A New Multigrid Method for Molecular Mechanics

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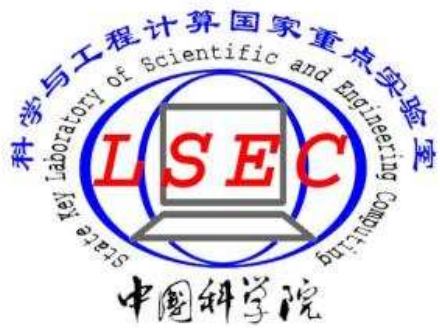

Joint with Jingrun Chen (ICMSEC)

&W. E (Princeton University)

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Outline

- Motivation and algorithm
- Examples
  - Homogeneous deformation:
    - tension in 1d
    - tension in Aluminum (3d)
    - shear in Aluminum
  - Inhomogeneous deformation:
    - Vacancy (Aluminum)
    - nanoindentation (Aluminum)
- Summary

Molecular Mechanics model of crystalline solids

$\boldsymbol{x}_i =$ position of i-th atom at undeformed state

$\boldsymbol{y}_i =$ position of i-th atom at deformed state

$$E^{\text{tot}}\{\boldsymbol{y}_1, \ldots, \boldsymbol{y}_N\} = \sum \left\{ V_2(\boldsymbol{y}_1, \ldots, \boldsymbol{y}_N) + V_3(\boldsymbol{y}_1, \ldots, \boldsymbol{y}_N) + \cdots \right\}$$

$$\{\boldsymbol{y}_1, \ldots, \boldsymbol{y}_N\} = \operatorname{argmin} \left\{ E^{\text{tot}}\{\boldsymbol{y}_1, \ldots, \boldsymbol{y}_N\} - \sum_{i=1}^{N} f(\boldsymbol{x}_i)\boldsymbol{y}_i \right\}$$

$f(\boldsymbol{x}_i) =$ external force at i-th atom

subject to certain boundary condition

Difficulty in solving molecular mechanics

- Total energy is nonconvex w.r.t. position of atoms; Fortunately, we aim for the physically relevant local minimizer instead of global minimizer (cf. geometry optimization problem: look for global minimization configuration)

- How to find the physically relevant local minimizer efficiently?
  - Larger the atomistic system, more the local minimizer
  - What is a good initial guess for iteration algorithms
  - Is there any $\mathcal{O}(N)$ algorithm=linear scaling algorithm

Local minimizer forest

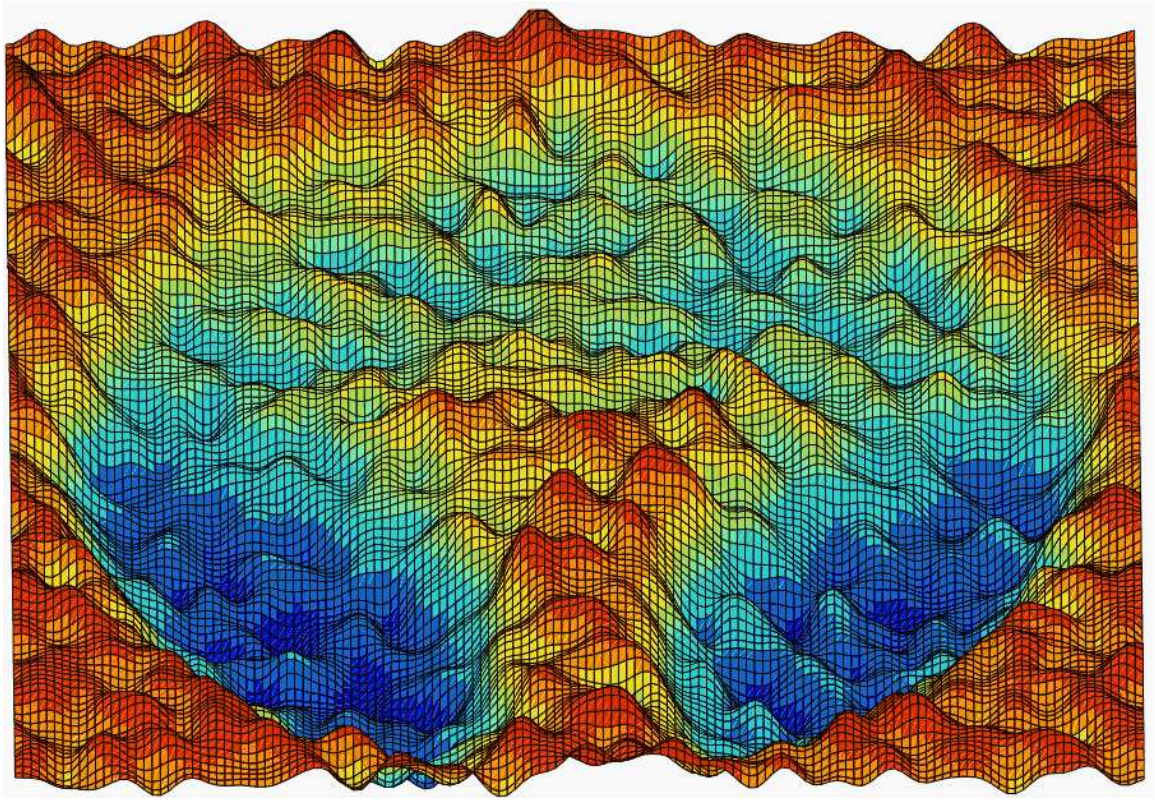


Fig. 1: Local minimizer forest (by courtesy of W.Q. Ren)

Conjugate gradient method is not linear scaling

- Iteration number of conjugate gradient method

$$n_{\text{it}} \simeq \sqrt{\kappa}|\log \text{TOL}| = \sqrt{\frac{\lambda_{\max}}{\lambda_{\min}}}|\log \text{TOL}| = \frac{\omega_{\max}}{\omega_{\min}}|\log \text{TOL}|$$

$\kappa =$ condition number TOL $=$ error tolerance

$\omega_{\min} =$ smallest phonon spectra frequency

$\omega_{\max} =$ highest phonon spectra frequency; either acoustic or optic

- For crystalline solids

$$n_{\text{it}} \simeq \frac{1/\epsilon}{1/L_{\max}}|\log \text{TOL}| = N^{1/d}|\log \text{TOL}|$$

$L_{\max} =$ largest linear dimensional of the system $L_{\max}^d \simeq N\epsilon^d$

$\epsilon =$ lattice constant; $N =$ total number of atoms

- Other iteration method: e.g., steepest descent method is even worse

Criteria for linear scaling algorithm

- The iteration number does not increase with the number of atoms
- The CPU time scales linearly
- Classic MG is actually a linear scaling method$\Longrightarrow$develop a MG like method for molecular mechanics model
- Compared with known method
  - Quasincontinuum method: more likes domain decomposition method; not linear scaling, only sublinear scaling; the same with HMM
  - Linear scaling method introduced by S. GOEDECKER, F. LANÇON & T. DEUTSCH, PHYS. REV. B 64(2001), 161102: first molecular mechanics + linear elasticity; no serious test

Algorithm

1. Solve Cauchy-Born elasticity model on the coarsest mesh; e.g., over $2 \times 2 \times 2$ in 3d

2. Interpolate solution on coarser mesh to finer mesh and solve Cauchy-Born continuum model or linearized model with this initial guess
   - CB elasticity for inhomogeneous deformation
   - Linearized model of CB elasticity (homogeneous deformation)

3. Using the CB elasticity solution as initial guess, solve atomistic model by conjugate gradient method or other iteration methods

4. All the linear systems are solved by Boomer AMG in Hypre: https://computation.llnl.gov/casc/linear_ssolvers/sls_hypre.html

Continuum model of solids

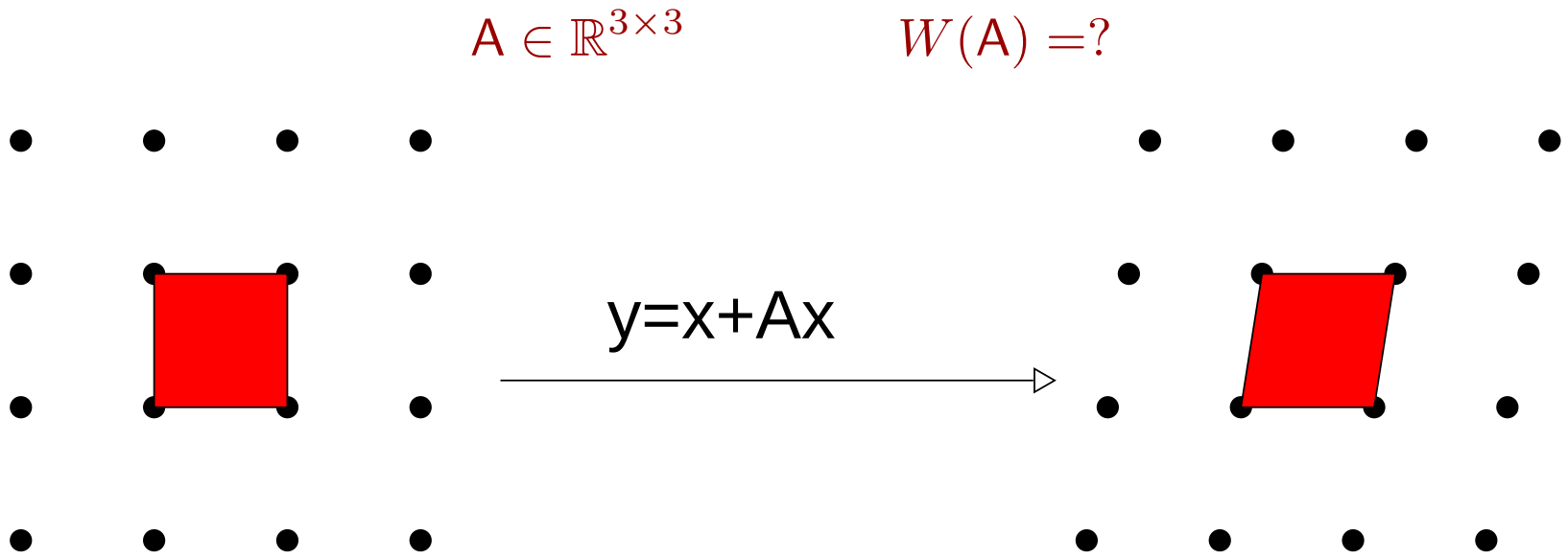
$\boldsymbol{u} : \Omega \to \mathbb{R}^3$, displacement field

$$I(\boldsymbol{u}) = \int_{\Omega} \Big(W\big(\nabla \boldsymbol{u}(\boldsymbol{x})\big) - \boldsymbol{f}(\boldsymbol{x})\boldsymbol{u}(\boldsymbol{x}) \Big) \, \mathrm{d}\boldsymbol{x}$$

$W =$ stored energy density $\quad$ $\boldsymbol{f} =$ external force

minimizing $I(\boldsymbol{u})$ in suitable space subject to certain boundary condition

Cauchy-Born rule: simple lattice



- $W(\mathsf{A}) =$energy of unit cell at the deformed configuration: uniformly deformed by A
- Example: 1d simple lattice with LJ potential

$$W_{\mathsf{CB}}(A) = \frac{\zeta^2(6)}{\zeta(12)}\left(|1+A|^{-12} - 2|1+A|^{-6}\right)$$

$\zeta =$ Riemann-zeta function

Molecular mechanics for tension in 1d

1d chain with Lennard-Jones potential

1. $\Omega = [0, 1]$
2. Initial guess is the undeformed state

$$x_i = (i-1)r^*, i = 1, \cdots, N-1, \qquad x_N = Nr^* + \delta$$

3. Boundary condition: ghost-atoms (2nd neighbor interaction)

$$x_0 = x_1 - r^* - \frac{1}{N}\delta \qquad x_{N+1} = x_N + r^* + \frac{N}{N-1}\delta$$

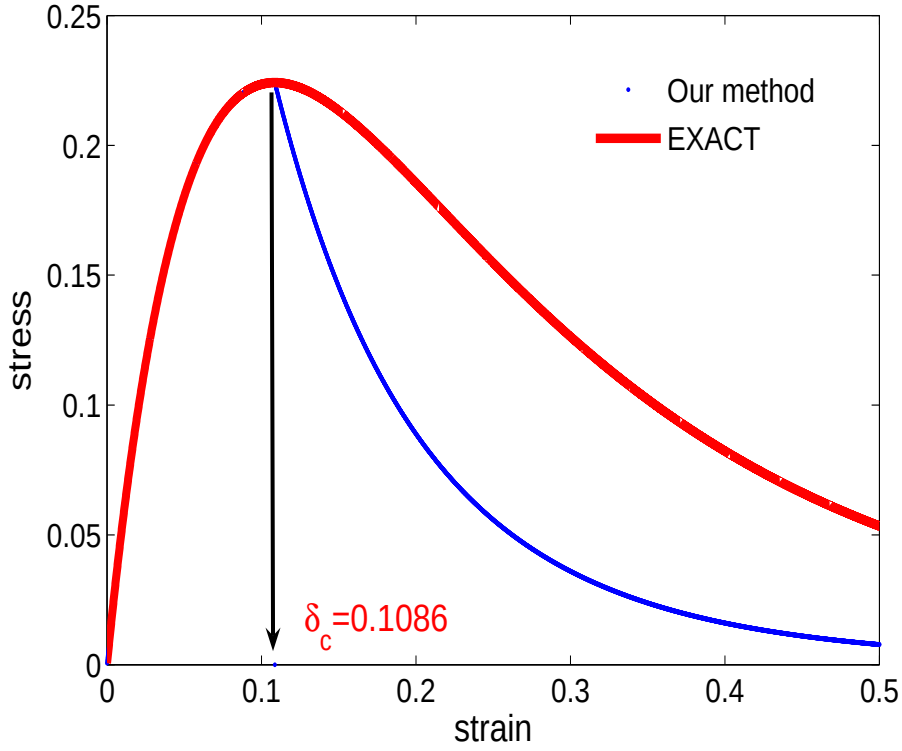
4. Iteration strategy: Newton method with line search: S.C. EISENSTAT & H.F. WALKER, SIAM J. SCI. COMPUT. 17(1996), 16–32
5. Parameter must be modified during the whole iteration process

neighbors	1st	2nd	3rd	4th	theoretic
δ_c	.0105	.0108	.0109	.0109	.1086

Our method

1. 4th neighbor interaction; Lennard-Jones potential; Newton method without line search for both continuum model and atomistic model

2. atom $N = 1024$; Mesh$H : 1/2 \Longrightarrow 1/256$



Red curve=Molecular Mechanics with elastic state as initial guess

Iteration number on different levels

strain(%)	≤ 2.13	≤ 7.19	≤ 9.34	≤ 10.31	≤ 10.72	≤ 10.86
$H = 1/2$	4	5	6	7	8	9
$H = 1/4$	1	1	1	1	1	1
$H = 1/16$	1	1	1	1	1	1
$H = 1/64$	1	1	1	1	1	1
$H = 1/256$	1	1	1	1	1	1
$N = 1024$	1	1	1	1	1	1

Total Iteration Number (compared with iteration number on atomistic scale)

$$\begin{aligned} \text{TIN} &\leq 9 \times \frac{2}{1024} + 1 \times \frac{4}{1024} + 1 \times \frac{16}{1024} \\ &\quad + 1 \times \frac{64}{1024} + 1 \times \frac{256}{1024} + 1 \\ &\leq 2 \end{aligned}$$

Linear scaling

The elastic state=$\delta \leq \delta_c$

N	1024	4096	16384	65536
TIN	≤ 2	≤ 2	≤ 2	≤ 2

Correct local minimizer or not

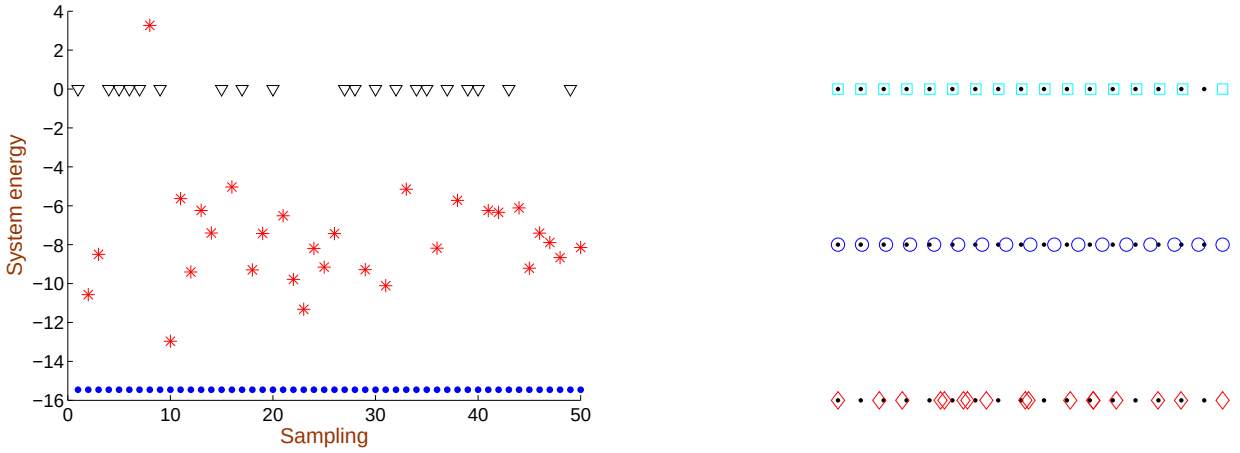


Fig. 2: Local minimizer or not; $E_\diamond = -8.4984; E_\circ = -15.4575; E_\square = -15.5492 =$ energy of initial state

3d: Aluminum

- Lattice=face-centered cubic (FCC)
- Potential=Embedded-atom method (EAM)

$$E = \sum_i \left(U_i(\rho_i) + \frac{1}{2} \sum_{j \neq i} \phi_{ij}(r_{ij}) \right)$$

$$\rho_i = \sum_{j \neq i} f_j(r_{ij}) = \text{electron density of ith atom}$$

$$\phi = \text{pairwise interaction potential}$$

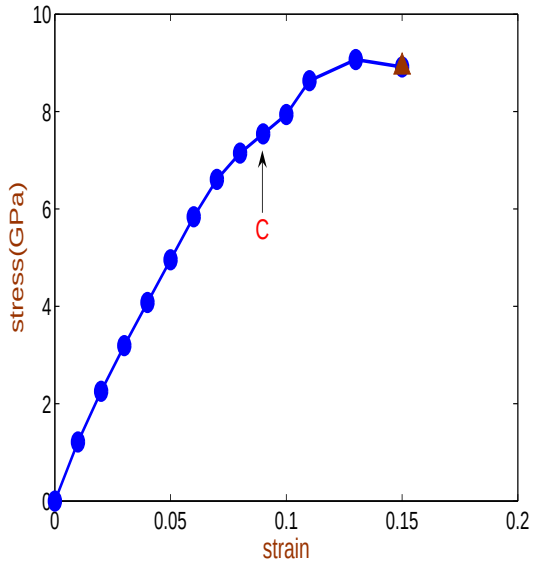
$$U_i = \text{glue function}$$

F. ERCOLESSI & J.B. ADAMS, EUROPHYS. LETT. 26 (1994), 583–588

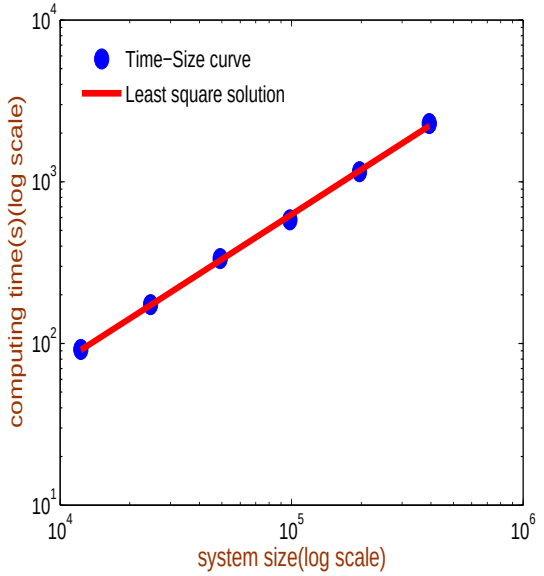
Tension in $[111]$ direction

- x axis=$[1\,1\,\bar{2}]$, y axis=$[\bar{1}\,1\,0]$, z axis=$[1\,1\,1]$
- Boundary condition: x, y= periodic boundary condition, z=tension, the same as $1-d$
- Solver: Newton-Raphson method for CB elasticity with line search
- Conjugate gradient (Fletcher-Reeves) method for molecular mechanics with line search (bisection method)

Stress strain curve for tension



(a) Stress-strain curve



(b) loglog plot for time vs. system size

- ● Linear scaling by our method
- Point C=Elasticity instability
- △ Linear scaling is lost
- mesh $= 2 \times 2 \times 2 \Longrightarrow 32 \times 32 \times 32$
- # of atom $= 1,572,864$

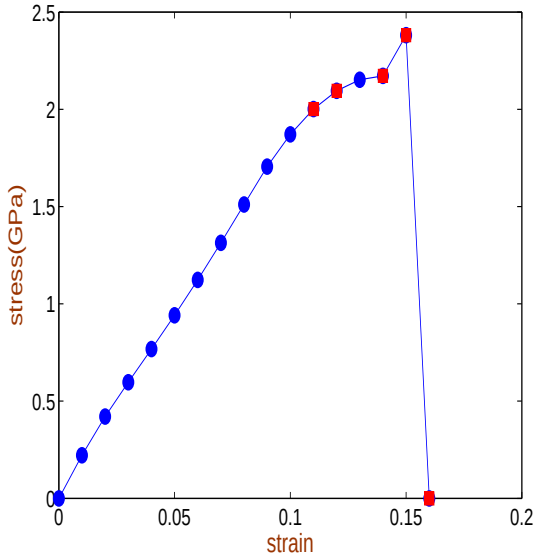
Linear scaling

$\delta \leq .08$=elastic state

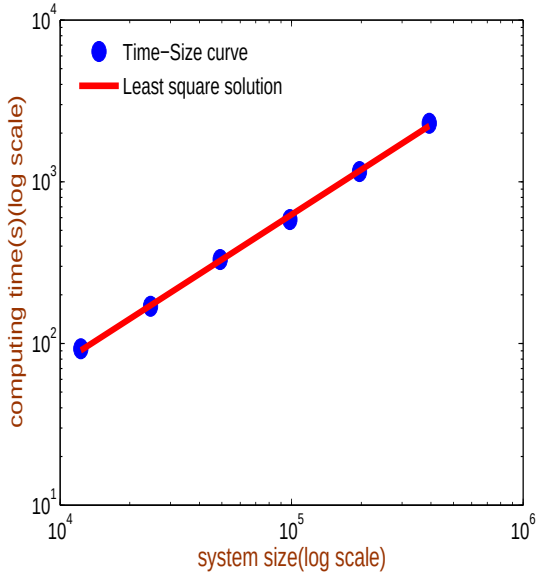
- TIN: total iteration number of Newton method
- TCG: total iteration number of conjugate gradient method

N	3072	24576	196608	1572864
TIN	≤ 2	≤ 2	≤ 2	≤ 2
TCG	≤ 2	≤ 2	≤ 2	≤ 2

Shear in $[11\bar{2}]$ direction



(c) Stress-strain curve



(d) loglog plot for time vs. system size

- ● Linear scaling
- □ Linear scaling is lost
- mesh $= 2 \times 2 \times 2 \Longrightarrow 32 \times 32 \times 32$
- # of atom $= 1,572,864$

Inhomogeneous deformation: vacancy under tension

- Set-up
  - simulation domain=$158 \times 91.2 \times 223.5$
  - $\#$ of atom=196608
  - tension $\delta = 2.235$
- mesh=$2 \times 2 \times 2 \Longrightarrow 16 \times 16 \times 16$
- Linear scaling is lost:

mesh	coarsest	other levels	atomistic scale
Iter. Num.	17	1	14

Table. 1: Iter. num. on different meshes & atomic scale

Localization

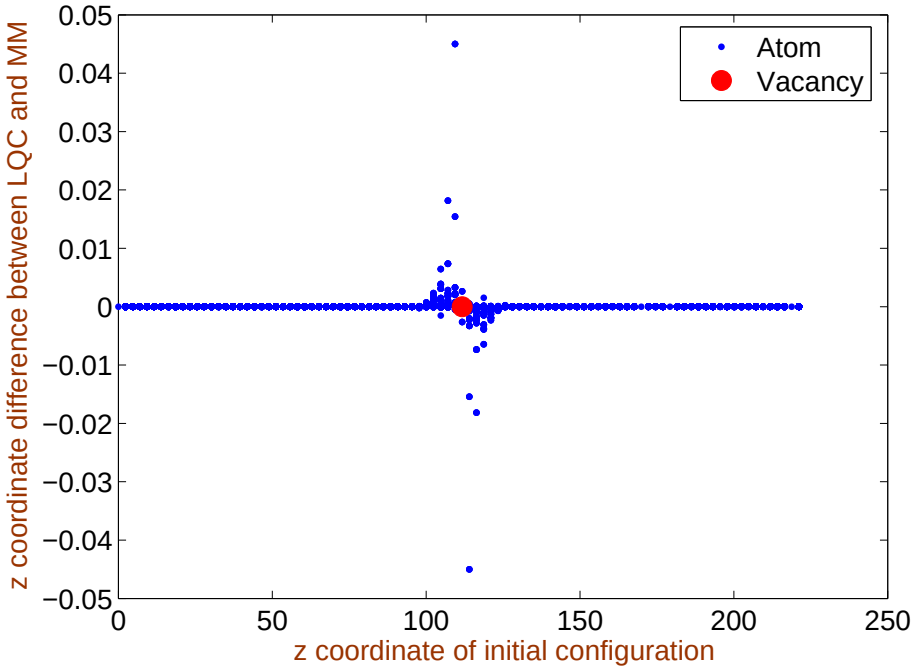


Fig. 3: Difference between LQC and MM in z direction under tension

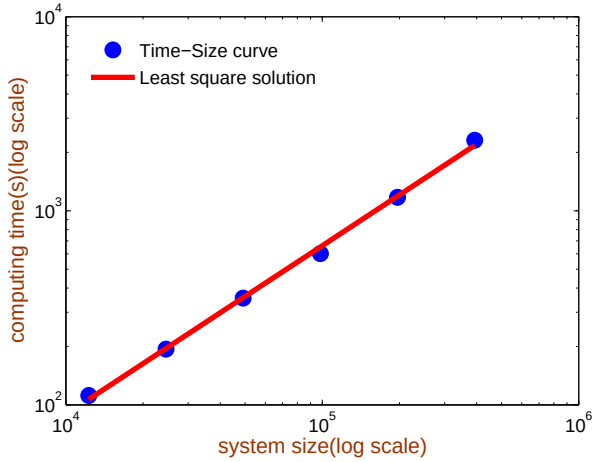
Error is localized !!!

Remedy: local correction

- Choose a box Ω around vacancy as simulation domain
- Use $\boldsymbol{y}_{\text{CB}}$ as the initial guess and carry out molecular simulation inside the box until to a given tolerance $\boldsymbol{y}_{\text{corr}}$
- Using

$$\boldsymbol{y} = \begin{cases} \boldsymbol{y}_{\text{CB}} & \text{if} \quad D \backslash \Omega \\ \boldsymbol{y}_{\text{corr}} & \text{if} \quad \Omega \end{cases}$$

as initial guess for molecular simulation



Inhomogeneous deformation: nanoindentation

x axis=$[111]$ y axis=$[1\,1\,\bar{2}]$ z axis=$[\bar{1}10]$

Ghost atoms setting: the same as tension in 3d

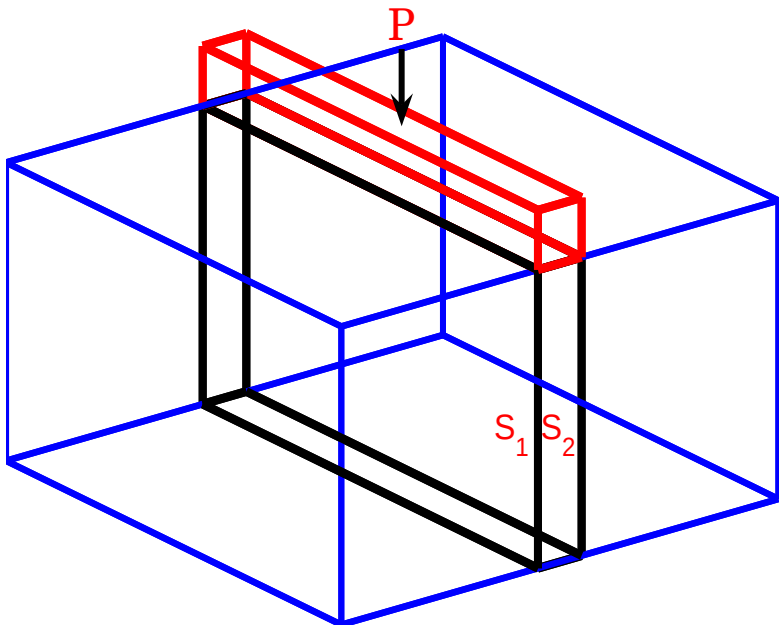
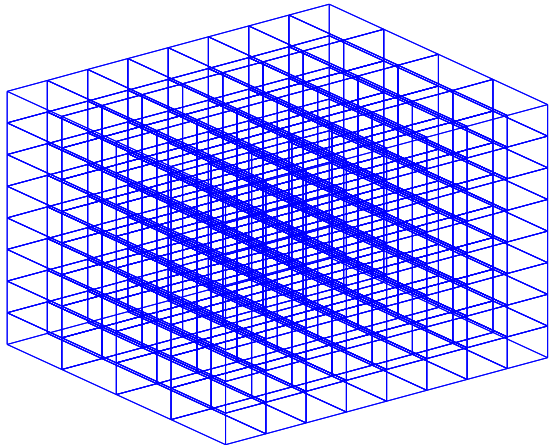
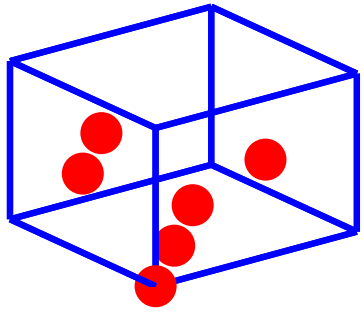


Fig. 4: system size$= 220$Å $\times\ 40$Å $\times\ 46$Å;indentor width$= 14$Å

Finite element mesh

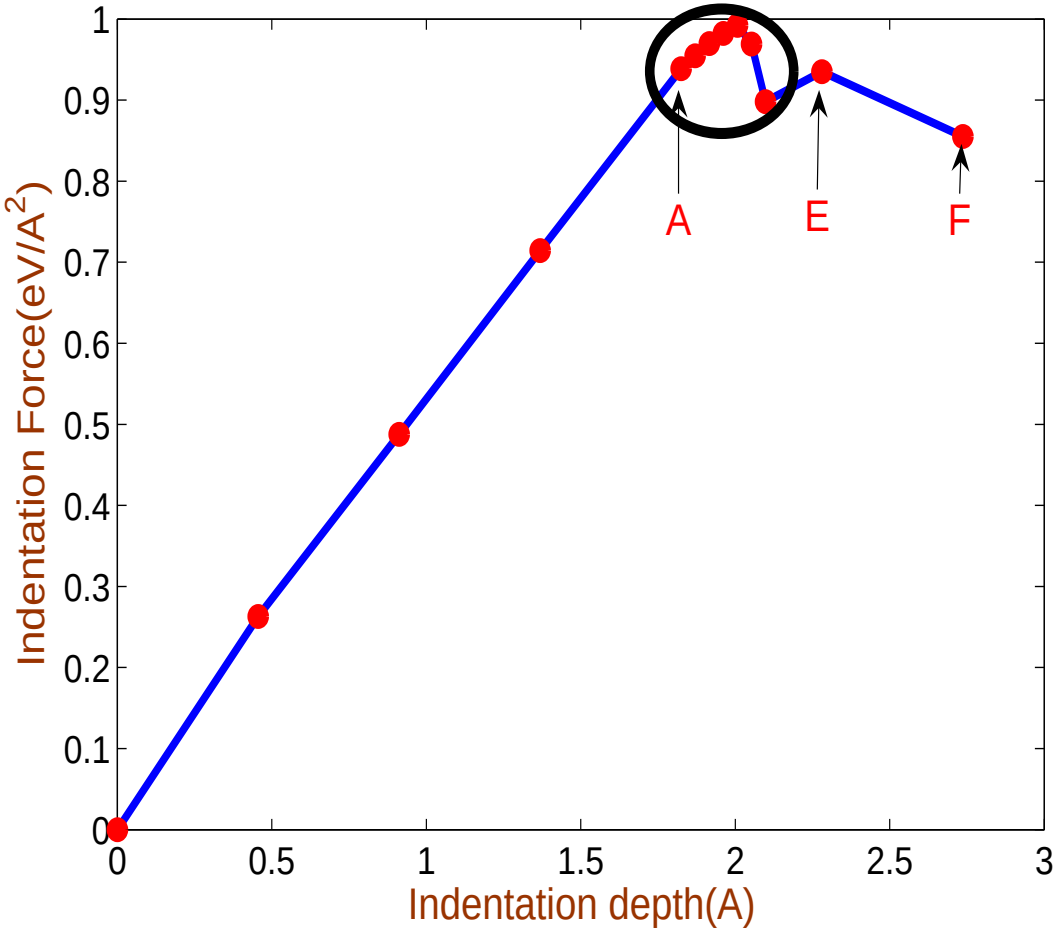
Schematic figure: Left, the finest mesh of LQC; Right, atoms(in red) in each element(in blue)



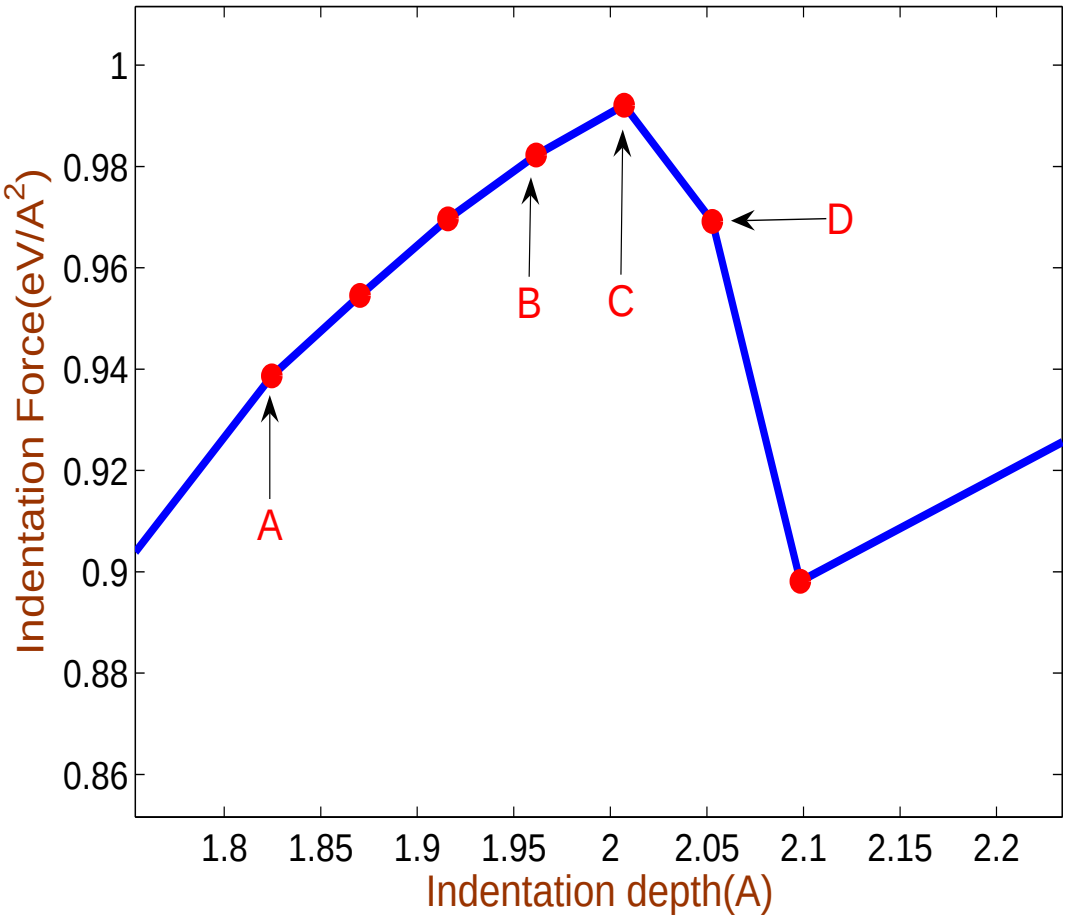


Load-displacement curve

System contains $24576 = 32 \times 8 \times 16 \times 6$ Al atoms



Zoom in


Indentation Force(eV/A^2)
1
0.98
0.96
0.94
0.92
0.9
0.88
0.86
1.8
1.85
1.9
1.95
2
2.05
2.1
2.15
2.2
Indentation depth(A)
A
B
C
D

Configurations A & B

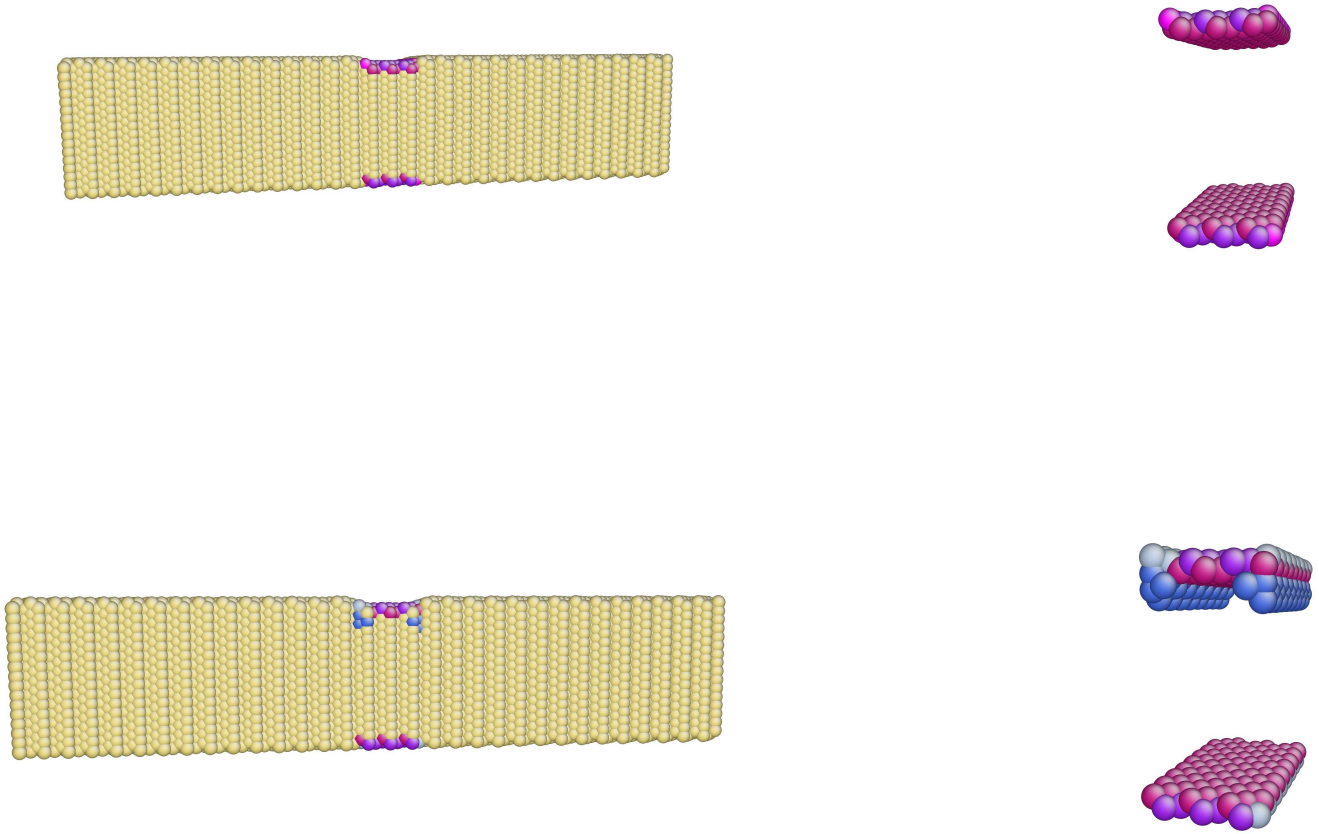


Fig. 5: Dislocation appears under the indentor

Configurations C & D

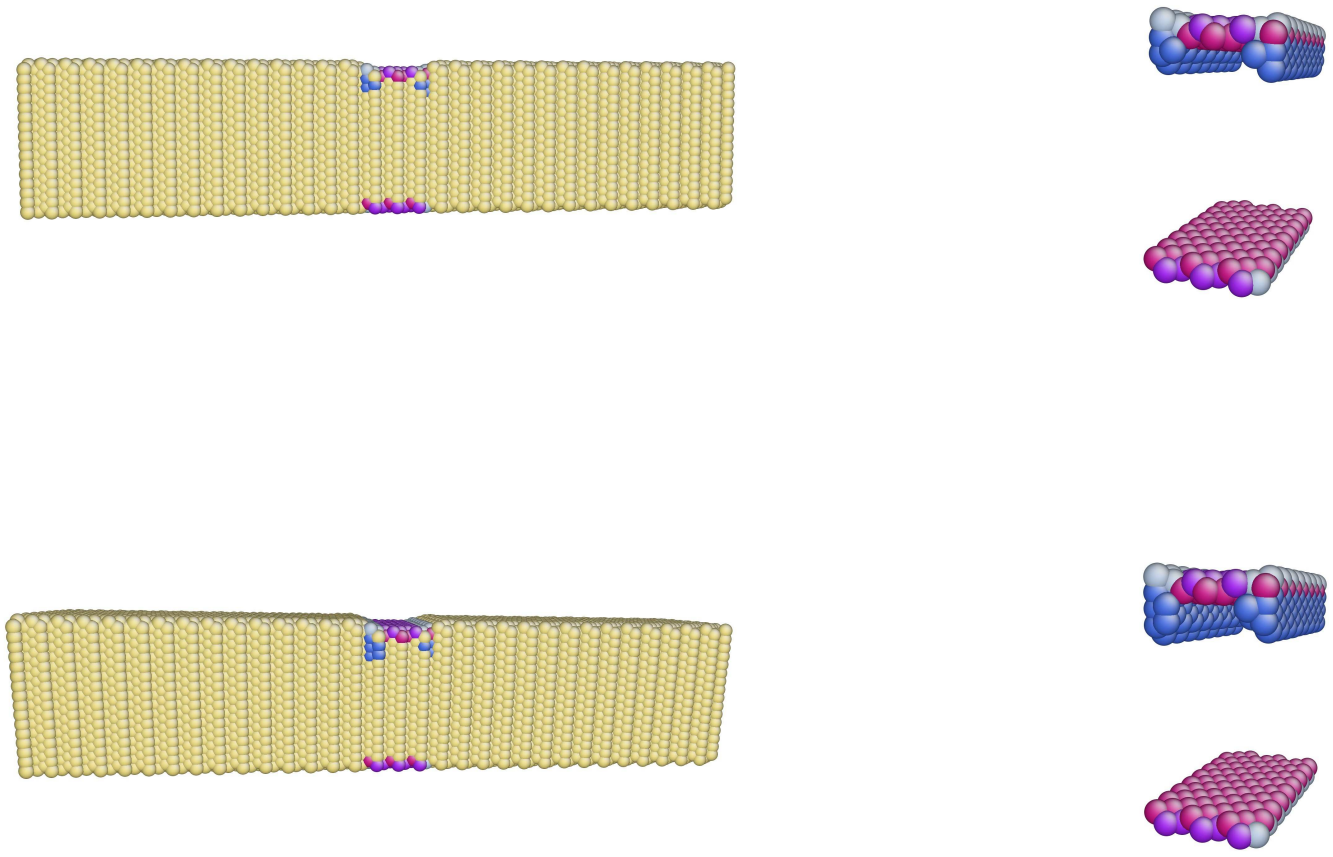


Fig. 6: Dislocation appears under the indentor

Configurations E & F

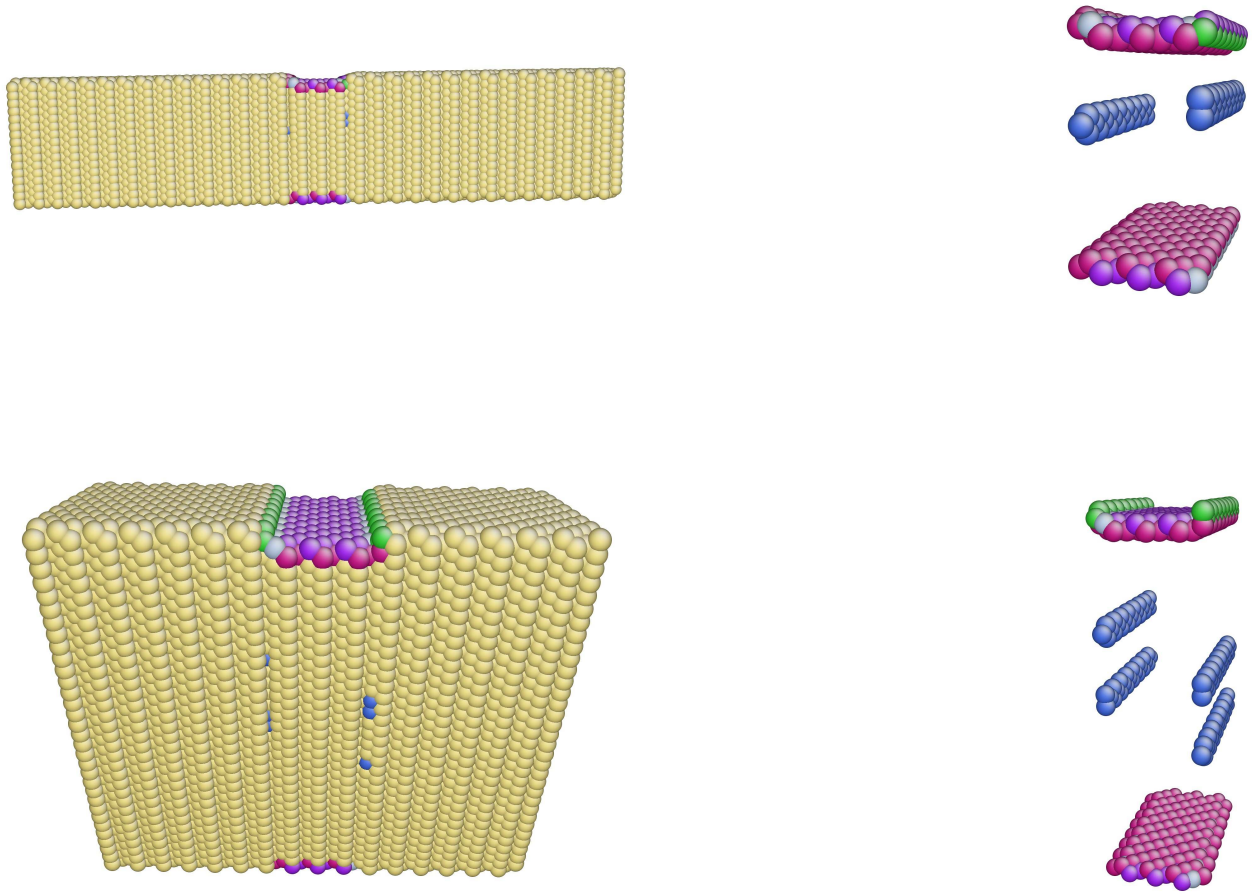


Fig. 7: Dislocation appears under the indentor

Discussion

Results

- Dislocation (B) comes in before the load-displacement curve decreases(d_C)
- Plasticity cannot be judged by load-displacement curve

Although the system is small, above results are conforming with with

- Numerical result: number of atom$= 2.5 \times 10^{11}$: (J. KNAP AND M. ORTIZ, PHYS. REV. LETT. 90 (2003) , 226102)
- Experiment result: (A.M. MINOR, S.A. SYED ASIF , Z.W. SHAN, E.A. STACH, E. CYRANKOWSKI, T.J. WYROBEK & O.L. WARREN, NATURE MATERIALS 5 (2006), 697-702))

Iteration numbers

Total Newton iteration numbers ≤ 2

TCG: Total CG iteration numbers on atomic scale

displacement(Å)	$\leq d_C$	$> d_C$
TCG	$46 \sim 85$	$113 \sim 188$

- Before d_C, TCG increases slowly and No jump for TCG
- After d_C, jump for TCG

Q Plasticity comes in after d_C?

Two stability regions

Define

$$\mathscr{R} := \{ \mathsf{A} \in \mathbb{R}^{d\times d} \mid \det(\mathsf{I}+\mathsf{A}) > 0 \}$$

Elasticity stiffness tensor $\mathsf{C}(\mathsf{A}) = D^2 W_{\mathsf{CB}}(\mathsf{A})$

$$\mathscr{O}_1(\Lambda) := \{ \mathsf{A} \in \mathscr{R} \mid \mathsf{C}(\mathsf{A})(\vec{\xi}\otimes\vec{\eta},\vec{\xi}\otimes\vec{\eta}) \geq \Lambda |\vec{\xi}|^2 |\vec{\eta}|^2 \}$$

$$\mathscr{O}_2(\Lambda_1,\Lambda_2) := \{ \mathsf{A} \in \mathscr{R} \mid \omega_a(\mathsf{A},\vec{k}) \geq \Lambda_1 |\vec{k}| \;\&\; \omega_o(\mathsf{A},\vec{k}) \geq \Lambda_2/\epsilon \}$$

$\vec{k} \in$ 1st Brillouin zone=foundamental domain

Theoretical result[E–M, ARMA, 07]

If $\vec{0} \in \mathscr{O}_1$ and $p > d, \exists K, R$ s.t. for any $\|\boldsymbol{f}\|_{L^p} \leq K$, $\exists |\boldsymbol{u}_{\text{CB}}$ of the continuum problem s.t. $\|\boldsymbol{u}_{\text{CB}}\|_{W^{2,p}} \leq R$, and $\boldsymbol{u}_{\text{CB}}$ is a $W^{1,\infty}-$local minimizer

If $\vec{0} \in \mathscr{O}_2$ and $p > d, \exists K$ s.t. for any $\boldsymbol{f} \in W^{6,p}(\Omega; \mathbb{R}^d)$ and $\|\boldsymbol{f}\|_{L^p} \leq K$, then the atomistic model has a local minimizer $\{\boldsymbol{y}^\epsilon\}$ that satisfies

$$\| \boldsymbol{y}^\epsilon - \boldsymbol{y}_{\text{CB}} \|_d \leq C\epsilon$$

where $\boldsymbol{y}_{\text{CB}} = \{\boldsymbol{y}_{\text{CB}}\}_j = \boldsymbol{x}_j + \boldsymbol{u}_{\text{CB}}(\boldsymbol{x}_j)$, $\epsilon =$ lattice constant, and $\| \cdot \|_d$ is a discrete H^1 norm

Summary of Our method

- In elastic regime
  - Physically reasonable configuration
  - Insensitive to parameters of nonlinear iteration method
  - Linear scaling of the computing complexity
  - Linear scaling is recovered if local correction is added (Vacancy)
- Out of this regime
  - Physical reasonable result (nanoindentation)
  - unphysical configuration (tension)
  - Linear scaling is lost (tension, nanoindentation)
- An efficient way to find physically relevant local minimizer; Hidden mechanism: automatically bypass many unphysically local minimizer

Conclusion

- Drawbacks of the algorithm:
  - Inefficient for inhomogeneous deformation
  - Adaptive FEM is required in solving Cauchy-Born elasticity
- More realistic applications are needed: e.g., other nanoindentation simulation; dislocation/fracture (Maradudin, Tewary)
- Theoretically understand of this alg., e.g.,
  - Mechanism for bypassing local minimizer
  - Rigorous prove the algorithm is linear scaling, at least for homogeneous deformation; vacancy is much harder

Extension and perspective

- Wider implementation: lattice equations in many other fields
  - Repetitive structure in solid mechanics (A.K. Noor)
  - Power grid (Babuska, Sauter)
  - Protein folding lattice model (Thumas, 1995)
  - Ising model
  - Quantum chromodynamics (lattice QCD)
  - Integer programming problem in operations research
  - Lattice-based cryptography and communication theory
- Difficulty
  - Do all the aforementioned problems concern local minima
  - Does there exists an efficient macroscopic model as CB elasticity in crystalline solids (e.g., finite difference homogenization for linear lattice equation)