# Magneto-structural coupling

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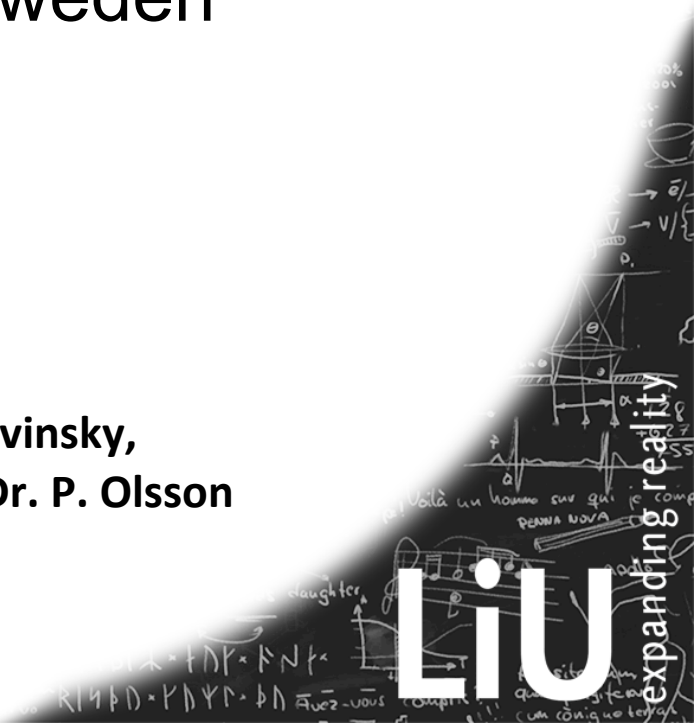
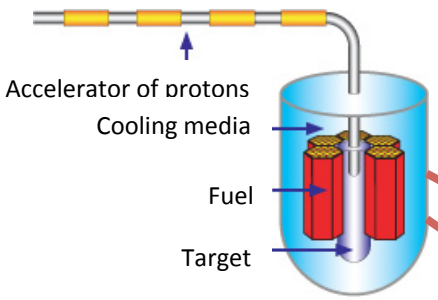
**Prof. A. V. Ruban, Prof. Yu. N. Gornostyrev, Prof. L. Dubrovinsky, Prof. N. Dubrovinskaya, Prof. L. Vitos, Prof. J. Wallenius, Dr. P. Olsson**
Swedish Foundation for Strategic Research (SSF),
Swedish Research Council (VR),
Swedish National Infrastructure for Computing (SNIC)



# Contents:

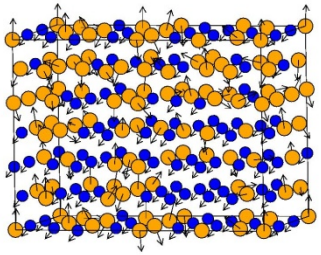
- Introduction. A*b initio* simulations of Fe-based alloys: theory of magneto-structural coupling and a new possibility for steel design.
- Mixing enthalpies of paramagnetic and ferromagnetic Fe-Cr-(X) alloys.
- Solution enthalpies of substitutional and interstitial impurities (C, N, V, Nb) in austenite.
- Magnetism and phase transition in $FeNi_3$ permalloy.
- Tuning magnetic interactions in high-pressure high-temperature synthesis of B2-$Fe_2Si$
- Conclusions

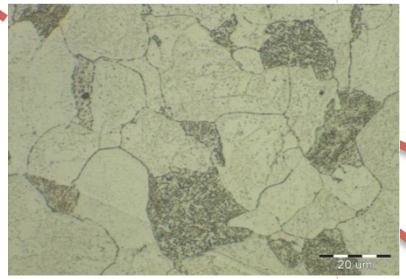
# Multiscale approach to materials modeling



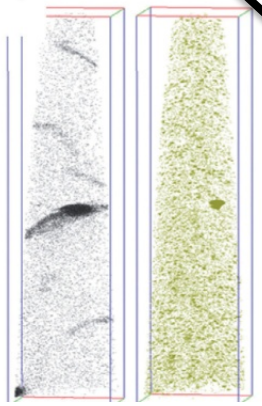

**Computed data-bases of materials parameters**

**Electrons (quantum mechanics)**
**≅ 2-100 Angstroms**
**Properties**
- Total energies
- Elastic properties
- Interatomic interactions

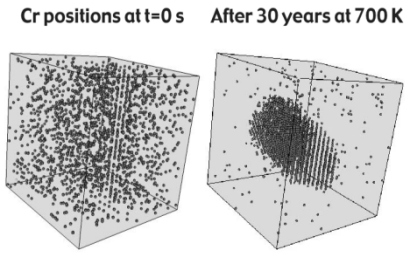




**Microstructure**
**- Grains**
**≅ 1 – 10 mm**
**Properties**
- High cycle fatigue
- Ductility



**Microstructure**
**-Phases**
**(continuum theories)**
**≅ 3-100 nanometers**
**Properties**
- Yield strength
- Ultimate tensile strength
- Low cycle fatigue
- Ductility




**Atoms (statistical mechanics/ thermodynamics)**
**≅ 10-100 Angstroms**
**Properties**
- Thermal Growth
- Free energies

**Goal:**
**theoretical calculations of relevant parameters at realistic conditions with accuracy comparable to experiment.**

**Tools:**
**novel techniques based on Density Functional Theory (Nobel Prize 1998)**

# 1978

x – experiment

• - theory

Magnetic elements

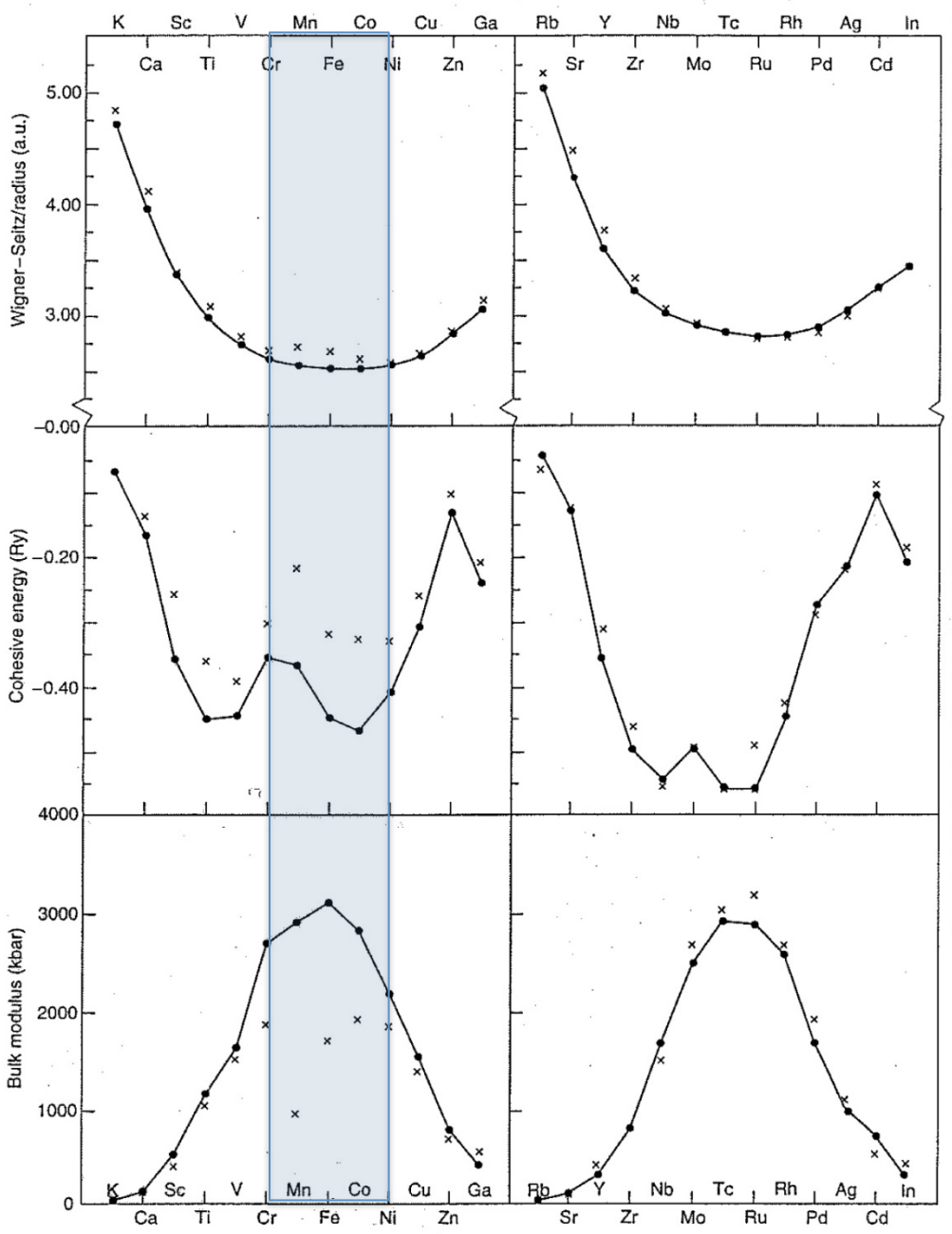
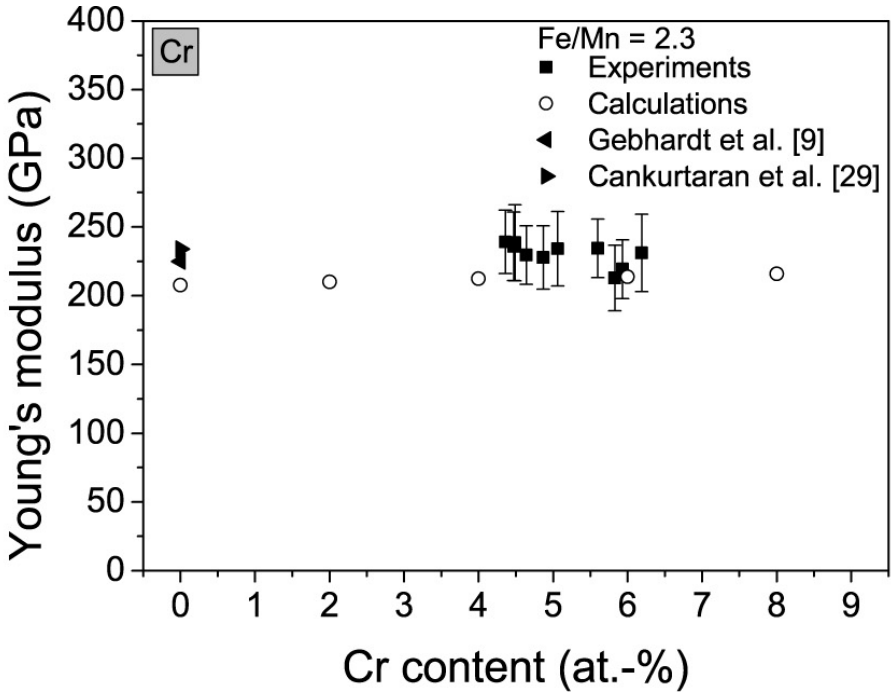
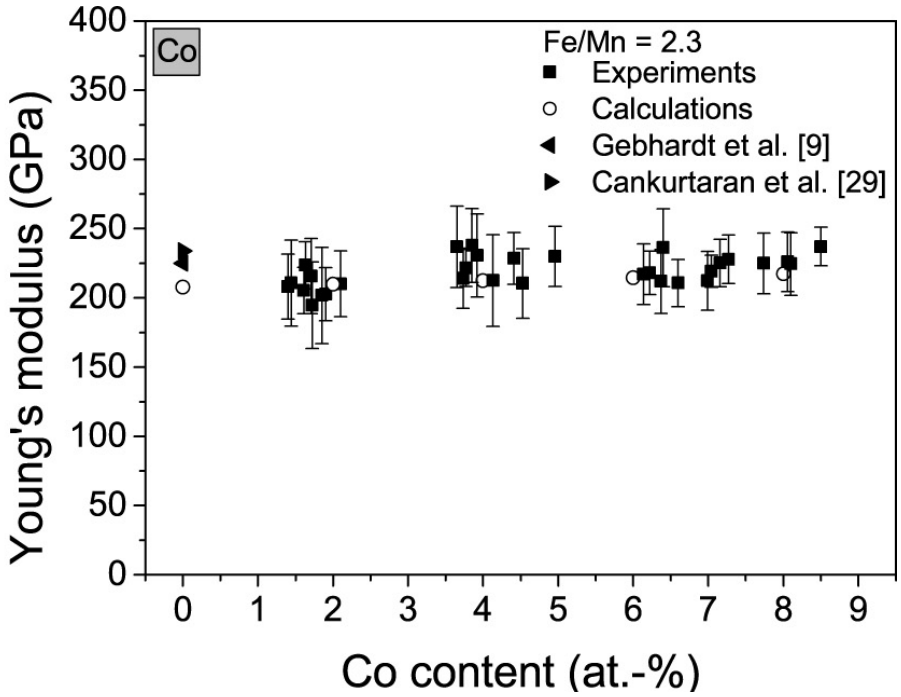


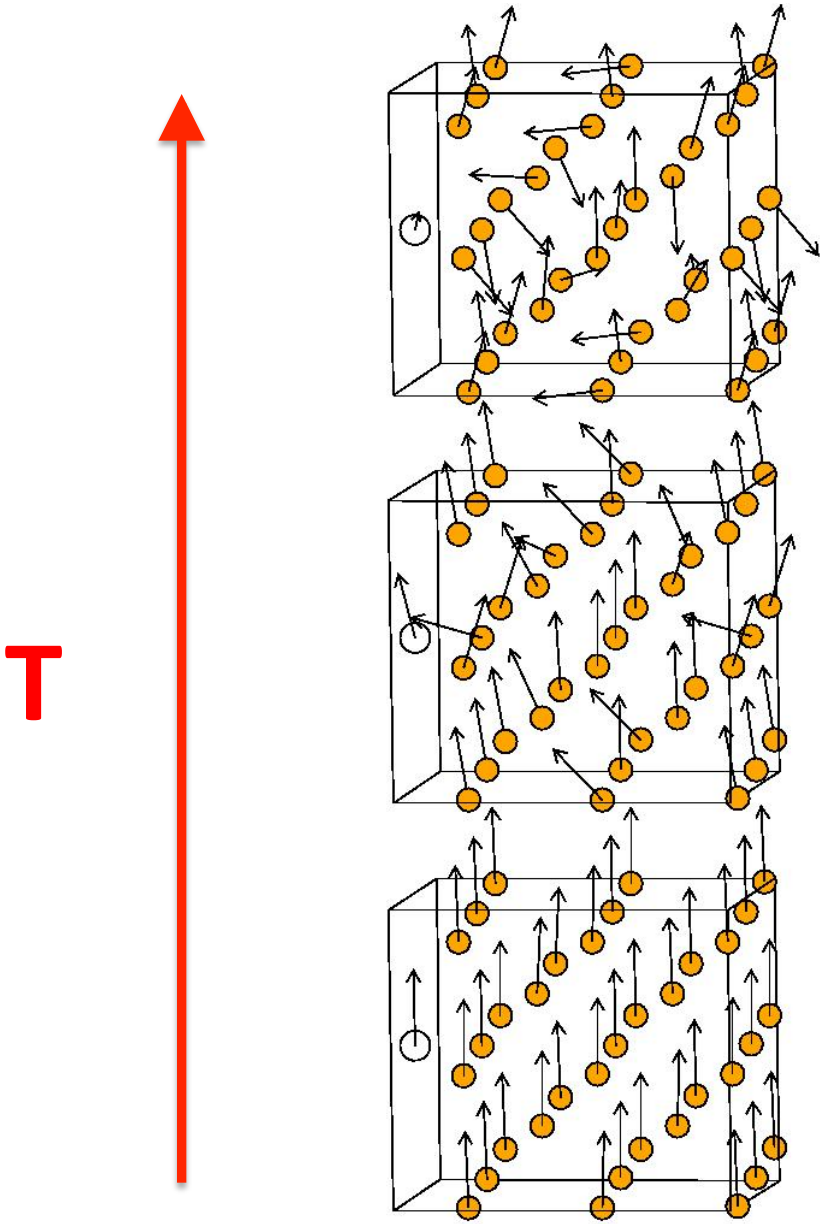

**Figure 2.3.** Calculated lattice constants and bulk moduli for the 3d and 4d series of transition metals. From Moruzzi, Janak, and Williams [106] (see also [107]).

# Young's modulus of Fe-Mn-X (X=Cr,Co) alloys

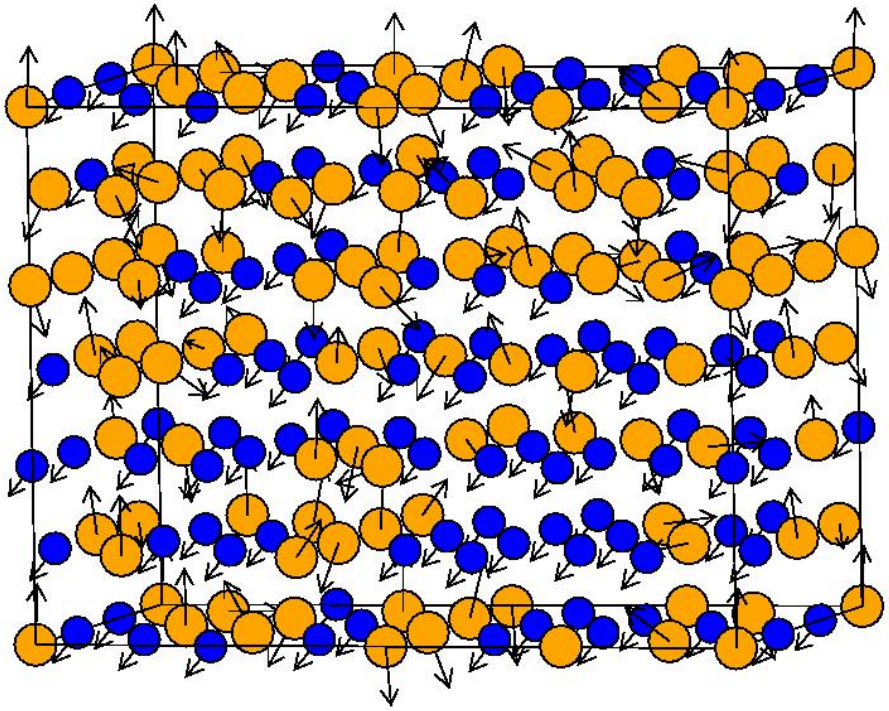


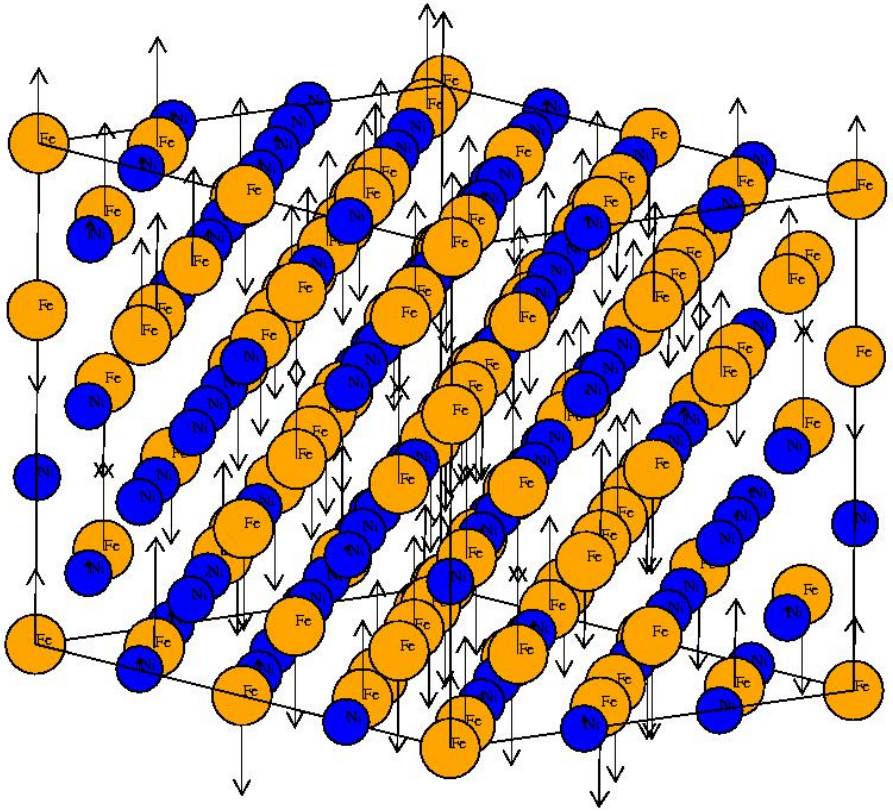




Calculated (open circles) and experimentally determined (filled rectangles) Young's modulus values for fcc Fe–Mn–Cr and Fe–Mn–Co with a Fe=Mn ratio of 2.3.
From S. Reeh, M. Kasprzak, C. D. Klusmann, F. Stalf**,** D. Music, M. Ekholm, I. A. Abrikosov, J. M. Schneider, J. Phys.: Condens. Matter **25**, 245401 (2013).


T

# Disordered Magnetism

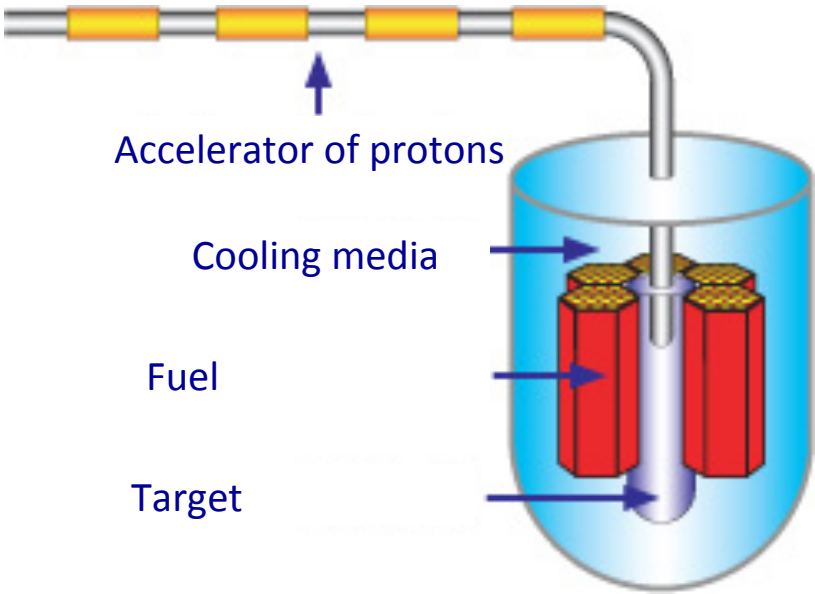


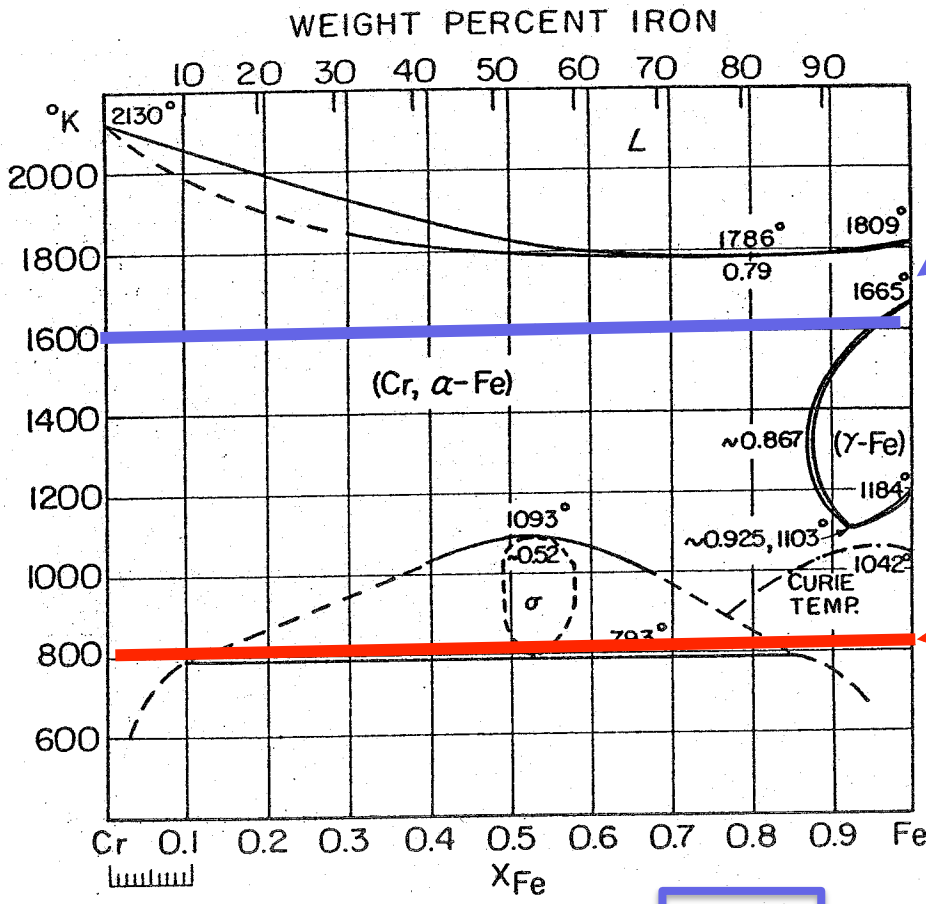



## Disordered Local Moment Model

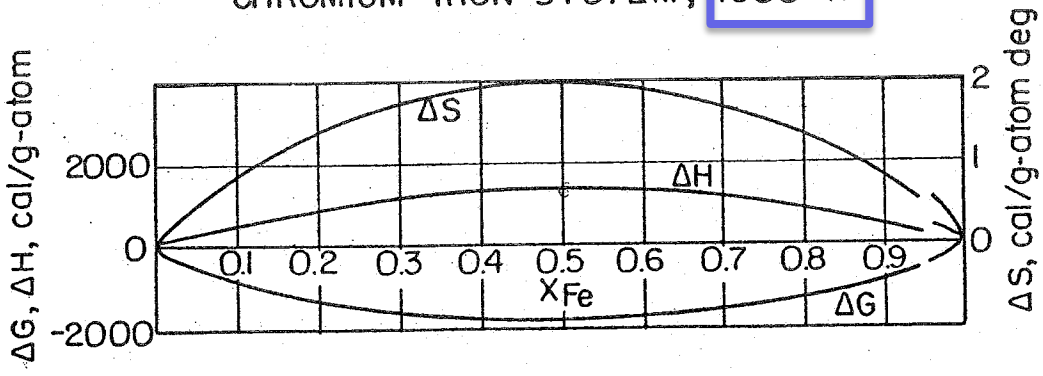
# Fe-Cr alloys

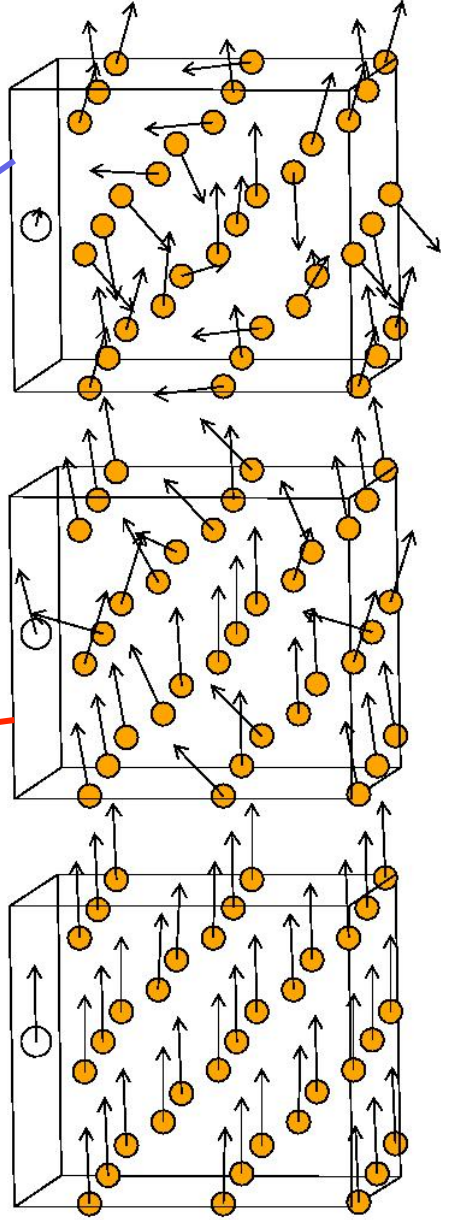
- Are the base for many important industrial steels
- Used as cladding material in fast neutron reactors
- Low  Cr steels, up to 10 % Cr, show:
  anomalous stability
  resistance to neutron
  radiation induced swelling
  corrosion resistance
  increased ductile to brittle
  transition temperature



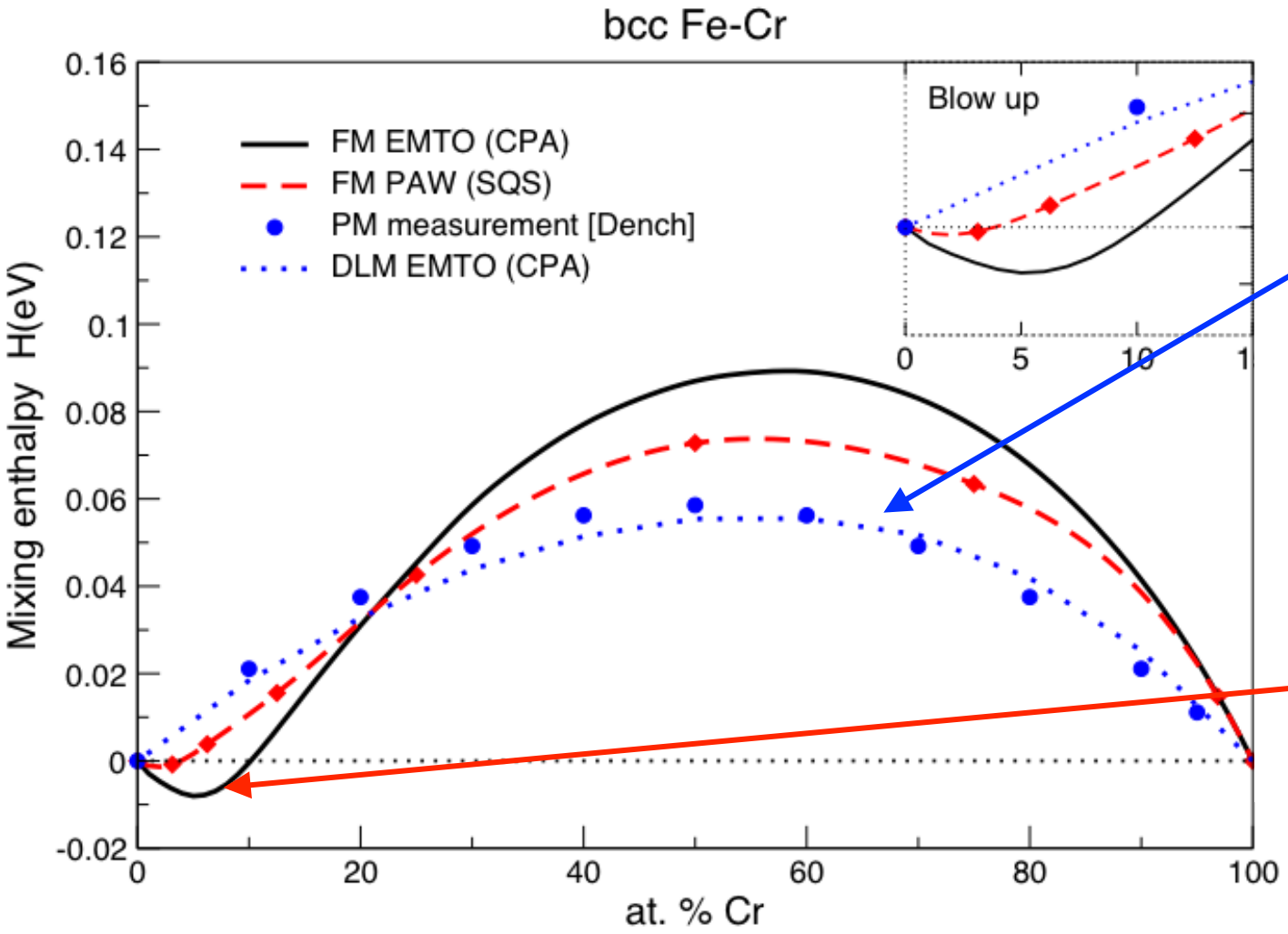


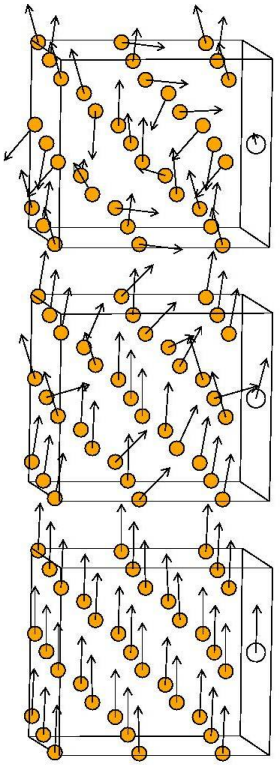

CHROMIUM -IRON SYSTEM, 1600 °K



# Mixing enthalpy of bcc Fe-Cr alloys

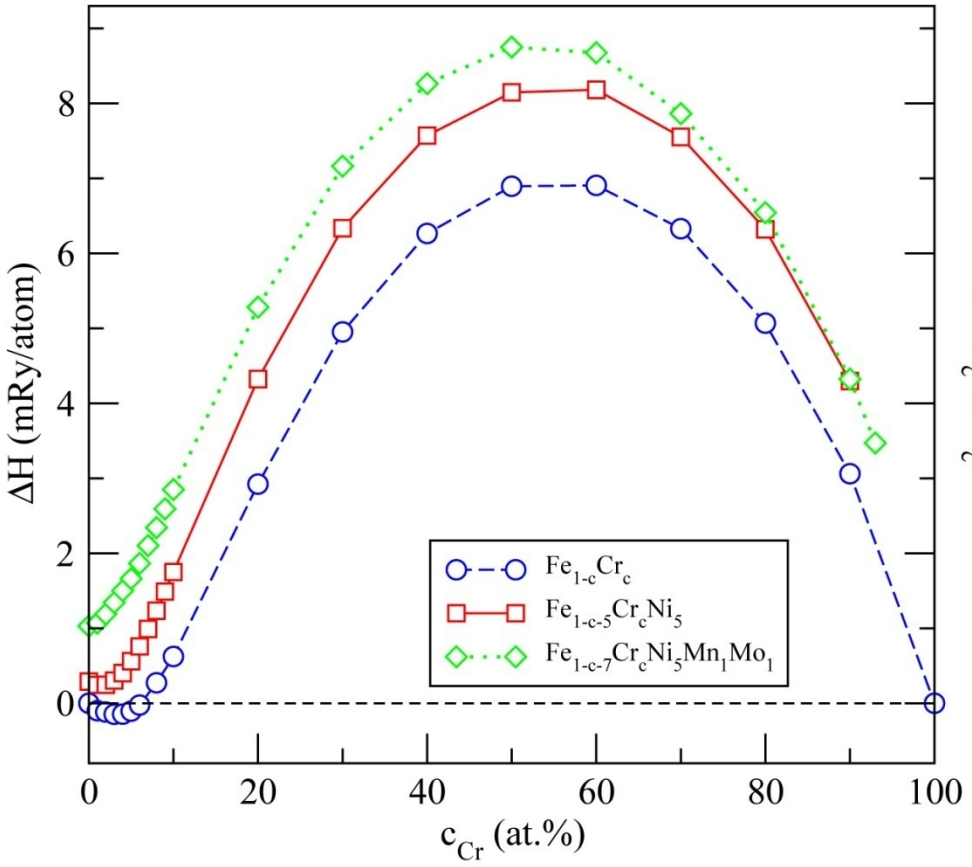


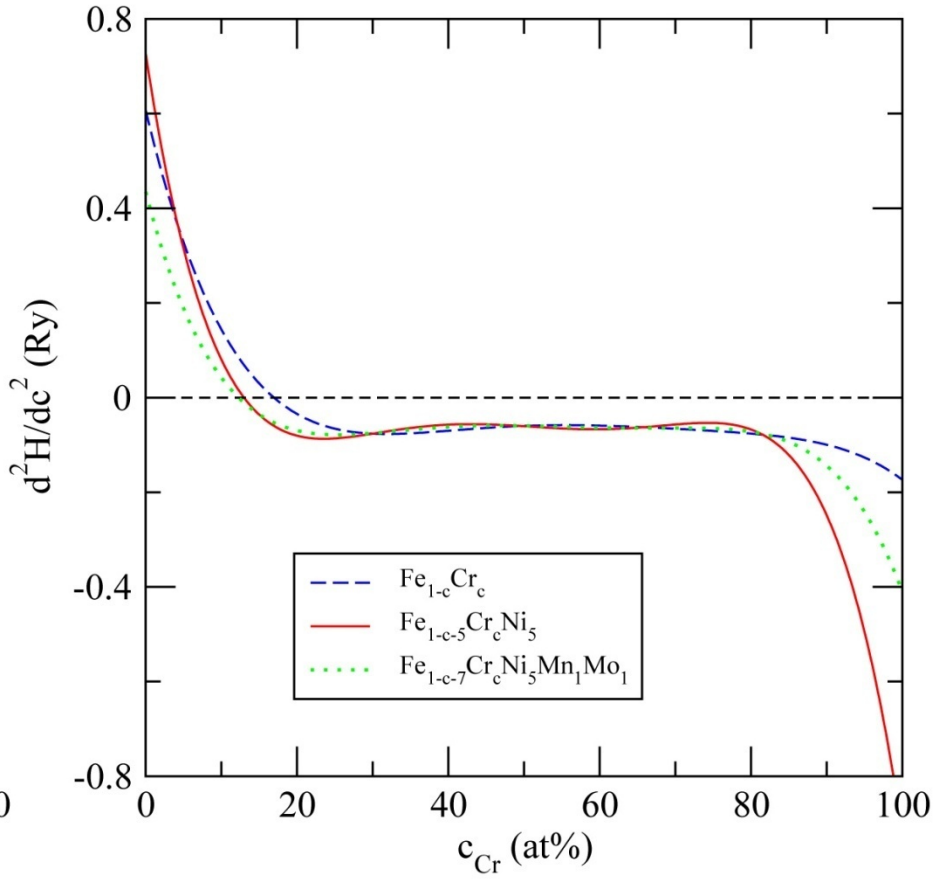




P. Olsson, I. A. Abrikosov, L. Vitos, and J. Wallenius, J. Nucl. Mater. 321, 84 (2003)

P. Olsson, I. A. Abrikosov, and J. Wallenius, Phys. Rev. B 73, 104416 (2006)

# Fe-Cr steels:
# Effect of multicomponent alloying



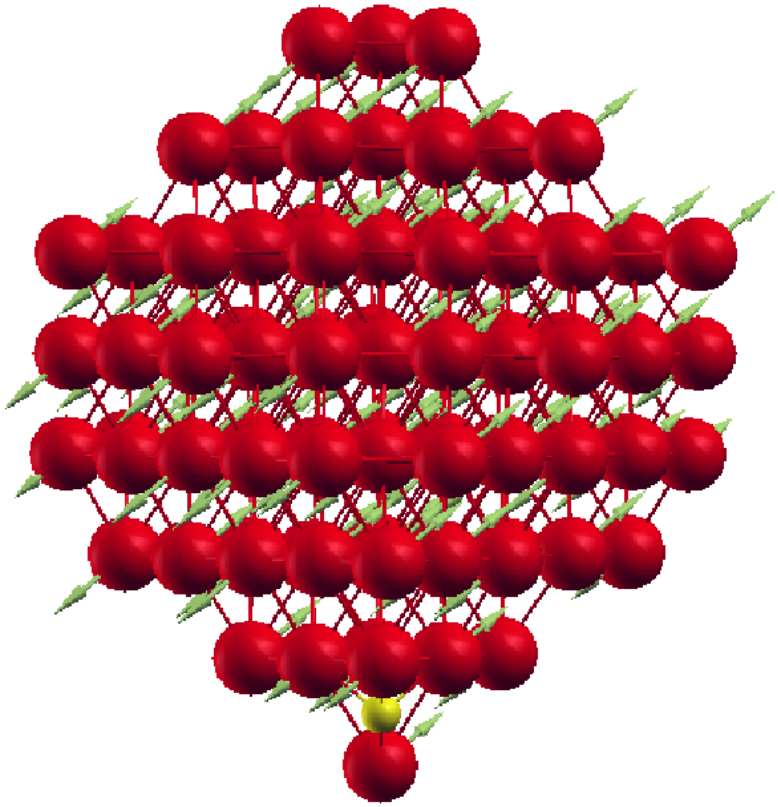

I. A. Abrikosov *et al.,* MRS Online Proceedings Library, 1514, mrsf12-1514-hh08-01 (2013). doi:10.1557/opl.2013.43

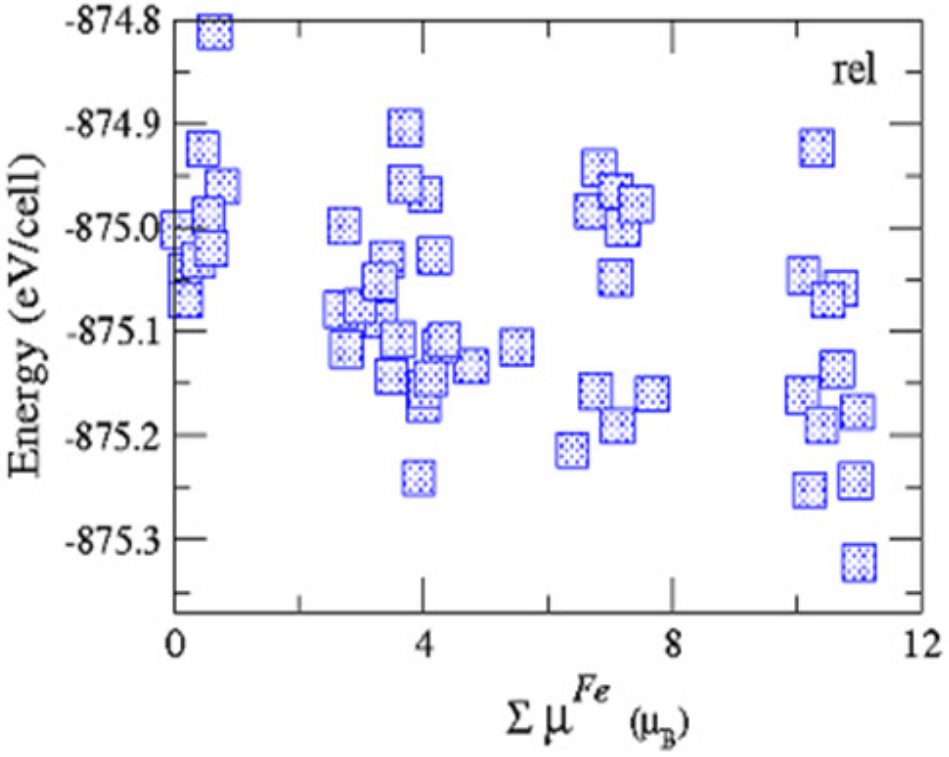
# C, N, Nb and V impurity solution energies in austenite

- Substitutional (Nb, V) and interstitial impurities (C, N) play an important role in carbonitride formation in pipe steel.
- Paramagnetic fcc Fe is an itinerant system.
- Magnetic interactions in fcc Fe are known to be long-ranged, at least in its magnetically ordered state

  [A. V. Ruban, M. I. Katsnelson, W. Olovsson, S. I. Simak, and I. A. Abrikosov, Phys. Rev. B **71**, 054402 (2005)].
- We deal with high-temperature paramagnetic state.
- Local lattice relaxations are expected to be very important, especially for interstitial impurities

# Structural and magnetic model






Supercell realization of the disordered local moment model using the magnetic sampling method [B. Alling, T. Marten, and I. A. Abrikosov, Phys. Rev. B **82**, 184430, (2010)]

Calculated energies of supercells with different impurity positions versus the total magnetic moment of Fe atoms located in the first coordination shell of the impurity.

A. V. Ponomareva, Yu.N. Gornostyrev, and I. A. Abrikosov, in manuscript.

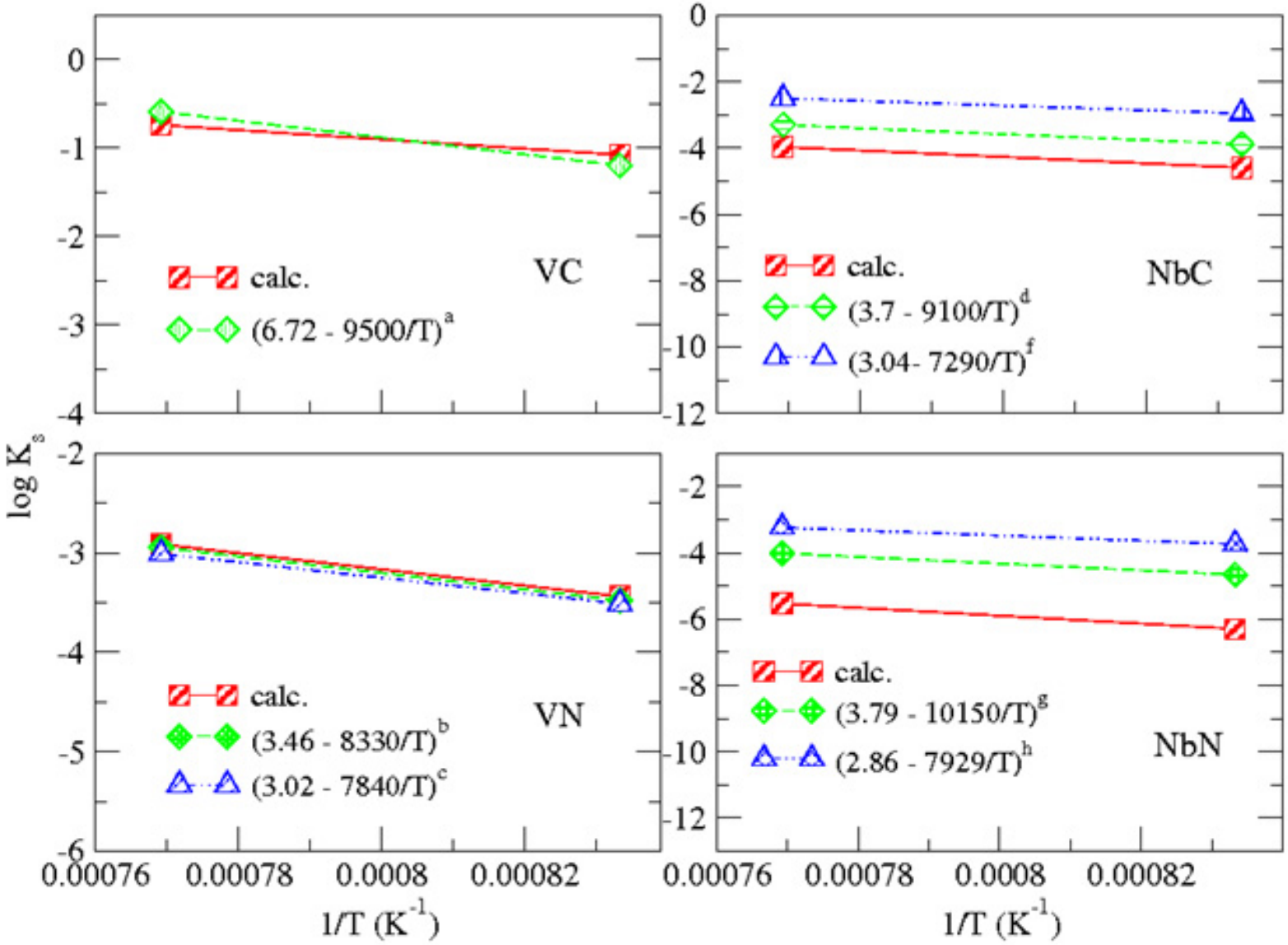
# Theoretical and experimental values of impurity solution energy in fcc iron (in eV)

| | Theory | | Experiment |
|---|---|---|---|
| | ideal positions | relaxed positions | |
| FeC | 1.50 | 0.20 | 0.40[Lobo_1976]<br>0.43 [Smith_1946]<br>0.44 [Chipman_1972]<br>0.41 [Ellis_1963] |
| FeN | 0.60 | -0.39 | -0.18 [Bouchard_1999, Hillert_1975] |
| FeV | -0.20 | -0.24 | |
| FeNb | 1.2 | 0.36 | |

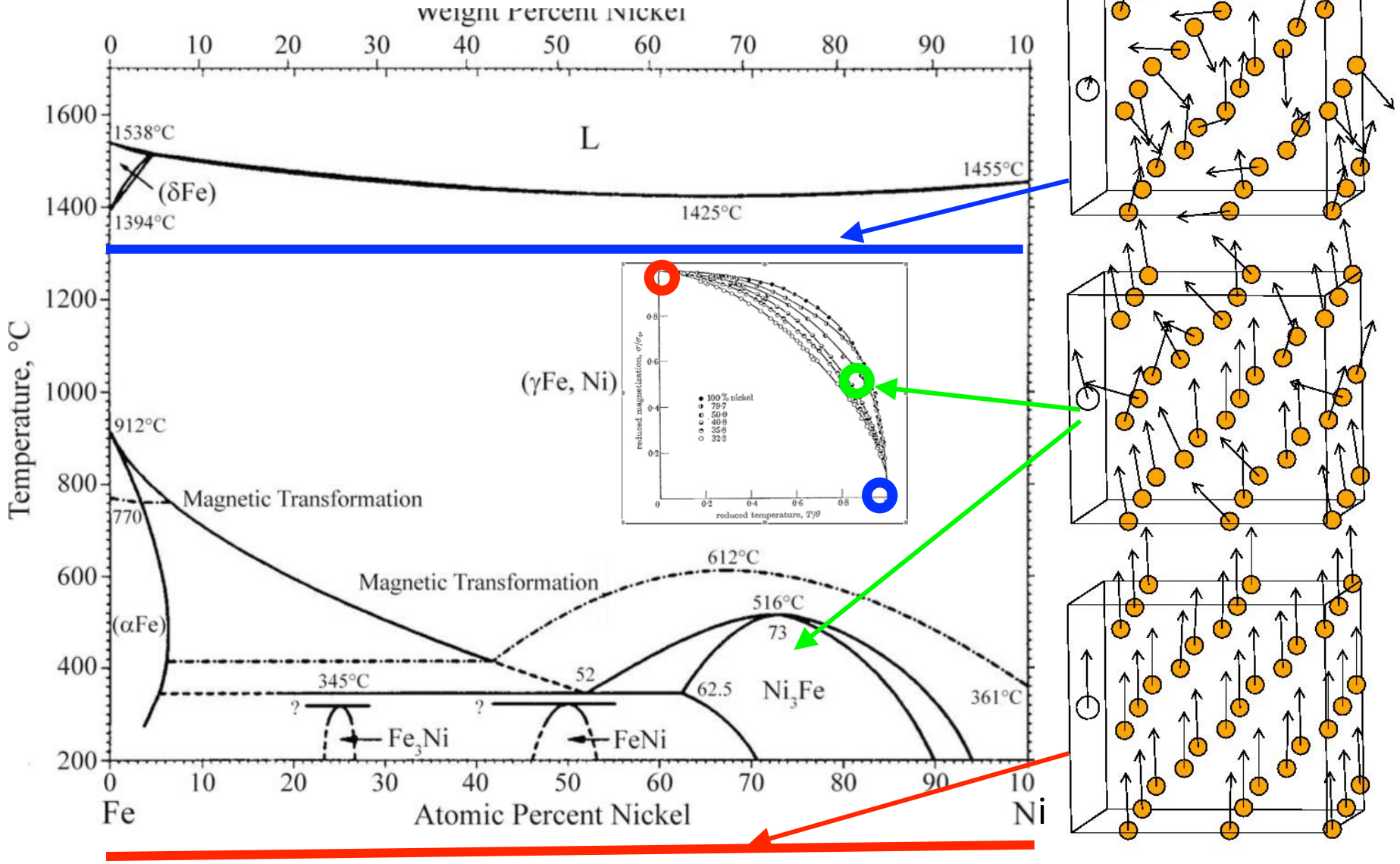
A. V. Ponomareva, Yu.N. Gornostyrev, and I. A. Abrikosov, in manuscript.

# Solubility product of VC, VN, NbC, NbN in austenite.

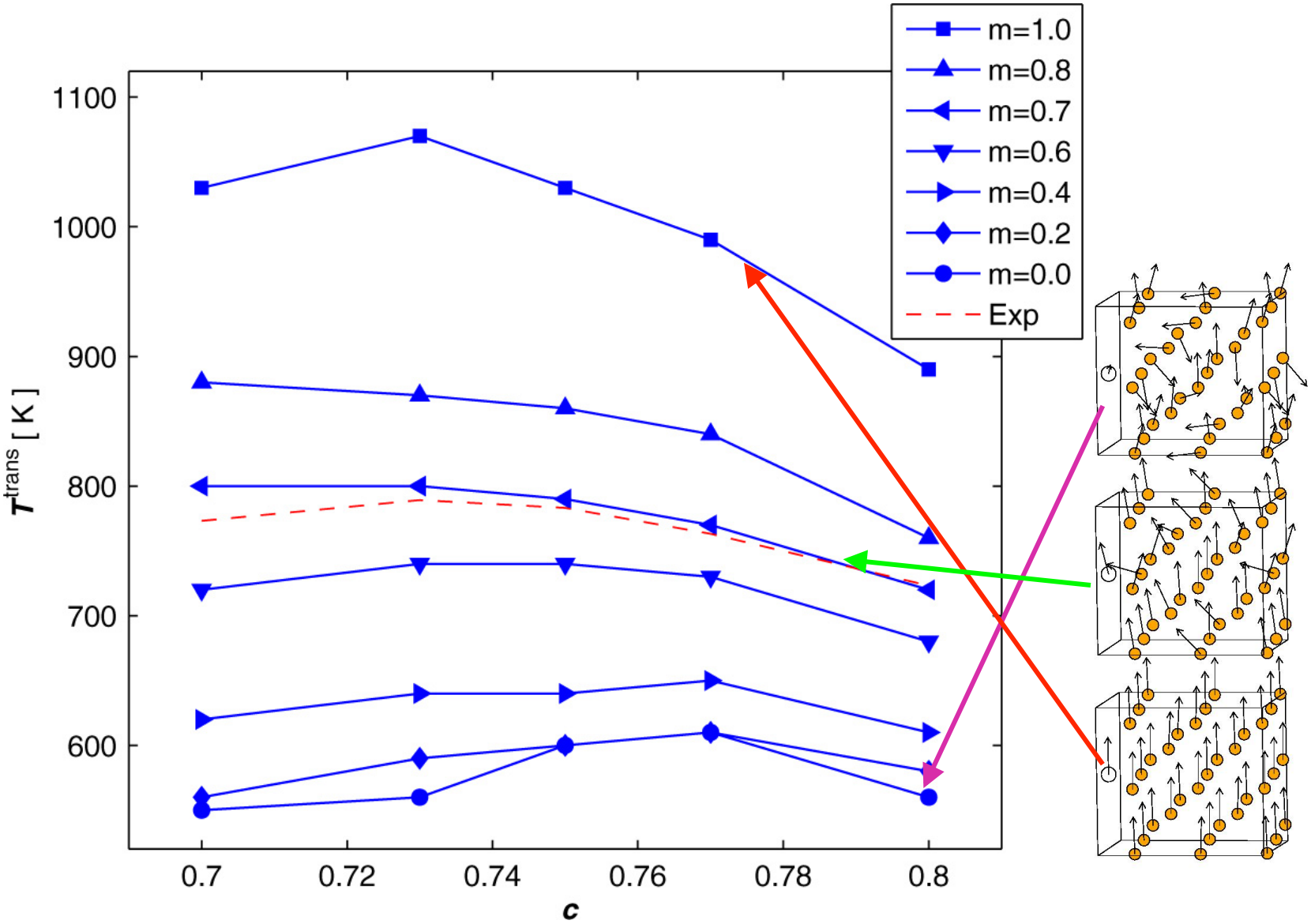
$$\log K_s \approx \log\left(\frac{10^4 m_{Me} m_X}{m_{Fe}^2}\right) + \frac{[\Delta E_f(MeX) - \langle E_{sol}\rangle(Me) - \langle E_{sol}\rangle(X)]}{\ln 10 \cdot k_B \cdot T}$$



# Experimental Fe-Ni phase diagram

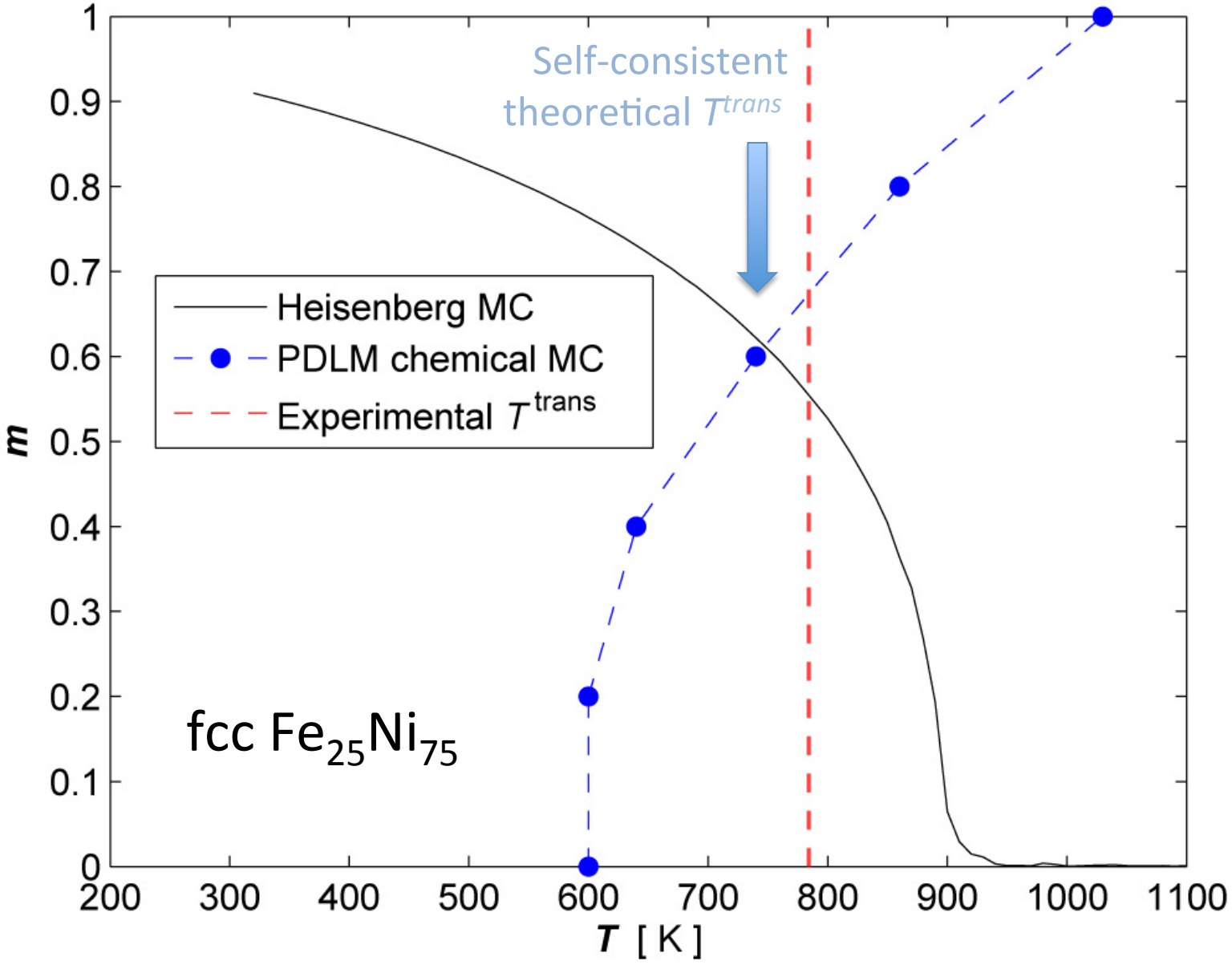


# Ni-rich part of Fe-Ni phase diagram calculated for different magnetic states of the system




M. Ekholm, …, IAA, Phys. Rev. Lett. **105**, 167208 (2010)

# *Ab initio* order-disorder transition temperature for fcc $Fe_{25}Ni_{75}$

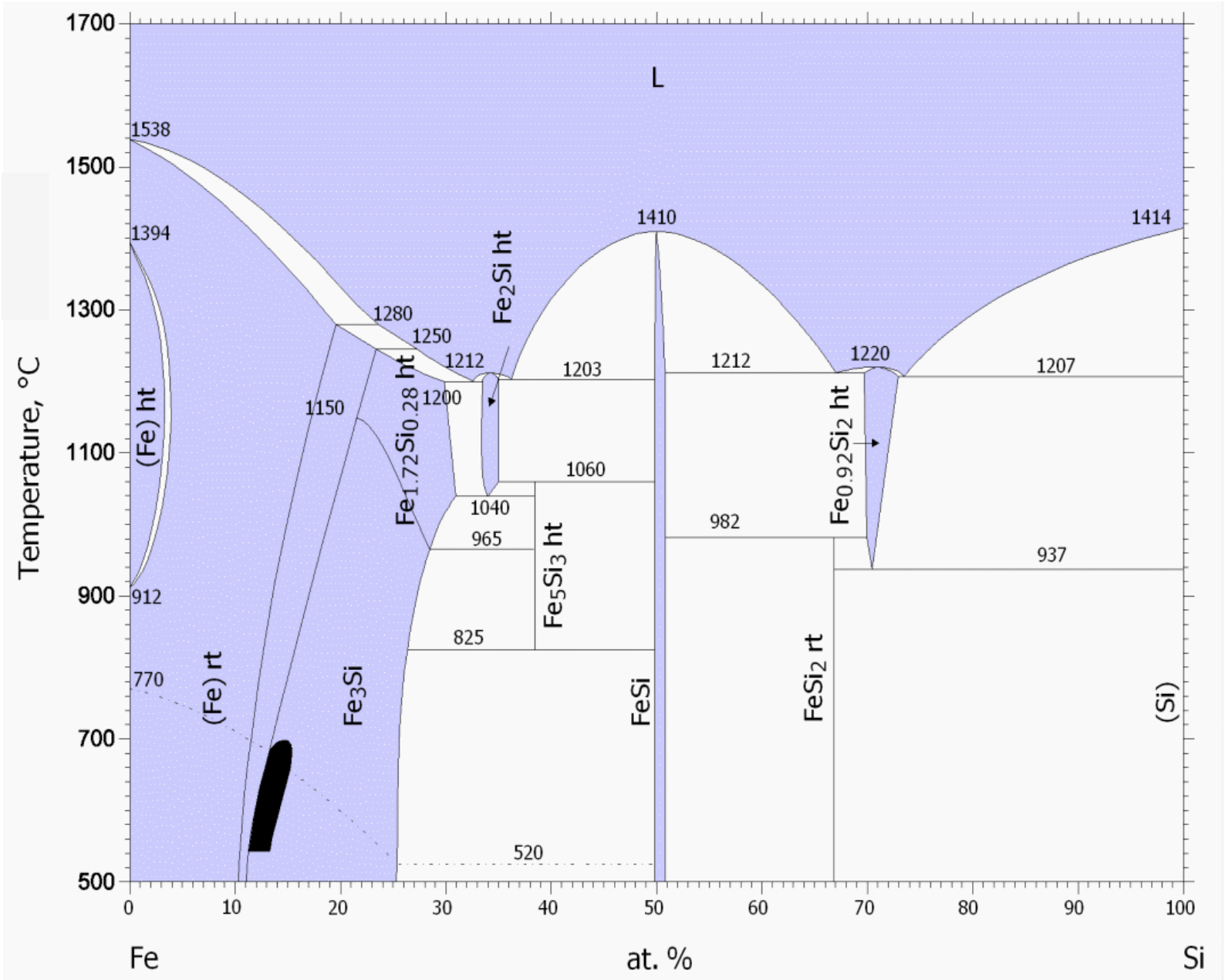



M. Ekholm, …, IAA, Phys. Rev. Lett. **105**, 167208 (2010)

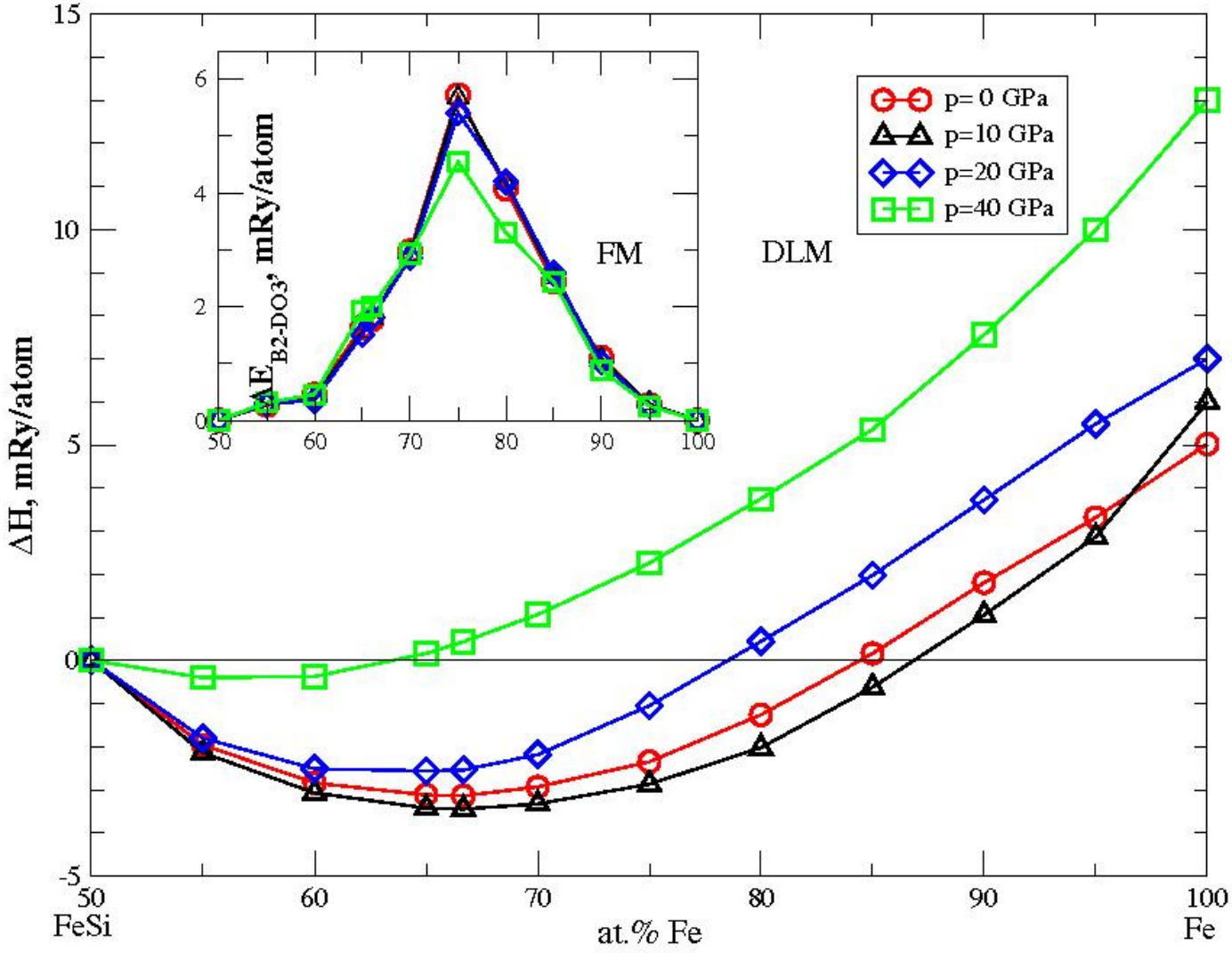
# Fe-Si system:

- At ambient pressure the phase diagram of Fe-Si system is complicated with different intermediate phases present.
- Moroni *et al. (*Phys. Rev. B **59,** 12860–12871, 1999) predicted the *B20* to *B2* structural transformation in FeSi at relatively low pressure.
- J.-F. Lin *et al.,* Iron-silicon alloy in Earth's core? *Science* **295**, 313-315 (2002).
- L. Dubrovinsky *et al.*, "Iron-silica interaction at extreme conditions and the electrically conducting layer at the base of Earth's mantle", Nature **422**, 58 (2003) .
- $Fe_3Si$ phase in the $DO_3$ structure is stabilized by the ferromagnetic order.
- Can a new phase in this system be synthesized by tuning its magnetic state?

# Fe-Si phase diagram

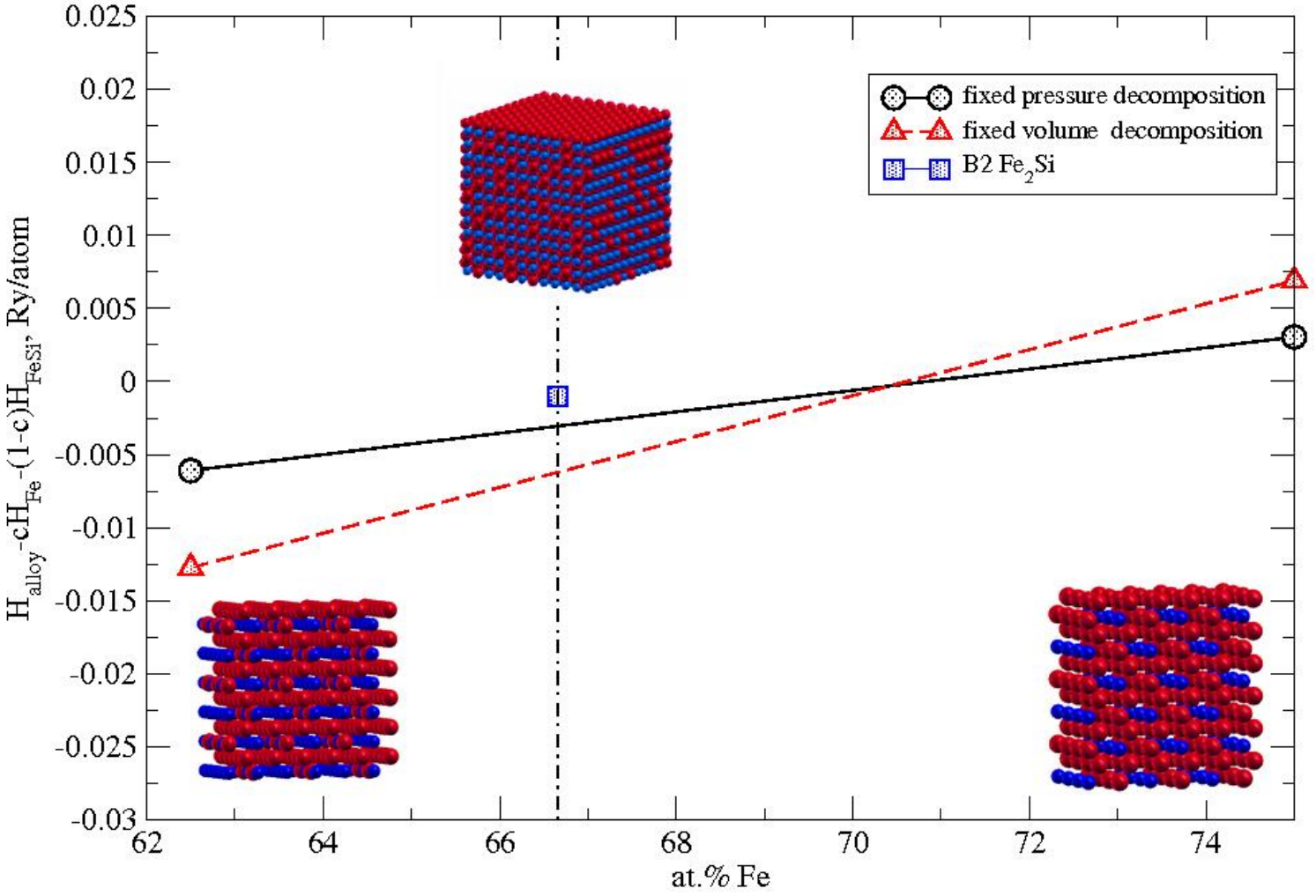


# Calculated mixing enthalpy of paramagnetic partially ordered *B2* Fe–Si alloy




A. V. Ponomareva, A.V. Ruban, N. Dubrovinskaya, L. Dubrovinskiy, and I. A. Abrikosov, Appl. Phys. Lett. **94**, 181912 (2009).

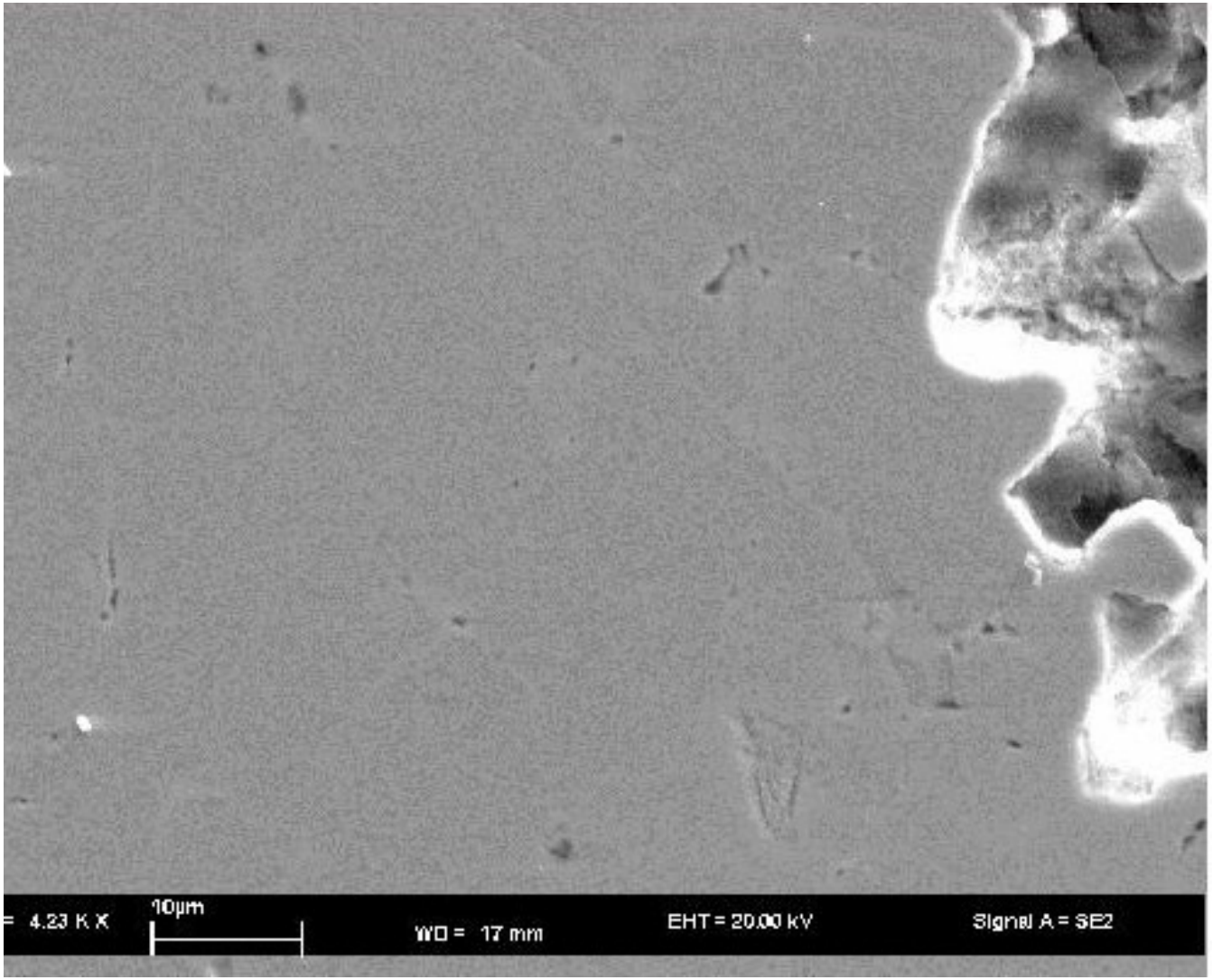
# Monte-Carlo simulations: results




Theory predicts a possibility of HPHT synthesis of metastable B2 phase with composition $Fe_2Si$, which should decompose into $Fe_3Si$ and $Fe_5Si_3$ in equlibrium

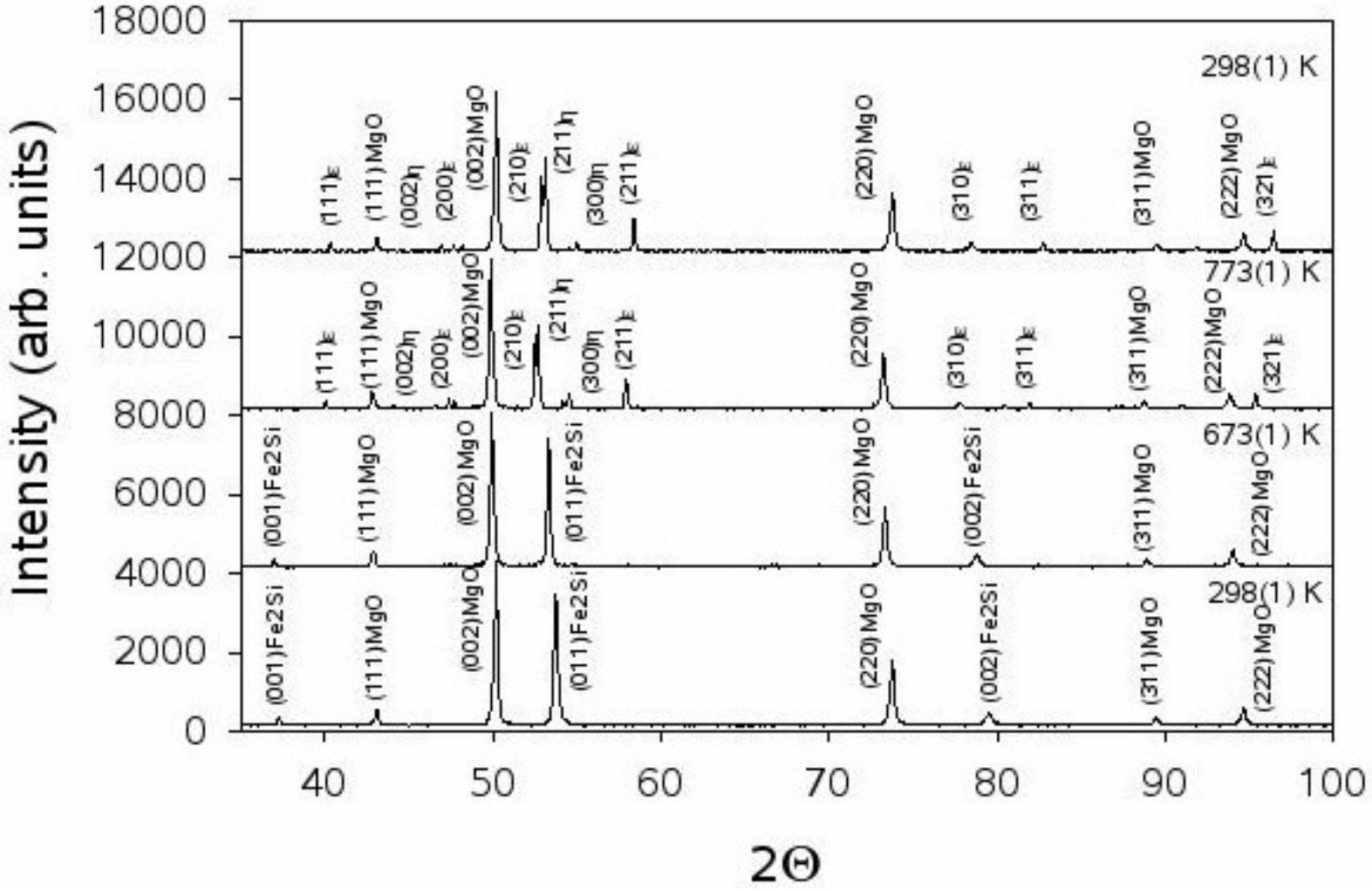
A. V. Ponomareva, A.V. Ruban, N. Dubrovinskaya, L. Dubrovinskiy, and I. A. Abrikosov, Appl. Phys. Lett. **94**, 181912 (2009).

# Experimental HPHT synthesis: results




Scanning electron microscopy secondary electron image of the polished quenched product of the experiment on Fe-Si (66.7:33.1 at.%) mixture treated for 20 minutes in large-volume press at 20 GPa and 2300 K.

# Experimental HPHT synthesis: results




Series of the diffraction patterns collected upon heating (a–c) and cooling back to room temperature (d) of the *B2*-structured $Fe_2Si$ compound.

A. V. Ponomareva, A.V. Ruban, N. Dubrovinskaya, L. Dubrovinskiy, and I. A. Abrikosov, Appl. Phys. Lett. **94**, 181912 (2009).

# Conclusions

- Relevant materials parameters can nowadays be calculated *ab initio* with an accuracy comparable to experimental one.
- It is essential to carry out calculations at realistic conditions.
- Temperature induced magnetic excitations influence interatomic interactions and affect materials parameters.
- It is possible to synthesize new materials by tuning the magnetic state.