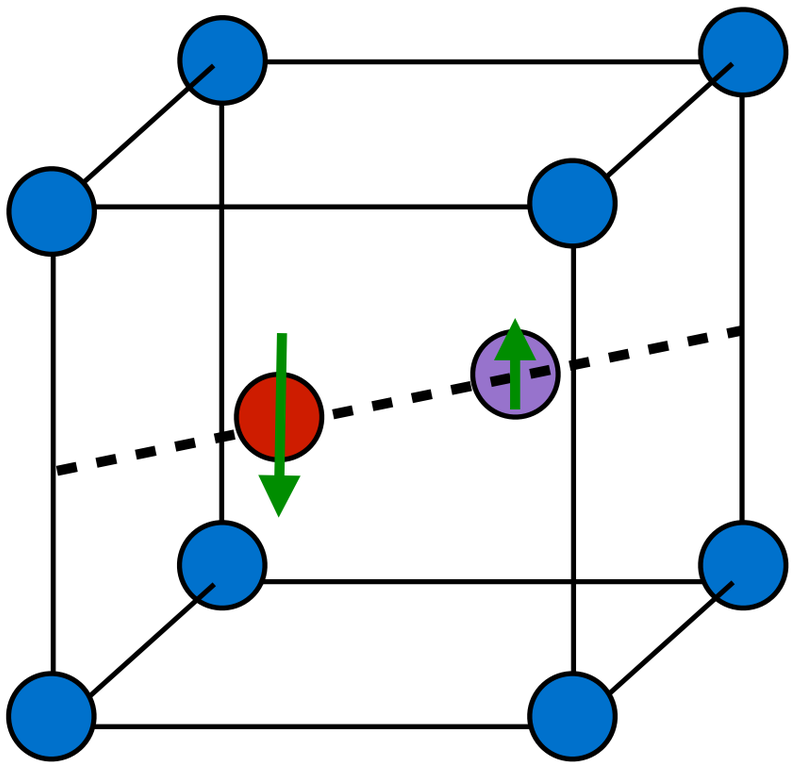
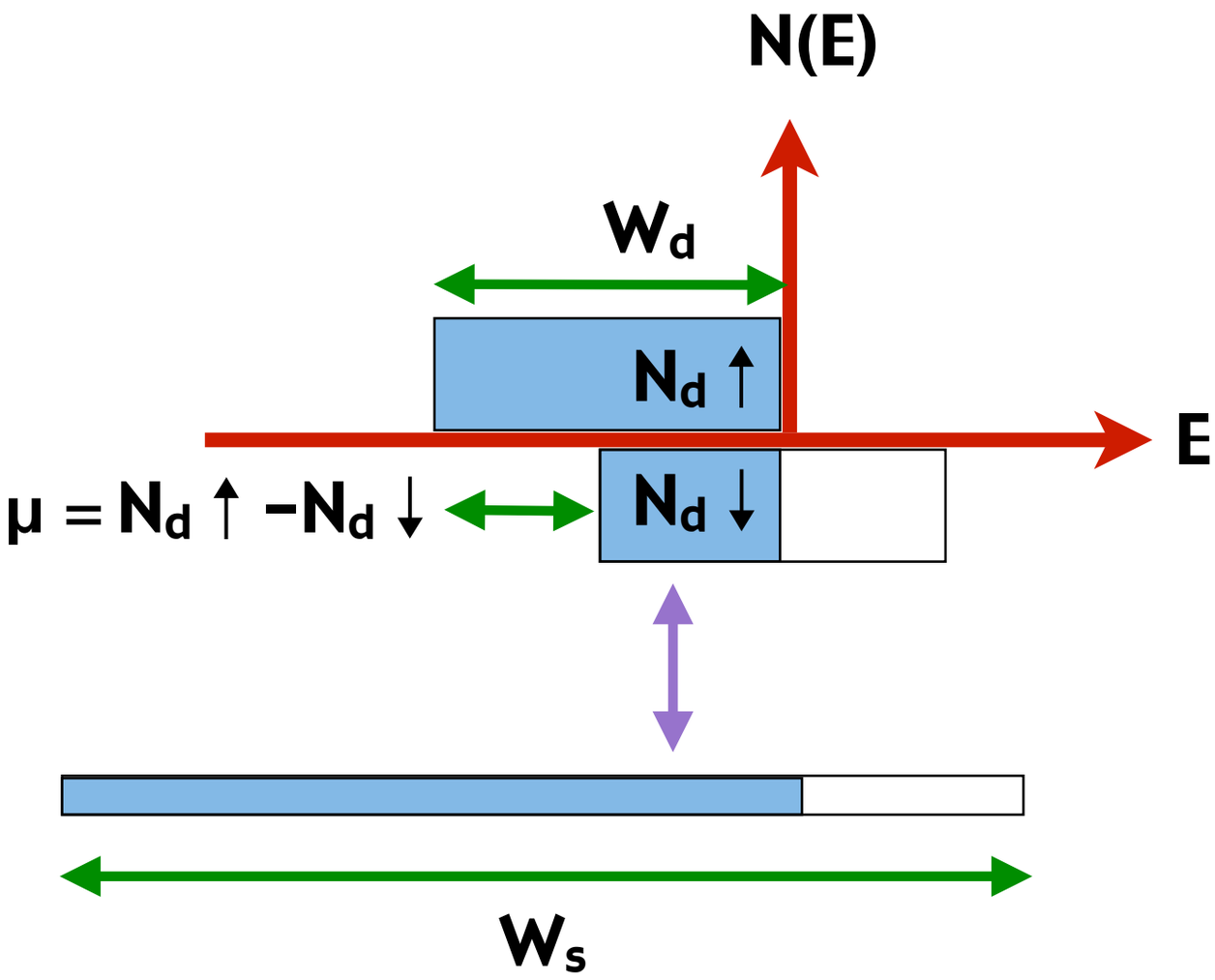
Finally, a functional spin-polarised potential for alpha and gamma iron?!!



Janne Wallenius & Graeme Ackland

22rd Fe-Cr workshop, Edinburgh, June 2013

A three-band 2nd moment model of Fe



- Density of states approximated by square bands for d-up, d-down and s-electrons
- Magnetic moment = $N_d\uparrow - N_d\downarrow$
- Transfer between s- and d-bands taken into account
- Cost for electron transfer: $E_{s\rightarrow d}$

Cohesive models

- Cohesive energy in 2nd moment models:

$$E_{coh}^{NM} = \left(\frac{N_d^2}{20} - \frac{N_d}{2} \right) W_d + \left(\frac{N_s^2}{4} - \frac{N_s}{2} \right) W_s$$

- The width is expressed as a square root of sums of pair interactions

$$W_b \propto \sqrt{\sum_n \phi_n(r_{ij})}$$

- 2nd moment expression for magnetic moment can be derived for a perfect lattice

$$\mu_i = \pm(10 - N_d) \sqrt{1 - \frac{2}{E_{sp}} \left(\frac{W_d}{5} + 2 \sum_j \left(\frac{1}{2} J_P + J_H \right) \right)}$$

Simplified model used for this fit

- Non-magnetic energy (only d-band retained)

$$E_i^{NM} = \left(\frac{N_d^2}{20} - \frac{N_d}{2} \right) W_{d,i} + \frac{1}{2} \sum_j V(r_{ij})$$

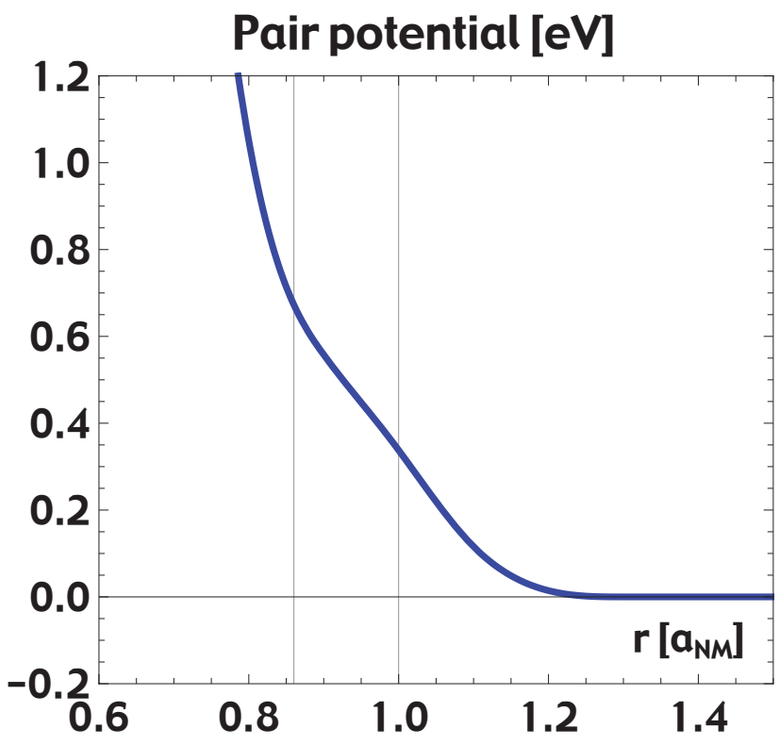
- Magnetic moment (single function for exchange-correlation interaction)

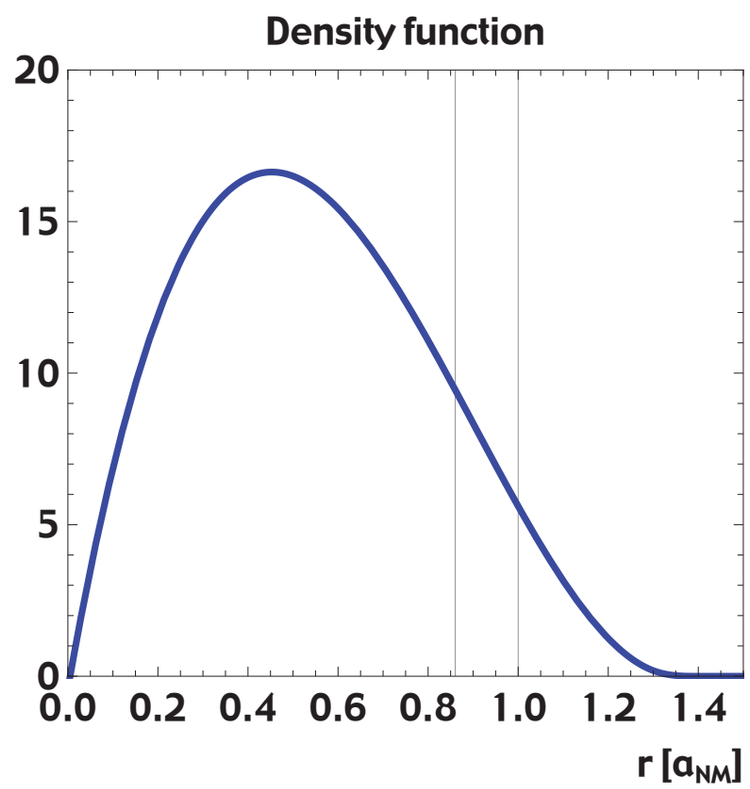
$$\mu_i = \pm (10 - N_d) \sqrt{1 - \frac{2}{E_{free}^{\mu}} \left(\frac{W_d}{5} + 2 \sum_j J_{XC}(r_{ij}) \right)}$$

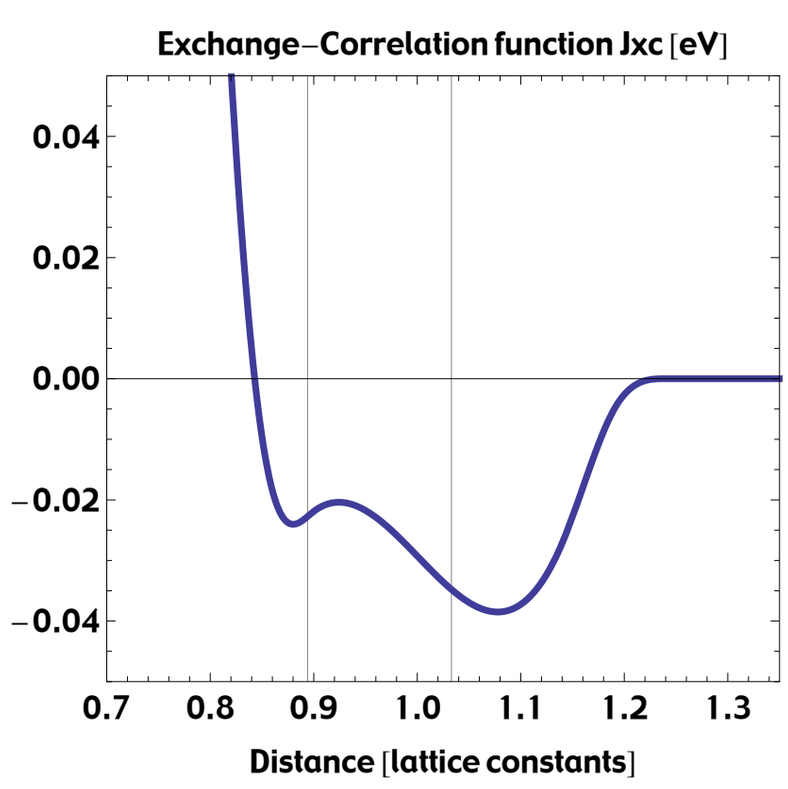
- Total energy

$$E_i^{NM} = \left(\frac{N_d^2 + \mu_i^2}{20} - \frac{N_d}{2} \right) W_{d,i} + \frac{1}{2} \sum_j V(r_{ij}) + \frac{1}{2} \sum_j J_{XC}(r_{ij}) \mu_i \mu_j - \frac{E_{free}^{\mu}}{8} \left(\mu_i^2 - \frac{\mu_i^4}{2\mu_{free}^2} \right)$$

Functions fitted to alpha and gamma iron

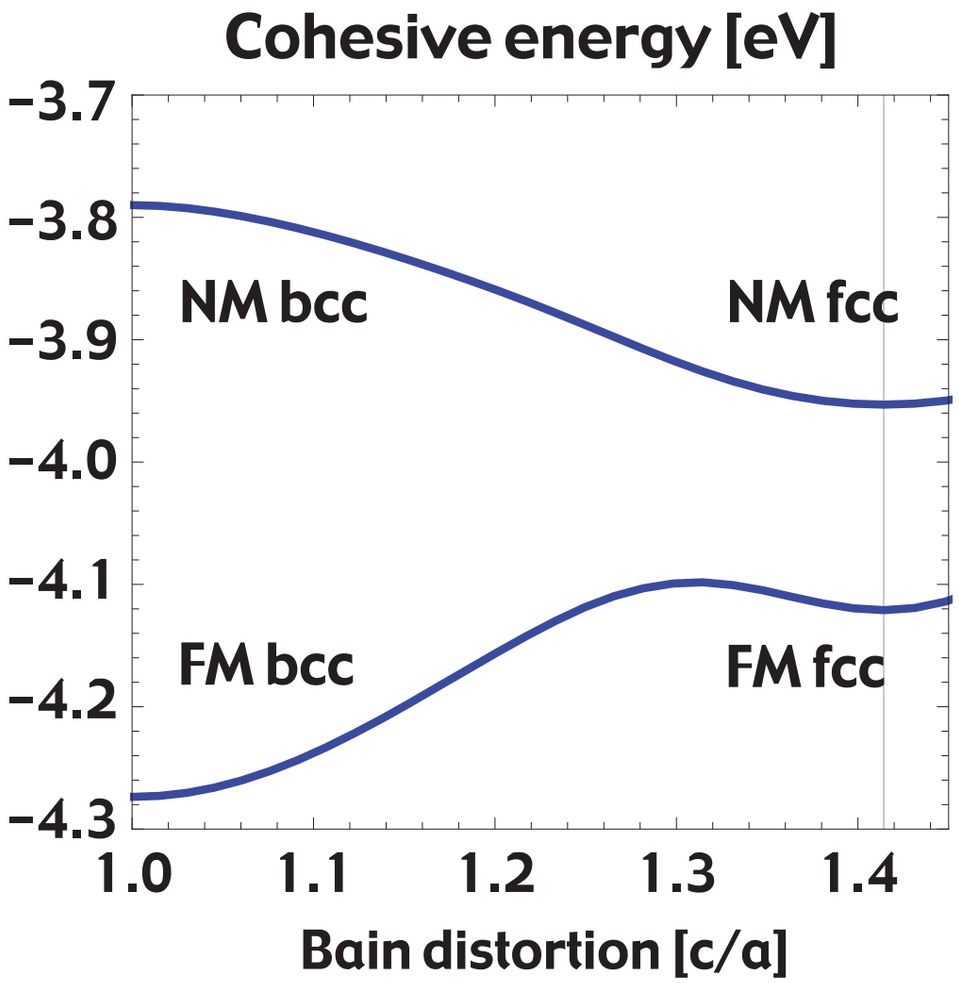






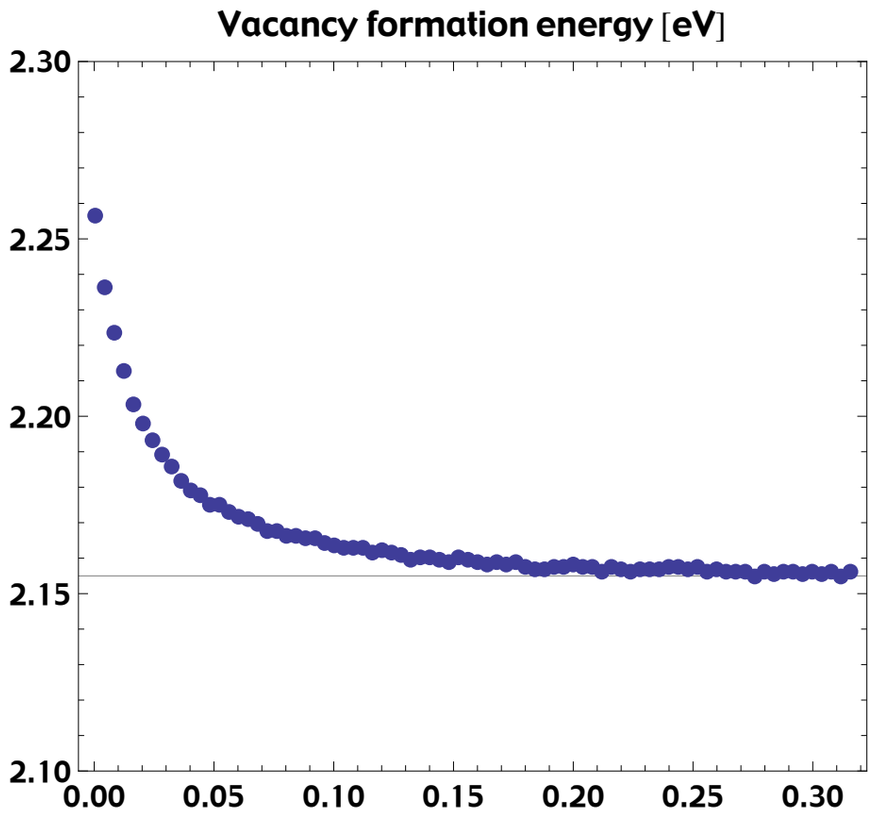
- Total of 13 splines fitted to properties of non-magnetic & ferro-magnetic iron
- Range of potential: 2nd nearest neighbour in bcc lattice (1st n.n. in fcc!)

Phase stability & Bain paths



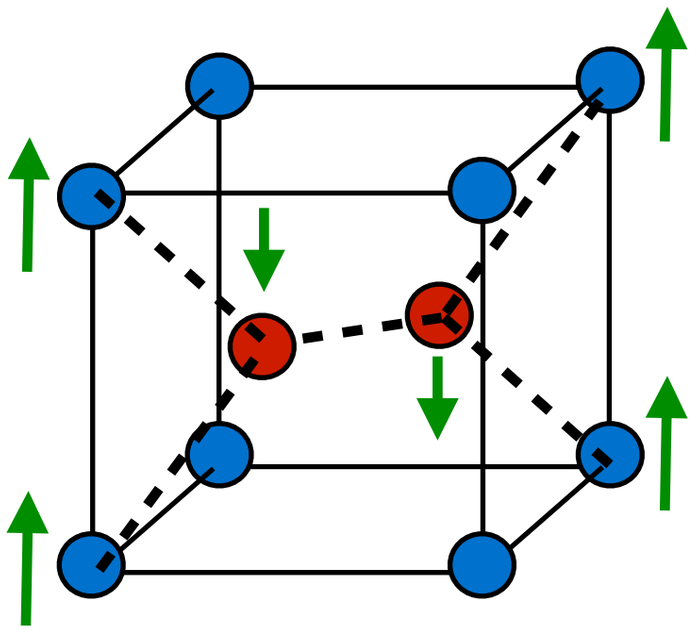
- The present potential provides
- Correct lattice parameters and elastic modulii of magnetic and non-magnetic bcc phases.
- Correct ordering between non-magnetic and ferro-magnetic bcc & fcc phases
- Meta-stability of ferro-magnetic fcc!
- Potential to correctly reproduce the phase diagram of Fe!

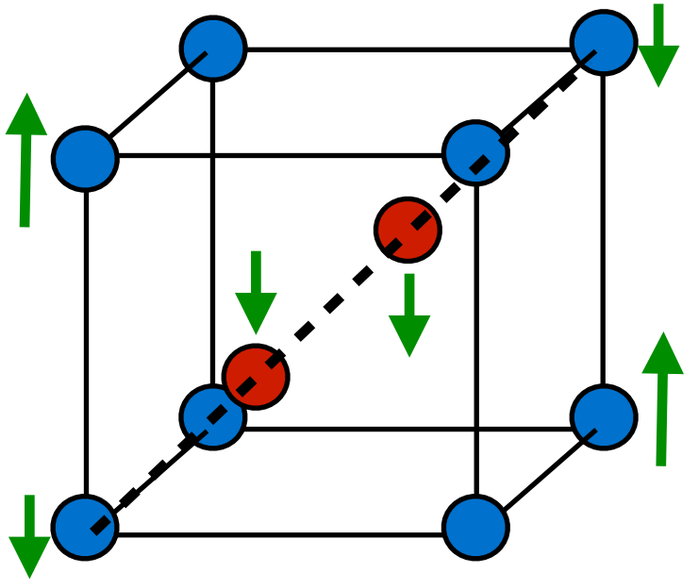
Vacancy formation energy (FM-bcc)



- Vacancy relaxation energy: -0.14 eV
- Ab initio: -0.24 eV
- 1st nn magnetic moment: +0.06 eV
- Ab initio: +0.21 eV

Interstitial defects: Quenching of moments





- ‹110› : each atom in SIA has 2 nearest neighbours, both with un-quenched moments
- ‹111› each atom in SIA has 1 nearest neighbour, with quenched moments
- Unquenched moments yield lower total energy, hence lower energy of the ‹110› configuration!

Interstitial defects: magnetic calculation without spin-flip

Property	Potential	DFT (VASP)
$E_{\langle 110 \rangle}$	4.90 eV	4.02 eV
$\mu_{\langle 110 \rangle}$	0.00 μ_B	-0.18 μ_B
$d_{\langle 110 \rangle}$	2.20 Å	1.93 Å
$E_{\langle 111 \rangle}$	7.02 eV	4.72 eV
$\mu_{\langle 111 \rangle}$	0.00 μ_B	-0.48 μ_B
$d_{\langle 111 \rangle}$	1.97 Å	1.86 Å

- Correct order of stability is observed
- SIA inter-atomic distance too large compared to DFT
- Box-size: 251 atoms
- Spin-flip at critical distance to be implemented, in order to achieve correct SIA distance and hence magnitude of formation energy.

Conclusions

- 2nd moment model of ferro-magnetism in alpha and gamma iron now exists.
- Ordering of SIA defects results directly from quenching of magnetic moments
- Implementation in MD codes imminent
- Next workshop: Anti-ferromagnetism, Curie temperature, diffusion and cascades?!