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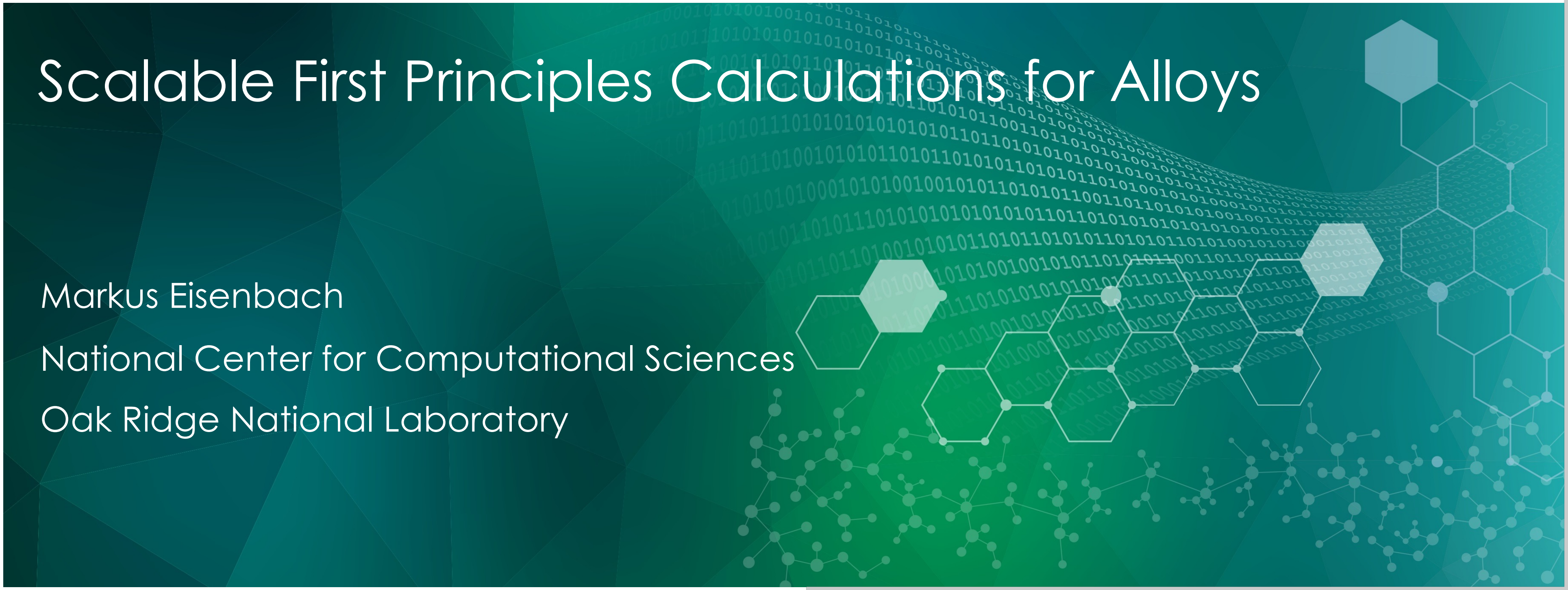

Scalable First Principles Calculations for Alloys

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U.S. DEPARTMENT OF
ENERGY

Multiple Scattering Theory

The Kohn-Sham equation is formally solved by $G(\epsilon) = (\epsilon - H)^{-1}$

Write Hamiltonian as $H = H_0 + V$
& free particle Green function: $G_0(\epsilon) = (\epsilon - H_0)^{-1}$

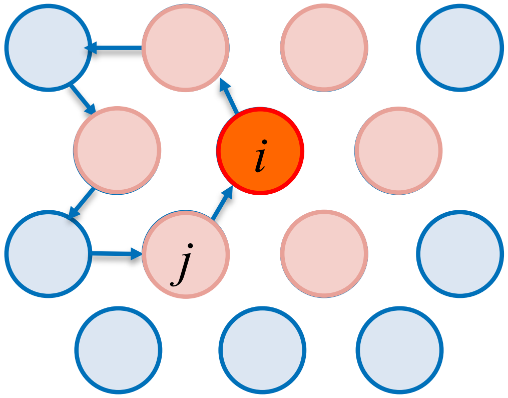
Green function for *H*: $G = G_0 + G_0 V G$
Define transition matrix *T*: $G = G_0 + G_0 T G_0$

The *T* matrix for the full system can be written in terms of the single site scattering *t* matrix and the scattering path matrix τ

$$T = \sum_{ij} \tau_{ij}$$

$$\tau_{ij} = (\delta_{ij} t_i^{-1} - G_0)^{-1}_{ij}$$

$$n(E) = -\frac{1}{\pi} \mathrm{Im}\ \mathrm{Tr} \int_\Omega G(E, r, r) dr$$



Korringa-Kohn-Rostocker Method

- Periodic solid
  - Pure metal, intermetallic compound, …..
  - Lattice Fourier transform
  - Scattering path matrix

$$\tau^{nn'}(\varepsilon) = \frac{1}{\Omega_{BZ}} \int_{BZ} [\mathbf{m} - \mathbf{G}(\mathbf{k};\varepsilon)]^{-1} e^{i\mathbf{k}\bullet\mathbf{R}_{nn'}} d^3k$$

$$\mathbf{G}(\mathbf{k};\varepsilon) = \frac{1}{N} \sum_{n \neq n'} \mathbf{G}(R_{nn'};\varepsilon) e^{i\mathbf{k}\bullet\mathbf{R}_{nn'}}$$

$$\mathbf{m} = \mathbf{t}^{-1}$$

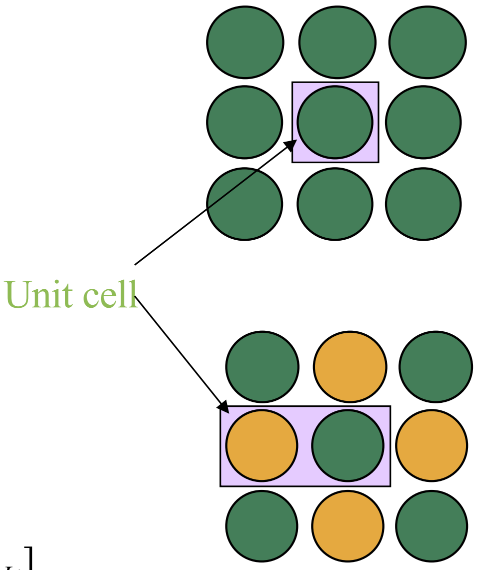
Energy bands given by poles of Green function

$$G(\mathbf{r},\mathbf{r};\varepsilon) = \sum_{LL'} [Z_L^n(\mathbf{r}_n;\varepsilon)\tau_{LL'}^{nn}(\varepsilon)Z_{L'}^n(\mathbf{r};\varepsilon) - Z_L^n(\mathbf{r}_>;\varepsilon)J_L^n(\mathbf{r}_<;\varepsilon)\delta_{LL'}]$$

- Poles of scattering path matrix

$$Det\,|\mathbf{m} - \mathbf{G}(\mathbf{k};\varepsilon)| = 0$$

- Korringa-Kohn-Rostoker method



The Dirac equation for single scatterer

- Dirac equation:

$$[-c\alpha + \frac{1}{2}\beta c^2 + V(\mathbf{r})]\psi(E,\mathbf{r}) = W\psi(E,\mathbf{r})$$

$$V(\mathbf{r}) = \begin{pmatrix} v(\mathbf{r}) + \sigma.\mathbf{B}(\mathbf{r}) & 0 \\ 0 & v(\mathbf{r}) - \sigma.\mathbf{B}(\mathbf{r}) \end{pmatrix}$$

- Free space solutions (V(r)=0):

$$J_\Lambda(E,\mathbf{r}) = \left(W/c^2 + \frac{1}{2}\right)^{1/2} \begin{pmatrix} j_l(pr)\chi_\Lambda(\hat{\mathbf{r}}) \\ \frac{ipcS_\kappa}{W+c^2/2} j_{\bar{l}}(pr)\chi_{\bar{\Lambda}}(\hat{\mathbf{r}}) \end{pmatrix}$$

$$N_\Lambda(E,\mathbf{r}) = \left(W/c^2 + \frac{1}{2}\right)^{1/2} \begin{pmatrix} n_l(pr)\chi_\Lambda(\hat{\mathbf{r}}) \\ \frac{ipcS_\kappa}{W+c^2/2} n_{\bar{l}}(pr)\chi_{\bar{\Lambda}}(\hat{\mathbf{r}}) \end{pmatrix}$$

The Dirac equation for single scatterer

- Ansatz: Expand solution in free space solutions using a phase integral:

$$\psi_\Lambda(E,\mathbf{r}) = \sum_{\Lambda'} S_{\Lambda'\Lambda}(E,\mathbf{r}) N_{\Lambda'}(E,\mathbf{r}) - C_{\Lambda'\Lambda}(E,\mathbf{r}) J_{\Lambda'}(E,\mathbf{r})$$

- This allows us to write coupled differential equations for the sine and cosine matrices ($S_{\Lambda'\Lambda}$ and $C_{\Lambda'\Lambda}$)

$$\frac{d}{dr} S_{\Lambda'\Lambda}(r) = \sum_{\Lambda''} a^{JN}_{\Lambda''\Lambda'}(r) S_{\Lambda''\Lambda}(r) - a^{JJ}_{\Lambda''\Lambda}(r) C_{\Lambda''\Lambda}(r)$$

$$\frac{d}{dr} C_{\Lambda'\Lambda}(r) = \sum_{\Lambda''} a^{NN}_{\Lambda''\Lambda'}(r) S_{\Lambda''\Lambda}(r) - a^{NJ}_{\Lambda''\Lambda}(r) C_{\Lambda''\Lambda}(r)$$

$$a^{XY}_{\Lambda''\Lambda'}(E,r) = r^2 \int p X^{+}_{\Lambda'}(E,\mathbf{r}) V(\mathbf{r}) Y_{\Lambda''}(E,\mathbf{r}) d\hat{\mathbf{r}}$$

The Dirac equation for single scatterer

- For r outside the scattering potential:

$$t_{\Lambda\Lambda'}(E) = -\frac{1}{p}\sum_{\Lambda''} S_{\Lambda\Lambda''}(E)(C_{\Lambda''\Lambda'}(E) - iS_{\Lambda''\Lambda'}(E))^{-1}$$

- Requires only the regular solution with the initial value condition

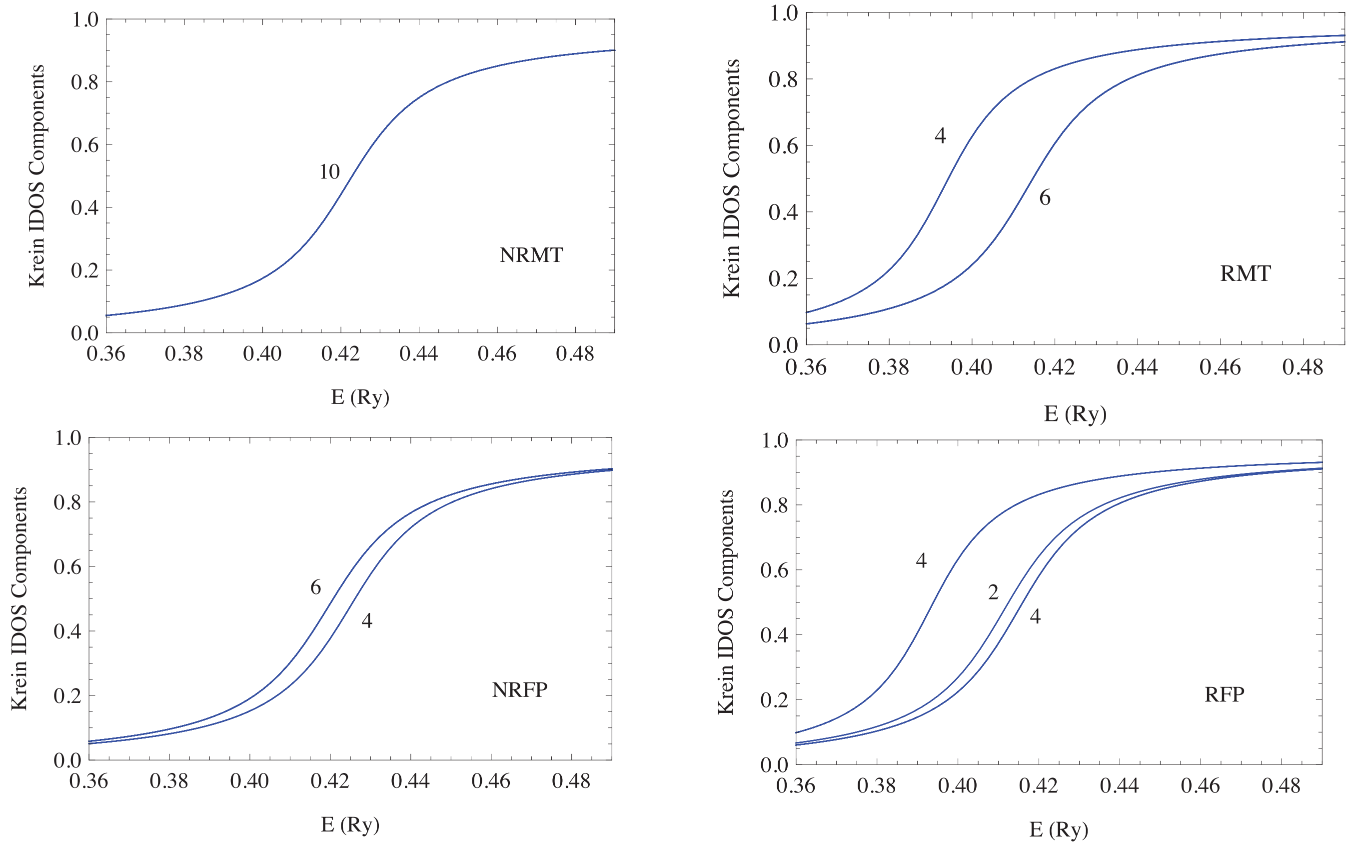
$$\psi_\Lambda(E, \mathbf{r})_{r\to 0} = J_\Lambda(E, \mathbf{r})$$

- i.e.

$$S_{\Lambda'\Lambda}(E, 0) = 0$$

$$C_{\Lambda'\Lambda}(E, 0) = -\delta_{\Lambda'\Lambda}$$

Example: single site Cu 3d scattering



Calculation of Densities

- When the Green’s function is known expectation values of operators can be calculated as traces
- I.e. the charge and magnetization densities are given by

$$n(\mathbf{r}) = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{E_F} \mathrm{Tr}G(\mathbf{r},\mathbf{r};\epsilon)d\epsilon$$

$$\mathbf{m}(\mathbf{r}) = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{E_F} \mathrm{Tr}G(\mathbf{r},\mathbf{r};\epsilon)\vec{\sigma}d\epsilon$$

- The calculation of the Green’s Function can be implemented as an order-N scaling method on parallel computers

Relativistic Multiple Scattering Theory

- Multiple scattering Green's function

$$G(E,r,r') = \sum_{\Lambda\Lambda'} Z_\Lambda(E,r)\tau_{\Lambda\Lambda'}(E)Z^+_{\Lambda'}(E,r') - Z_\Lambda(E,r)\mathcal{J}^+_\Lambda(E,r')\delta_{\Lambda\Lambda'}$$

- Z are the regular solutions and J the irregular solutions of the single site scattering equation and scattering path matrix

$$\tau = (t^{-1} - G_0)^{-1}$$

- As $r \to 0$, J diverges as r^{-l-1}. This leads to numerical instability of the solver for the coupled ODEs for S and C.

Relativistic Multiple Scattering Theory

- Split the Green's function into single scatterer and remaining multiple scattering part:

$$G(E,r,r') = G_s(E,r,r') + G_m(E,r,r')$$

$$G_s(E,r,r') = \sum_{\Lambda\Lambda'} Z_\Lambda(E,r) t_{\Lambda\Lambda'}(E) Z^+_{\Lambda'}(E,r') - \sum_{\Lambda} Z_\Lambda(E,r) \mathcal{J}^+_\Lambda(E,r')$$

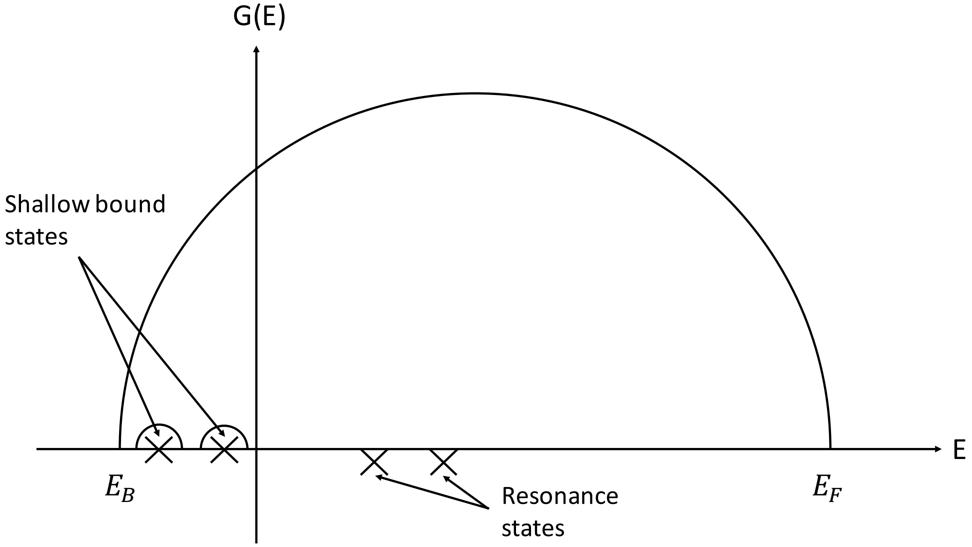
$$G_m(E,r,r') = \sum_{\Lambda\Lambda'} Z_\Lambda(E,r) \left(\tau_{\Lambda\Lambda'}(E) - t_{\Lambda\Lambda'}\right) Z^+_{\Lambda'}(E,r')$$

- For real E, $Z_\Lambda(E,r)\mathcal{J}^+_\Lambda(E,r')$ is real, so it can be ignored for calculations of total Energy, charge density, DOS, etc

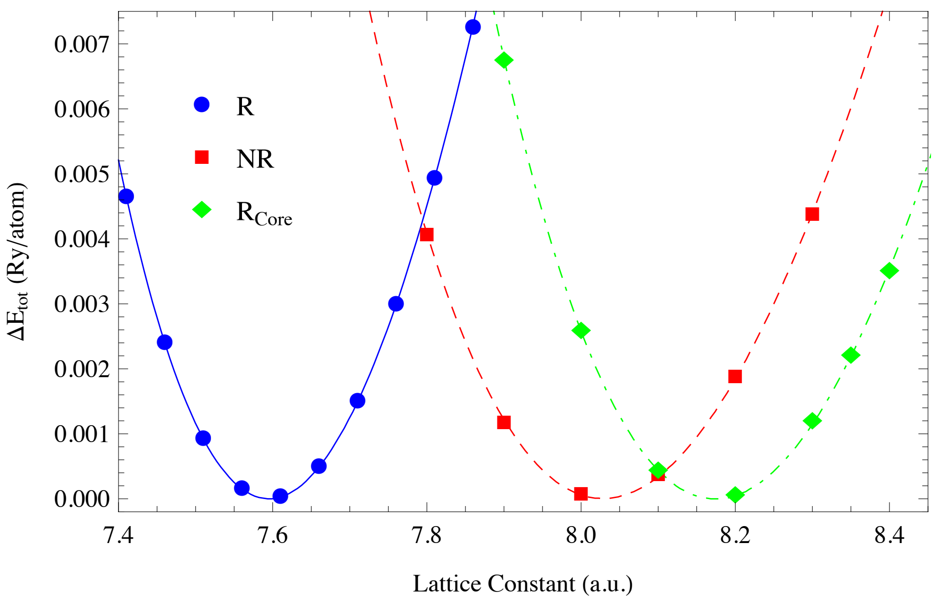
$$n(E) = -\frac{1}{\pi} \mathrm{Im}\ \mathrm{Tr} \int_\Omega G(E,r,r)dr$$

Instability of the irregular solution

- Integrate G_m along contour in complex plane
- Integrate G_s on real axis taking into account zeros of the KKR matrix.



Lattice Constant of Au

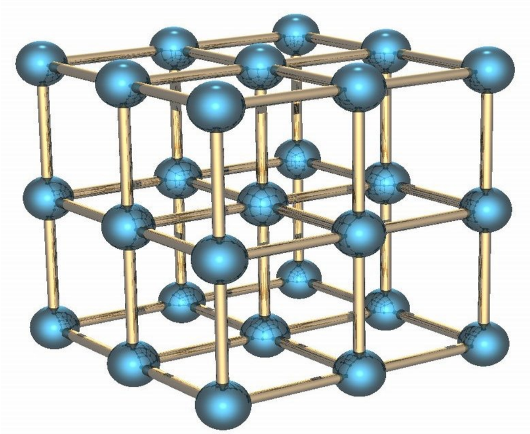


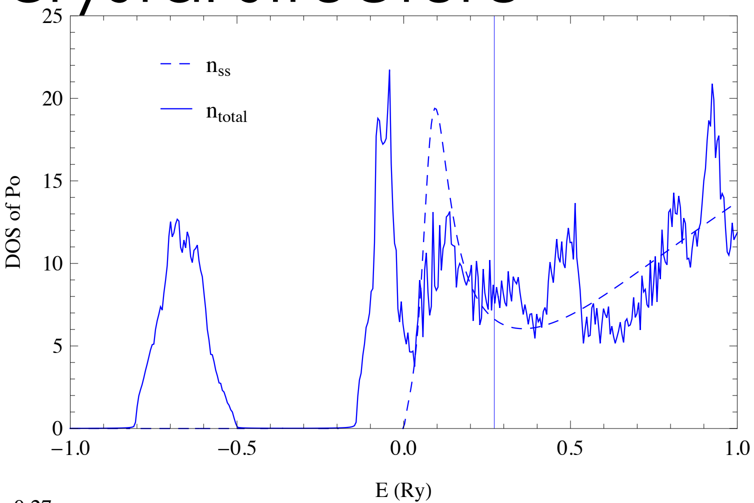
	Non-Rel	Rel Core	Full Rel.	Exp.
Lattice Const.	8.03Å	8.17Å	7.60Å	7.71Å
Bulk Modulus	112 GPa	124 GPa	208 GPa	180 GPa

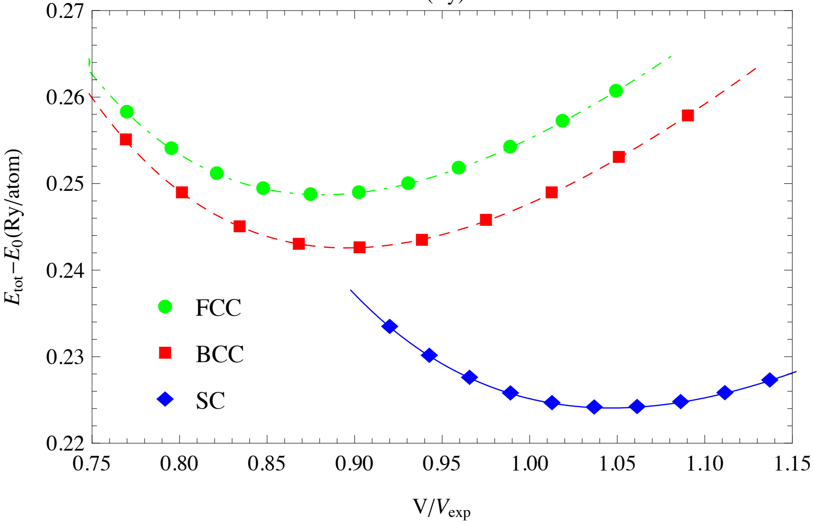
Example: Polonium crystal structure

α -Polonium is unique in having a simple cubic crystal structure.

Lattice Constant:
Exp.:   3.345Å
This work: 3.397Å





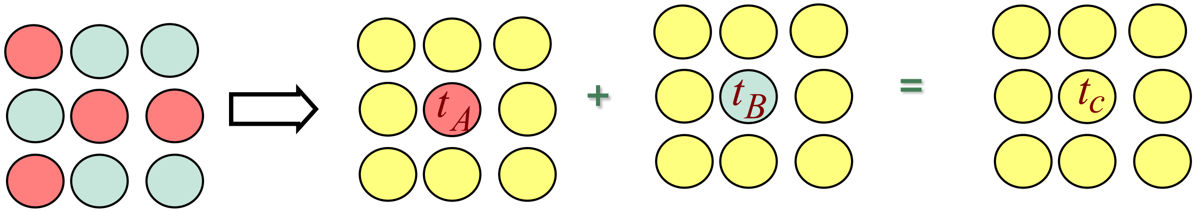


Disordered Systems

- Many systems are not perfectly periodic crystals.
- Disorder is important for understanding materials such as alloys.
  - Steel, Bronze, Brass, etc.
- Approaches for dealing with disordered and complex systems:
  - Coherent Potential Approximation: average probabilistic distribution of atoms
  - Locally Self-Consistent Multiple Scattering: real space large simulation cell to represent the actual placement of atoms

Coherent Potential Approximation

- Korringa-Kohn-Rostoker coherent-potential approximation (KKR-CPA)
  - Effective “coherent potential” medium
  - Characterized by effective t-matrix
  - Determine ‘best’ effective t-matrix
- KKR-CPA condition

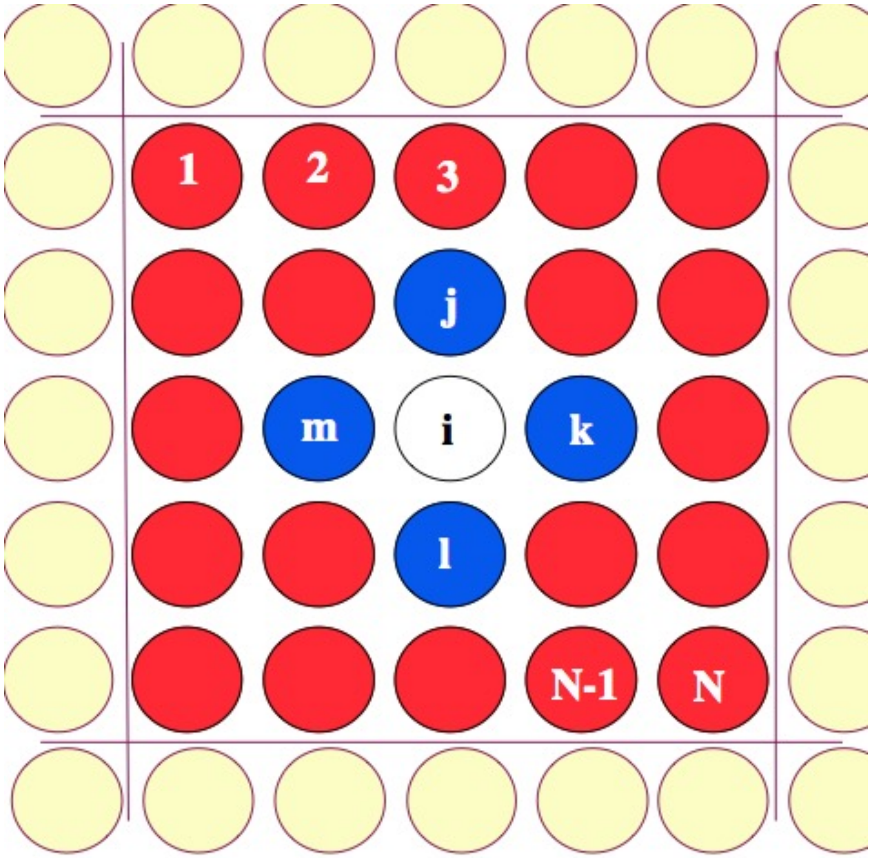


$$\sum_{\alpha} c_{\alpha} \tau^{\alpha,nn}(\varepsilon) = \tau^{C,nn}(\varepsilon)$$

$$\tau_{\alpha}^{c,nn} = [\mathbf{1} + \tau_{c}^{nn}(\mathbf{m}_{\alpha} - \mathbf{m}_{c})]^{-1} \tau_{c}^{nn}$$

$$\left\langle G(\mathbf{r},\mathbf{r}';\varepsilon) \right\rangle_{\alpha} = \tilde{G}^{\alpha}(\mathbf{r},\mathbf{r}';\varepsilon) = \sum_{LL'} [Z_{L}^{\alpha,n}(\mathbf{r};\varepsilon) \tau_{\alpha,LL'}^{c,nn}(\varepsilon) Z_{L'}^{\alpha,n}(\mathbf{r}';\varepsilon) - Z_{L}^{\alpha,n}(\mathbf{r}_{>};\varepsilon) J_{L}^{\alpha,n}(\mathbf{r}_{<};\varepsilon) \delta_{LL'}]$$

Locally Self-Consistent Multiple Scattering

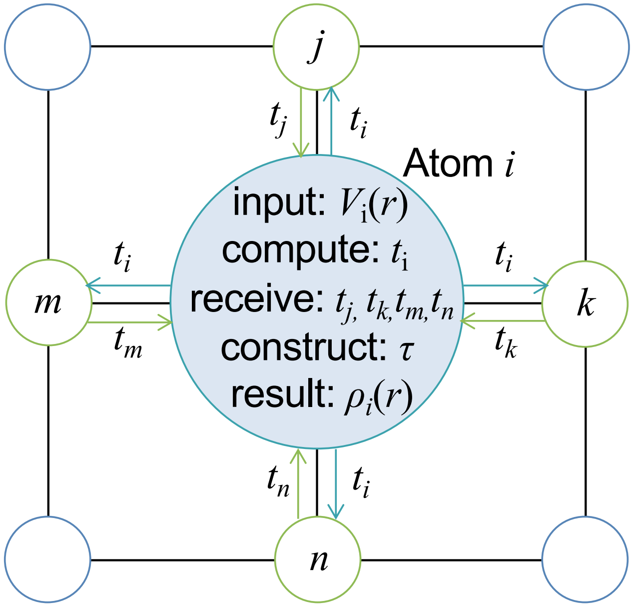


- Nearsightedness of electronic matter - Prodan & Kohn, PNAS **102**, 11635 (2005)
  - *Local electronic properties such as density depend on effective potential only at nearby points.*
- Locally self-consistent multiple scattering method - Wang et al., PRL **75**, 2867 (1995)
  - *Solve Kohn-Sham equation on a cluster of a few atomic shells around atom for which density is computed*
  - *Solve Poisson equation for entire system - long range of bare coulomb interaction*

Locally Self-Consistent Multiple Scattering

- An *O*[*N*] algorithm
- Massively parallel and scalable, achieves nearly perfect weak scaling
- Solve Kohn-Sham equation on a cluster of a few atomic shells around atom for which electron density, $\rho(r)$, is computed
- Solve Poisson equation for entire system (long range coulomb interaction)

$$\nabla^2 V_e(r) = n_\uparrow(r) + n_\downarrow(r)$$



$V_i(r)$: potential

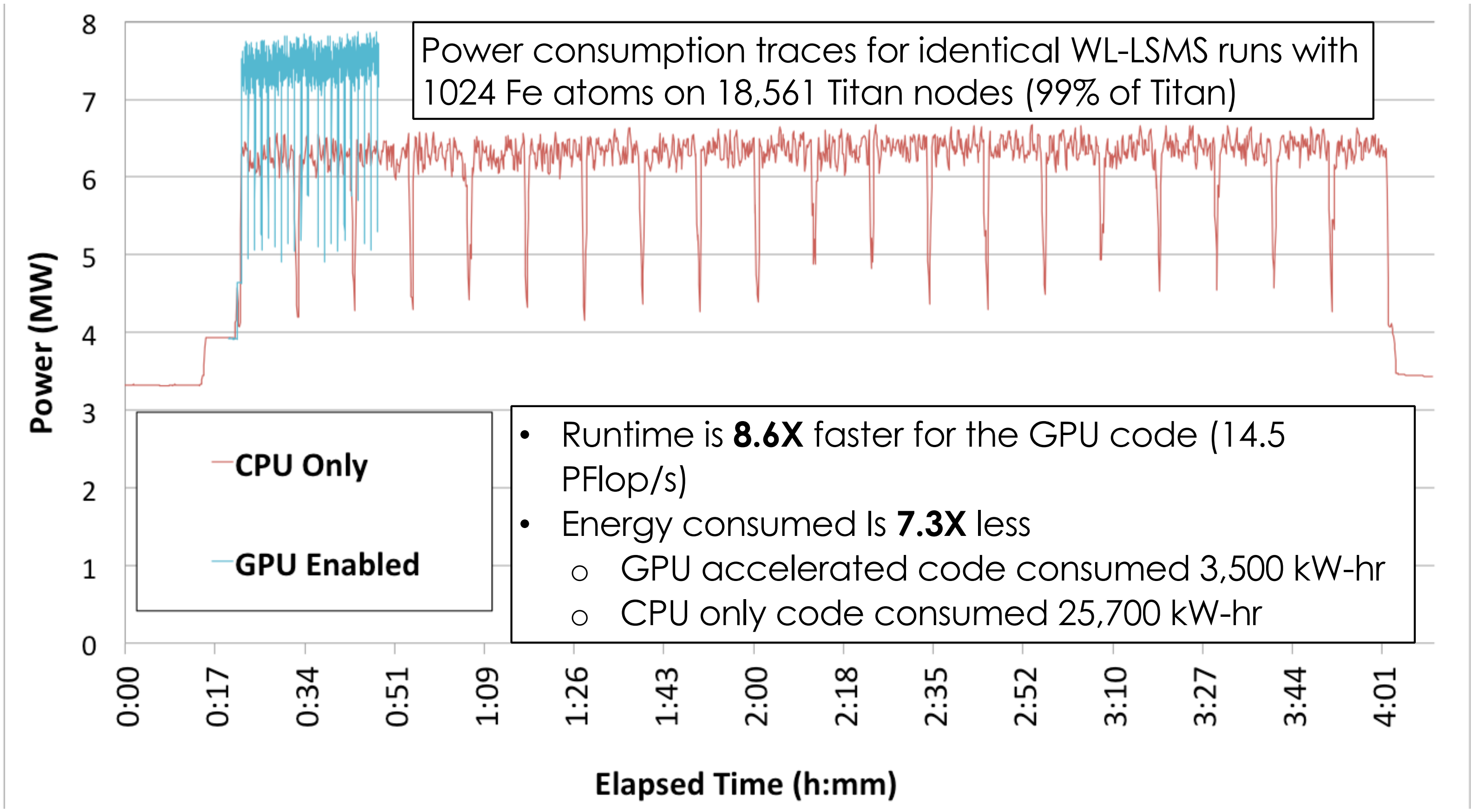
t_i : single site scattering matrix

τ : scattering path matrix

$\rho_i(r)$: charge density

Y. Wang *et. al.*, Phys. Rev. Lett. **75**, 2867 (1995)

WL-LSMS Power Efficiency on Titan

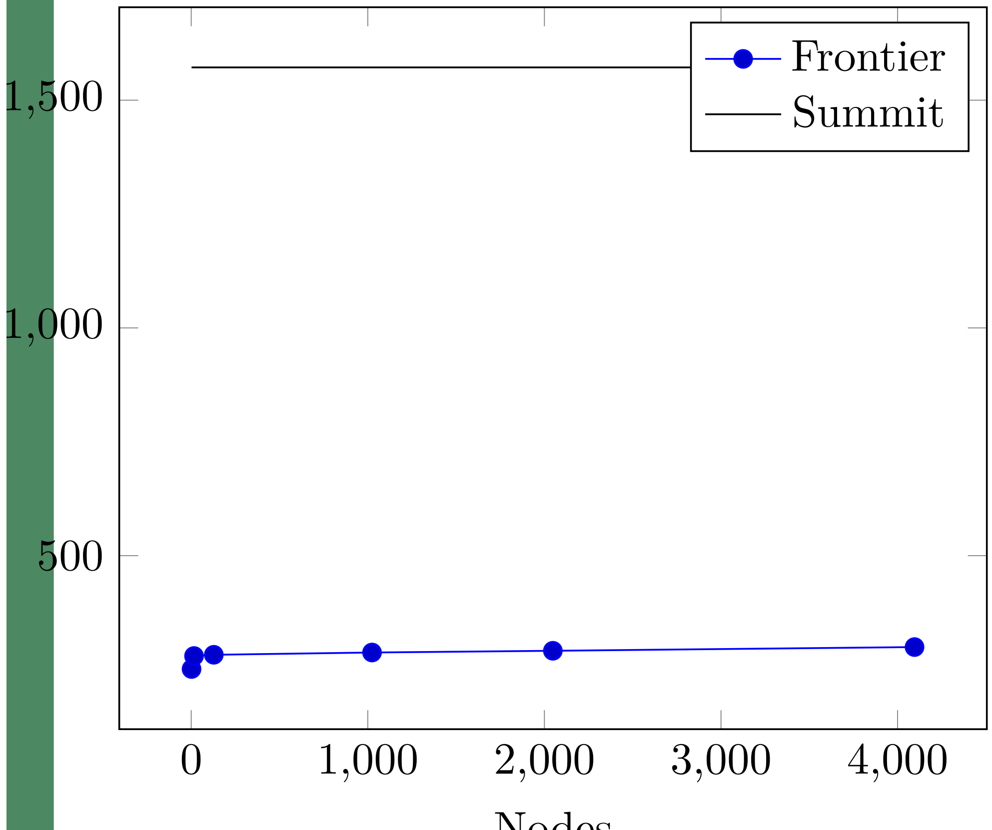


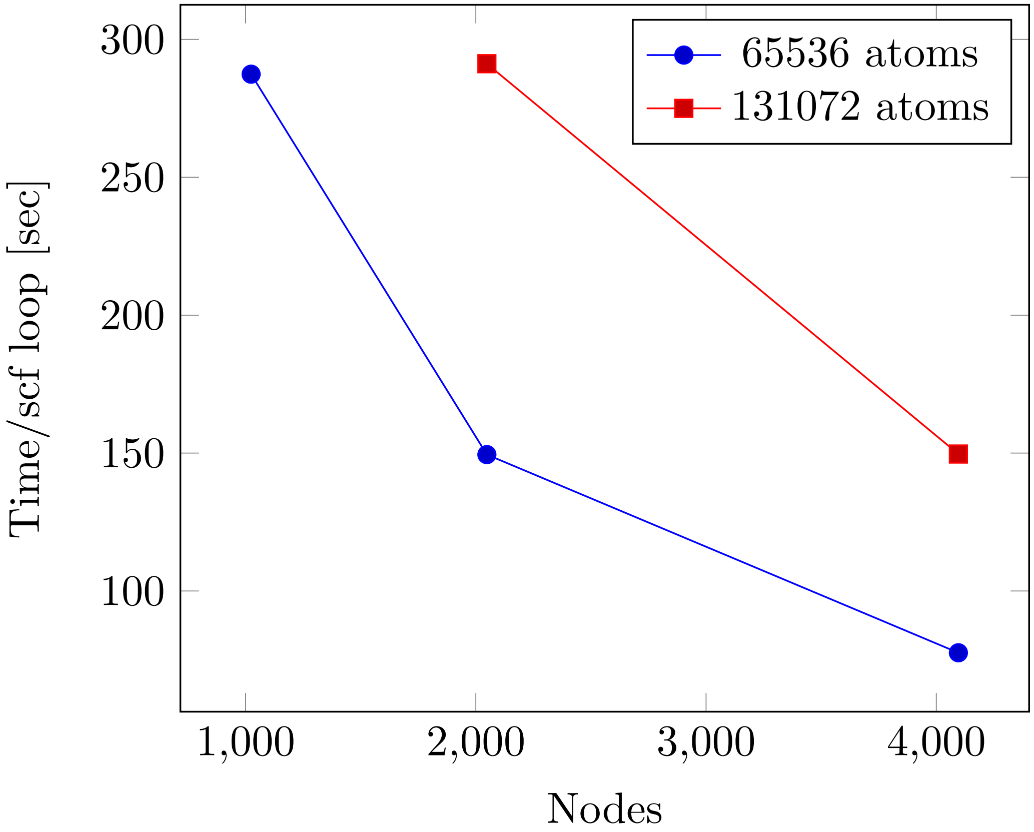
Power consumption traces for identical WL-LSMS runs with 1024 Fe atoms on 18,561 Titan nodes (99% of Titan)

- Runtime is **8.6X** faster for the GPU code (14.5 PFlop/s)
- Energy consumed Is **7.3X** less
  - GPU accelerated code consumed 3,500 kW-hr
  - CPU only code consumed 25,700 kW-hr

M. Eisenbach et al. CPC **212**, 2 (2017)

Scalability on Frontier

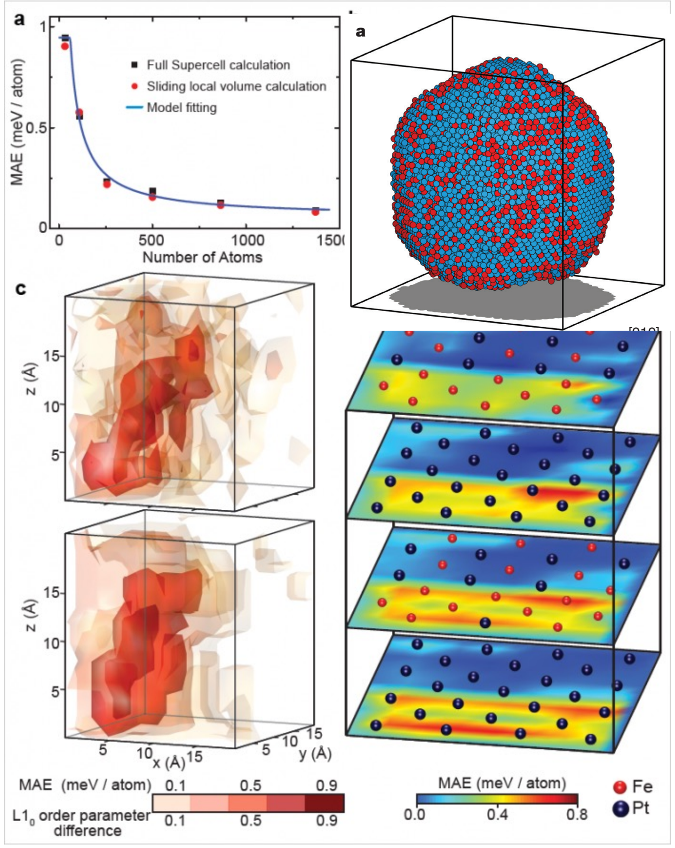




- Left: Weak scaling for FePt with 64 atoms / node from 2 to 4096 Frontier nodes. Right: Strong scaling on Frontier from 1024 to 4096 nodes

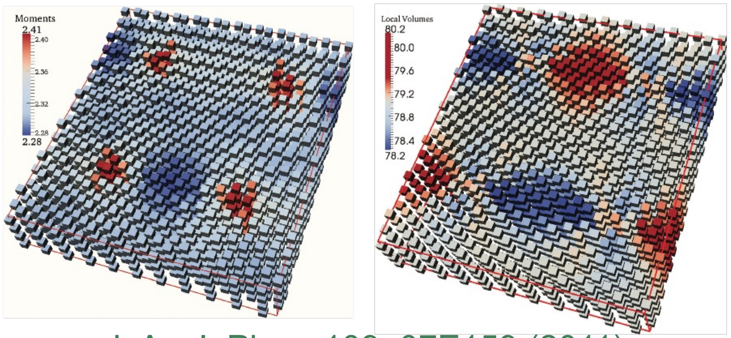
Example of LSMS calculations

Magnetic anisotropy in FePt nanoparticle

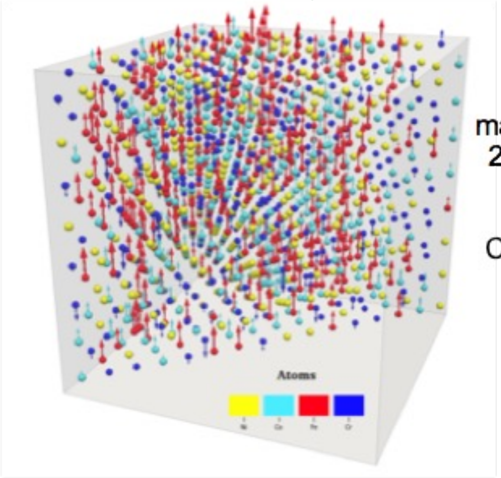


Nature 542, 75 (2017)

Screw Dislocation in bcc Fe with LSMS



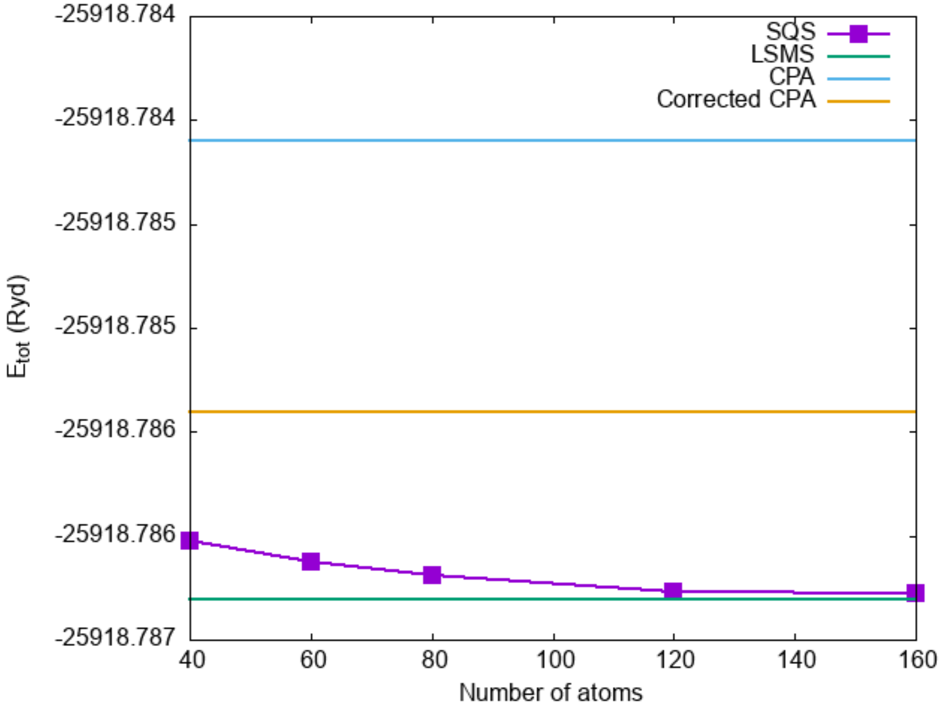
J. Appl. Phys. 109, 07E159 (2011)

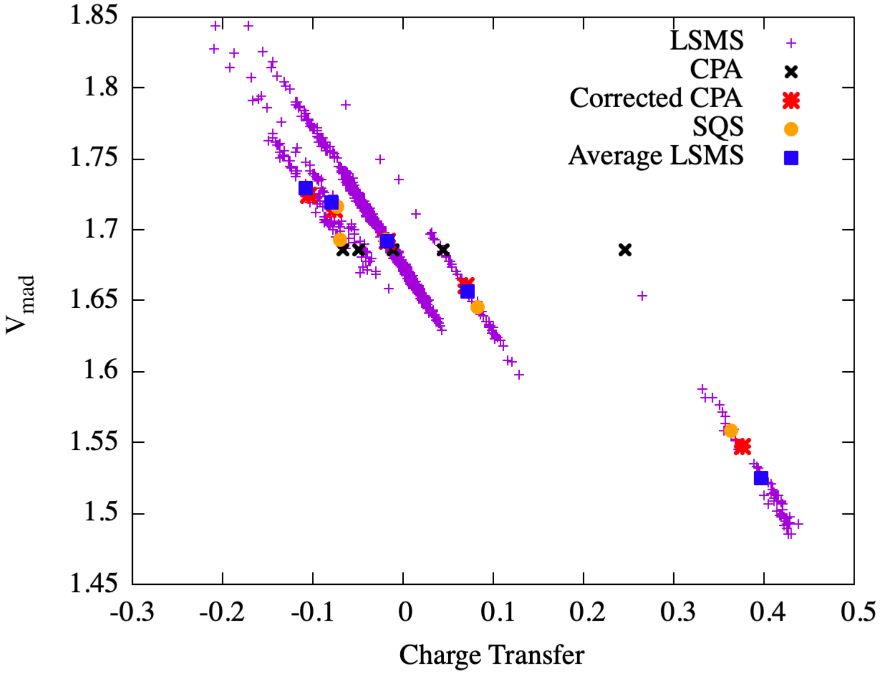


Ground state local magnetic moments in a 2048-atom super-cell model of the High Entropy Alloy $Cr_{0.25}Fe_{0.25}Co_{0.25}Ni_{0.25}$

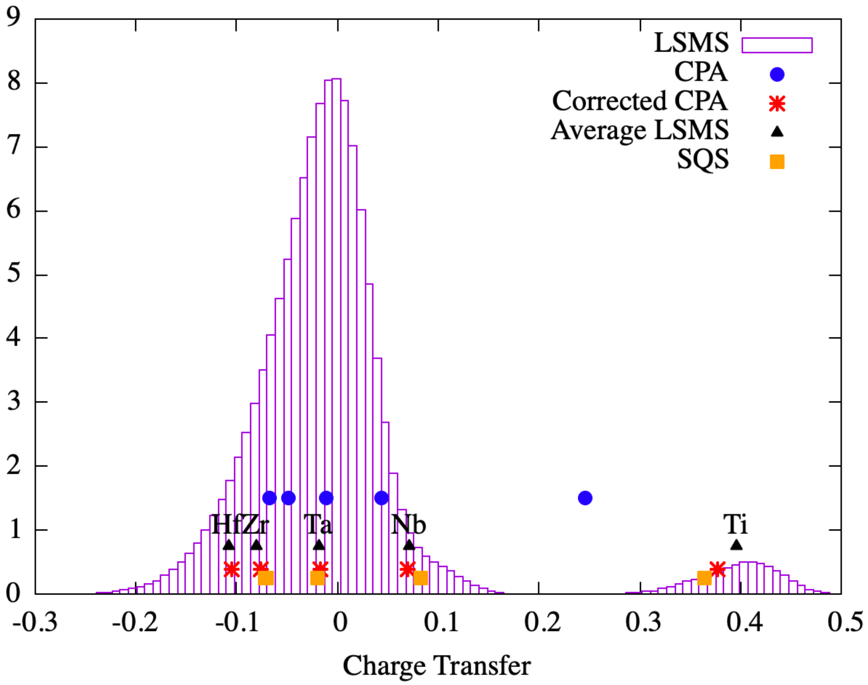
Comparison of LSMS and CPA

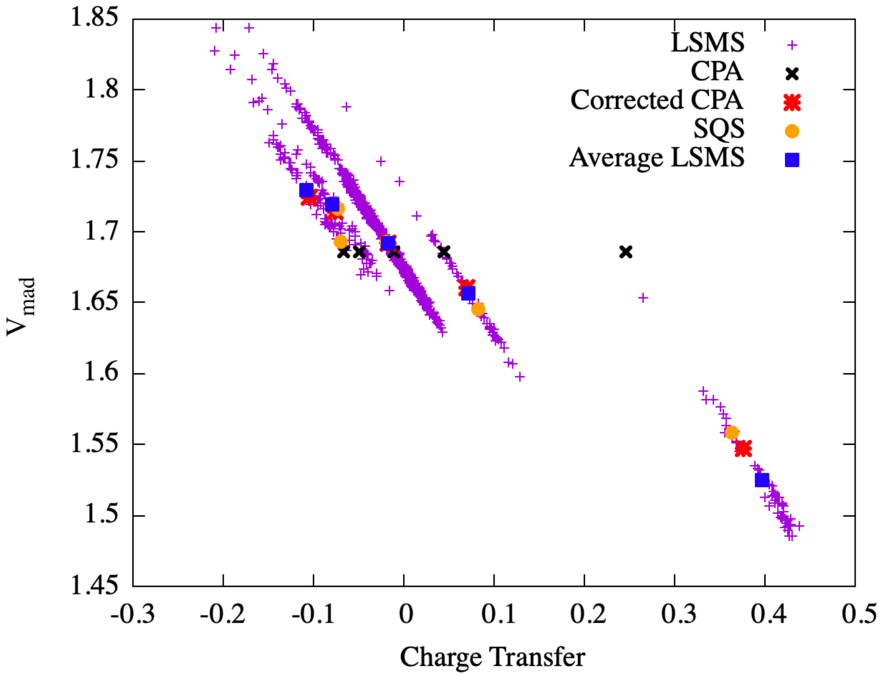
- We compare $Hf_{0.05}Nb_{0.05}Ta_{0.8}Ti_{0.05}Zr_{0.05}$ using
  - CPA, CPA with Madelung potential correction, LSMS for SQS structures (40-160 atom supercells) and LSMS for a 1120 atom random supercell





Changing local environments in alloys





Standard CPA assumes constant Madelung potential for all species

This is not true: A possible correction is charge screening by the average net charge per species

$$\Delta V_\alpha^{\mathrm{CS}} = -e^2 \frac{\Delta Q_\alpha}{R_1},$$

Statistical Mechanics

- Describes the behavior of a large ensemble of constituent particles that gives rise to macroscopic observables: thermodynamics.
- A physical system is described by phase space coordinates $\xi \in \Omega$ that describe the state of each constituent of the particles, e.g. position, momentum, atomic species, magnetic moment, etc.
- The dimension of Ω is huge: for a system describing the position and velocities of N atoms $\Omega=\mathbb{R}^{6N}$.
- The behavior of the system is determined by its Hamiltonian $H(\xi)$, or energy, that maps $\Omega \longrightarrow \mathbb{R}$

Statistical Mechanics

- At finite, non zero, temperature T (or inverse temperature $\beta=1/k_B T$) a system is in state ξ with a probability that is given by the Boltzmann distribution:

$$p(\xi;\beta) = \frac{e^{-\beta H(\xi)}}{\int_\Omega e^{-\beta H(\xi')} d\xi'}$$

- Observables of a system are measured as the averages over this probability density:

$$\langle A \rangle_\beta = \int_\Omega A(\xi) p(\xi;\beta) d\xi$$

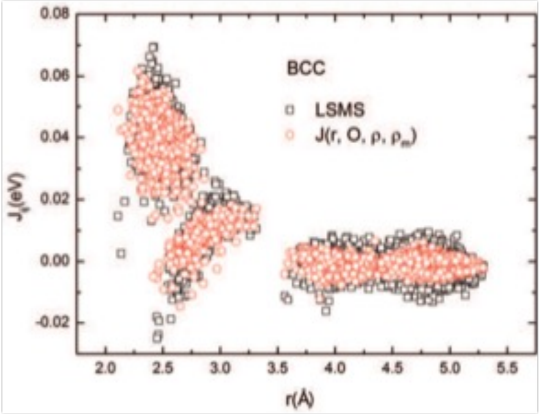
- The evaluation of this integral is the original application of the Metropolis algorithm

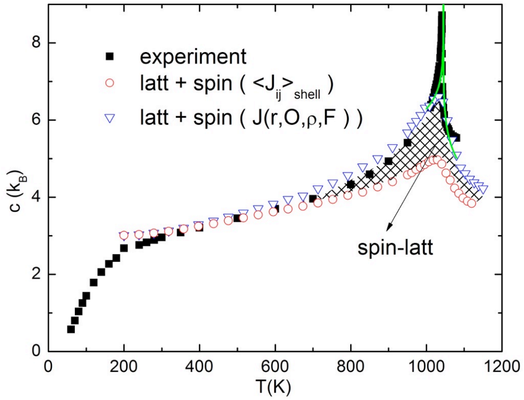
Statistical Mechanics of Materials from First Principles

- Traditionally MC sampling had been applied to model systems where the energy is fast and easy to evaluate.
- We are combining the statistical mechanics with first principles density functional theory calculations, where the energy of the system is the smallest eigenvalue of a partial differential equation with constraints that are given by the state of the system.
- Thus, $H(\xi)$ requires multiple CPU-hours to evaluate a single energy.
- ⇒ any reduction in the number of energy evaluations will save significant computational resources.
- ⇒ Build a surrogate model from first principles

Statistical Mechanics of Materials from First Principles

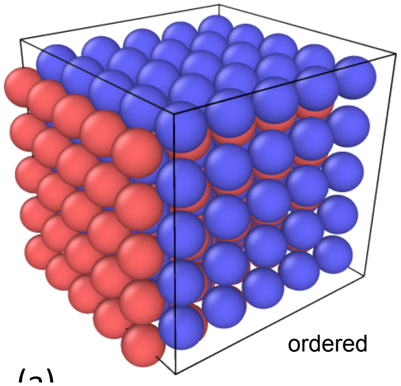
- Traditionally: perform first principles calculations and construct a model that fits well to describe the desired physics. E.g. Magnetism & lattice vibrations in Iron

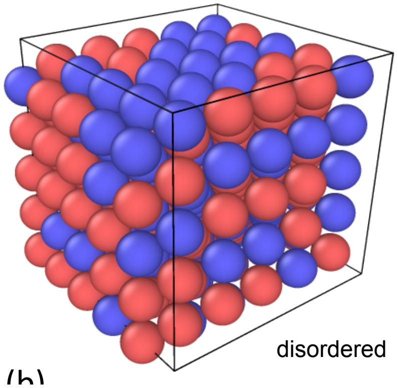


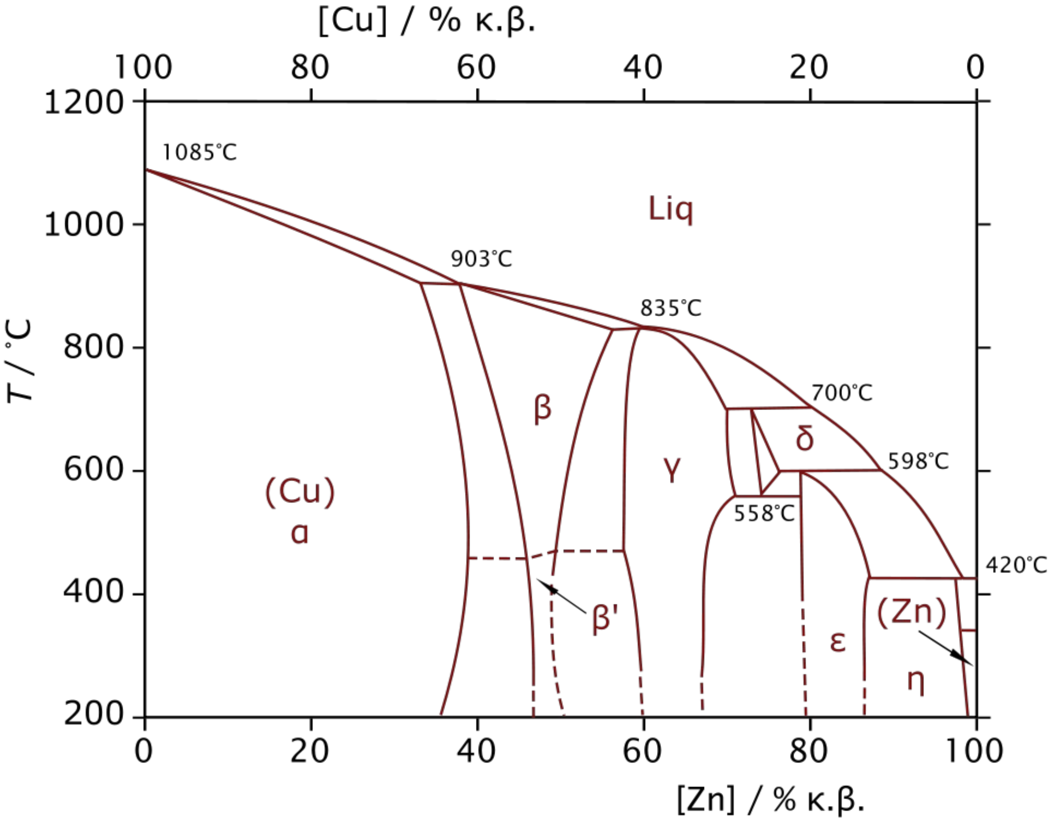


$$
\begin{aligned}
J_{ij}(r, O, \rho, \rho_m) = {} & (-7.2036956\text{E-}4\rho + 8.2453018\text{E-}3\rho_m - 4.9133771\text{E-}3\rho_m^2 \\
& + 9.7308351\text{E-}4\rho_m^3 - 1.0581546\text{E-}3r_{ij} + 2.2441192\text{E-}3 \\
& - 7.5827276\text{E-}4\rho\rho_m)(6.0 - r_{ij})^3\Theta(6.0 - r_{ij}) \\
& + (-1.7559806\text{E+}1r_{ij} + 5.4676556\text{E+}0r_{ij}^2 - 5.3687858\text{E-}1r_{ij}^3 \\
& + 2.8872662\text{E+}0\rho + 1.4496825\text{E+}1\rho_m - 4.3811389\text{E+}0\rho_m^2 \\
& + 3.1372442\text{E-}1\rho_m^3)O
\end{aligned}
$$

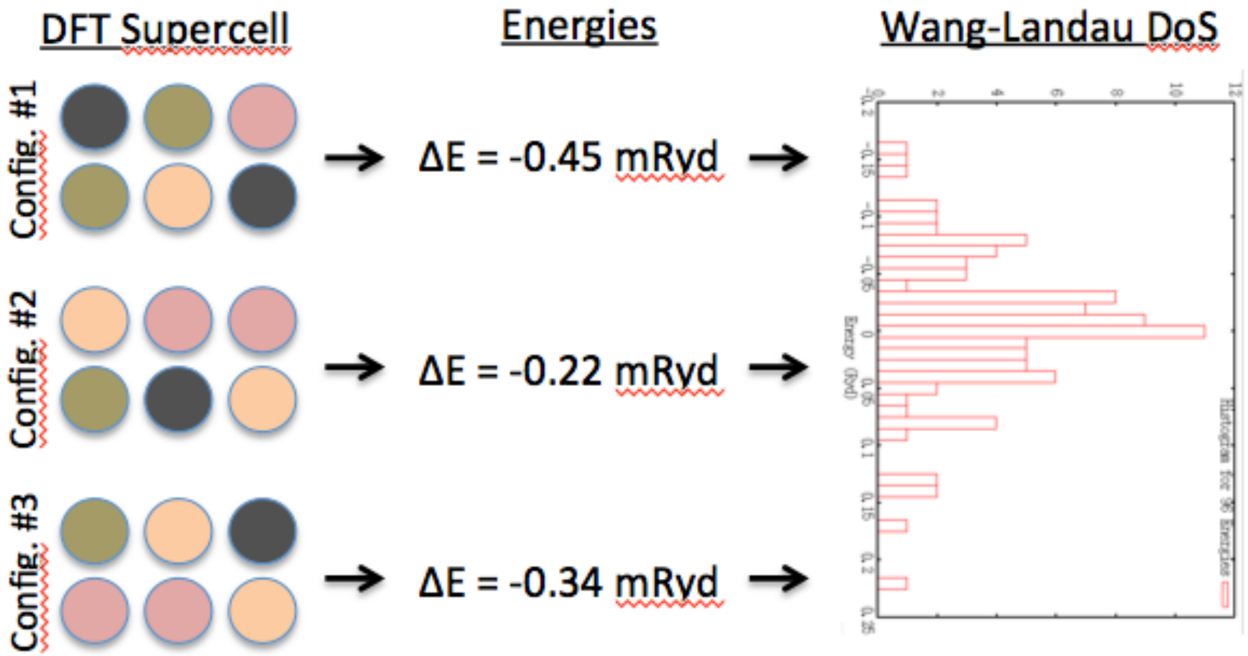
Order/Disorder Transitions – CuZn (bcc)







Sampling of chemical order using Wang-Landau



DFT Supercell

Energies

Wang-Landau DoS

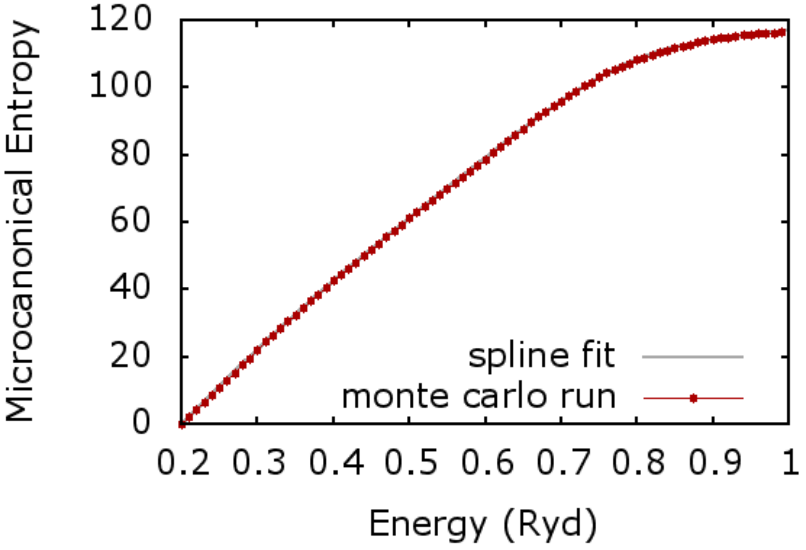
Config. #1 → ΔE = -0.45 mRyd →

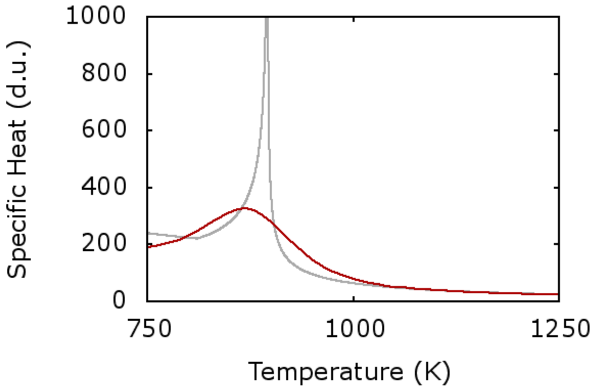
Config. #2 → ΔE = -0.22 mRyd →

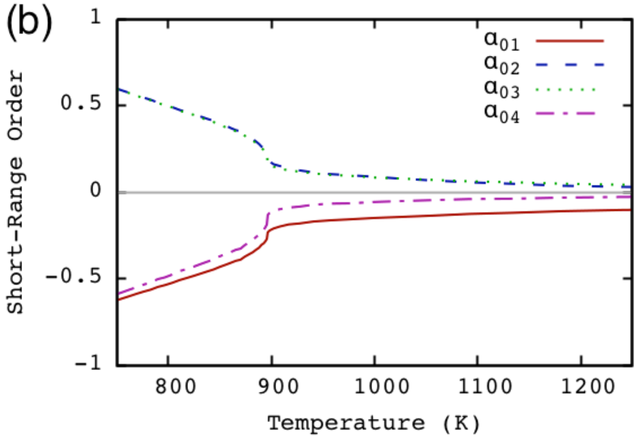
Config. #3 → ΔE = -0.34 mRyd →

Order/disorder transition in CuZn

- 250 site bcc lattice occupied by Cu and Zn atoms
- Monte-Carlo moves swap occupations of randomly selected pairs
- fully self consistent energy calculation for each configuration
- Calculated phase transition: 870K (exp. 750K)





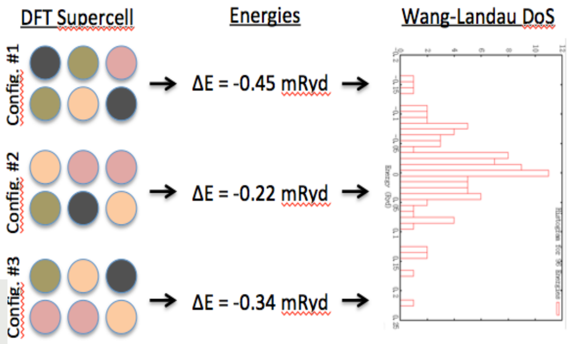


Example: First Principles Wang-Landau sampling of alloys

Monte Carlo-ab initio simulation of finite temperature statistical physics of magnets and alloys

Ultimately: **Reliably calculate material properties at finite temperature**

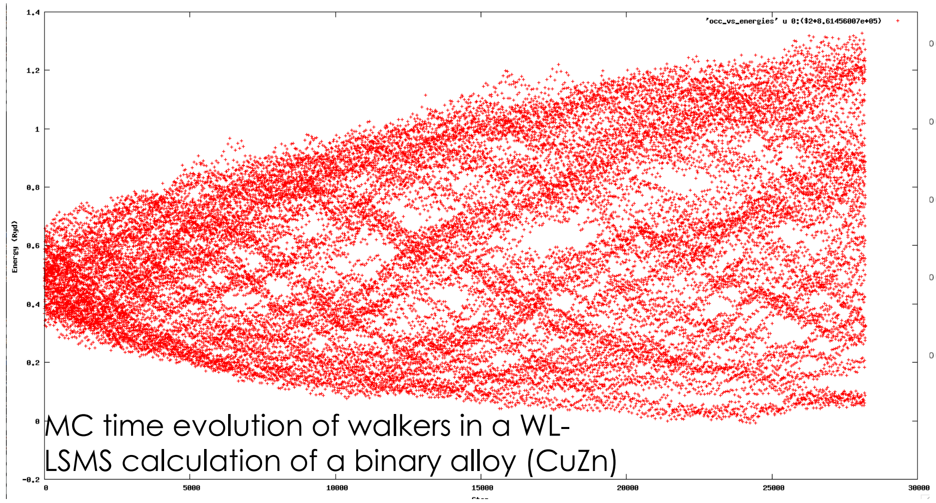
Each dot is a 250-atom DFT calculation !!
> 600,000 accepted, > 60% rejected moves



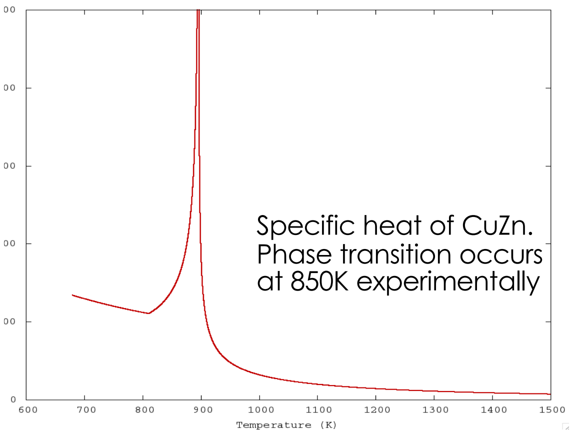
ΔE = -0.45 mRyd

ΔE = -0.22 mRyd

ΔE = -0.34 mRyd



MC time evolution of walkers in a WL-LSMS calculation of a binary alloy (CuZn)



Specific heat of CuZn. Phase transition occurs at 850K experimentally

S. Khan and M. Eisenbach, Phys. Rev. B **93**, 024203 (2016).

Goal: Accelerate Monte Carlo Stochastic Sampling

Total computational cost:

- T_1: Calculate the change in energy
  $$p(x_i)$$
- T_2: "Random walk" with probability
  $$A(x_i, x^*)$$

$$\min \quad T = \psi(T_1, T_2)$$

1. Generate a new configuration x^* from x_i with probability $q(x_i, x^*)$
2. Calculate the change in energy, ΔE
3. Accept with probability:
   $$p(E \to E + \Delta E) = \min\left\{1, e^{-\Delta E/k_B T}\right\}$$
4. Calculate and accumulate physical observables
5. Repeat steps 1-3 until a desired number of steps

Minimize the number of expensive evaluations of model functions

- decrease the time-to-solution ratio for stochastic simulations

Formulate a machine learning approach with two model levels:

$$A(x_i, x^*) = p(x^*)q(x^*, x_i)/p(x_i)q(x_i, x^*)$$

Level 2 - Predict a classifier model based on acceptance probability $A(x_i, x^*)$ to directly generate Monte Carlo moves

Level 1 - Predict a regression model for the probability density distribution $p(x)$

Supervised Learning

- **Goal**: predict the total energy associated with a lattice configuration for an alloy system (e.g. Cu-Au, Fe-Pt)
- **Idea**:
    1. Generate multiple lattice configurations and estimate the related total energy with accurate DFT calculations
    2. Use the generated lattice configurations and estimated total energy as training set and test set for a neural network
- **Challenges**:
    - Risk of overfitting (low quality estimate for new incoming data)
    - Training of a neural network is a global non-convex optimization problem (between multiple local and global minima, only physically admissible ones are of interest – naive training could result in models with non-physical behavior)

Local Atomic Energy

The previous presented results used the total energy of the system as calculated by DFT.

The total Energy is taken to be the sum of local site contributions: $E_t = \sum_i E_i.$

With $E_i = H(\vec{\sigma}_i) + \epsilon$ and $\vec{\sigma} = (\sigma^0, \sigma^1, \cdots, \sigma^{N_L - 1})$ the vector of site occupations within the local interaction zone.

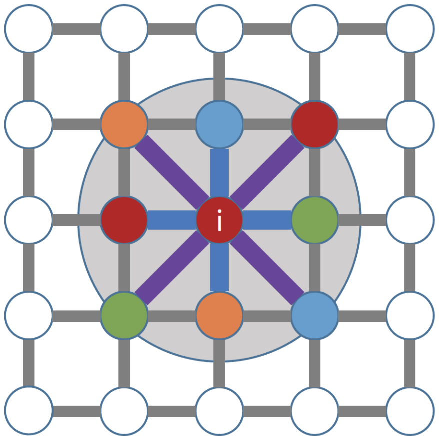
A common approach is cluster expansion:

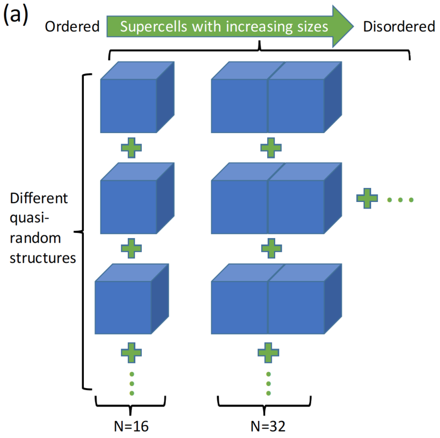
$$E(\sigma) = J_0 + \sum_i J_i\sigma_i + \sum_{i,j} J_{ij}\sigma_i\sigma_j + \sum_{i,j,k} J_{i,j,k}\sigma_i\sigma_j\sigma_k + \ldots$$

The EPI model is a special case with

$$E_i = \sum_f V^f \Pi^f(\vec{\sigma}_i) + V^0 + \epsilon$$

Where f is the combined feature index (species, shell) and Π is the count of sites with this feature.





Neural Networks for local atomic energy

As the local energy training data set can be significantly larger than the total energy data sets, we can explore more complex models that can allow the local energies to be non-linear functions.

An example would be a quadratic pair interaction term:

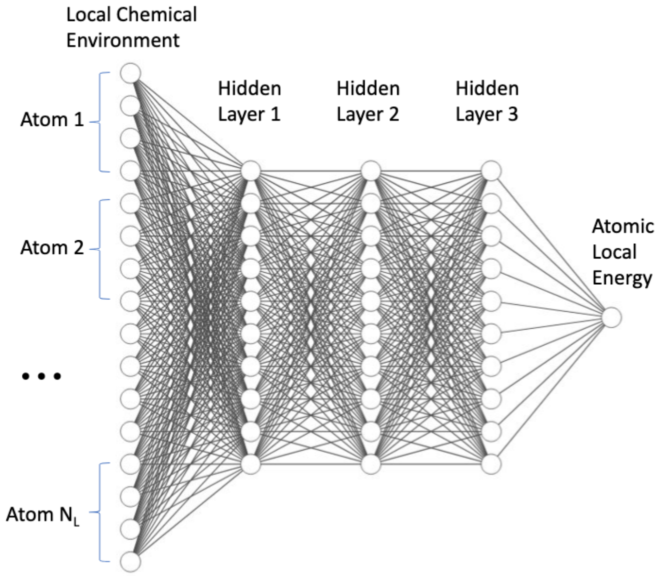
$$E_i = \sum_f V^f \Pi^f(\vec{\sigma}_i) + \sum_{f,f' \leq f} V^{ff'} \Pi^f(\vec{\sigma}_i) \Pi^{f'}(\vec{\sigma}_i) + V^0 + \epsilon.$$

The calculation of the Π takes into account the lattice symmetry. It is possible to write the local energy directly in configuration space, e.g.

$$E_i = \sum_{j \in \mathrm{LIR}} V^j \sigma_i^j + \sum_{j,k>j \in \mathrm{LIR}} V^{jk} \sigma_i^j \sigma_i^k + V^0 + \epsilon$$

This forms the basis of a neural network to predict the local energy.

The input layer represents the site occupation by a binary label.



Supervised Learning: Neural Networks (cont. 1)

Approach: introduce physical constraints in the optimization process to train the neural network

$$\begin{cases} \underset{\mathbf{w}}{\operatorname{argmax}} \| E_{\text{true}} - E_{\text{predicted}}(\mathbf{w}) \| \\ \text{s.t.} \quad \mathbf{c}_{\text{true}} = \mathbf{c}_{\text{predicted}}(\mathbf{w}) \end{cases}$$

Objective function

Physical constraint

Possible quantities for the physical constraint are charge density or magnetization vectors

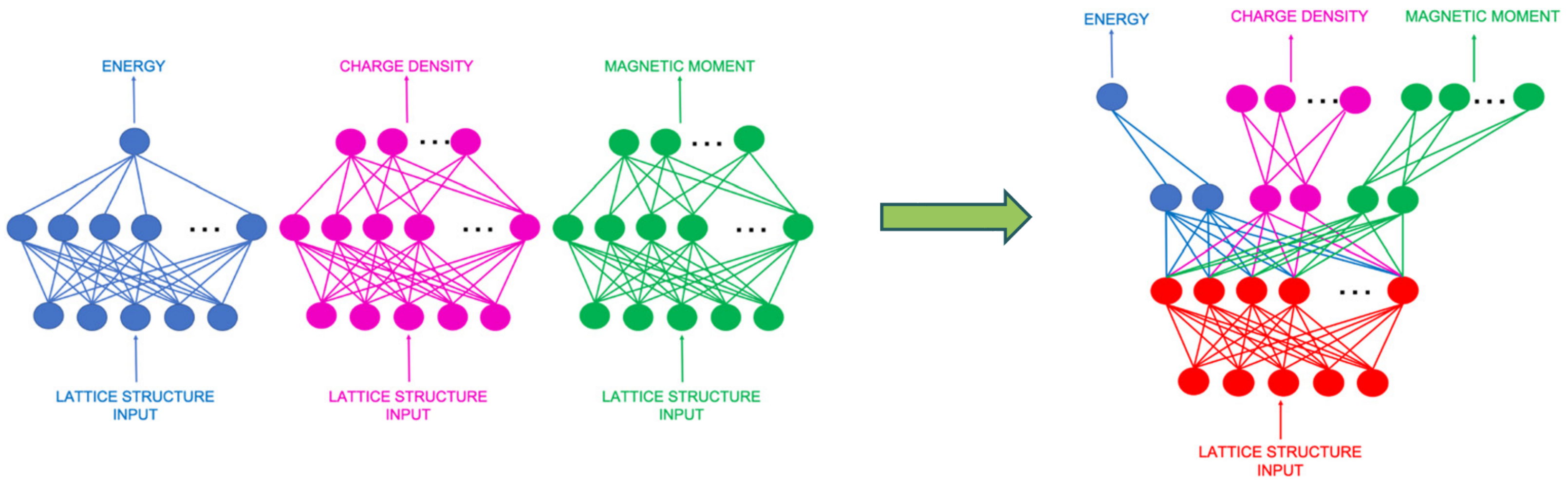
The constrained optimization problem can be recast as a global optimization problem by interpreting the constraint as a Lagrangian term in the objective function

Joint training or multitasking: train a neural network to simultaneously predict multiple targets associated with the same input

$$\underset{\mathbf{w}}{\operatorname{argmax}} \| E_{\text{true}} - E_{\text{predicted}}(\mathbf{w}) \| + \lambda \| \mathbf{c}_{\text{true}} - \mathbf{c}_{\text{predicted}}(\mathbf{w}) \|$$

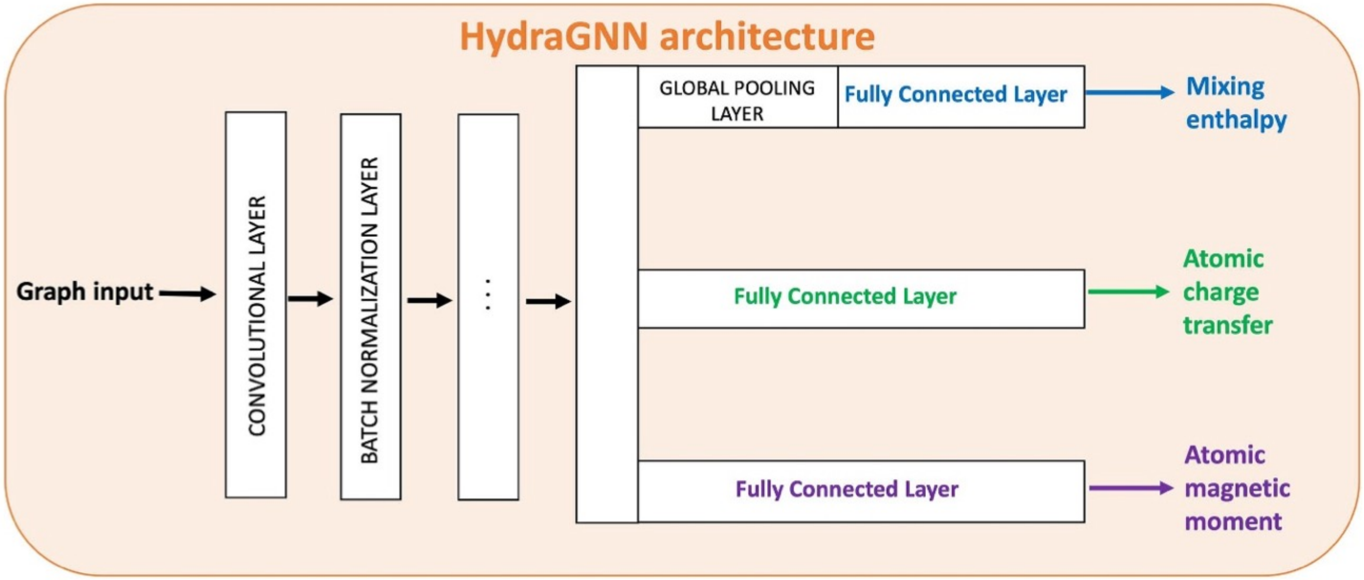
HydraGNN

- Graph convolutional neural network:
  - atoms are interpreted as nodes and interatomic bonds are interpreted as edges, and outputs total (graph-level) and atomic (node-level) physical properties



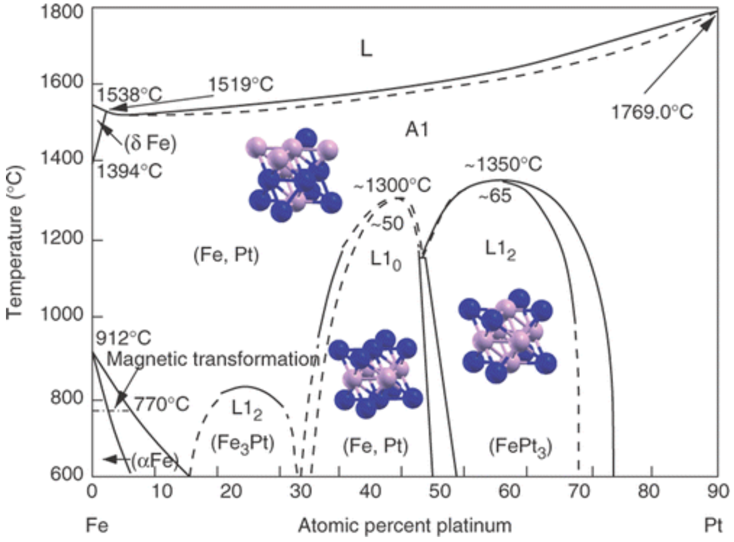
HydraGNN

- The model takes the atomic number for each position and generates the enthalpy as well as vectors of the charge and magnetic moments at each site



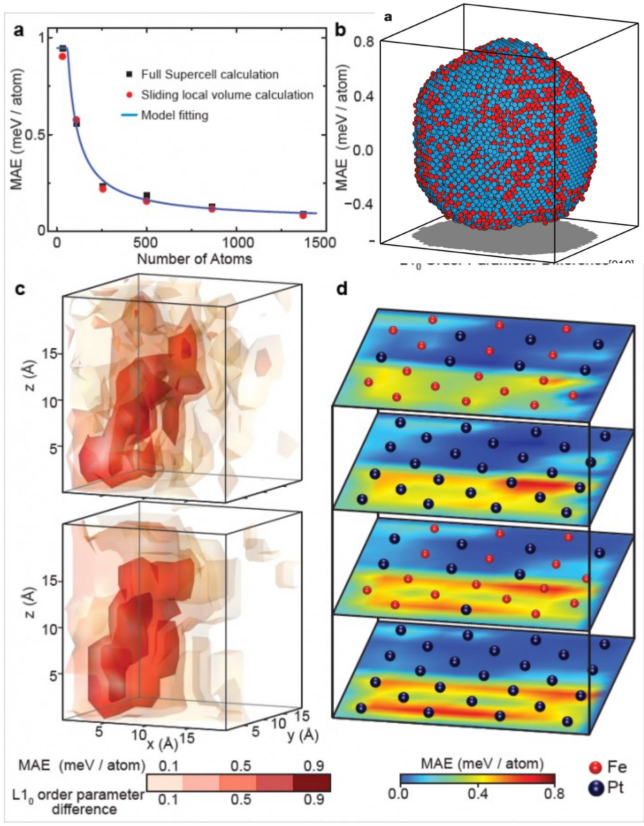
Iron-Platinum

- FePt is of interest for hard magnetic materials
- The magnetic properties depend significantly on the chemical order



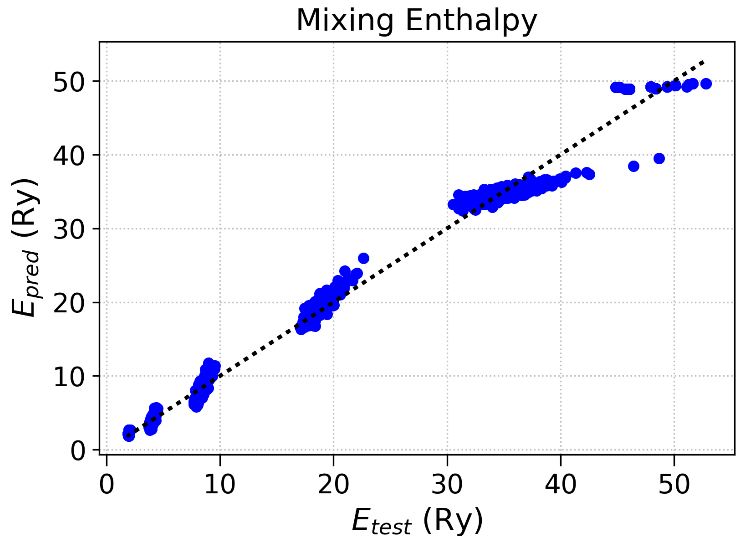
Phil. Mag. 88, 2725 (2008)

Magnetic anisotropy in FePt nanoparticle

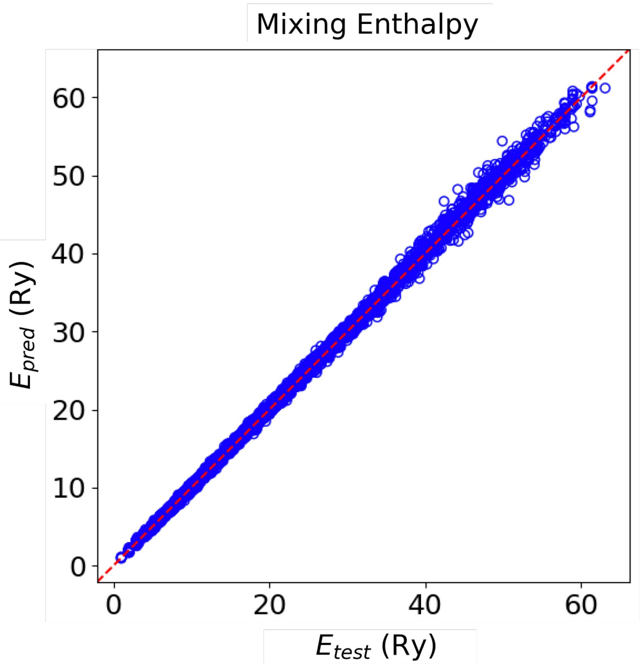


Nature 542, 75 (2017)

Numerical Result - Enthalpy



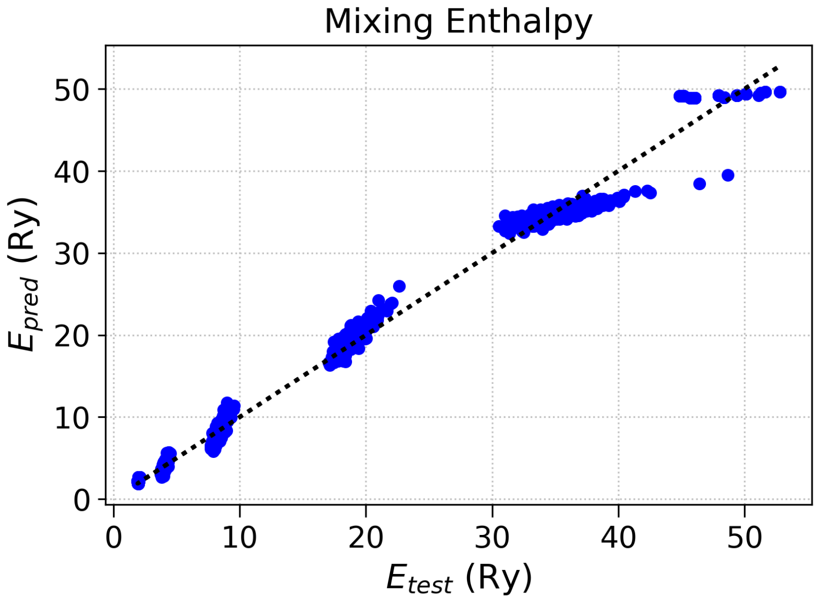
Linear Model



HydraGNN

Numerical Result - Enthalpy

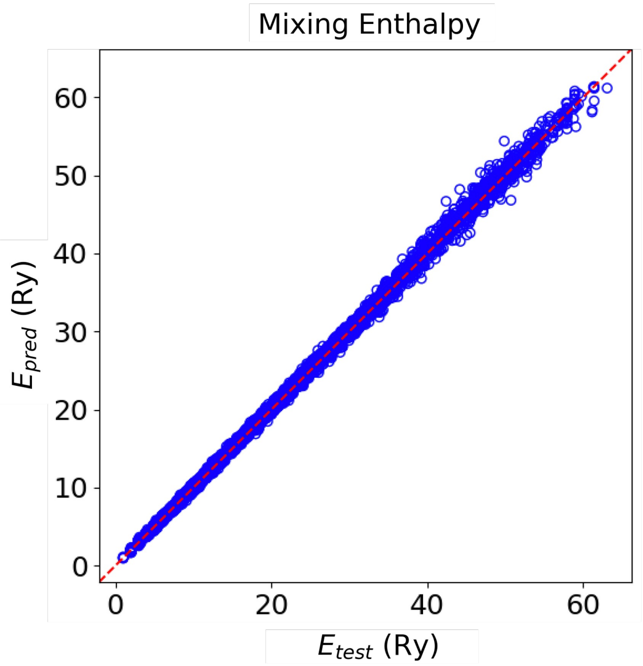
- The EPI model fails to capture the non-linear behavior of the mixing enthalpy
- The model shows clustering of its behavior for different concentrations
- A good model could be obtained for a fixed concentration, but not across all concentration
- This indicated the non-linear contribution arising from magnetism in the FePt system



Linear Model

Numerical Result - Enthalpy

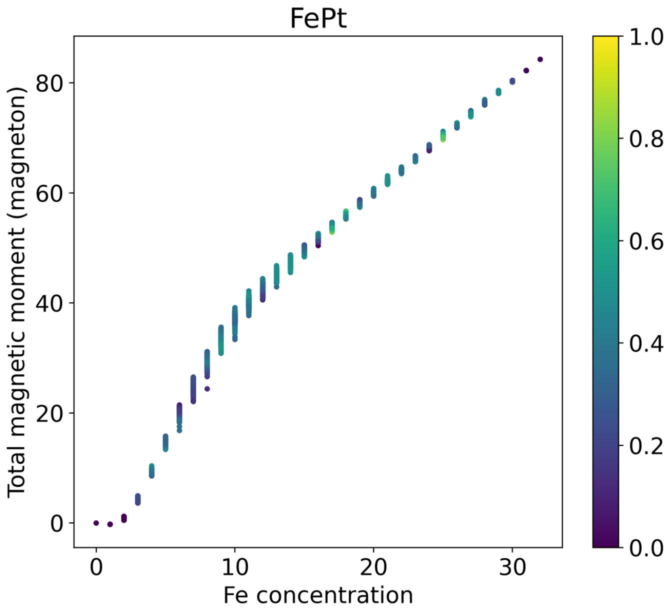
- HydraGNN accurately predicts the mixing enthalpy for different compositions and atomic configurations associated with each composition
- It is an effective non-linear predictive model
- It accurately captures the dependence of the mixing enthalpy as a function of a binary ferromagnetic solid solution alloy



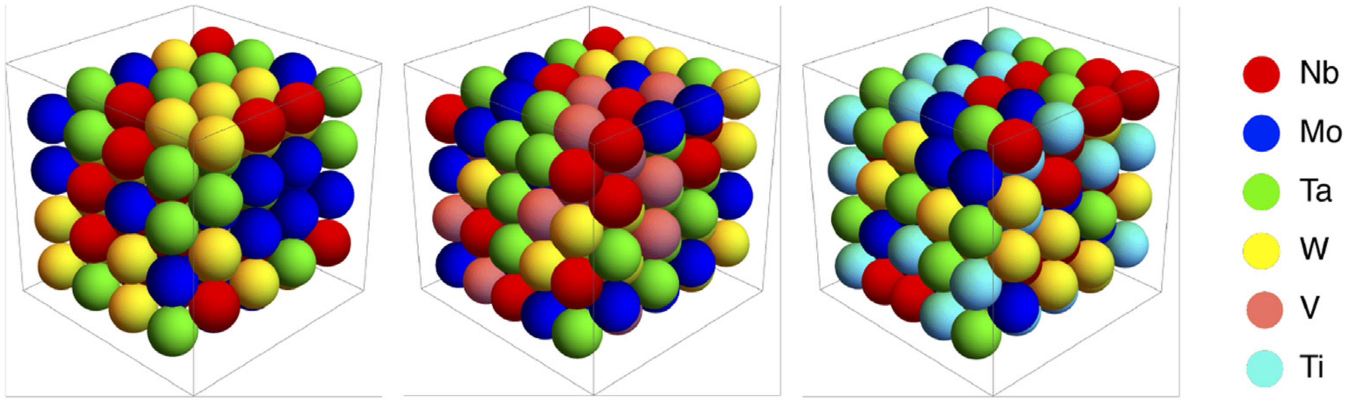
HydraGNN

Interpretation

- Pure Fe is magnetic, and Pt is non-magnetic
- Formation of magnetic moments in alloys is driven by the collective behavior of electrons in the alloy
- in the FePt, the magnetic moment associated with the Pt sites depend on their environment and the Fe concentration
- This behavior can be captured by the HydraGNN model, while the EPI model fails to account for this concentration dependent behavior.



Modeling High Entropy Alloys



NbMoTaW NbMoTaWV NbMoTaWTi

High Entropy Alloys (concentrated solid solution alloys) consist of four or more principal alloying element. They exhibit a wide range of interesting properties. The chemical order is important to understand these.

Due to the large design space, modeling is essential in guiding and understanding the experimental work and for gaining insight into the stability of these alloys.

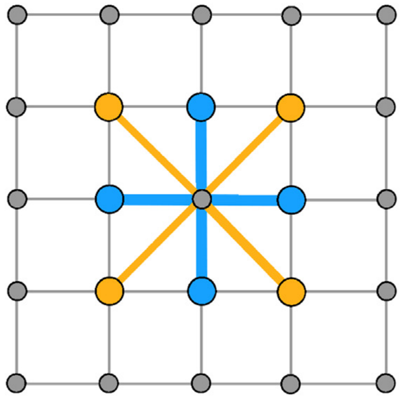
Traditional cluster expansion approaches of force fields become challenging for these multicomponent alloys.

Modeling High Entropy Alloys

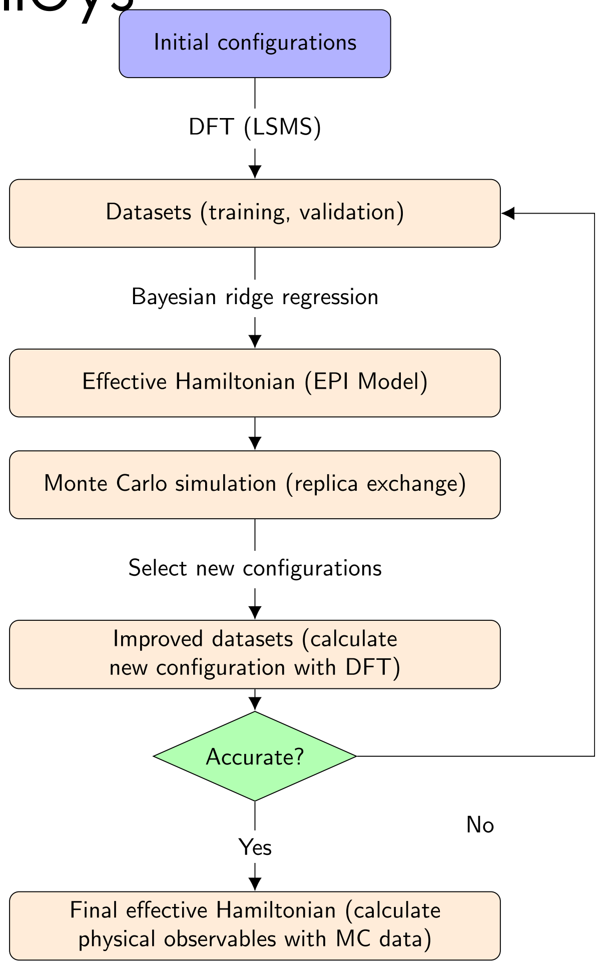
Ansatz: consider only pair interactions

$$\tilde{E}(\boldsymbol{\sigma}) = NJ_0 + N \sum_{X,Y,m} J_m^{X,Y} \sigma_m^{X,Y}$$

X,Y: chemical species
m: coordination shell



Use the Bayesian information criterion (BIS) to select the optimal number of shells. (m)



Initial configurations

DFT (LSMS)

Datasets (training, validation)

Bayesian ridge regression

Effective Hamiltonian (EPI Model)

Monte Carlo simulation (replica exchange)

Select new configurations

Improved datasets (calculate new configuration with DFT)

Accurate?

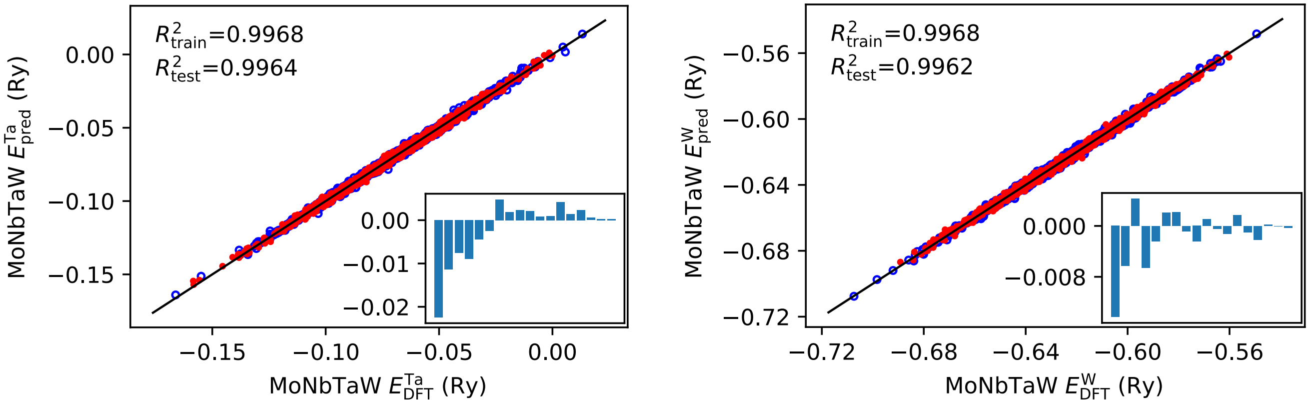
No

Yes

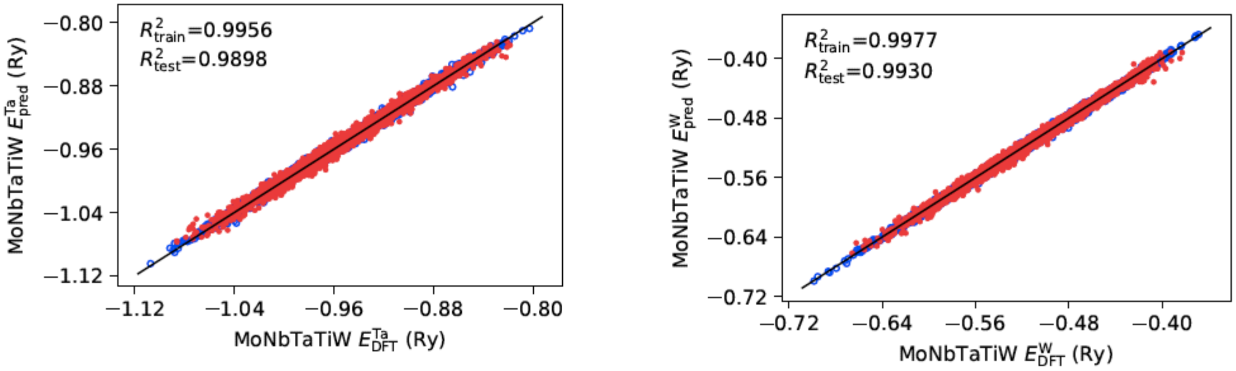
Final effective Hamiltonian (calculate physical observables with MC data)

Results

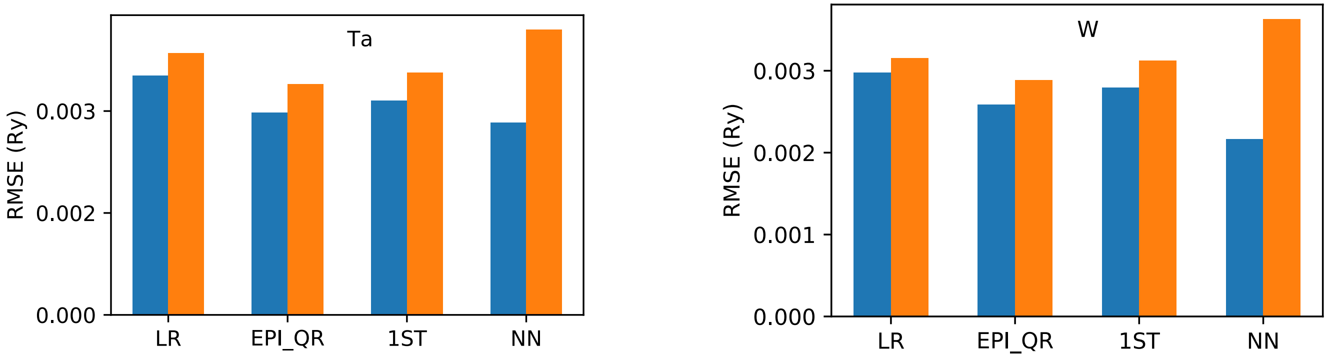
Linear EPI:



Neural Network:



Results

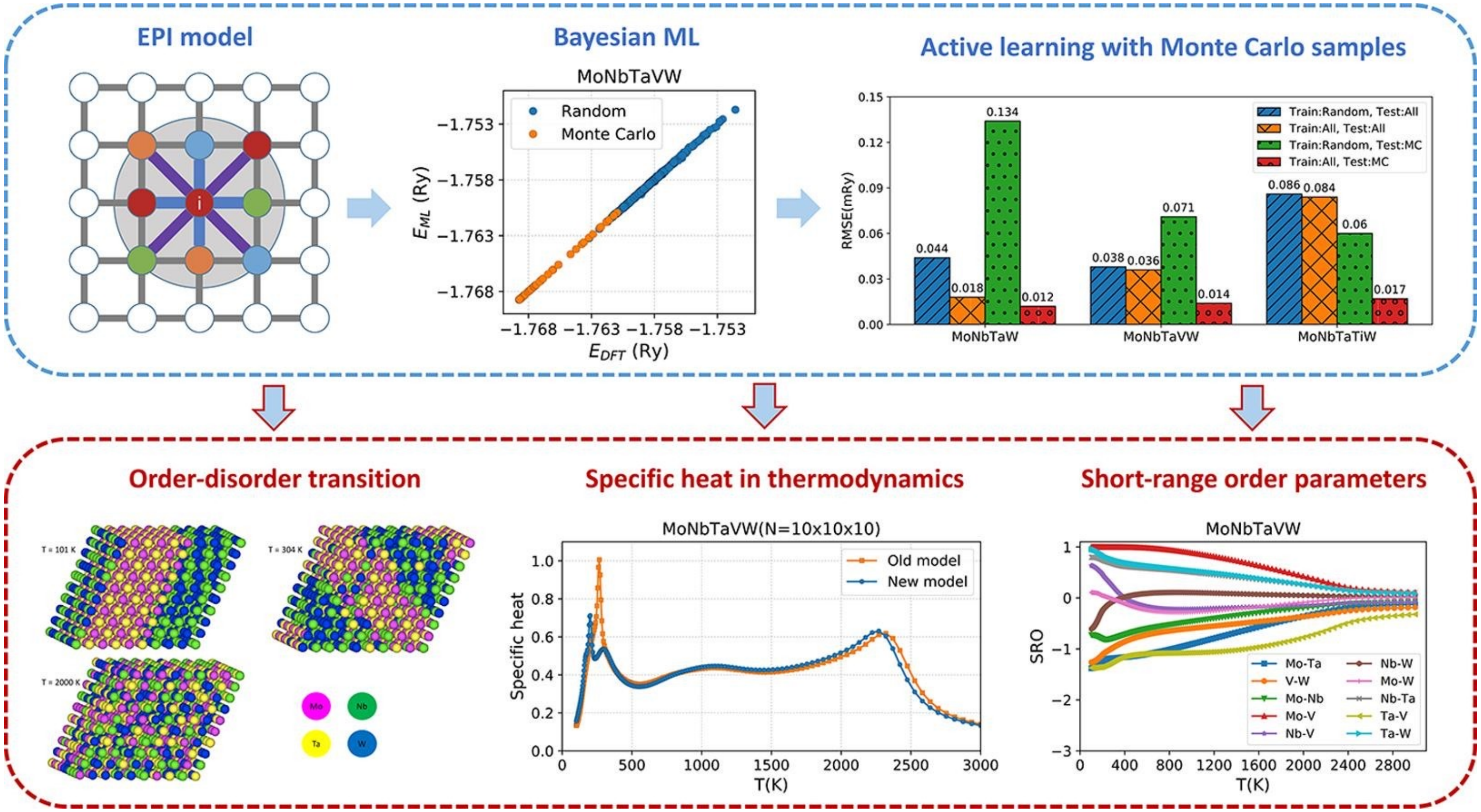


Training (blue) vs testing (orange) for the various models with the same training data set. (38,400 data points)

It can be seen that compared to the linear model (LR), non linear contributions (EPI_QR) are more important than triplet interactions (1ST).

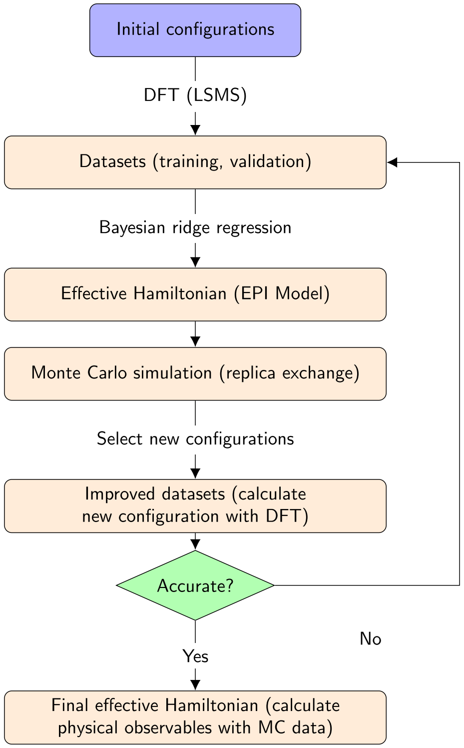
From the training results, the NN model demonstrates the capability to represent the data noticeably better than the other models, but due to the significantly larger number of parameters the training set size is not large enough to avoid some overfitting as indicated by the testing results.

Statistical mechanics with HPC & ML



Conclusion

- Using HPC to generate sufficient amount training data from first principles with high accuracy using scalable code (LSMS)
- Training of Surrogate Model
- Statistical Mechanics Monte-Carlo simulation
- **Active refinement of the model with new HPC generated data driven by the simulation**
- **⇒ Experimentally verifiable prediction of material specific finite temperature behavior of complex materials**
- **Enabled by tight coupling of HPC & ML & MC workflow**



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- Our open source codes: github.com/mstsuite

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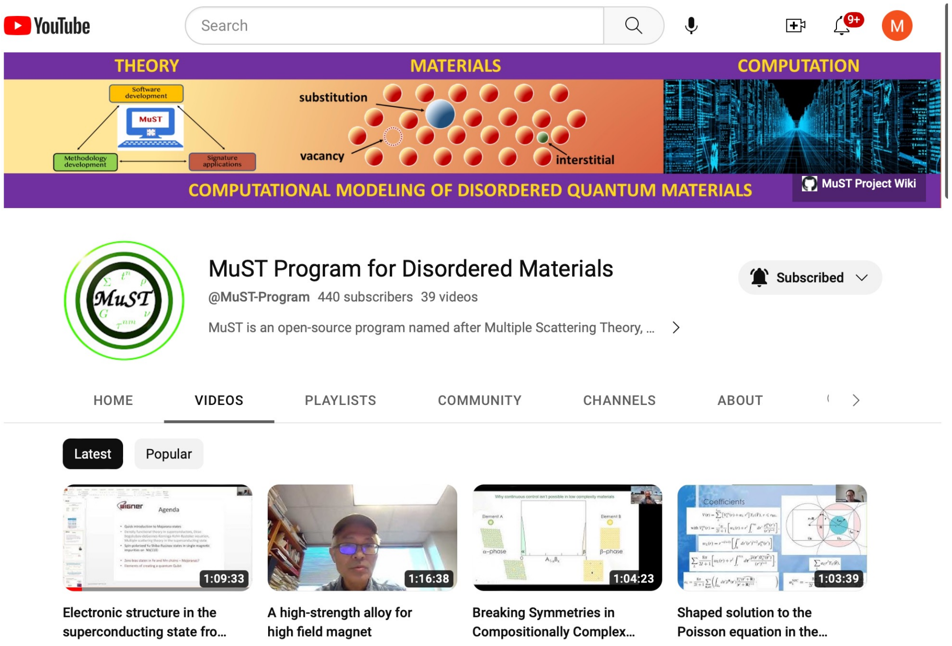
Our multiple scattering codes are available at:

https://github.com/mstsuite

Resources



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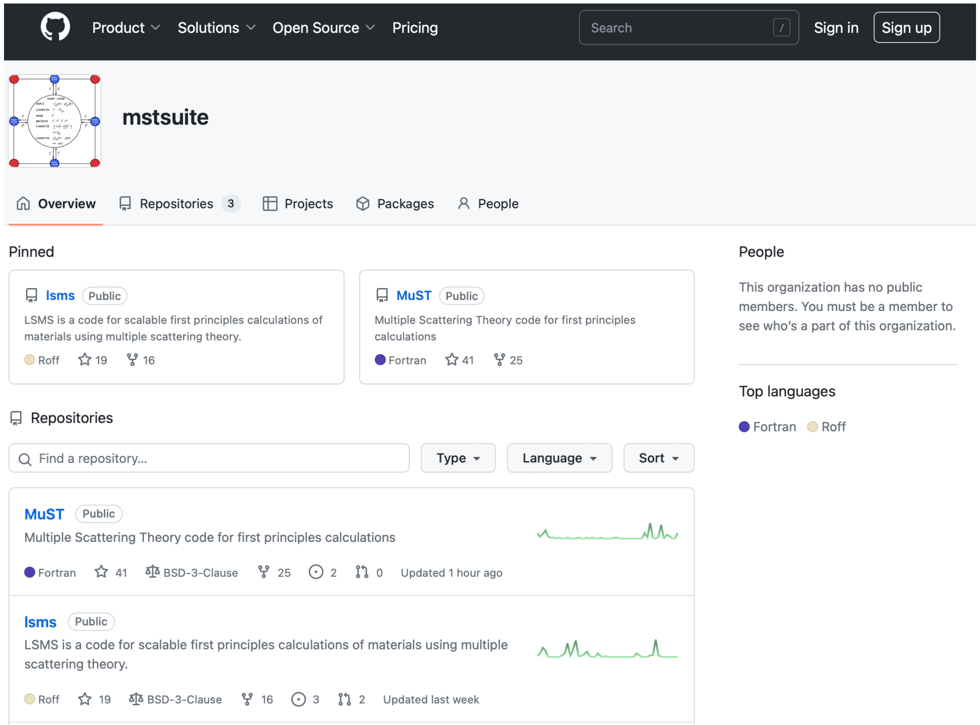

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