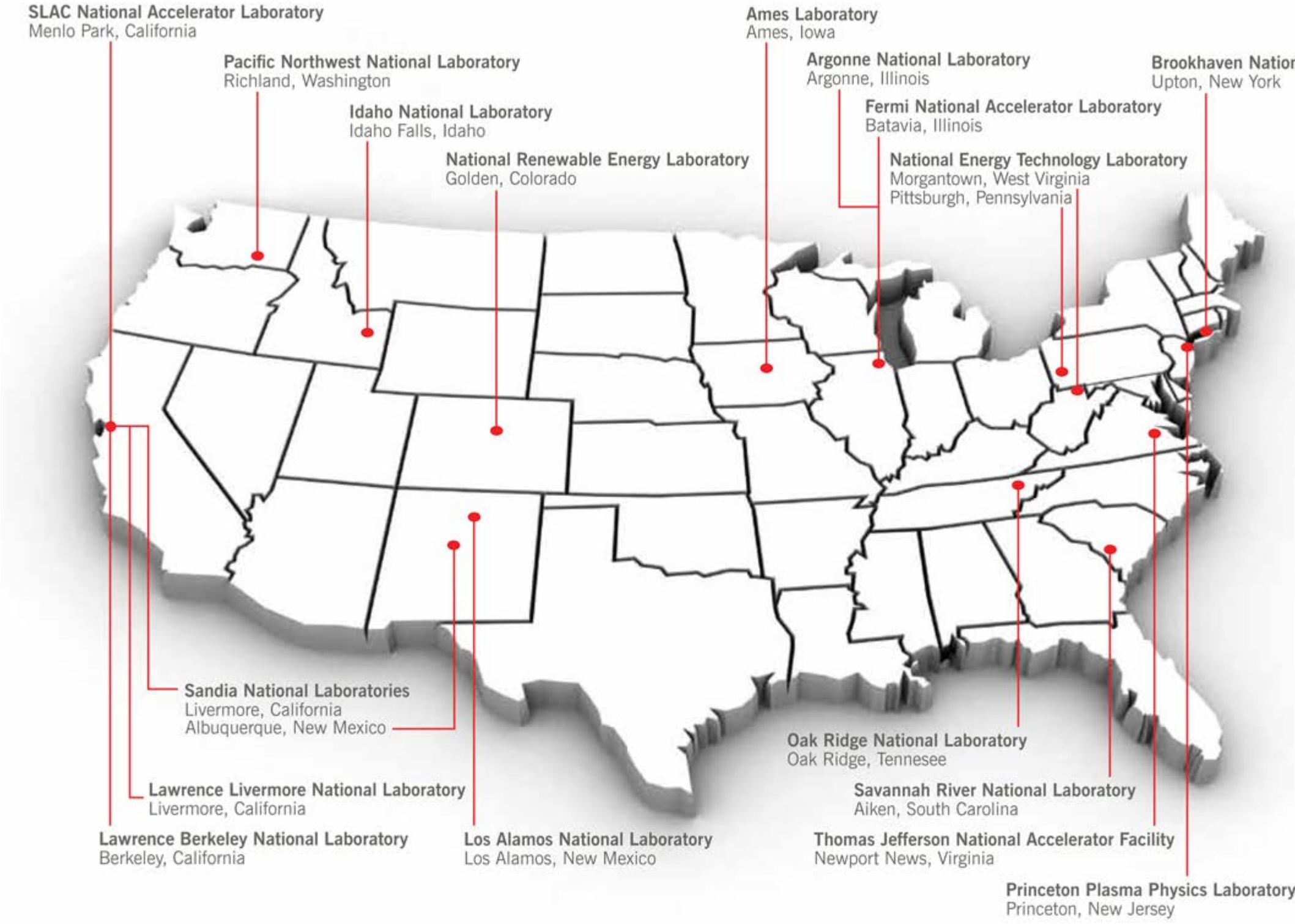
Adaptive Genetic Algorithm Method for Crystal Structure Prediction and Materials Discovery

C. Z. Wang
Ames Laboratory – USDOE, Iowa State University, Ames, Iowa 50011

In collaboration with K. M. Ho (Ames), M. Ji (Ames), S. Q. Wu (Ames & Xiamen Univ, China), M. C. Nguyen(Ames),K. Umemoto (Univ Minnesota & Ames), R. Wentzcovitch (Univ Minnesota)

Work at Ames Lab was supported by U. S. DOE
Work at University of Minnesota was supported by NSF/EAR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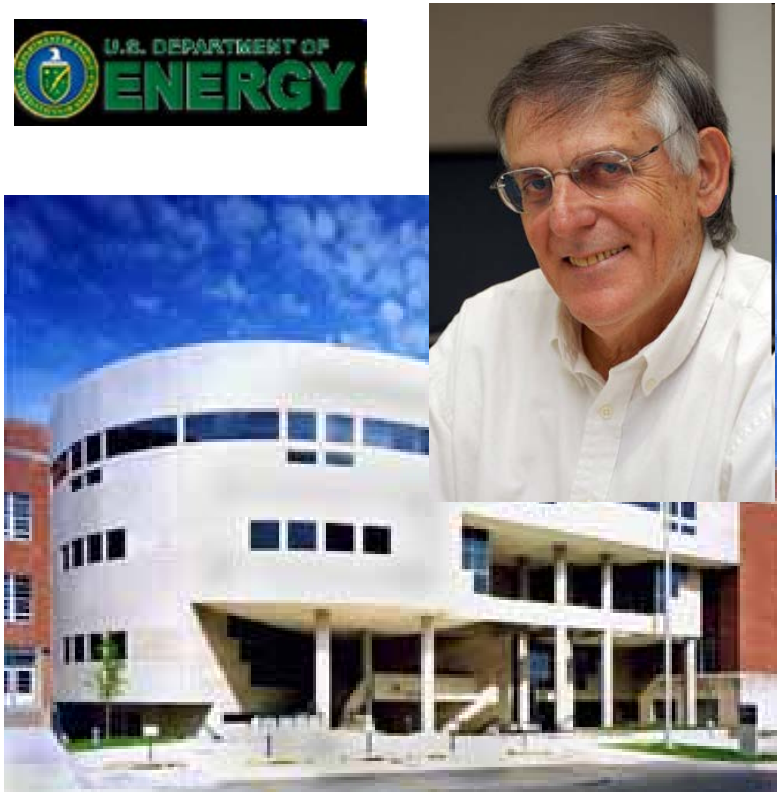

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The Nobel Foundation has named Dan Shechtman of the U.S. Department of Energy's Ames Laboratory, Iowa State University and Israel's Technion winner of the 2011 Nobel Prize in Chemistry.

Kai-Ming Ho received 2012 APS Aneesur Rahman Prize for Computational Physics

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- Applied Mathematics & Computational Sciences
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Current Research Project in our group

- Exploratory Development of Theoretical Methods
  - First-principles methods for strongly correlated electron systems
  - Tight-binding potentials and tight-binding molecular dynamics
  - Cluster alignment method for medium-range order in metallic glass
  - **Adaptive genetic algorithm for structure prediction and material discovery**
- Surface Structure Far-from-Equilibrium
  - Metal on graphene and semiconductor surfaces
- Structure and Dynamics of Condensed Systems
  - Short and medium-range orders in metallic liquids and glasses and their effects on phase selection and growth
- Beyond Rare Earth Permanent Magnets
  - Crystal structure prediction and phase diagram exploration

Postdoc position(s) available

Contact: wangcz@ameslab.gov

kmh@ameslab.gov

Outline

1. Why need adaptive GA?
2. How adaptive GA works?
3. Parallel adaptive GA on peta-scale computers

Motivation

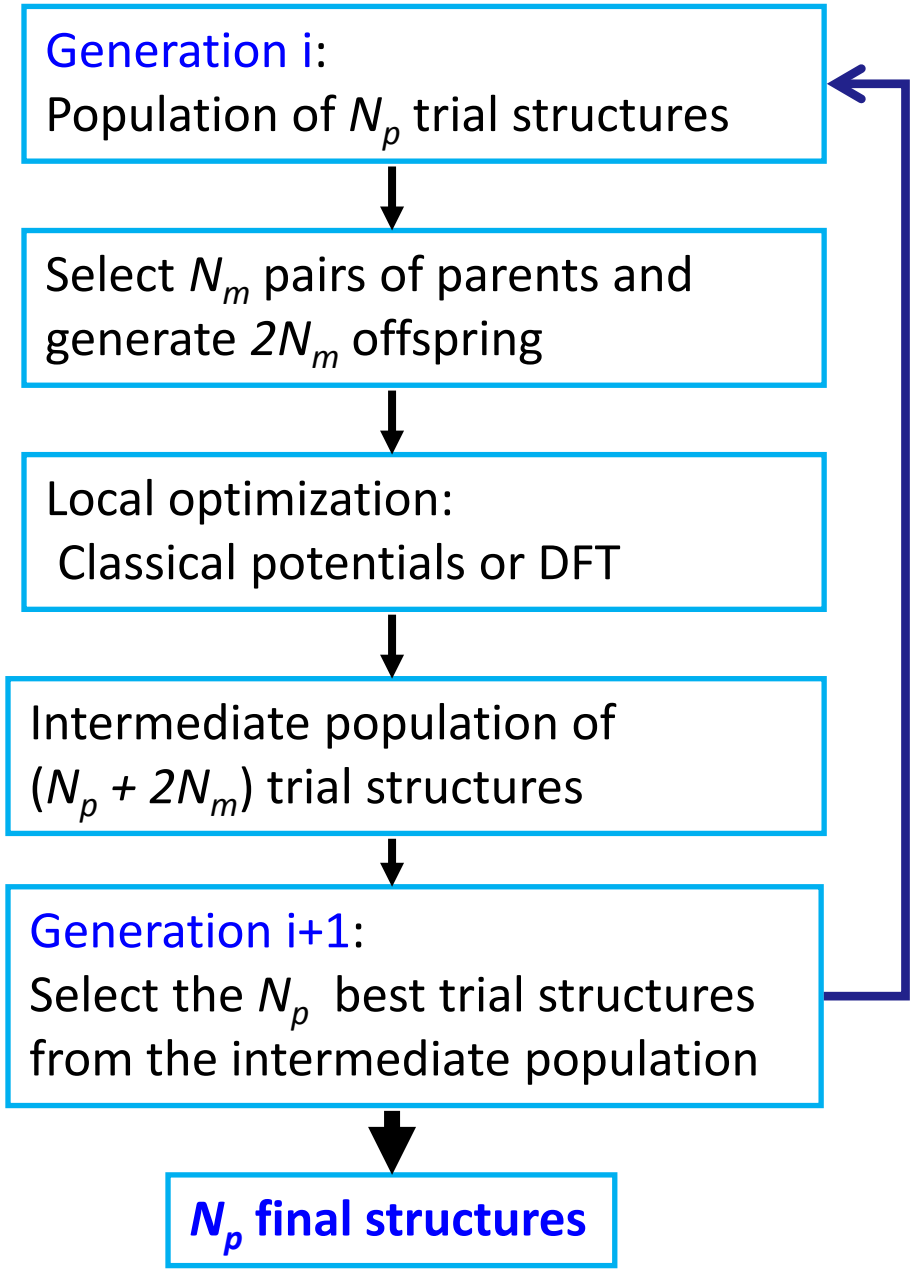
The urgent demand for new energy technologies has greatly exceeded the capabilities of today's materials and chemical processes

Accurate and fast theoretical structure/property determinations will complement the traditional experimental efforts in material design and accelerating the pace of technological advances.

- Predict the atomistic structures of materials for given chemical compositions

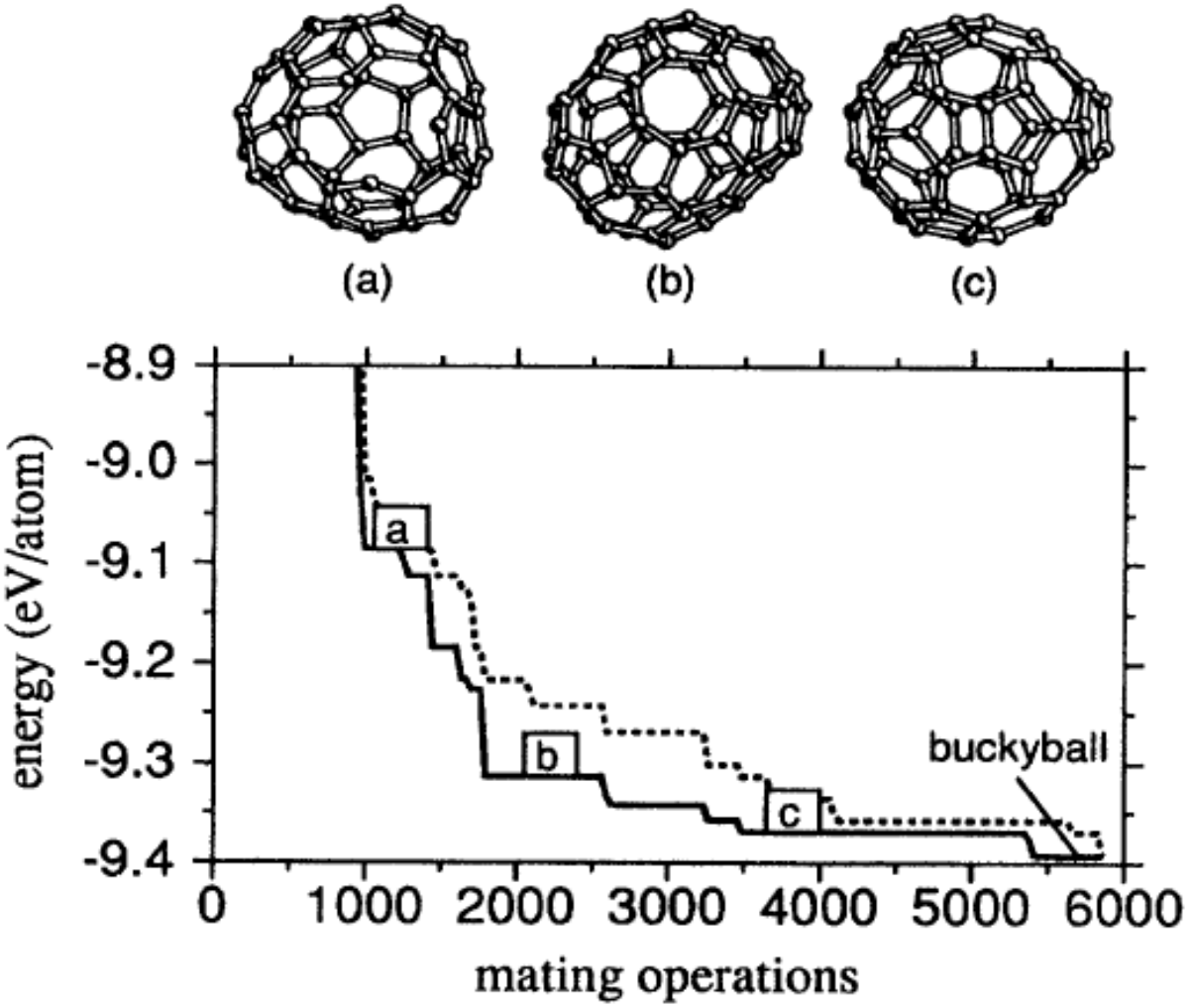
- Help new material discovery and synthesis to meet various needs in energy applications

Genetic Algorithm (GA)



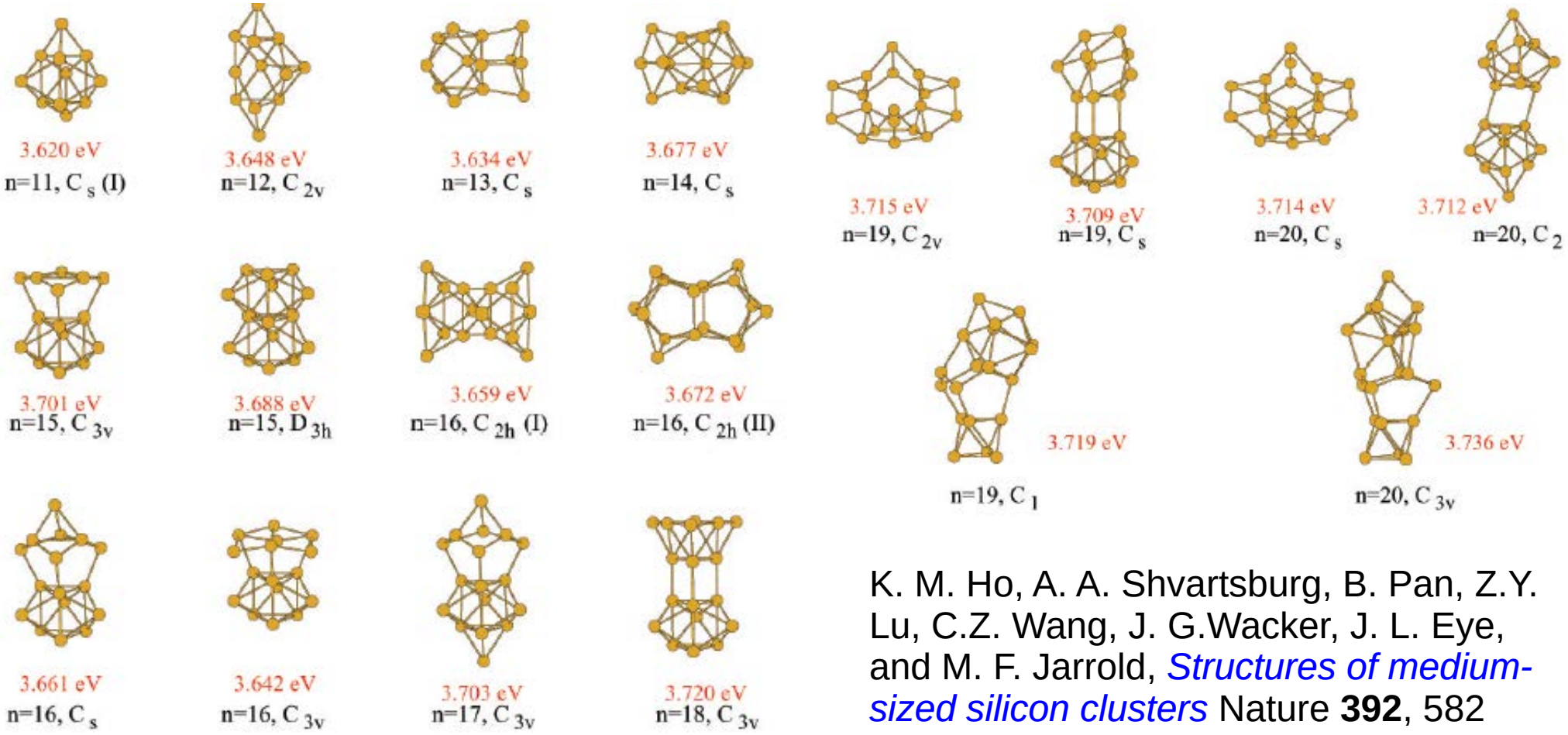
- GA is an optimization strategy inspired by the Darwinian evolution process. Here it is used for atomistic structure optimization
- GA approach has been applied to
  - Atomic clusters
  - Surface & Interfaces (grain boundaries)
  - Nanowires
  - Crystal structures (Catlow& Woodley, Aganov, Zunger, …)

GA/TBMD optimization of C_{60}



D. Deaven and K.M. Ho *Molecular Geometry Optimization with a Genetic Algorithm* PRL 75 288 (1995)

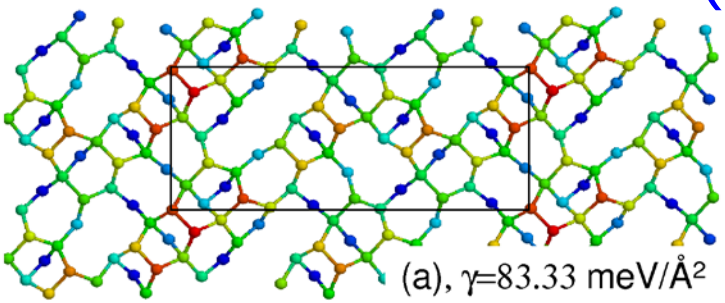
GA/TBMD optimization of Si clusters



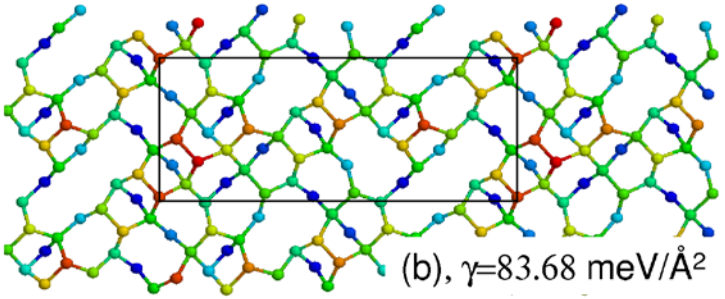
K. M. Ho, A. A. Shvartsburg, B. Pan, Z.Y. Lu, C.Z. Wang, J. G.Wacker, J. L. Eye, and M. F. Jarrold, *Structures of medium-sized silicon clusters* Nature **392**, 582 (1998)

Si(105) 2x2

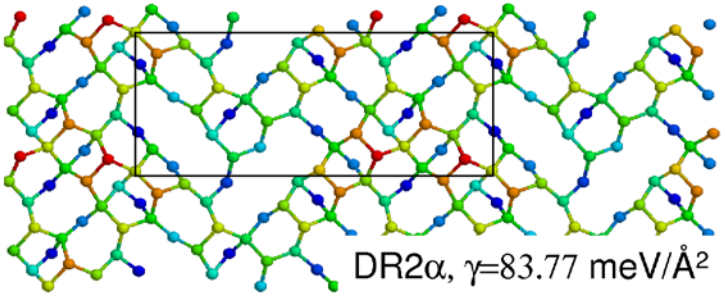
GA

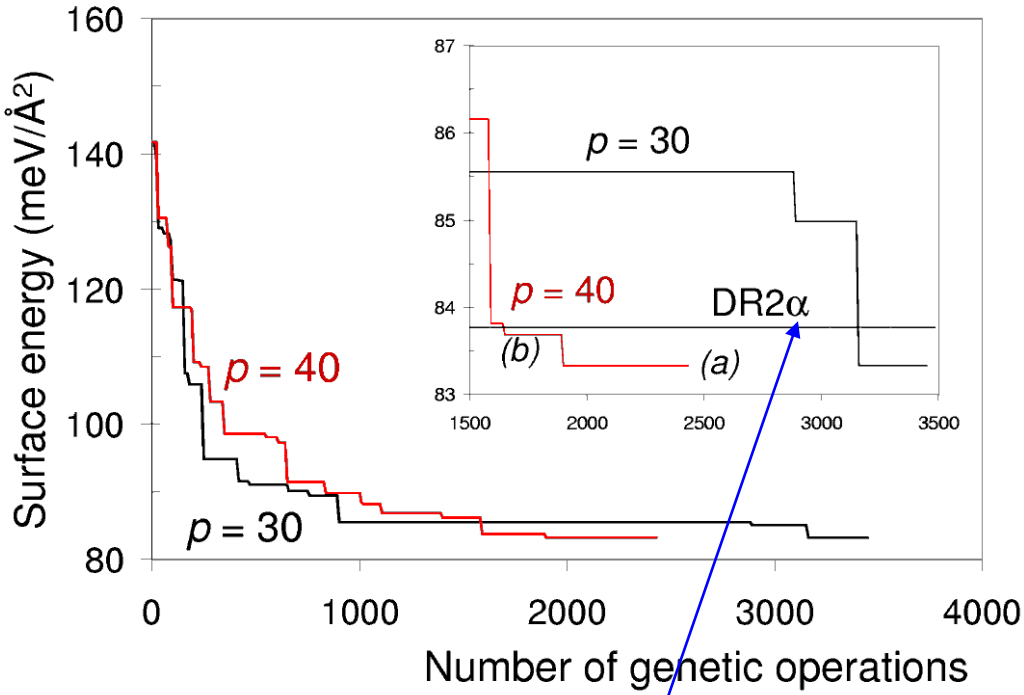


GA



PTMC





Lowest-energy structure obtained using PTMC Method

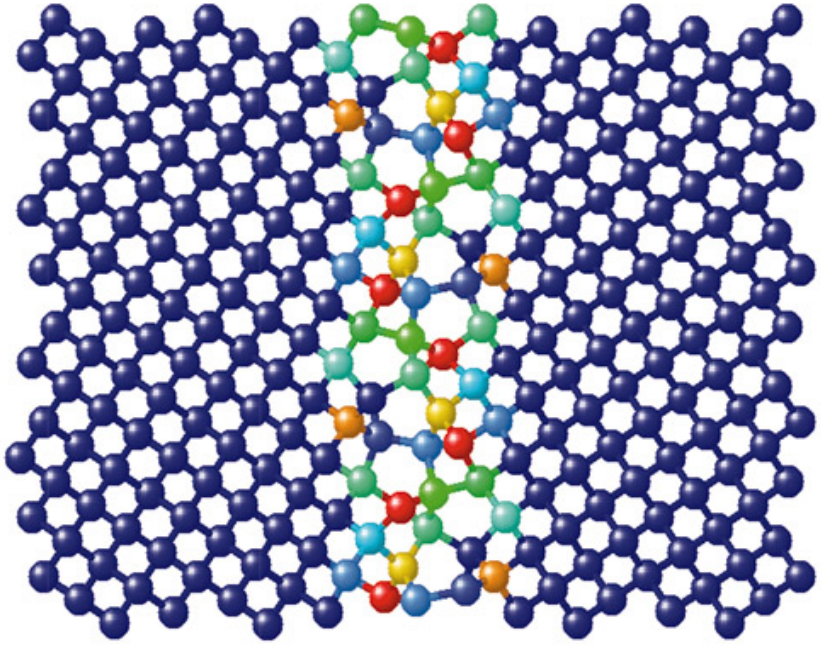
PTMC – Parallel Tempering Monte Carlo

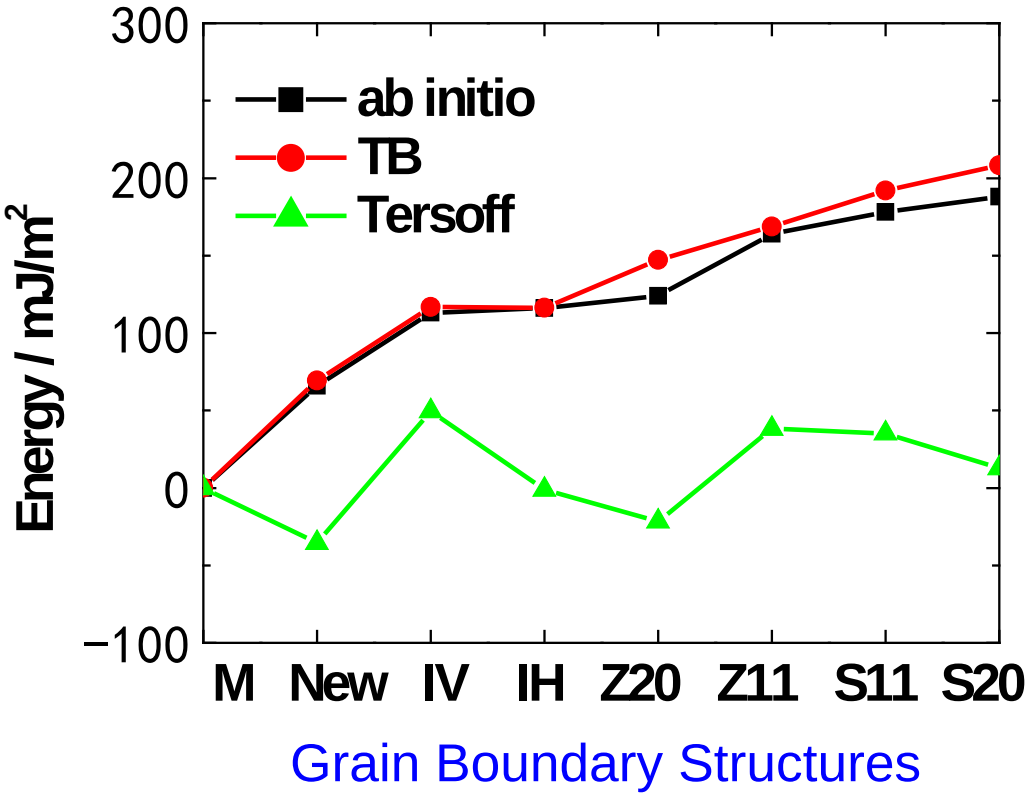
FC Chuang, CV. Ciobanu, V.B. Shenoy, CZ Wang and KM Ho *Finding the reconstructions of semiconductor surfaces via a genetic algorithm*, Surf. Sci.**573**, L375 (2004)

GA gets lower energy structures and faster than PTMC Method

GA for grain boundaries in Silicon

(510) Tilt boundary

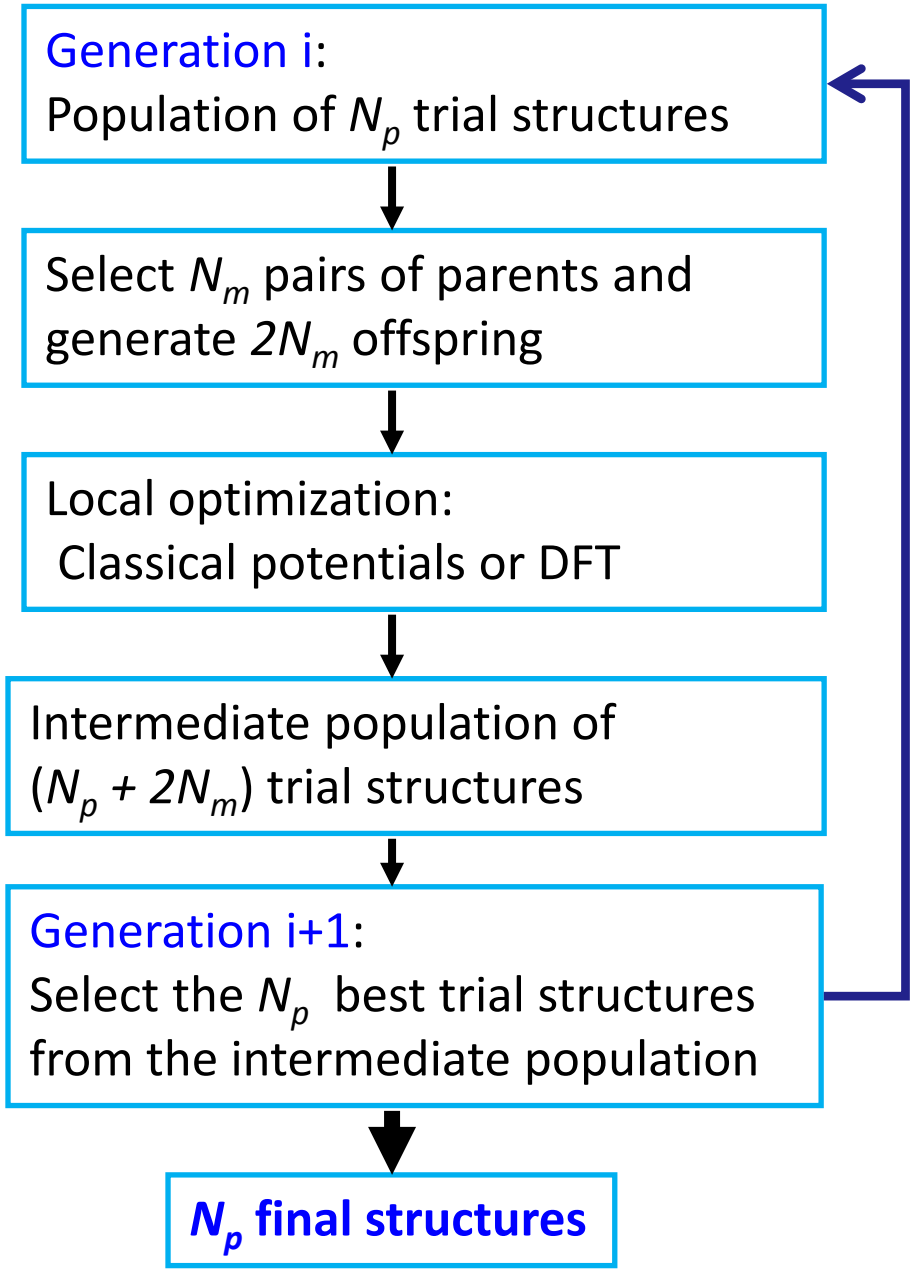




J. R. Morris, Z.-Y. Lu, D. M. Ring, J.-B. Xiang, K.-M. Ho, C. Z. Wang, and C.-L. Fu
Phys. Rev. B **58**, 11241-11245 (1998)

J. Zhang, C. Z. Wang and K. M. Ho, PRB, **80**, 174102 (2009)

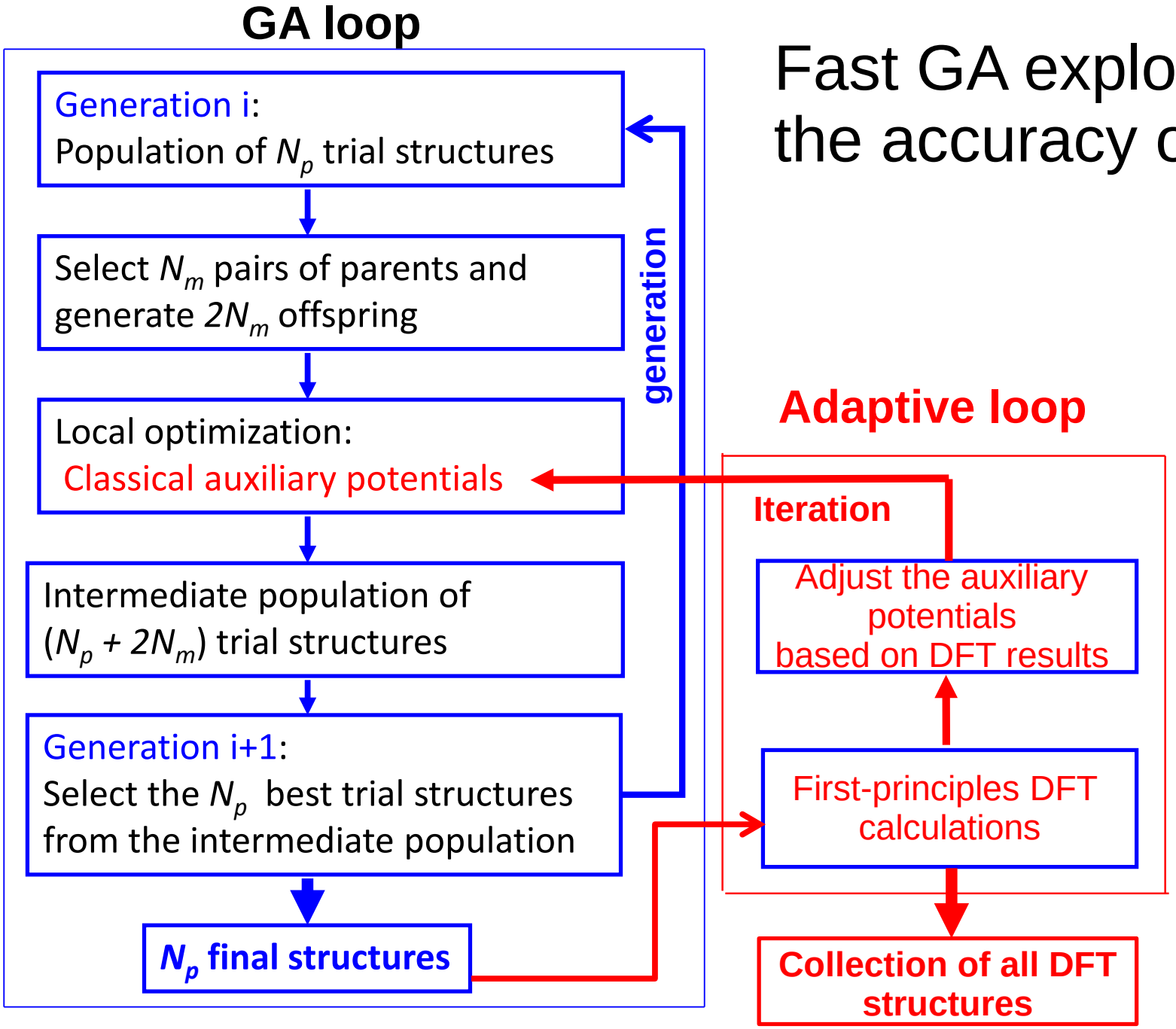
Genetic Algorithm (GA)



Challenges:

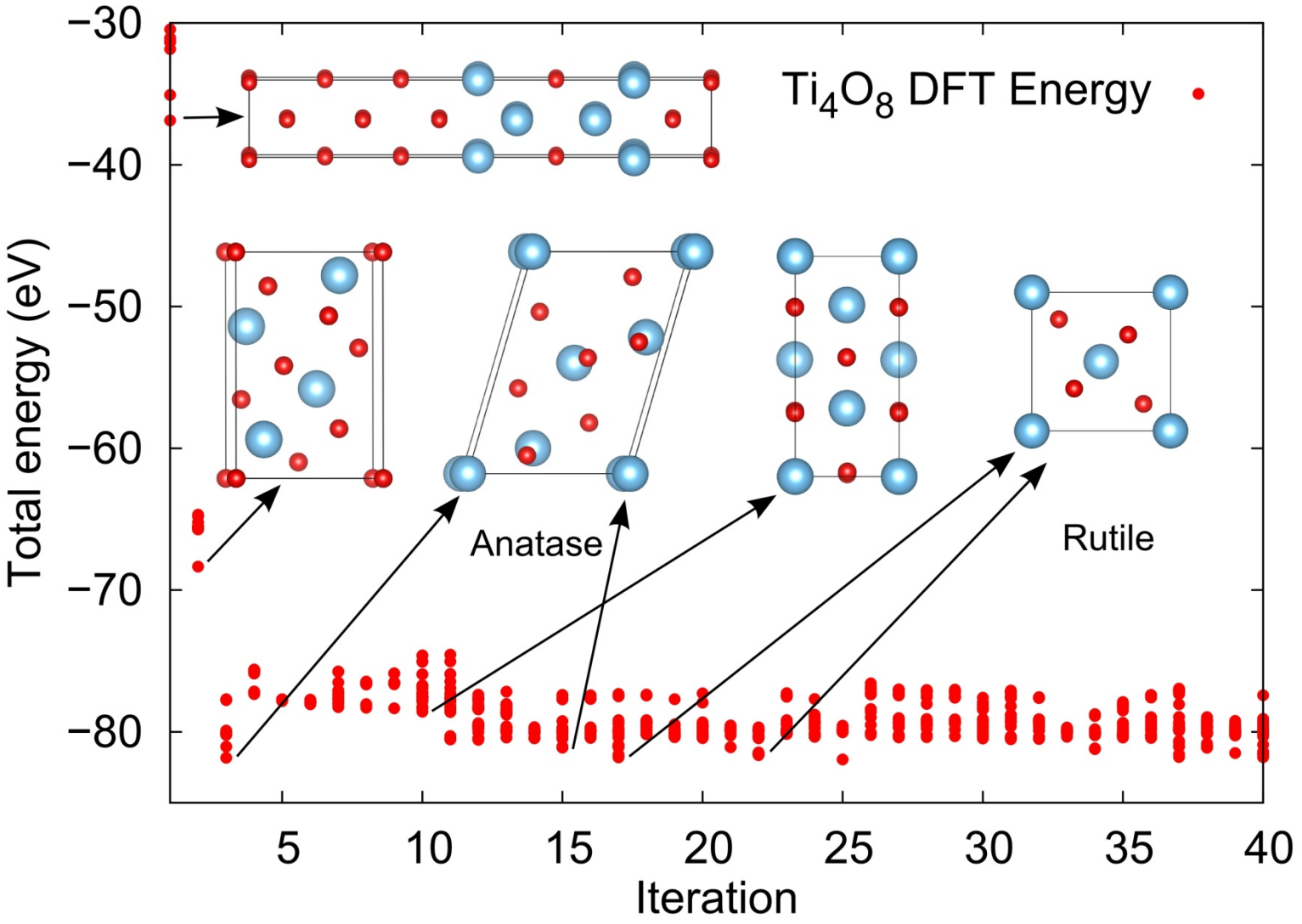
- Multi-component systems have complex structures and a huge configuration space
- *Ab initio* calculations are accurate but limited by computer power
- Classical potentials are fast but limited by availability and accuracy

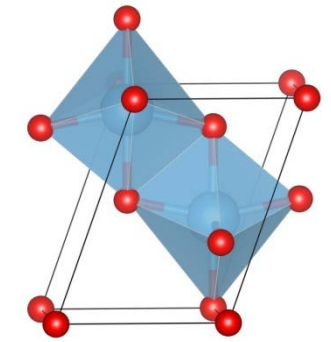
Adaptive Genetic Algorithm

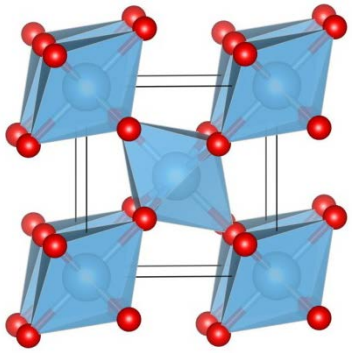


Fast GA explorations with the accuracy of DFT

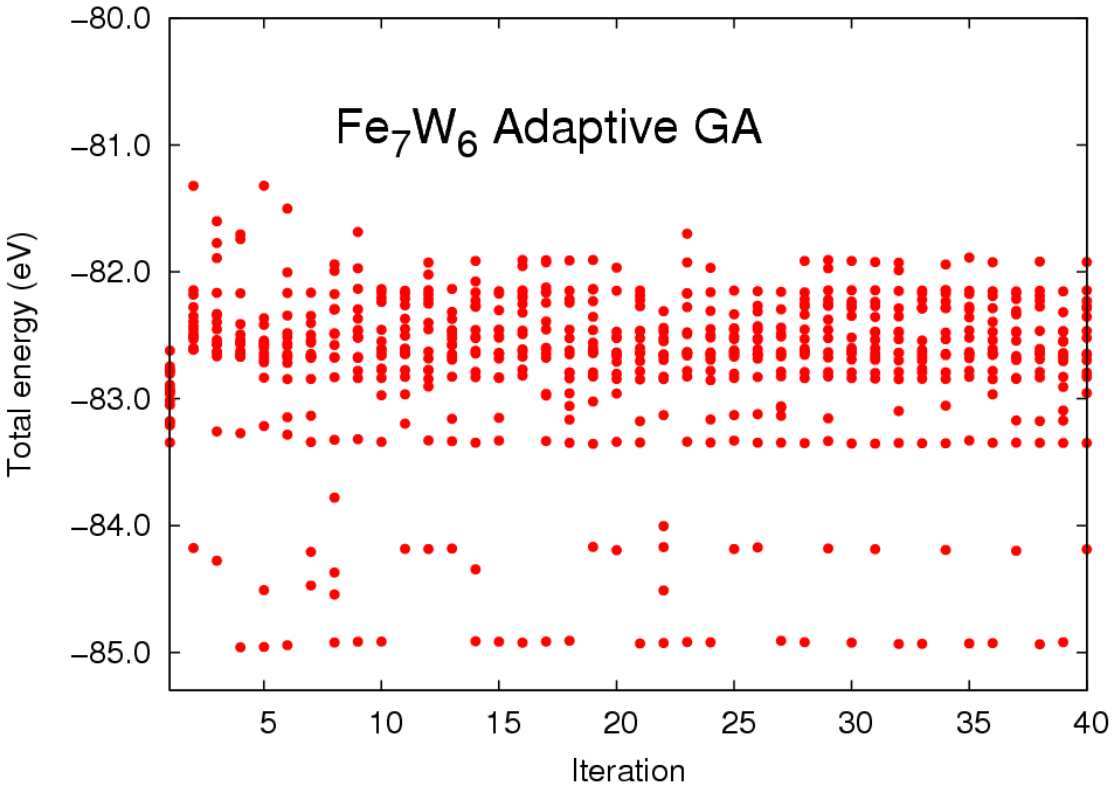
Prediction of TiO_2 crystal structures by adaptive GA using EAM auxiliary potentials and 12-atom supercell

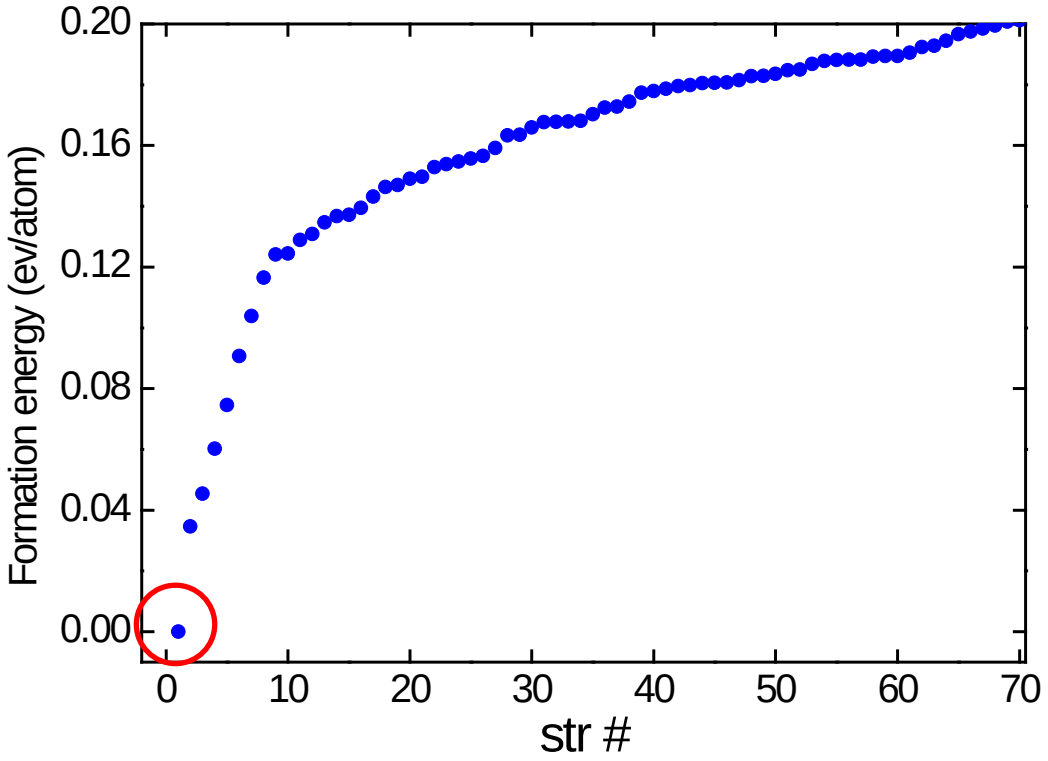






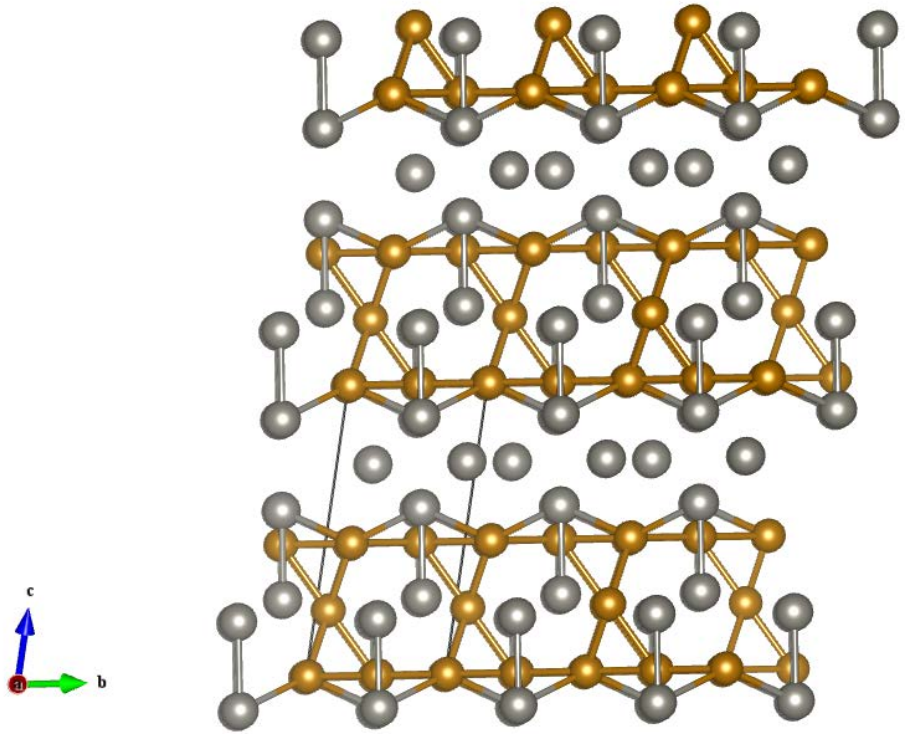
The crystal structures of TiO_2 can be predicted correctly using even EAM auxiliary potentials





Fe_7W_6: Adaptive GA search

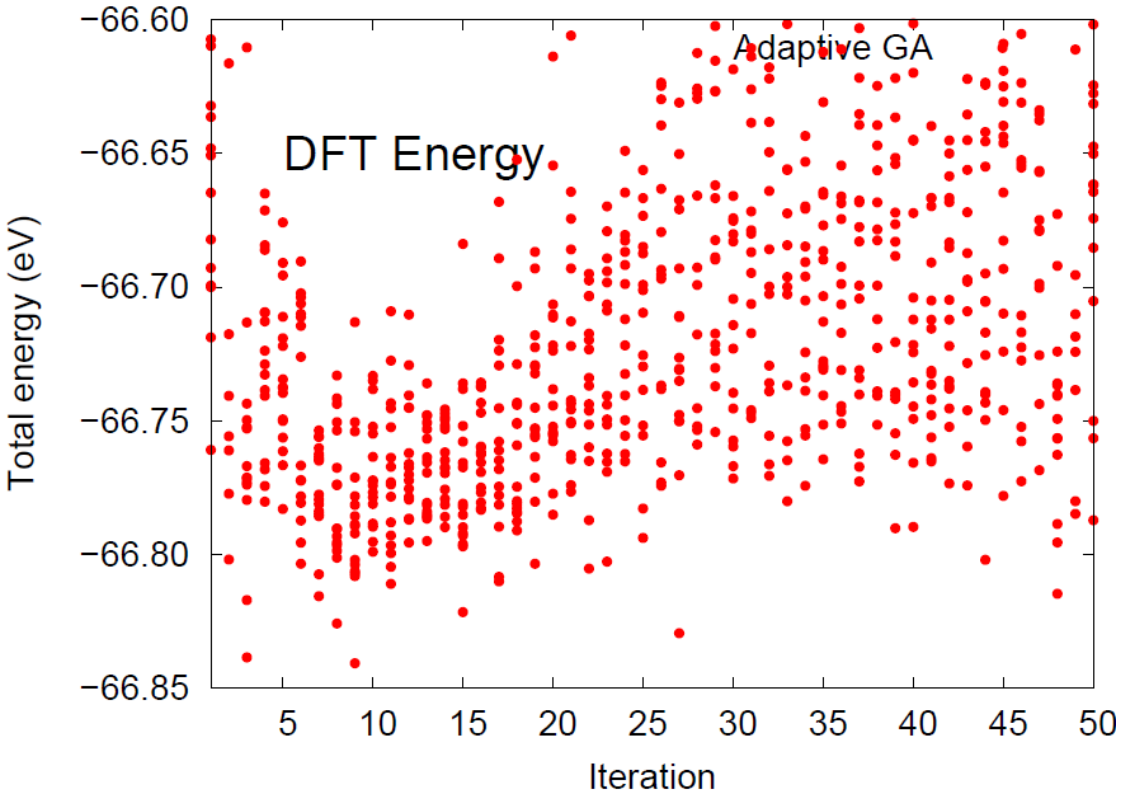
Cell size: 13 atoms and 26 atoms

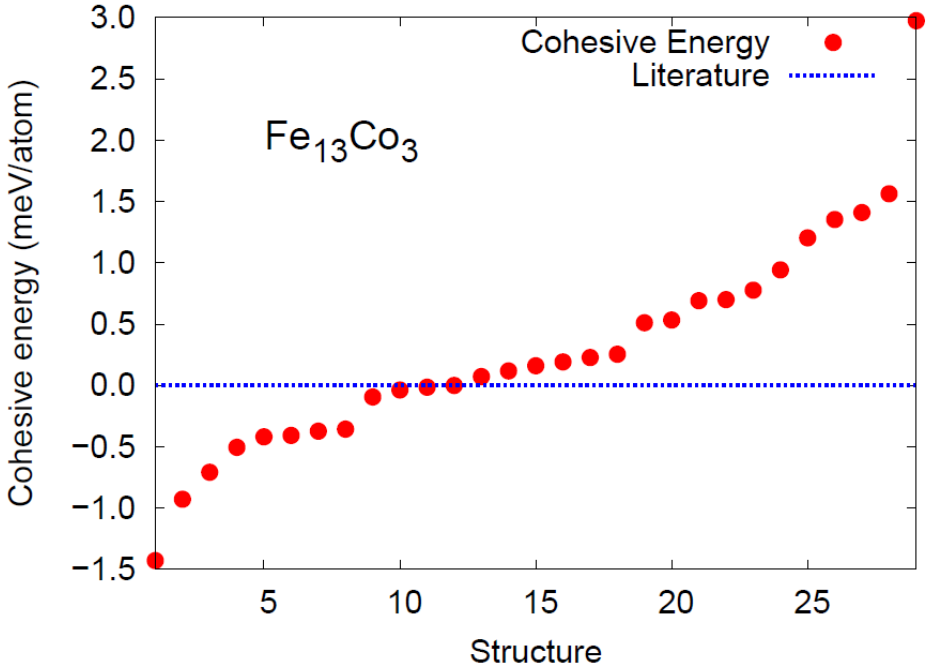


Structure of Fe_7W_6

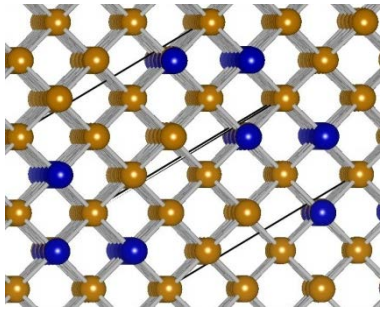
Space group (166), R3m

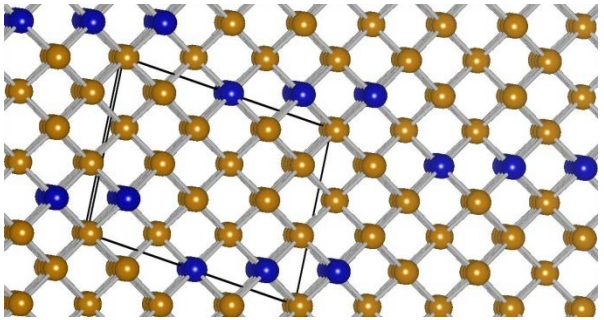
$Fe_{13}Co_3$: 16-32 atoms/cell; variable unit cell

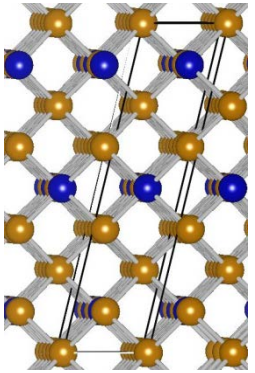




Lowest energy structures with energy window of 5 meV/atom.



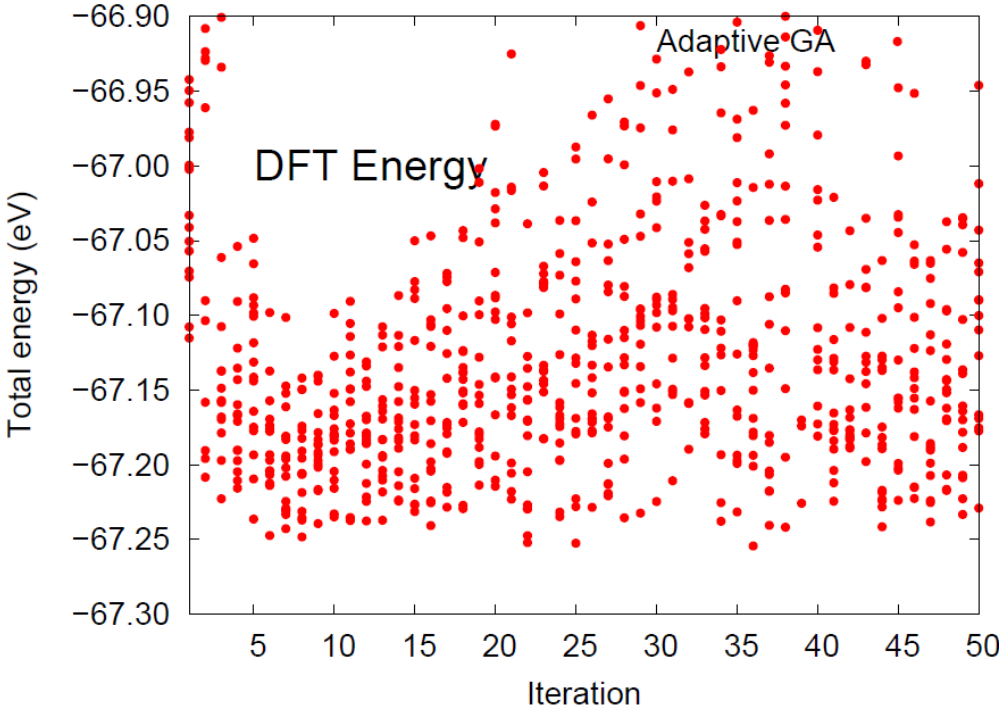


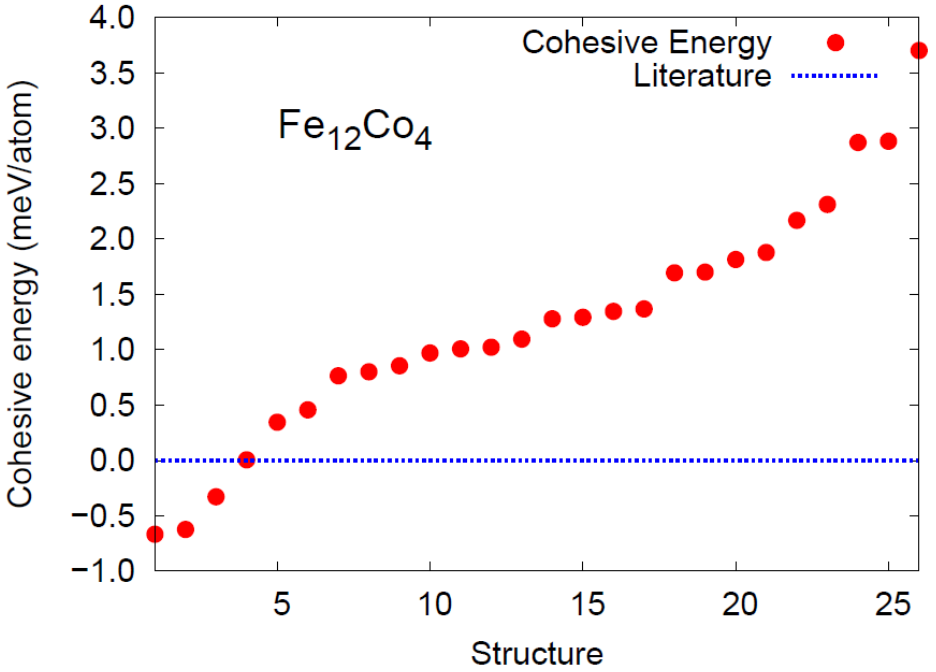


New low energy structures have been predicted using EAM auxiliary potentials

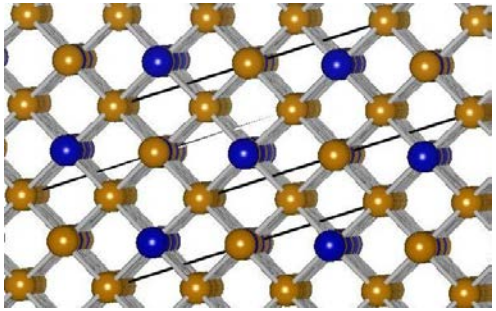
All low energy structures have BCC lattice

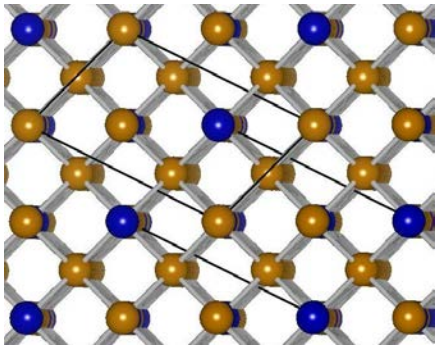
$Fe_{12}Co_4$: 16-32 atoms/cell

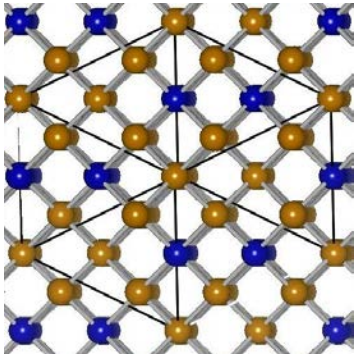


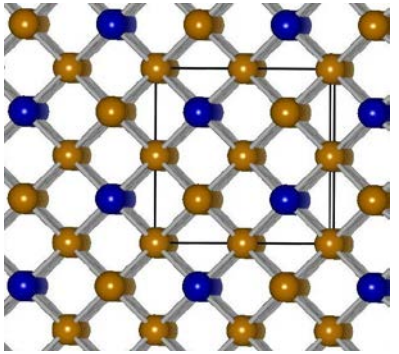


Lowest energy structures with energy window of 5 meV/atom.









New low energy structures have been predicted using EAM auxiliary potentials

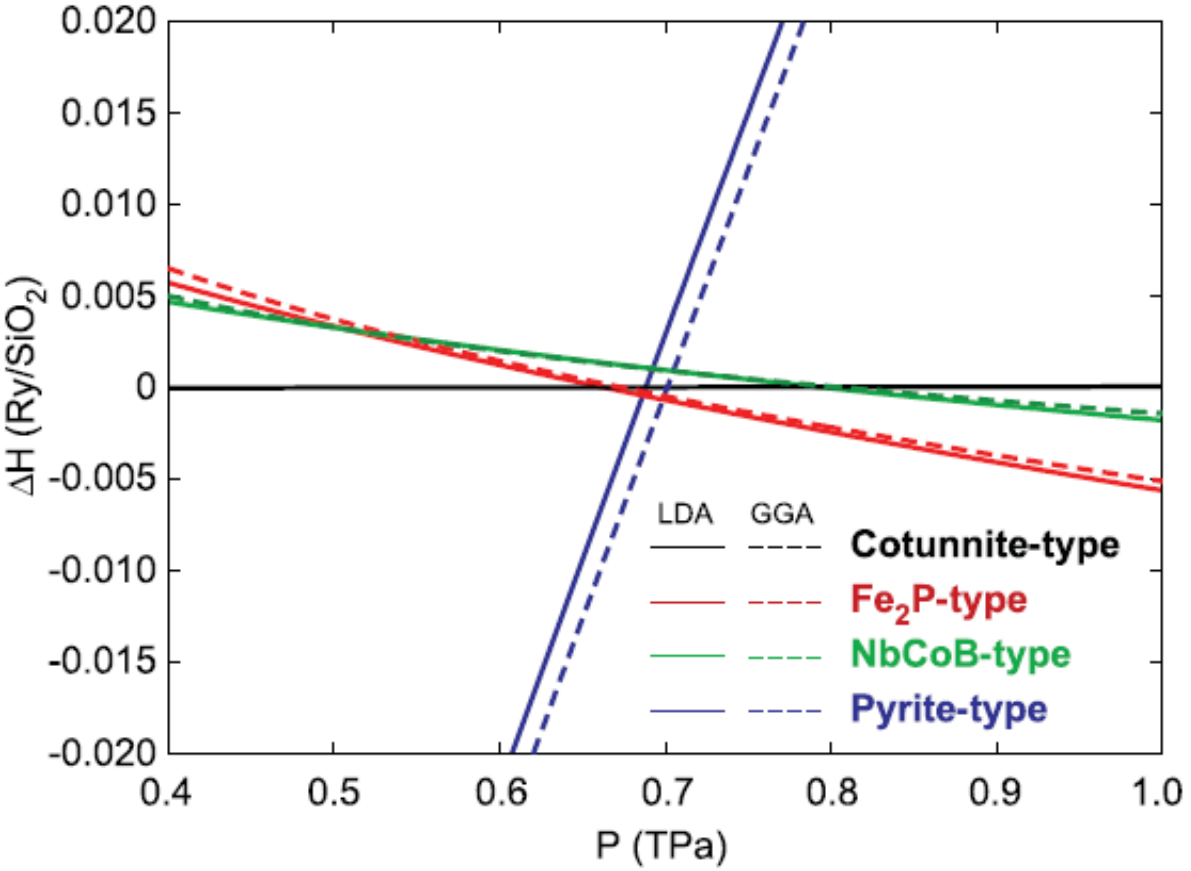
All low energy structures have BCC lattice

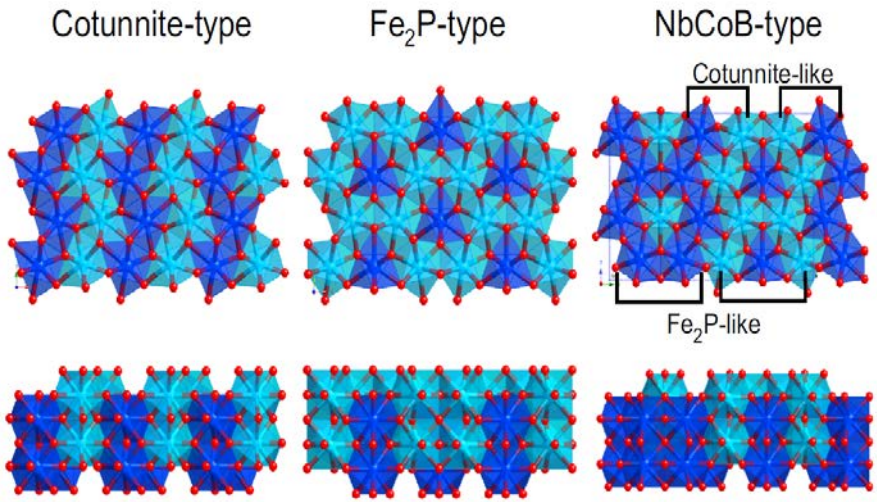
Crystal structures under ultra-high pressures

Super earth?

In collaboration with K. Umemoto and
R. Wentzcovitch ,Univ Minnesota

Identification of post-pyrite phase transitions in SiO_2 by genetic algorithm



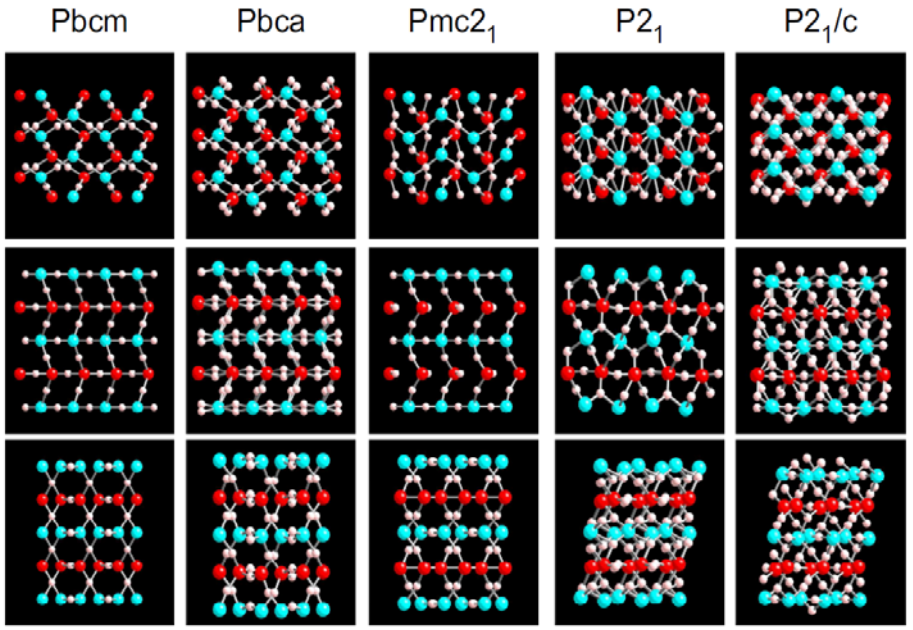


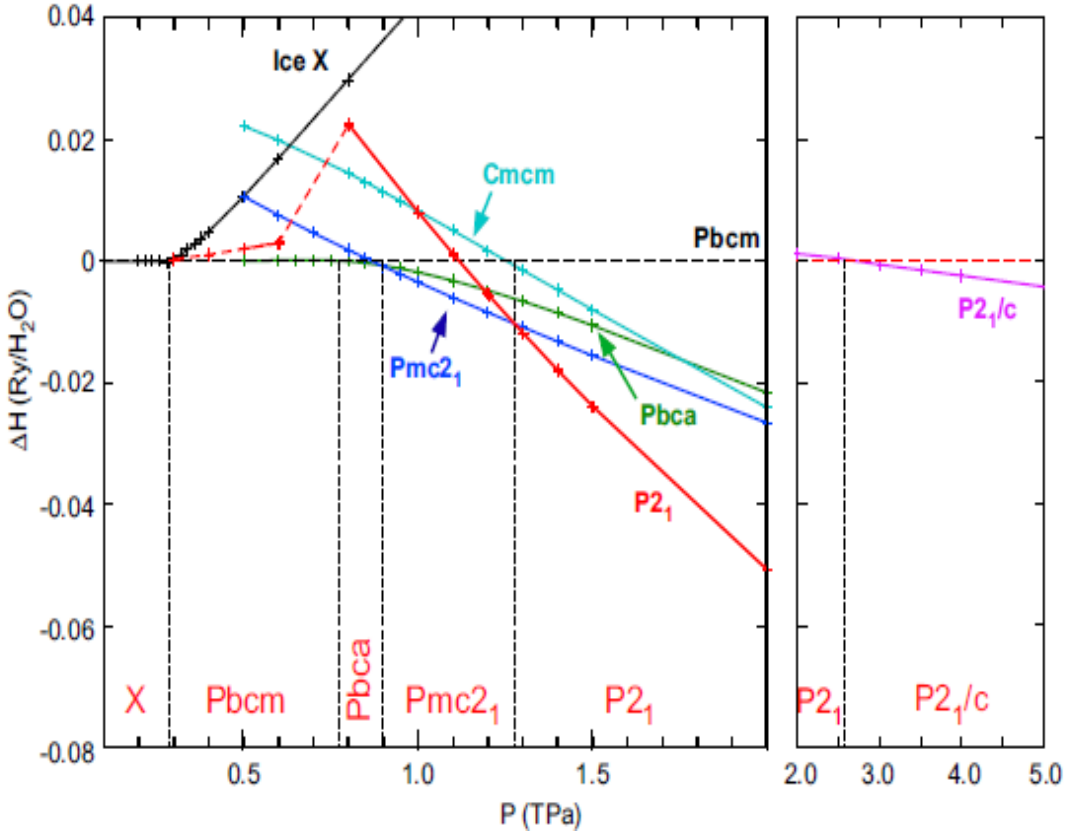
Using Lennard-Jones auxiliary potentials, we predict an Fe_2P phase is the first post-pyrite phase of SiO_2 at low temperatures

S. Q. Wu, K. Umemoto, M. Ji, C. Z. Wang, K. M. Ho and R. M. Wentzcovitch, Phys. Rev. B **83**, R184102(2011).

New ultrahigh pressure phases of H_2O-ice using an adaptive genetic algorithm

Min Ji, K. Umemoto, C. Z. Wang, K. M. Ho and R. M. Wentzcovitch, PRB, **84**, 220105(R), (2011).

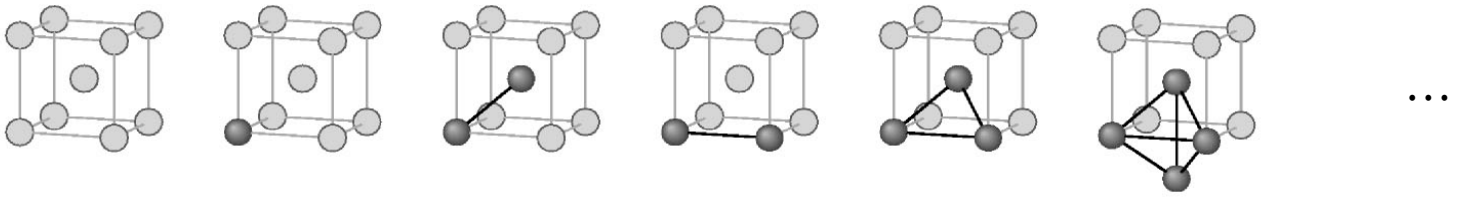




New ultrahigh pressure phases of H_2O-ice are predicted using Lennard-Jones auxiliary potentials

Cluster Expansion

Short-range CE: Figures

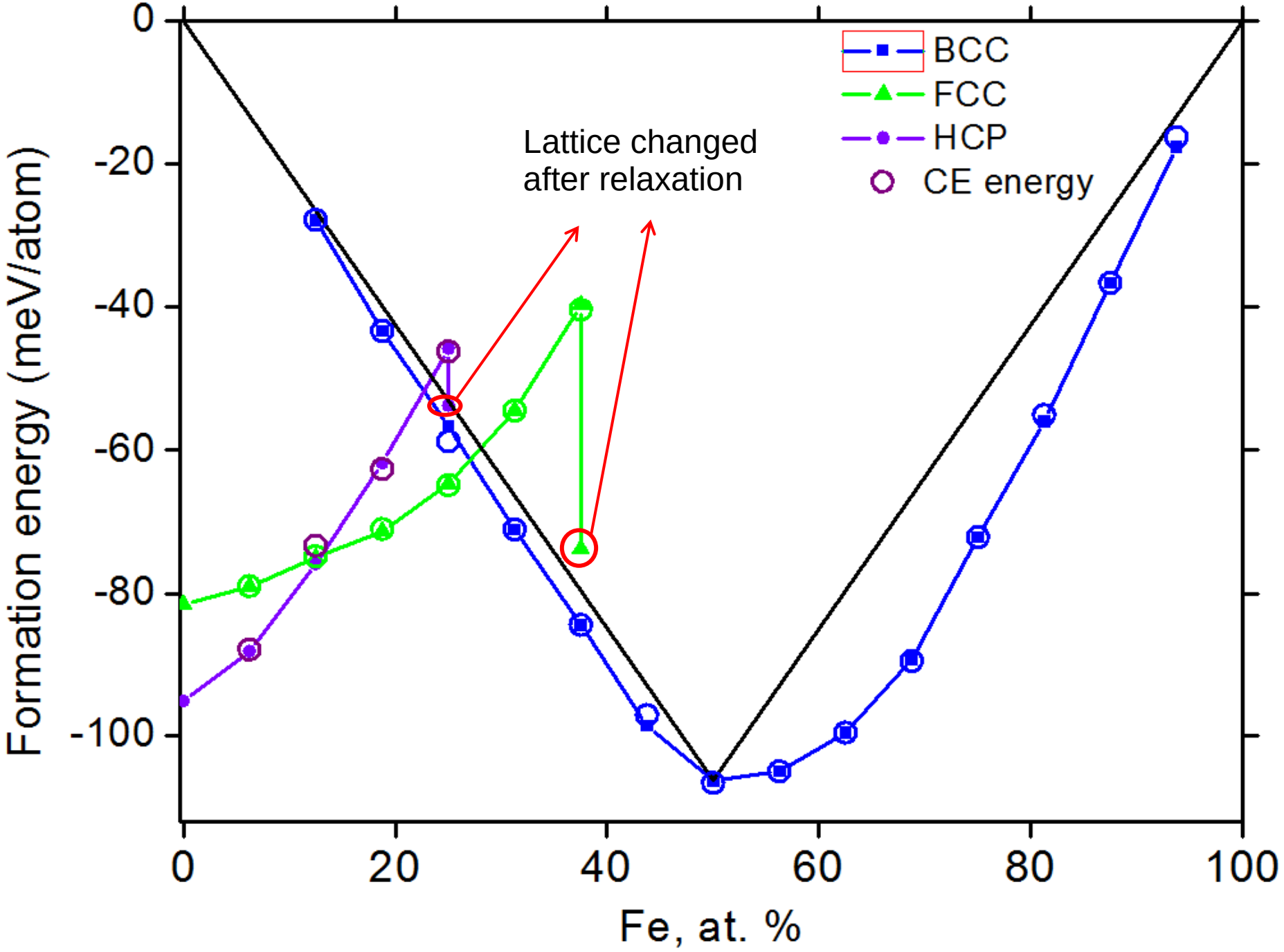


empty figure | point figure | smallest pair | 2nd smallest pair | smallest triangle | smallest tetrahedron | …

$$E_{\mathrm{CE}}(\sigma) = J_0 + \sum_{\text{sites}} J_i \hat{S}_i + \sum_{\text{pairs}} J_{ij} \hat{S}_i \hat{S}_j + \sum_{\text{triplets}} J_{ijk} \hat{S}_i \hat{S}_j \hat{S}_k + \cdots$$

- Where J is cluster interaction energy; S_i=-1 (+1) if lattice site i is occupied by A (B) for a binary system.
- This expansion is exact if all nonequivalent pairs and many-body interaction types (MBIT) on the lattice are taken into account.
- In practice, the method relies on there being only a finite number of non-negligible interactions.
- Fit to a small number of *ab initio* calculated energies for different configurations → J's → energy of new configuration can be easily predicted.

Results – lowest energy structures



Summary

- Adaptive GA provides a fast scheme for crystal structure prediction at the accuracy of DFT
- The performance of Adaptive GA is demonstrated: TiO_2, $Fe_{1-x}Co_x$, SiO_2, Ice, $MgSiO_3$
- Our adaptive GA code can scale well on massively parallel computers
- Combination with cluster expansion approach further extend the power of adaptive GA for material discovery

Gutzwiller density functional theory

- LDA is highly effective, but fails for systems with strong electron correlations
- In the Kohn-Sham approach, **kinetic energy functional** is introduced in reference to a **non-interacting** electron system
- We introduce new kinetic energy functional in reference to an **interacting** electron system with exact treatment of onsite kinetic energy and Coulomb repulsion while using the **Gutzwiller approximation** for interactions between localized and delocalized electrons
- New energy functional yields a set of self-consistent one-particle Schrödinger equations analogous to LDA
- Our scheme includes additional variational degrees of freedom corresponding to ***occupation of local electron configurations***
- Compare to other methods (*e.g., dynamical mean field theory (DMFT), LDA+U, LDA+DMFT, SIC-LDA, LDA+Gutzwiller*), our scheme is a first-principles **parameter-free** theory: no need for U parameter

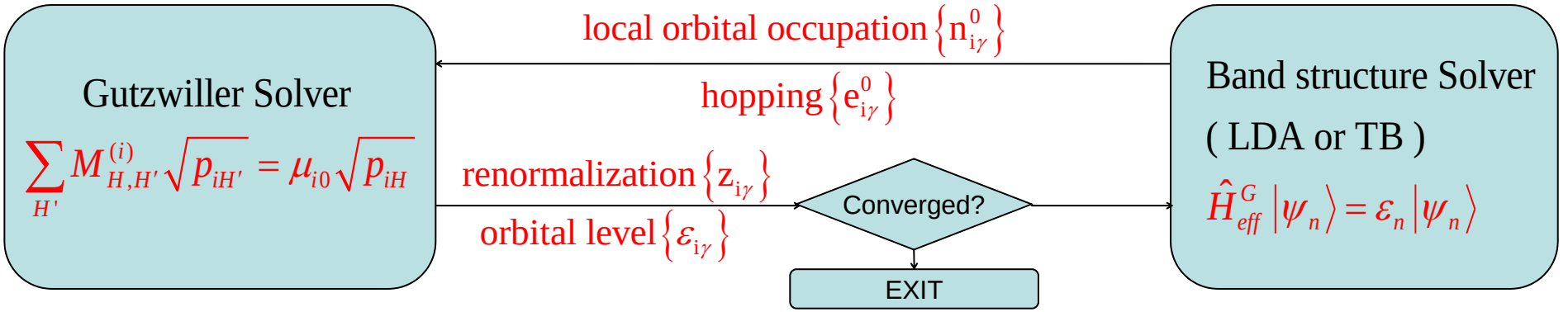
- ❑ First-principles density functional theory (DFT) incorporating electron correlations explicitly through Gutzwiller wave function (GWF)
- ❑ GWF reduces the weights of those energetically unfavorable many body configurations so the total energy of the system can be minimized

K. M. Ho, J. Schmalian, C. Z. Wang, PRB **77**, 073101 (2008)

Comparison between Kohn–Sham DFT (LDA) and Gutzwiller DFT

	LDA	Gutzwiller DFT
Reference system	Non-interacting electron system with Hartree wavefunction, $\|\Psi_0\rangle = \prod_{n=1}^{N} \psi_n(\vec{r}_n)$	Interacting electron system with Gutzwiller wavefunction $\|\Psi_G\rangle = \hat{G}\|\Psi_0\rangle,\ \ G = \prod_i \left(\sum_H g_{iH} \|H\rangle\langle H\| \right)$
Energy functional	$\langle \Psi_0 \| \hat{T} + V_{ion} \| \Psi_0 \rangle + E_H + E_{xc}^{LDA}$	$\langle \Psi_G \| \hat{T} + \hat{V}_{ion} \| \Psi_G \rangle + E_{Coul.} + \tilde{E}_{xc}$
Kinetic energy	$\langle \Psi_0 \| \hat{T} \| \Psi_0 \rangle$	$\langle \Psi_G \| \hat{T} \| \Psi_G \rangle$ with exact local onsite *K.E.*
Coulomb energy	$E_H = \frac{1}{2}\iint \rho v \rho$	$E_{Coul.} = \frac{1}{2}\iint \rho v \rho + \langle \Psi_G \| \hat{U}_s \| \Psi_G \rangle - \frac{1}{2}\iint \rho_l v \rho_l$
Ex.–corr. energy	$E_{xc}^{LDA} = \int \rho \varepsilon_{xc}^{LDA}(\rho)$	$\tilde{E}_{xc} = \int \rho \varepsilon_{xc}^{LDA}(\rho) - \int \rho_l \varepsilon_{xc,l}(\rho, \rho_l)$
Variational parameters	$\|\Psi_0\rangle$	$\|\Psi_0\rangle$, and local configuration (H) probabilities $\{p_{iH}\}$
Self–consistent equations	$\hat{H}_{eff}^{LDA} \|\psi_n\rangle = \varepsilon_n \|\psi_n\rangle$ $\hat{H}_{eff}^{LDA} = \hat{T} + V_{ion} + V_H + \hat{\mu}_{xc}^{LDA}$	$\hat{H}_{eff}^{G} \|\psi_n\rangle = \varepsilon_n \|\psi_n\rangle;\ \ \sum_{H'} M_{H,H'}^{(i)} \sqrt{p_{iH'}} = \mu_{i0} \sqrt{p_{iH}}$ $\hat{H}_{eff}^{G} = \tilde{\tilde{T}} + V_{ion} + V_H + \tilde{\tilde{\mu}}_{xc}^{LDA} - P_l \left(V_H^l + \mu_{xc}^l \right) P_l + \sum_{i\gamma} \varepsilon_{i\gamma} P_{i\gamma}$ $M_{H,H'}^{(i)} = \sum_{\gamma} \frac{e_{i\gamma}}{2\sqrt{n_{i\gamma}(1-n_{i\gamma})}} \left(\left\| \langle H \| h_{i\gamma}^{+} \| H' \rangle \right\|^2 + \left\| \langle H' \| h_{i\gamma}^{+} \| H \rangle \right\|^2 \right)$ $+ \delta_{H,H'} \left(E_{iH} - \sum_{\gamma \in H} \mu_{i\gamma} \right)$

Code structure



Thank You