The Quasicontinuum Method at Finite Temperature
&
A Dynamic Finite Temperature Coupled Atomistic and Discrete Dislocation Model

Ronald E. Miller
Carleton University, Ottawa, Canada

QC Goals:

- Rigorous treatment of the continuum at finite T
- A “replacement” for thermostatted MD

QC Issues:

- Entropy of Coarse-Grained region

CADD Goals:

- Atomistic region can be “far from equilibrium”
- Don’t want artificial damping due to a thermostat
- Continuum treatment of dislocations

CADD Issues:

- Wave reflections at the atomistic/continuum interface
- Heat dissipation

The Quasicontinuum Method at Finite Temperature

Ronald E. Miller
Carleton University, Ottawa, Canada

and

Ellad Tadmor, Technion, Haifa, Israel
Laurent Dupuy, Lawrence Livermore National Labs, USA
Rob Phillips, CalTech, USA

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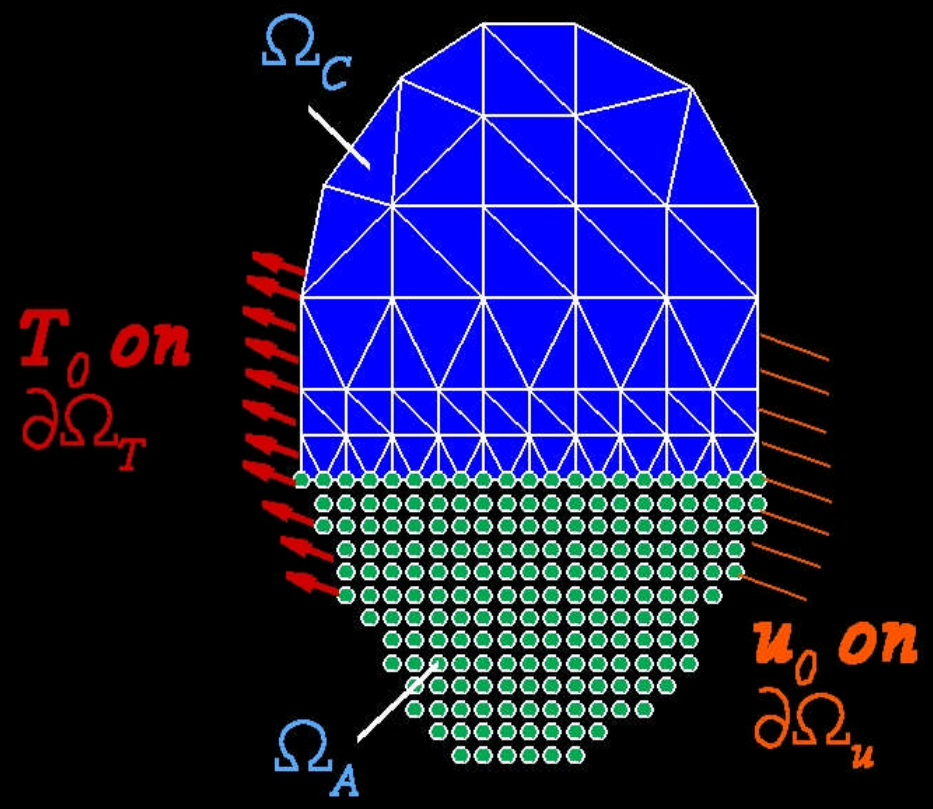
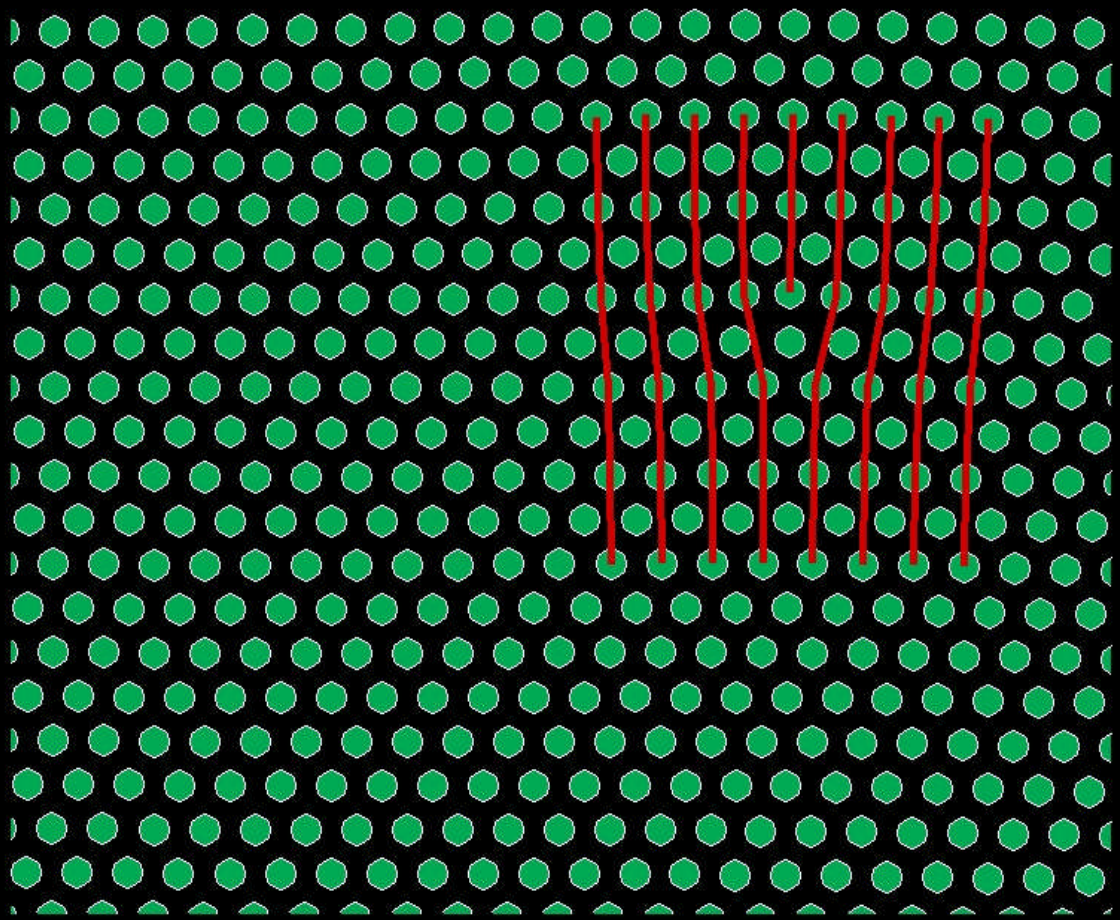
Concurrent Multiscale Modeling

- One such method is the Quasicontinuum (QC) Method
  - Originally developed to solve quasi-static, 2D, single crystal deformation problems (Tadmor, Ortiz and Phillips, 1996).
- Extended to:
  - Polygrains (Shenoy et al 1998)
  - 3D (Rodney and Phillips, 1999)
  - Finite T (Shenoy, Shenoy and Phillips, 1999, Dupuy et al 2005)



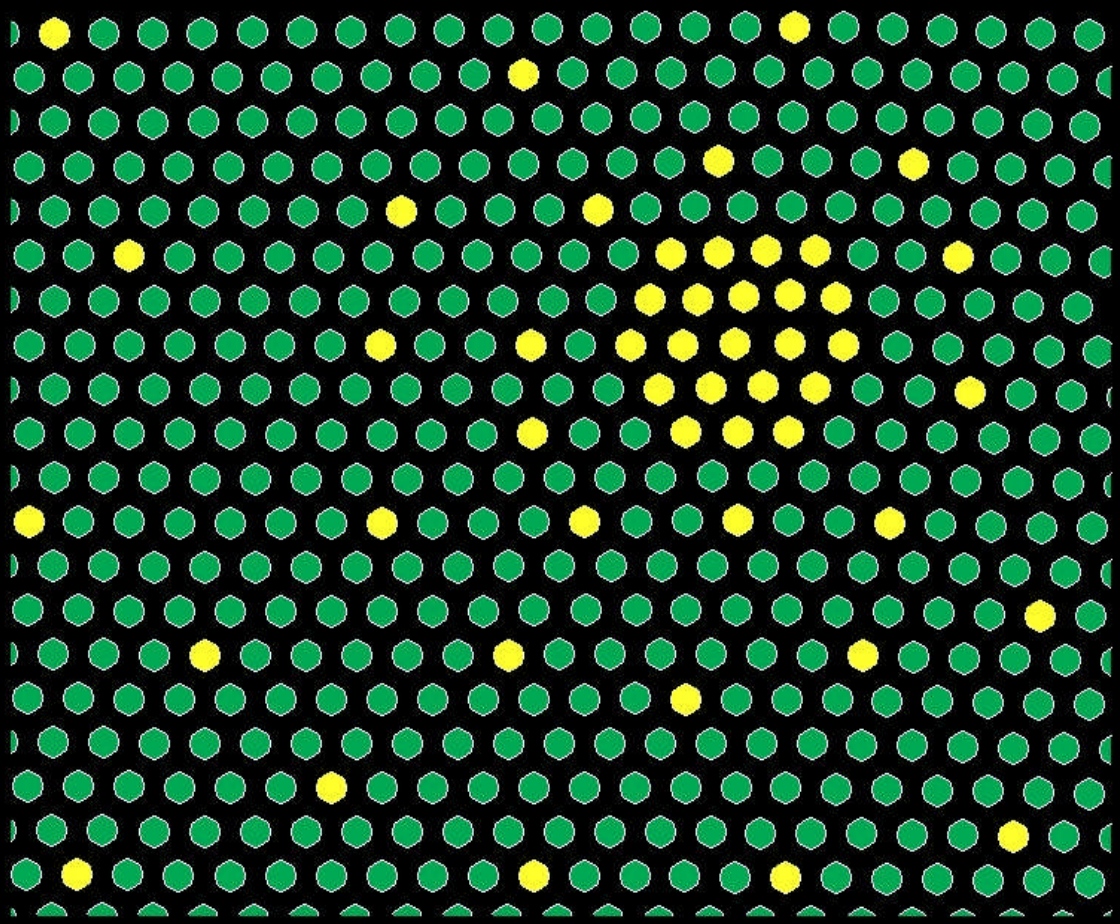
The Quasicontinuum (QC) Method

- Identify regions of slowly varying deformation.
- Note that environment of each atom in these regions is about the same



The Quasicontinuum (QC) Method

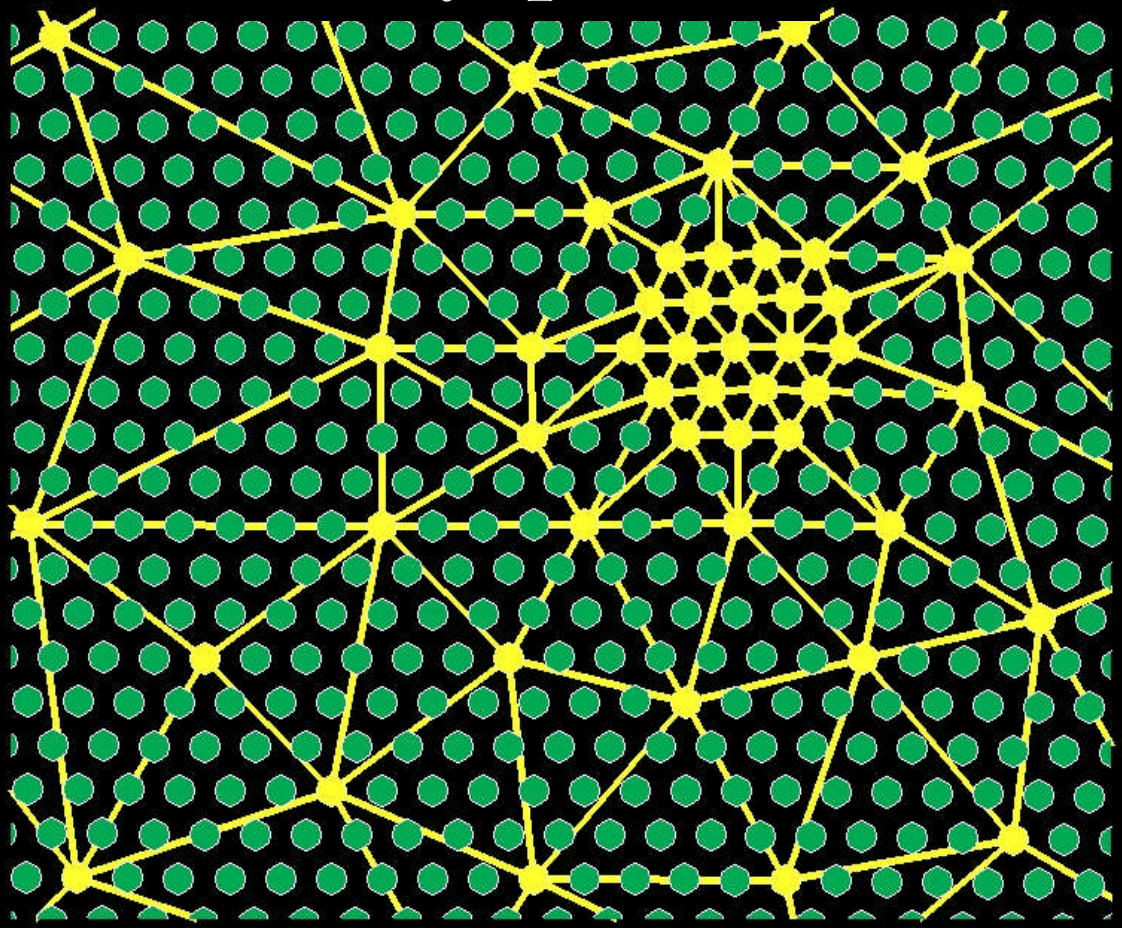
- "Representative atoms" are chosen



The Quasicontinuum (QC) Method

- Finite Element mesh to constrain the position of all other atoms

$$\mathbf{q}_j^c = \sum_{i=1}^{N_r} S_{ij} \mathbf{q}_i^r$$



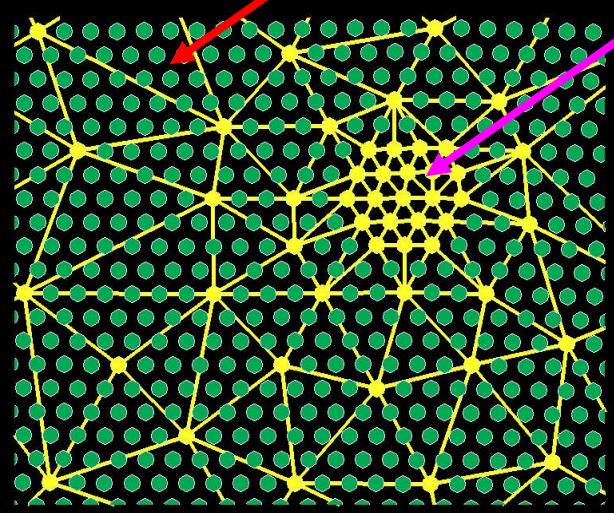
The Quasicontinuum (QC) Method

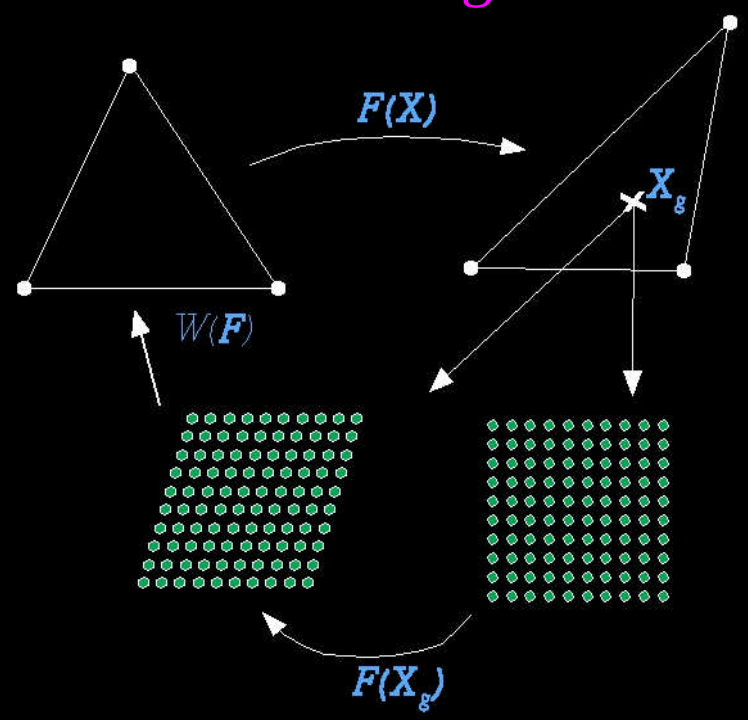
- The potential energy is approximated as:

$$E = \sum_{i=1}^{N} E_i \approx \sum_{e}^{N_e} n_e E_{CB}(\mathbf{F}_e) + \sum_{i \in nl} E_i$$

"Local" or continuum regions

"Nonlocal" or atomistic regions





Extension to Finite Temperature

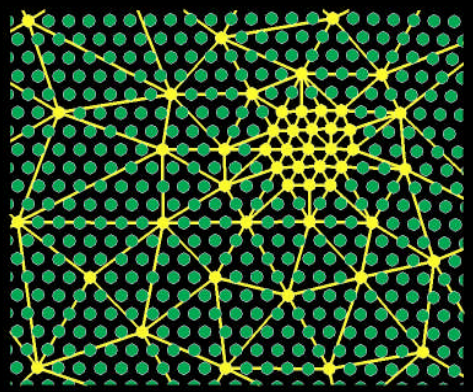
- Our goal:
  - Canonical (constant T) ensemble, multiscale approach
- Our "gold standard":
  - a thermostatted, full MD simulation with the same underlying interatomic potentials.
- *Our criteria to quantify the success of the method:*
  - *Nonlocal (atomistic) regions behave as in full MD*
  - *behaviour of the overall system compared to full MD:*

$$\langle A(\{\mathbf{q}^r\})\rangle_{\mathrm{QC}} \equiv \langle A(\{\mathbf{q}^r\})\rangle_{N_r,V,T}$$
$$= \langle A(\{\mathbf{q}^r\})\rangle_{\mathrm{N,V,T}}$$

e.g.: thermal expansion, elastic constants

Repatoms and Constrained Atoms

- Divide into repatoms, $\{\boldsymbol{q}^r, \boldsymbol{p}^r\}$ and constrained atoms $\{\boldsymbol{q}^c, \boldsymbol{p}^c\}$
- Determine the *mean position* of the constrained atoms using FE shape functions



$$\left\langle \mathbf{q}_j^c \right\rangle = \sum_{i=1}^{N_r} S_{ij} \mathbf{q}_i^r$$

- We seek a Hamiltonian that depends only on the repatom positions and momenta:

$$H_{QC}(\boldsymbol{q}^r, \boldsymbol{p}^r) = V_{QC}(\boldsymbol{q}^r) + \sum_i^{N_r} \frac{\boldsymbol{p}_i^r \cdot \boldsymbol{p}_i^r}{2m_i^r}$$

- The masses, $\boldsymbol{m}_i^r$ and the potential V_{QC} will ultimately determine the dynamics of our system

The Helmholtz Free Energy

- The Hamiltonian we choose must give an accurate approximation to the *Helmholtz Free Energy*

$$F(\beta) = -\frac{1}{\beta}\ln\int\frac{d\boldsymbol{p}d\boldsymbol{q}}{h_p^{3N}h_q^{3N}}e^{-\beta K(\boldsymbol{p})}e^{-\beta V(\boldsymbol{q})}$$

- *F is separable into the momentum and positional parts*

$$F = F_p + F_q$$

$$F_p(\beta) = -\frac{1}{\beta}\ln\int\frac{d\boldsymbol{p}}{h_p^{3N}}e^{-\beta K(\boldsymbol{p})}$$

$$F_q(\beta) = -\frac{1}{\beta}\ln\int\frac{d\boldsymbol{q}}{h_q^{3N}}e^{-\beta V(\boldsymbol{q})}$$

Try to accurately approximate both contributions

The Momentum Free Energy

- For all atoms of the same mass, *m*

$$F_{p,exact} = -\frac{1}{\beta}\ln\left(\frac{2\pi m}{\beta h_p^2}\right)^{3N/2}$$

- The QC momentum free energy is

$$F_{p,QC} = -\frac{1}{\beta}\ln\prod_{i=1}^{n_r}\left(\frac{2\pi m_i^r}{\beta h_p^2}\right)^{3/2}$$

- these can be made to exactly agree if we choose the masses correctly

$$m_i^r = \alpha^{n_i-1}m$$

$$h_p = \sqrt{\frac{2\pi m}{\alpha\beta}} \qquad \sum_{i=1}^{N_r}\alpha^{n_i-1} = N$$

The Positional Free Energy

$$F_q(\beta) = -\frac{1}{\beta}\ln\int\frac{d\boldsymbol{q}^r d\boldsymbol{q}^c}{h_q^{3N}}e^{-\beta V(\boldsymbol{q}^r,\boldsymbol{q}^c)}$$

- Formally integrate out the constrained atoms

$$F_q(\beta) = -\frac{1}{\beta}\ln\int\frac{d\boldsymbol{q}^r}{h_q^{3N}}e^{-\beta V_{QC}(\boldsymbol{q}^r)}$$

- Where the QC potential energy is

$$V_{QC}(\{\boldsymbol{q}^r\},\beta) = -\frac{1}{\beta}\ln\int d\boldsymbol{q}^c e^{-\beta V(\boldsymbol{q}^r,\boldsymbol{q}^c)}$$

But it is not practical to compute V_{QC} this way

Quasi-Harmonic Approximation

- V_{QC} is comprised of two parts:
  - (1) Zero T contribution with atoms at their mean positions
  - (2) Contribution due to the thermal fluctuations of atoms about mean positions
- *Treat using the Quasi-Harmonic Approximation of LeSar et al (1989):*

$$V_{QC}\left(\{\mathbf{q}^r\},\beta\right) = \sum_{i=1}^{N} E_i\left(\{\mathbf{q}^r\}\right) + \sum_{i=1}^{N-N_r} \frac{1}{2\beta} \ln \frac{\left\|\mathbf{D}_i\left(\{\mathbf{q}^r\}\right)\right\|}{\left(2\pi/\beta\right)^3}$$

T=0 contribution

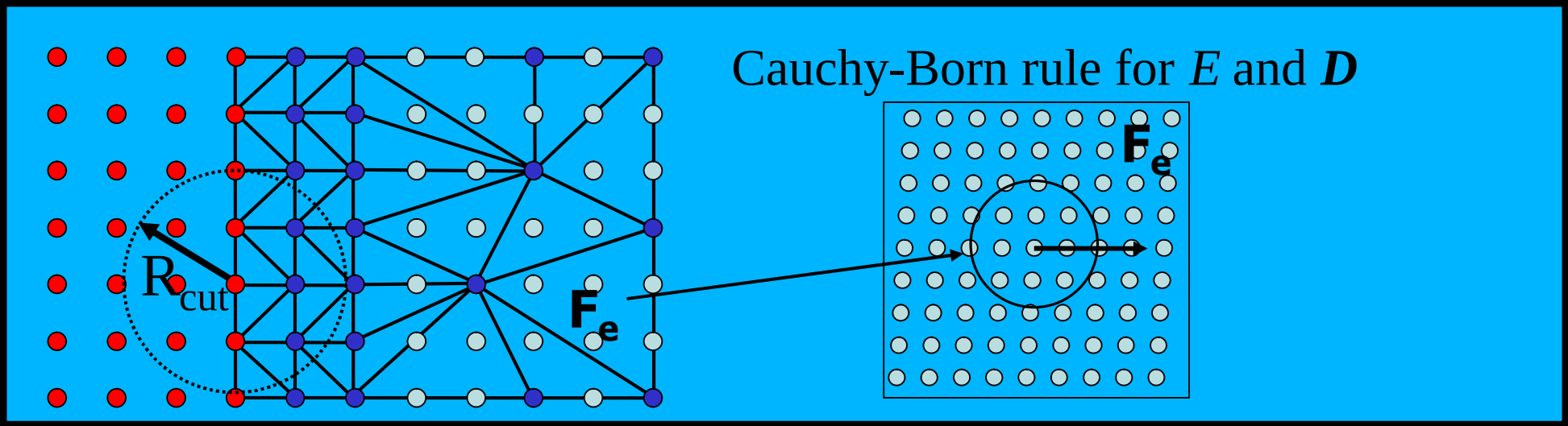
Entropic contribution, TS

Quasi-Harmonic Approximation

$$V_{QC}\left(\{\mathbf{q}^r\},\beta\right) = \sum_{i\in\mathrm{NL}} E_i + \sum_e \left[n_e E_{\mathrm{CB}}\left(\mathbf{F}_e\right) + \frac{n_e^c}{2\beta} \ln \frac{\left\|\mathbf{D}_{\mathrm{CB}}\left(\mathbf{F}_e\right)\right\|}{\left(2\pi/\beta\right)^d} \right]$$

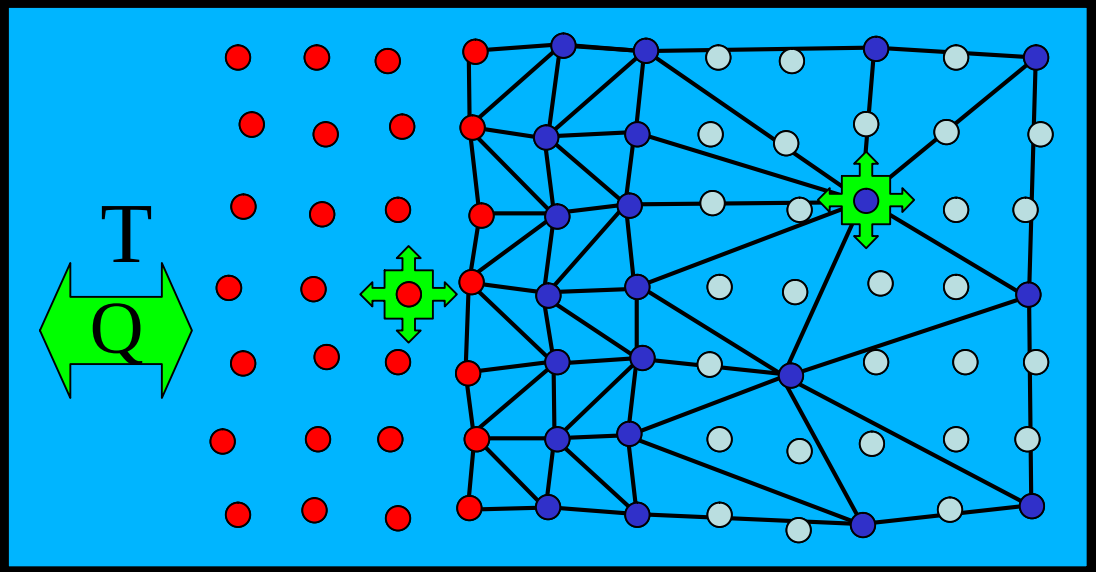
zero T energy contributions

Thermal motion of constrained atoms



Nosé-Poincaré Thermostat

- Couple the repatoms to a heat bath
- Problem:
  - motion of local repatoms is really the motion of the *mean positions* of the atoms.
  - Thus, the repatoms acquire an unphysical vibrational entropy of their own - "*ghost entropy*"
- This is negligible if $d_{elements}$ & d_{atoms}



Correcting the "Ghost Entropy"

- Ghost Entropy comes from an error in the Helmholtz Free Energy

$$F_{QC}(\beta) = \hat{F}_{QC}(\beta) + TS_{ghost}(\beta)$$

- Introduce a correction to V_{QC} :

$$\hat{V}_{QC}(\{\boldsymbol{q}^r\}, \beta) = V_{QC}(\{\boldsymbol{q}^r\}, \beta) - TS_{corr}(\{\boldsymbol{q}^r\}, \beta)$$

such that it approximately cancels out the ghost entropy.

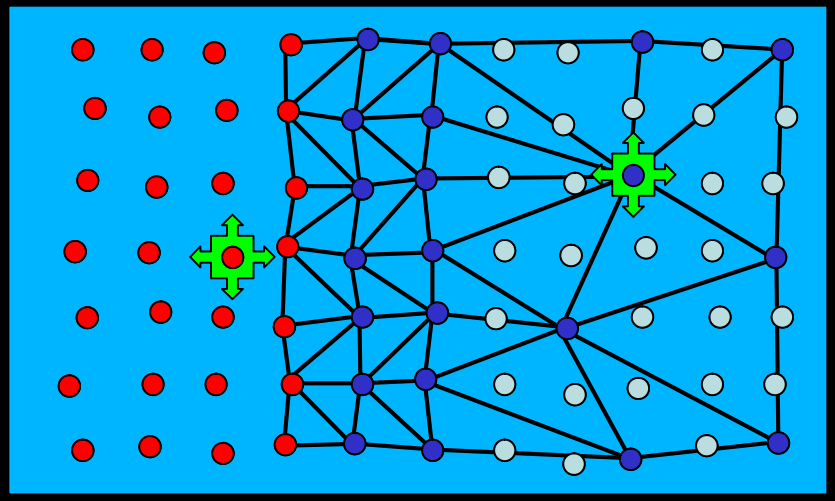
Correcting the "Ghost Entropy"

- Again, appeal to the quasi-harmonic model.
- Approximate ghost entropy as coming from harmonic flucutations about mean positions $\{\boldsymbol{q}^r\}$

$$S_{corr}(\{\boldsymbol{q}^r\},\beta) = -\frac{k_B}{2}\ln\frac{\|\mathbf{K}(\{\boldsymbol{q}^r\})\|}{(2\pi/\beta)^{3N_L}}$$

Stiffness Matrix from the FE

Number of local repatoms



Final Form of QC Potential Energy

$$\hat{V}_{QC}\left(\{\mathbf{q}^r\},\beta\right) =$$

$$\sum_{i\in\mathrm{NL}} E_i + \sum_e \left[n_e E_{\mathrm{CB}}\left(\mathbf{F}_e\right) + \frac{n_e^c}{2\beta} \ln \frac{\|\mathbf{D}_{\mathrm{CB}}\left(\mathbf{F}_e\right)\|}{\left(2\pi/\beta\right)^d} \right]$$

$$-\frac{1}{2\beta} \ln \frac{\|\mathbf{K}(\{\mathbf{q}^r\})\|}{\left(2\pi/\beta\right)^{3N_L}}$$

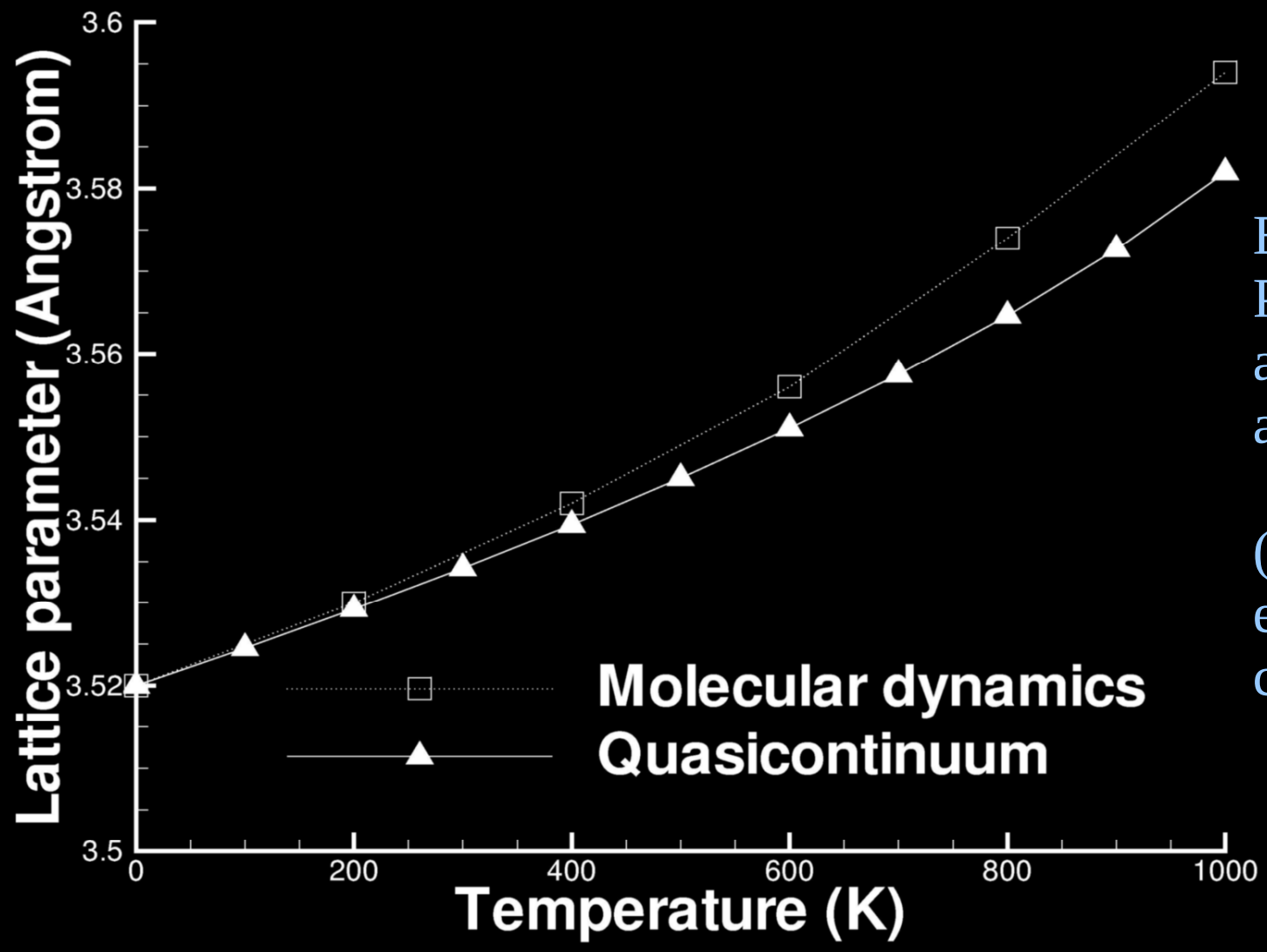
- Completes the definition of the QC Hamiltonian:

$$H_{QC}(\boldsymbol{q}^r, \boldsymbol{p}^r) = \hat{V}_{QC}(\boldsymbol{q}^r) + \sum_i^{N_r} \frac{\boldsymbol{p}_i^r \cdot \boldsymbol{p}_i^r}{2m_i^r}$$

Gives an accurate Helmholtz Free Energy:

$F_{QC}(T)=F_{MD}(T)+\mathcal{O}(T^2)$

Thermal Expansion: FCC Ni

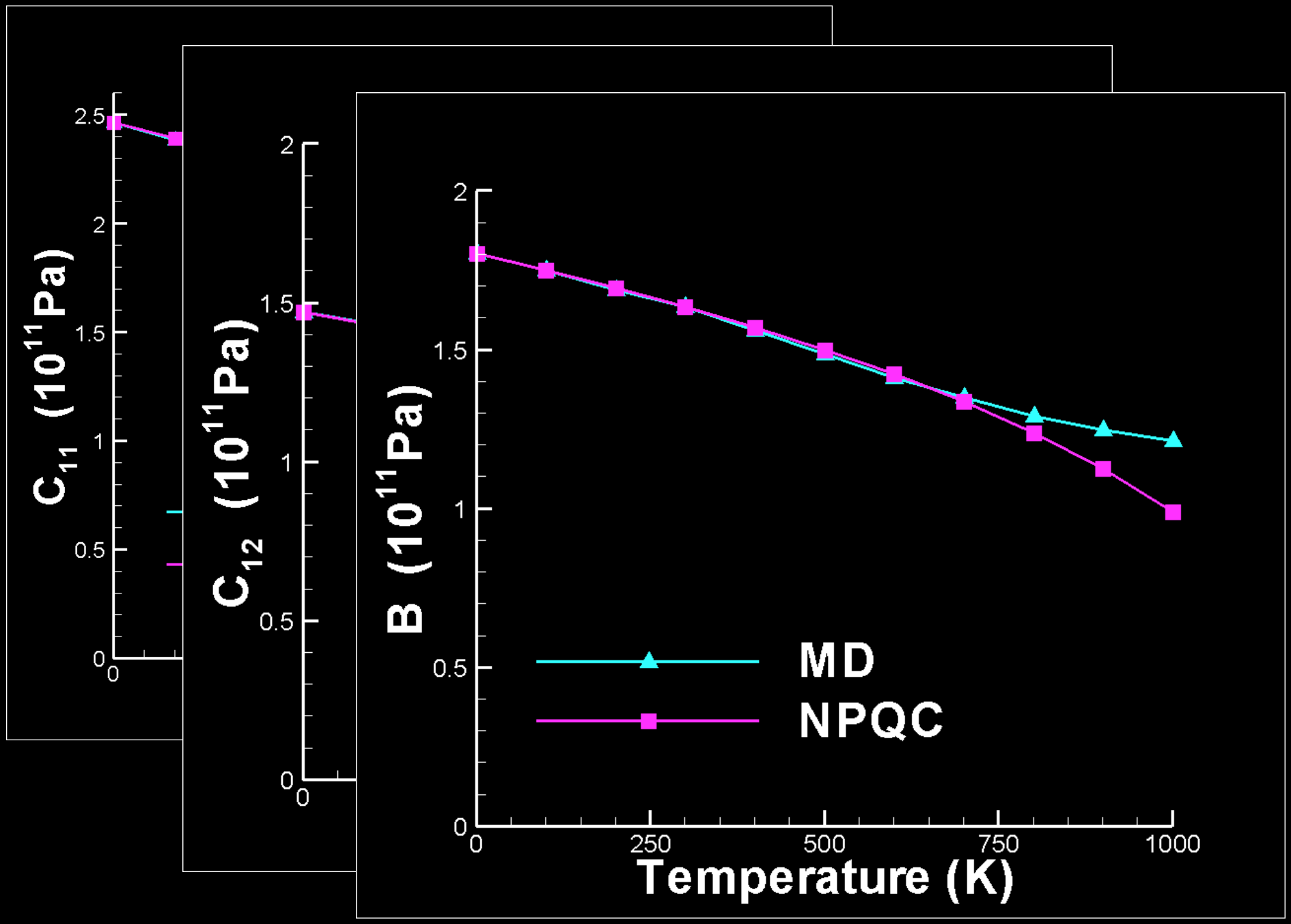


Error in lattice Parameter is about 0.5% at 1000K

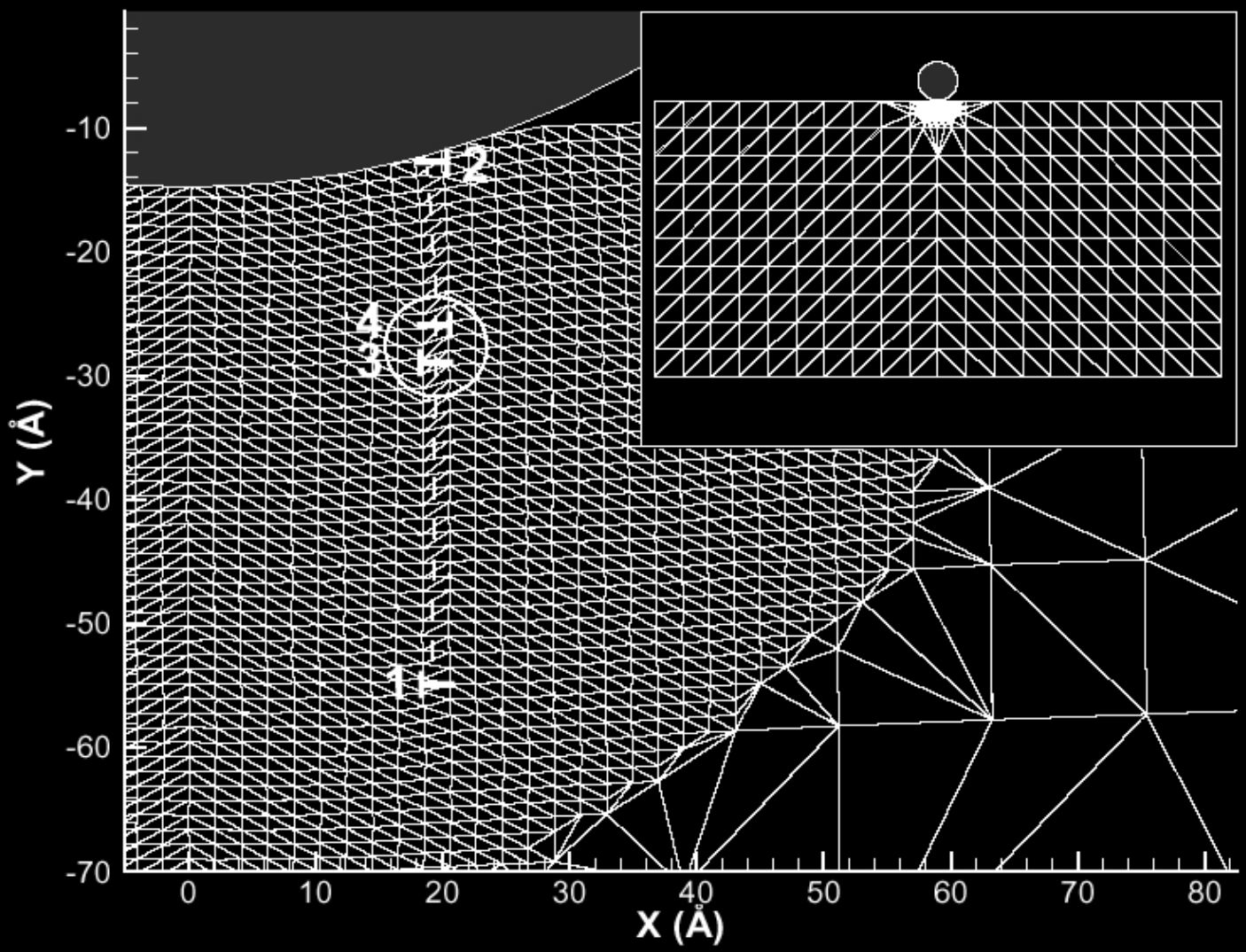
(10% error in expansion coefficient)

(Melting T is 1478K)

Elastic Constants

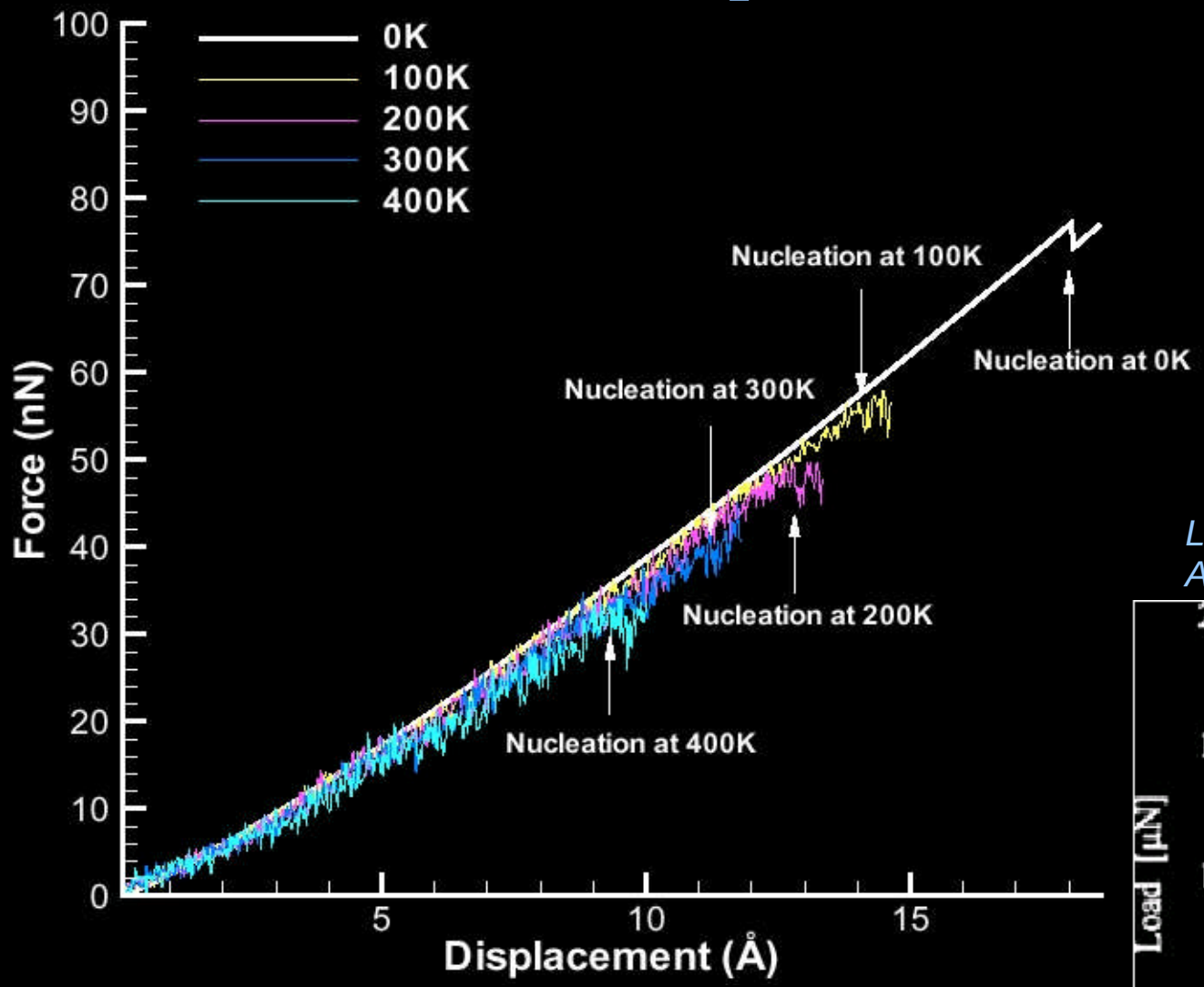


Finite Temperature Indentation

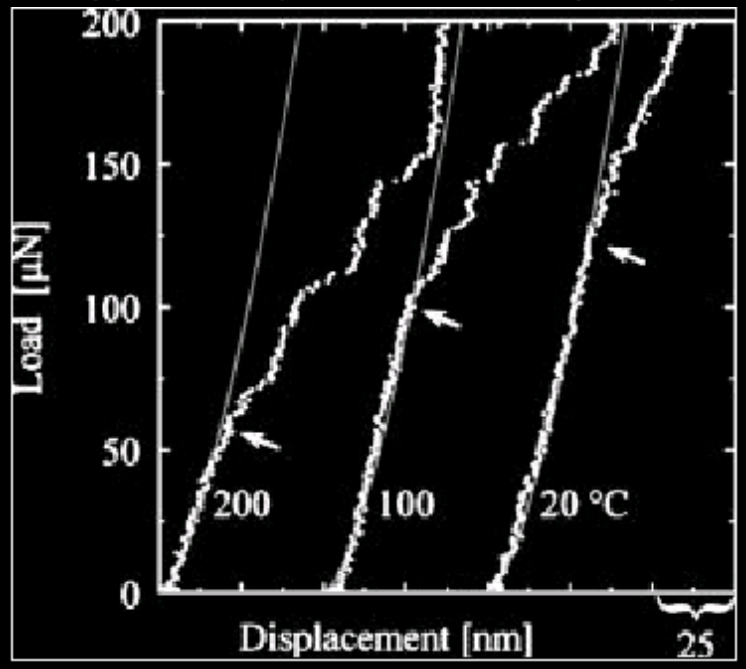


- Spherical Indenter, R=7nm
- Ni EAM Potential
- 5000 repatoms vs. 10^7 atoms
- Speed up approx. 10^3

Finite Temperature Indentation

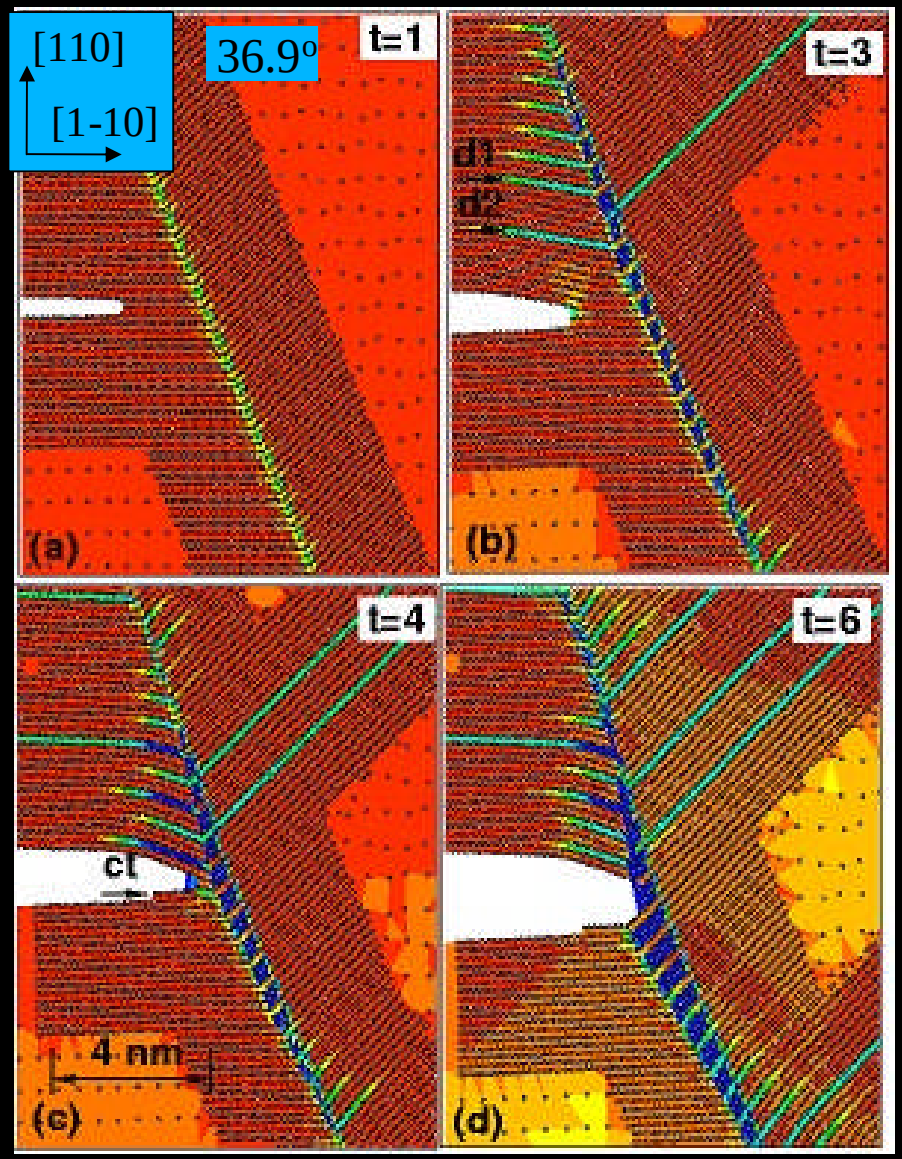


Lund, Hodge, and Schuh, Applieds Physics Letters 85 (2004)



Next Steps

- An important feature of QC: Automatic Adaption
  - Algorithm to determine if a region is “local” or “nonlocal”
  - Algorithm to determine if an element must be refined
- Non-equilibrium dynamics
  - Thermostat only the local region



(Miller et al, 1998)

Extension to Finite Temperature

- The original QC method is an equilibrium or "molecular statics" approach.
- We wish to extend this method *in a rigorous way to finite temperature.*

Thermostatted MD without all the atoms

- *We are not considering issues related to "far from equilibrium" MD/continuum coupling:*
  - *wave reflections*
  - *absorbing boundary conditions*