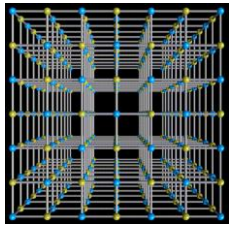
Coarse Graining of Electric Field Interactions with Materials



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Adviser

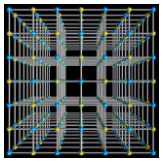
Dr. Kaushik Dayal

Funded by
Army Research Office

Research Talk

Indian Institute of Technology Madras

*Figure : http://www.themolecularuniverse.com/mile/mile1.htm



About me

- **B.E. from Government Engineering College, Raipur, Chattisgarh 2006 - 2010**
- **M.E. from Indian Institute of Science, Bengaluru, Karnataka 2010 - 2012**

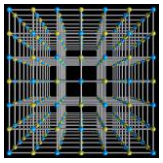
  Adviser: Dr. C. S. Jog, Department of Mechanical Engineering

  Thesis: A monolithic strategy for fluid-structure interaction in compressible flow.
- **Ph.D. from Carnegie Mellon University, Pittsburgh, USA 2012 – 2016**

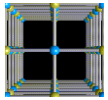
  Adviser: Dr. Kaushik Dayal, Civil and Environmental Engineering

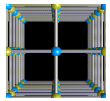
  Thesis: Coarse graining of electric field interactions with materials.
- **Post-doc from Louisiana State University, Baton Rouge, USA**

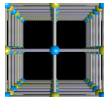
  Adviser: Dr. Robert Lipton, Department of Mathematics

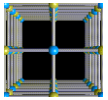


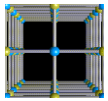
Overview of the talk

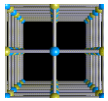

Goal and introduction


Continuum limit calculations


Multiscale formulation

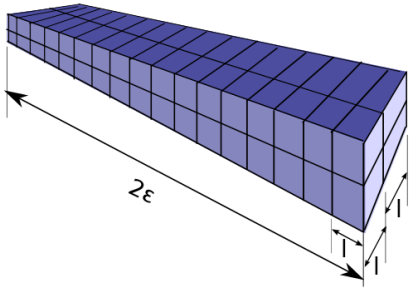

Results

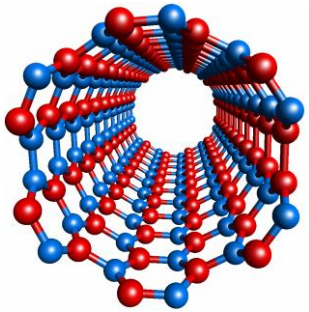

Discussions


Future work

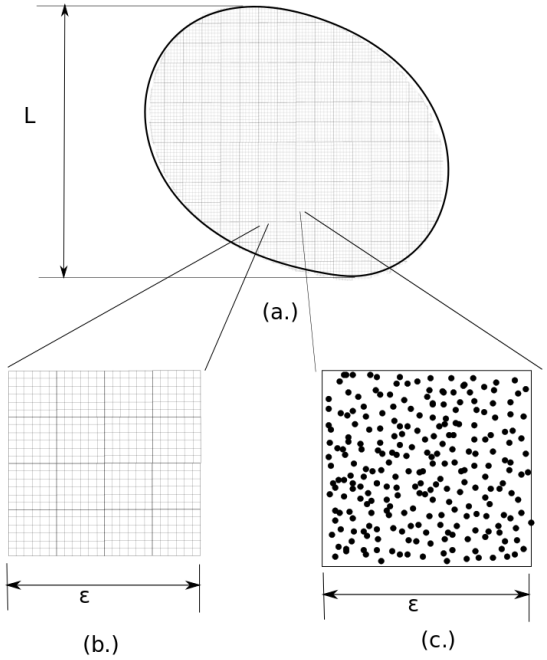
Goal

1. Electrostatics in nanostructures

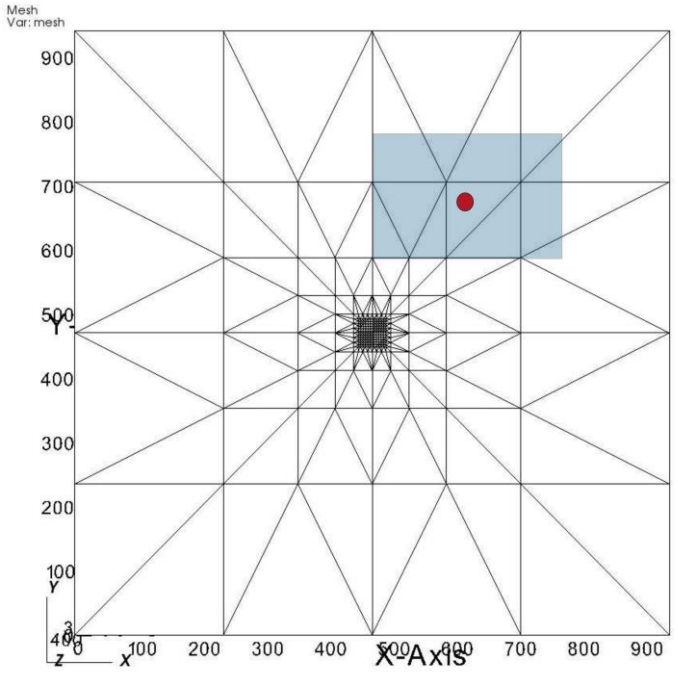




2. Electrostatics in random media



3. Multiscale method for ionic solids at finite temperature

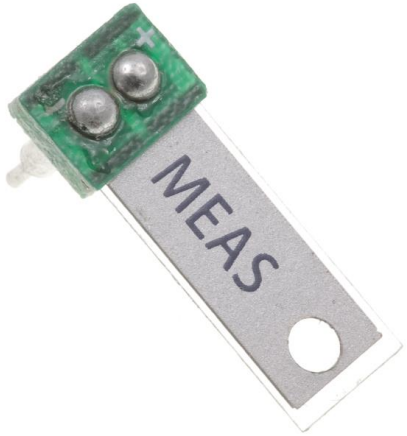


Motivation

- Electrostatics interaction
  - Storage devices
  - Ferroelectric RAM
  - Piezoelectric sensors
- Finite temperature
  - Thermal fluctuations of atoms
  - Coupling of deformation, electric field with temperature

(a) Hard drive

(b) Ferroelectric RAM



(c) Piezoelectric sensor

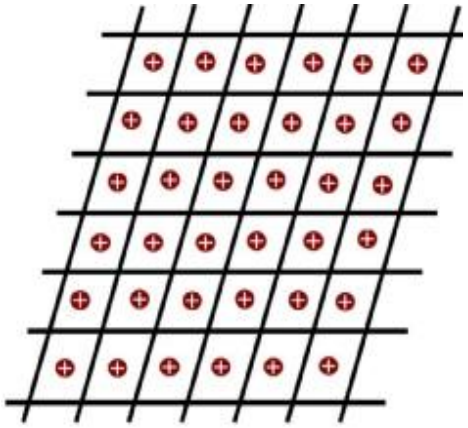
(a) http://phys.org/news/2009-10-hard_1.html
(b) http://abdulmoeez55.blogspot.com/2015/12/ferroelectric-ram.html
(c) http://www.meas-spec.com/product/piezo/MiniSense_100NM.aspx

Long range interactions

$$\text{Energy density at X} = \int_Y G(X,Y) f(Y) dY$$

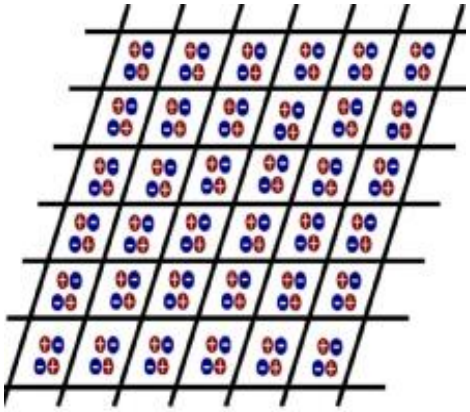
Field at X due to charge/dipole at Y

Charge/dipole at Y



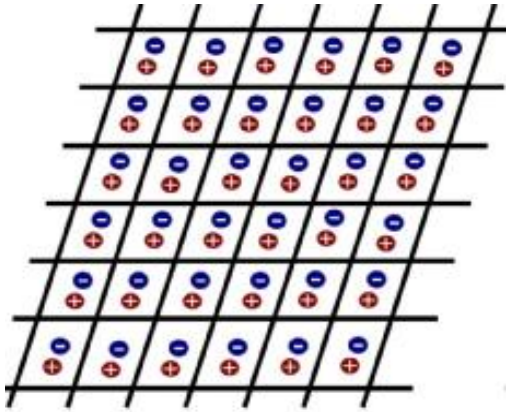
Charge distribution

$$W \approx \sum_{r=1}^{\infty} 1/r \times r^2 = \sum_{r=1}^{\infty} r$$



Quadrupole distribution

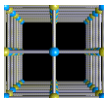
$$W \approx \sum_{r=1}^{\infty} 1/r^5 \times r^2 = \sum_{r=1}^{\infty} 1/r^3$$



Dipole distribution

$$W \approx \sum_{r=1}^{\infty} 1/r^3 \times r^2 = \sum_{r=1}^{\infty} 1/r$$

Figure: Jason Marhsall and Kaushik Dayal



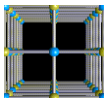
Long range interactions…

Linear Elasticity → $W(\boldsymbol{x}) = \frac{1}{2}\boldsymbol{\epsilon}(\boldsymbol{x}) \cdot \mathbb{C}\boldsymbol{\epsilon}(\boldsymbol{x})$

Electrostatics → $W(\boldsymbol{x}) = \boldsymbol{\nabla}\phi(\boldsymbol{x}) \cdot \boldsymbol{\nabla}\phi(\boldsymbol{x})$

$$\boldsymbol{\nabla} \cdot \boldsymbol{\nabla}\phi = \boldsymbol{\nabla} \cdot \boldsymbol{p}$$

Energy density depends on polarization field over whole material domain



Long range interactions…

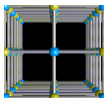
$$\mathrm{E} = V(\boldsymbol{q}) + \frac{1}{2}\sum_{i=1}^{N}\sum_{j=1}^{n}\frac{Q_i Q_j}{|\boldsymbol{q}_i - \boldsymbol{q}_j|}$$

Continuum limit of electrostatic energy

$$\mathrm{E} = V(\boldsymbol{q}) + \frac{1}{2}\int_{\mathbb{R}^3} |\boldsymbol{\nabla}\phi|^2$$

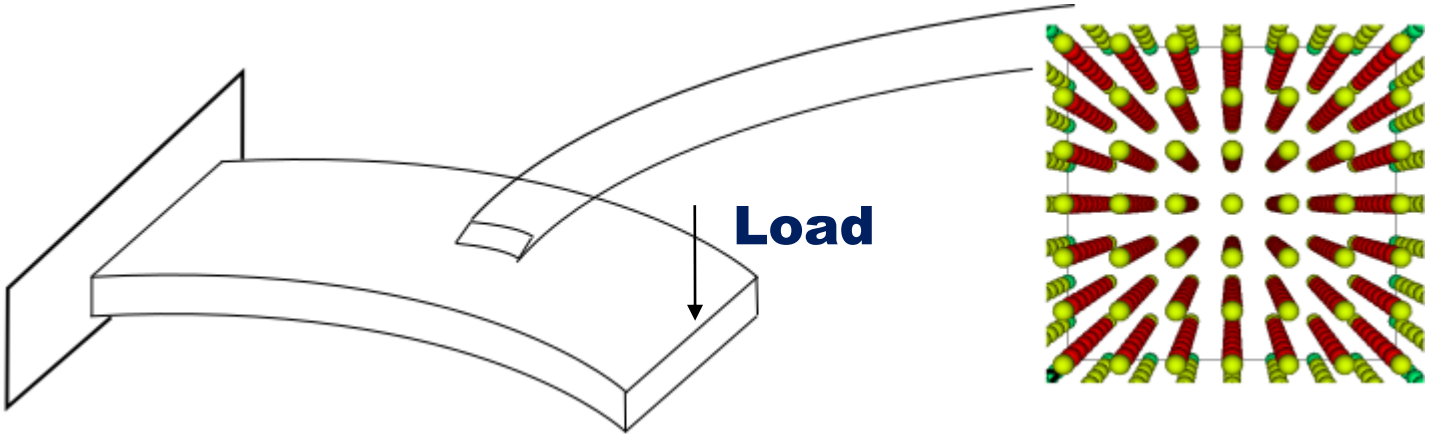
$$\nabla^2\phi = \boldsymbol{\nabla}\cdot\boldsymbol{p} \quad \in \mathbb{R}^3, \boldsymbol{p} = \mathbf{0} \ \in \mathbb{R}^3 - \Omega$$

$\boldsymbol{p}$: polarization field in a material



Multiscale in a material

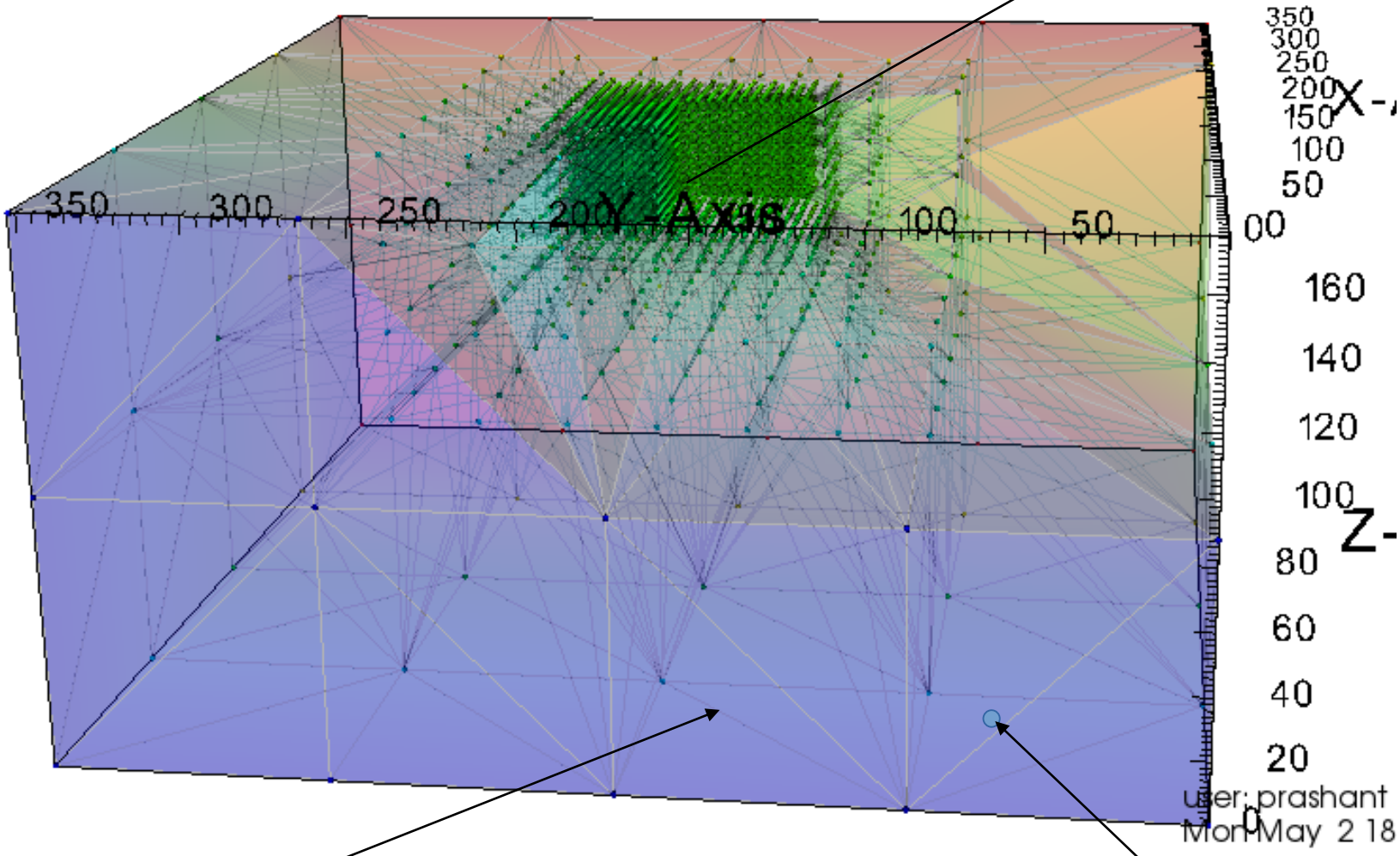
(a) macroscale (b) atomic scale



Piezoelectric material

- Deformation is slowly varying field
- Displacement of charges cause change in electric field
- Change in electric field causes deformation of material
- Except near loading, variation of deformation field is at higher scale than the scale at which atoms displace

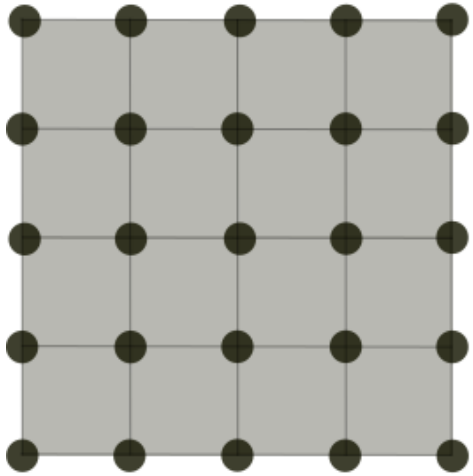
Multiscale method



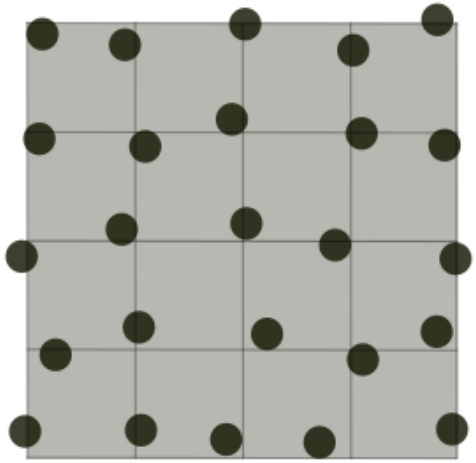
Coarse mesh as we move away from defect

We use interpolation for atoms within element

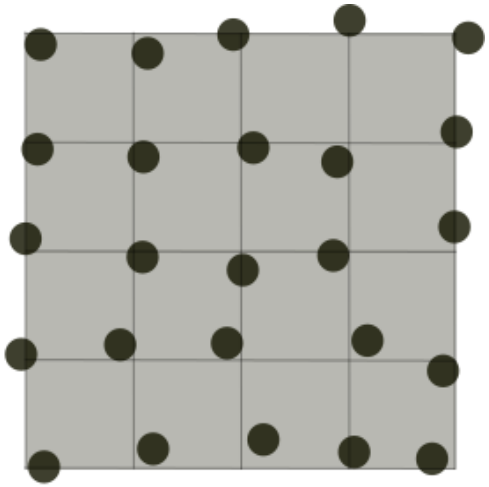
Finite temperature



Time t_1



t_2



t_3

- Observation of property at time scale >> time scale at which system change state
- Phase average → need probability distribution function p
- for each state → p is the probability of system being at that state

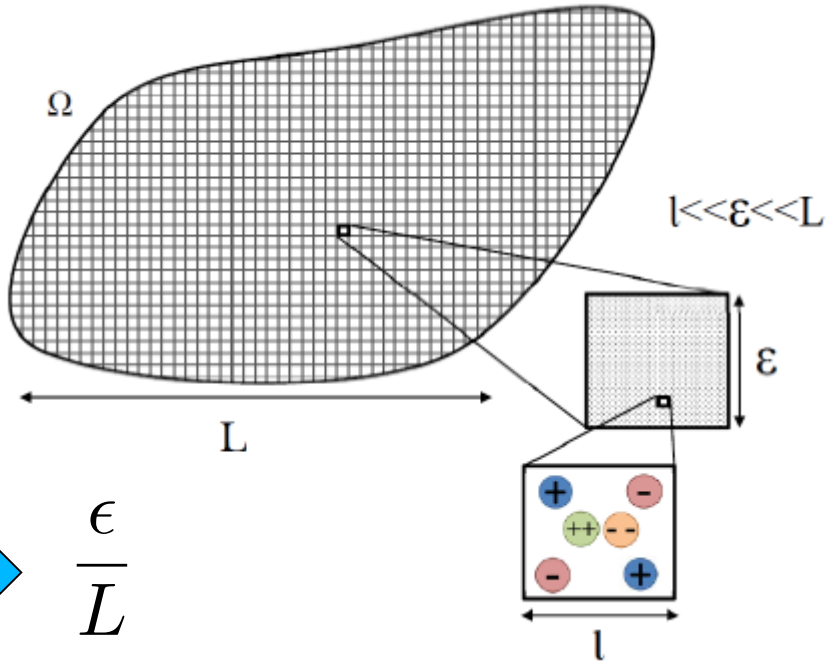
$$f_{observed} := \int_{\Gamma} f(\mathbf{q},\mathbf{p})p(\mathbf{q},\mathbf{p})d\mathbf{q}d\mathbf{p} \qquad \mathrm{p}(\mathbf{q},\mathbf{p}) = \exp[-\frac{H(\mathbf{q},\mathbf{p})}{\beta T}]$$

Position of all atoms

Momenta of all atoms

Length scales

- Continuum Length scale : L
- Size of material point : ϵ
- Atomic spacing : l



Macroscopic field vary at the scale ⇒ $\frac{\epsilon}{L}$

Interested in limit

- **Continuum mechanics** → $\epsilon << L$

  Fields vary at fine scale compared to size of material

- **Continuum limit approximations** → $l << \epsilon$

  Atomic spacing is fine compared to scale at which fields vary

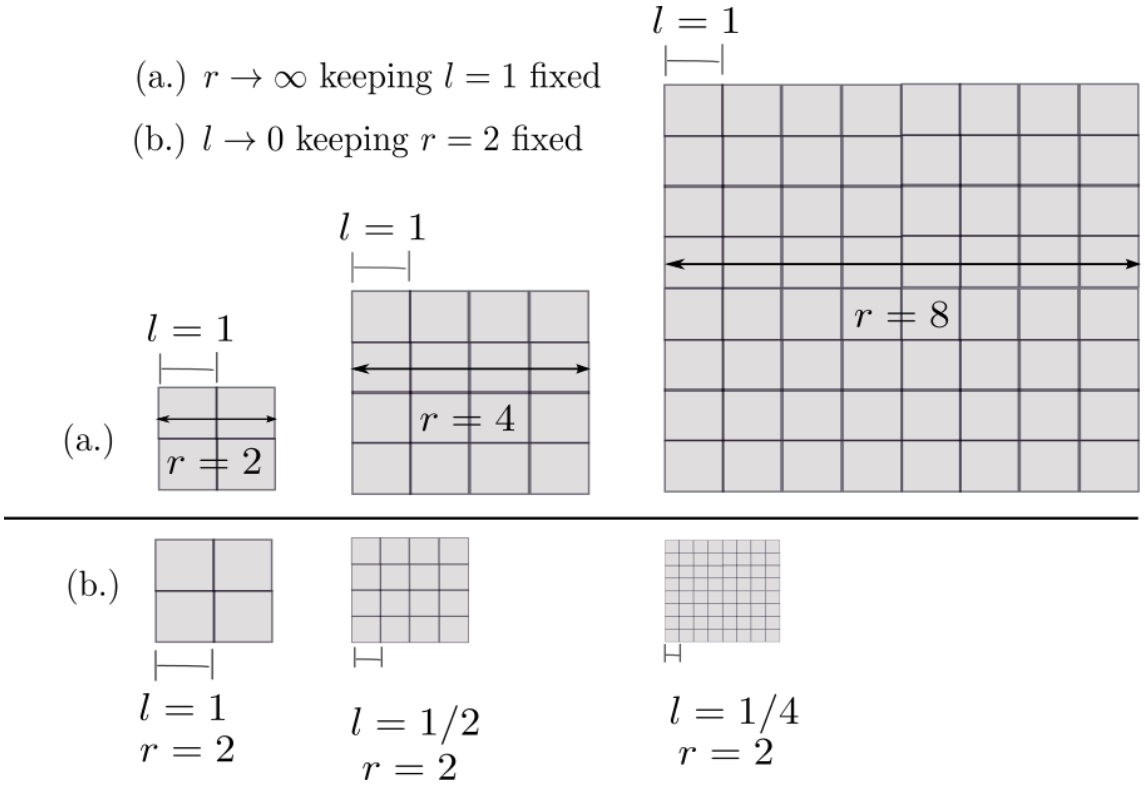
Figure: Jason Marhsall and Kaushik Dayal

Continuum limit

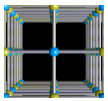
$$E_{limit} = \lim_{r\to\infty} \left\{ \frac{1}{vol(B_r(\mathbf{0}))} \sum_{i,j} \Phi(\boldsymbol{x}_i - \boldsymbol{x}_j) \right\}$$

→ Average energy of atoms in Sphere $B_r(0)$

Two equivalent approach →



Scaled potential → $\Phi_l(\boldsymbol{x}) = \Phi\left(\frac{\boldsymbol{x}}{l}\right)$



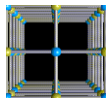
Continuum limit…

→ **Energy of domain**

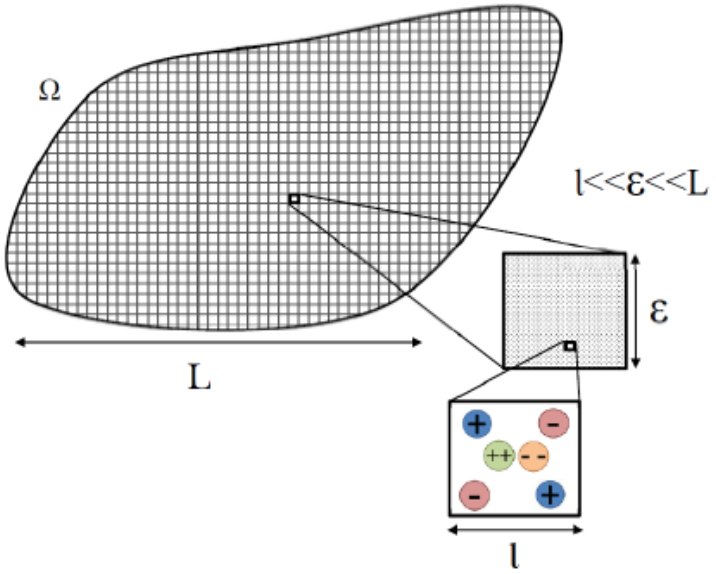
$$E(\Omega) \approx vol(\Omega) \times E_{limit}$$

→ **Accuracy increases as**

$\frac{diam(\Omega)}{l}$ **increases**



Electrostatics energy



- $\rho : \Omega \times \mathbb{R}^3 \to \mathbb{R}$ charge density field
- small scale dependence : $\rho_l(\boldsymbol{x}, \boldsymbol{y}) = \rho(\boldsymbol{x}, \boldsymbol{y}/l)$
- Electrostatics energy

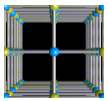
$$E = \sum_a E(a)$$

Local energy

E(a) = [energy due to interactions of charges within material point a]

+ [energy due to interactions of charges outside material point a]

Non-Local energy



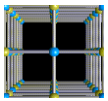
Random media: Charge density field

- $\rho : \Omega \times \mathbb{R}^3 \times D \to \mathbb{R}$ random field
- stationary : $\bar{\rho} : \Omega \times D \to \mathbb{R} \longrightarrow \rho(\boldsymbol{x}, \boldsymbol{y}, \omega) = \bar{\rho}(\boldsymbol{x}, T_{\boldsymbol{y}}\omega)$
- ρ is ergodic
- scaled charge density field

$$\rho_l(\boldsymbol{x}, \boldsymbol{y}, \omega) = \rho(\boldsymbol{x}, \boldsymbol{y}/l, \omega) = \bar{\rho}(\boldsymbol{x}, T_{\boldsymbol{y}/l}\omega)$$

we find later: scaling is not correct

$$\text{need } \rho_l(\boldsymbol{x}, \boldsymbol{y}, \omega) = \frac{\rho(\boldsymbol{x}, \boldsymbol{y}/l, \omega)}{l}$$



Random media: Local energy

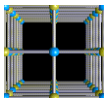
$$E_{local} = \frac{4\pi}{3} \sum_{\boldsymbol{x} \in \Omega_\epsilon} \epsilon^3 l^2 \left(\frac{1}{\left| B_{\epsilon/l}(\boldsymbol{x}) \right|} \int_{\boldsymbol{z} \in B_{\epsilon/l}(\boldsymbol{x})} \rho(\boldsymbol{x}, \boldsymbol{z}, \omega) h(\boldsymbol{x}, \boldsymbol{z}, \omega) dV_{\boldsymbol{z}} \right)$$

Ergodic theorem

We don't want energy to go to zero or infinity trivially

$$\text{Correct scaling : } \rho_l(\boldsymbol{x}, \boldsymbol{y}, \omega) = \frac{\rho(\boldsymbol{x}, \boldsymbol{y}/l, \omega)}{l}$$

*we had assumed earlier $\rho_l(\boldsymbol{x}, \boldsymbol{y}, \omega) = \rho(\boldsymbol{x}, \boldsymbol{y}/l, \omega)$



Random media: Non-local energy

After change of variable and dividing and multiplying $vol(B_{\epsilon/l}(\boldsymbol{x}))vol(B_{\epsilon/l}(\boldsymbol{x}'))$

$$E_{nonlocal} = \left(\frac{4\pi}{3}\right)^2 \sum_{\substack{\boldsymbol{x},\boldsymbol{x}'\in\Omega_\epsilon,\\ \boldsymbol{x}\neq\boldsymbol{x}'}} \epsilon^6 \left(\frac{1}{l^2} \frac{1}{|B_{\epsilon/l}(\boldsymbol{x})|} \frac{1}{|B_{\epsilon/l}(\boldsymbol{x}')|} \int_{\substack{\boldsymbol{z}\in B_{\epsilon/l}(\boldsymbol{x}),\\ \boldsymbol{z}'\in B_{\epsilon/l}(\boldsymbol{x}')}} \underbrace{\frac{\rho(\boldsymbol{x},\boldsymbol{z},\omega)\rho(\boldsymbol{x}',\boldsymbol{z}',\omega)}{|\boldsymbol{x}+l\boldsymbol{z}-\boldsymbol{x}'-l\boldsymbol{z}'|}} dV_{\boldsymbol{z}} dV_{\boldsymbol{z}'} \right)$$

Taylor's series expansion

$$\frac{1}{|\boldsymbol{x}+l\boldsymbol{z}-\boldsymbol{x}'-l\boldsymbol{z}'|} = \frac{1}{|\boldsymbol{x}-\boldsymbol{x}'|} + \left[\frac{\partial}{\partial \boldsymbol{y}}\frac{1}{|\boldsymbol{y}|}\right]_{\boldsymbol{y}=\boldsymbol{x}-\boldsymbol{x}'} l\cdot(\boldsymbol{z}-\boldsymbol{z}') + \left[\frac{\partial^2}{\partial \boldsymbol{y}^2}\frac{1}{|\boldsymbol{y}|}\right]_{\boldsymbol{y}=\boldsymbol{x}-\boldsymbol{x}'} l^2 : (\boldsymbol{z}-\boldsymbol{z}')\otimes(\boldsymbol{z}-\boldsymbol{z}') + O(l^3)$$

Zeroth order term

$$\frac{1}{l^2}\left\{\frac{1}{|B_{\epsilon/l}(\boldsymbol{x})|}\int_{\boldsymbol{z}\in B_{\epsilon/l}(\boldsymbol{x})}\rho(\boldsymbol{x},\boldsymbol{z},\omega)dV_{\boldsymbol{z}}\right\} \times \left\{\frac{1}{|B_{\epsilon/l}(\boldsymbol{x}')|}\int_{\boldsymbol{z}'\in B_{\epsilon/l}(\boldsymbol{x}')}\rho(\boldsymbol{x}',\boldsymbol{z}',\omega)dV_{\boldsymbol{z}'}\right\}$$

Goes to infinity, unless the term in bracket is zero

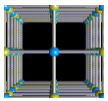
Second order term

Charge neutrality condition

$$\lim_{\epsilon/l\to\infty}\frac{1}{|B_{\epsilon/l}(\boldsymbol{x})|}\int_{\boldsymbol{z}\in B_{\epsilon/l}(\boldsymbol{x})}\rho(\boldsymbol{x},\boldsymbol{z},\omega)dV_{\boldsymbol{z}} = 0 \quad \forall \boldsymbol{x}\in\Omega$$

By Ergodic theorem

$$\mathbb{E}[\rho(\boldsymbol{x},\boldsymbol{y},)] = 0 \quad \forall \boldsymbol{x}\in\Omega, \boldsymbol{y}\in\mathbb{R}^3$$



Random media: Result

Assume that ρ is ergodic and stationary, and also satisfies charge neutrality condition. Let ρ_l be scaled field. Then, electrostatics energy, in the limit is given by

$$E = E_{local} + E_{nonlocal} \tag{1}$$

$$E_{local} = \mathbb{E}\left[\int_{\boldsymbol{x}\in\Omega}\left(\int_{\mathbb{R}^3}\frac{\rho(\boldsymbol{x},\boldsymbol{0},\cdot)\rho(\boldsymbol{x},\boldsymbol{z}',\cdot)}{|\boldsymbol{0}-\boldsymbol{z}'|}dV_{\boldsymbol{z}'}\right)dV_{\boldsymbol{x}}\right] \tag{2}$$

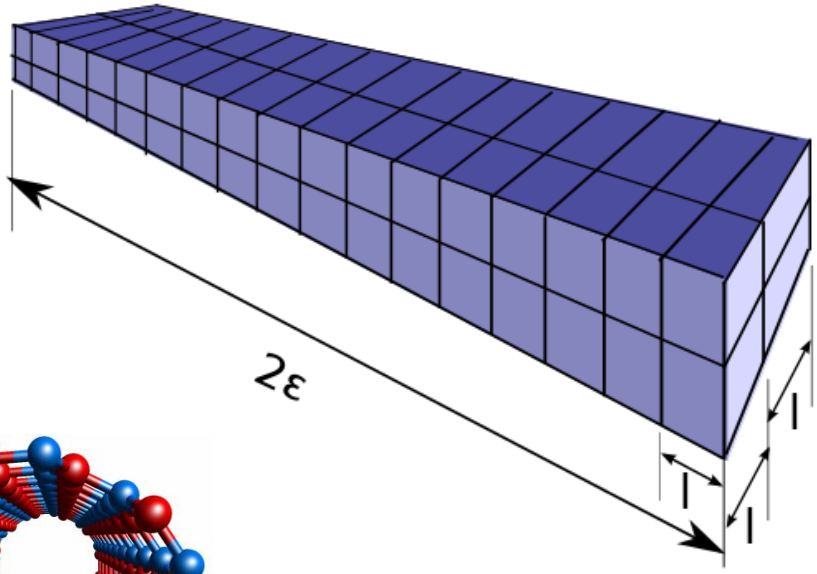
$$E_{nonlocal} = \int_{\substack{\boldsymbol{x},\boldsymbol{x}'\in\Omega,\\ \boldsymbol{x}\neq\boldsymbol{x}'}}\mathbb{K}(\boldsymbol{x}-\boldsymbol{x}') : \hat{\boldsymbol{p}}(\boldsymbol{x})\otimes\hat{\boldsymbol{p}}(\boldsymbol{x}')dV_{\boldsymbol{x}}d_{\boldsymbol{x}'} \tag{3}$$

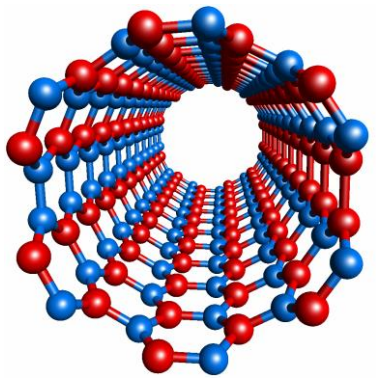
where $\hat{\boldsymbol{p}}(\boldsymbol{x})$ is dipole moment at $\boldsymbol{x}$ and is independent of ω.

$$\hat{\boldsymbol{p}}(\boldsymbol{x}) = \boldsymbol{p}(\boldsymbol{x},w) = \lim_{r\to\infty}\frac{1}{|B_r(\boldsymbol{x})|}\int_{\boldsymbol{z}\in B_r(\boldsymbol{x})}\rho(\boldsymbol{x},\boldsymbol{z},\omega)\boldsymbol{z}dV_{\boldsymbol{z}}$$

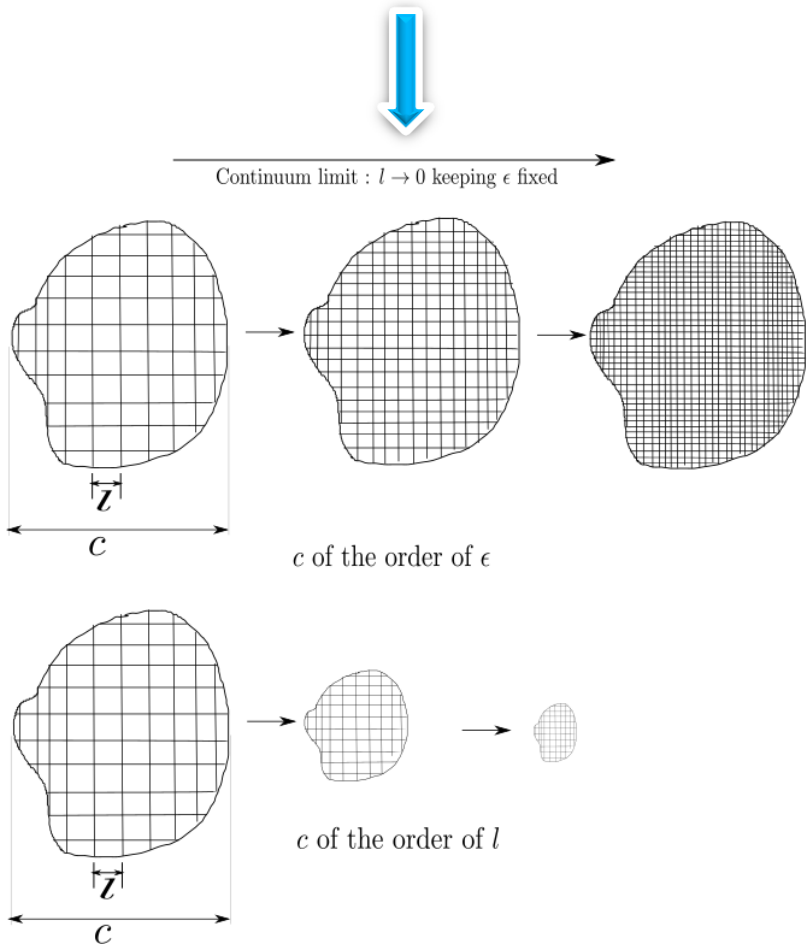
Nanostructures

- Cross-section is of few atomic thickness
- Long in axial direction
- Translational, and/or rotational symmetry

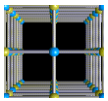




Nanostructure and macroscopically thick structures in a continuum limit

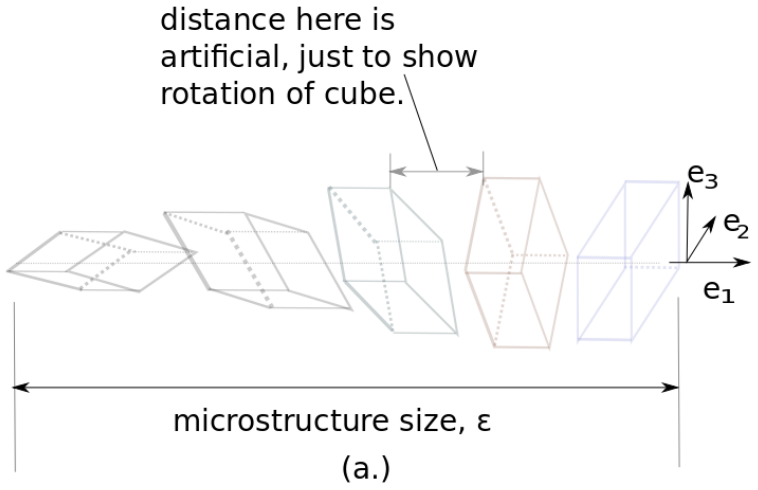


Nanotube Image: http://nano-bio.ehu.es/areas/nanostructures-and-nanotubes



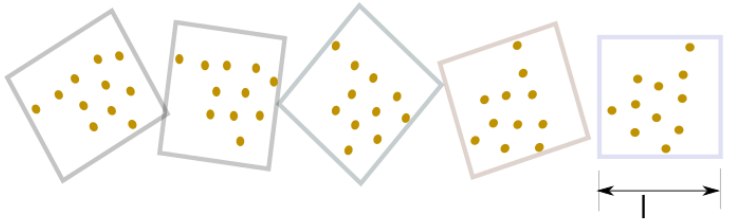
Nanostructures: Geometry

- cross-section is $[0, l]^2$
- Let $\boldsymbol{Q}$ be rotation and $\boldsymbol{e}_1$ be unit translation
- for periodic nanorod: $\boldsymbol{Q} = \boldsymbol{I}$



Symmetry

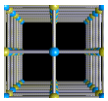
$$\rho(\boldsymbol{x}, \boldsymbol{Q}^k \boldsymbol{y} + k\boldsymbol{e}_1) = \rho(\boldsymbol{x}, \boldsymbol{y})$$



Scaling

$$\rho_l(\boldsymbol{x}, \boldsymbol{y}) = \rho(\boldsymbol{x}, \boldsymbol{y}/l)$$

Correct scaling will be determined by condition that local energy is finite in the limit



Nanostructures: Result

- $$E = \int_{x \in \Omega} E_{local}(x) dl_x + \int_{\substack{x,x' \in \Omega, \\ x \neq x'}} E_{nonlocal}(x, x') dl_x dl_{x'}$$

- $$E_{nonlocal}(x, x') = \frac{q(x)q(x')}{|x\boldsymbol{e}_1 - x'\boldsymbol{e}_1|} = 0$$

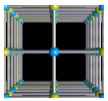
  → **If net charge in unit cell is zero**

- net charge $q(x) := \int_{\boldsymbol{u} \in x\boldsymbol{e}_1 + [0,1]^3} \tilde{\rho}(x, \boldsymbol{u}) dV_{\boldsymbol{u}} = 0$

- we assume there exist $\tilde{\rho}$ such that

$$\rho_l(x, \boldsymbol{y}) = \frac{\tilde{\rho}(x, \boldsymbol{y}/l)}{l^2}$$

No long-range interaction



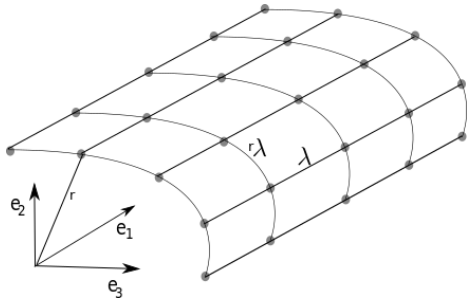
Nanostructures/thin films behave differently

Field at $\boldsymbol{x}$ due to dipole $\boldsymbol{d}$ at origin is $\boldsymbol{K}(\boldsymbol{x})\boldsymbol{d}$ → $\boldsymbol{K}(\boldsymbol{x}) = -\frac{1}{4\pi|\boldsymbol{x}|^3}\left\{\boldsymbol{I} - 3\frac{\boldsymbol{x}}{|\boldsymbol{x}|}\otimes\frac{\boldsymbol{x}}{|\boldsymbol{x}|}\right\}$

Estimate of dipole energy for 1-D, 2-D and 3-D materials

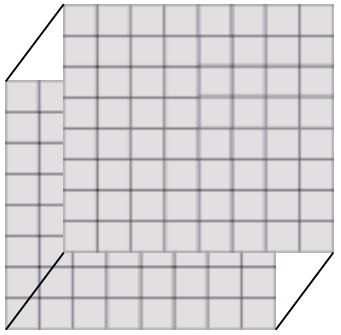
$$W = \sum_{r=1}^{\infty}\frac{1}{r^3}\times 1 = \sum_{r=1}^{\infty}\frac{1}{r^3}$$

At distance r
net dipole is 1



$$W = \sum_{r=1}^{\infty}\frac{1}{r^3}\times r = \sum_{r=1}^{\infty}\frac{1}{r^2}$$

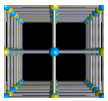
Along the circumference of
circle of r, net dipole is 2*pi*r



$$W = \sum_{r=1}^{\infty}\frac{1}{r^3}\times r^2 = \sum_{r=1}^{\infty}\frac{1}{r}$$

At the surface of sphere of
radius r, net dipole is 4*pi*r*r

Dipole field kernel decays fast for 1-D and 2-D materials

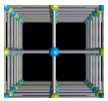


Phase average of a function

- $H(\mathbf{q},\mathbf{p})$: Hamiltonian of system
- $\mathbf{q}$: position vector of all atoms
- $\mathbf{p}$: momenta vector of all atoms
- $p_{exact}(\mathbf{q},\mathbf{p})$: exact probability density function
- F_{exact}: exact free energy
- $f(\mathbf{q},\mathbf{p})$: phase function

Canonical ensemble

- $\langle f \rangle = \frac{1}{N!h^{3N}} \int_{\Gamma} f(\mathbf{q},\mathbf{p}) p_{exact}(\mathbf{q},\mathbf{p}) d\mathbf{q} d\mathbf{p}$
- $p_{exact}(\mathbf{q},\mathbf{p}) = \frac{1}{Z_{exact}} \exp[-\frac{H(\mathbf{q},\mathbf{p})}{k_B T}]$
- $F_{exact} = -k_B T \log[Z_{exact}]$



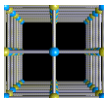
Monte Carlo approximation

- $(\mathbf{q}^0, \mathbf{p}^0)$: initial state of system
- For $(n+1)^{\text{st}}$ step

  Let $(\mathbf{q}, \mathbf{p})$ be randomly choosen state and x random number in $[0, 1]$

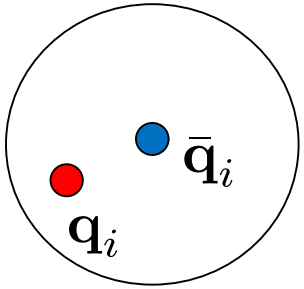
$$(\mathbf{q}^{n+1}, \mathbf{p}^{n+1}) = \begin{cases} (\mathbf{q}, \mathbf{p}) & \text{if } H(\mathbf{q}, \mathbf{p}) - H(\mathbf{q}^n, \mathbf{p}^n) \leq 0, \\ (\mathbf{q}, \mathbf{p}) & \text{if } H(\mathbf{q}, \mathbf{p}) - H(\mathbf{q}^n, \mathbf{p}^n) > 0 \\ & \text{and } \exp[-\dfrac{H(\mathbf{q}, \mathbf{p}) - H(\mathbf{q}^n, \mathbf{p}^n)}{k_B T}] \geq x, \\ (\mathbf{q}^n, \mathbf{p}^n) & \text{if } H(\mathbf{q}, \mathbf{p}) - H(\mathbf{q}^n, \mathbf{p}^n) > 0 \\ & \text{and } \exp[-\dfrac{H(\mathbf{q}, \mathbf{p}) - H(\mathbf{q}^n, \mathbf{p}^n)}{k_B T}] < x \end{cases}$$

- $$\langle f \rangle_{Monte-Carlo} = \frac{1}{N} \sum_{i=1}^{N} f(\mathbf{q}^i, \mathbf{p}^i) N p(\mathbf{q}^i, \mathbf{p}^i)$$



max-ent approach

We use *max-ent* method developed by Kulkarni, Knapp and Ortiz*



1. **Mean position and mean momenta**

$$\langle \mathbf{q}_i \rangle = \bar{\mathbf{q}}_i$$

$$\langle |\mathbf{q}_i - \bar{\mathbf{q}}_i|^2 \rangle = 3\tau_i^2$$

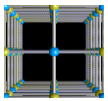
2. **Maximum entropy principle → Probability density function**

$$S[p] = -\frac{k_B}{N!h^{3N}} \int_\Gamma p \log p d\Gamma$$

3. **Variational mean field theory → minimization problem**

$$F_p := \langle H(\mathbf{q}, \mathbf{p}) \rangle_p - TS[p] \geq F_{exact}$$

* Kulkarni, Y., Knapp, J., and Ortiz, M.: A Variational approach to coarse graining of equilibrium and non-graining atomistic description at finite temperature. *J. Mech. and Phys. of Solids,* 56 (2008).



Minimization problem

- **Assumption:** quasi-static problem

$$\bar{\mathbf{p}}_i = \mathbf{0}$$

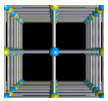
- **Free energy**

$$\begin{aligned} F_p :=& F_p(\bar{\mathbf{q}}, \omega, T) \\ =& \langle H \rangle_p - TS[p] \\ =& \sum_i \frac{3}{2} k_B T + \sum_i < V_i >_p \\ &+ \sum_{i \neq j} \frac{1}{2} < \frac{Q_i Q_j}{|\mathbf{q}_i - \mathbf{q}_j|} >_p - \sum_i TS_i \end{aligned}$$

- **Determine the mean state**

$$\min_{\mathbf{q},\omega} F_p(\mathbf{q}, \omega; T) + F_{ext}(\mathbf{q}, \omega; T)$$

$$\omega_i := \frac{\sigma_i}{\tau_i} \quad \Leftarrow \quad \text{mean frequency of atom } i$$



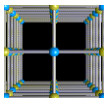
Quasi-harmonic approximation*

- $$V(\{\boldsymbol{q}_i\}) = V(\{\bar{\boldsymbol{q}}_i\}) + \sum \frac{\partial V(\{\boldsymbol{q}_i\})}{\partial \boldsymbol{q}_i} \Big|_{\boldsymbol{q}_i=\bar{\mathbf{q}}_i} \cdot(\boldsymbol{q}_- \bar{\boldsymbol{q}}_i) + \frac{1}{2}\sum_i \underbrace{\frac{\partial^2 V(\mathbf{q})}{\partial \boldsymbol{q}_i^2}\Big|_{\boldsymbol{q}_i=\bar{\boldsymbol{q}}_i}}_{\boldsymbol{H}_{ii}} : (\boldsymbol{q}_i - \bar{\boldsymbol{q}}_i)\otimes(\boldsymbol{q}_i - \bar{\boldsymbol{q}}_i)$$

Phase average

- $$< V >= V(\{\bar{\boldsymbol{q}}_i\}) + \frac{1}{2}\sum_i \frac{\sigma_i^2}{\omega_i^2}\boldsymbol{I} : \boldsymbol{H}_{ii}$$
- For Coulombic interaction $\boldsymbol{I} : \boldsymbol{H}_{ii} = 0$
- $$\min_{\omega} F_p(\mathbf{q}, \omega, T) \quad \Rightarrow \quad \omega_i^2 = \frac{1}{3}\boldsymbol{I} : \boldsymbol{H}_{ii}$$

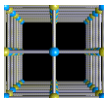
* Kulkarni, Y., Knapp, J., and Ortiz, M.: A Variational approach to coarse graining of equilibrium and non-graining atomistic description at finite temperature. *J. Mech. and Phys. of Solids,* 56 (2008).



QC code

- Extended Jason Marshall's code[1] to finite temperature
- Object oriented
- New more efficient algorithm to compute phase average of EAM like potential

1 Marshall, J. and Dayal, K.: Atomistic to continuum multiscale modeling with long range electrostatic interaction In ionic solids. *J. Mech. and Phys. of Solids, 1 (2013).*

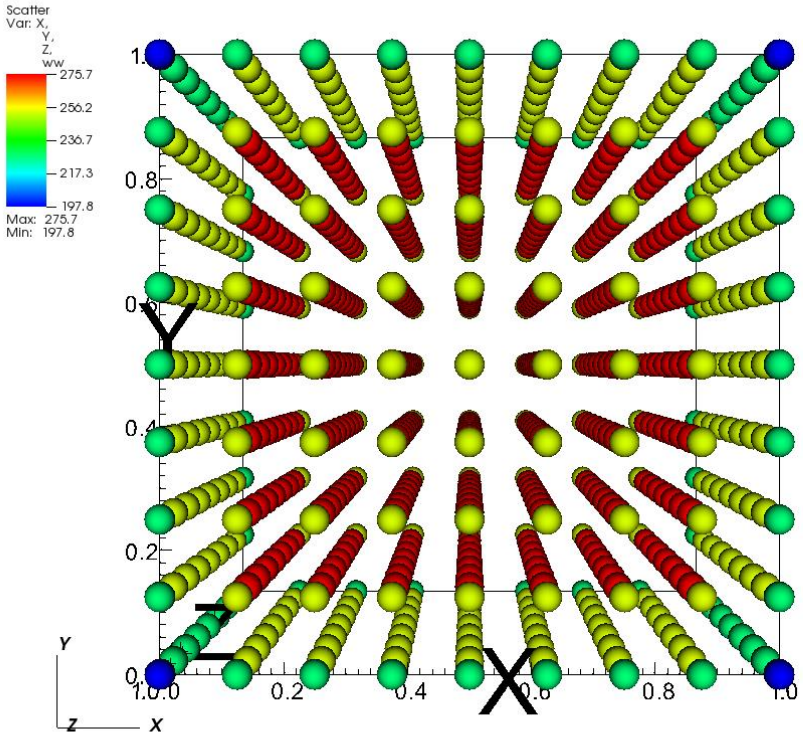


Quasi-harmonic approximation: QC code

Ar Lennard Jonnes

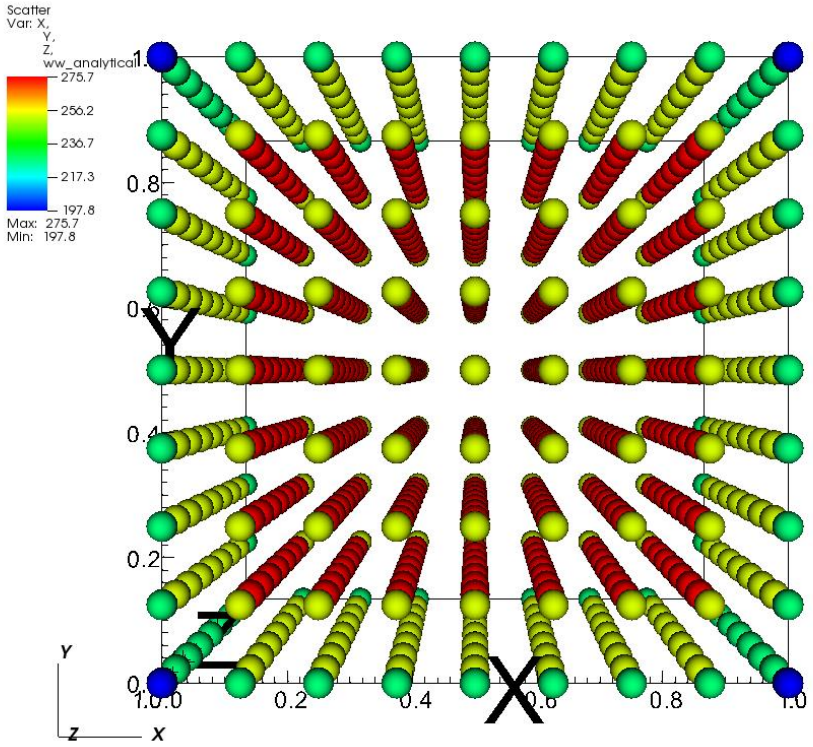
Size		Type	Constant a	Potential				Temperature	Initial freq.
Full	Atomistic			Type	σ_0	ϵ_0	r_{cut}		
	8x8x8	SC	3.6697304	LJ	3.4	0.0104	8.5	100K	288.2

DB: node_quasi_1_load_number_00000.plt.gz

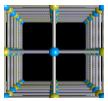


QC Code minimization

DB: node_quasi_1_load_number_00000.plt.gz



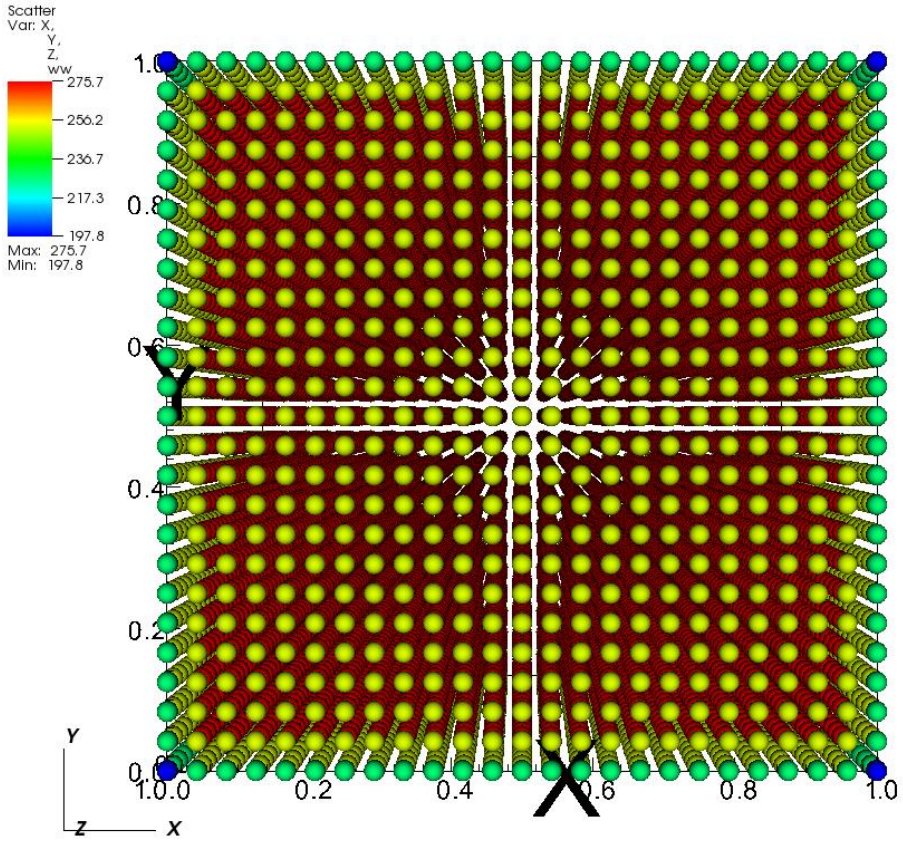
Analytical frequency



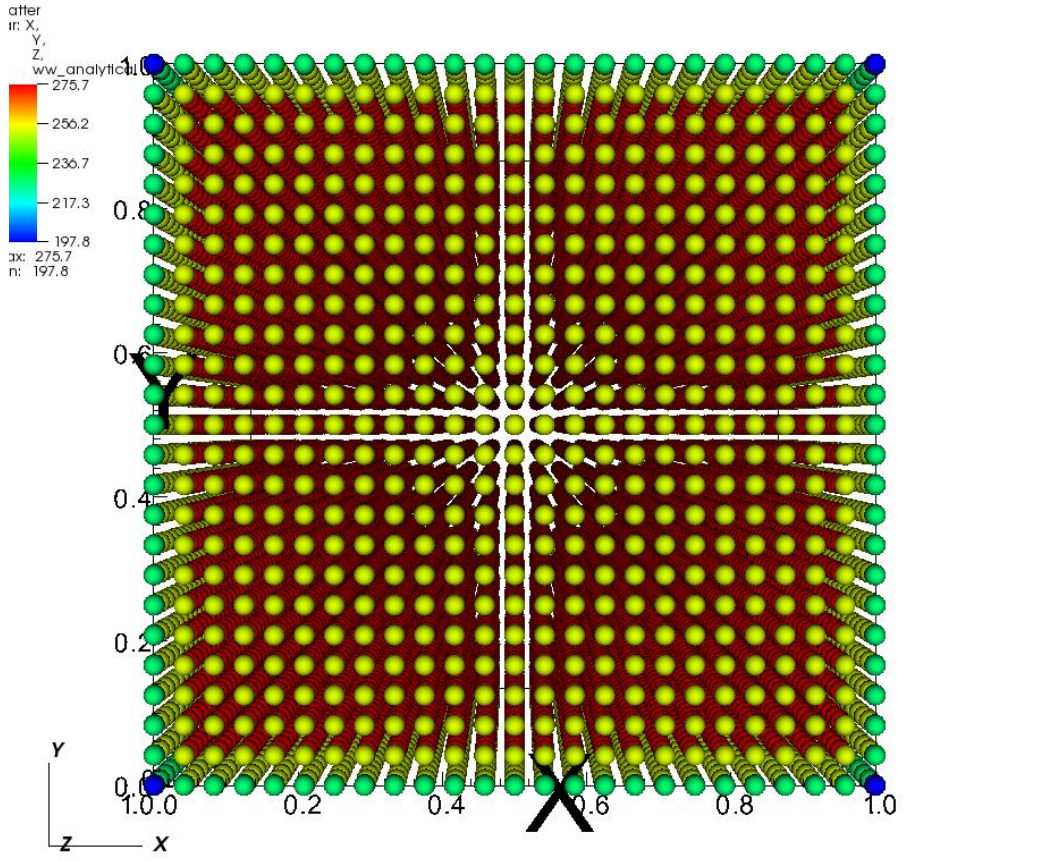
Quasi-harmonic approximation: QC code

Ar Lennard Jonnes

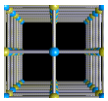
Size		Type	Constant a	Potential				Temperature	Initial freq.
Full	Atomistic			Type	σ_0	ϵ_0	r_{cut}		
	24x24x24	SC	3.6697304	LJ	3.4	0.0104	8.5	100K	288.2



QC Code minimization

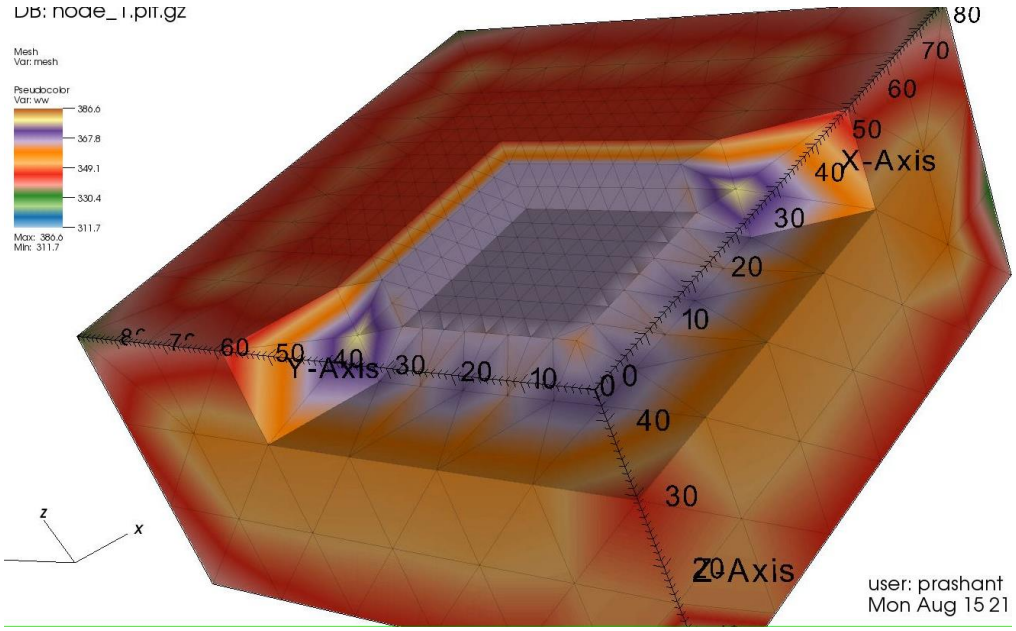


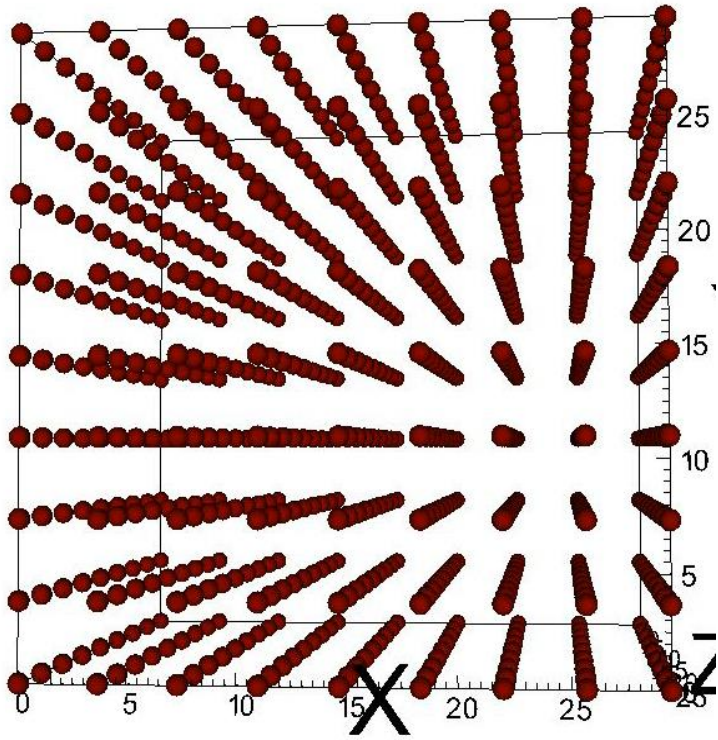
Analytical frequency



Frequency minimization

Frequency which minimizes free energy should be independent of initial value





Mesh: 24x24x12 – 6x6x6

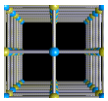
Initial frequency

1. 288.2
2. 230.5
3. 192.1
4. 164.7

Mesh: 8x8x8

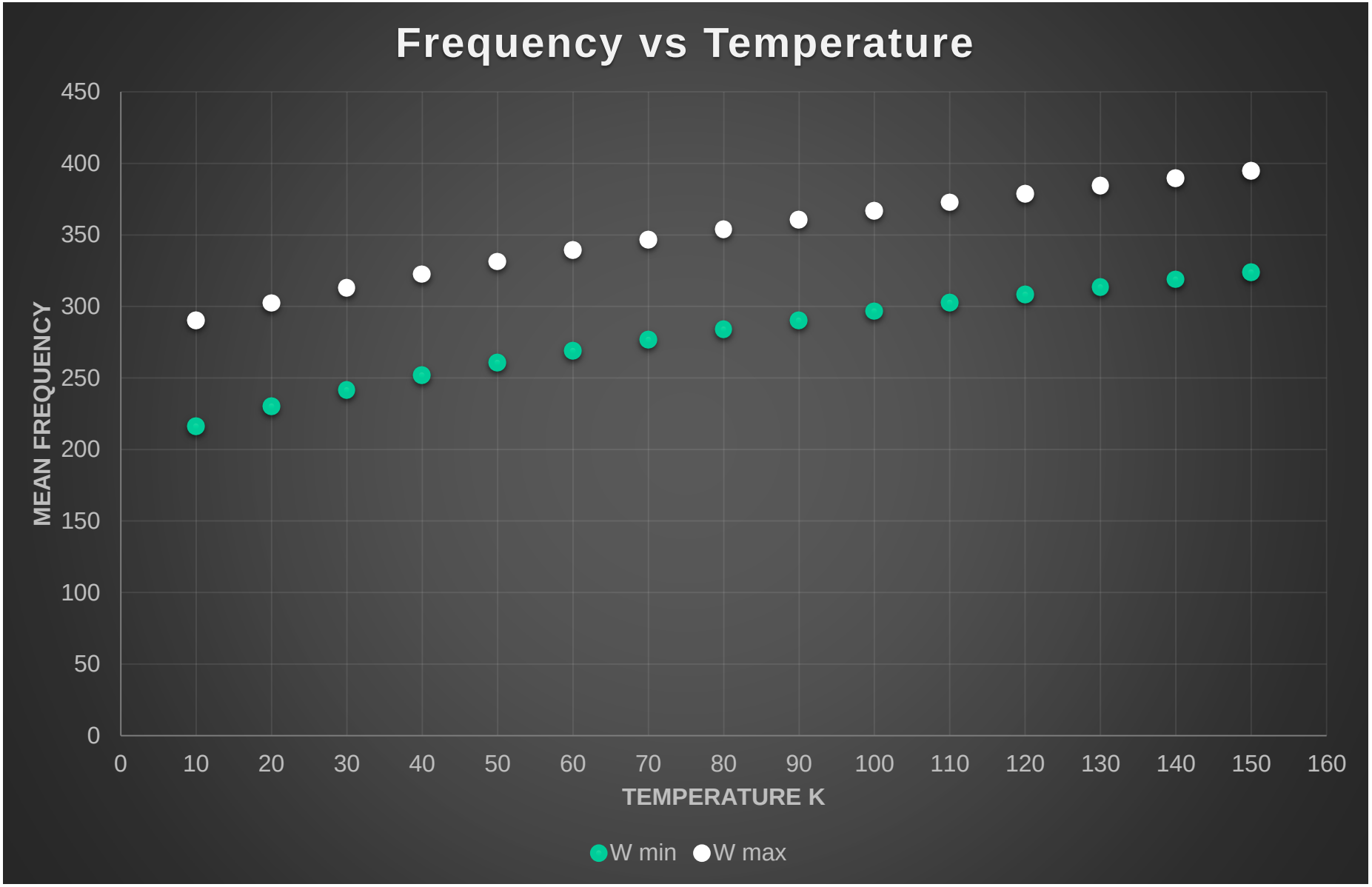
Initial frequency

1. 576.3
2. 230.5
3. 192.1
4. 144.1
5. 115.1
6. 96.05

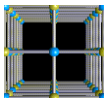


Frequency minimization

Mean frequency should increase with the temperature

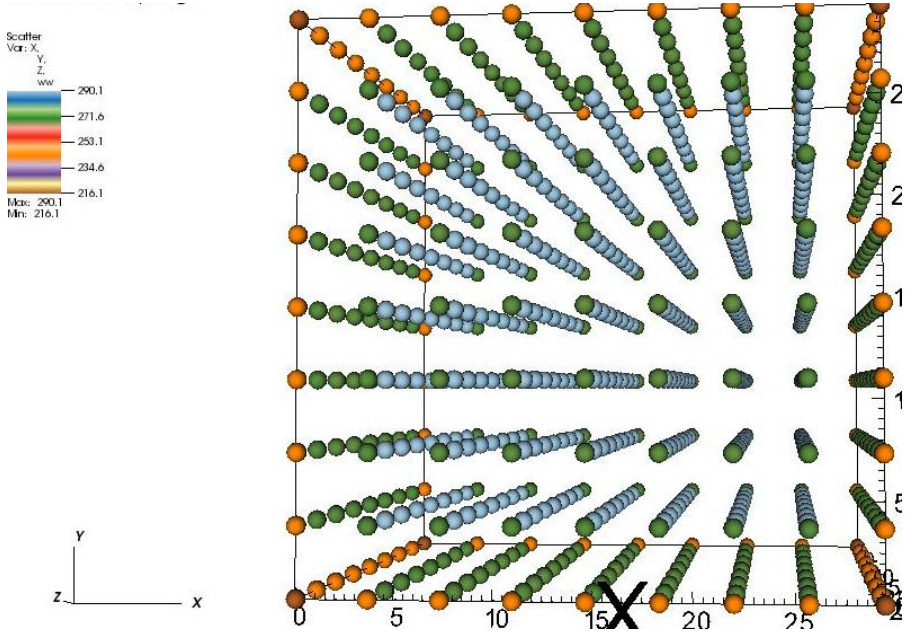


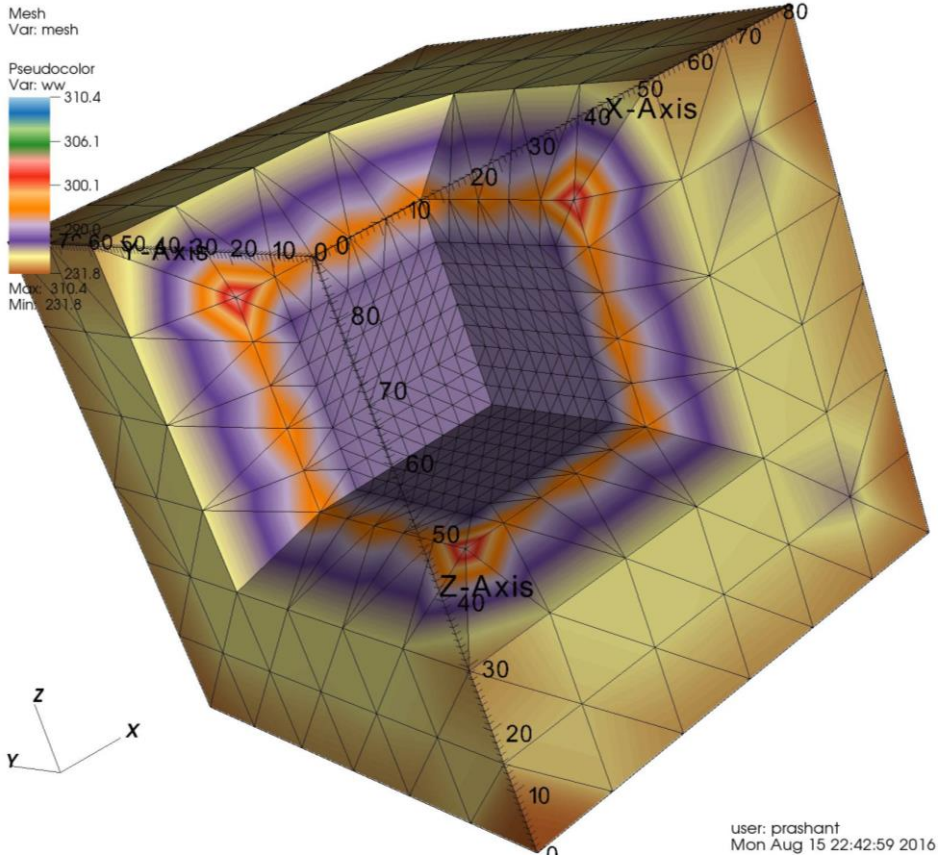
Mesh: 8x8x8



Frequency minimization

Mean frequency should increase with the temperature

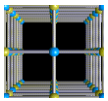




Mesh: 8x8x8

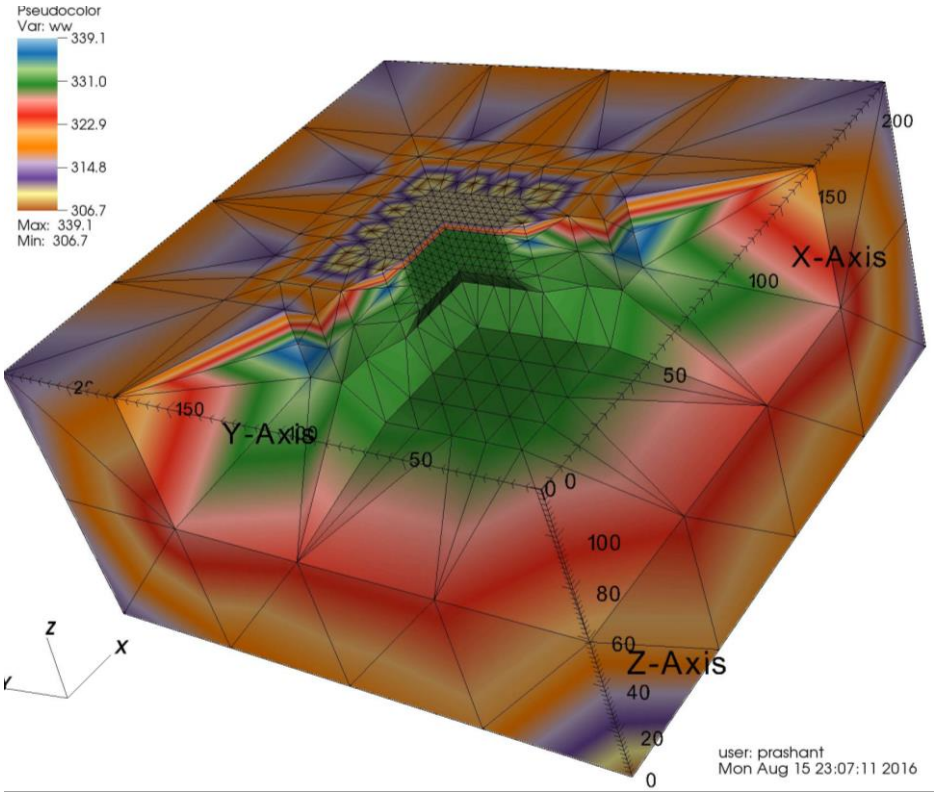
Mesh: 24x24x24-6x6x6

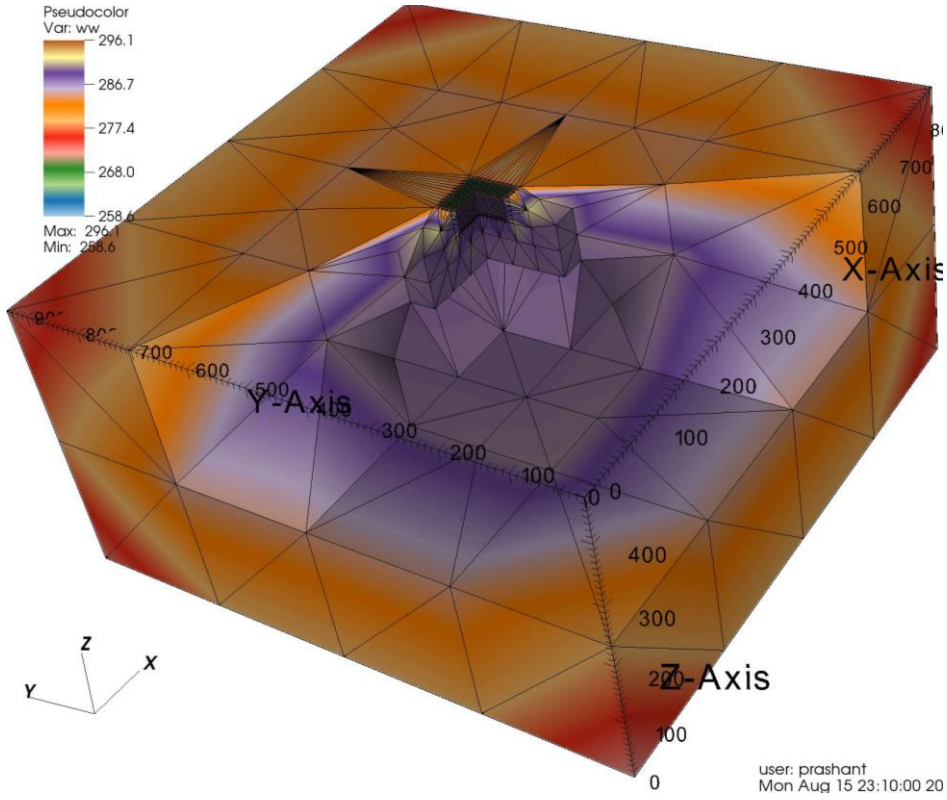
Temperature: {10K, 20K, …, 150K}



Frequency minimization

Mean frequency should increase with the temperature



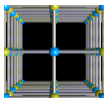


Mesh: 64x64x32-6x6x6

Temperature
{10K, 30K, 50K, 70K, 90K, 100K, 120K, 150K}

Mesh: 256x256x128-10x10x10

Temperature
{50K, 80K, 90K, 100K}



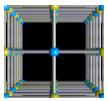
Frequency minimization: Discussion

- $\omega = \frac{\sigma}{\tau},\ \sigma = \sqrt{2k_B T}$

- if τ is very small
  f_ω due to entropy dominates and it is uniform

- if τ is very large
  f_ω due to interatomic potentials dominates and it is very large

- we find that when initial frequency is such that f_ω due to interatomic potential and entropy is of the same order the code converges.

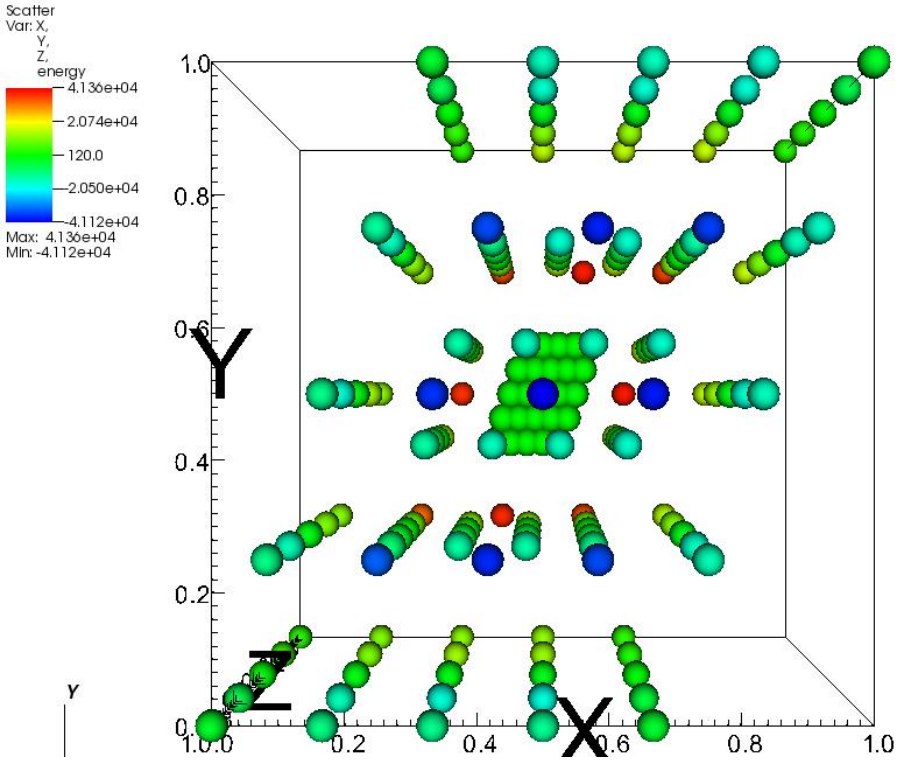


Electrostatics implementation

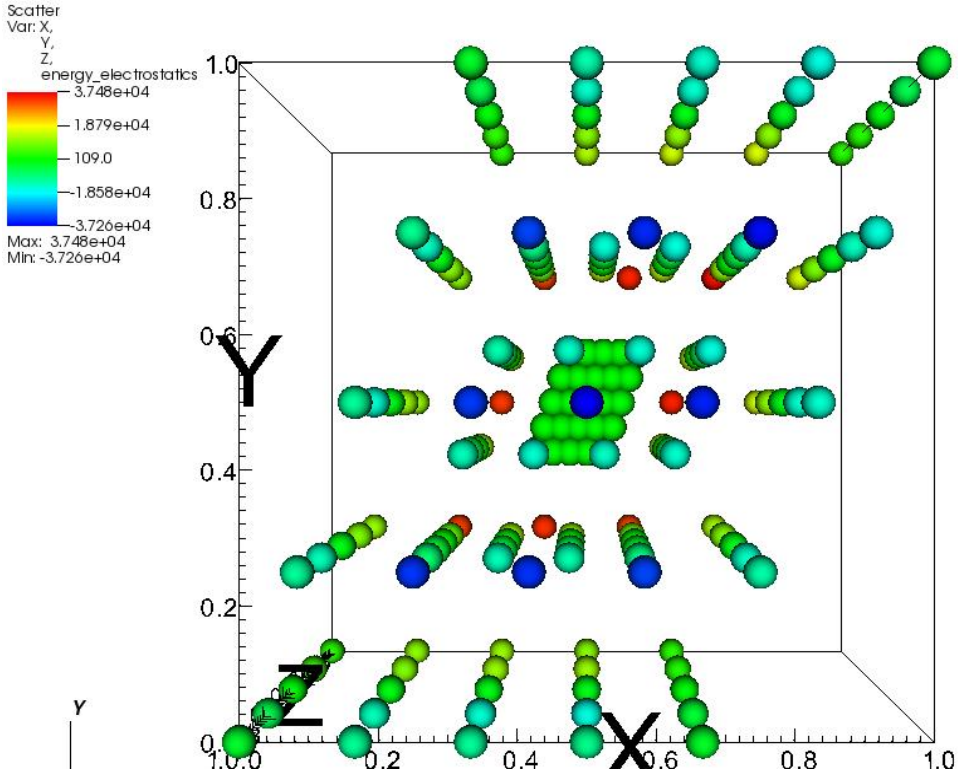
- For small τ, phase average of energy would be very close to the energy at mean configuration!

Gallium nitride 6-lattice core-shell model

Size		Type	Constant	Potential	Temperature	Initial freq.
Full	Atomistic		a			
24x24x24	2x2x2	Wurtzite	---	Core-shell 6 lattice model*	300K	----

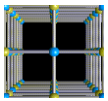


Zero temp QC (Old Code)



Finite temp QC (New code), tau = 0.01

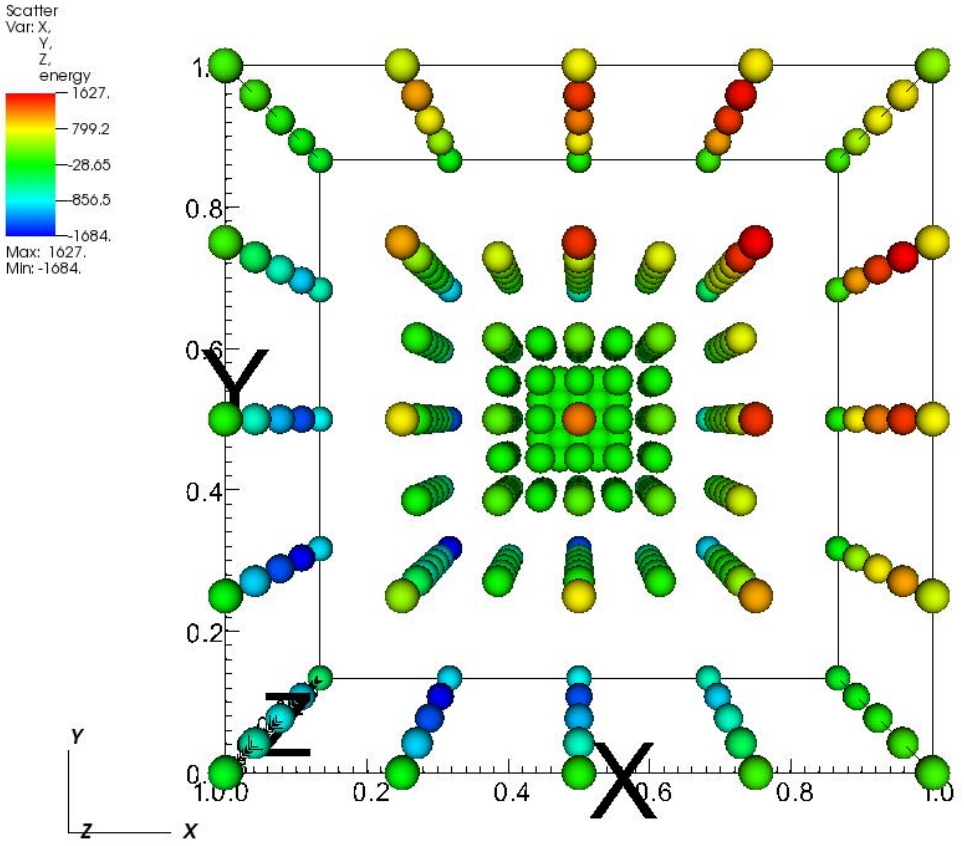
* Zapol, P., Pandey, R., and Gale, J. D..: An interatomic potential study of the properties of gallium nitride. *J. of Phys.: Condensed Matter, 9(44):9517 (1997)*



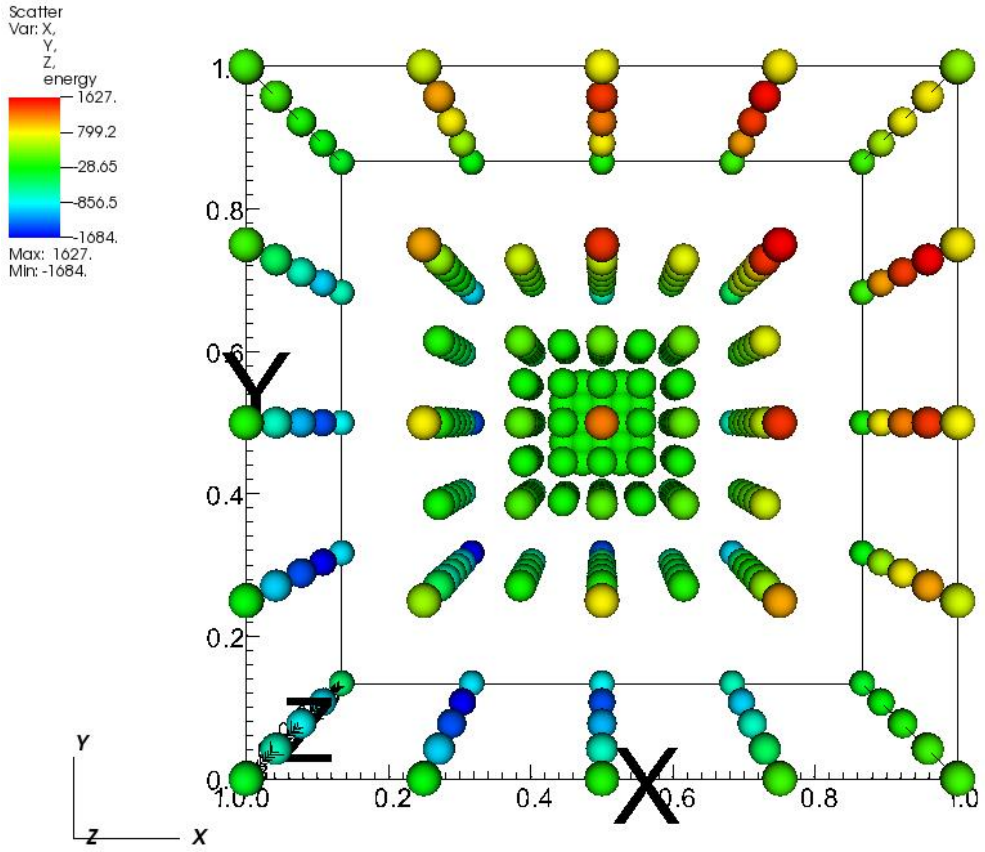
Electrostatics implementation

NiAl: Artificial charge +1 at Ni and -1 at Al

Size		Type	Constant	Potential	Temperature	Initial freq.
Full	Atomistic		a			
32x32x32	2x2x2	SC	---	MishinNiAl *	300K	----

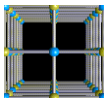


Zero temp QC (Old Code)



Finite temp QC (New code), tau = 0.001

* Mishin, Y., Mehl, M., and Papaconstantopoulos, D..: Embedded-atom potential for b 2-nial. *Physical Review B,* 65(22):224114 (2002)



Discussion

- No long range interactions in nanostructures and thin films

  Agrees with Gioia and James calculation for thin film

- In case of random media, we find that nonlocal energy, does not depend on fluctuations.

  - Fluctuations are happening at the scale of l

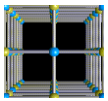
    Whereas nonlocal energy is due to the interaction between material points which are ϵ apart.

  - Coulombic interaction is linear.

- Our QC calculation show that initial frequency should be in range such that frequency force from different interactions is of the same order

- We also show that minimizing frequency is independent of initial frequency.

Gioia, G. and James, R. D.: Micromagnetics of very thin films. *Proc. R. Soc. Lond. A, 453* (1997)



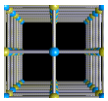
Future works

- Point defects plays an important role in semiconductor devices. We would like to model the single charge point defect in a large crystal and see how it interacts with surrounding.

- The multi-scale formulation is for finite constant temperature problems. Doing non-equilibrium in a multiscale framework is still a challenge.

  Groups like Tadmor group and Knapp group are working on this challenge.

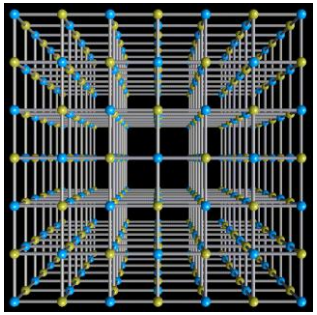
- For non-equilibrium temperature problem, we may have to revisit the ergodic and stationary assumption on charge density field.
  - If charge density field is not ergodic then computation of dipole moment **p(x)** is not clear.
  - If there is a gradient of temperature, the charge density field may not be stationary, as stationarity requires that statistical properties, e.g. mean, should be independent of spatial location.



Future works…

- Experiments can be carried out to find the critical ratio of length of nanotube to the size in cross-section, such that above the critical ratio, nanotube does not show long-range electrical interactions. This will be useful if goal is to develop multiscale models for nanostructures.

- We can also estimate the rate at which difference between actual electrostatics energy, and continuum limit of electrostatics energy, goes to zero with respect to ratio macroscopic length and atomic spacing.



Thank you!