al: CRSS, dislocation decomposition, strain-stress curves

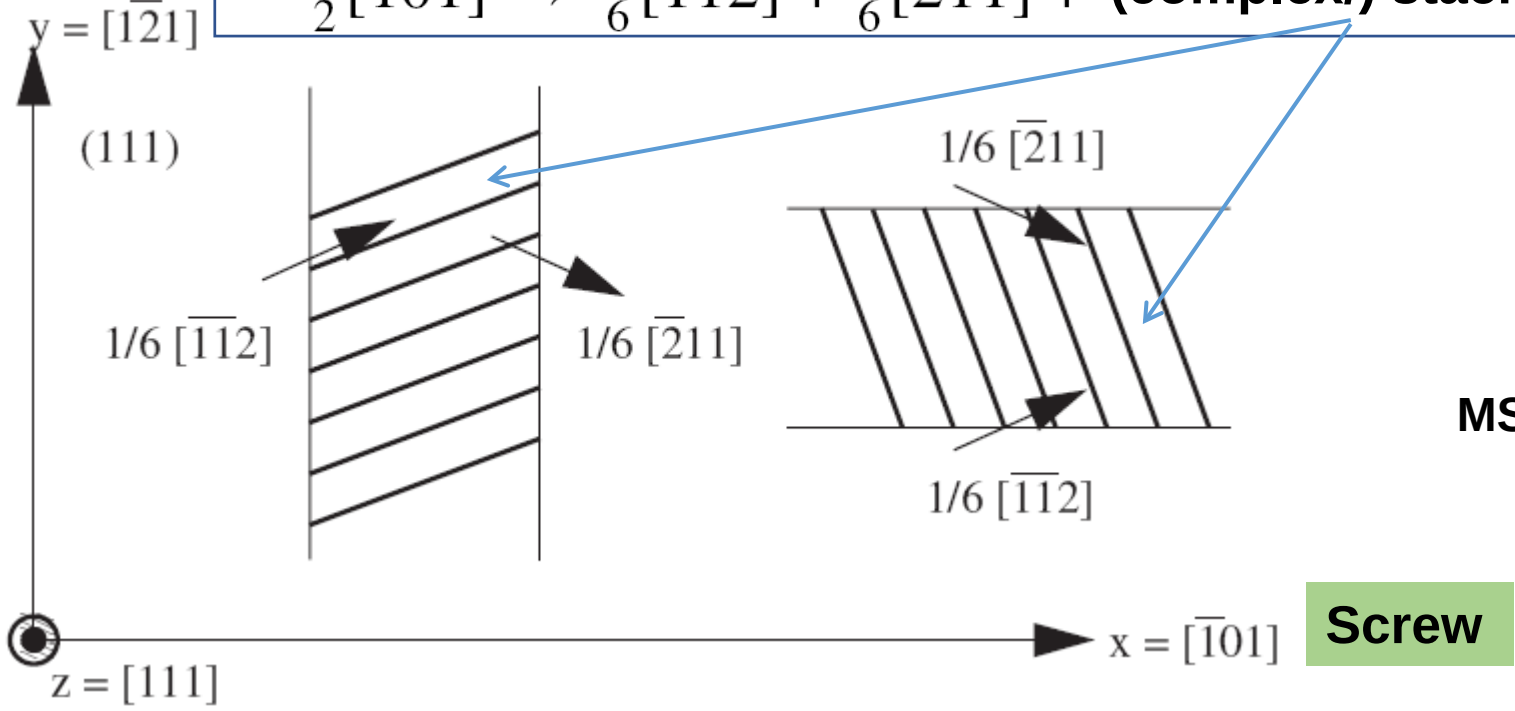
Summary





Edge

$$\tfrac{1}{2}[\bar{1}01] \rightarrow \tfrac{1}{6}[\bar{1}\bar{1}2] + \tfrac{1}{6}[\bar{2}11] +$$ **(complex/) stacking fault (C/SF)**



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Screw