Non-Equilibrium Thermodynamics and Multiscale Modeling of Dynamic Void Growth in Crystals

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Intl. Symp. on Current Problems in Solid Mechanics in honor of

Professor Rodney J. Clifton

June 24th – 29th, 2012, Symi, Greece

Michael Ortiz
RJC 06/12

To Professor Rodney J. Clifton

Scientist, teacher, mentor

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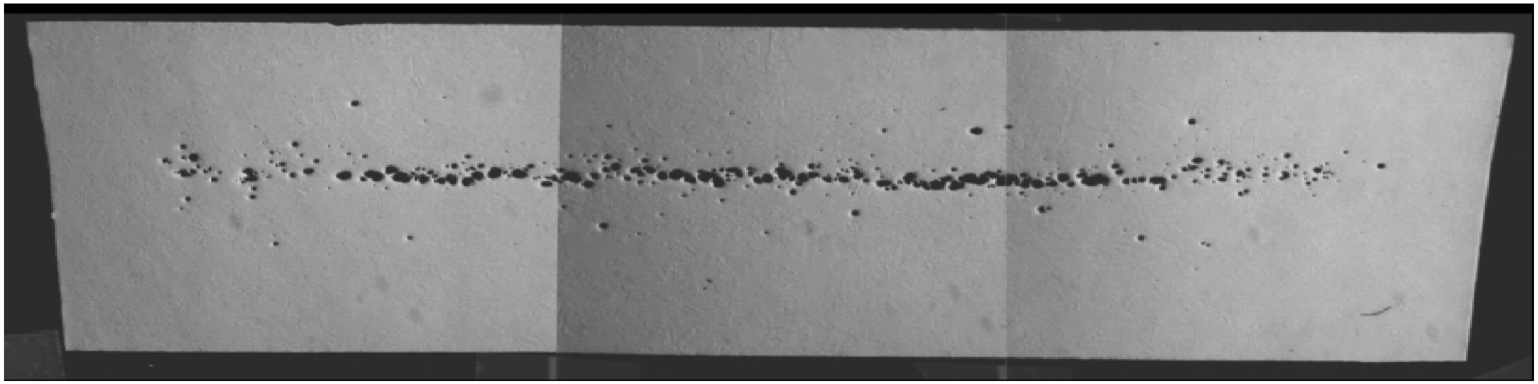
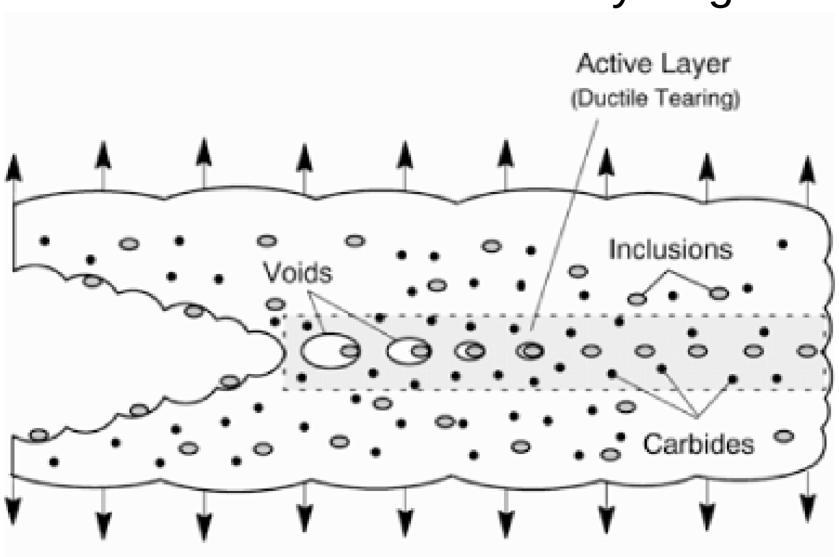
Atomistic-to-continuum - Outline

- The essential difficulty: Multiple scales,
  - *Atomic level rate-limiting processes: Thermal vibrations, lattice defects, transport (thermal, mass…)*
  - *Macroscopic processes of interest: Ductile fracture, GB embrittlement, irradiation damage, aging…*
- Time-scale gap: From molecular dynamics (MD) (femptosecond) to macroscopic (seconds-years)
- Spatial-scale gap: From lattice defects (Angstroms) to macroscopic (mm-m)
- Application to nanovoid plastic cavitation in Cu at intermediate strain rates (e.g., $p_c(a,T,d\varepsilon/dt)$):
  - *Dislocation emission mediates cavitation*
  - *MD limited to extreme strain rates (10^8-10^{12} 1/s)*
  - **Question**: *Intermediate strain rates? (< 10^8 1/s)*

Motivation – Ductile fracture in metals

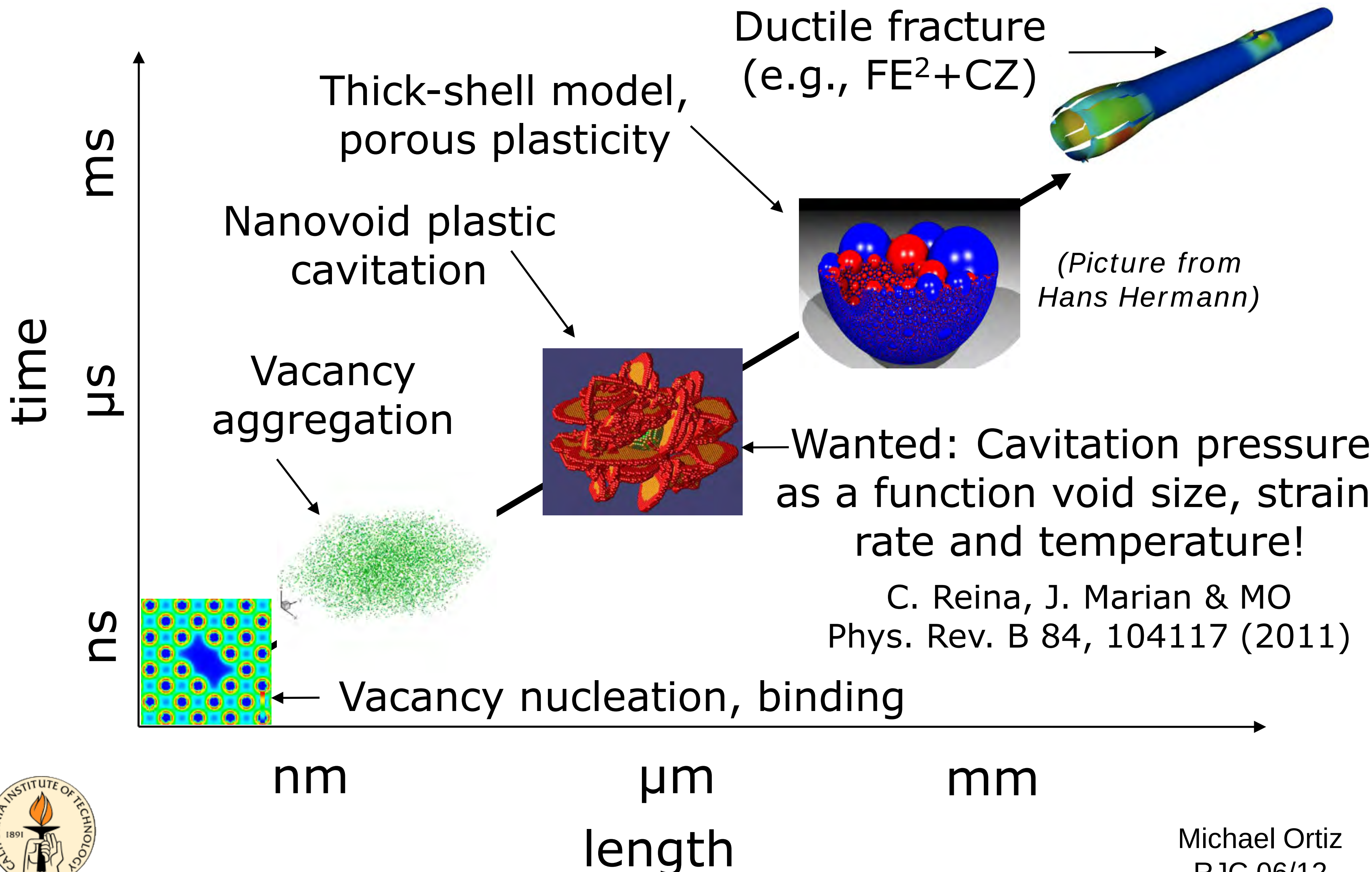


R. Becker "How Metals Fail", *Science and Technology Review*, LLNL, July/August 2002



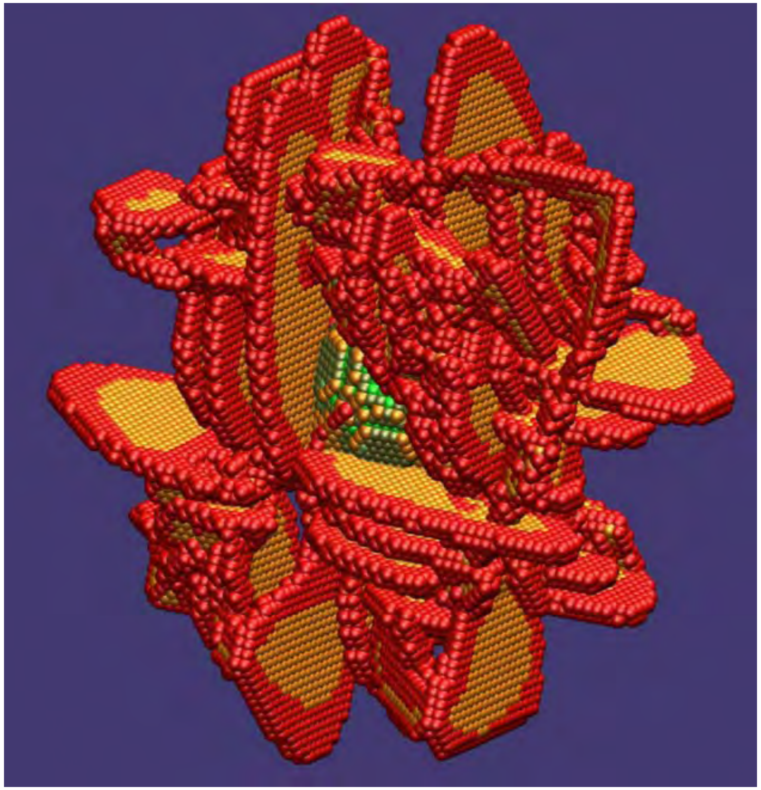
C. Ruggieri, J. of the Braz. Soc. of Mech. Sci. & Eng., Vol. XXVI, No. 2 (2004) 190-198.

Motivation – Ductile fracture in metals



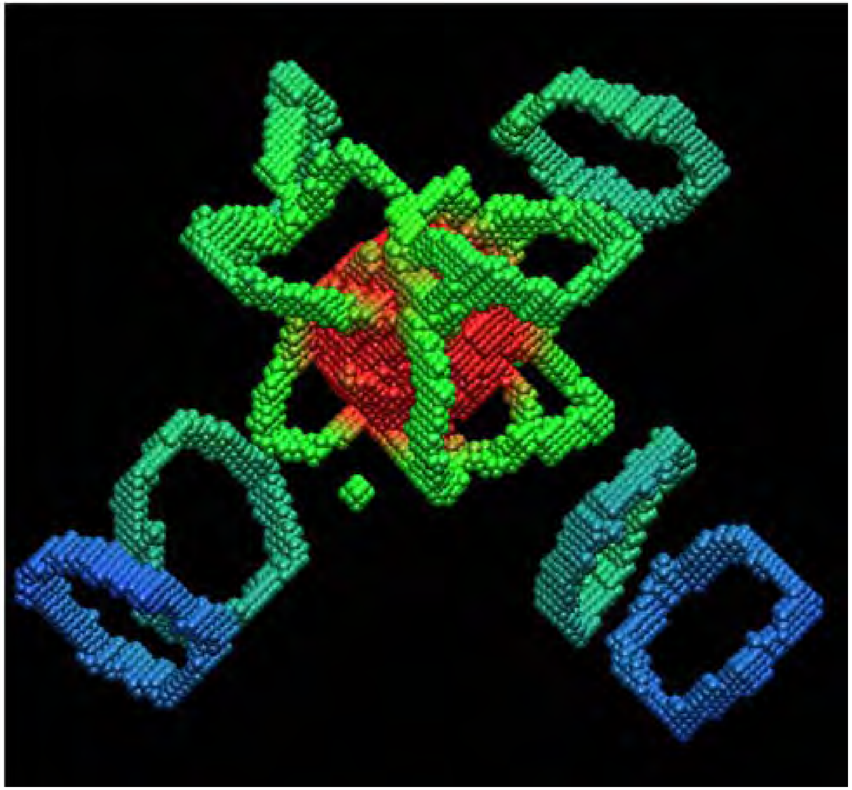
Nanovoid cavitation – MD work

T=0, dε/dt < 10^8 1/s



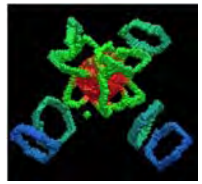
Marian J., Knap J. and MO, "Nanovoid deformation in aluminum under simple shear", *Acta Materialia*, **53**:2893, 2005

T>0, dε/dt < 10^8-10^{12} 1/s

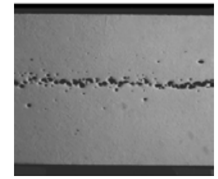


Tang, Y., Bringa, E.M., Remington, B.A., and Meyers, M.A., "Growth and collapse of nanovoids in Ta monocrystals", *Acta Materialia*, **59**:1354, 2011

Atomistic-to-continuum



Tang, Y. *et al.*, *Acta Mater.*, **59**:1354, 2011.



R. Becker, *S&T. Rev.*, (LLNL) July/Aug 2002

	Atomistic	**Coarse-grained**
Configuration space	Phase space (micro)	•Phase space (meso) •Temperature •Concentrations
Governing equations	ΣF=ma	•ΣF=ma (averaged) •Diffusive transport
Spatial resolution	Atomic lattice	•Lattice defects •Temperature grads. •Concentration grads.
Temporal resolution	•Thermal vibrations •Transition states	•Elastic waves (meso) •Diffusional transients
Time-scale bridging	Non-equilibrium statistical mechanics	
Spatial-scale bridging	Quasicontinuum method	

Equilibrium SM – Max-ent formulation

- Gran-canonical ensemble, N atoms, M species:
  - State: $(\{\boldsymbol{q}\}, \{\boldsymbol{p}\}, \{\boldsymbol{n}\}) \in \mathbb{R}^{3N} \times \mathbb{R}^{3N} \times \mathcal{O}_{NM}$
  - Atomic positions: $\{\boldsymbol{q}\} = \{\boldsymbol{q}_1, \dots, \boldsymbol{q}_N\}$
  - Atomic momenta: $\{\boldsymbol{p}\} = \{\boldsymbol{p}_1, \dots, \boldsymbol{p}_N\}$
  - Occupancy: $n_{ik} = \begin{cases} 1, & \text{site } i \text{ occupied by species } k, \\ 0, & \text{otherwise.} \end{cases}$
- Ensemble average of observable $A(\{\boldsymbol{q}\}, \{\boldsymbol{p}\}, \{\boldsymbol{n}\})$:

$$\langle A \rangle = \sum_{\{\boldsymbol{n}\} \in \mathcal{O}_{NM}} \int A(\{\boldsymbol{q}\}, \{\boldsymbol{p}\}, \{\boldsymbol{n}\}) \, p(\{\boldsymbol{q}\}, \{\boldsymbol{p}\}, \{\boldsymbol{n}\}) \, dq \, dp$$

- $p(\{\boldsymbol{q}\}, \{\boldsymbol{p}\}, \{\boldsymbol{n}\}) \equiv$ Gran-canonical pdf

Equilibrium SM – Max-ent formulation

- Information-theoretical entropy: $\mathcal{S}[p] = -k_B \langle \log p \rangle$

- Principle of maximum entropy: $\mathcal{S}[p] \to \max!$

  subject to: $\langle H \rangle = E, \quad \langle \sum_{i=1}^{N} n_{ik} \rangle = N_k$

- Lagrangian: reciprocal temperature (β)

$$\mathcal{L}[p, \beta, \boldsymbol{\Gamma}] = \mathcal{S}[p] - k_B \beta \langle H \rangle - k_B \sum_{k=1}^{M} \Gamma_k \langle \sum_{i=1}^{N} n_{ik} \rangle .$$

chemical potentials (Γ_k)

- Gran-canonical distribution:

$$p = \frac{1}{\Xi} \mathrm{e}^{-\beta H - \boldsymbol{\Gamma}^T \sum_{i=1}^{N} \boldsymbol{n}_i},$$

$$\Xi = \sum_{\{\boldsymbol{n}\} \in \mathcal{O}_{NM}} \int \mathrm{e}^{-\beta H - \boldsymbol{\Gamma}^T \sum_{i=1}^{N} \boldsymbol{n}_i} \, dq \, dp$$

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Equilibrium SM – Macroscopic equilibrium

- Macroscopic variables $\equiv \boldsymbol{X}$ (e.g., volume, strain...)
- Parametrized Hamiltonians: $H(\{\boldsymbol{q}\}, \{\boldsymbol{p}\}, \{\boldsymbol{n}\}, \boldsymbol{X})$
- Gran-canonical thermodynamic potentials:
  - Internal energy: $U(S, \Gamma, \boldsymbol{X}) = -\frac{\partial \log \Xi}{\partial \beta}$
  - Free energy: $F(\beta, \Gamma, \boldsymbol{X}) = -\frac{1}{\beta} \log \Xi$
  - Free entropy: $\Phi(\beta, \Gamma, \boldsymbol{X}) = k_B \log \Xi$
- Equivalent statements of macroscopic equilibrium:

$U(S, \Gamma, \cdot) \to \min!$ (internal energy principle)

$F(\beta, \Gamma, \cdot) \to \min!$ (free energy principle)

$\Phi(\beta, \Gamma, \cdot) \to \max!$ (free entropy principle)

Equilibrium SM – Meanfield theory

- Space of trial Hamiltonians: $\mathcal{H}_0$
- Free-entropy inequality: For all $H_0 \in \mathcal{H}_0$,

  i) $\Phi \geq \Phi_0 - k_B \beta \langle H - H_0 \rangle_0 \equiv -\mathcal{F}[H_0, \beta, \mathbf{\Gamma}]$

  ii) $\Phi = -\mathcal{F}[H_0, \beta, \mathbf{\Gamma}] \Leftrightarrow H_0 = H$

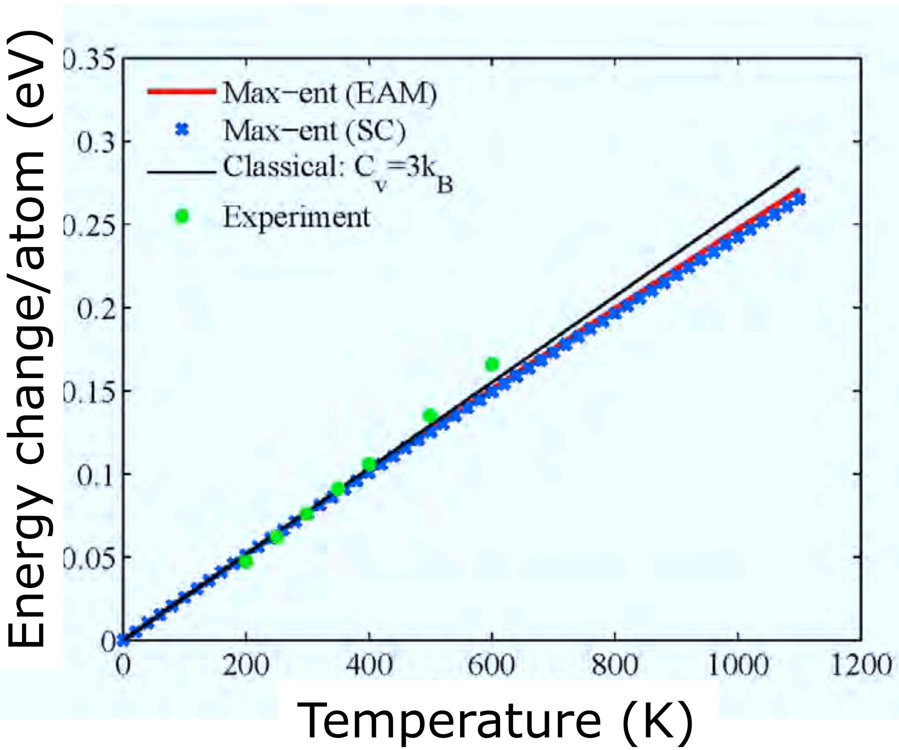
- Best approximation: $\mathcal{F}[\cdot, \beta, \mathbf{\Gamma}] \to$ min! over $\mathcal{H}_0$
- Example: $\mathcal{H}_0 \equiv$ uncoupled harmonic oscillators,

$$H_0 = \sum_{i=1}^{N} \left(\frac{1}{2m(\boldsymbol{n}_i)} |\boldsymbol{p}_i - \bar{\boldsymbol{p}}_i|^2 + \frac{m(\boldsymbol{n}_i)\omega_i^2}{2} |\boldsymbol{q}_i - \bar{\boldsymbol{q}}_i|^2 \right)$$

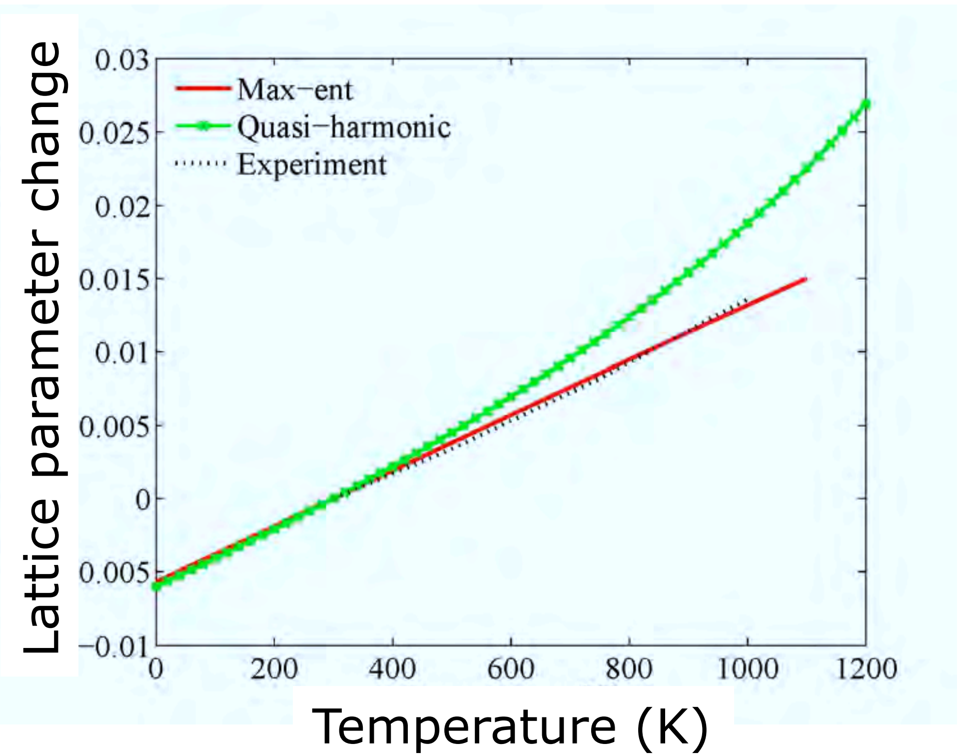
- Minimize $\mathcal{F}(\{\pi\}, \beta, \mathbf{\Gamma})$ w.r.t. $\{\pi\} \equiv (\{\bar{\boldsymbol{q}}\}, \{\bar{\boldsymbol{p}}\}, \{\omega\})$
- Compute $\langle \cdot \rangle_0$ using Gaussian quadrature

Equilibrium SM – Meanfield validation

Specific heat



Thermal expansion



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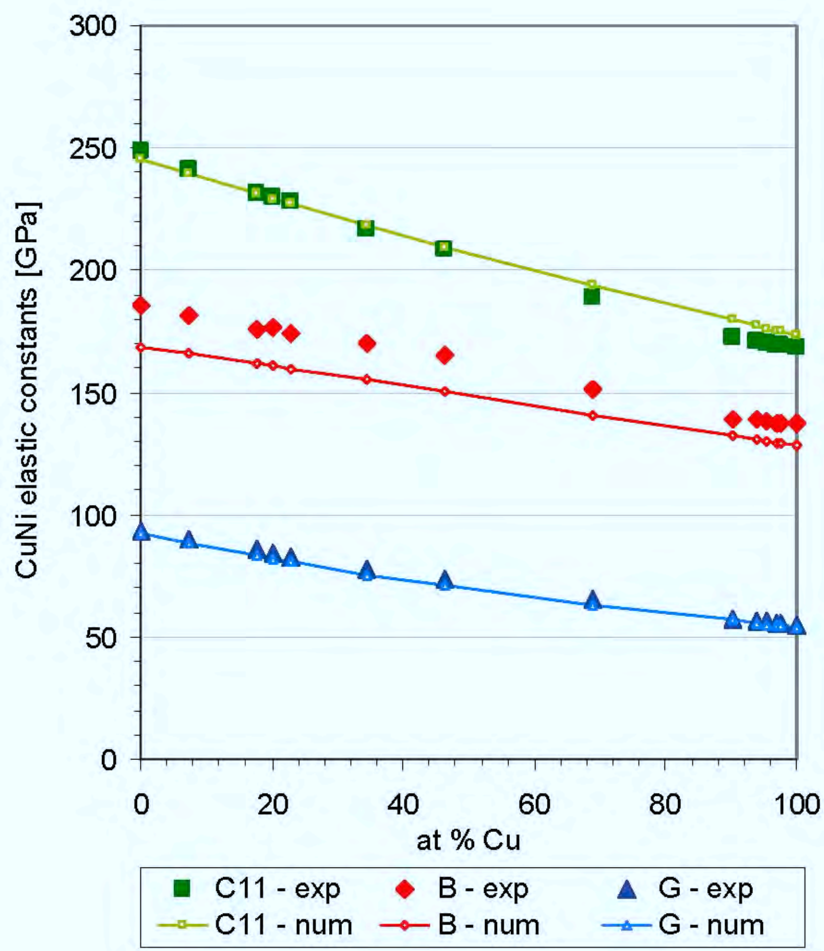
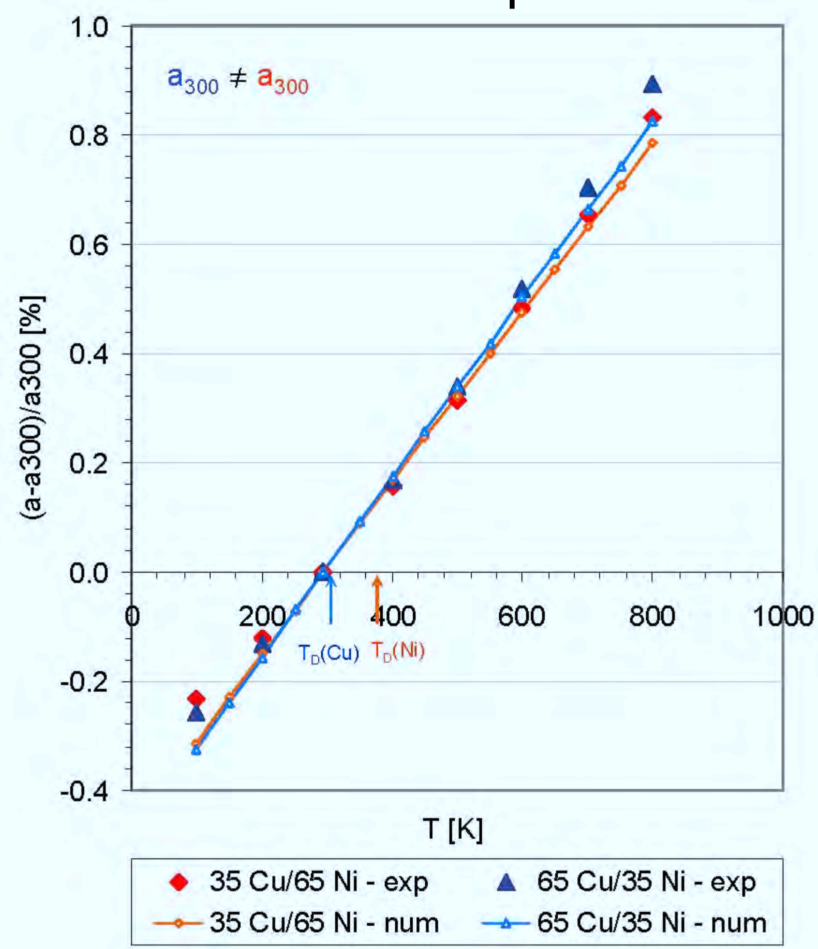
Equilibrium SM – Meanfield validation

Elastic constants



Thermal expansion



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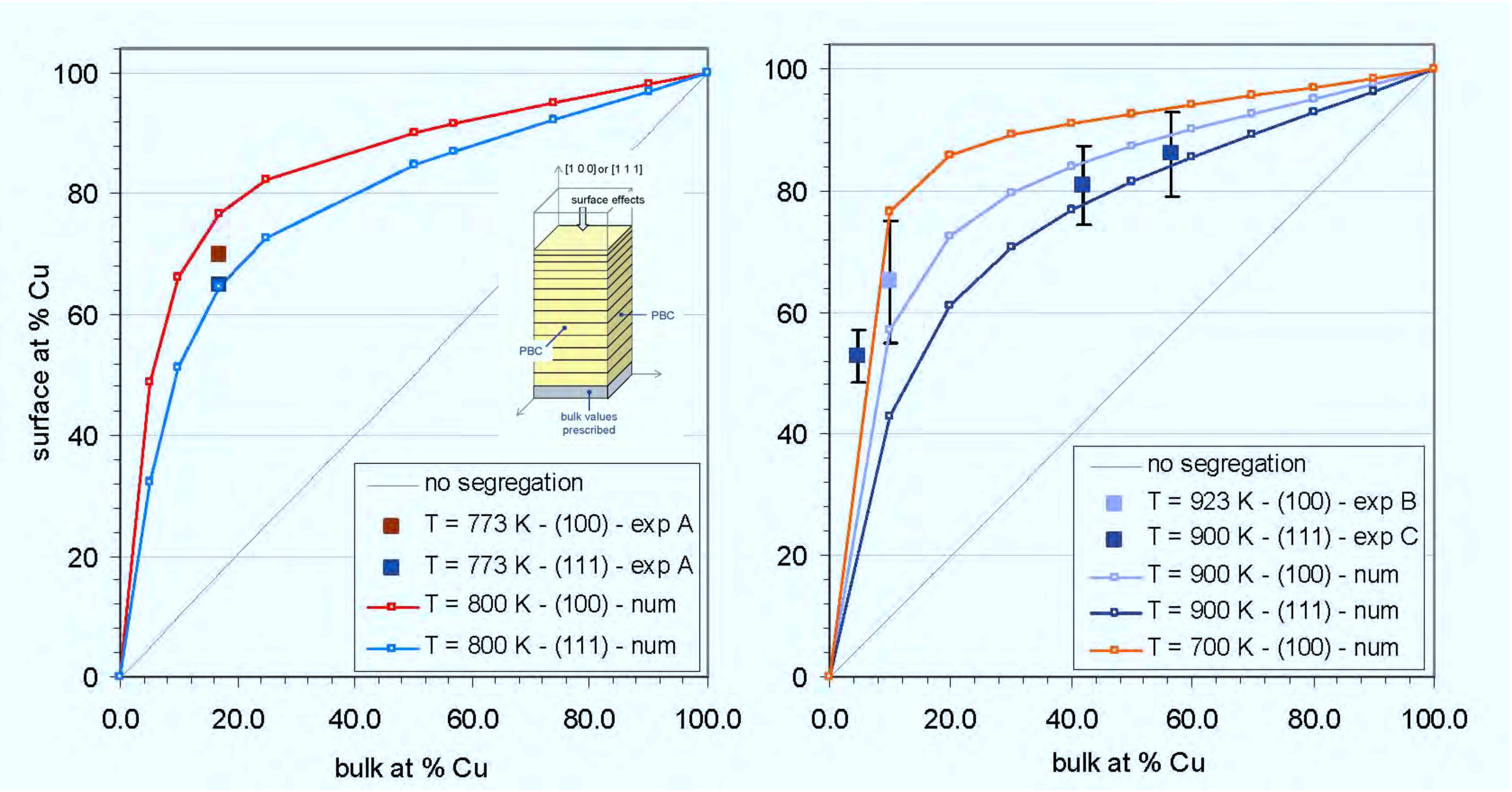
Equilibrium SM – Meanfield validation



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Venturini, G., *Doctoral Dissertation*, Caltech, 2011

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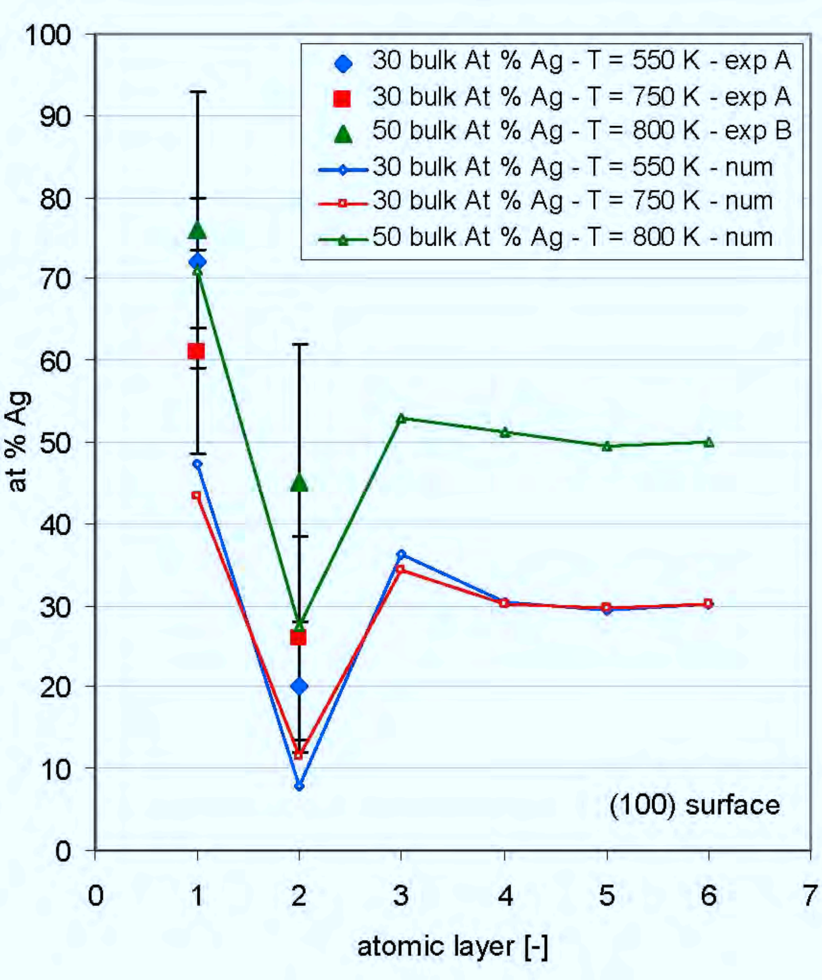
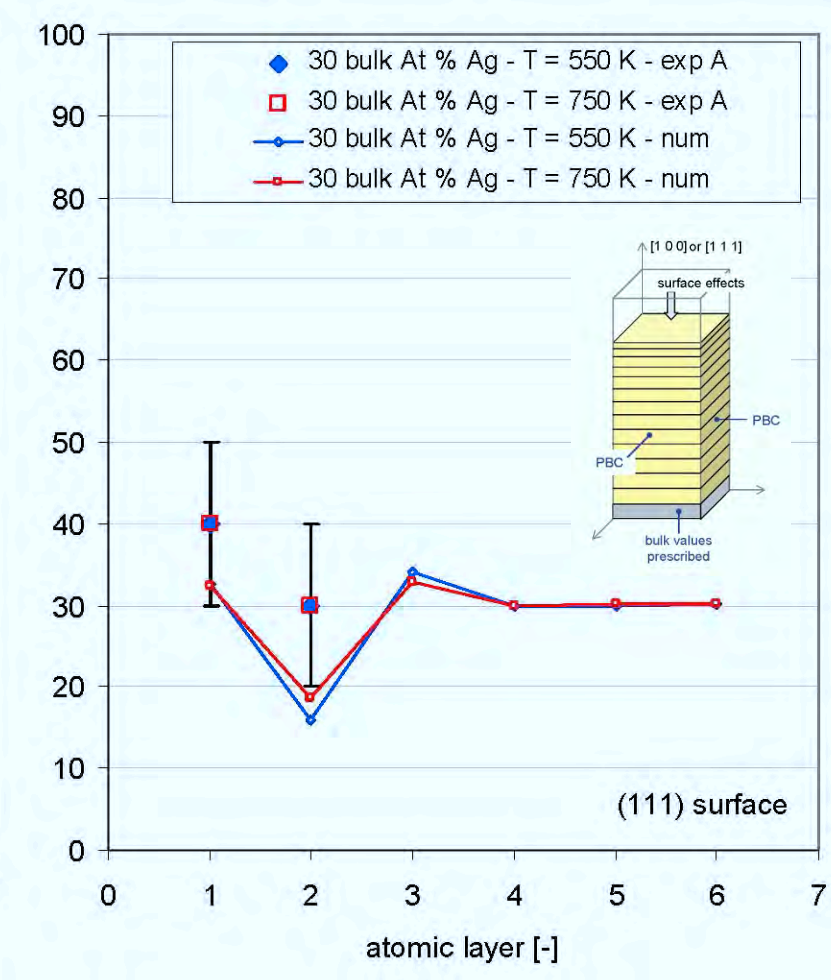
Equilibrium SM – Meanfield validation





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Venturini, G., *Doctoral Dissertation*, Caltech, 2011

Extension to non-equilibrium SM

- Assume $H = \sum_{i=1}^{N} h_i$, otherwise arbitrary.
- Principle of maximum entropy: $\mathcal{S}[p] \to \max!$

  subject to: $\langle h_i \rangle = e_i, \quad \langle n_{ik} \rangle = x_{ik}$

- Lagrangian: reciprocal temperature field

$$\mathcal{L}[p, \{\beta\}, \{\boldsymbol{\gamma}\}] = \mathcal{S}[p] - k_B \{\beta\}^T \{\langle h \rangle\} - k_B \{\boldsymbol{\gamma}\}^T \{\langle \boldsymbol{n} \rangle\}$$

- Gran-canonical distribution: chemical potential field

$$p = \frac{1}{\Xi} \mathrm{e}^{-\{\beta\}^T \{h\} - \{\boldsymbol{\gamma}\}^T \{\boldsymbol{n}\}}$$

$$\Xi = \sum_{\{\boldsymbol{n}\} \in \mathcal{O}_{NM}} \int \mathrm{e}^{-\{\beta\}^T \{h\} - \{\boldsymbol{\gamma}\}^T \{\boldsymbol{n}\}} \, dq \, dp$$

Non-equilibrium SM – Macroscopic equilibrium

- Macroscopic variables $\equiv \boldsymbol{X}$ (e.g., volume, strain...)
- Parametrized Hamiltonians: $H(\{\boldsymbol{q}\}, \{\boldsymbol{p}\}, \{\boldsymbol{n}\}, \boldsymbol{X})$
- Gran-canonical thermodynamic potentials:
  - Free entropy: $\Phi(\{\beta\}, \{\gamma\}, \boldsymbol{X}) = k_B \log \Xi$
- Statement of macroscopic equilibrium:

  $\Phi(\{\beta\}, \{\gamma\}, \cdot) \to$ max! (free entropy principle)

Non-equilibrium SM – Meanfield theory

- Space of trial *local Hamiltonians*: $\mathcal{H}_0$
- Free-entropy inequality: For all $\{h_0\} \in \mathcal{H}_0$,

  i) $\Phi \geq \Phi_0 - k_B \{\beta\}^T \{\langle h - h_0 \rangle_0\} \equiv -\mathcal{F}[\{h_0\}, \{\beta\}, \{\gamma\}]$

  ii) $\Phi = -\mathcal{F}[\{h_0\}, \{\beta\}, \{\gamma\}] \Leftrightarrow \{h_0\} = \{h\}$

- Best approximation: $\min \mathcal{F}[\{h_0\}, \{\beta\}, \{\gamma\}]$ over $\mathcal{H}_0$
- Example: $\mathcal{H}_0 \equiv$ local harmonic oscillators,

$$h_{0i} = \frac{1}{2m(\boldsymbol{n}_i)} |\boldsymbol{p}_i - \bar{\boldsymbol{p}}_i|^2 + \frac{m(\boldsymbol{n}_i)\omega_i^2}{2} |\boldsymbol{q}_i - \bar{\boldsymbol{q}}_i|^2$$

- Minimize $\mathcal{F}[\{h_0\}, \{\beta\}, \{\gamma\}]$ w.r.t. $\{\pi\} \equiv (\{\bar{\boldsymbol{q}}\}, \{\bar{\boldsymbol{p}}\}, \{\omega\})$
- Compute $\langle \cdot \rangle_0$ using Gaussian quadrature

Non-equilibrum SM – Kinetics (Onsager)

- Need equations of evolution for $\{e\}$ and $\{\gamma\}$.
- Free entropy: $\Phi = k_B \log \Xi(\{\beta\}, \{\gamma\})$
- Thermodynamic driving-force identities:

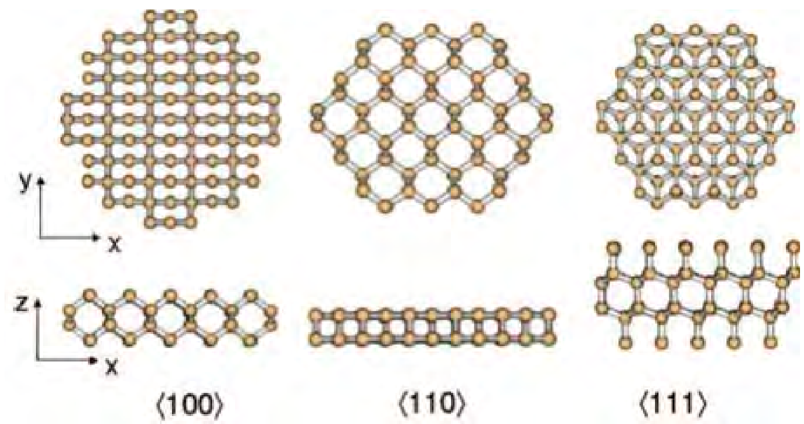
$$\beta_i = \frac{1}{k_B}\frac{\partial \Phi^*}{\partial e_i}(\{e\}, \{\boldsymbol{x}\}), \quad \gamma_{ik} = \frac{1}{k_B}\frac{\partial \Phi^*}{\partial x_{ik}}(\{e\}, \{\boldsymbol{x}\})$$

- General kinetic relations (*a la* Onsager):

$$\underbrace{\dot{e}_i = \frac{\partial \psi}{\partial \beta_i}(\{\beta\}, \{\boldsymbol{\gamma}\}),}_{\text{Discrete Fourier law}} \qquad \underbrace{\dot{x}_{ik} = \frac{\partial \psi}{\partial \gamma_{ik}}(\{\beta\}, \{\boldsymbol{\gamma}\})}_{\text{Discrete Fick's law}}$$

- ψ formulated using discrete differential operators

Non-equilibrium SM – Validation



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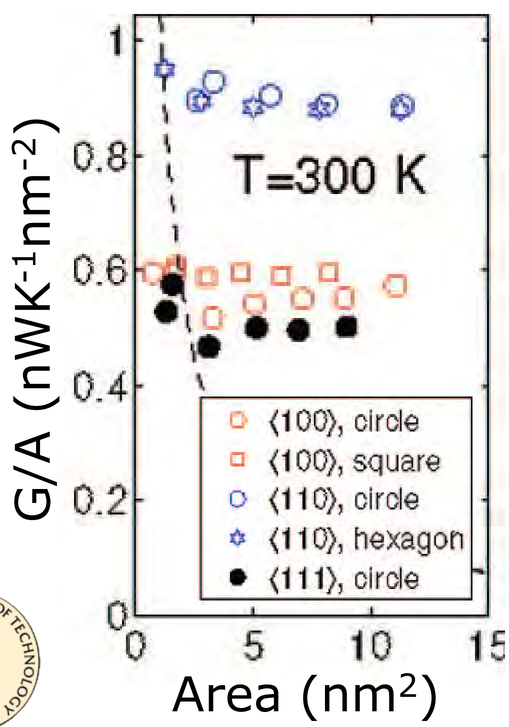
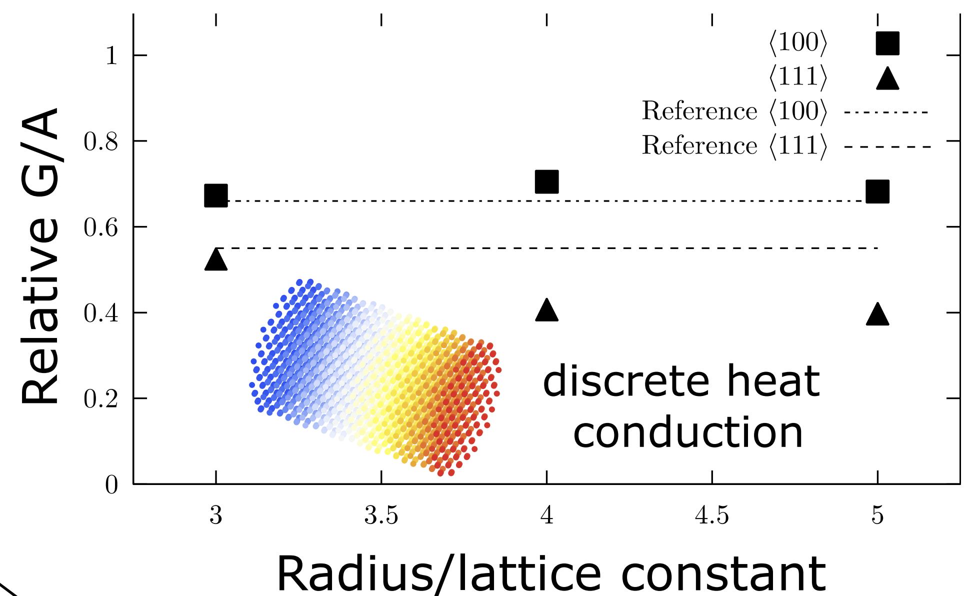
"Heat Conductance Is Strongly Anisotropic for Pristine Silicon Nanowires", *Nano Letters*, **8**(11):3771, 2008.





Markussen, T., *et al*., 2008.

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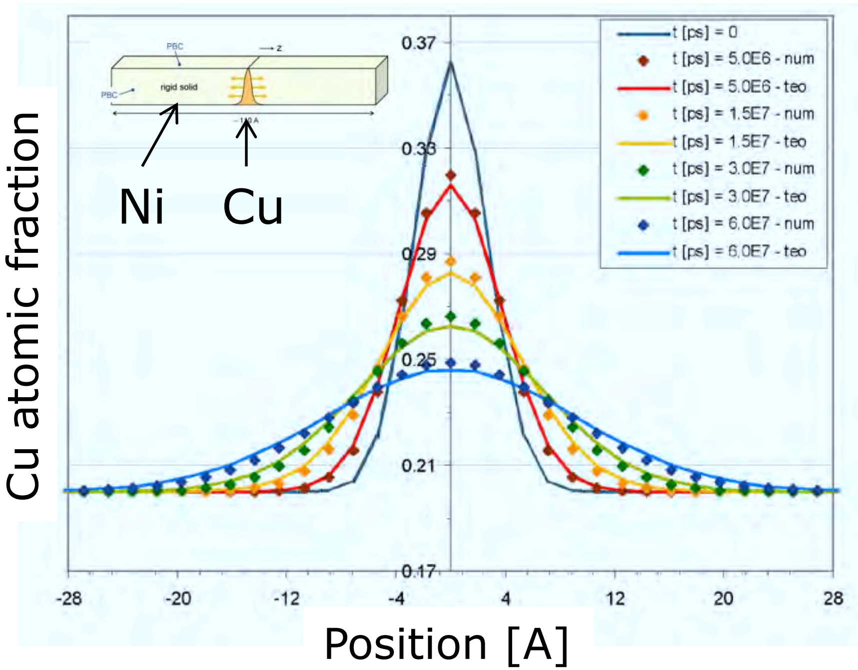
Non-equilibrium SM – Validation



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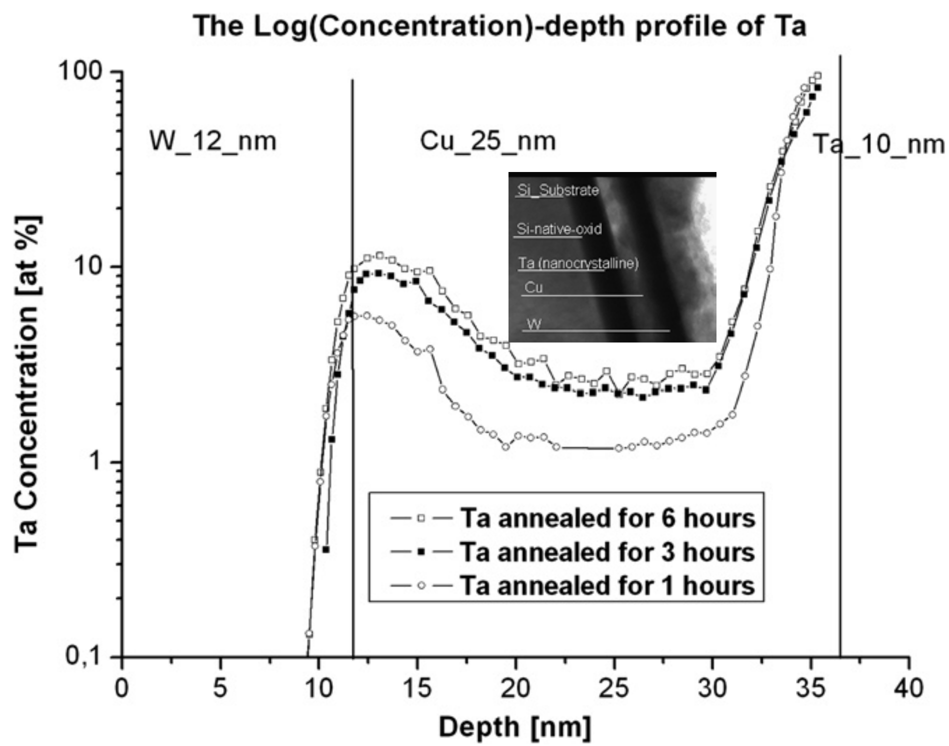

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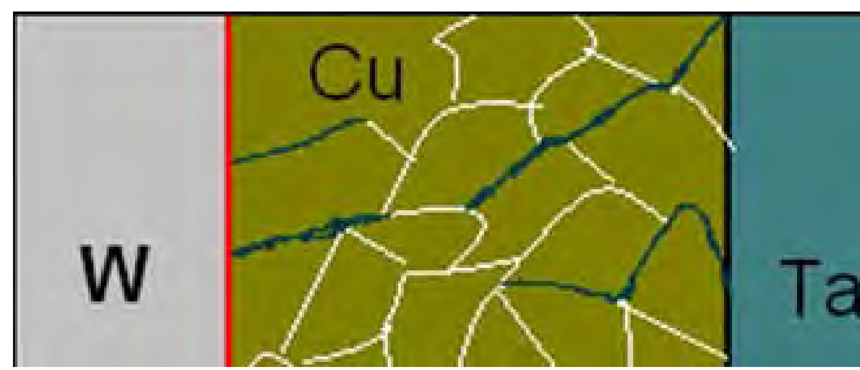
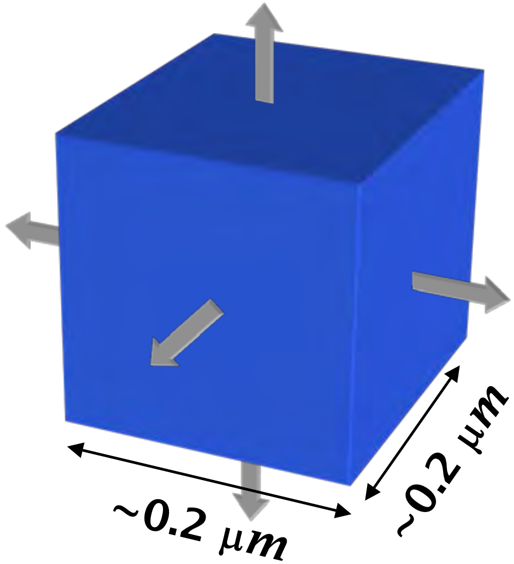
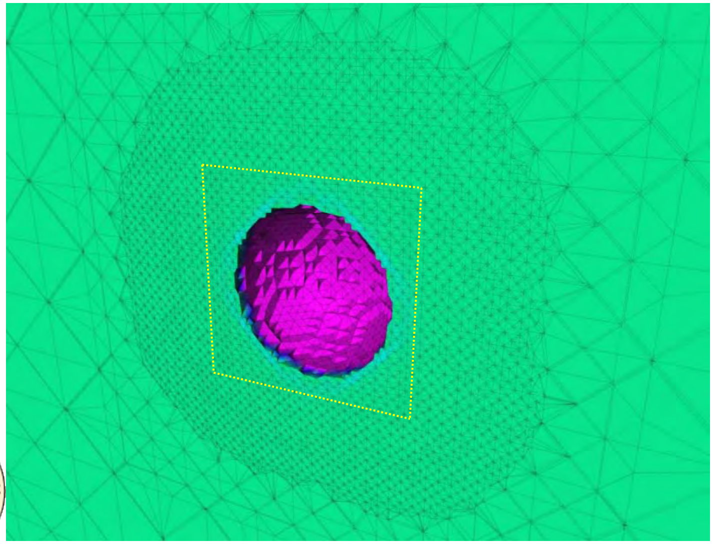


Application to nanovoid plastic cavitation





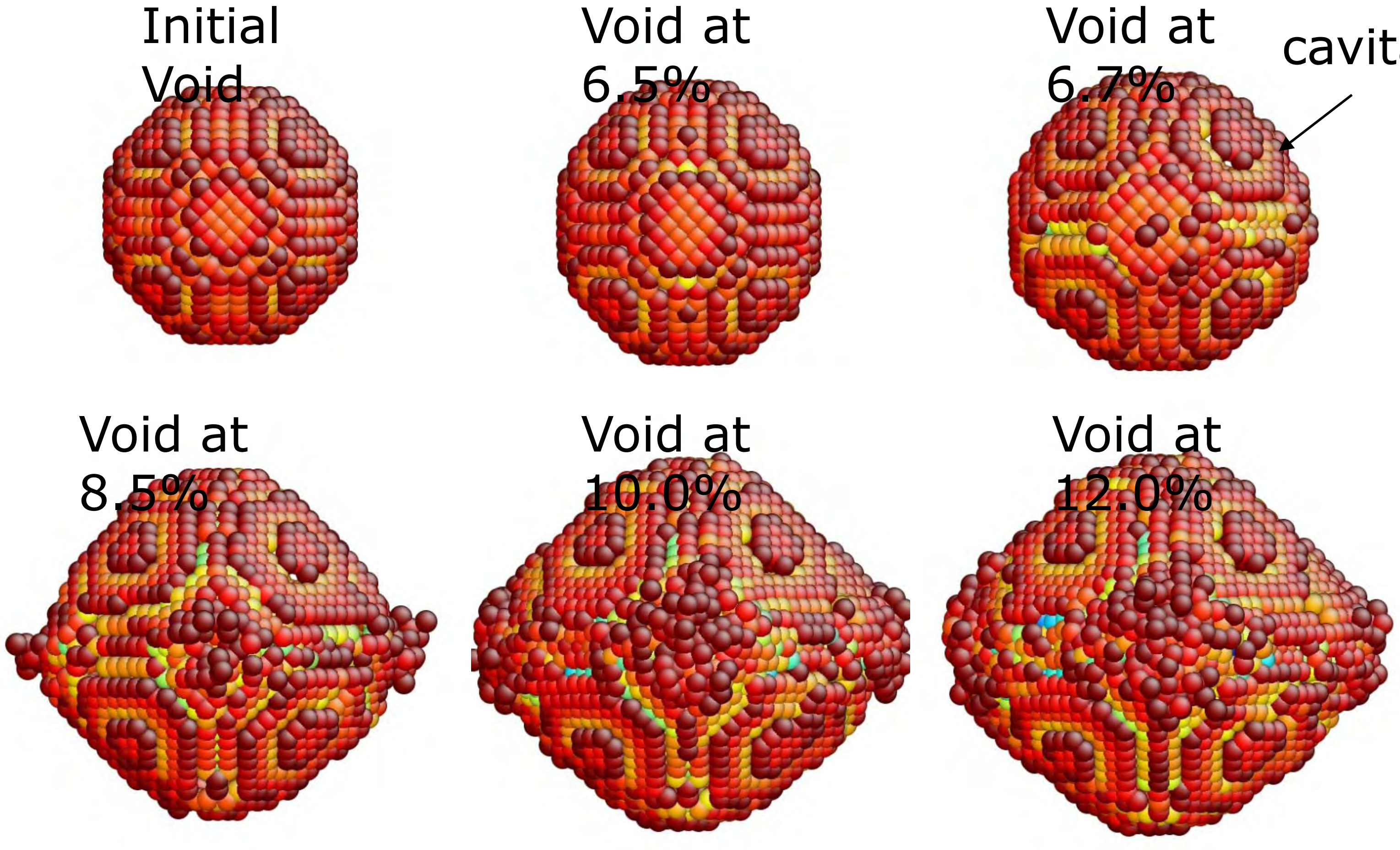
- Parameters:
  - *T_0 = 300K*
  - *Full Size = $72a_0$*
  - *Atomistic Zone = $14a_0$*
  - *Diameter = $12a_0$*
  - *Strain Rate = <10^{10} 1/s*
- Loading:
  - *Triaxial, uniaxial*
- Material:
  - *Copper, Mishin, Y., Phys. Rev. B. 2001,*

Initial quasicontinuum mesh with full atomistic resolution near void

Nanovoid plastic cavitation in Cu

Uniaxial loading

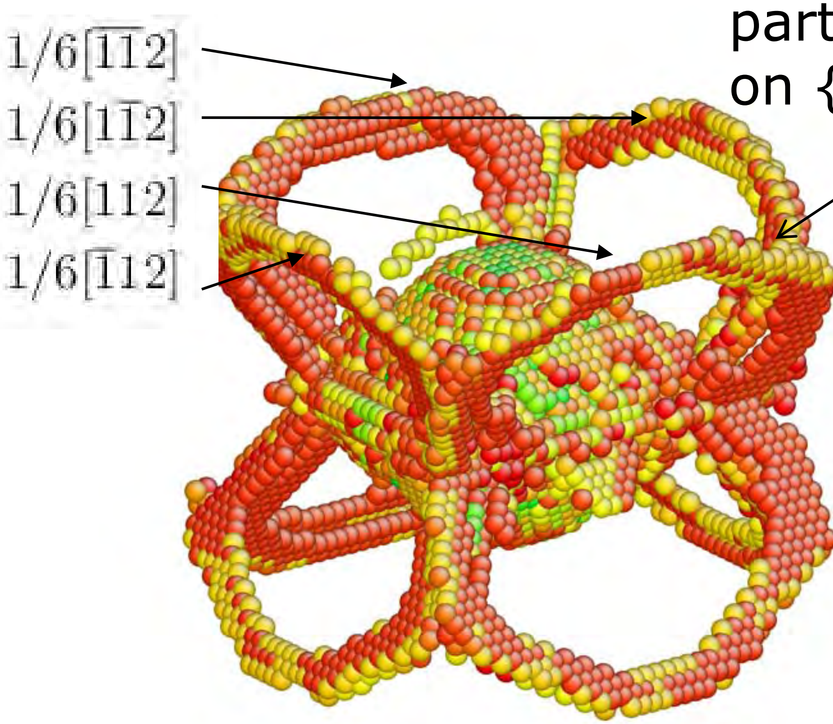
Growth on transversal
direction when
dislocation are emitted

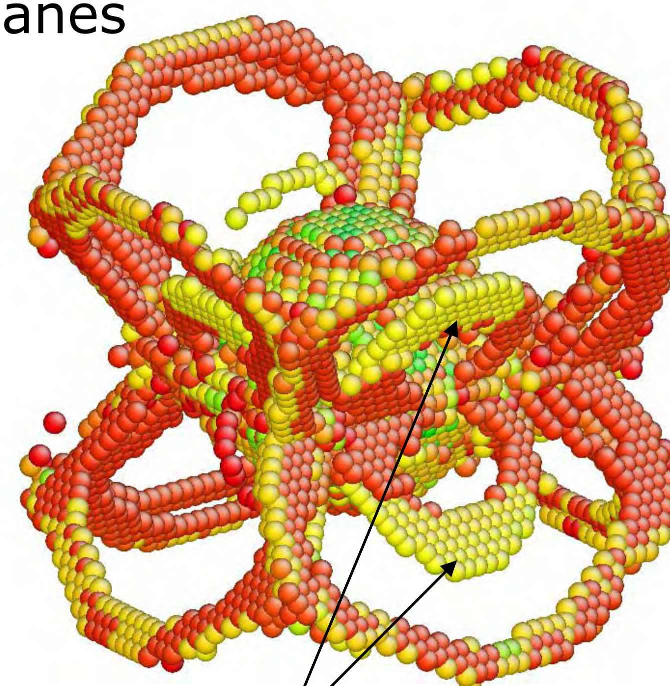


Final shape of void,
octahedral on 8 planes on {111}

Nanovoid plastic cavitation in Cu

Leading Shockley partial dislocations on {111} planes





Uniaxial loading, ε = 6.6%

Trailing Shockley partial dislocations on {111} planes

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Nanovoid plastic cavitation in Cu

Shear to Prismatic loop reactions:

On <110> directions

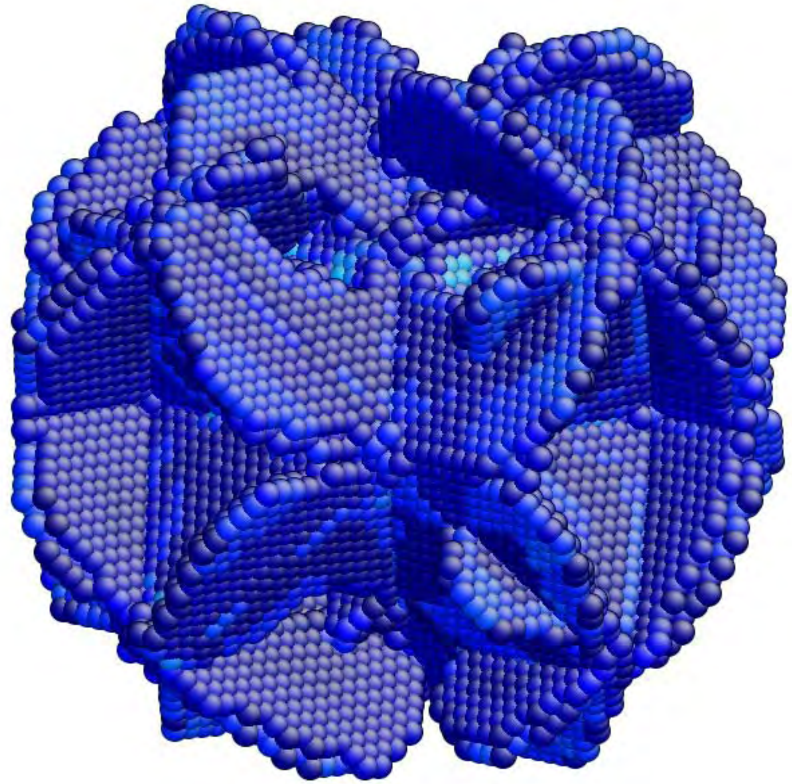
$$1/6[\bar{2}11] + 1/6[\bar{2}1\bar{1}] + 1/6[\bar{1}21] + 1/6[\bar{1}2\bar{1}] \rightarrow [\bar{1}10]$$
$$1/6[\bar{2}\bar{1}\bar{1}] + 1/6[\bar{2}\bar{1}1] + 1/6[\bar{1}\bar{2}\bar{1}] + 1/6[\bar{1}\bar{2}1] \rightarrow [\bar{1}\bar{1}0]$$
$$1/6[1\bar{2}1] + 1/6[1\bar{2}\bar{1}] + 1/6[2\bar{1}1] + 1/6[2\bar{1}\bar{1}] \rightarrow [1\bar{1}0]$$
$$1/6[211] + 1/6[12\bar{1}] + 1/6[21\bar{1}] + 1/6[121] \rightarrow [110]$$

On <110> directions

$$1/6[\bar{2}\bar{1}\bar{1}] + 1/6[\bar{1}\bar{1}\bar{2}] + 1/6[\bar{2}1\bar{1}] + 1/6[\bar{1}1\bar{2}] \rightarrow [\bar{1}0\bar{1}]$$
$$1/6[1\bar{1}\bar{2}] + 1/6[2\bar{1}\bar{1}] + 1/6[21\bar{1}] + 1/6[11\bar{2}] \rightarrow [10\bar{1}]$$
$$1/6[2\bar{1}1] + 1/6[211] + 1/6[1\bar{1}2] + 1/6[112] \rightarrow [101]$$
$$1/6[1\bar{1}2] + 1/6[\bar{2}11] + 1/6[\bar{2}\bar{1}1] + 1/6[\bar{1}\bar{1}2] \rightarrow [\bar{1}01]$$

On <110> directions

$$1/6[\bar{1}21] + 1/6[\bar{1}12] + 1/6[\bar{1}21] + 1/6[112] \rightarrow [011]$$
$$1/6[1\bar{2}\bar{1}] + 1/6[\bar{1}\bar{2}\bar{1}] + 1/6[1\bar{1}\bar{2}] + 1/6[\bar{1}\bar{1}\bar{2}] \rightarrow [0\bar{1}\bar{1}]$$
$$1/6[\bar{1}\bar{2}1] + 1/6[1\bar{2}1] + 1/6[\bar{1}\bar{1}2] + 1/6[1\bar{1}2] \rightarrow [0\bar{1}1]$$
$$1/6[1\bar{2}1] + 1/6[11\bar{2}] + 1/6[\bar{1}1\bar{2}] + 1/6[12\bar{1}] \rightarrow [01\bar{1}]$$



Triaxial loading

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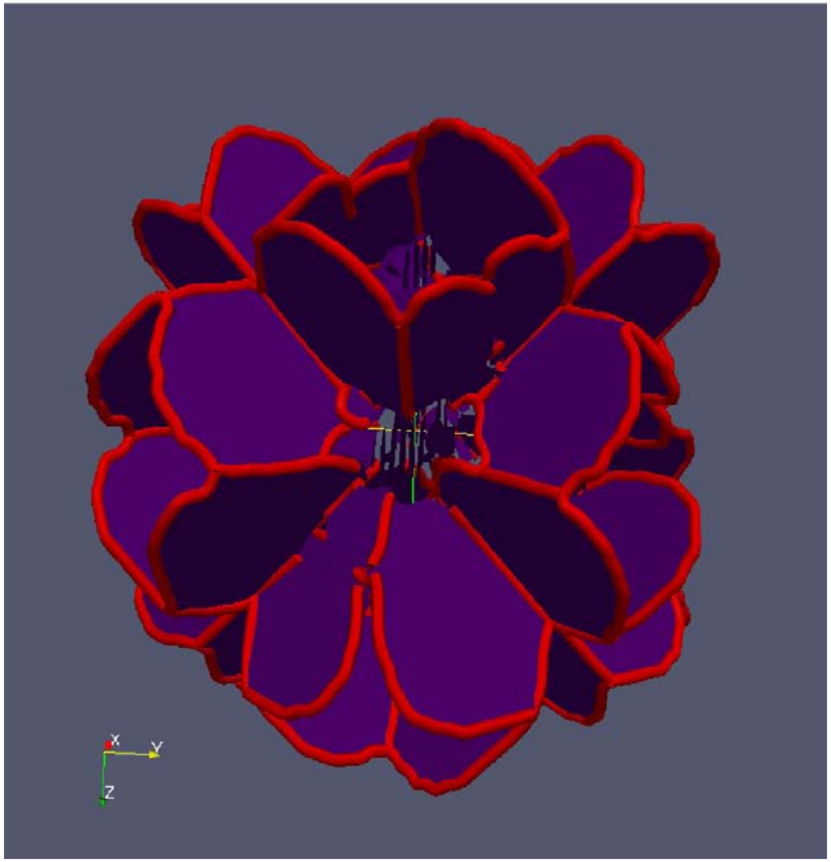
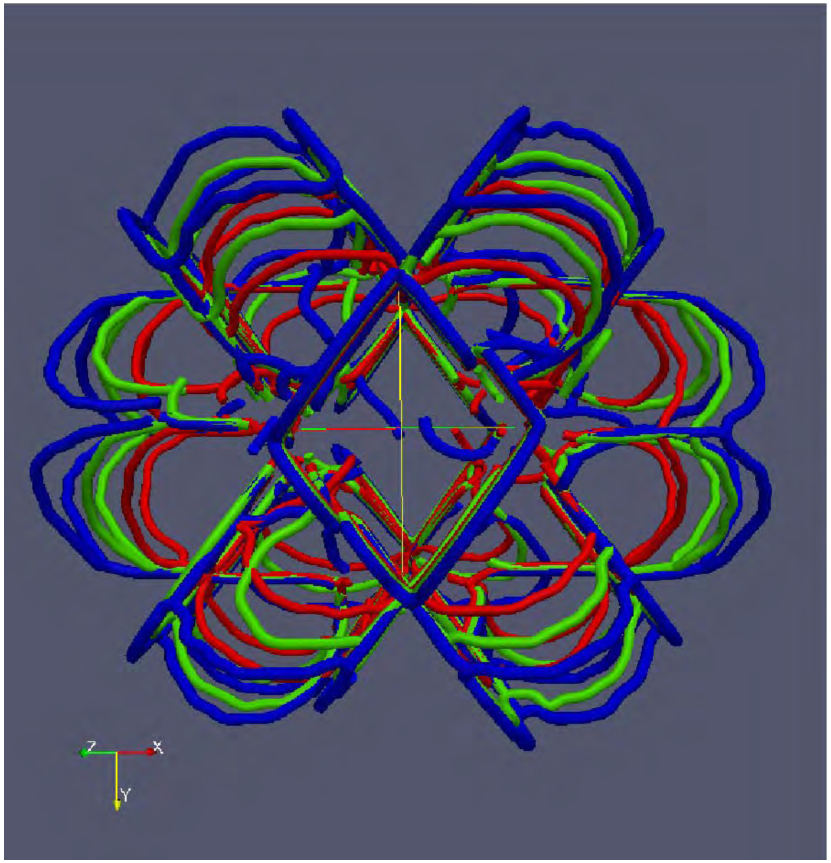
Nanovoid plastic cavitation in Cu



Prismatic loop structure,
triaxial loading

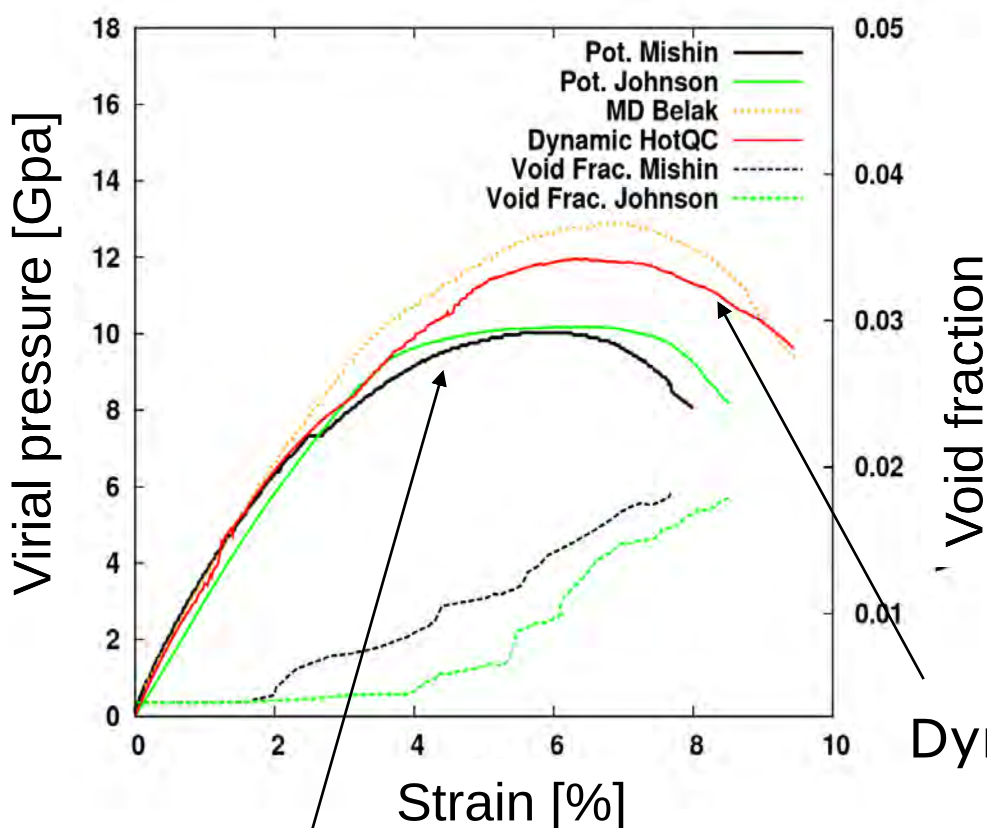


Prismatic loop evolution
(ε = 5, 6, 7%)

(Images obtained with **DXA** and **Paraview**)

Nanovoid plastic cavitation in Cu

Triaxial loading ($d\varepsilon/dt = 10^{10}$ 1/s)



Dynamic

Quasistatic

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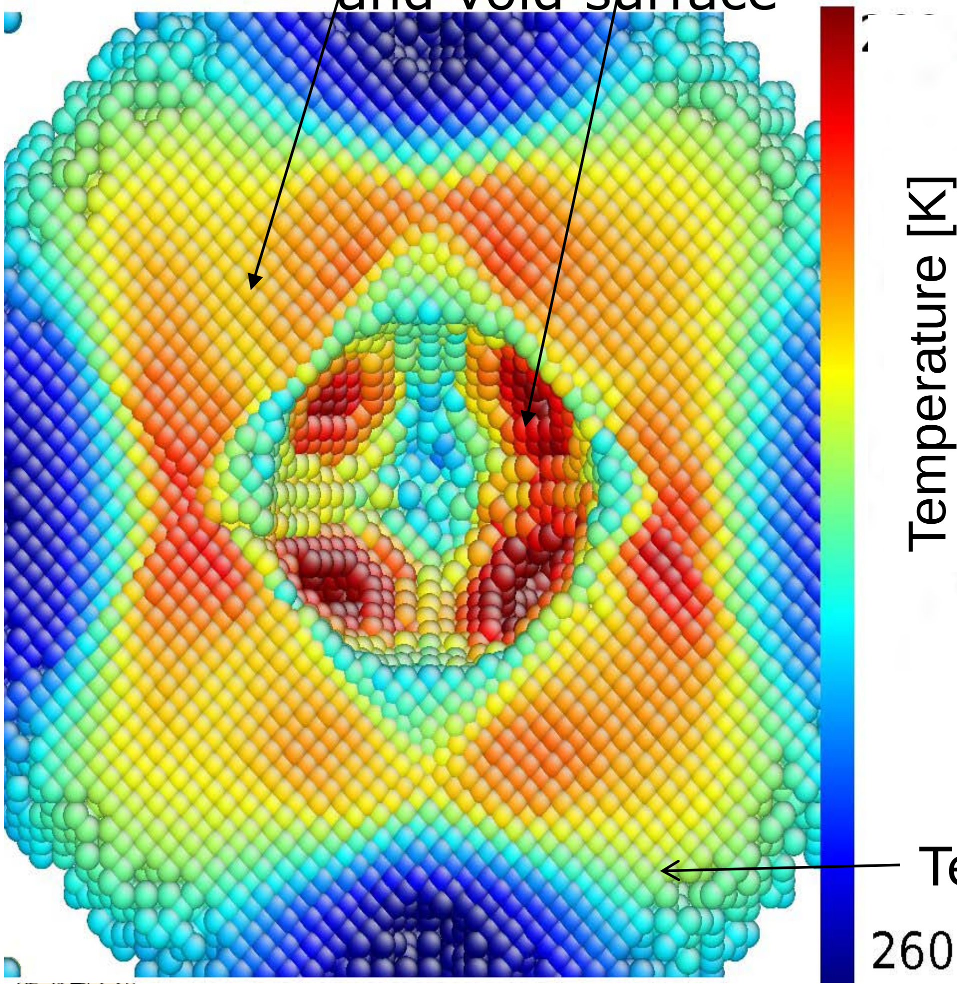
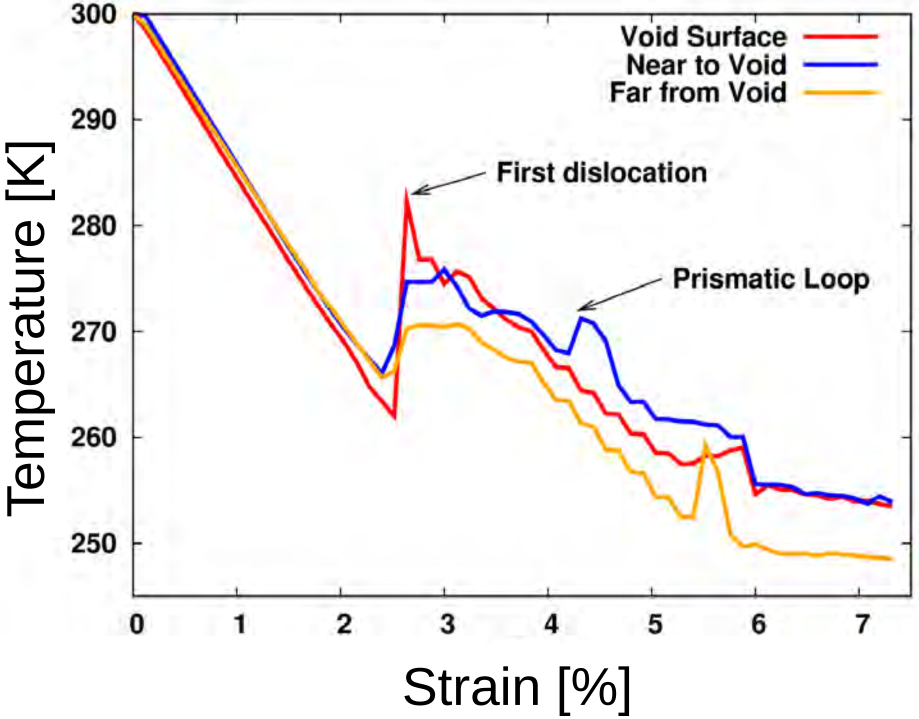
Nanovoid plastic cavitation in Cu

Temperature increase on [110] directions and void surface



Temperature field @ ε=2.7%

Temperature evolution



To Professor Rodney J. Clifton

Scientist, teacher, mentor

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