Atomistic Exploration of Impurity Effects on the Intrinsic Brittleness of Silicon Carbide

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Project Overview

Timeline:

3 year project - initiated September 15, 2011

Primary Objective:

Utilize advanced atomistic modeling techniques to investigate the role of impurity elements and nonstoichiometry on high temperature SiC crack tip behavior

Potential Outcomes:

1. Improve understanding of fracture mechanisms in SiC
2. Identify impurity elements that are influential to crack growth in SiC
3. Provide guidance in choosing crack tip constitutive relations
4. Develop a foundation for future atomic-scale investigations of fracture in chemically complex materials

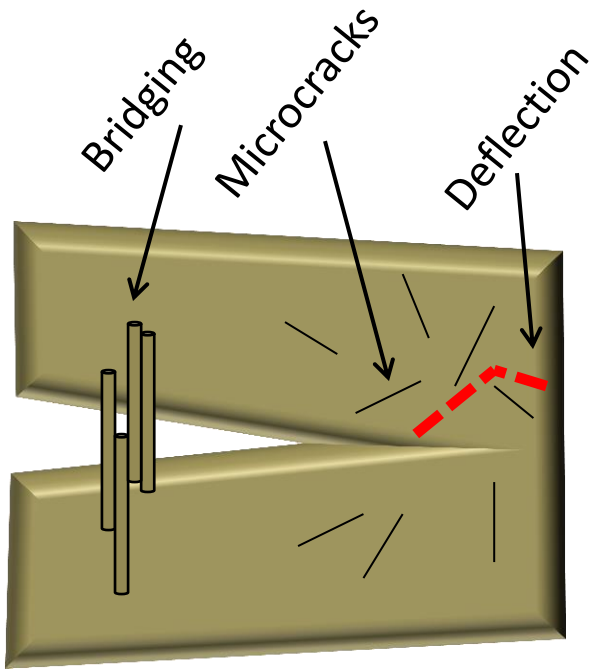
Context

Why silicon carbide?

→ Large literature

→ Significant technological motivation

- High-T structural applications
- High-T electronic applications



Fracture mechanisms in silicon carbide

→ Improve crack growth resistance via microscopic mechanisms

→ Atomic-scale mechanisms can serve as a “valve” controlling microscopic mechanisms

Approach

1. **Identify key crack tip mechanisms and important regions in configuration space**
   - Large 3D atomistic simulations
   - Penny shaped cracks with empirical potentials
   - Investigate crystallography, crack tip geometry, loading type, & temperature
2. **Examine key mechanisms and configurations more carefully** with regard to spatial scale, time scale, interatomic potentials
   - Concurrently coupled atomistic - discrete dislocation framework
   - *Specific mechanisms*: Variational transition state theory via finite temperature string method
   - *Specific configurations:* Parallel replica + hyperdynamics
3. **Examine the effect of impurities and nonstoichiometry** on the key mechanisms and configurations
   - QM-based continuum models

Some Experimental and Simulation Results from Literature

- **Amelinckx 1960: 6H SiC**
  - Glide traces as a result of plastic flow with optical microscopy and x-ray diffraction microscopy
- **Pilyankevich 1984: 6H SiC**
  - Partial dislocations and stacking fault with TEM after high-temperature plastic deformation

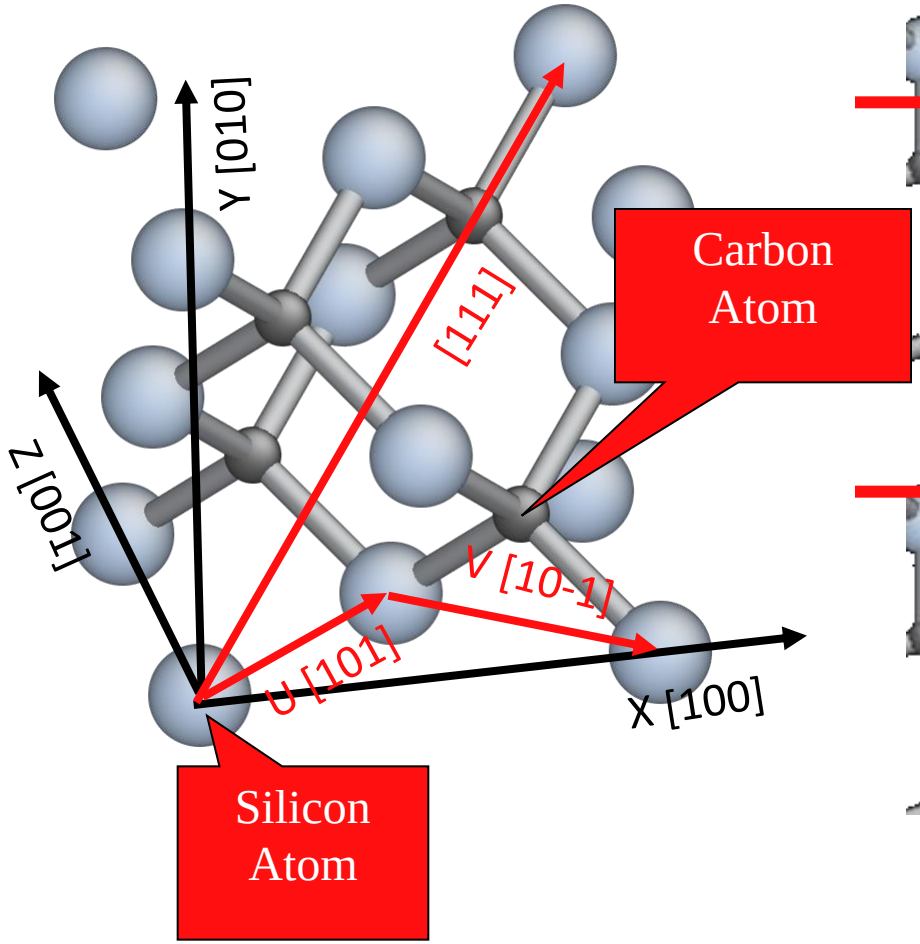
- **Henshall 1977: 6H SiC**
  - γs=11.5 J/m2 below 1000 K and γs=25 J/m2 above 1400 K obtained from 3-point bend tests on single edge-notched beam specimens
  - Fracture surfaces with plastic deformation, with large cleavage steps
- **Campbell 1989: sintered and hot pressed SiC**
  - Below 1700 K, the material fails by brittle crack extension. Above 1700 K, the material become ductile
- **Pirouz 1999: 6H and 4H SiC**
  - (T < Tc) deformation proceeds by the nucleation and glide of single partial dislocations
  - (T > Tc) deformation proceeds by the formation and glide of perfect, dissociated, dislocations

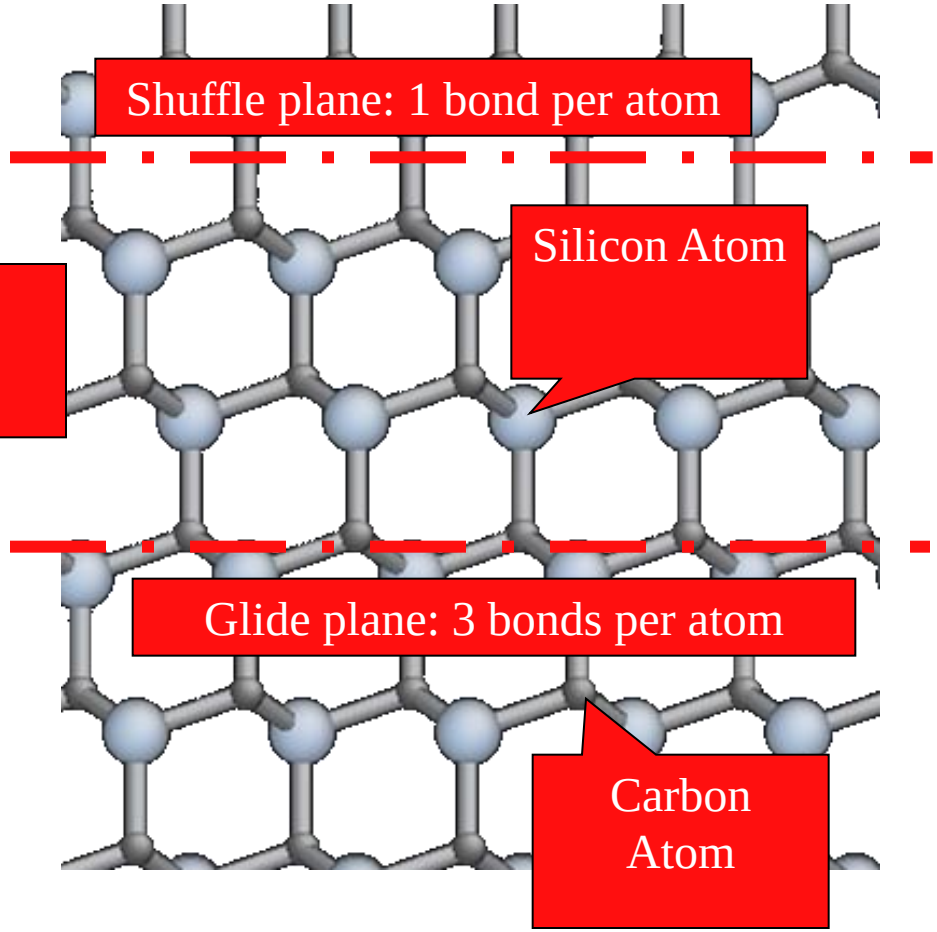
- **Kikuchi 2005: 3C SiC**
  - MD simulation 100 K, 111 fracture involves slip in the 111 planes
  - γs=2.1 J/m2

Presentation Outline

- **Part I:** Atomistic model
  - Direct Molecular dynamics (MD) simulation of SiC crack tip
- **Part II:** Analytic continuum model
  - Competition between crack blunting by plastic deformation and crack propagation
  - Griffith's fracture model [1]: strain energy release rate for fracture
  - Rice's dislocation model [2]: strain energy release rate for dislocation nucleation
  - Dislocation nucleation as a function of temperature and strain rate
  - Calculate elastic constants, surface energies and unstable stacking fault energies
- **Part III:** Compare the direct MD simulation results to the continuum fracture/dislocation model

Silicon Carbide Terminology

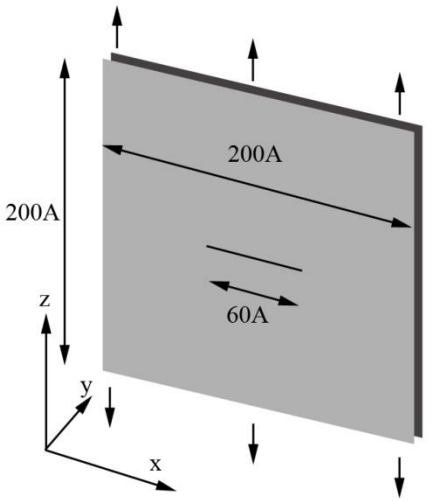




Part I:
Molecular dynamics Simulations

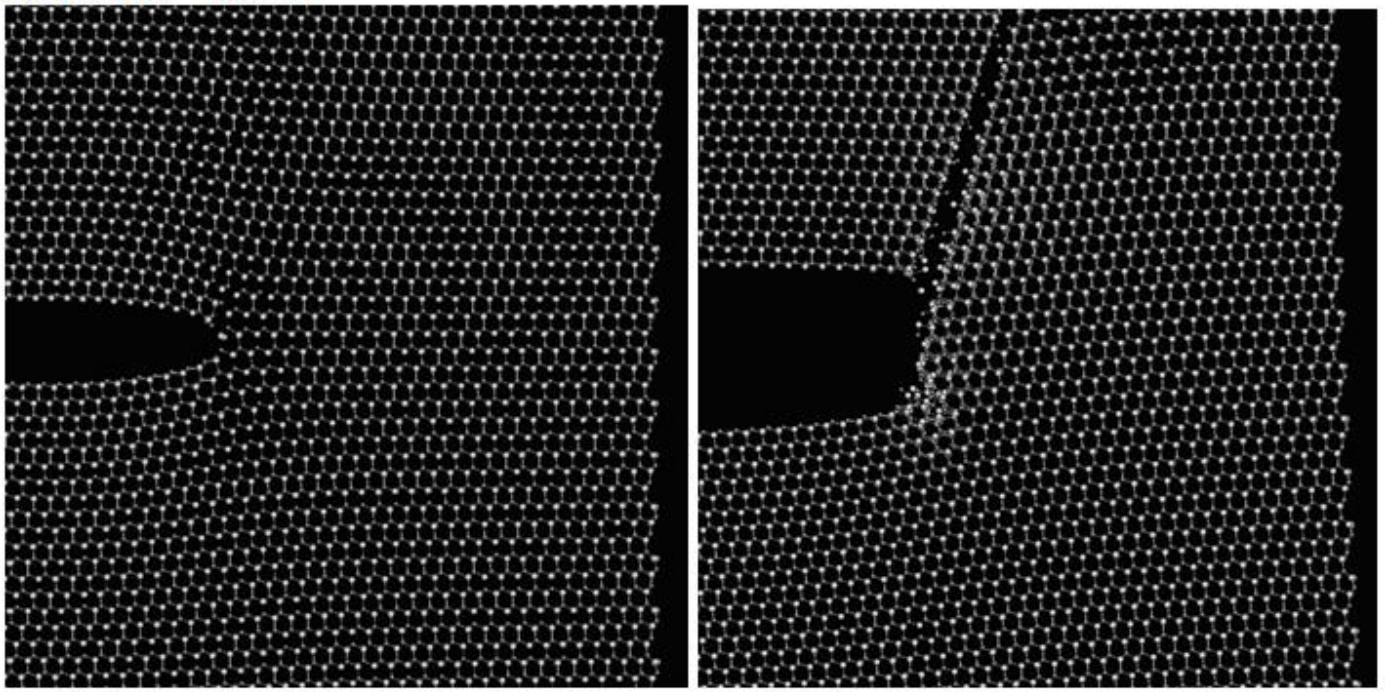
Simulation Setup (Traditional MD)

- Mode I loading through crack simulations conducted with LAMMPS package
- 200A x 200A x 20A (x x z x y) with a 60A through crack
- Traction free boundary condition was applied in x and y direction and mode I tensile loading was applied in the z direction, corresponding to the <111> direction of the crystal
- Potentials:
  - Devanathan Tersoff potential
  - Erhart Tersoff potential
- Temperature: 0 K to 800 K with 200 K increment
- Strain rate: 10^7/s to 10^9/s



MD Results

Erhart potential
Temperature = 0 K
Strain rate = 1e7 /s

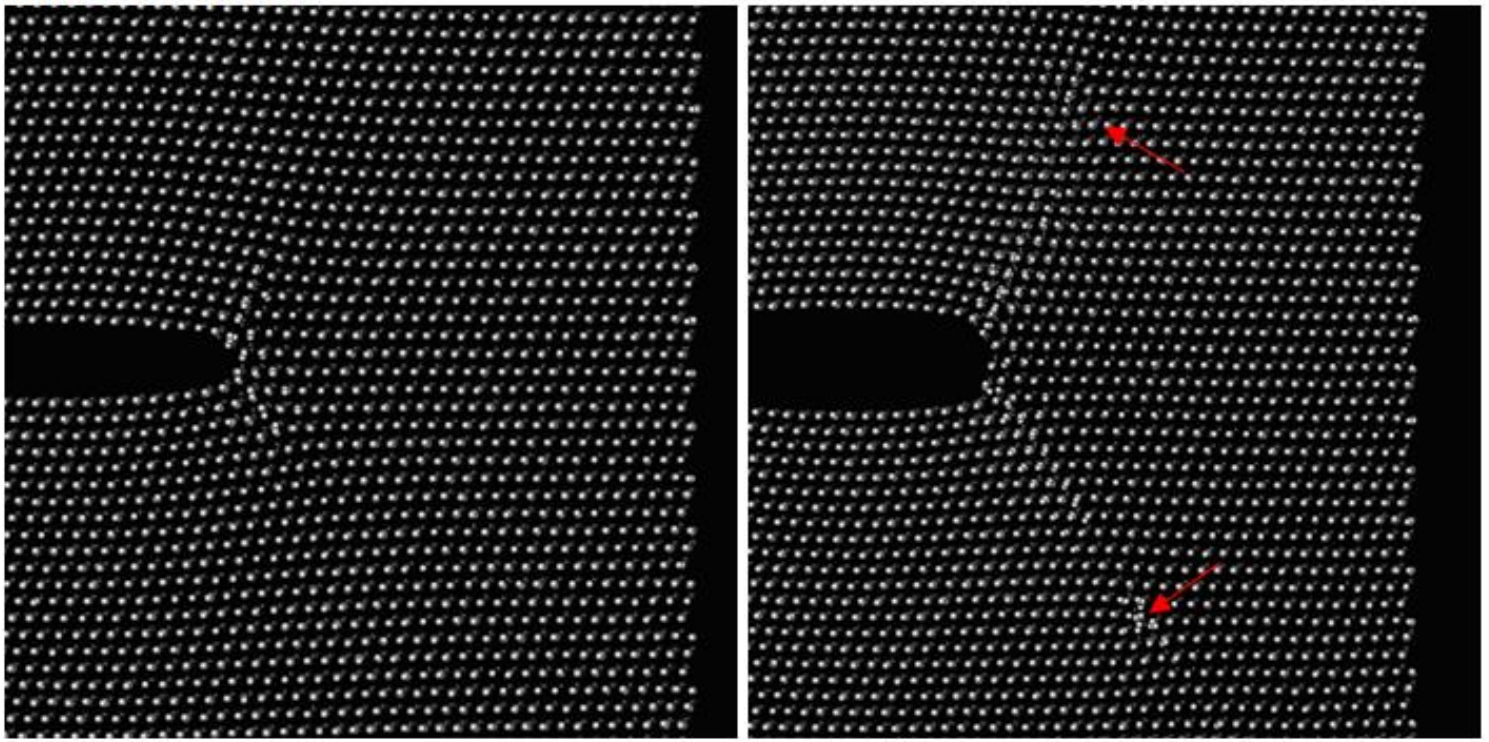


ε = 0.047 ε = 0.063

Crack does not have ductile response. Crack initiates on {111} plane perpendicular to loading direction and continues to propagate on the inclined {111} planes.

MD Results

Erhart potential
Temperature = 400 K
Strain rate = 1e7 /s

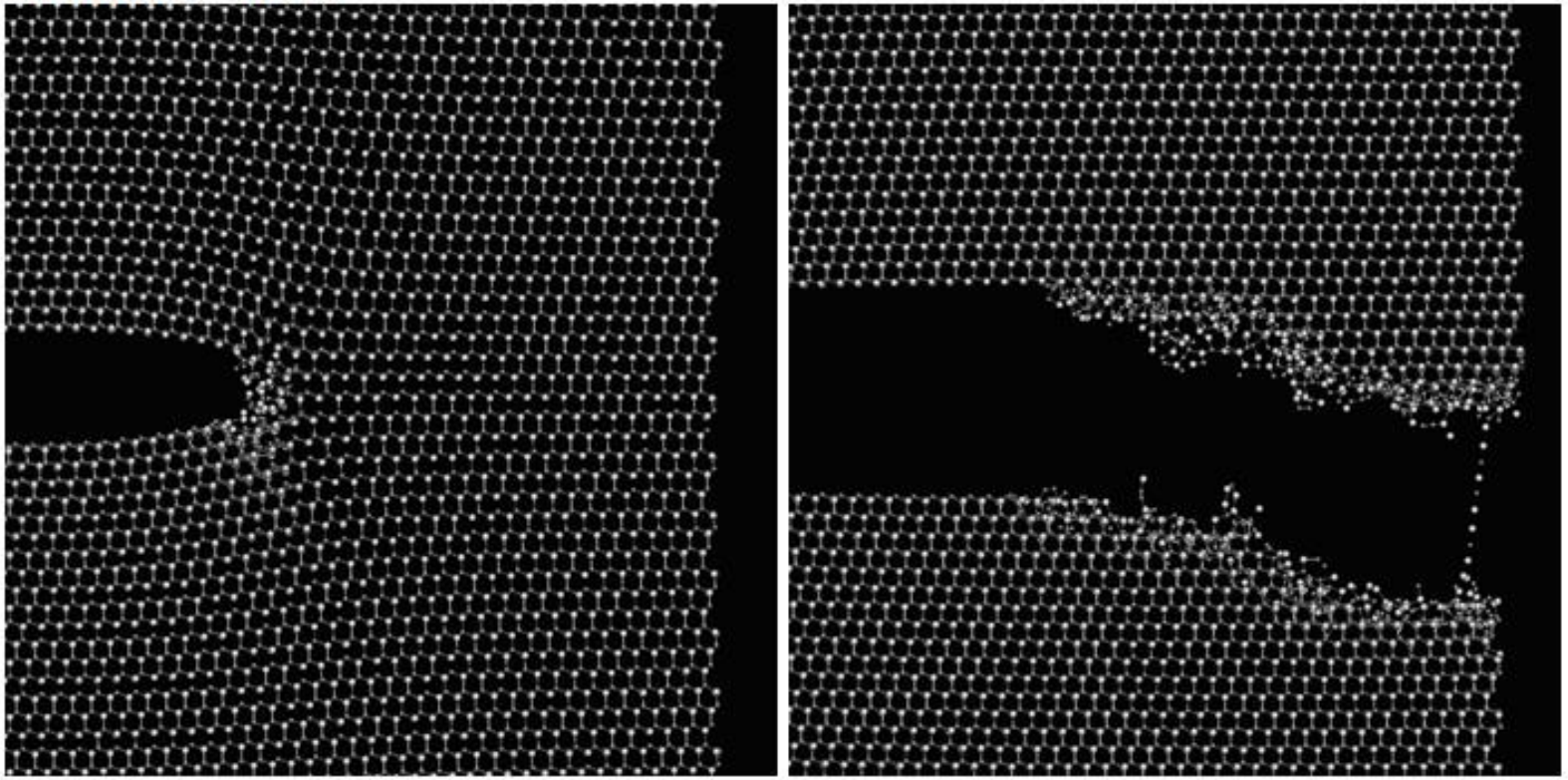


ε = 0.039 ε = 0.055

Ductile crack response by nucleation of dislocations on {111} and {100} planes. Leading partial and trailing partial are formed on {111} plane. Full {100} dislocation is mobile at 400 K. No twinning is observed.

MD Results

Devanathan potential
Temperature = 0 K
Strain rate = 1e7 /s

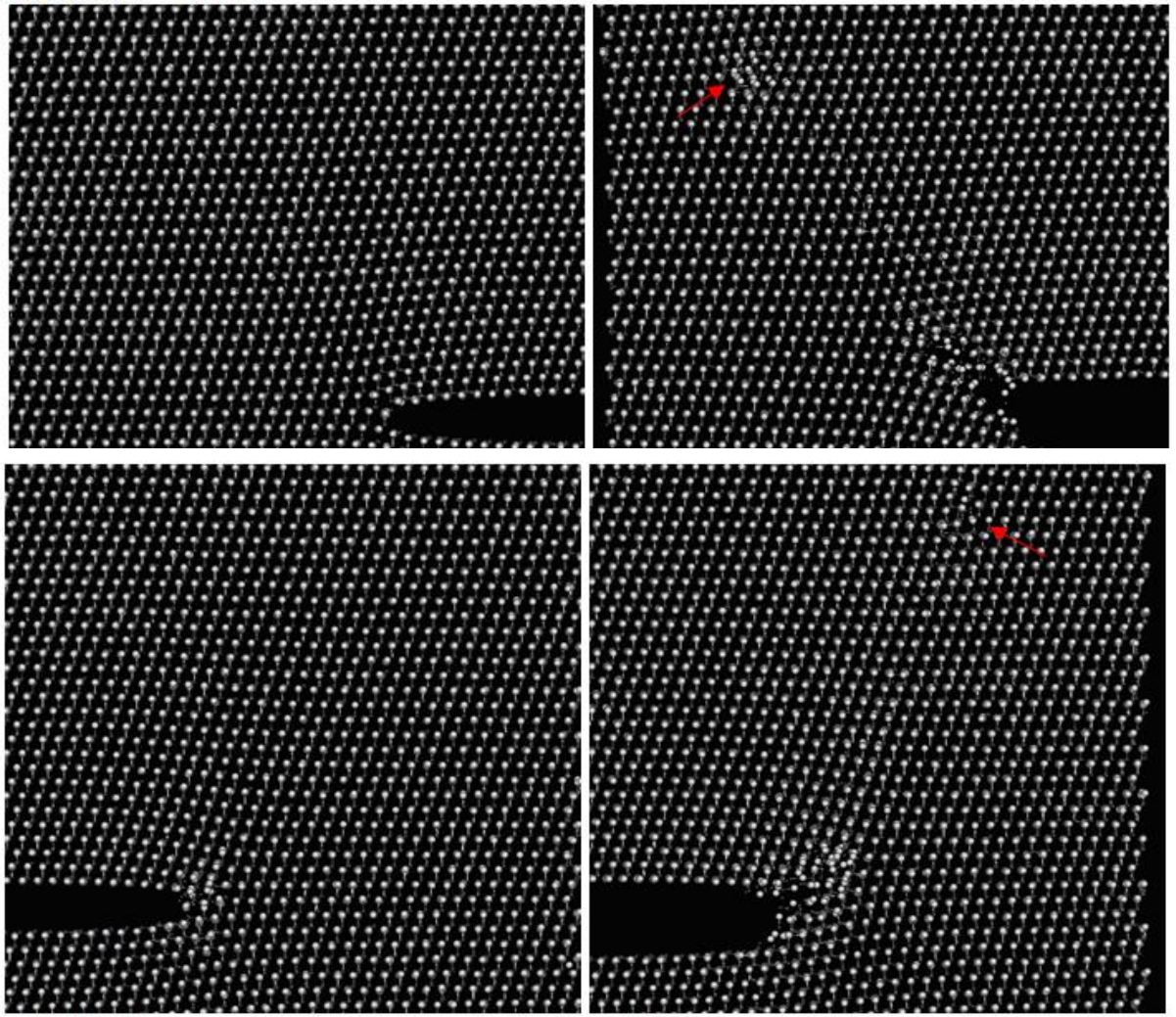


ε = 0.056 ε = 0.068

Brittle crack initiates on {111} plane and continues to grow catastrophically

MD Results

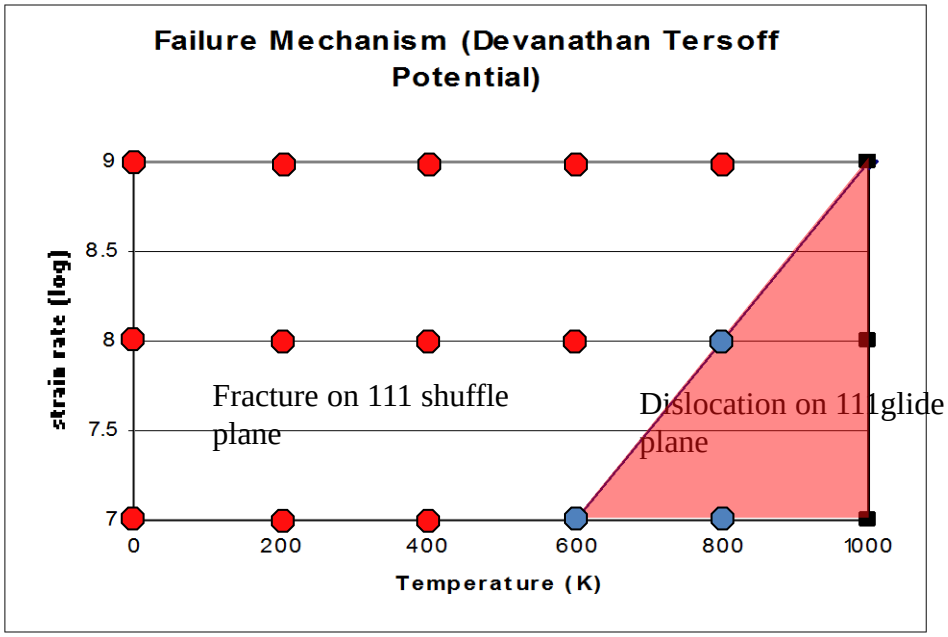
Devanathan potential
Temperature = 1000 K
Strain rate = 1e7 /s

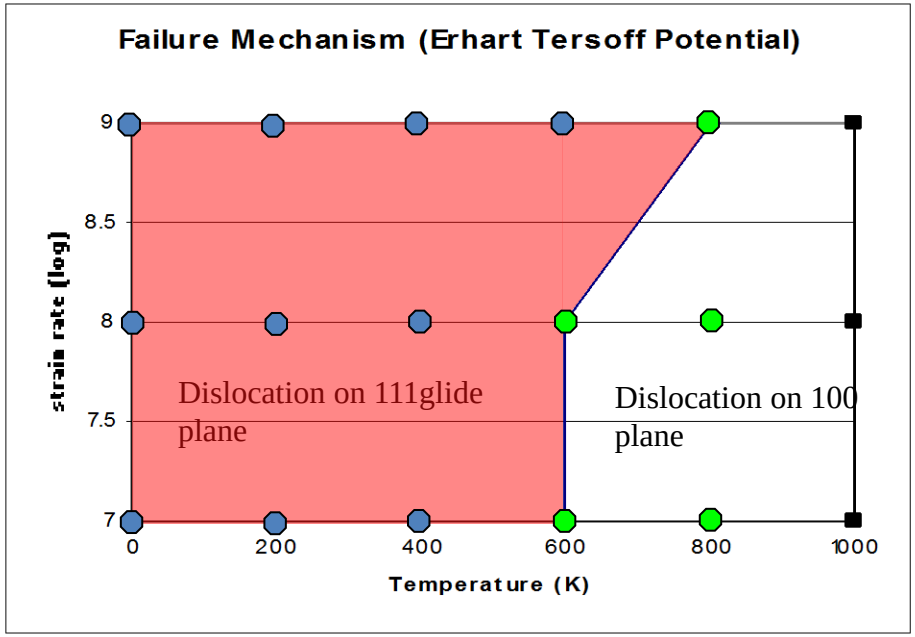


ε = 0.028 ε = 0.036

Dislocation nucleates on both {111} and {100} planes.

MD Results

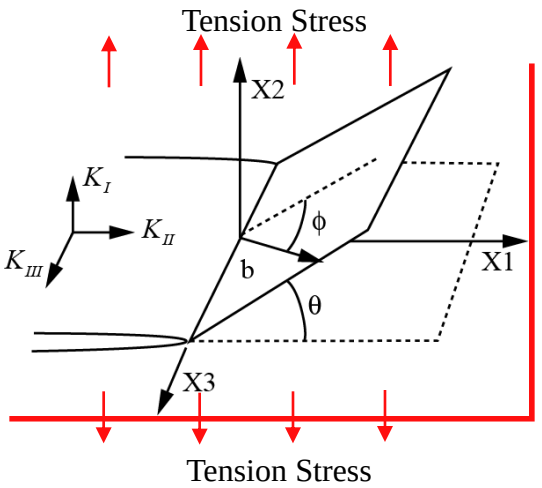

Failure Mechanism (Devanathan Tersoff Potential)
strain rate (log)
9
8.5
8
7.5
7
0
200
400
600
800
1000
Temperature (K)
Fracture on 111 shuffle plane
Dislocation on 111glide plane


Failure Mechanism (Erhart Tersoff Potential)
strain rate (log)
9
8.5
8
7.5
7
0
200
400
600
800
1000
Temperature (K)
Dislocation on 111glide plane
Dislocation on 100 plane

Part II:
Analytic Continuum Model

Continuum Modeling

	Mode I	Mode II
fracture	■ Consider a mode I crack, if the strain energy release rate is greater than the surface energy, the crack will propagate $G_I = (1-\upsilon)K_I^2 / 2\mu > 2\gamma_s$	■ Consider a mode II crack, new surface is formed when Griffith criterion for fracture is fulfilled: GI > g(γs) $G_{II} = (1-\upsilon)K_{II}^2 / 2\mu > 2\gamma_s$
dislocation	■ For mode I crack loading, the dislocation nucleation criterion GI > g(γus) $G_I = \frac{(1-\upsilon)K_I^2}{2\mu} > 8\frac{1+(1-\upsilon)\tan^2\phi}{(1+\cos\theta)\sin^2\theta}\gamma_{us}$	■ If the strain energy release rate is greater than the unstable stacking fault energy, dislocation will nucleate: GII > g(γus) $G_{II} = (1-\upsilon)K_{II}^2 / 2\mu > 2\gamma_{us}$



Strain Rate and Temperature Dependence

- Activation energy for nucleating a dislocation in 3 dimension [1]

$$Q_{3D} = 5b_p{}^3 0.287\mu F \frac{(\cos^2\varphi + (1-\nu)\sin^2\varphi)}{(1-\nu)}\left(1 - \frac{K_{II}^{eff}}{K_{IIcrit}^{eff}}\right)^{\frac{3}{2}}$$

- The effective KII stress intensity factor is given by the applied KI and the geometry

$$K_{II}^{eff} = K_I \cos\phi \cos^2\frac{\theta}{2}\sin\frac{\theta}{2}$$

- The critical KII stress intensity factor is a function of the elastic constants and the unstable stacking fault energy

$$K_{IIcrit}^{eff} = \sqrt{\frac{2\mu}{1-\nu}(\cos^2\varphi + (1-\nu)\sin^2\varphi)\gamma_{us}}$$

- Finally, the critical KI stress intensity factor for dislocation nucleation can be solved numerically at given temperature and strain rate [2]

$$\frac{Q_{3D}}{k_B T} = \ln\left(\frac{k_B T N \omega_0}{-\dot{K}_I \frac{dQ_{3D}}{dK_I}}\right)$$

- Analytic continuum fracture and dislocation model:
  - Elastic constants

    $C_{11}(T)$, $C_{12}(T)$, $C_{44}(T)$
  - Surface energies

    $\gamma_s(T)$
  - Unstable Stacking fault energies

    $\gamma_{us}(T)$

[1] J Rice, 1994
[2] Zhu. et al., 2008

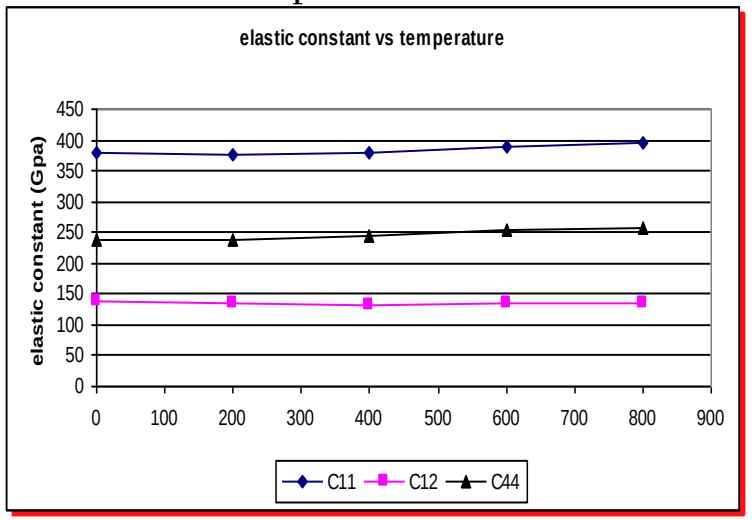
Elastic Constants

- Elastic constants input for continuum model
- Direct calculation of elastic constants with Lammps
- 0K elastic constants compared to experiments

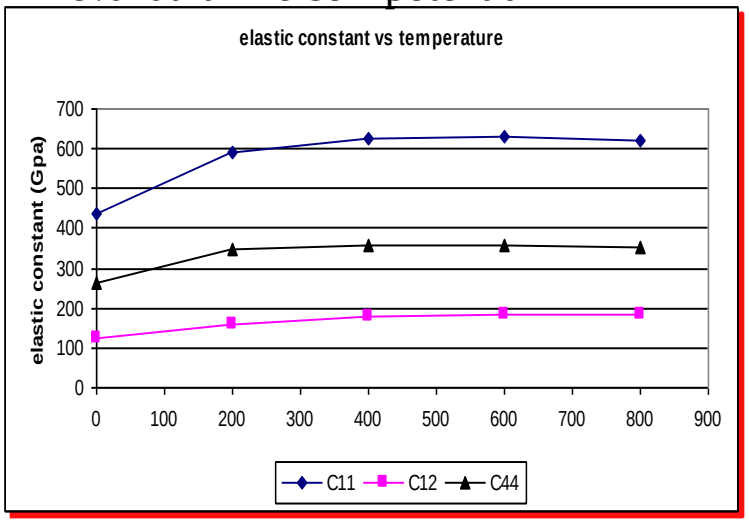
	Experiment [1]	Experiment [2]	This work Erhart	This work Devanathan
a0	4.3596		4.3596	4.2799
C11	390	352	380	437
C12	142	140	138	126
C44	256	233	239	265

- Elastic constants as a function of temperature with NPT
- Elastic constants increase with temperature for Devanathan Tersoff potential, also observed in [3]

Erhart Tersoff potential



Devanathan Tersoff potential



[1] - W. R. L. Lambrecht, 1991d.
[2] - H. Ullmeier, 2001d.
[3] - J. David Schall, 2008

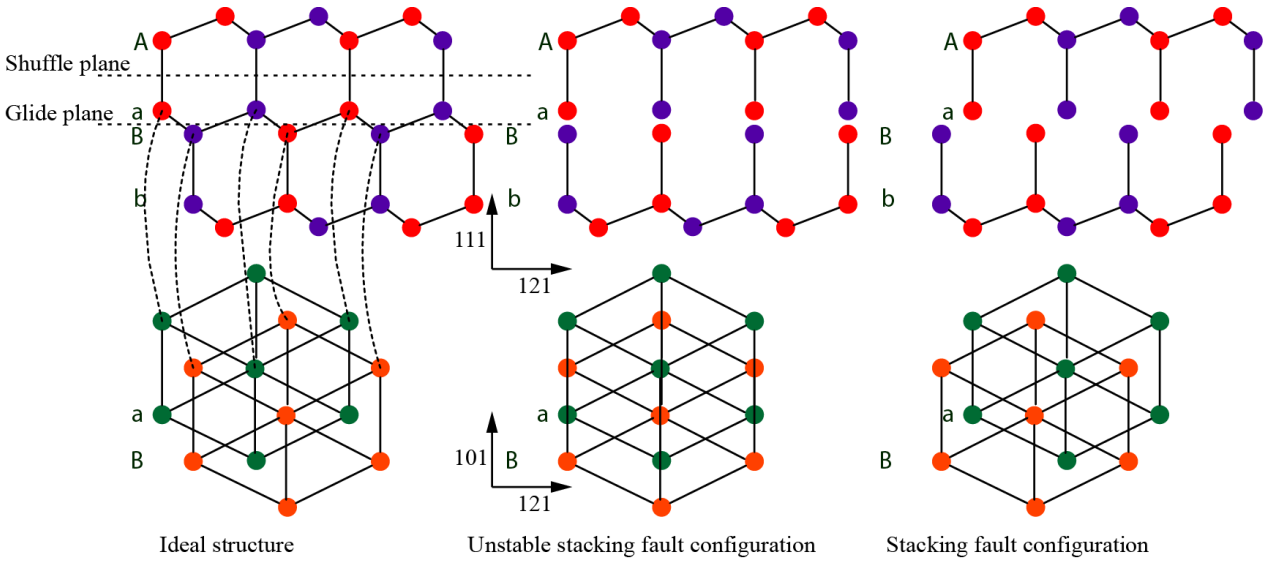
Surface Energies

	T(K)	Devanathan Tersoff Potential energy (J/m2)	Erhart Tersoff Potential energy (J/m2)	DFT Perdew-Zunger (LDA) exch-corr energy (J/m2)
111 glide plane	0.00	15.61	13.86	13.82
	800.00 (enthalpy)	10.17	9.31	-
111 shuffle plane	0.00	6.80	5.08	8.28
	800.00 (enthalpy)	6.40	4.84	-
100 plane	0.00	11.37	9.75	10.10
	800.00 (enthalpy)	9.60	8.73	-

- Surface energies input for fracture criterion
- Direct calculation of surface energies with Lammps
- γ(111 shuffle plane) < γ(100 plane) < γ(111 glide plane)
- 111 shuffle plane is the most preferable fracture plane
- The 111 shuffle plane surface energy computed with DFT is higher

Unstable Stacking Fault Energies

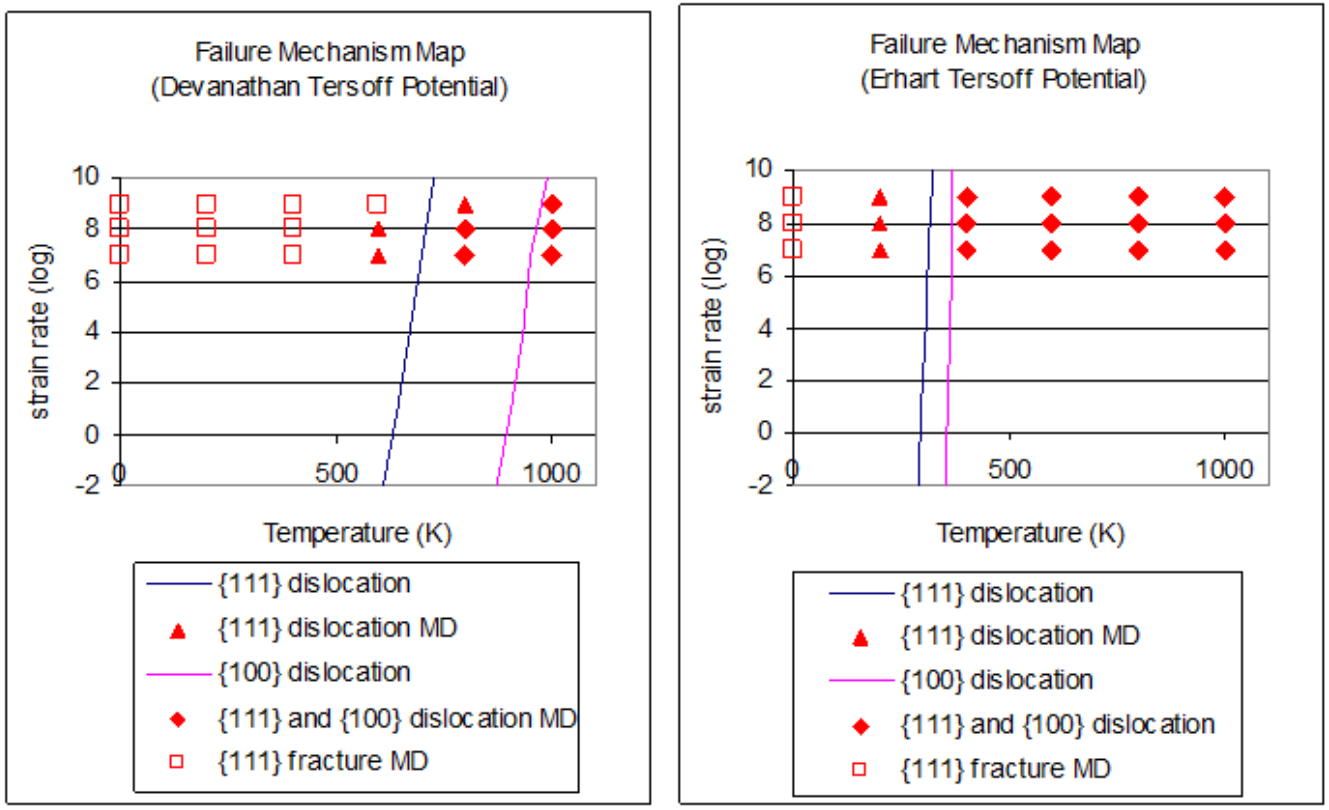
- Input for dislocation criterion
- moving one half of the crystal rigidly in the direction of the Burgers vector
- Most preferable dislocation planes are 111 glide plane and 100 plane for Erhart potential



	T(K)	Devanathan Tersoff Potential energy (J/m2)	Erhart Tersoff Potential energy (J/m2)	DFT TPSS (Tao-Perdew-Staroverov-Scuseria) GGA
111 glide plane (partial)	0.00	6.15	2.82	3.00
	800.00	5.82	2.76	-
111 shuffle plane	0.00	6.72	6.51	4.00
	800.00	6.20	3.93	-
100 u direction	0.00	9.12	7.86	7.57
	800.00	9.03	7.76	-
100 v direction	0.00	12.22	7.86	-
	800.00	8.19	2.18	-

Part III:
MD Model vs Analytic Model

Failure Mechanism Map



Failure mechanism map for Devanathan and Erhart Tersoff potential
The continuum model predicts fracture on the {111} plane to the left of the blue curve. Between the blue and magenta curve, the model predicts dislocation initiates only on {111} glide plane. To the right of the magenta curve, both {111} and {100} dislocations are predicted. Open squares denote fracture in MD simulation. Triangle and diamond denote dislocation on {111} plane and both {111} and {100} planes respectively.

Summary

- Three competing crack tip mechanisms:
  - Cleavage on 111 plane
  - Dislocation nucleation on {111} planes (glide set)
  - Dislocation nucleation on {100} planes
- The competition between these mechanisms was critically examined using atomistic simulation analytic continuum modeling as a function of temperature and strain rate
- Crack tip mechanism map for SiC – cleavage at low temperature and high strain rate and dislocation nucleation at high temperature and low strain rate
- Future directions:
  - Mobility of different types of dislocation vs brittle ductile transition
  - The impurity effect on cleavage and dislocation nucleation

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Overview

Primary Objective: Utilize advanced atomistic modeling techniques to investigate the role of impurity elements and nonstoichiometry on high temperature SiC crack tip behavior

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Initiated September 15, 2011

Potential Outcomes:
1. Improve understanding of fracture mechanisms in SiC
2. Identify impurity elements that are influential to crack growth in SiC
3. Provide guidance in choosing crack tip constitutive relations
4. Develop a foundation for future atomic-scale investigations of fracture in chemically complex materials

Technical Challenges: Four longstanding challenges exist for performing meaningful atomistic simulations of deformation and fracture processes
1. Spatial scaling
2. Temporal scaling
3. Interatomic interactions
4. Vast configuration space

Approach

1. Identify key crack tip mechanisms and important regions in configuration space:
   - Large 3D atomistic simulations
   - Penny shaped cracks with empirical potentials
   - Investigate effects of crystallography, crack tip geometry, loading type, & temperature
2. Examine key mechanisms and configurations more carefully with regard to spatial scale, time scale, interatomic potentials
   - Concurrently coupled atomistic - discrete dislocation framework
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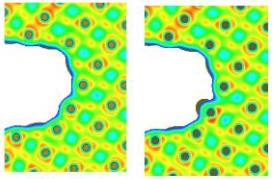
3. Examine the effect of impurities and nonstoichiometry on the key mechanisms and configurations
   - QM-based continuum models

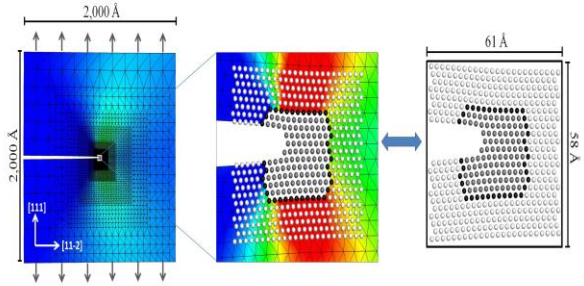
The images to the right show the difference in electron density at an aluminum crack tip when a single hydrogen element is present

Previous Work



Highlighted Tools

Concurrently Coupled Multiscale Simulation



Quantum Mechanics Based Coupled Atomistic Discrete Dislocation Method (QM-CADD)
- Force balance coupling → simple quantum-continuum mechanical coupling
- No ghost forces + quantifiable & controllable coupling errors

Accelerated Timescale Atomistic Simulations
We use several techniques to extend the timescale which our simulation can probe. The below image depicts the hyperdynamics approach