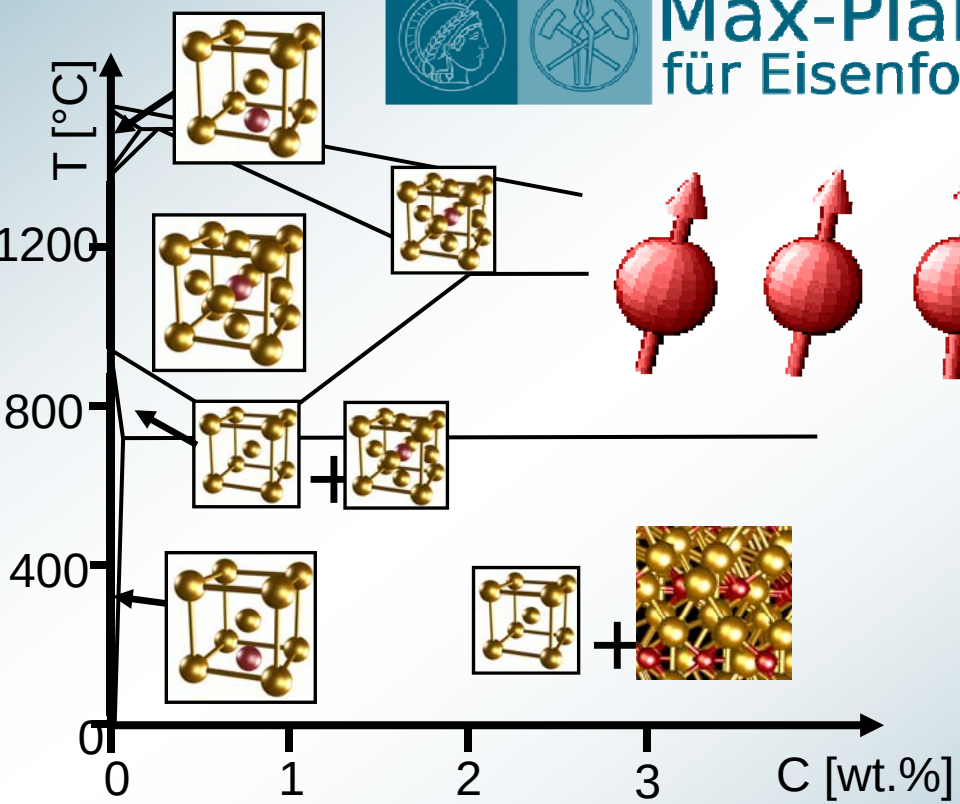
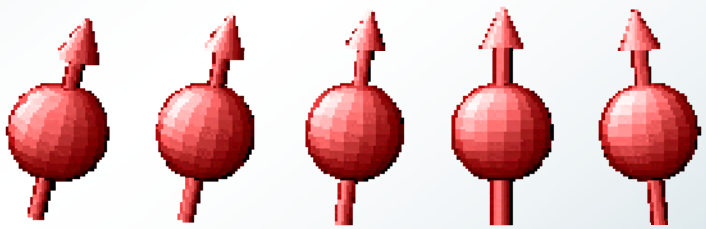
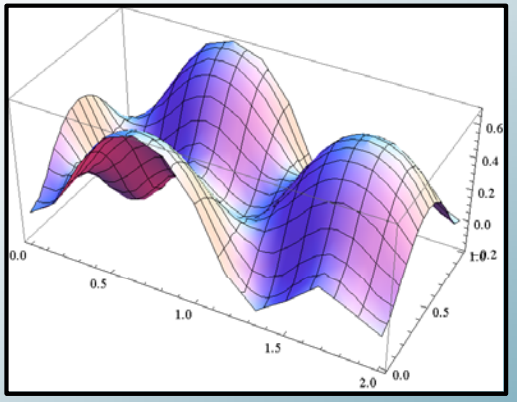
Ab initio investigation of austenitic steels: The interplay of composition, magnetism and mechanical behaviour

Tilmann Hickel, F. Körmann, I. Bleskov, J. Neugebauer

Max-Planck-Institut für Eisenforschung GmbH



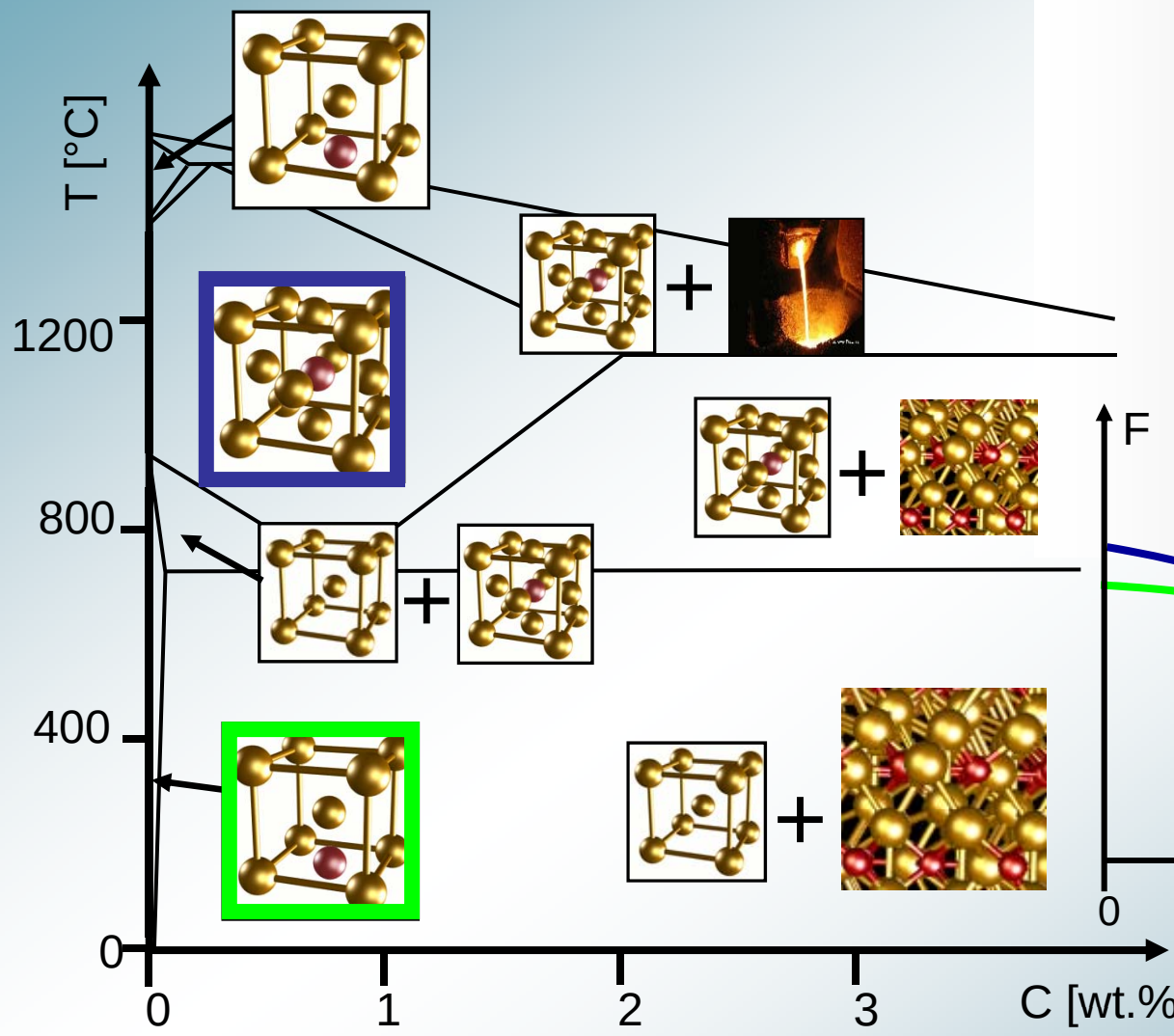


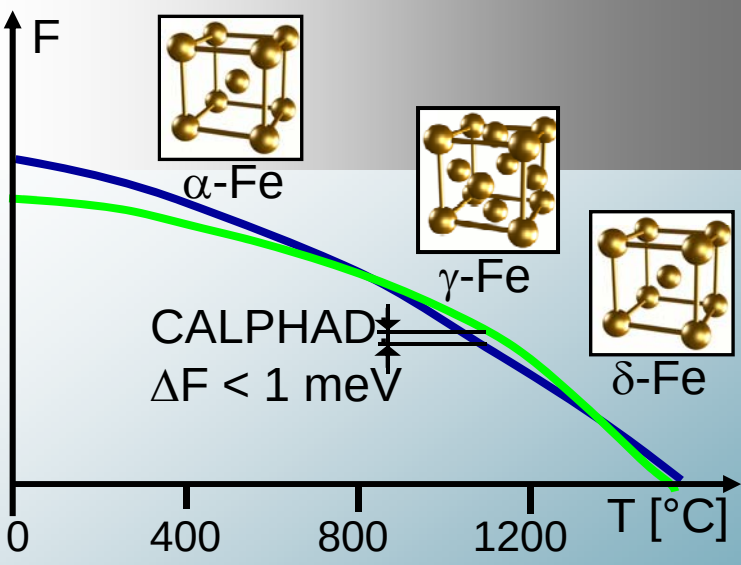


SFE

4th nFAME and 22nd Fe-Cr workshop
Edinburgh, 04/05 June 2013

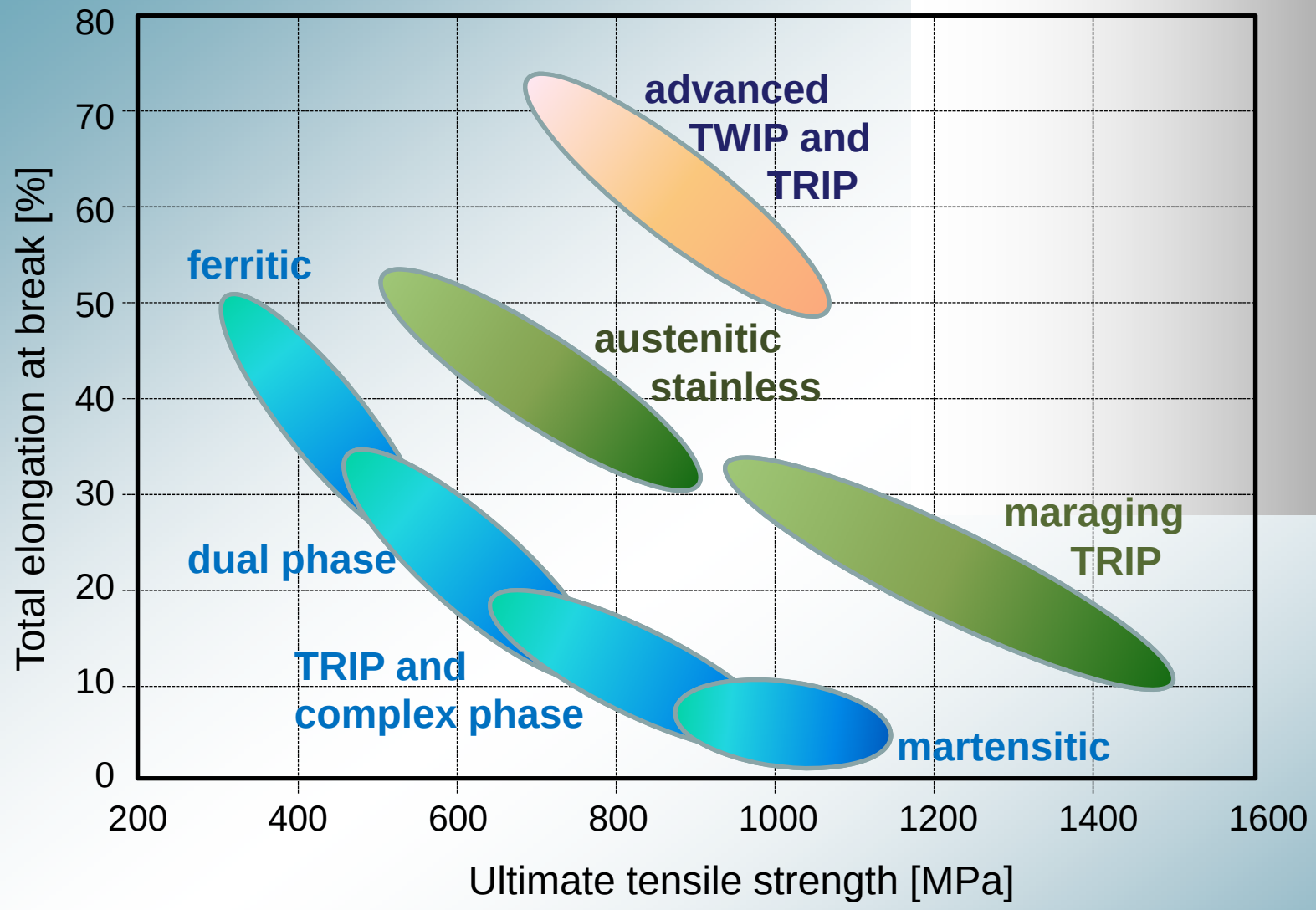
Phase diagrams: The Fe-C system



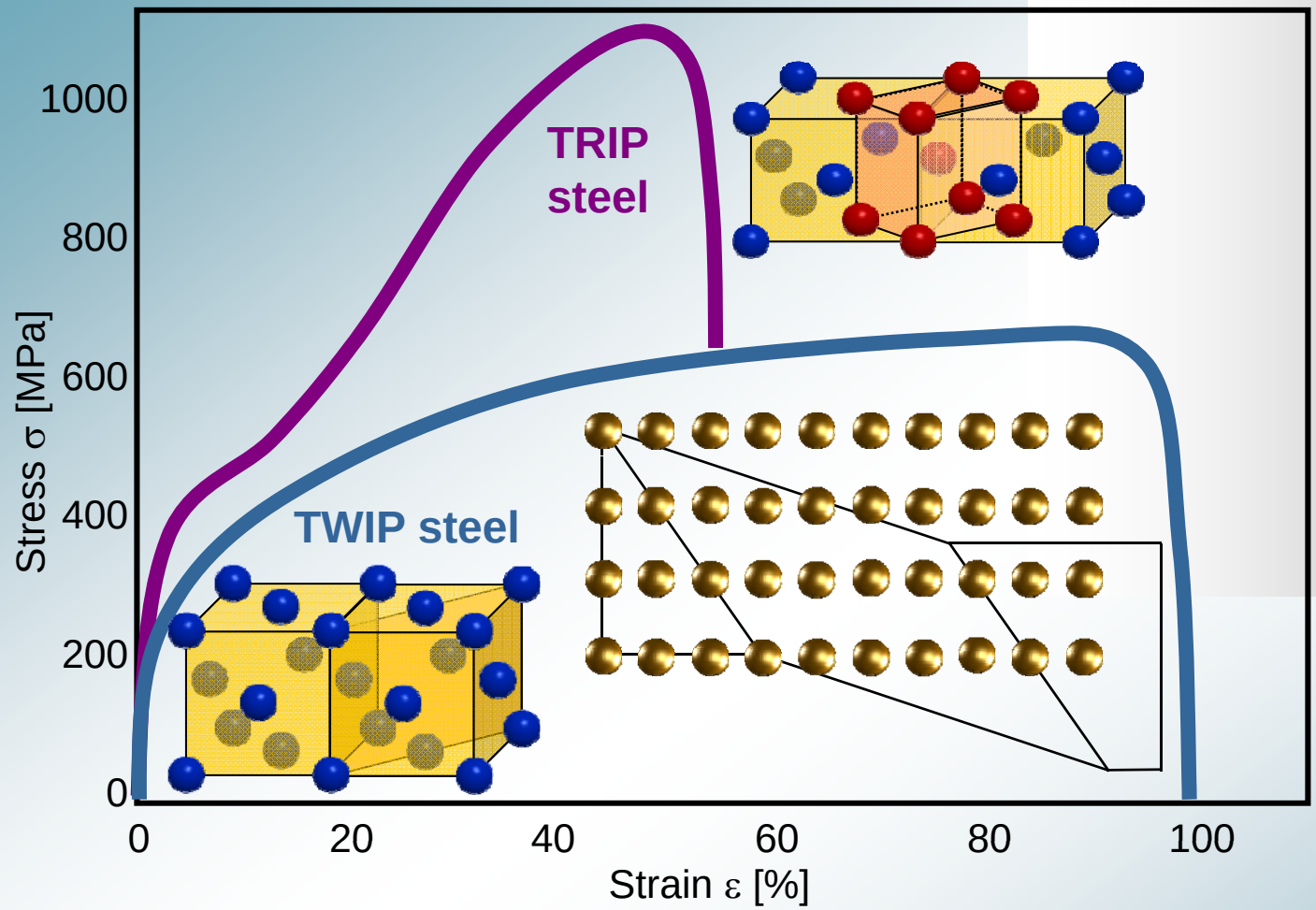


- accuracy 1000x smaller than typical binding energies required

Materials design for innovative steels



Materials design for innovative steels



- deformation processes in innovative steels: TRIP and TWIP
- controlled by stacking fault energy
- based on free energy calculations

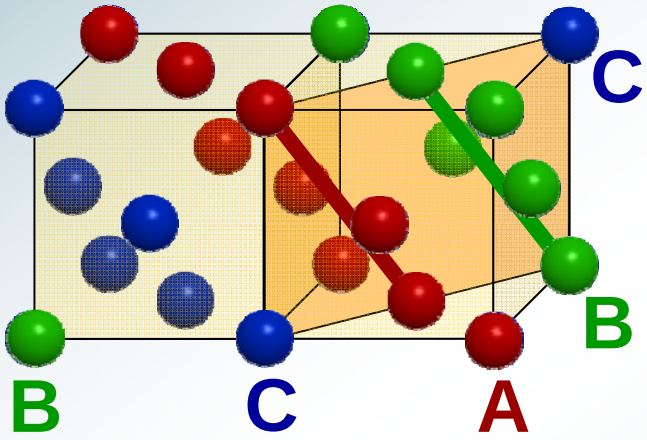
Stacking sequences

TRIP

phase transformation

$\gamma \rightarrow \varepsilon \rightarrow \alpha'$

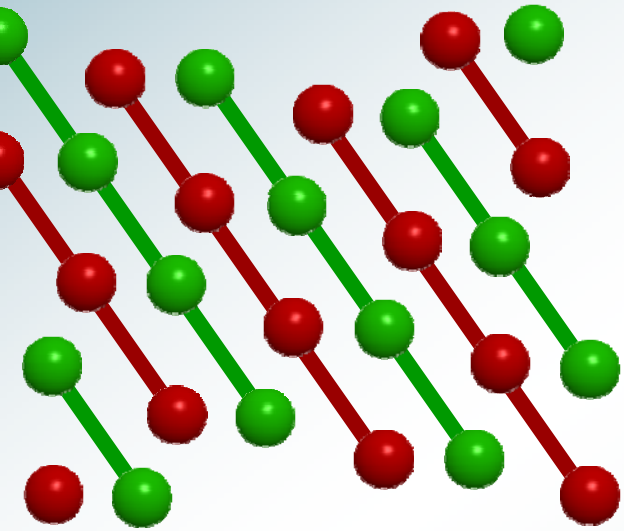
Austenite



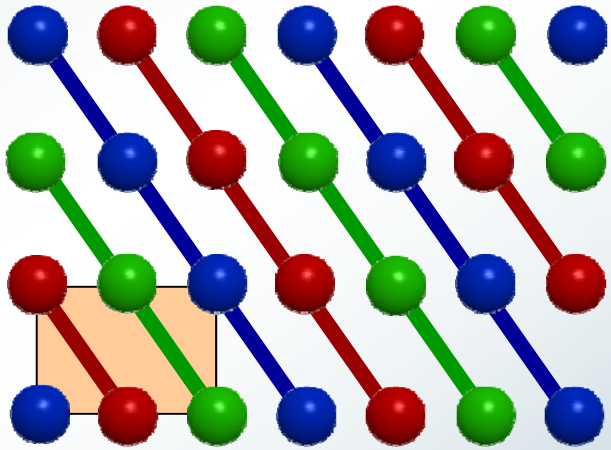
TWIP

twinning formation

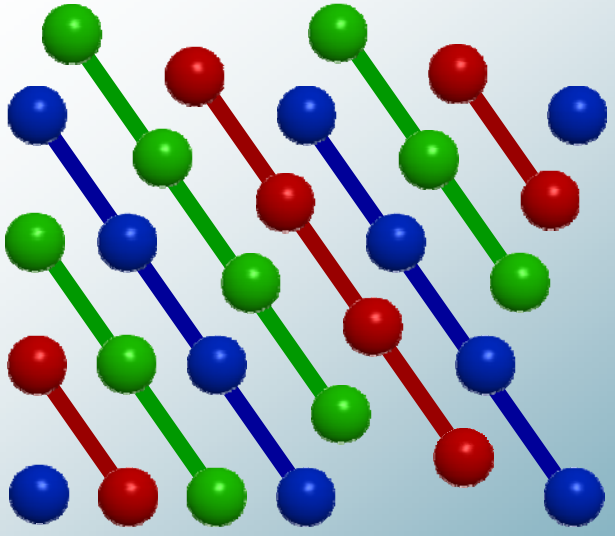
Σ_3 twin boundaries



A B A B A B A



C A B C A B C



C A B C B A C

- stacking faults are decisive for mechanical behaviour
- stacking fault energy (SFE) determines importance of TRIP, TWIP

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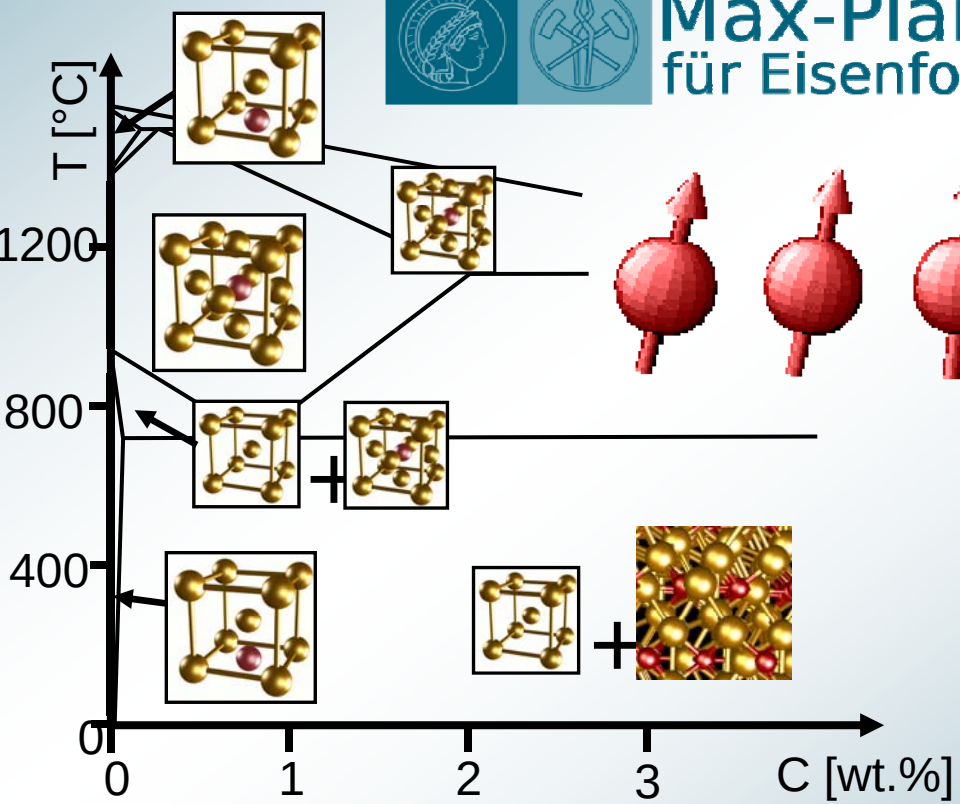

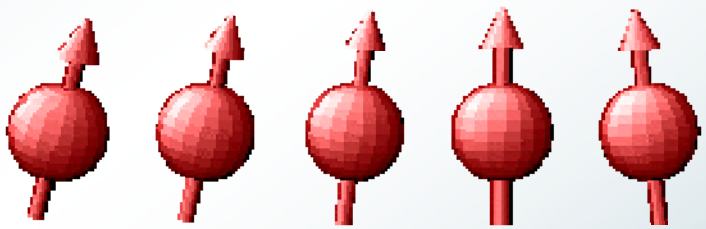

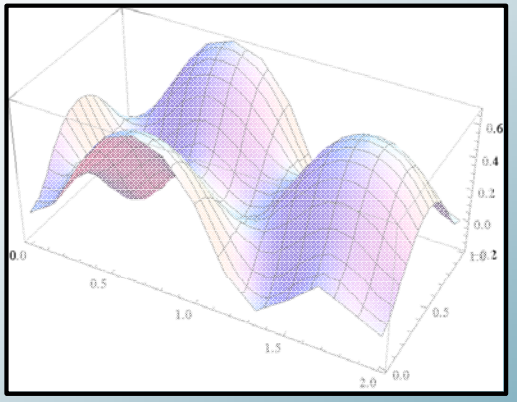

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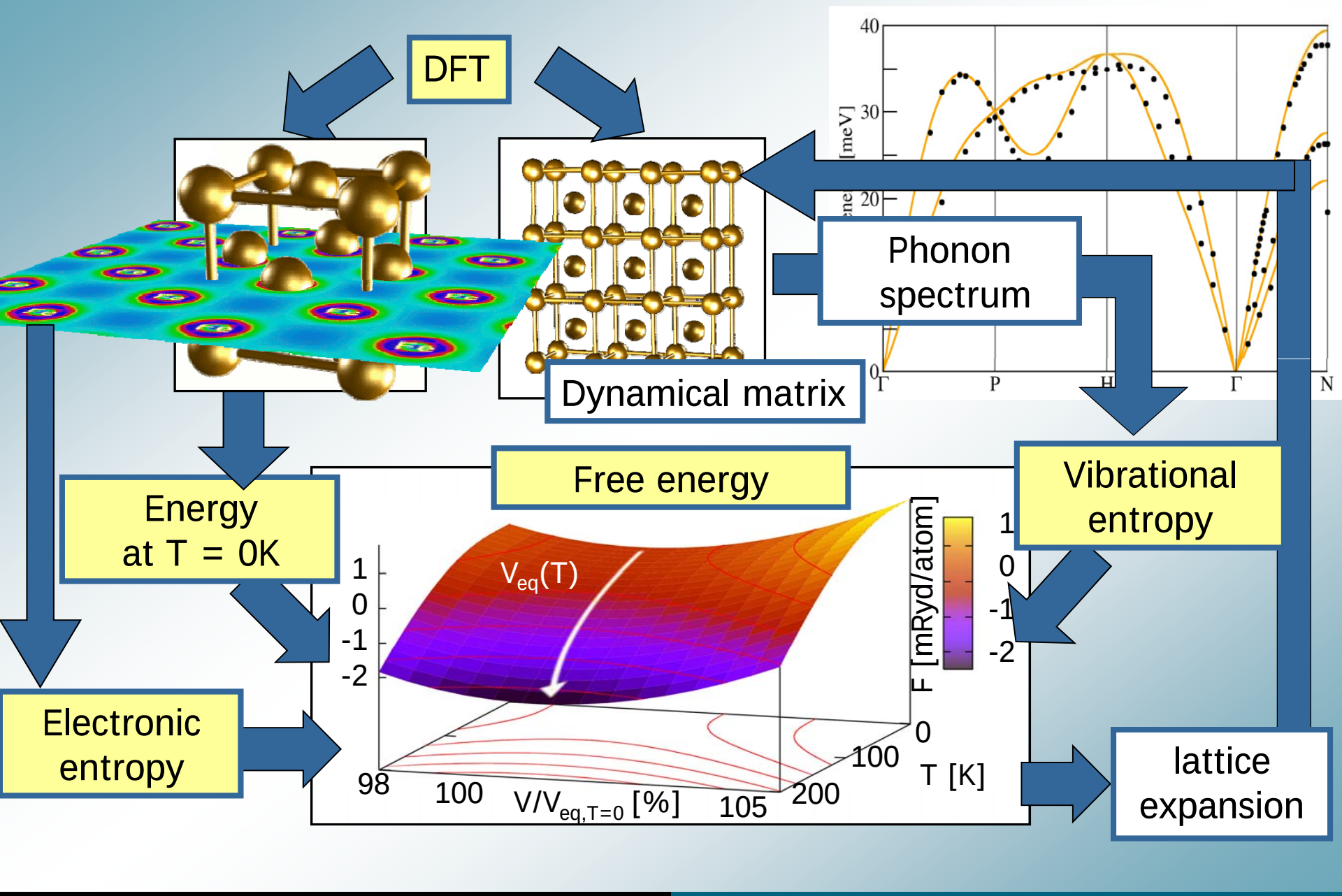
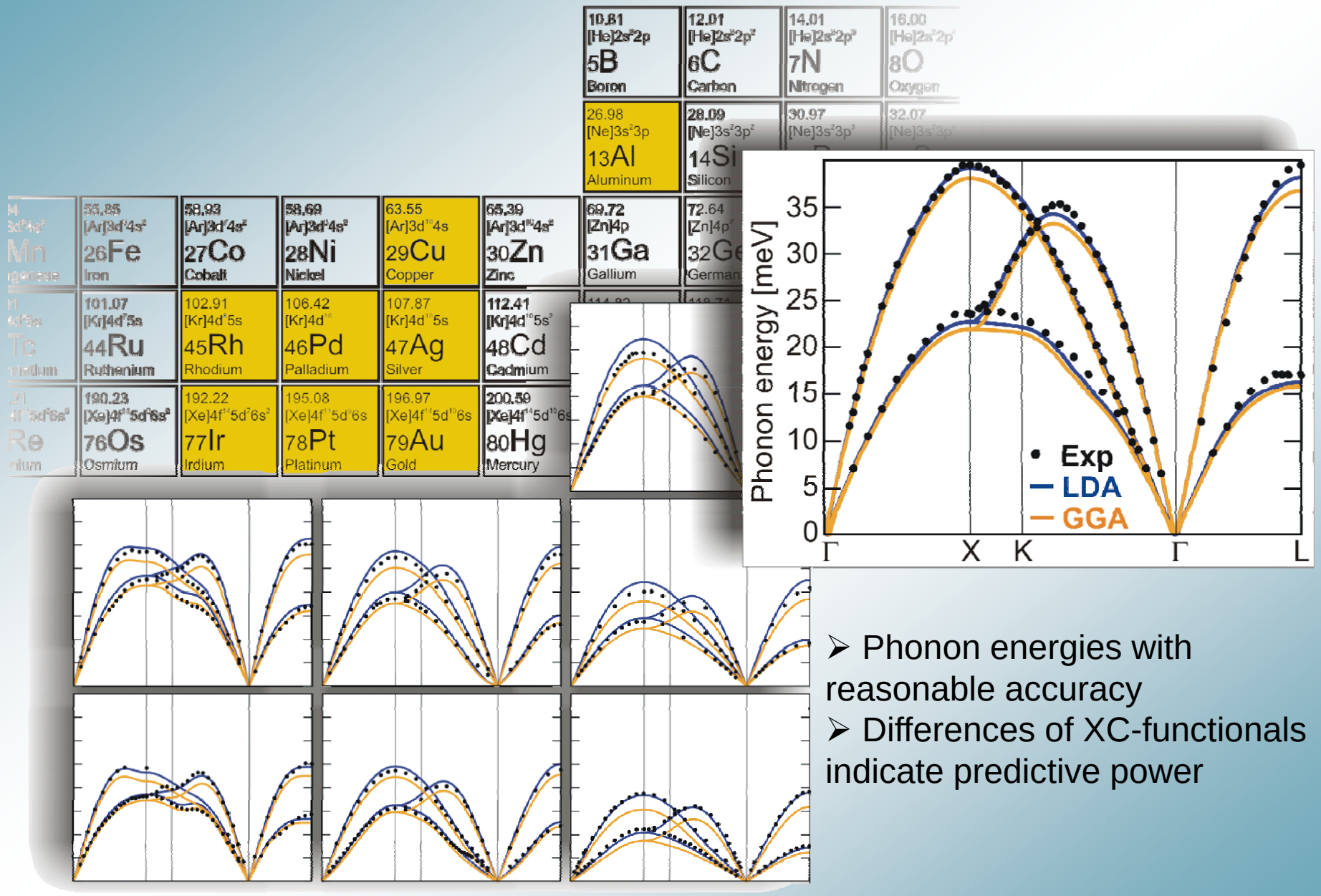
Ab initio for finite temperatures

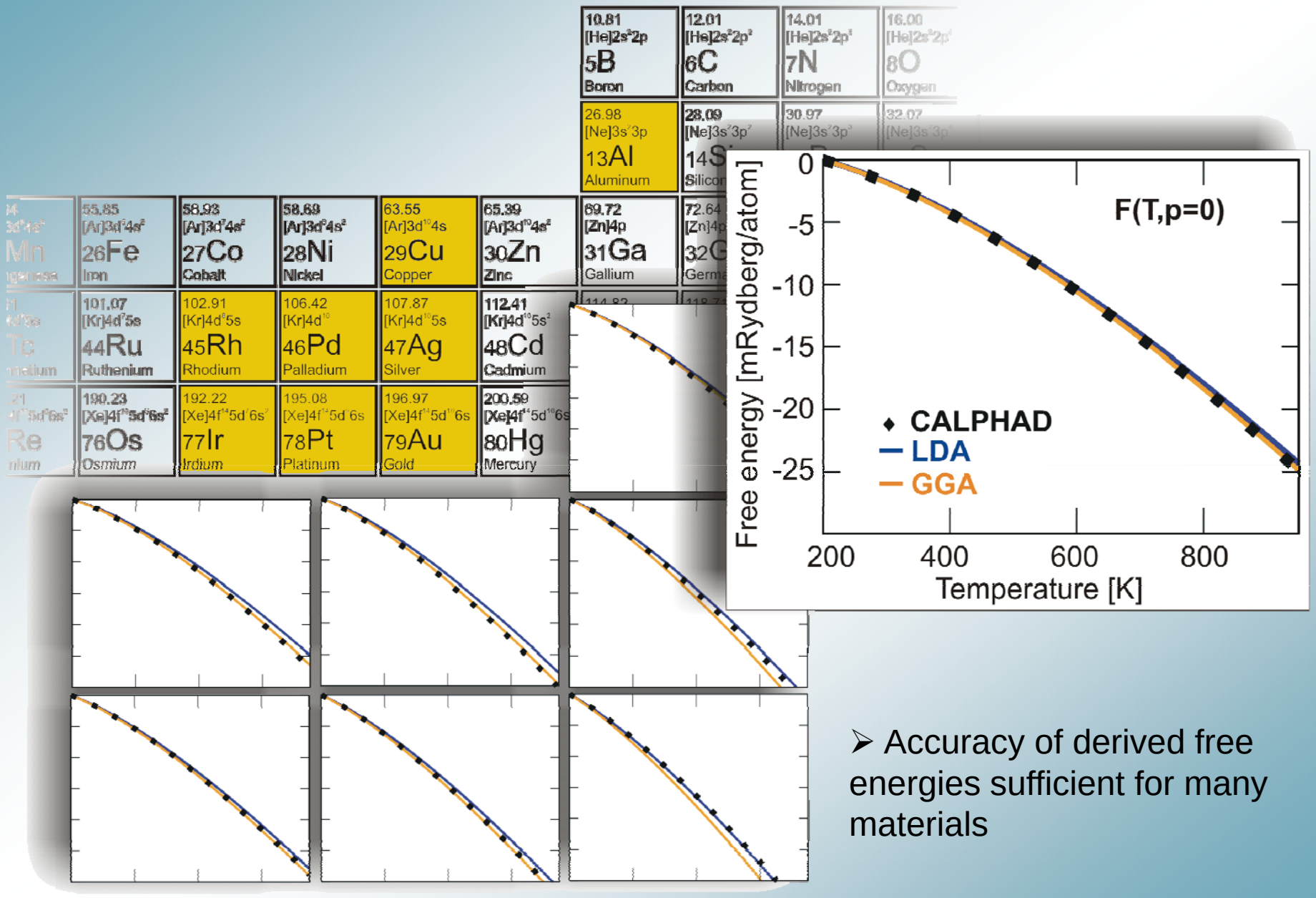


Performance for phonon energies



- Phonon energies with reasonable accuracy
- Differences of XC-functionals indicate predictive power

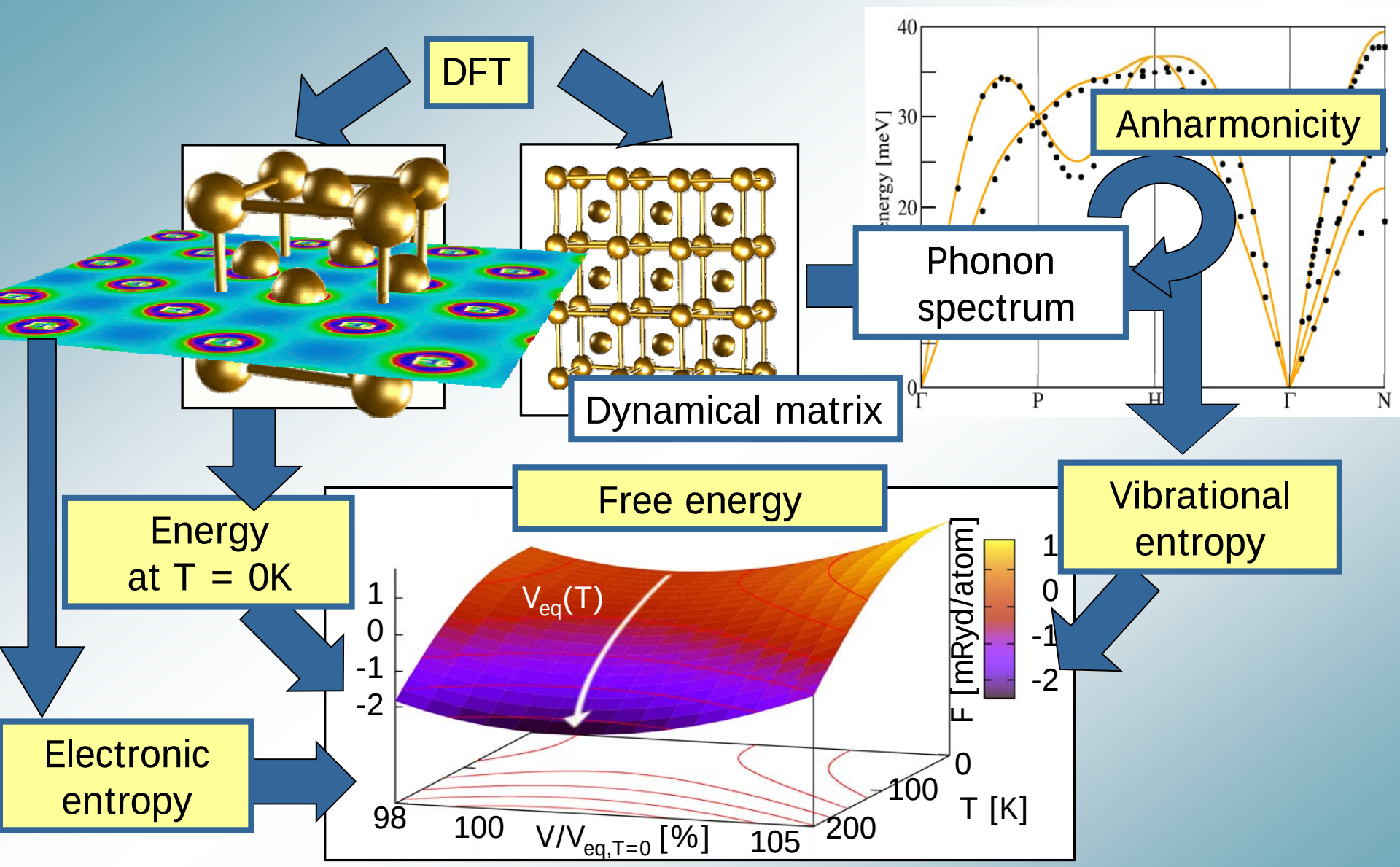
Performance for free energies



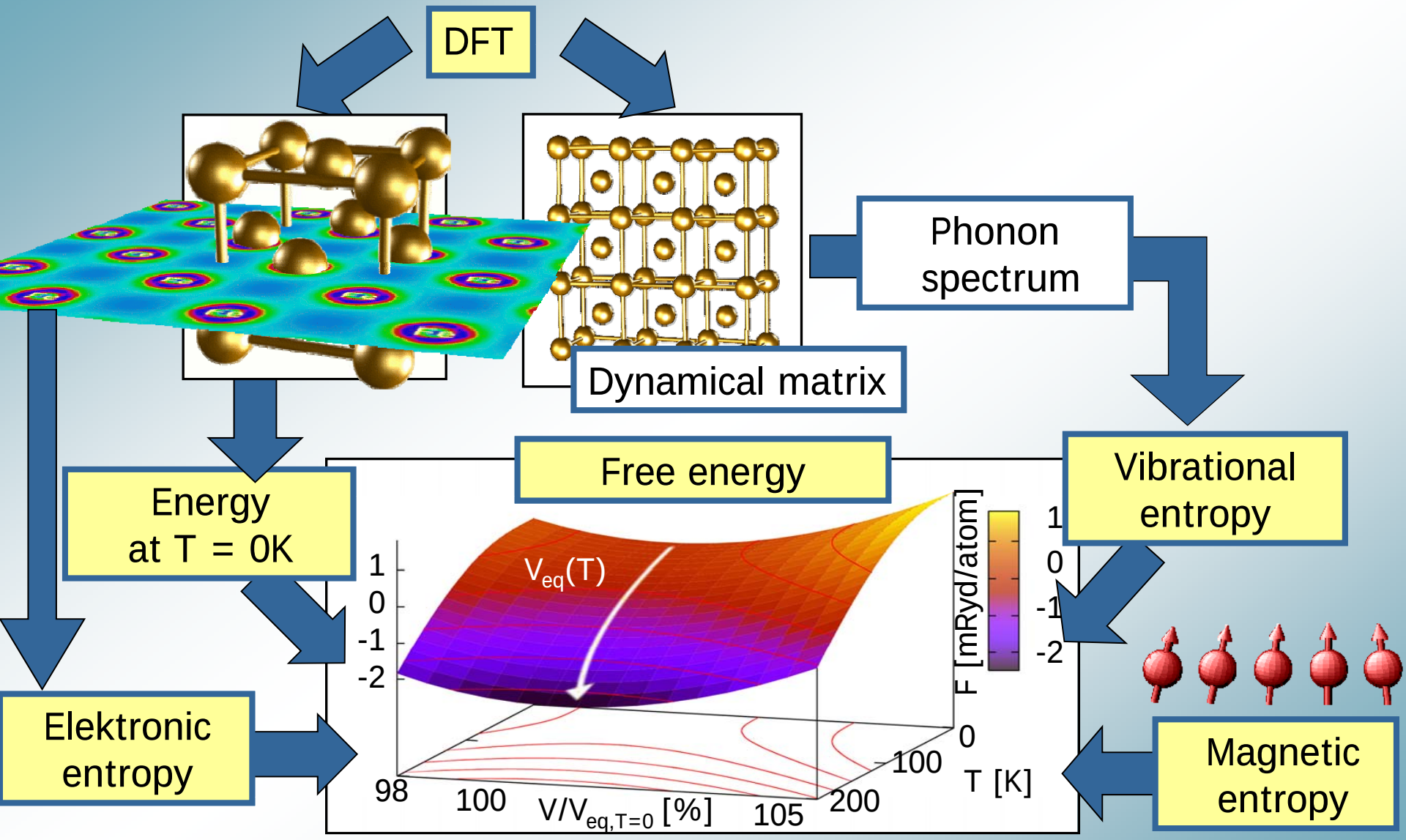
➢ Accuracy of derived free energies sufficient for many materials

B. Grabowski, TH, JN, PRB **76**, 024309 (2007).

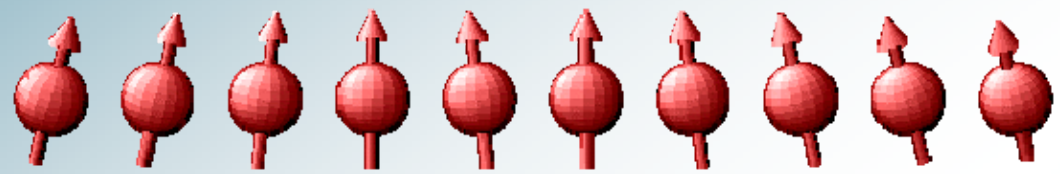
Ab initio for finite temperatures



Ab initio for finite temperatures



Treatment of magnetic excitations



- DFT for magnons

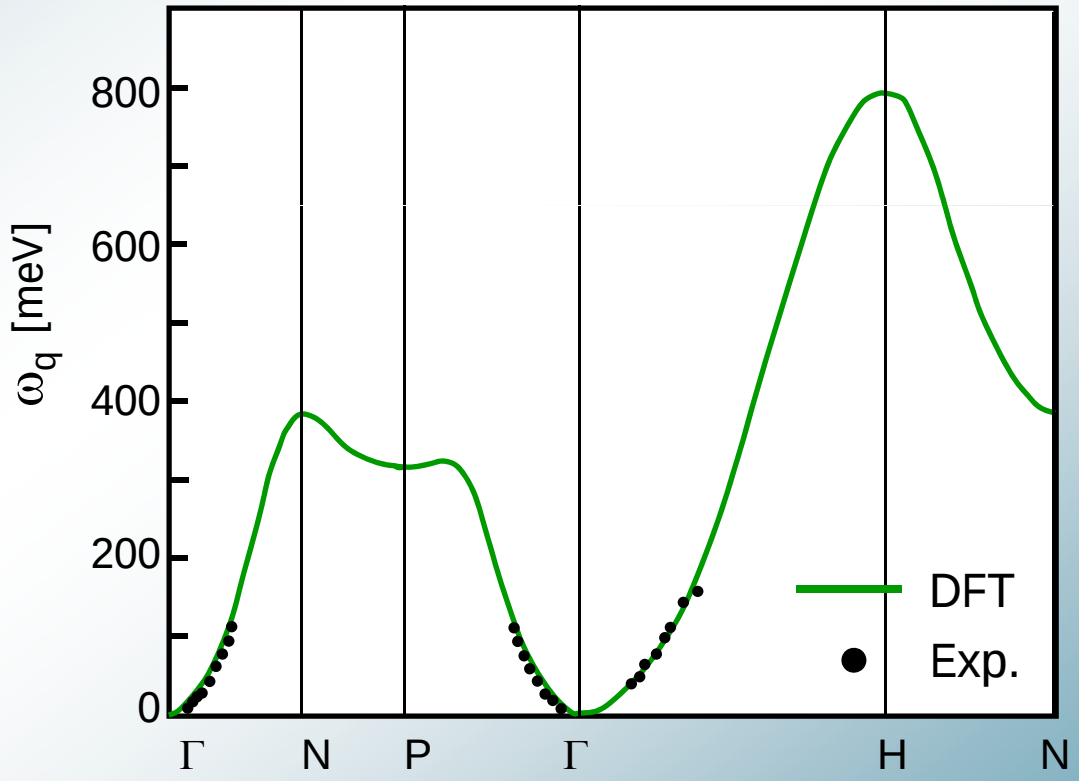
$$\omega_q = \frac{4}{M} \lim_{\theta \to 0} \frac{\Delta E(q,\theta)}{\sin^2 \theta}$$

- calculated and measured magnons in good agreement

$$H = -\sum_{i,j} J_{i,j}\, \mathbf{s}_i \cdot \mathbf{s}_j$$

- exchange integrals derived from magnon spectrum

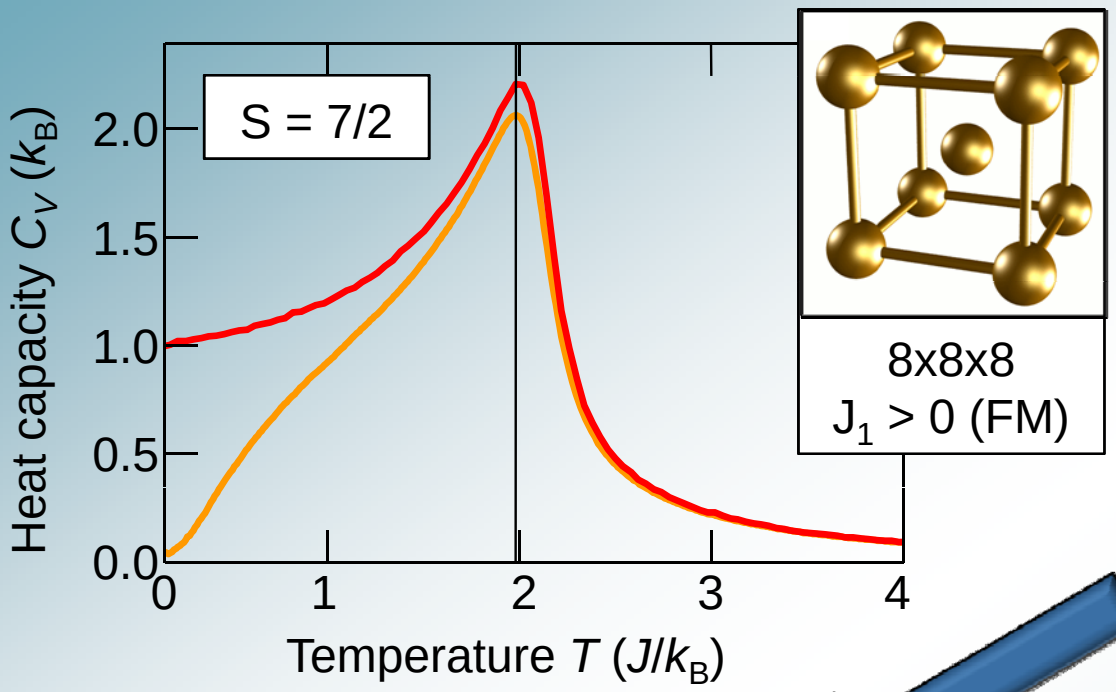
$$\omega_q = \frac{4}{M}\left(J_0 - J_q\right)$$



J. Lynn, PRB **11**, 2624 (1974).
C. Loong et al., JAP **55**, 1895 (1984).

F. Körmann, TH, *et al.*, PRB **78**, 033102 (2008).

Simulation of Heisenberg model systems



- well defined (finite size) crystal structure
- only nearest-neighbor magnetic interaction

$$H = -\sum_{ij} J_{ij} \hat{S}_i \hat{S}_j$$

Quantum solution

$\hat{S}_j$ operators

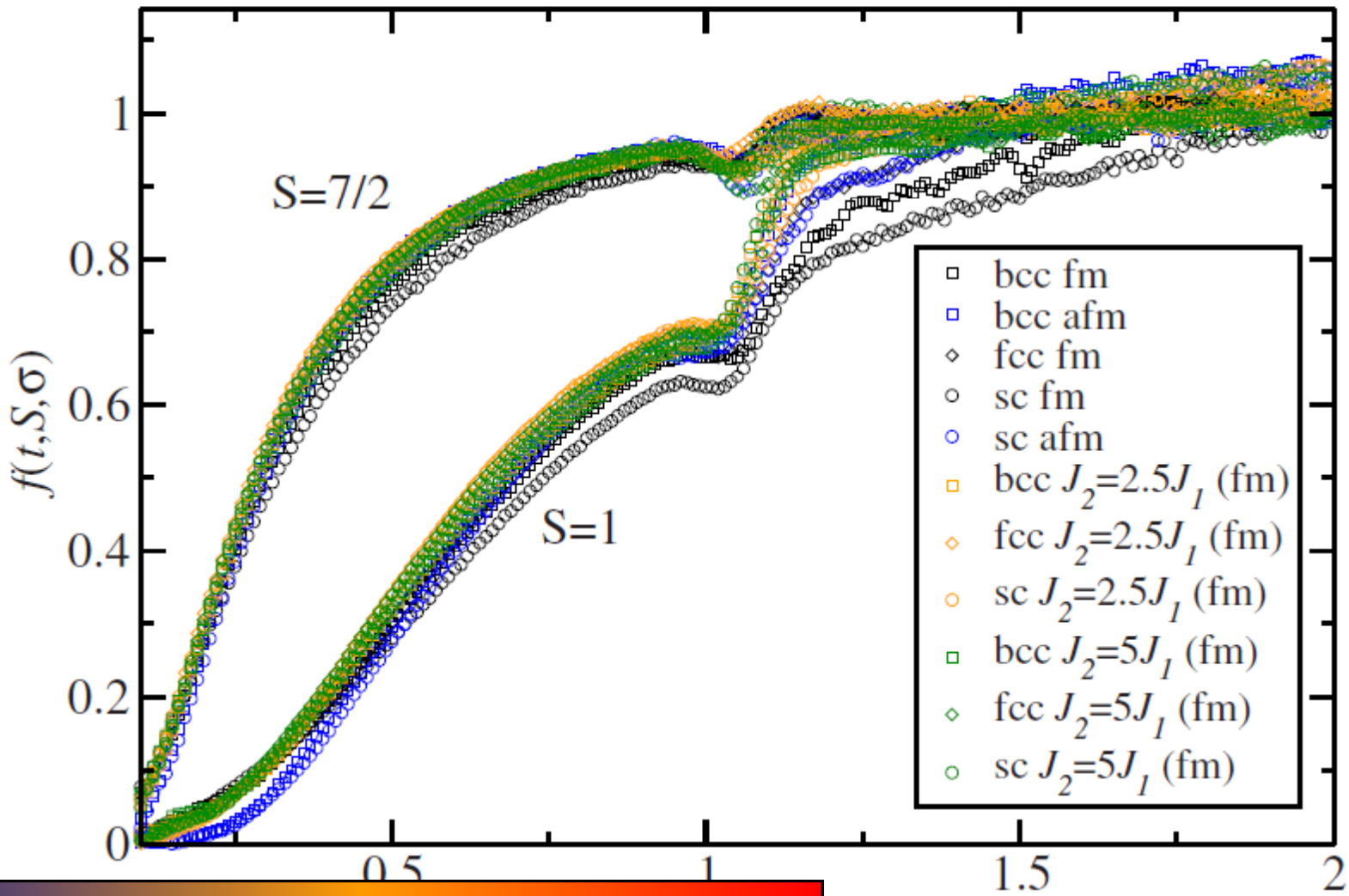
$$\langle A \rangle_{QM} = \mathrm{Tr}\left\{ A e^{-\beta H} \right\} / \mathrm{Tr}\left\{ e^{-\beta H} \right\}$$

Classical solution

$\hat{S}_j$ continuous vectors

$$\langle A \rangle_{CL} = \sum_c A_c e^{-\beta E_c} / \sum_c e^{-\beta E_c}$$

Analysis of different MC approaches



$$f(t,S,\sigma) = C_V^{qm}(t,S) / C_V^{cl}(t)$$

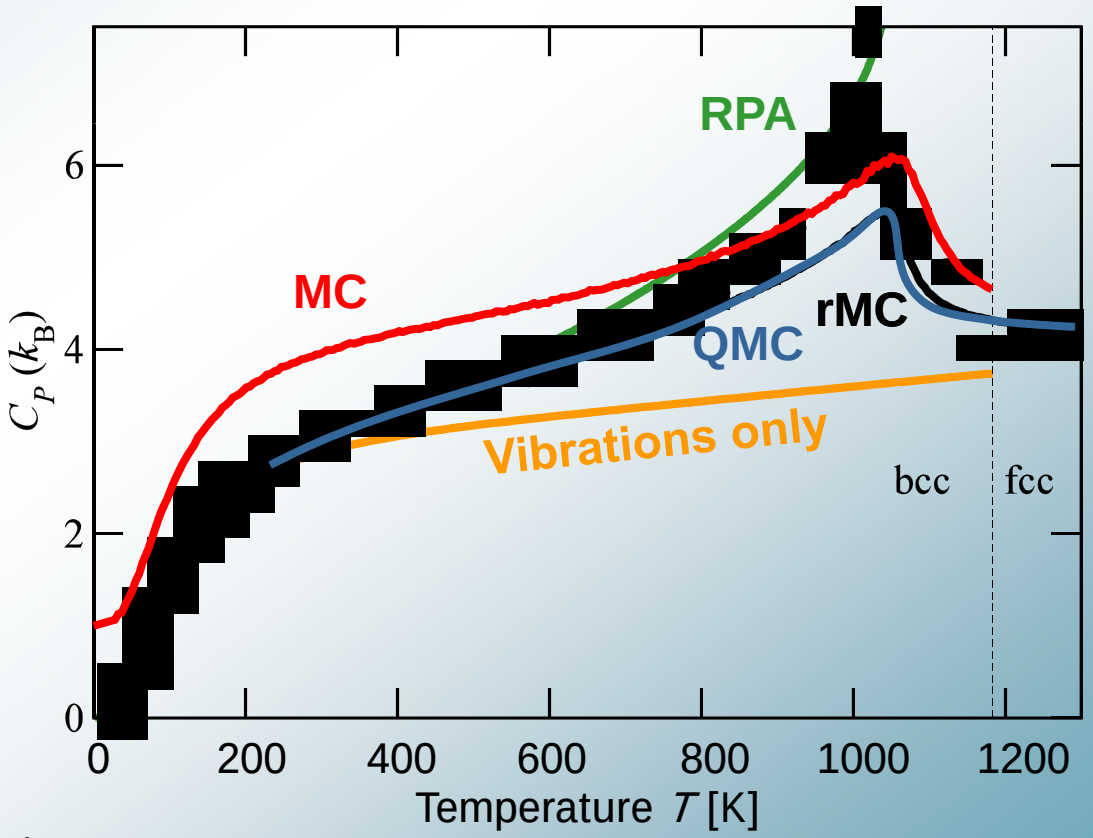
F. Körmann, TH, *et al.*, PRB **81**, 134425 (2010).

Magnetic entropy for bcc iron

Magnetic entropy important contribution to free energy of bcc iron
Shortcomings of approaches:
→ analytical approach (**RPA**): cannot cover complete temperature range
→ classical Monte-Carlo (**MC**): error at low temperatures due to quantum effects
→ quantum Monte-Carlo (**rMC**): negative sign problem

Effective nn approach (QMC):

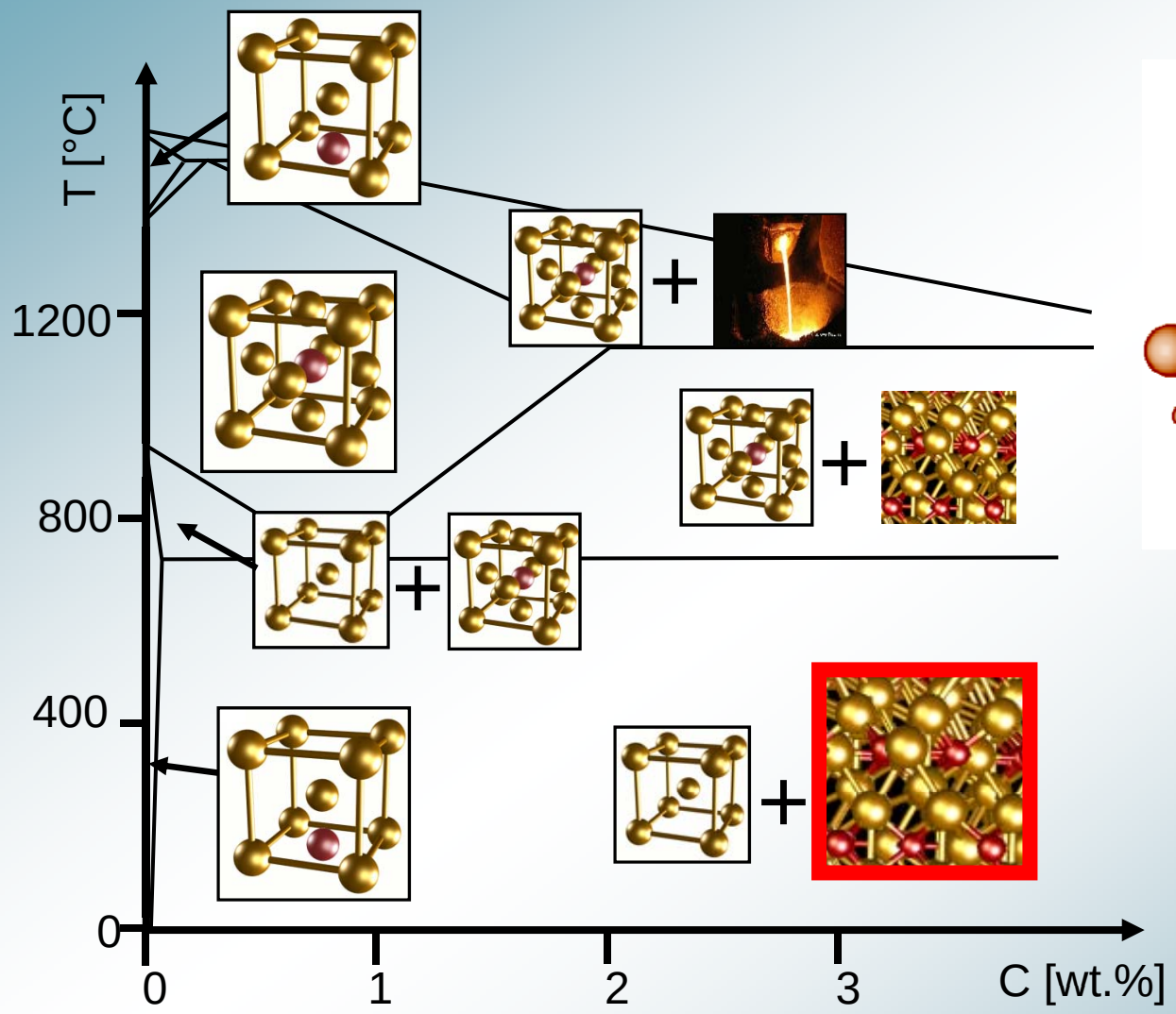
- Define an effective Heisenberg model with only nearest-neighbor interactions
- Ensure the correct critical temperature
- Perform QMC calculations for this model
- Derive free energy by intergration down from the exact high-temperature limit



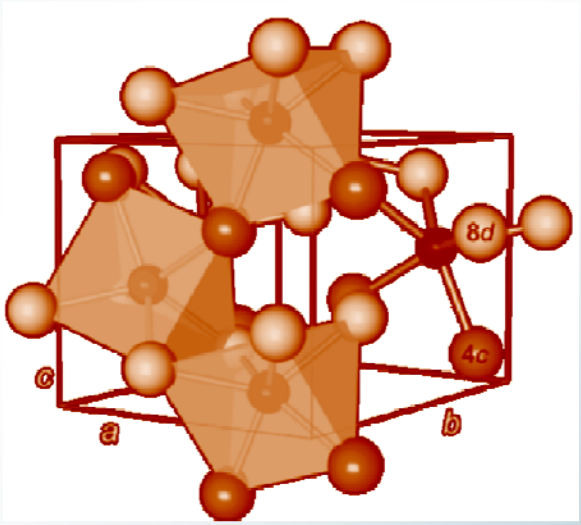
Numerics performed with ALPS simulation package.

F. Körmann, TH, *et al.*, PRB **81**, 134425 (2010).

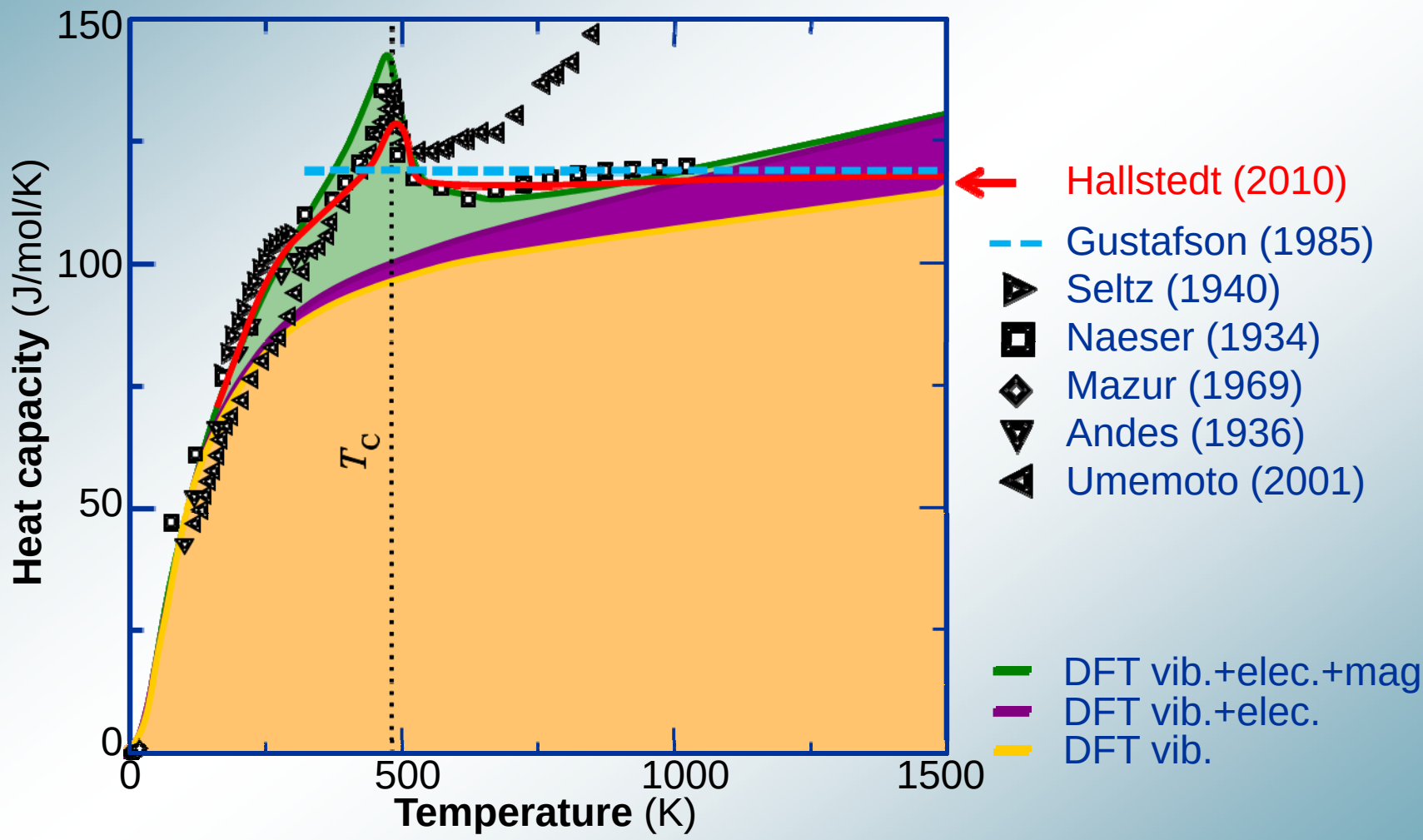
The Fe-C system



Cementite Fe_3C



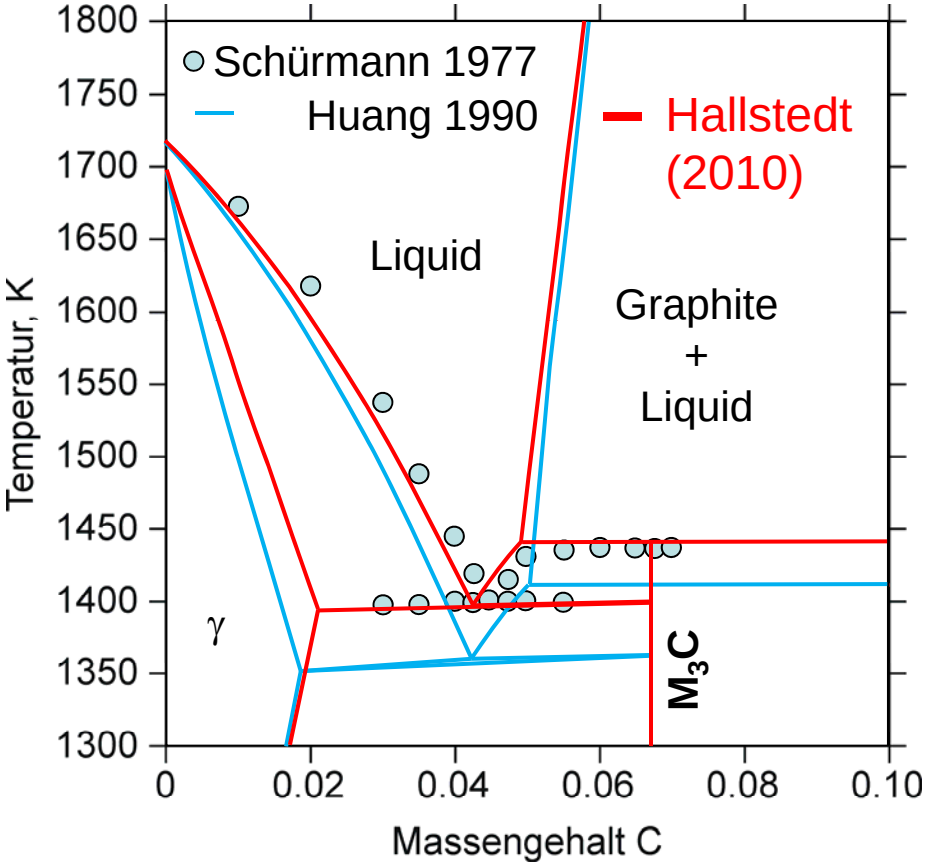
Heat capacity of Fe_3C



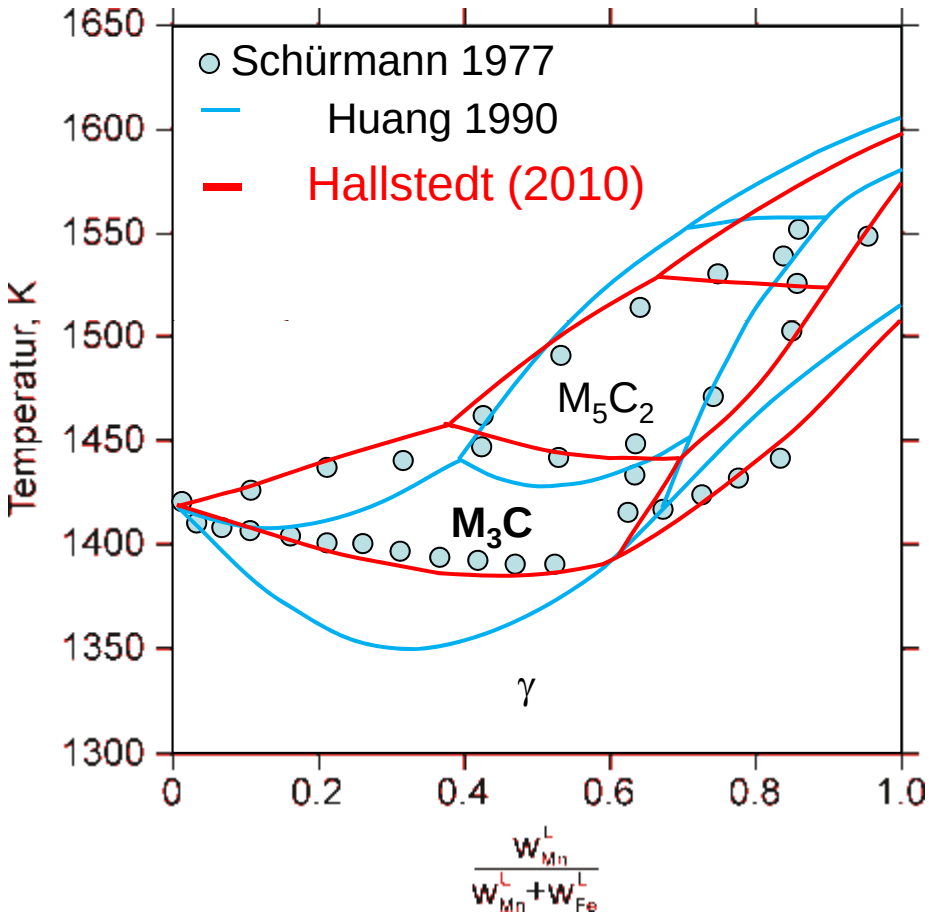
B. Hallstedt, D. Djurovic, J. von Appen, R. Dronskowski, A. Dick, F. Körmann, T. Hickel, J. Neugebauer, *Calphad*, **34**, 129 (2010).

Relevance for phase diagram

Phase diagram isopleth for 20% Mn



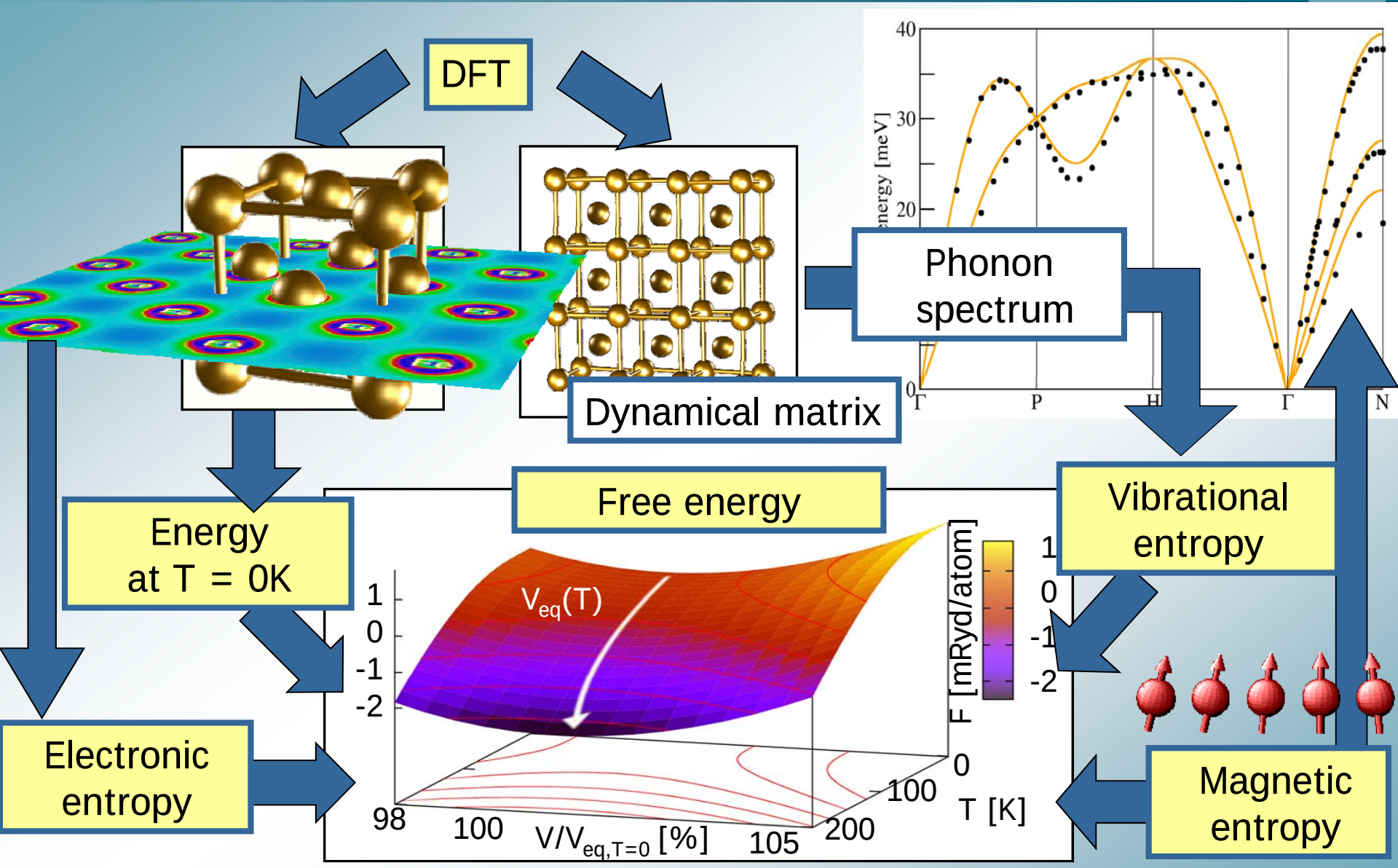
Liquidus surface



D. Djurovic, B. Hallstedt, J. v. Appen, R. Dronskowski: CALPHAD **34**, 279 (2010)

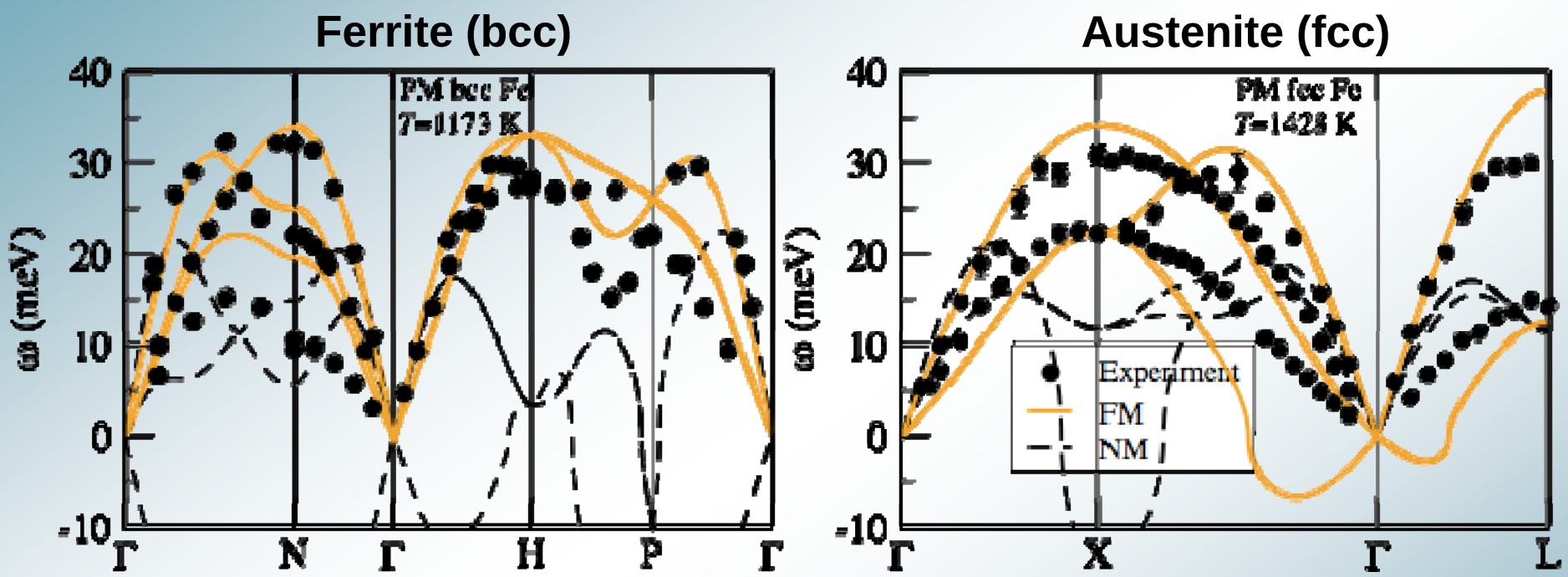
Dick *et al.* Phys. Rev. B **84**, 125101 (2011)

Ab initio for finite temperatures



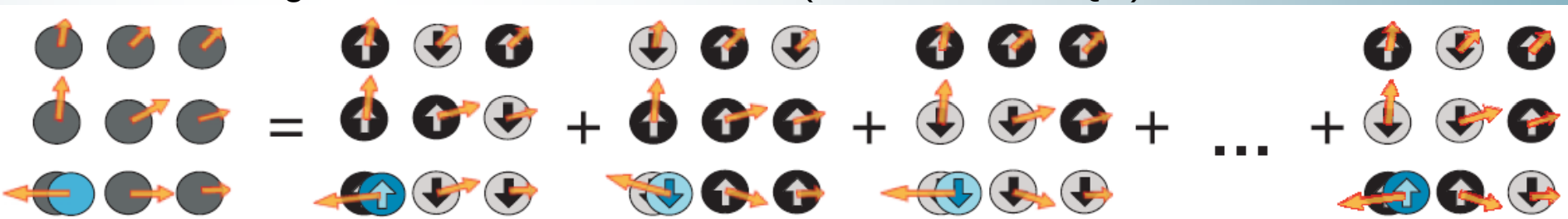
Influence of magnetism on phonons of Fe

Phonons of iron above the Curie temperature



Spin space average (SSA) technique

Adiabatic approximation: assuming magnetic degree of freedom faster than atomic motion, i.e. at high T each atom „feels“ same (disordered = SQS) environment



F. Körmann *et al.*, PRB **85**, 125104 (2012).

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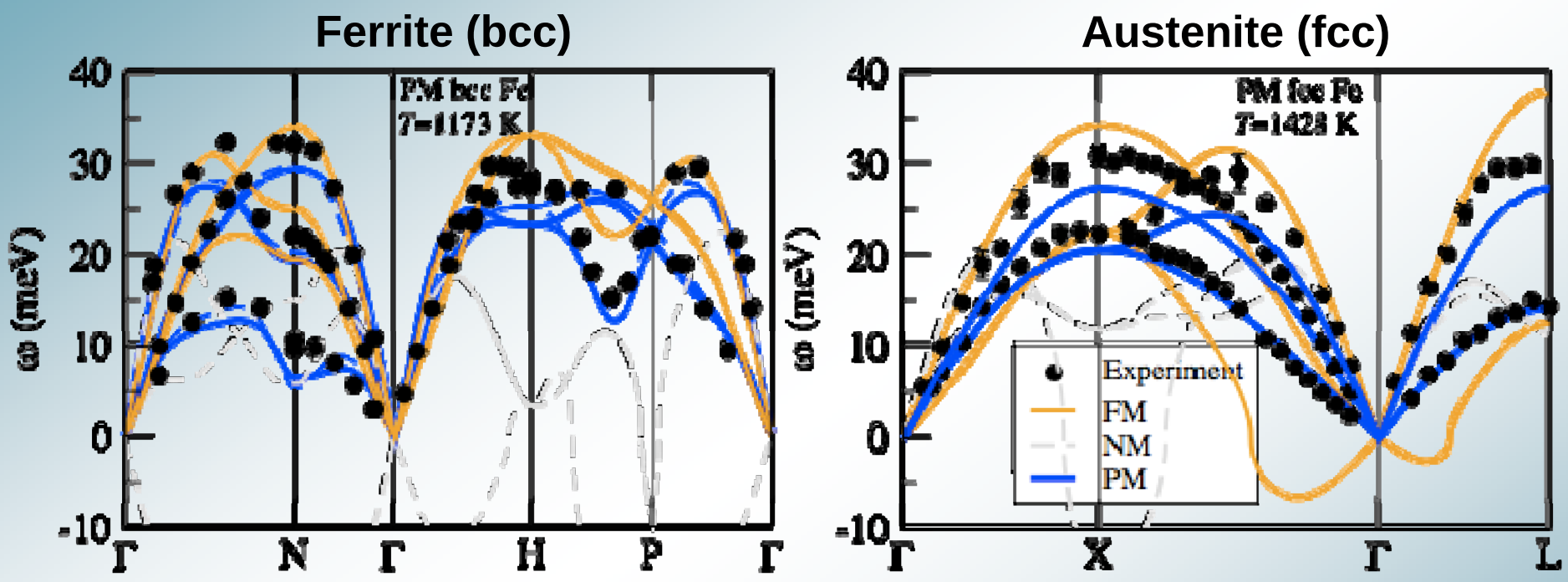
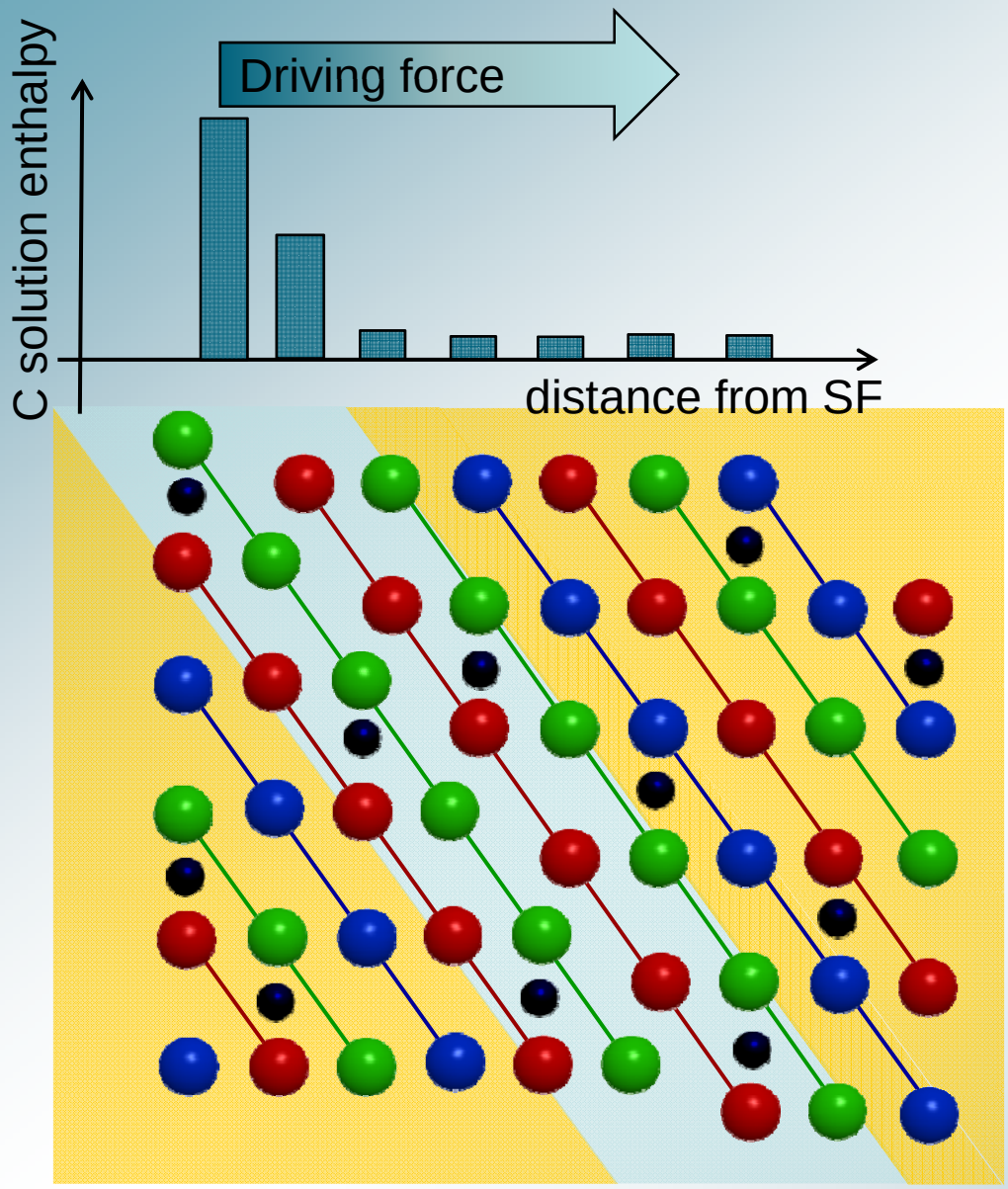


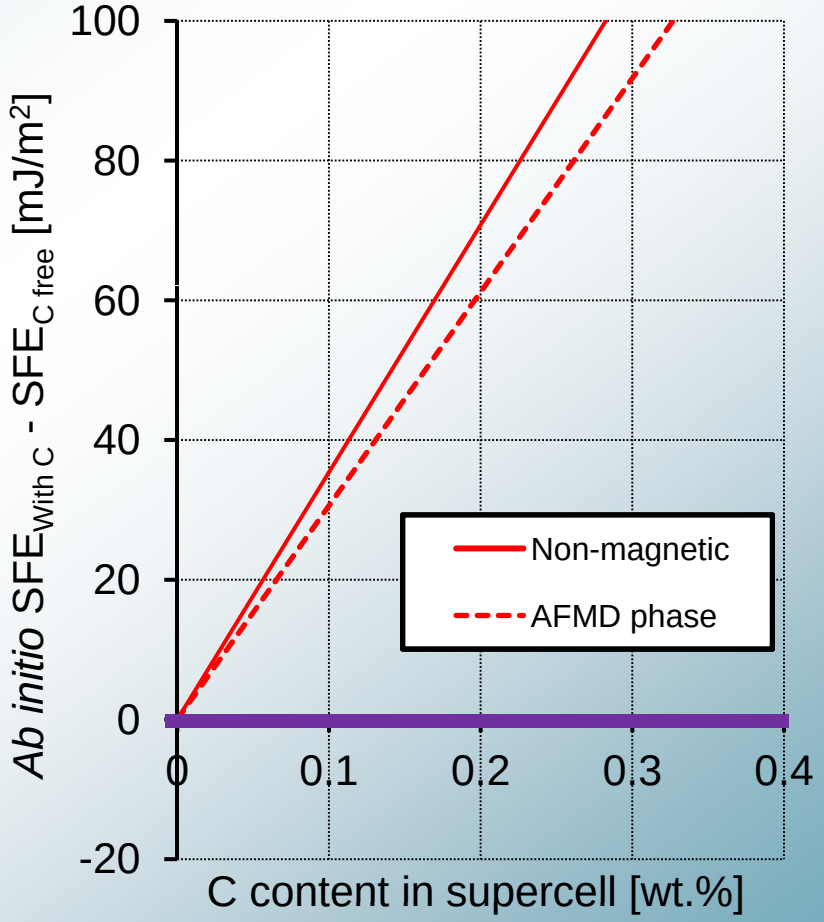
- PM calculated with newly developed spin space average (SSA) technique,
  → average over multiple spin Born-Oppenheimer surfaces

- Stability of fcc Fe due to spin excitations

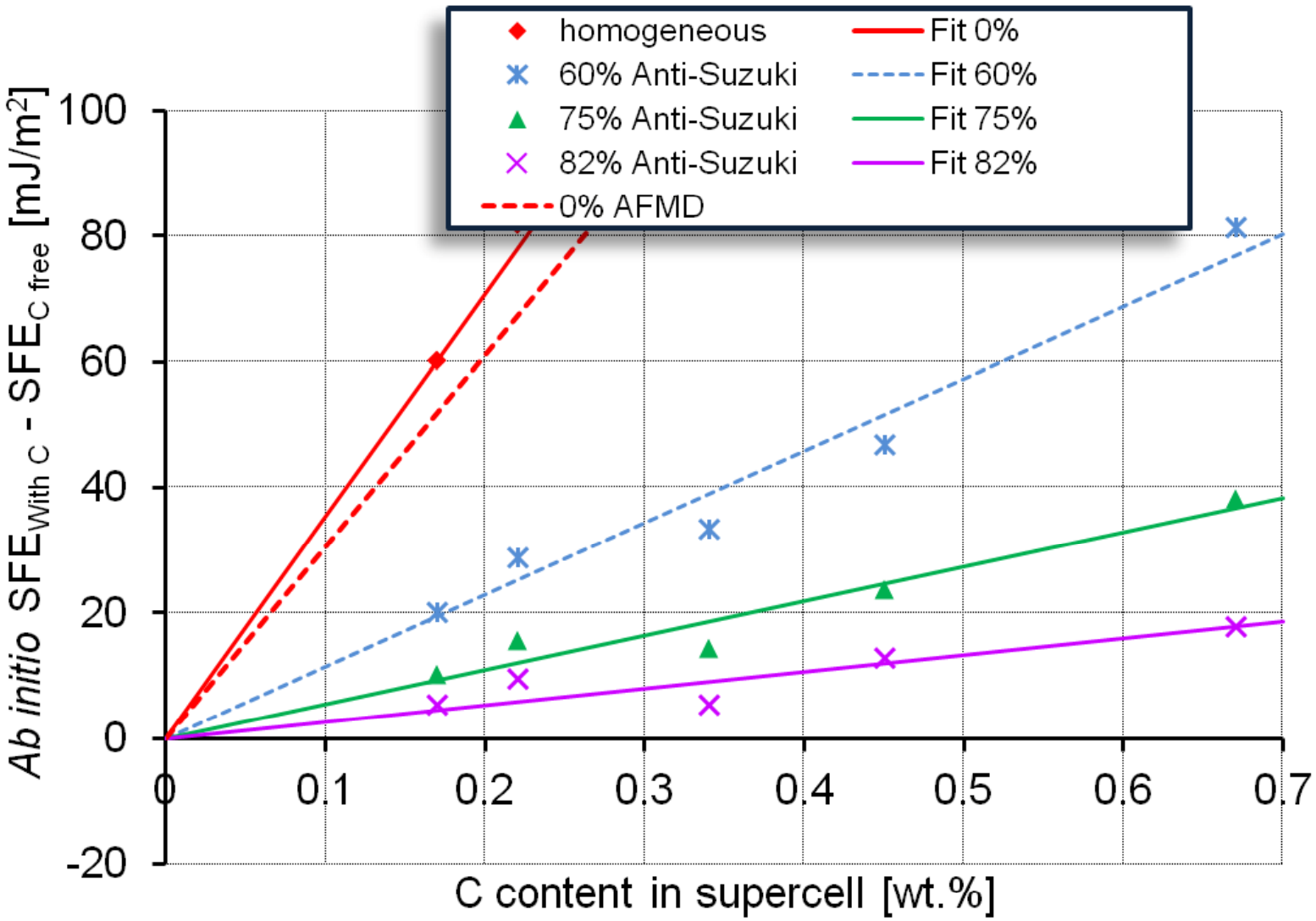
F. Körmann *et al.*, PRB **85**, 125104 (2012).

Dependence on C content

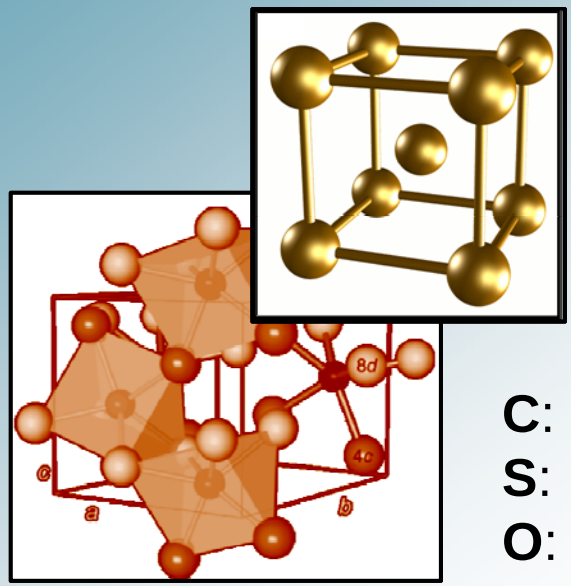




Dependence on C content

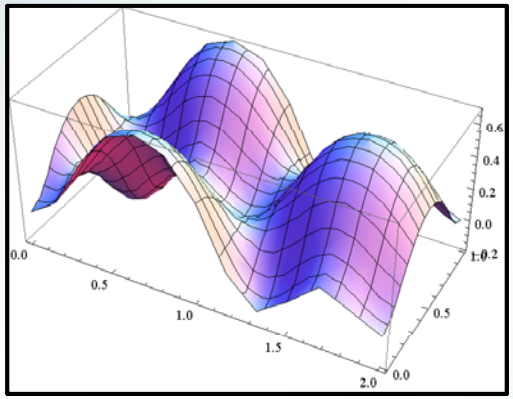


Conclusions (Challenge, Status, Outlook)



C: Small energy difference between phases
S: Quantum-effects in magnetic data
S: Phonon spectra in PM regime
O: Accurate description of phase transition

C: Magnetic structure with inequivalent atoms
S: Identification of reliable experimental C_p data
O: Formation energy at T = 0K



C: Strong dependence of SFE on magnetic order
S: Application to Fe-Mn alloys
O: Generalization to PM configurations