Importance of Madelung potential for magnetism of alloys

Disordered FePt studied via CPA and via supercells

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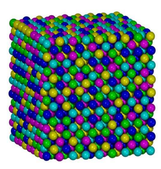
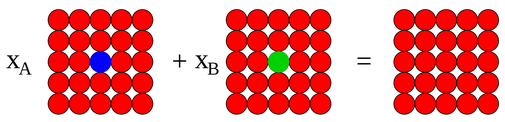
Dealing with substitutional alloys

1. Simulating random occupation of sites via supercells



- If sufficiently large number of configurations taken into account, provides an exact description.
- However, it may be computationally expensive.

2. Coherent potential approximation (CPA)

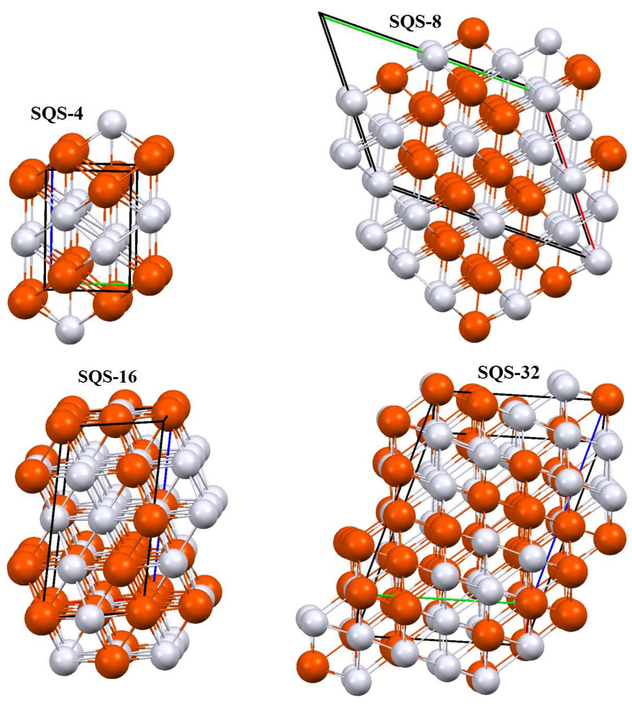


- Mean field theory: disorder simulated by auxiliary effective medium.
- Computationally cheap.
- Trends of properties with alloy composition often described very well.
- Fluctuations in the local environment neglected (single-site method).
- Madelung contribution to the potential cannot be included (within the standard CPA).

Generating supercells efficiently: special quasirandom structures (SQS's)

Periodic structures created so that they have the same correlation functions as random alloys up to a certain coordination shell.

Disordered FePt via SQS's of 4, 8, 16, and 32 atoms in unit cell:



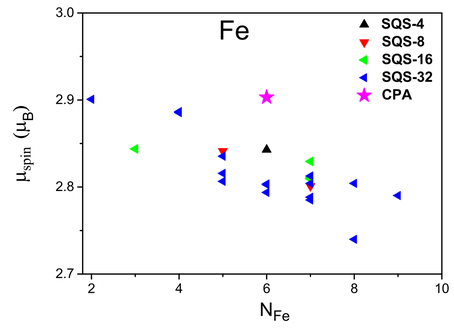
CPA and supercells calculations differ

Average spin and orbital magnetic moments for Fe and Pt atoms in disordered FePt alloy:

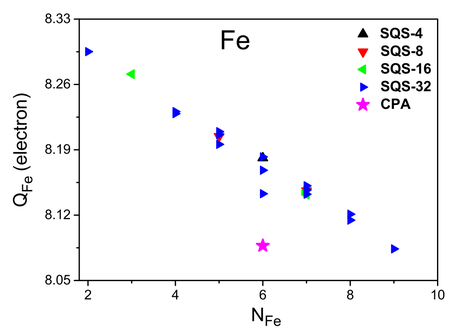
		$\langle\mu_{\text{spin}}\rangle$	$\langle\mu_{\text{orb}}\rangle$
SQS-4	Fe	2.843	0.043
	Pt	0.253	0.027
SQS-8	Fe	2.821	0.069
	Pt	0.286	0.051
SQS-16	Fe	2.823	0.066
	Pt	0.263	0.042
SQS-32	Fe	2.816	0.073
	Pt	0.264	0.043
⟨SQS-4,8,16,32⟩	Fe	2.821	0.069
	Pt	0.266	0.043
CPA	Fe	2.903	0.070
	Pt	0.239	0.039

Even for the largest SQS, a small but distinct difference in magnetic moments obtained via the supercells and via the CPA remains!

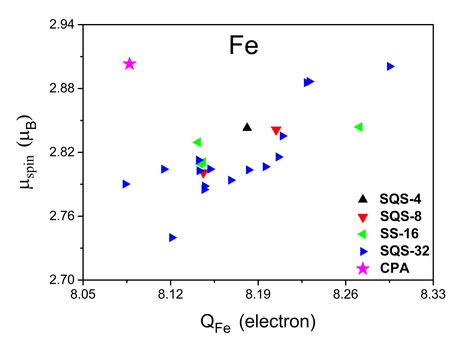
More detailed view on the differences between CPA and supercells



Dependence of spin moments at Fe sites on the number of Fe atoms in the first coordination shell N_{Fe}.



Dependence of electronic charge at Fe sites on N_{Fe}.



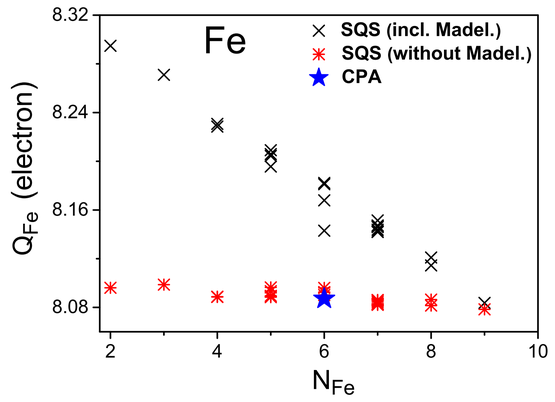
Dependence of spin moment μ_{spin}(Fe) on the charge Q_{Fe}.

Where does the difference between CPA and supercells come from?

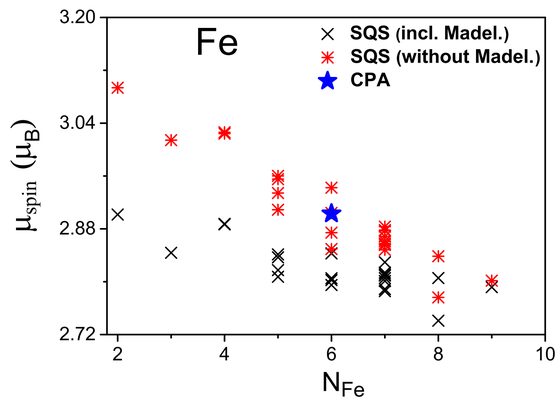
- The difference in description of magnetism is linked to the difference in description of electronic charge.
- CPA cannot account for the Madelung potential. If supercells are used, the Madelung potential is normally included.

Effect of the Madelung potential

Apply the supercells method, either with the Madelung potential or without it. See how the results compare with values obtained via the CPA.



Dependence of the electronic charge at Fe sites on the coordination number N_{Fe}, obtained for supercell calculation either with the Madelung potential or without it. The CPA result shown for comparison.



Dependence of spin moment at Fe sites on coordination number N_{Fe}. Analogous to the figure above.

Charges Q and magnetic moments μ_{spin} obtained by averaging over all sites in all four SQS's, obtained either with the Madelung potential or without it.

	Fe		Pt	
	Q_{Fe}	μ_{spin}	Q_{Pt}	μ_{spin}
⟨SQS⟩ (incl. Madel.)	8.174	2.821	9.825	0.266
⟨SQS⟩ (no Madel.)	8.088	2.907	9.912	0.245
CPA	8.087	2.903	9.913	0.239

Computational method

- *Ab initio* KKR Green's function formalism, as implemented in the SPRKKR code [Ebert *et al.* Rep. Prog. Phys. 2011].
- Fully-relativistic, full-potential, LDA.
- FLAPW method as implemented in the WIEN2k code used for additional supercell calculations.

Executive summary

- By increasing the number of atoms N in special quasirandom structures SQS-N, the results approach the CPA.
- However, a small but distinct residual difference remains in magnetic moments.
- This difference is due to the neglect of the Madelung potential in the CPA.
- Khan *et al.* Phys. Rev. B **95**, 014408 (2017) for more.