Materials design using smooth interpolation in the periodic table

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Normal DFT calculation

- Input: Location and types of atoms
- Output: Energy, electron density, potential, ...

What we want

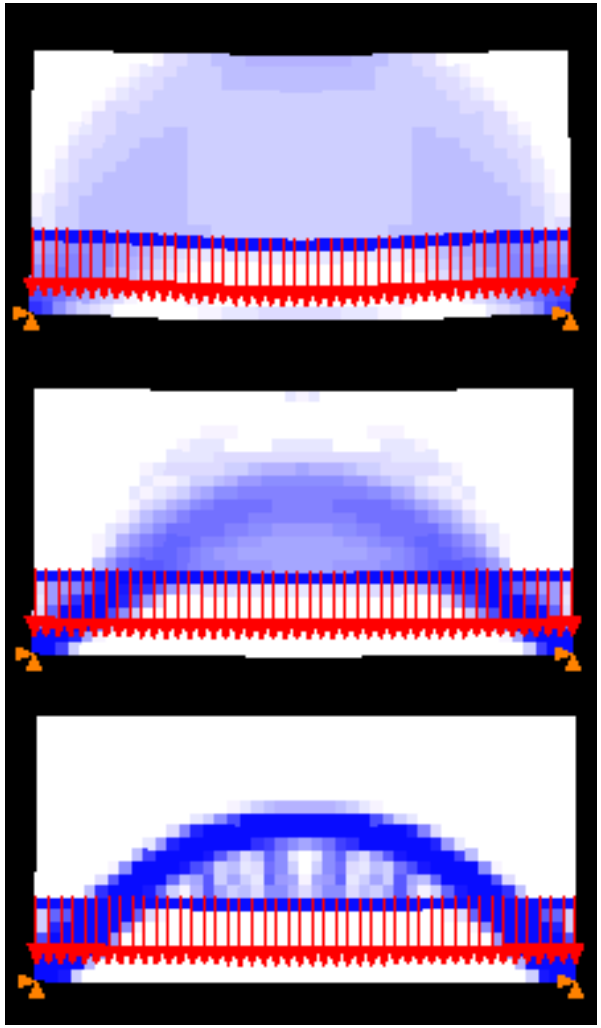
- Input: "[quantity] should be [optimal]"
- Output: Chemical composition

Method

- Formulate a model where an atom can vary smoothly between different elements
- Use an optimization algorithm to find the best chemical composition for a given problem by interpolating elements
- Optimize binding energies, band gaps (in the future), ...

Topology optimization

- Used in mechanical engineering
- Domain starts in an initial unphysical, mixed ("grey") state
- Grey areas are penalized during optimization, forcing system into definite state
- Gradient-based optimization – efficient even with large numbers of parameters



HGH pseudopotentials

In a calculation, each atom is represented by a pseudopotential given by various radial functions and coefficients:

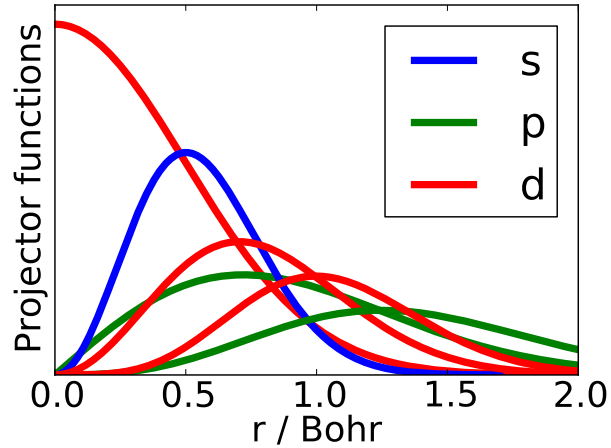


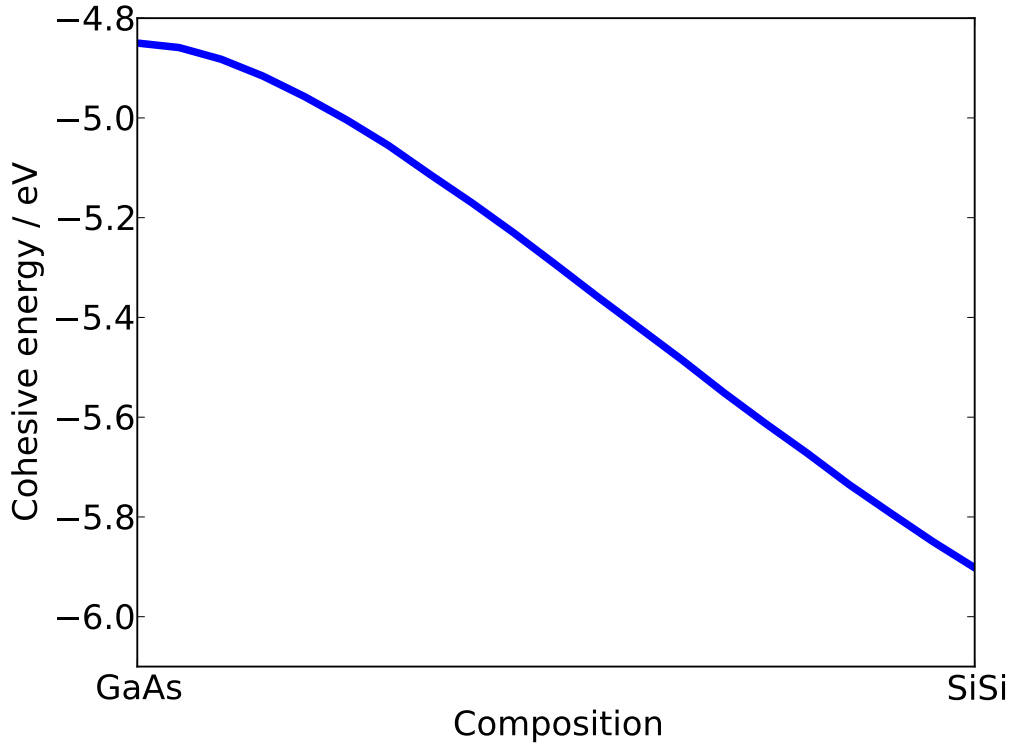
Figure: Projectors of Cr

$$\hat{v} = v_{\text{charge}}(r) + v_{\text{local}}(r) + \sum_{ij} |p_i\rangle h_{ij} \langle p_j|$$

Parameters

- Number of valence electrons Z
- Radius and shape of potentials, projectors
- Hamiltonian coefficients h_{ij} of projectors
- Around 10-15 parameters per element
- We can interpolate between elements by varying these

Interpolated cohesive energy of GaAs-SiSi



Left	Ga	As
Between	$(1-w)$ Ga $+w$ Si	$(1-w)$ As $+w$ Si
Right	Si	Si

Calculated using GPAW

Interpolation

Express an atom as a linear combination of elements, e.g.
$\mathrm{X} = w_1 \times \mathrm{Si} + w_2 \times \mathrm{As} + w_3 \times \mathrm{Pu}$

Derivatives

The energy E is then a function of the weights, whose derivatives $\frac{\partial E}{\partial w_i}$ can be evaluated by means of the Hellman-Feynman force theorem after a self-consistent DFT calculation

Optimization

Use the MMA (method of moving asymptotes), to optimize the weights w_i, finding the lowest energy.

Penalization

After finding the optimum, mixed atoms are penalized so that they converge to definite elements

Penalization of mixed configurations

After finding the optimum, apply one of the following to make the atoms converge to definite elements:

- Direct penalization of the objective – optimize:

$$E(w_1, ..., w_n) + \alpha \sum_i w_i(1 - w_i)$$

- Global constraint – optimize $E(w_1, ..., w_n)$ subject to:

$$g_M(w_1, ..., w_n) = \sum_i w_i(1 - w_i) - M < 0$$

An actual (simple) optimization problem

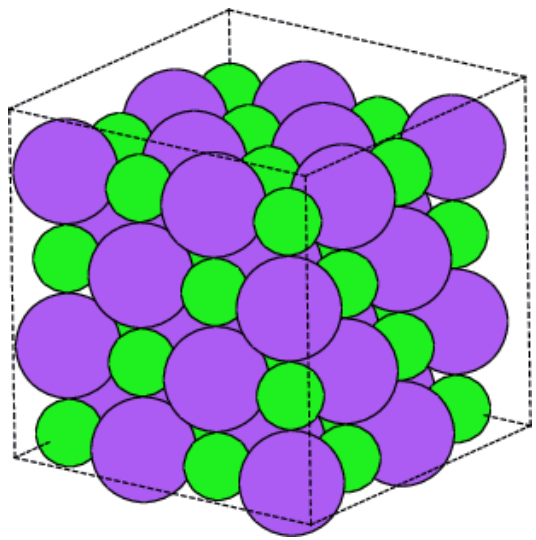
Consider $4 \times 4 \times 4$ atoms in a cubic lattice, each of which can be a combination of Na and Cl, specifically:

$$\alpha_i = (1 - w_i) \times \mathrm{Na} + w_i \times \mathrm{Cl}$$

We want to optimize the cohesive energy:

$$E^{\mathrm{crystal}}[w_1, ..., w_n] - \sum_i E^{\mathrm{atom}}[w_i]$$

Can we get Na and Cl right from a random starting guess?



64 atoms of Na or Cl

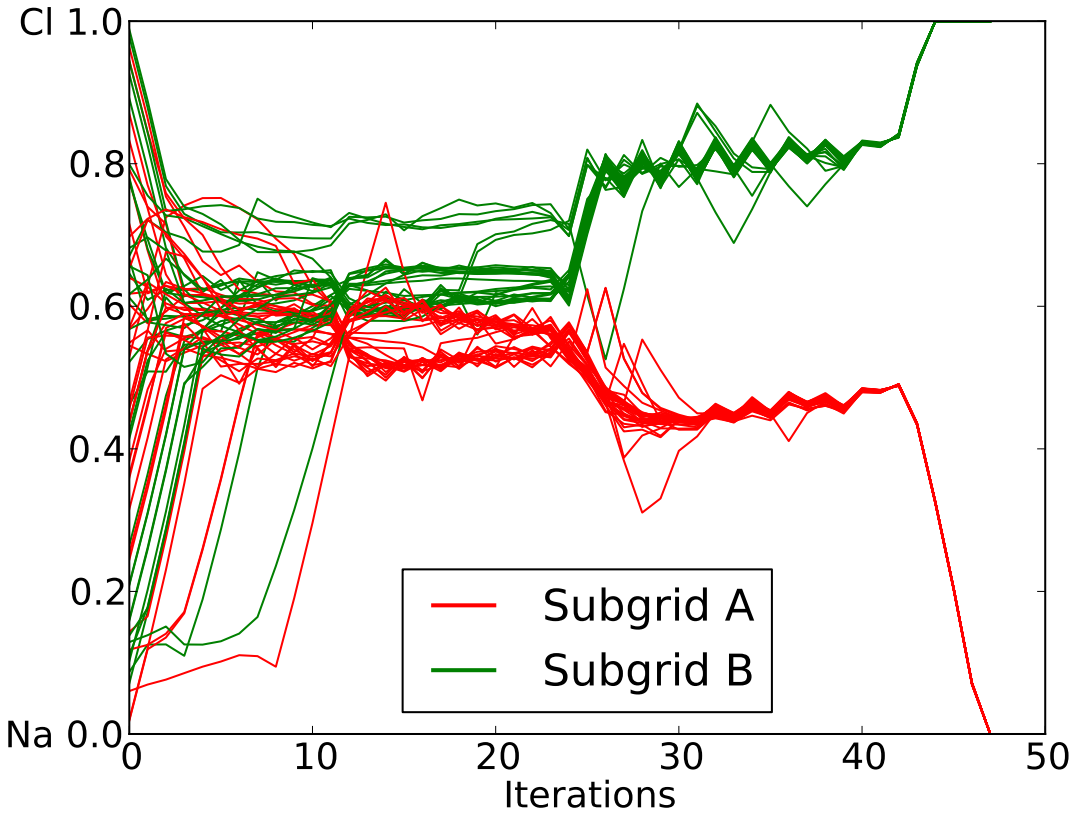


Figure: 64 NaCl, optimization with stepwise penalization.

A more complicated problem: Stability of FCC Ni_3Al

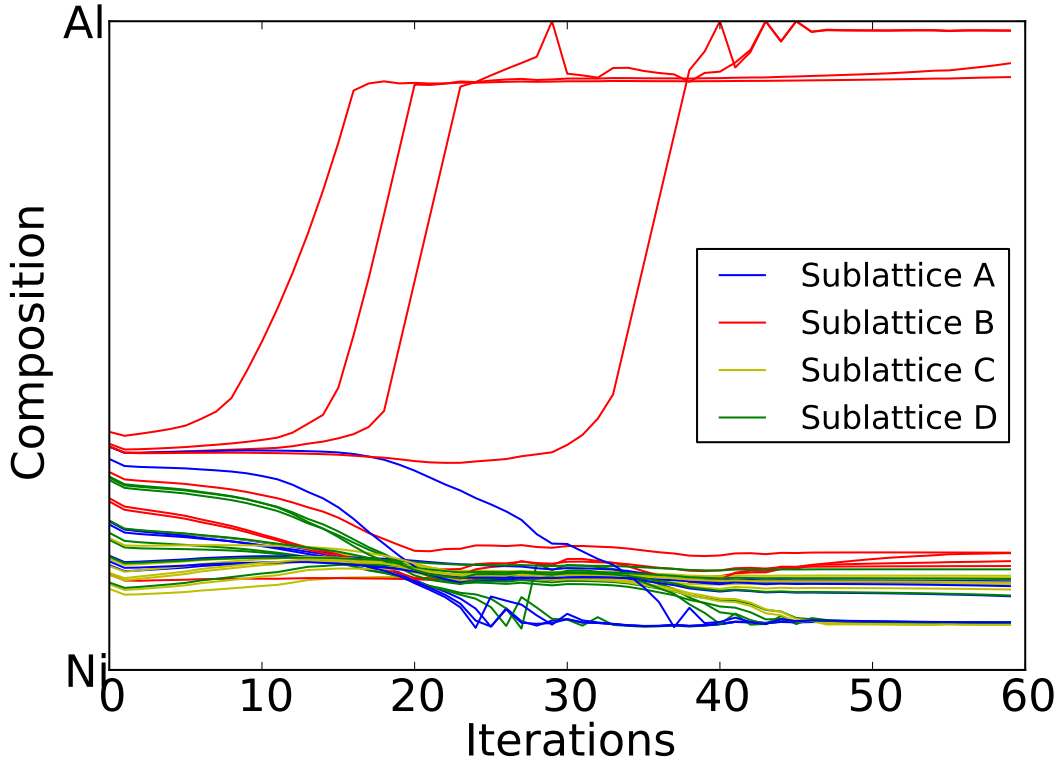
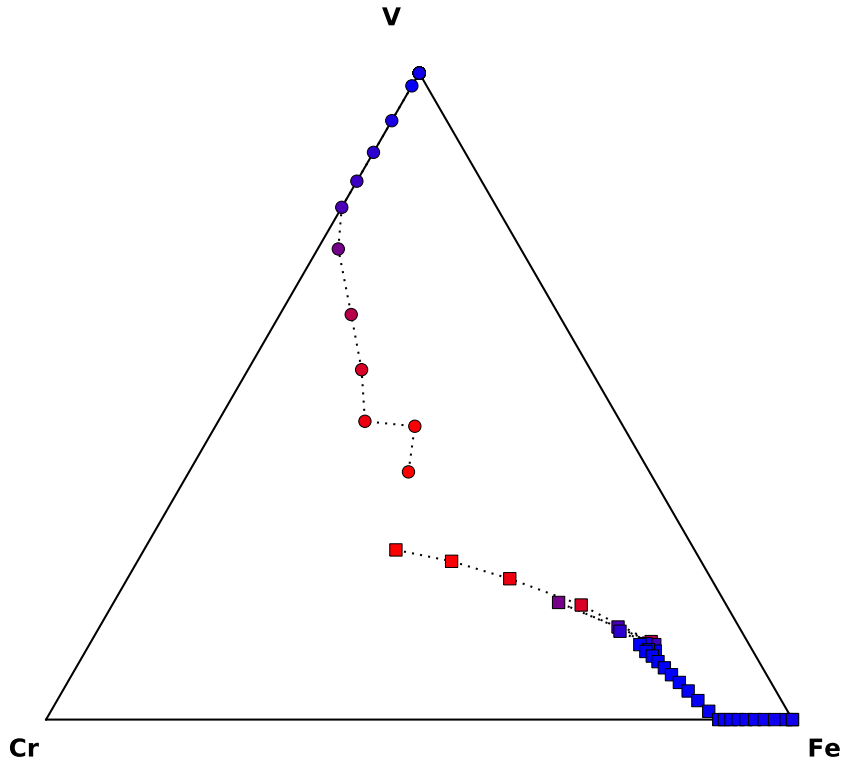


Figure: 32 atoms, 25% Al, 75% Ni, unpenalized optimization

Another problem: Stability of bcc Fe-V-Cr



Two atoms α, β, each can be any combination of Fe, V and Cr – maximize the alloy stability:

$$E[\alpha, \beta] - \tfrac{1}{2}E[\alpha, \alpha] - \tfrac{1}{2}E[\beta, \beta]$$

Thank you for listening