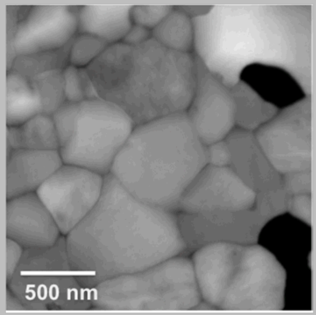
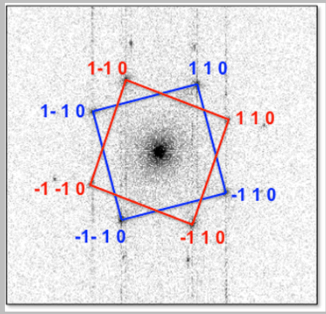
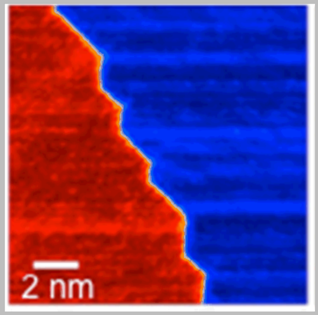
SAND2014-20675PE

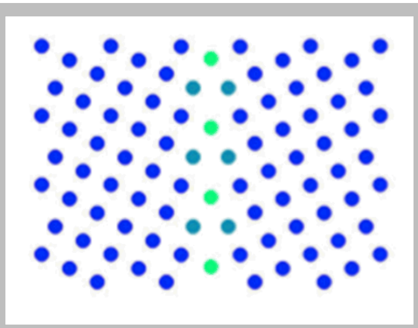
Exceptional service in the national interest

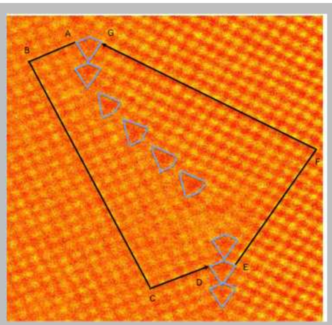
Sandia National Laboratories











Grain Boundaries: Experiment and Modeling

Doug Medlin (8341), Jon Zimmerman (8342),

Khalid Hattar (1111), Fadi Abdeljawad (1814),

Stephen Foiles (1814)

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. SAND NO. 2011-XXXXP

Interfaces: key to linking microstructural variability to performance and reliability

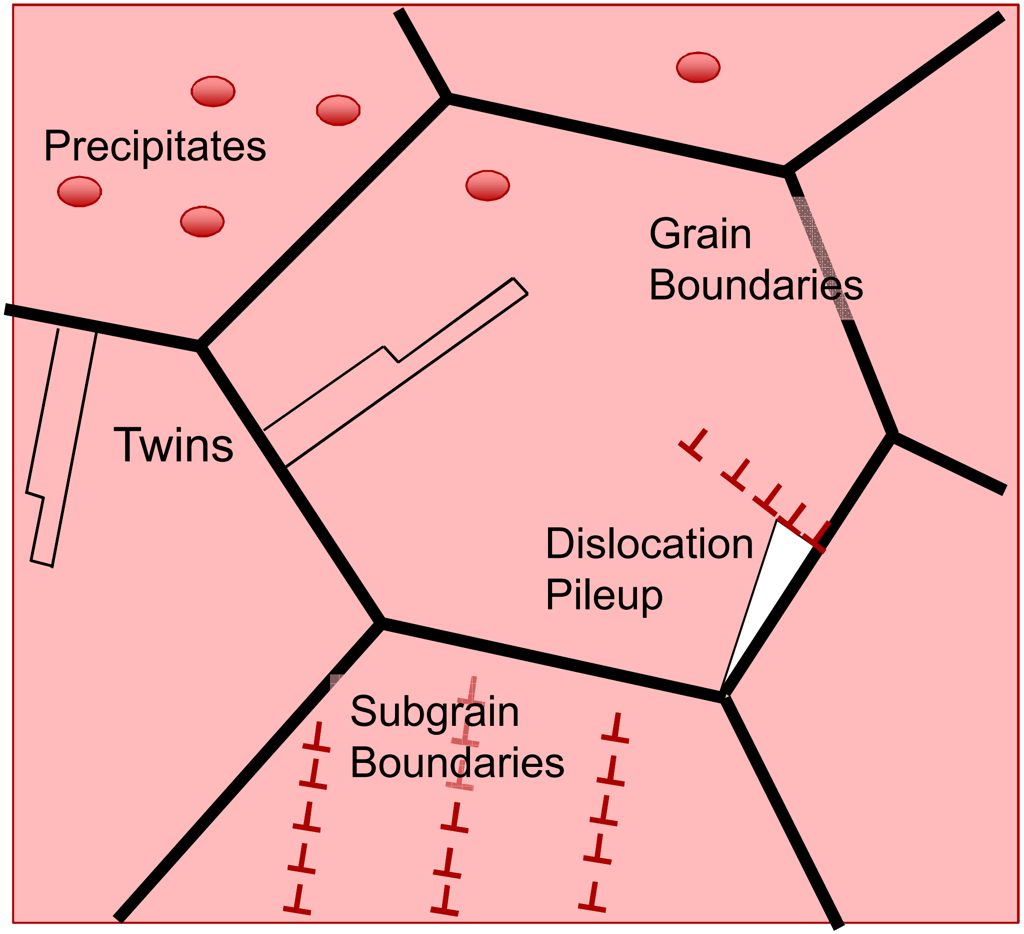
-Barriers to slip.
- -Hardening.
- -Stress concentration
- -Intergranular void nucleation, fracture.

-Softening at high T.
- -e.g., gb sliding, creep

-Sinks for point defects, dislocations
- -e.g. recrystallization

-Heterogeneous nucleation sites

-Compositional segregation, diffusional pathways.



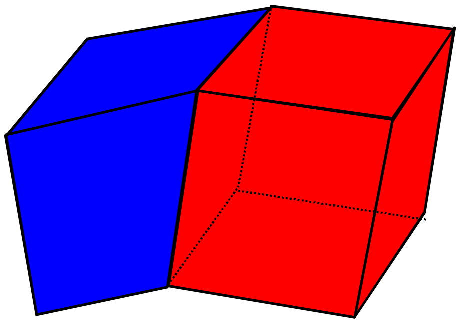
Our work under task 1 is increasingly focused on elementary defect processes at materials interfaces.

Our Emphasis and Approach:

•Elementary mechanisms relating structure to behavior and properties of interfaces at the atomistic and nano scale.

- Strain-localization, Slip Transfer
  - -Dependence on interface crystallography?
- How to generalize?
- How to capture interfacial variability?

This is hard: huge phase space



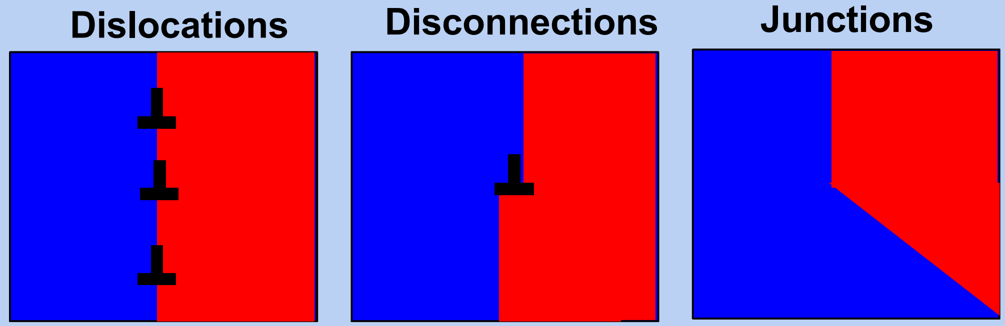
Grain boundary geometry characterized by 5 "macroscopic" degrees of freedom

misorientation (3 dof)
inclination (2 dof)

•Experiment: (Medlin and Hattar)
-Atomic resolution and diffraction contrast S/TEM
-Orientation imaging
-*in situ* straining
-Film growth

•Theory and Modeling:
(Zimmerman, Abdeljawad)
-atomistic simulations
-mesoscale: phase-field and elasticity

Interfacial line defects—a unifying concept



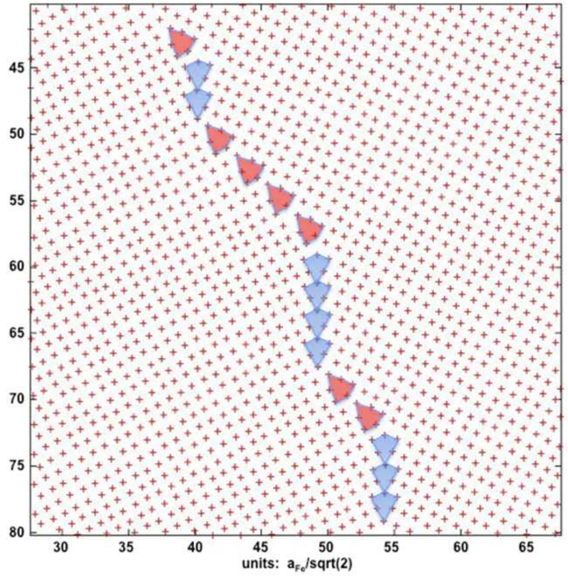
- generalize across misorientation-inclination space
-link between atomistic and continuum interface descriptions

Focus for today's talk:

•**Observations and calculations of atomic structure at a grain boundary in BCC Fe.**

-In contrast to grain boundaries in FCC metals, we know relatively little about the fundamental structure of BCC GBs.

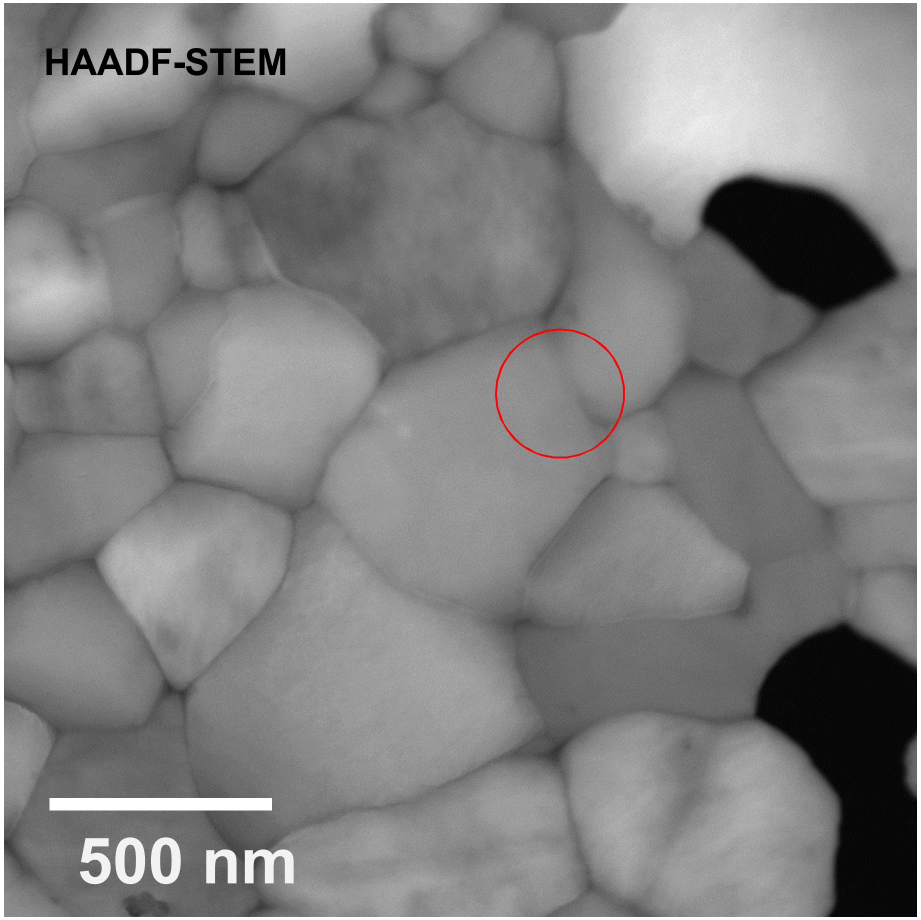
-Detailed analysis of facet and defect structure provides insight concerning interplay between structure, inclination/misorientation, and strain.

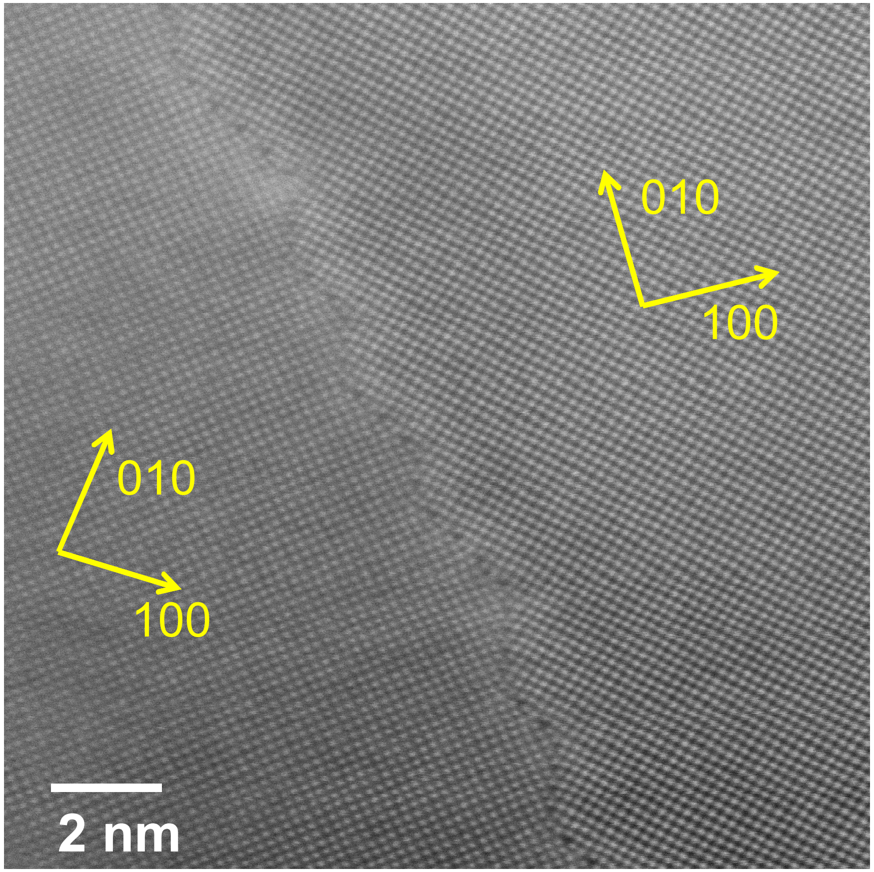


•**Discussion of upcoming activities and plans under Task 1.**

-Begin addressing impact of grain boundary structure on dislocation interactions and slip transmission processes.

Observations: Polycrystalline BCC Fe film

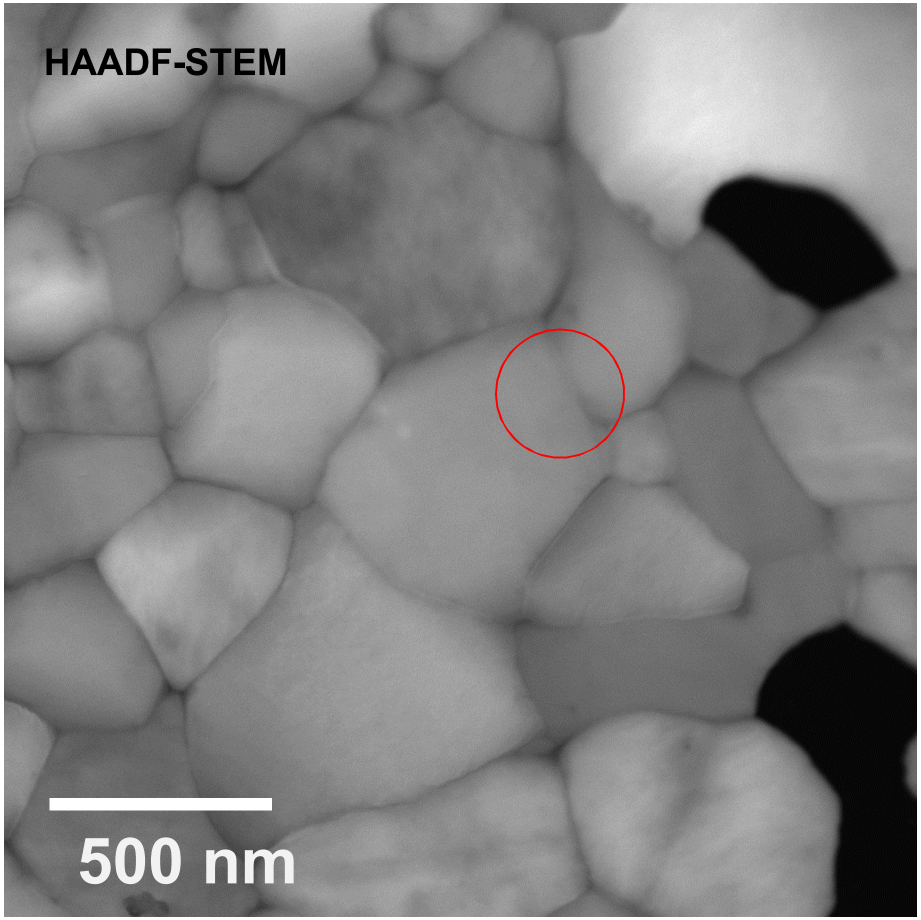




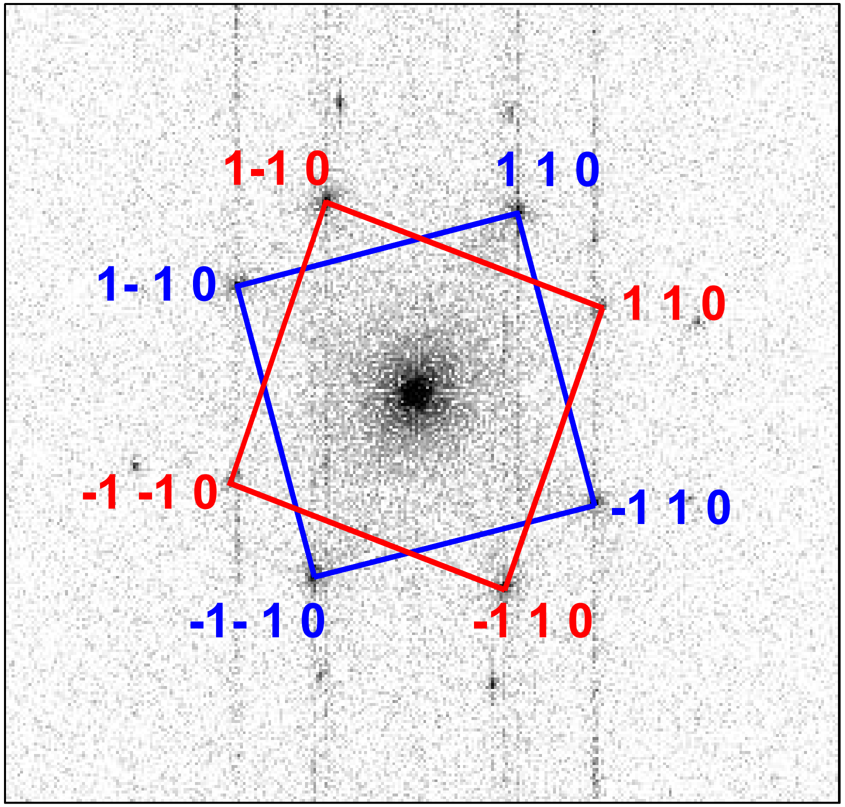
Pulsed Laser Deposited Fe on Rocksalt (NaCl). 36 nm thickness. Specimen released and annealed on Mo grid 675°C, 2 hours. under vacuum

HAADF-STEM
FEI-200 keV probe corrected Titan

Observations: polycrystalline Fe thin film

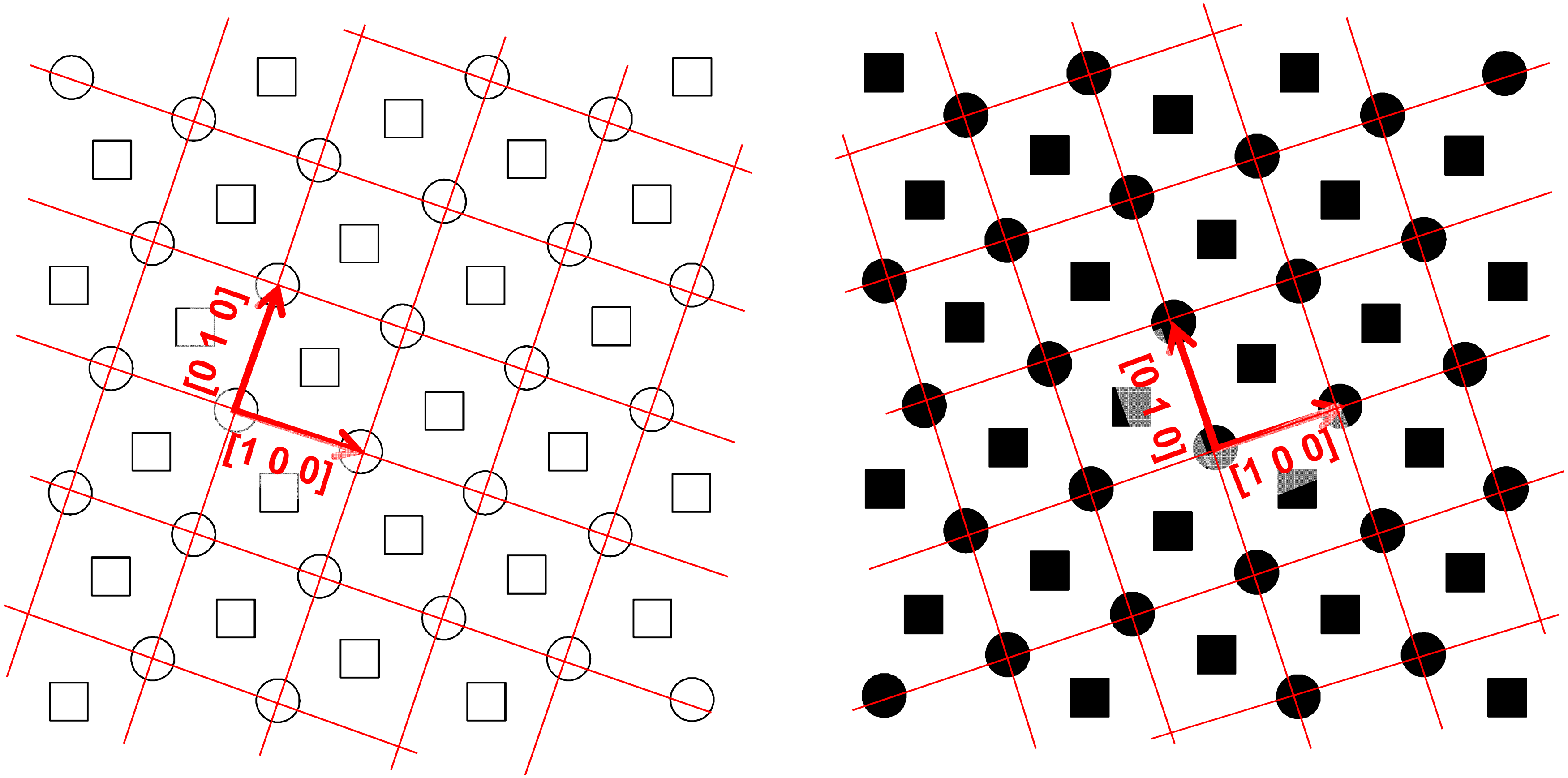


Pulsed Laser Deposited Fe on Rocksalt (NaCl). 36 nm thickness. Specimen released and annealed on Mo grid 675°C, 2 hours. under vacuum



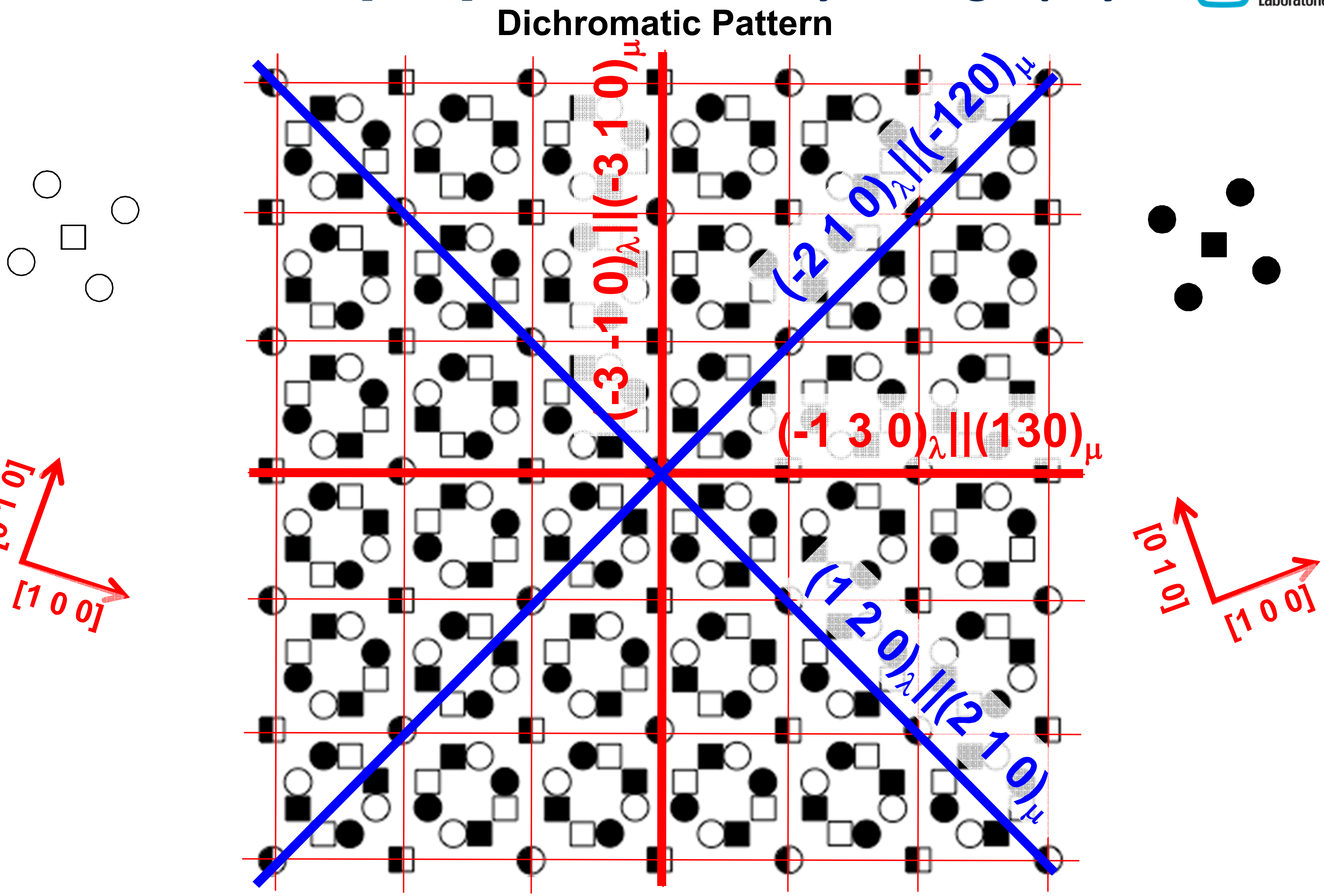
Measured misorientation: 34.49° ± 0.7°
Very close to Σ=5: $\theta_{\Sigma=5}$=36.87°
$\Delta\theta$= -2.38°

BCC Σ=5 [001]: Interfacial Crystallography



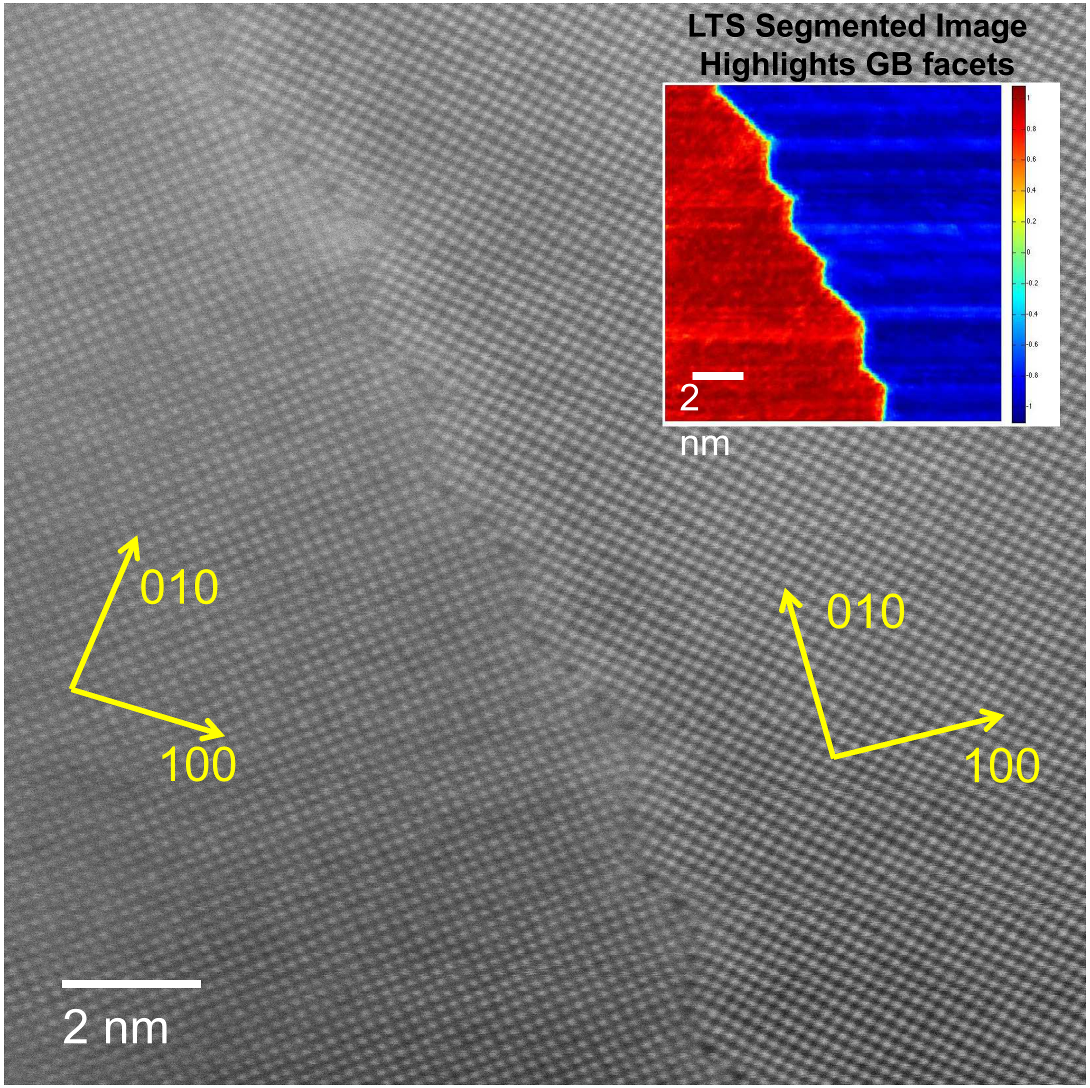
36.87° Rotation about [001]

BCC Σ=5 [001]: Interfacial Crystallography

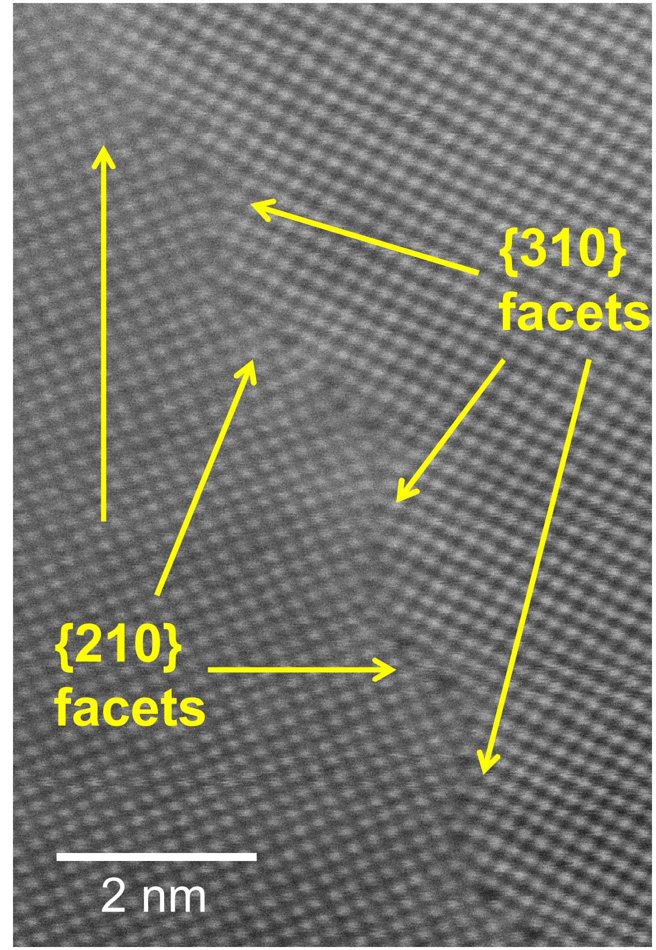


HRSTEM shows nanoscale faceting at Grain boundary

HAADF-STEM Σ=5 <001> Boundary in Fe



Boundary is faceted on
{210} and {310} type inclinations



Faceting: Signature of anisotropic interfacial energy

- **Driving force (μ) for interface evolution:**

H: mean curvature
V_m: molar volume
γ: interface energy

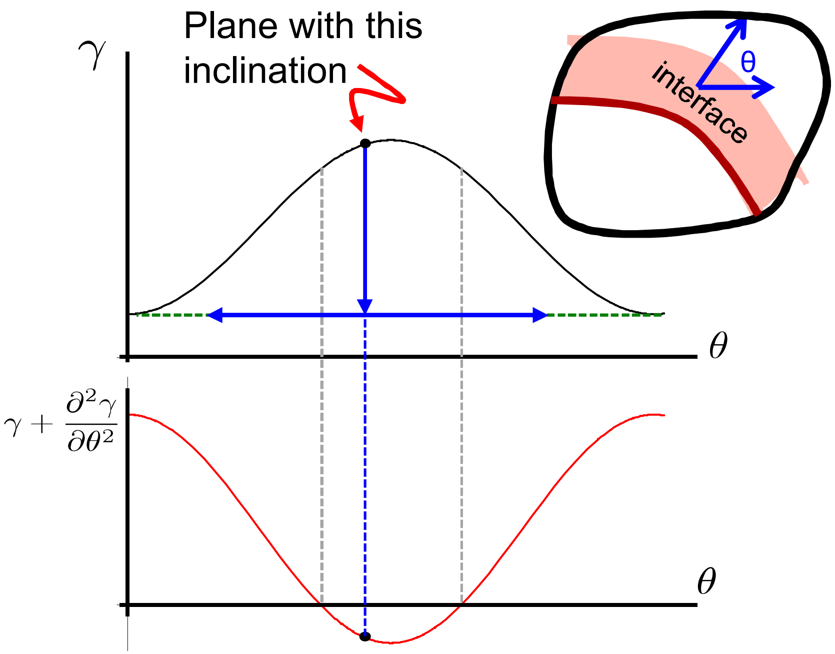
$$\mu \sim v_m \left(\gamma + \frac{\partial^2 \gamma}{\partial \theta^2} \right) H$$

Interface stiffness

W. W. Mullins (1963)

-Inclinations with negative interface stiffness break into facets with minimum energy orientations.

-"interface spinodals": analogous to phase separation in bulk materials.

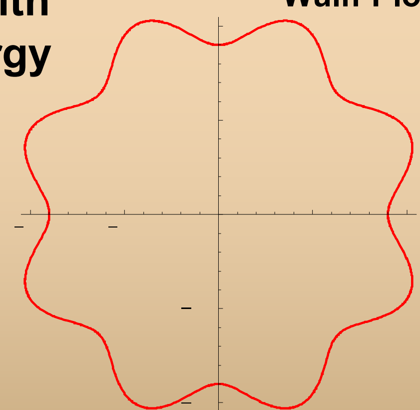


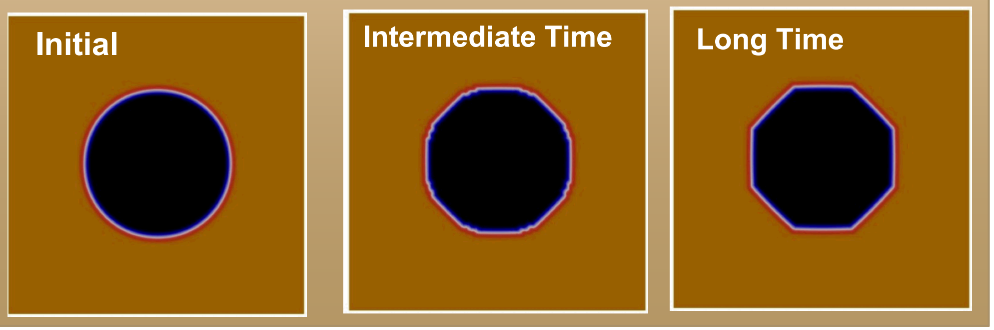
Frank (1963), Cabrera (1964), Stewart (1992), Liu (1993)

Example: GB Evolution with anisotropic interfacial energy

-Wulff surface with minima every 45°

-Phase-field simulation.





Faceting: Signature of anisotropic interfacial energy

- **Driving force (μ) for interface evolution:**

H: mean curvature
V_m: molar volume
γ: interface energy

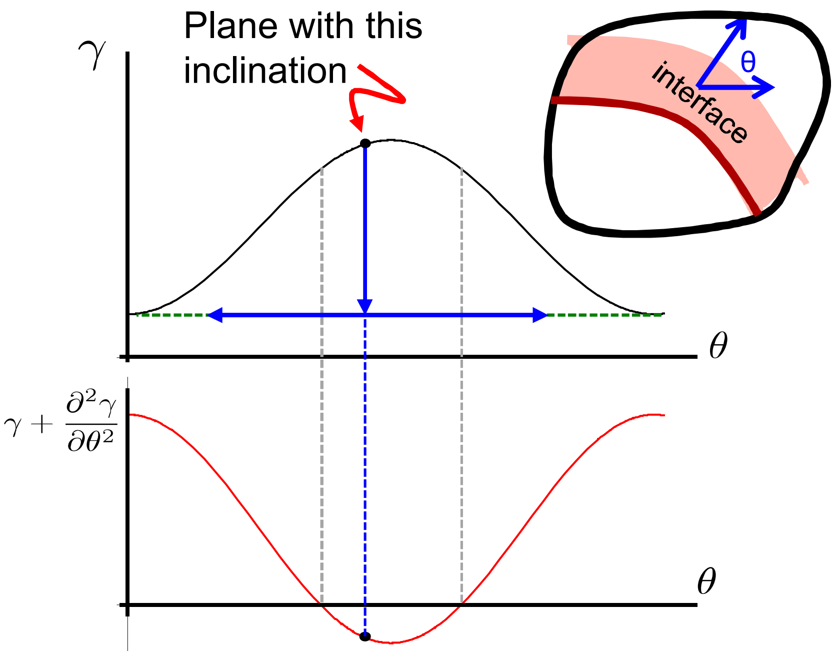
$$\mu \sim v_m \left(\gamma + \frac{\partial^2 \gamma}{\partial \theta^2} \right) H$$

W. W. Mullins (1963)

Interface stiffness

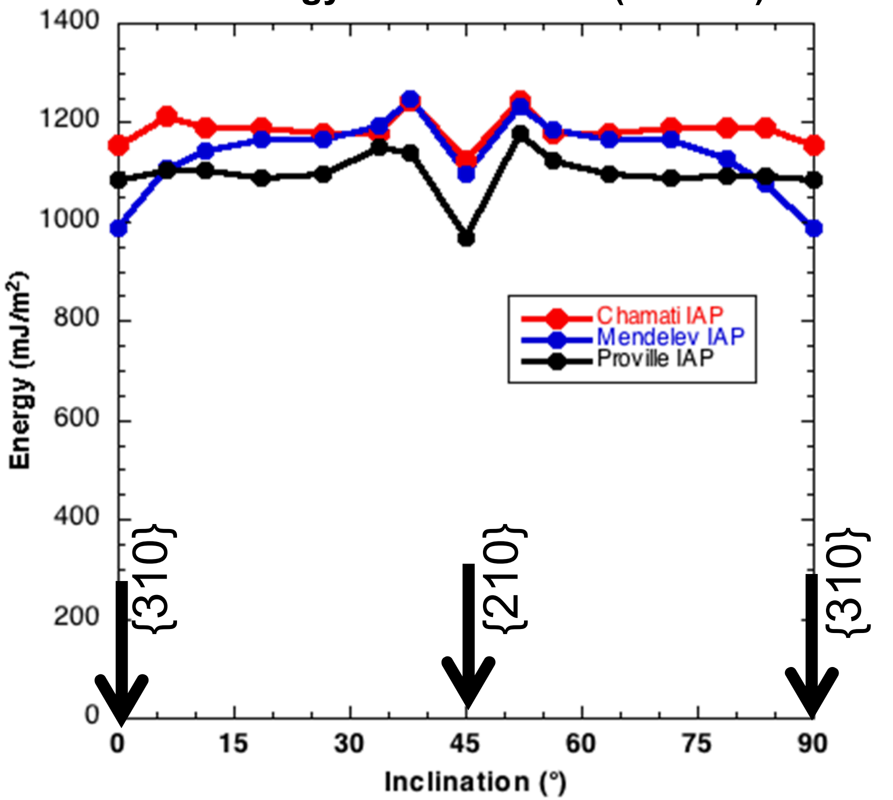
-Inclinations with negative interface stiffness break into facets with minimum energy orientations.

-"interface spinodals": analogous to phase separation in bulk materials.



Frank (1963), Cabrera (1964), Stewart (1992), Liu (1993)

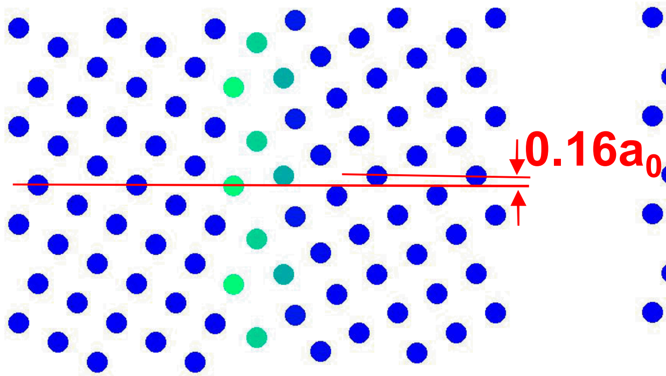
GB Energy vs. Inclination (Fe Σ=5)



Σ=5 {310} Structures with different Potentials

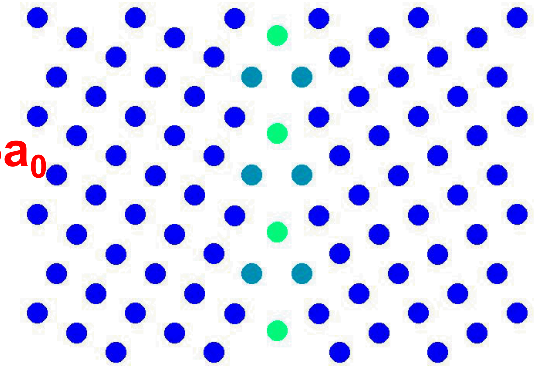
Sandia National Laboratories

Asymmetric



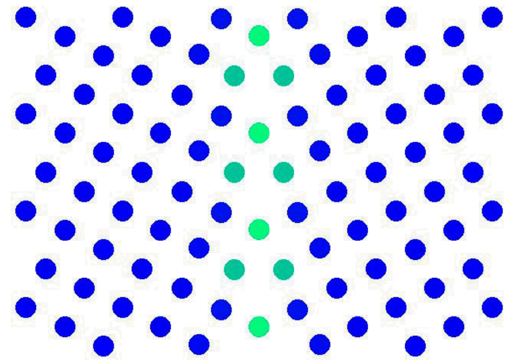
Potential: Chamati, 2006

Symmetric



Potential: Mendelev, 2003

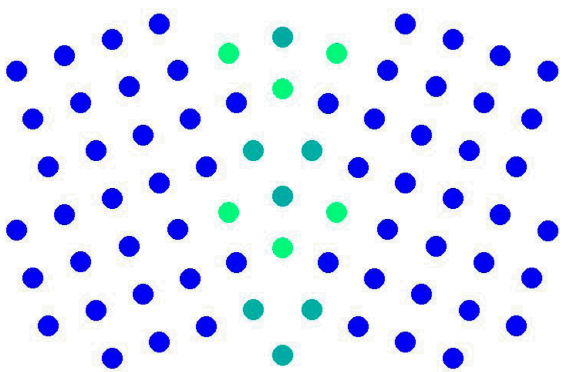
Symmetric



Potential: Proville, 2012

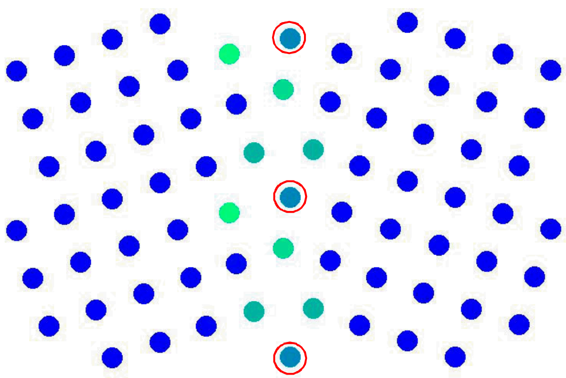
Σ=5 {210} Structures with different Potentials

Symmetric



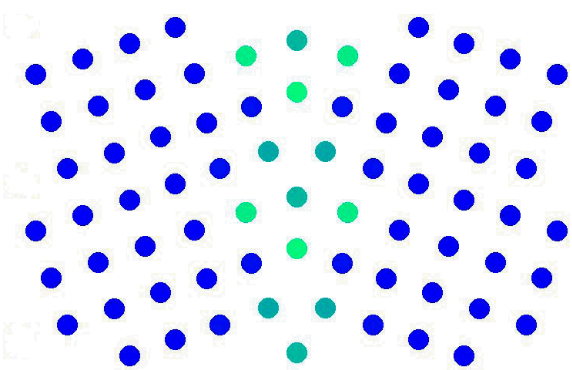
Potential: Chamati, 2006

Asymmetric



Potential: Mendelev, 2003

Symmetric

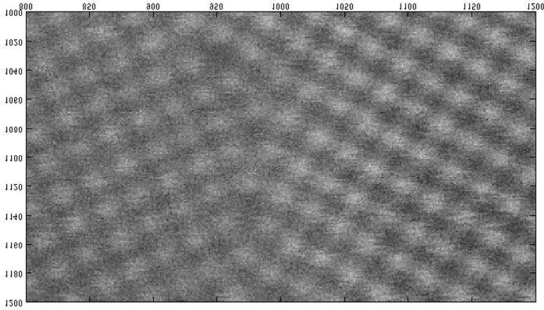


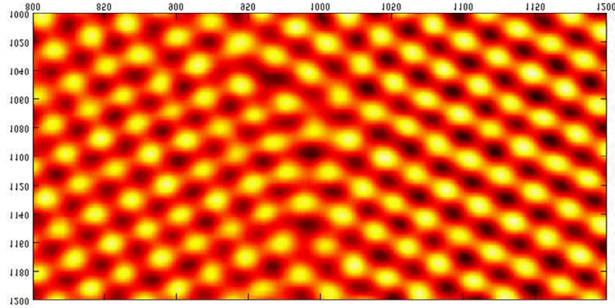
Potential: Proville, 2012

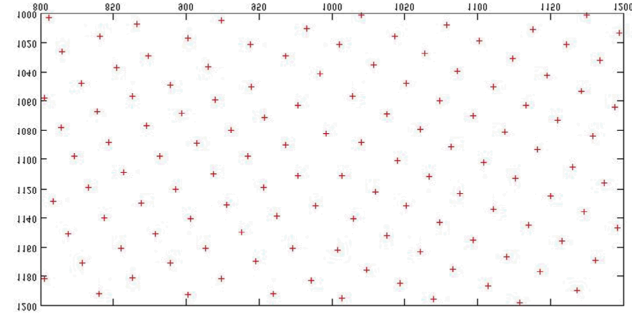
Atoms shaded by centrosymmetry parameter

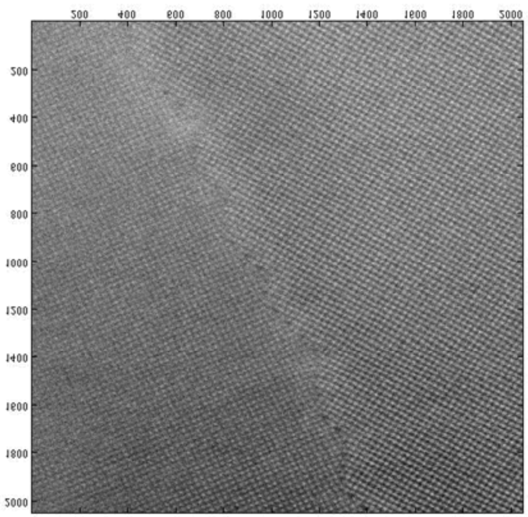
Quantifying the GB Images: Peak Location

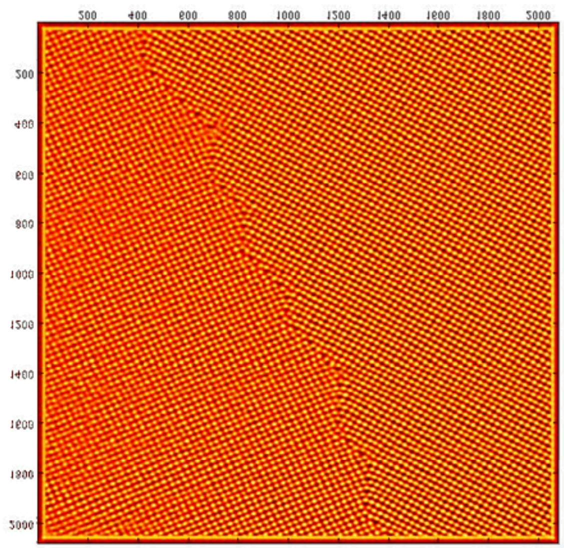
Raw HAADF STEM Image | **Correlation Image-Guassian** | **Peak Positions**

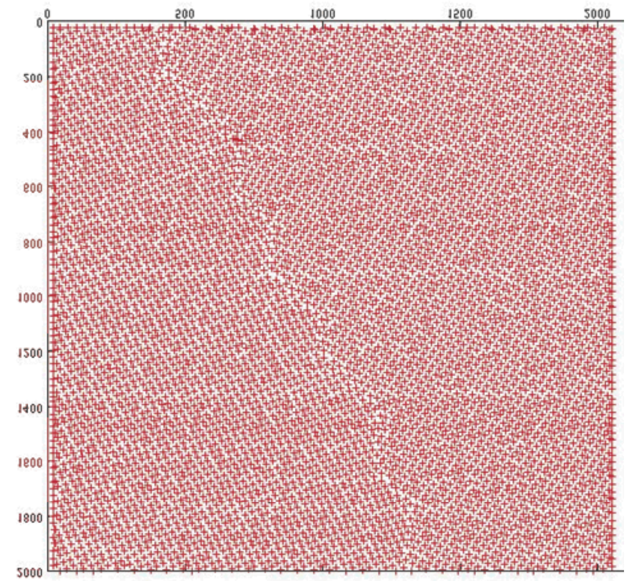




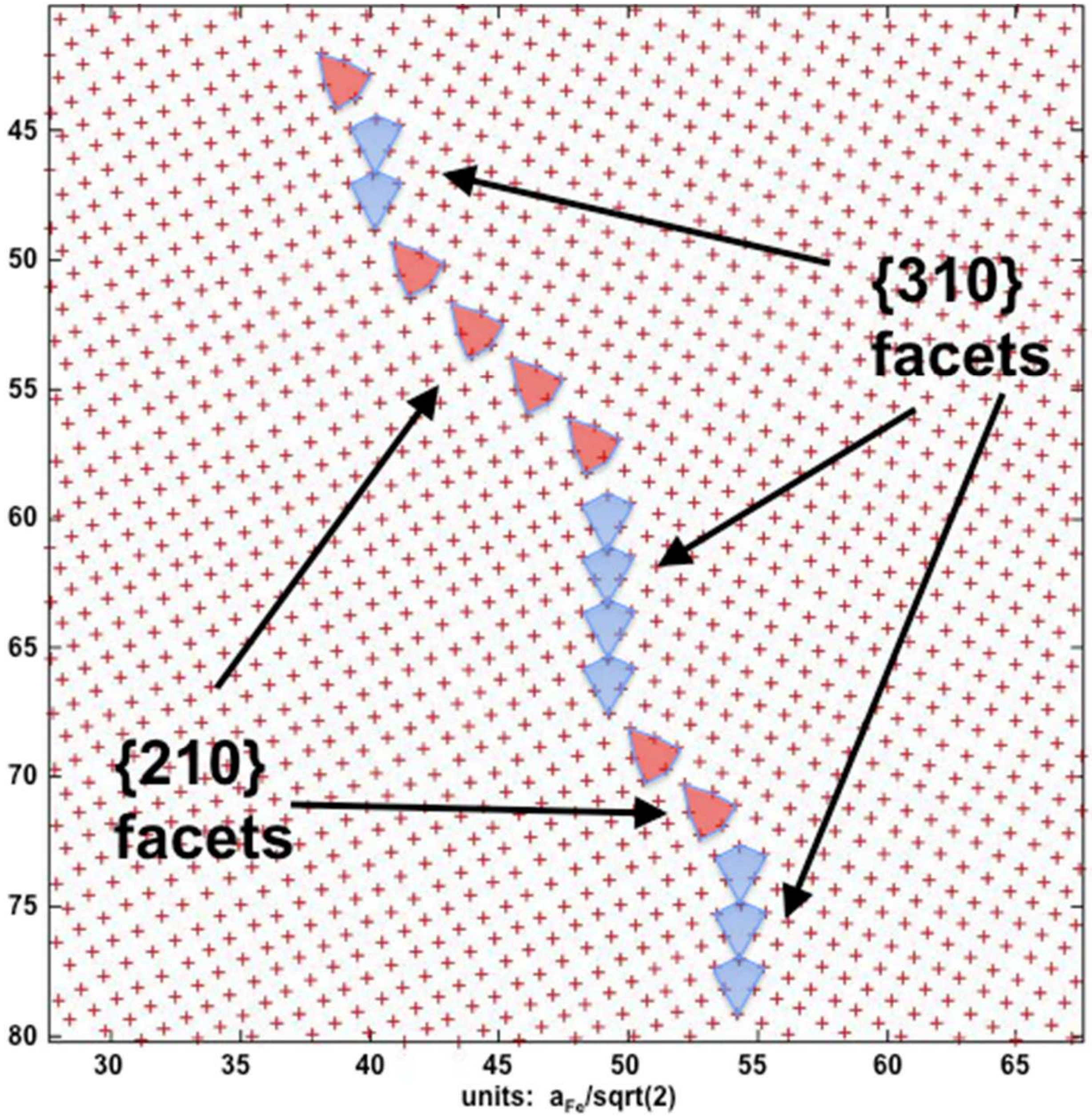








Shear distortion due to specimen drift during image acquisition.
Corrected by affine transformation to peak position array.

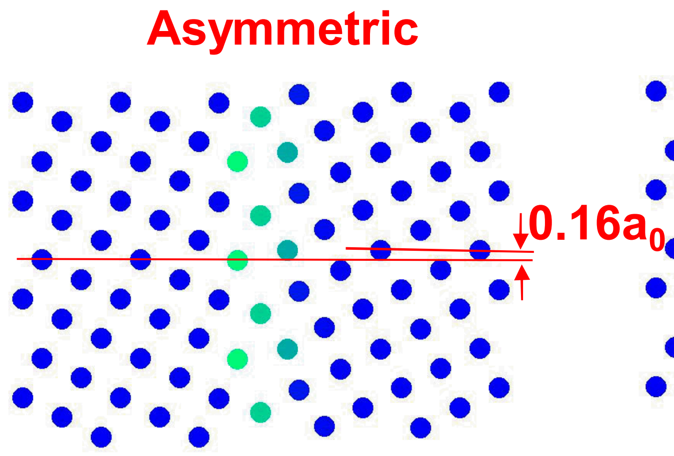


Intensity peak positions from HAADF-STEM of Fe Σ=5 grain boundary

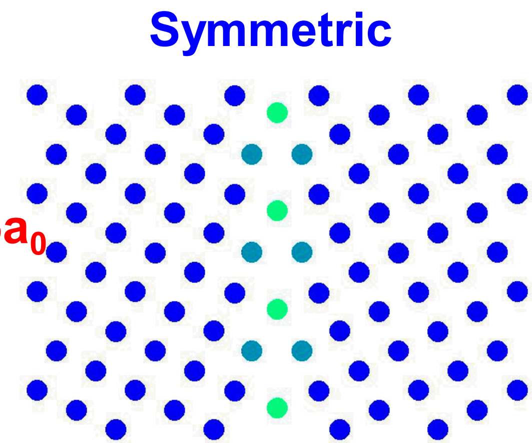
How do the {310} and {210} structural units compare with atomistic predictions?

Σ=5 {310} Structures with different Potentials

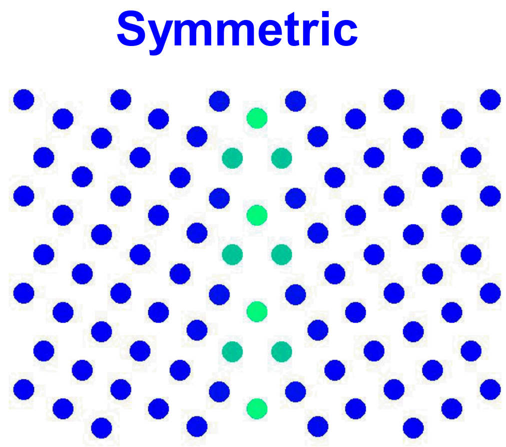
Sandia National Laboratories



Potential: Chamati, 2006



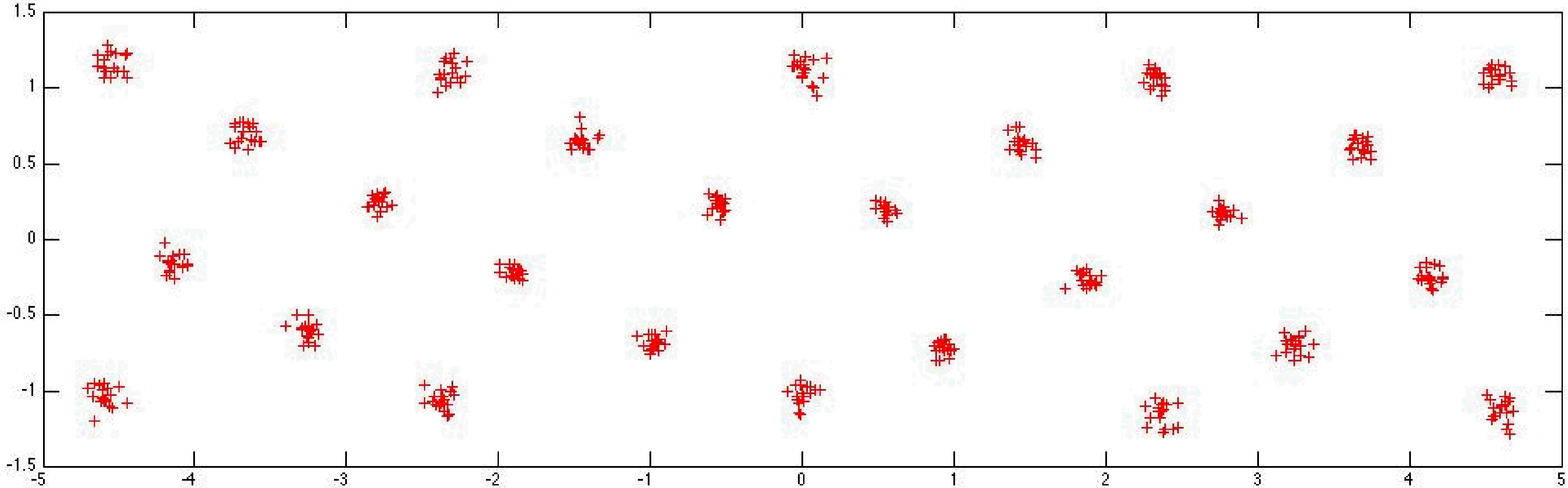
Potential: Mendelev, 2003



Potential: Proville, 2012

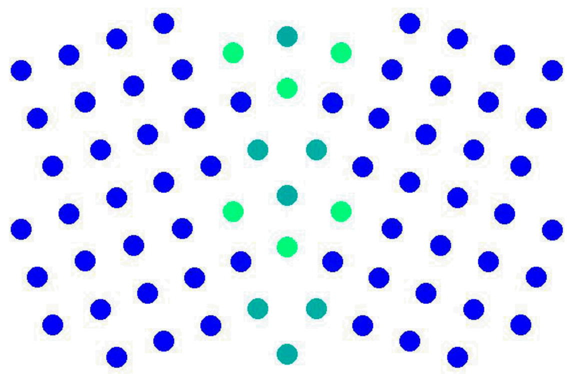
Experimental Peak Positions (HAADF STEM)

$\Delta y = -0.015 \pm 0.036\ a_0$

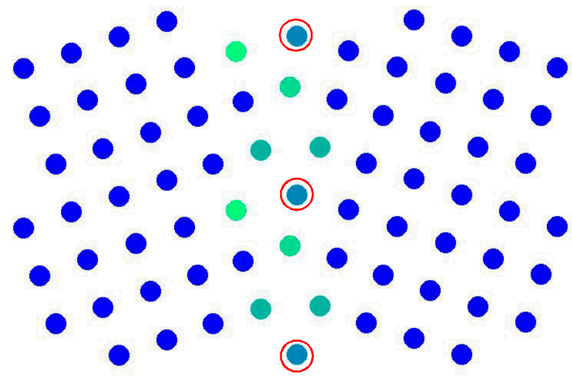


Σ=5 {210} Structures with different Potentials

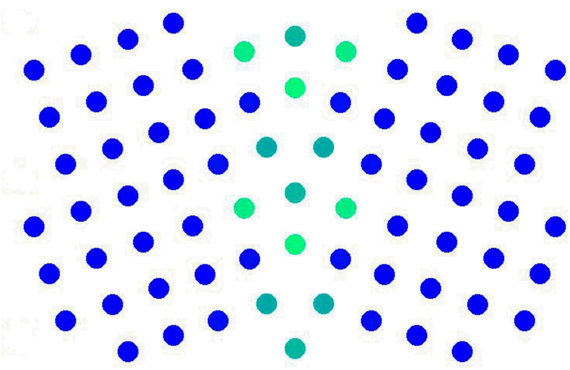
Sandia National Laboratories



Potential: Chamati, 2006

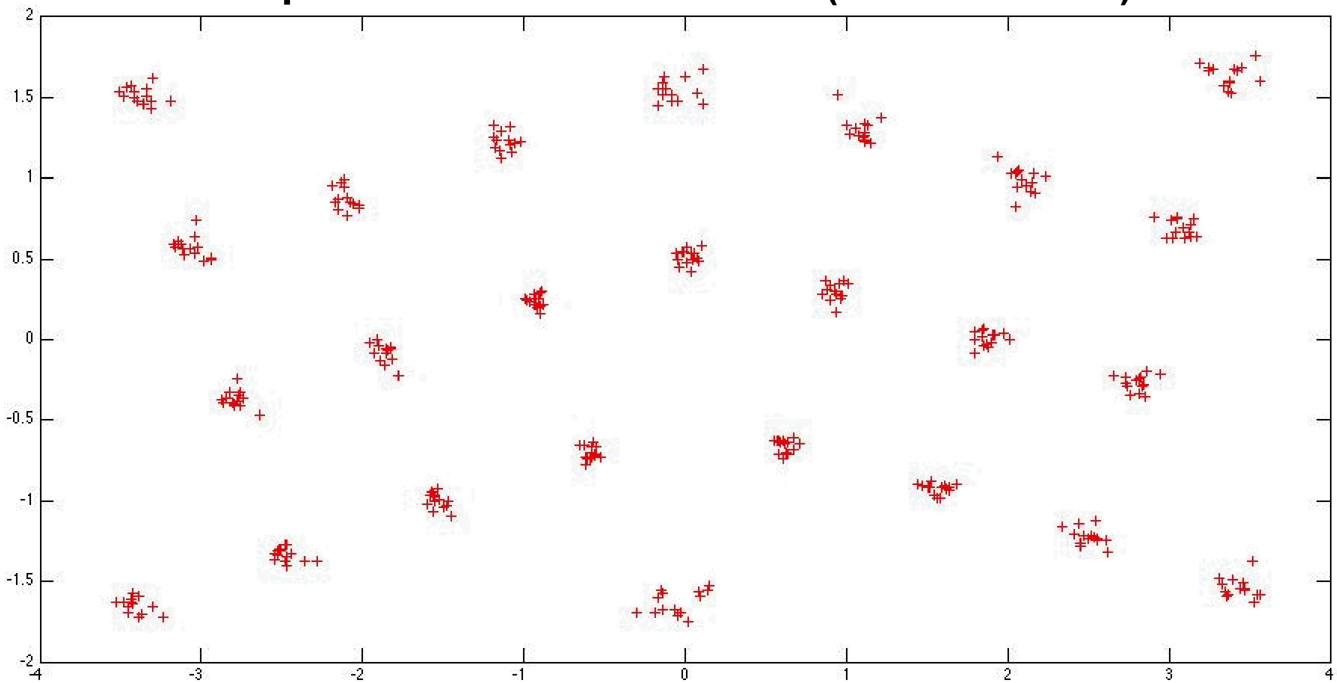


Potential: Mendelev, 2003



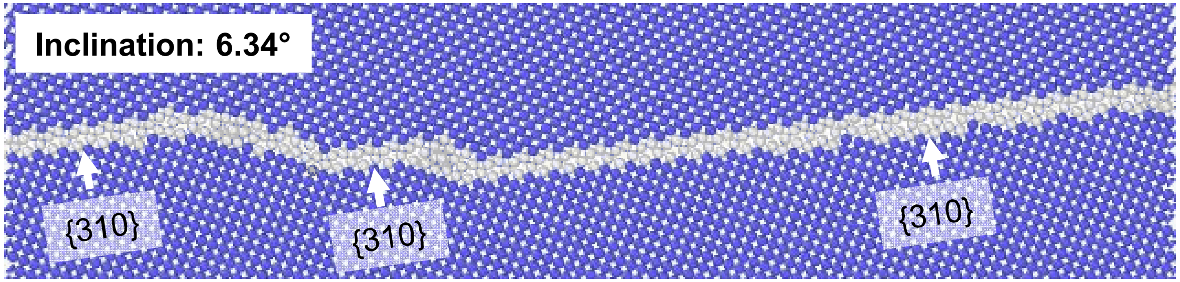
Potential: Proville, 2012

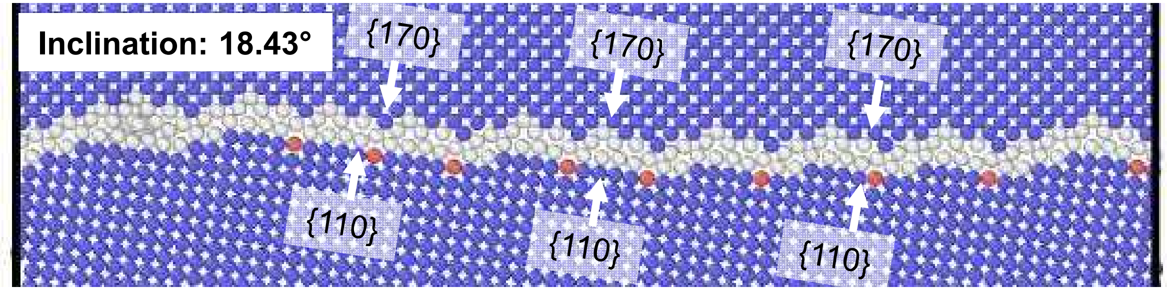
Experimental Peak Positions (HAADF STEM)

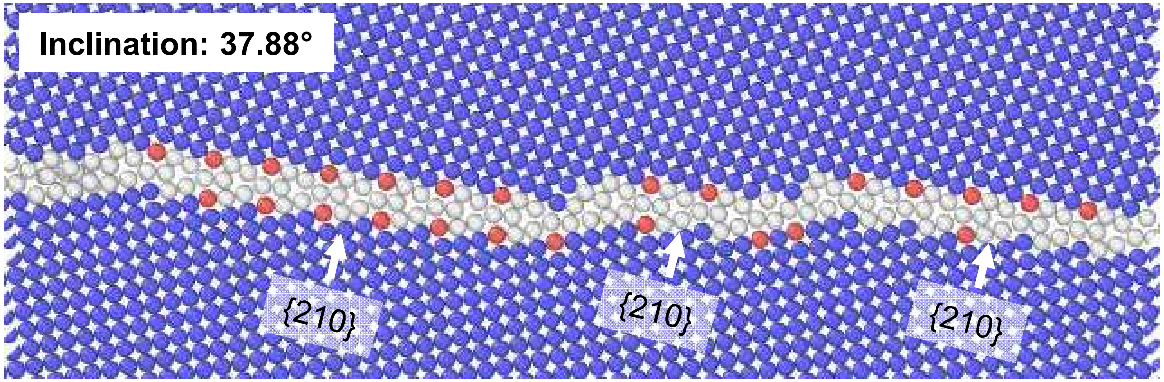


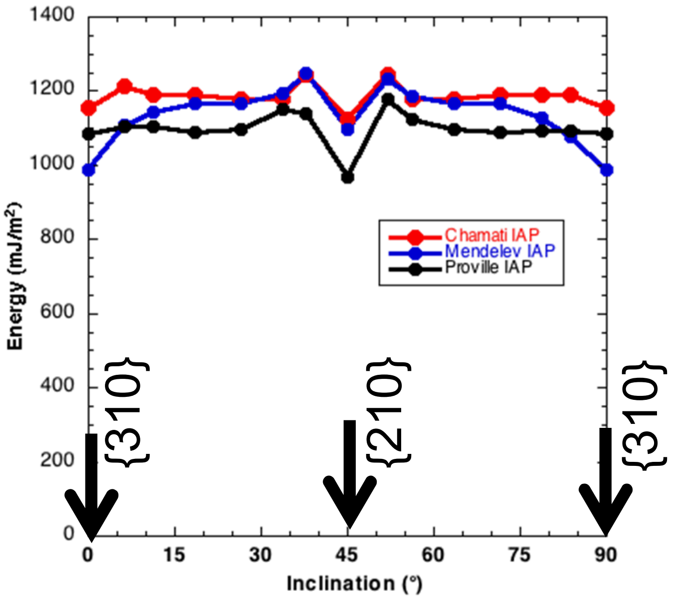
$\Delta y = 0.035 \pm 0.015\ a_0$

What about the junction structure? MD simulations









Surprisingly, the atomistics show {110}/{170} facets rather than coexisting {310} and {210} facets at intermediate inclinations.

-Insufficient kinetics for {310} and {210} facets to develop?

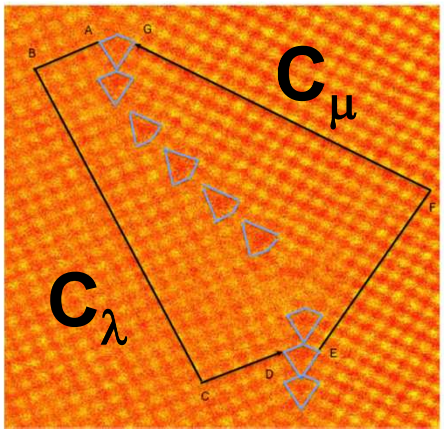
-Connection to grain boundary dislocations and degree of misorientation?

Atoms shaded by CNA

Are Grain Boundary Dislocations Present?

Boundary is misoriented from exact Σ=5 ($\Delta\theta$=-2.38°)

Determine defect content by Circuit Mapping over all facet junctions



Path in μ crystal

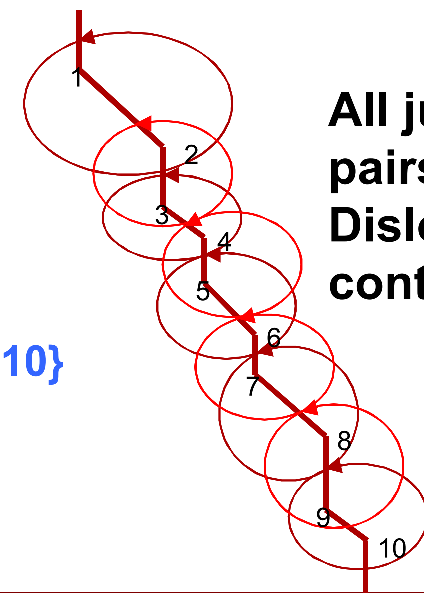
Path in λ crystal

$$\mathbf{b} = -(C_\lambda + \mathbf{P}C_\mu)$$

Burgers vector

Re-express μ path in λ crystal coordinates.

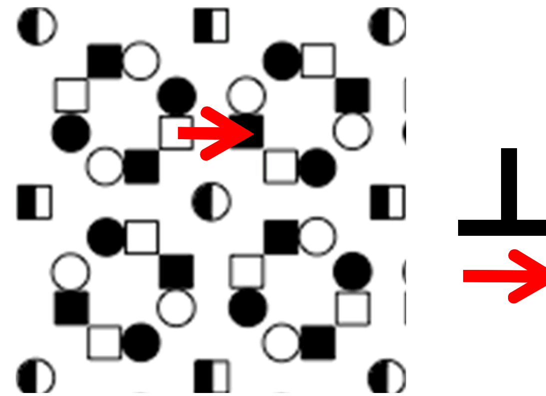
-Circuits must cross at equivalent GB sites
-Every circuit then includes 2 junctions.
-Alternate between circuits on {210} and {310} inclinations



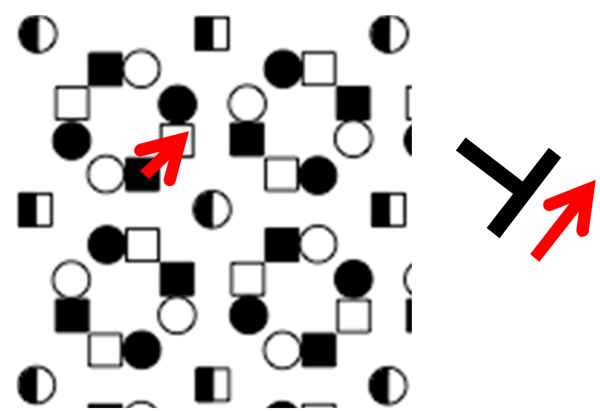
All junction pairs exhibited Dislocation content

Two types of defect observed:

b=(1/5)[3,1,0]

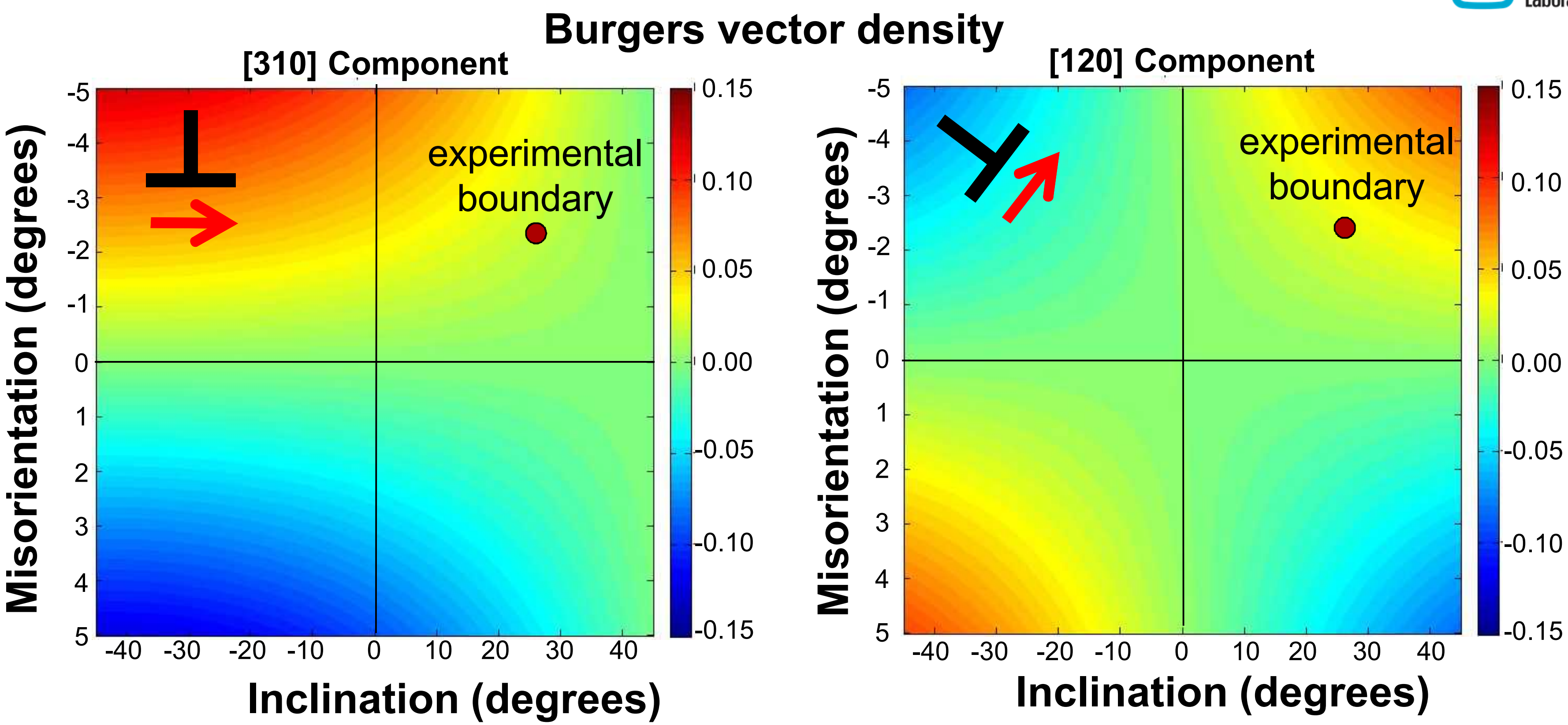


b=(1/5)[1,2,0]



Defect content tied to misorientation and inclination

Sandia National Laboratories



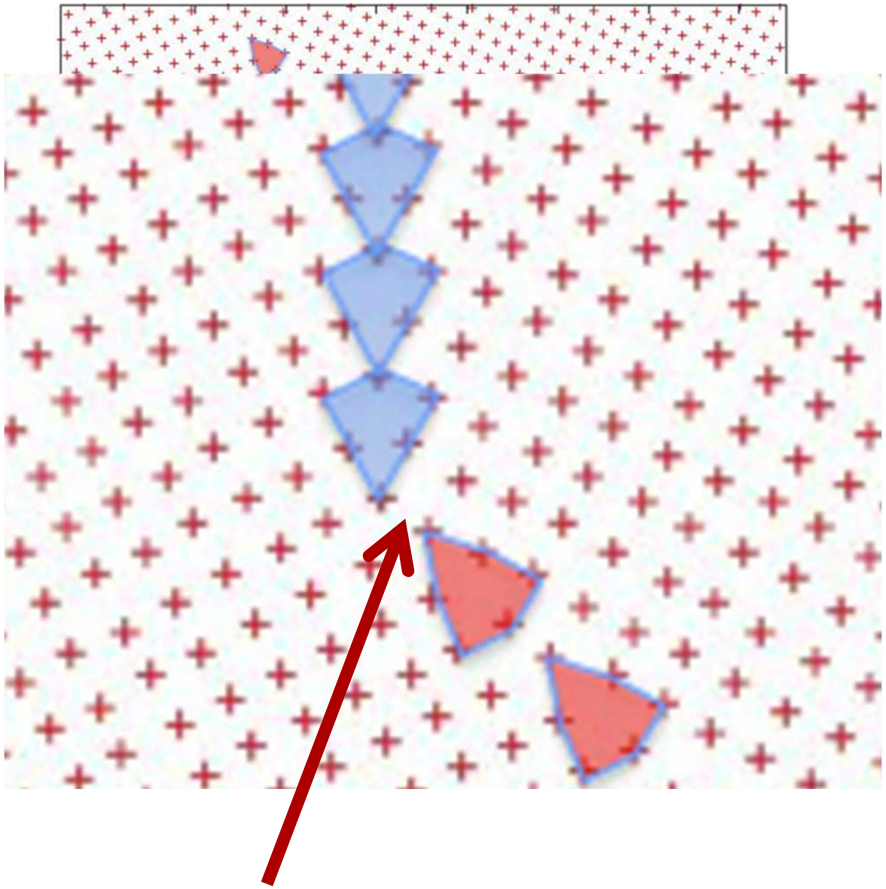
- **Burgers vector density related to misorientation and inclination through Frank-Bilby Equation: $B=(I-P^{-1})\, v$**

Experimental		Frank-Bilby equation (θ=-2.38°±0.8°,ϕ=25.9±1.0°	
<310> component:	0.0323	<310> component:	0.0180 ±0.006
<120> component:	0.0152	<120> component:	0.027 ± 0.010

- **For inclinations away from {310}, b_{120} component required to accommodate interfacial coherency strains.**

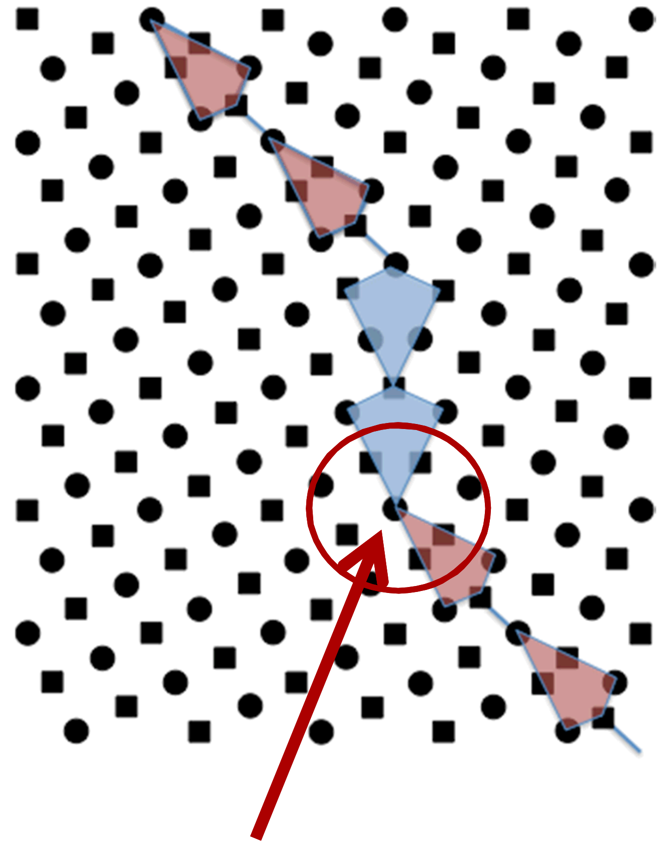
How are the grain boundary dislocations manifested in the junction structure?

Experimental Junctions
b=(1/5)(120) and (1/5)(310)



Kites Offset

Geometric construction for junctions with b=0

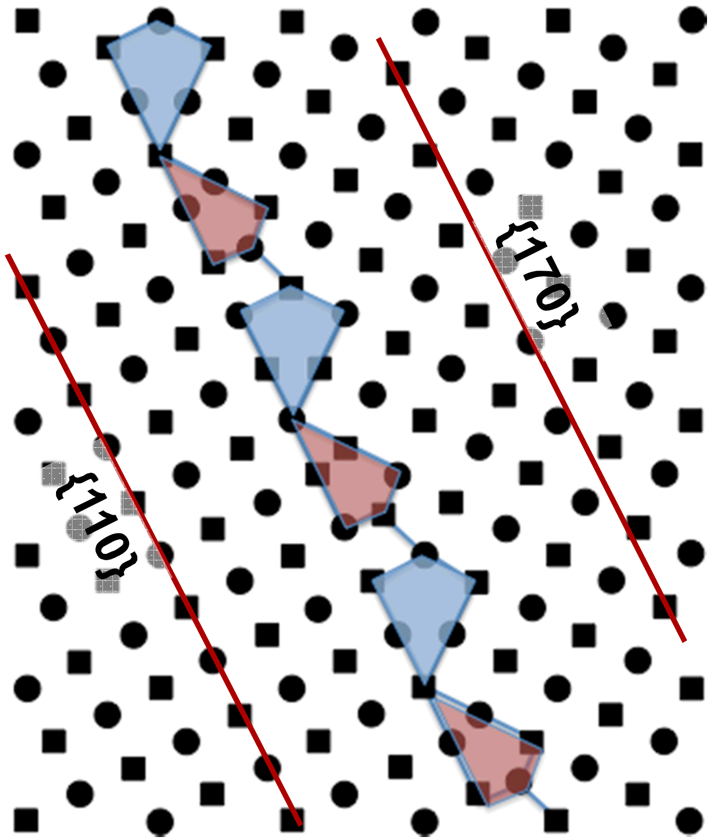


Kites Joined

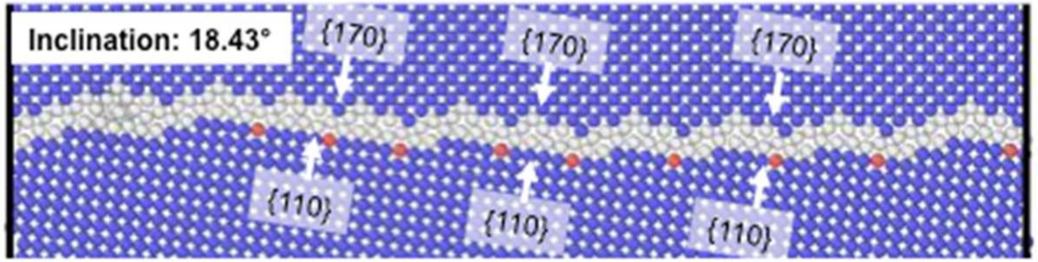
Analysis helps explain the unexpected {170}/{110} facets found in atomistic simulations:

Sandia National Laboratories

Geometric construction (b=0)
{310}:{120} = 1:1



Example from simulated annealing



Adjacent {310} and {120} kites give {110}/{170} facets in absence of grain boundary dislocations.

Consistent with hypothesis that the simulated structures have not yet produced coarsened facet structures.

Next steps:
-Relax geometric constructions with and without grain boundary dislocations

Future directions and challenges

Sandia National Laboratories

- **The Σ=5 Fe boundary illustrates the complex interplay between atomic structure and macroscopic geometrical parameters of grain boundaries.**

- **Some challenges moving forward:**

-Linking to GB properties at larger length-scales
-Extending across GB orientation space.

Research Thrusts for FY15

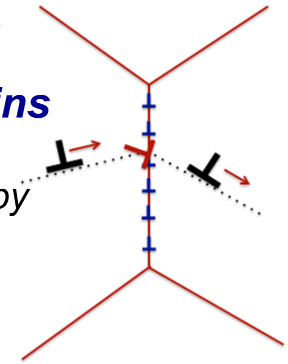
Statistical variation of GB structure and defect arrangements
-PLD Fe films as a "model" material.
-Convenient columnar grain structure.
-higher-throughput HRSTEM analysis procedures.
-New precession diffraction/ACOM capability.
-*in situ* straining to explore defect interactions.

Implementing elasticity-based model for grain boundary dislocation arrays
-Validation through *in situ* and post-mortem characterization
-Initial focus on dislocation/twin interactions in 304L stainless steel.

Our work will begin addressing mechanisms of slip transmission

Transmission controlled by:

-Orientation of adjacent grains
-Interface compatibility stresses due to elastic and plastic anisotropy
-Alignment of slip planes and interface inclination.
-Conservation of ***b****.*



-Interfacial structure
*-**short range** stress fields from intrinsic GBDs*
*-**long range** stress fields from extrinsic defects not yet incorporated into the intrinsic GB structure.*
*-**Reconfigurability** of interface to absorption and emission of dislocations.*

Sensitive to the discrete, atomistic details of interface

New Precession Electron Diffraction Capability

Orientation imaging in TEM:

-Coupling of orientation imaging with complementary TEM methodologies.

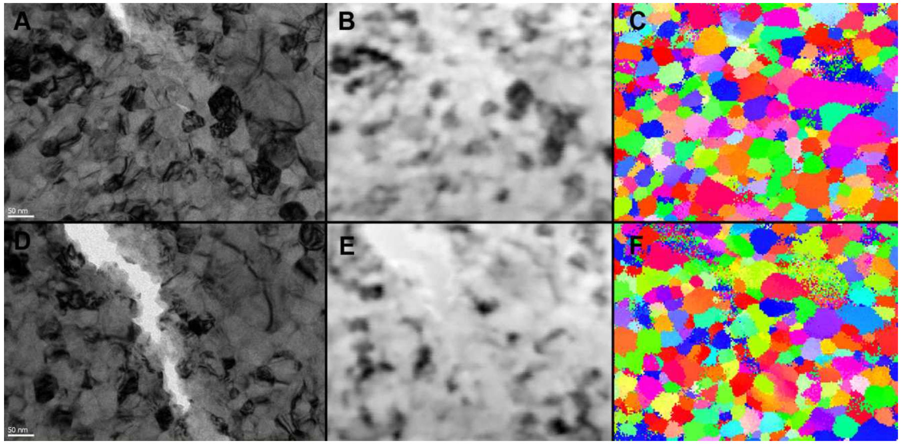
-Track orientational changes during *in situ* straining.

-Relate local orientation distributions to dislocation mechanisms through TEM diffraction contrast imaging.

-Screen and identify GB regions for follow-on atomic and strain-constrast imaging, assessing statistical distributions.

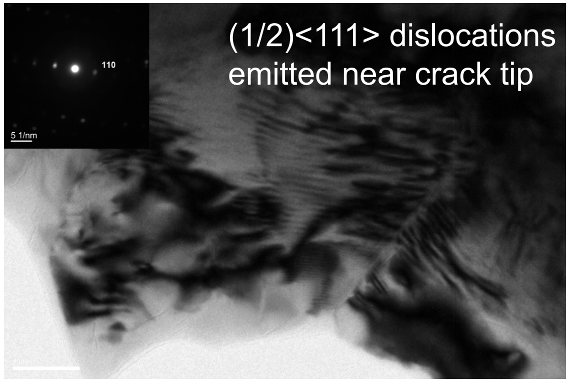
Status:
-Hardware installed (JEOL 2100 TEM)
-Awaiting final software configuration (expected early CY 2015)

Example: Crack formation during *in situ* straining of BCC Fe film



Vetterick, Marshall, Baldwin, Misra, Hattar, Taheri (2014, in prep)

Collaboration between Sandia, LANL, Drexel



GB dislocation arrays: interactions and strain distributions

Model and Characterize strain distributions resulting from arrays of intrinsic and extrinsic grain boundary dislocations.

-Geometric constructions for relating dislocation content to misorientation and inclination.

-Develop solutions for near and far-field elastic strain fields.

-Build off pioneering work by Forwood & Clarebrough

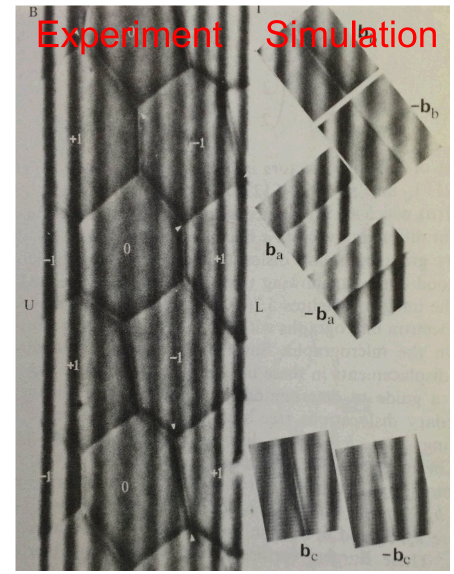
-Input to diffraction contrast simulations for comparison with S/TEM diffraction contrast simulations.

-Public domain codes: M. DeGraef CTEMsoft

-Initial focus on twin/dislocation interactions in 304L.

-static observation and *in situ* straining.

Example: Observation and simulation of GBD strain contrast in TEM



Forwood & Clarebrough 1991

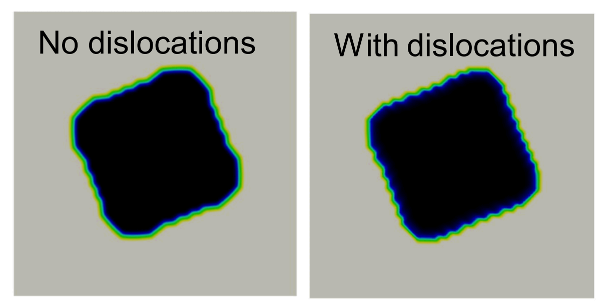
Develop Mesoscale Phase Field approaches to link dislocation content to developing GB morphology

$$\mathcal{F}_1 = \int d\mathbf{r} \left[f(\phi) + \frac{1}{2}|\kappa(\theta)\nabla\phi|^2 + f_{coup}(\rho_{ij}, \phi, \Gamma) \right]$$

Bulk

Interface energy + anisotropy

Coupling to
1) **dislocation density tensor:** ρ_{ij}
2) **elastic fields (Airy stress function):** Γ



Final Comments

Linking interfaces to materials performance variability will require that they be treated much more deeply than simply as geometric dividing surfaces.

Example of the Fe Σ=5 grain boundary illustrates the complex interplay between interfacial energy and crystallography, defect structure, and morphology.

A key challenge is in understanding how this atomistic scale interfacial complexity connects up to fundamental interfacial properties.

Interfacial line defects provide the natural elementary building blocks to bridge between atomistic and continuum interface descriptions.

Extra

Local Translational Symmetry Segmentation Algorithm:

Sandia National Laboratories

Approach:

(1) At each pixel, compute cross-correlation of local image region with its immediate surroundings (within ~2 nearest neighbor distanc

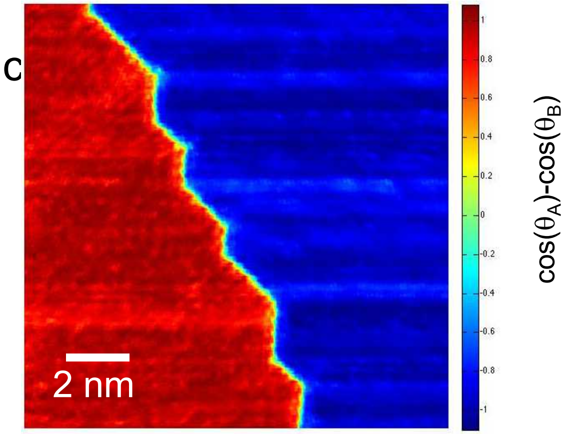
> ***Why?*** This provides an measure of the local translational symmetry and orientation that is invariant at all points within an undistorted crystal (i.e., constant regardless of whether pixel is on or off an atomic column)

(2) Compare the cross-correlation image with templates obtained from cross-correlation images averaged over region of bulk crystal on either side of interface.

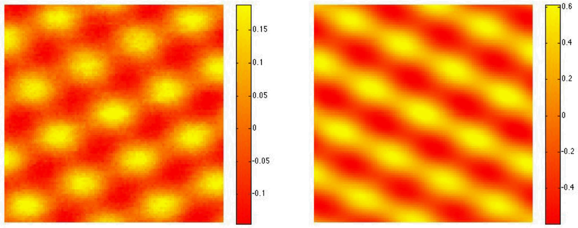
(3) Determine similarity, pixel-by-pixel, by computing angle, θ, between n-dimensional vectors for the reference correlation image templates and the correlation image of the raw image.

$$\cos(\theta_A) = \frac{P \bullet P_A}{\|P\| \|P_A\|} \qquad \cos(\theta_B) = \frac{P \bullet P_B}{\|P\| \|P_B\|}$$

Facets at Σ=5 Fe GB

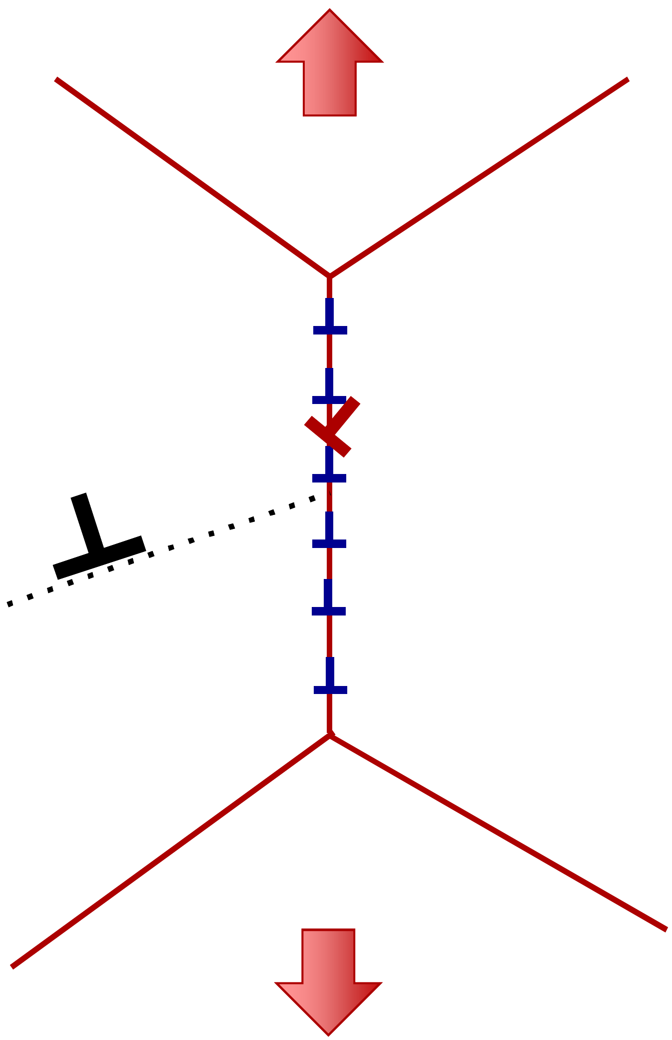


Reference Cross-Correlation Templates



-cross-correlate central 32x32 pixel region with outer 128x128 region
-average over reference regions in left and right grains.

Dislocation-Grain Boundary Interactions



Peach-Koehler Equation:

$$\boldsymbol{F}/L = (\boldsymbol{b} \cdot \boldsymbol{\sigma}) \times \boldsymbol{\xi}$$

Contributions to stress on dislocation:

$\sigma_{applied}$: macroscopic loading

$\sigma_{compatibility}$: anisotropic elastic and plastic strain in abutting crystals

$\sigma_{GB\text{-}intrinsic}$: short-range strain field due to intrinsic periodic GB dislocation structure.

$\sigma_{GB\text{-}extrinsic}$: long-range strain field from defects not yet incorporated into intrinsic GB structure.

Controlled by discrete, atomistic details of interface