Elemental Partitioning Characteristics Across Transformation Interfaces During Thermo-Mechanical-Magnetic Processing - Challenges and Future Direction

S. S. Babu
Ohio State University, Columbus, OH
R. A. Jaramillo
Special Metals Corporation, Huntington, WV
G. M. Ludtka and M. K. Miller
Oak Ridge National Laboratory, Oak Ridge, TN

Acknowledgements

- ISIJ and Prof. Enomoto for invitation and support
- Office of Basic Energy Sciences, Division of Materials Science, U. S. Department of Energy
- Office of Energy Efficiency and Renewable Energy, U. S. Department of Energy
- University of Cambridge, UK
- Ohio State University, Industrial Welding and Systems Engineering
- Oak Ridge National Laboratory
- National High Magnetic Field Laboratory
- Edison Welding Institute

Office of Basic Energy Sciences

U.S. Department of Energy
Energy Efficiency and Renewable Energy

OAK RIDGE NATIONAL LABORATORY
Managed by UT Battelle for the Department of Energy

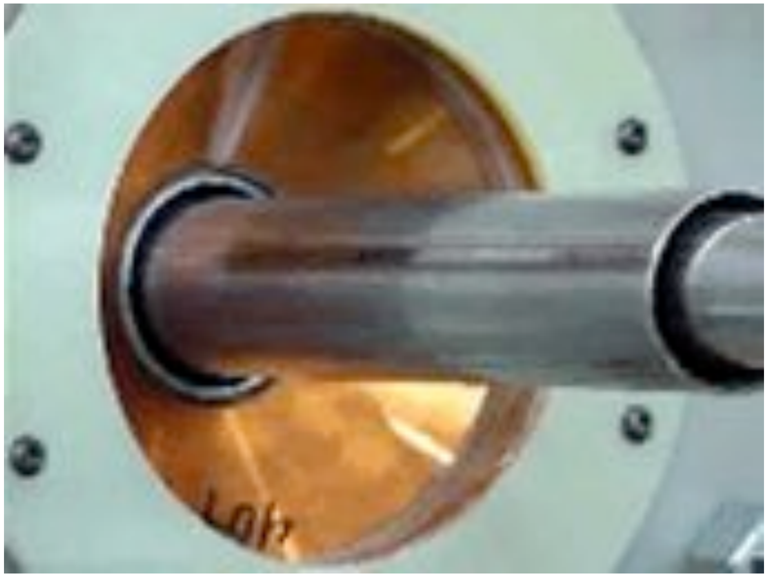
MAGNET LAB
NATIONAL HIGH MAGNETIC FIELD LABORATORY
FLORIDA STATE UNIVERSITY · LOS ALAMOS NATIONAL LABORATORY · UNIVERSITY OF FLORIDA

Outline

- Background & Motivation
- Thermo-Magnetic Processing
  - Alloys
  - Continuous and Isothermal Heat Treatments
- Characterization of Microstructure
  - Optical and Hardness Mapping
  - Transmission Electron Microscopy
  - Local Electrode Atom Probe Microscopy
- Towards Understanding the Mechanisms of Phase Transformation
- Challenges & Future Directions
- Summary

Non-equilibrium (or high driving force) phase transformations are prevalent in materials processing

- Driving Forces (static & dynamic)
  - Composition, Thermal & Mechanical
  - Now electromagnetic conditions
- Immediate Need for Joining Point of View
  - Microstructure evolution under magnetic pulse welding
- Focus of the current paper is to use magnetic fields to accelerate the transformation and compare with computational thermodynamic and kinetic framework



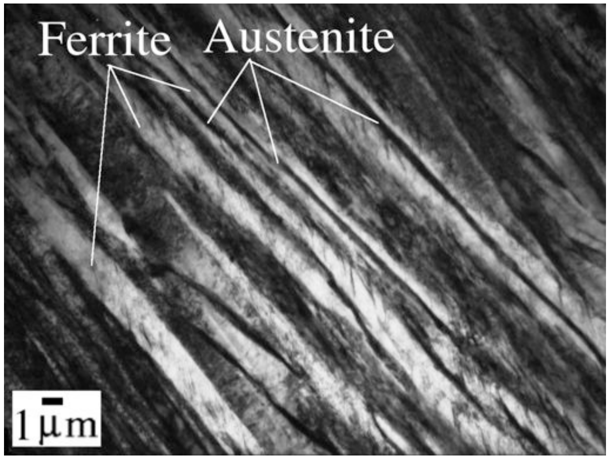
Courtesy: Zhang et al (EWI)

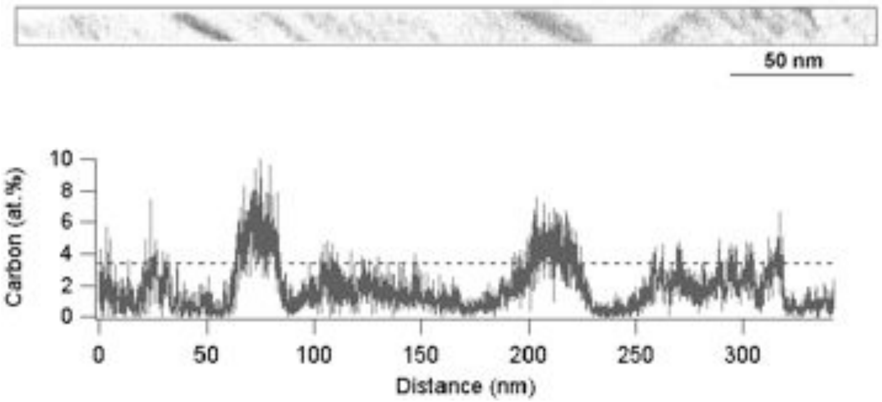
Current focus: Phase Transformations and Alloying Element Partitioning During Thermo-Magnetic Processing

Collaborative Research with Oak Ridge National Laboratory and University of Cambridge

Recently, a bainitic steel with tensile strength greater than 2 GPa has been developed.

- **Based on Fe-Si-Mn-Cr-Co system with high carbon (>0.7 wt.%) and low-temperature-transformation (< 300°C).**
- **Substantial trapping of carbon in ferrite was measured by atom probe field ion microscopy.**
- **Motivation is to accelerate the above transformation by applying high magnetic field ~ 30 Tesla**





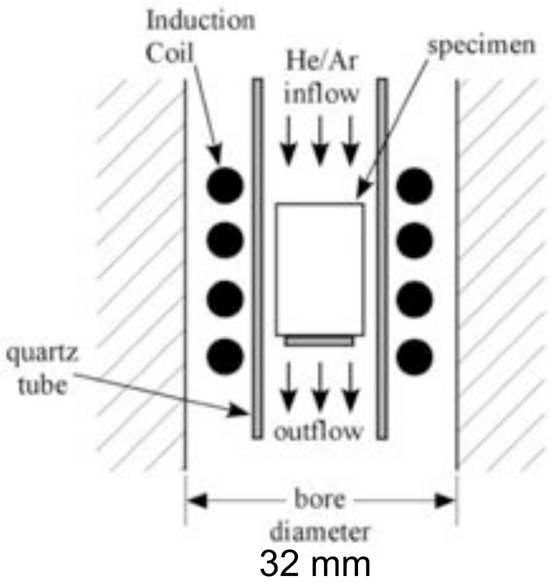
Composition of steels used in these experiments

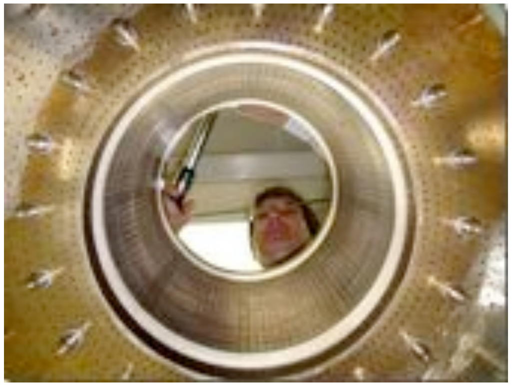
Alloy	C	Si	Mn	Mo	Cr	Co	Ni	Al	Fe
SK (wt.%)	0.75	1.63	1.95	0.28	1.48			0.01	Balance
SK (at.%)	3.34	3.10	1.89	0.16	1.52			0.02	Balance
FK (wt.%)	0.78	1.60	2.02	0.24	1.01	3.87		1.37	Balance
FK (at.%)	3.43	3.01	1.94	0.13	1.03	3.47		2.68	Balance
FeNiC (wt.%)	0.34	1.82	0.01	0.01	0.32	0.01	15.11	0.01	Balance

- **High-carbon steels were used in the isothermal and continuous cooling experiments**
  - **SK: Slow Kinetics; FK: Fast Kinetics**
- **Fe-Ni-C steels were used in isothermal experiments only**

Heat-Treatments were performed within the magnet bore

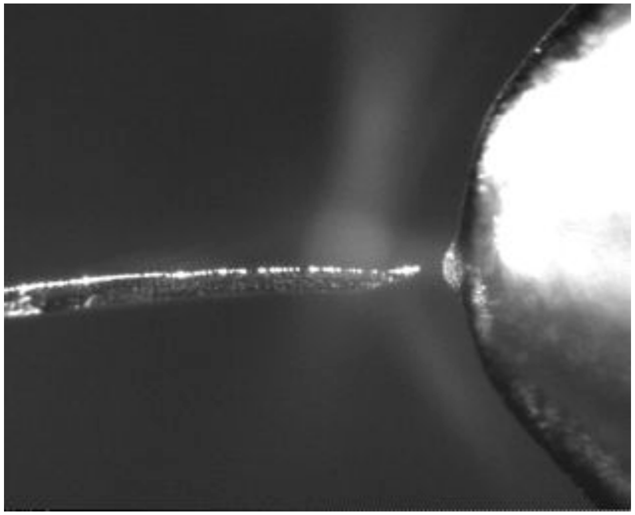
- Controlled induction heating and gas cooling.
  - Austenitization: 1000 °C /3min
- Continuous cooling (0 and 30 Tesla):
  - 1 K/s
- Isothermal heat treatment (0 and 30 Tesla):
  - 740°C / 5 min
  - 300°C / 10 min





Wide range of characterization techniques were used:

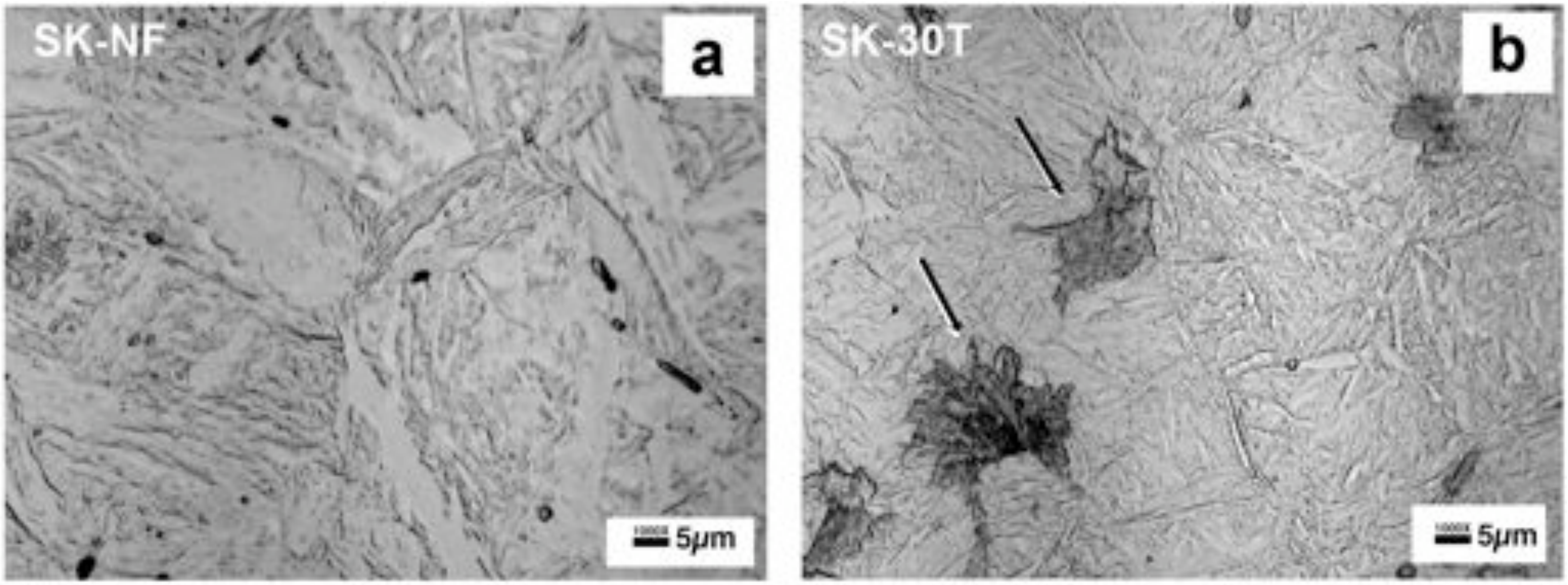
- Optical Microscopy
- Hardness Maps
- Orientational Imaging Microscopy
- Transmission Electron Microscopy
  - TEM (Philips: Tecnai 20
  - 200 kV
- LEAP (imago)
  - 60 K specimen temperature
  - 200 kHz pulse repetition rate
  - Faster data acquisition and large field of view



Results from SK and FK (Fe-C-Si-Mn-Cr-Co-Al) steels

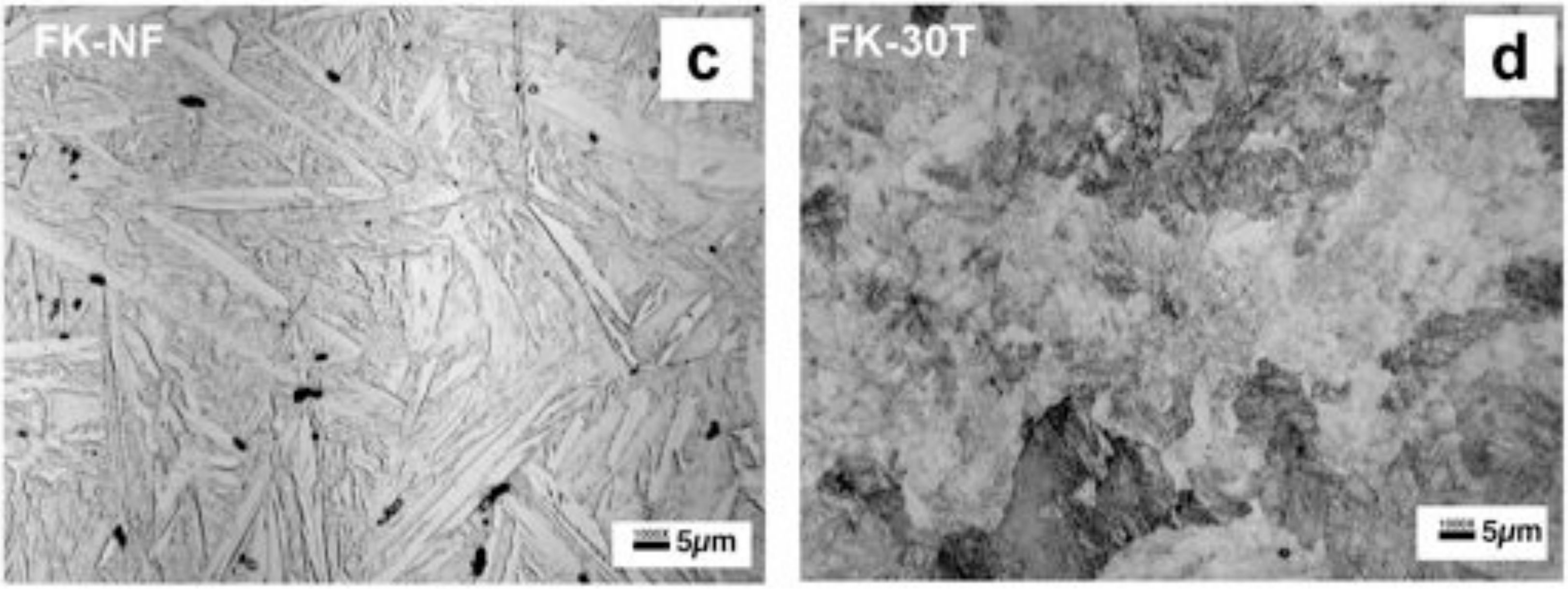
Motivation: Accelerate the bainitic transformation during continuous cooling (1 K/s)!

Steel (SK) with no ferrite stabilizing element showed some differences



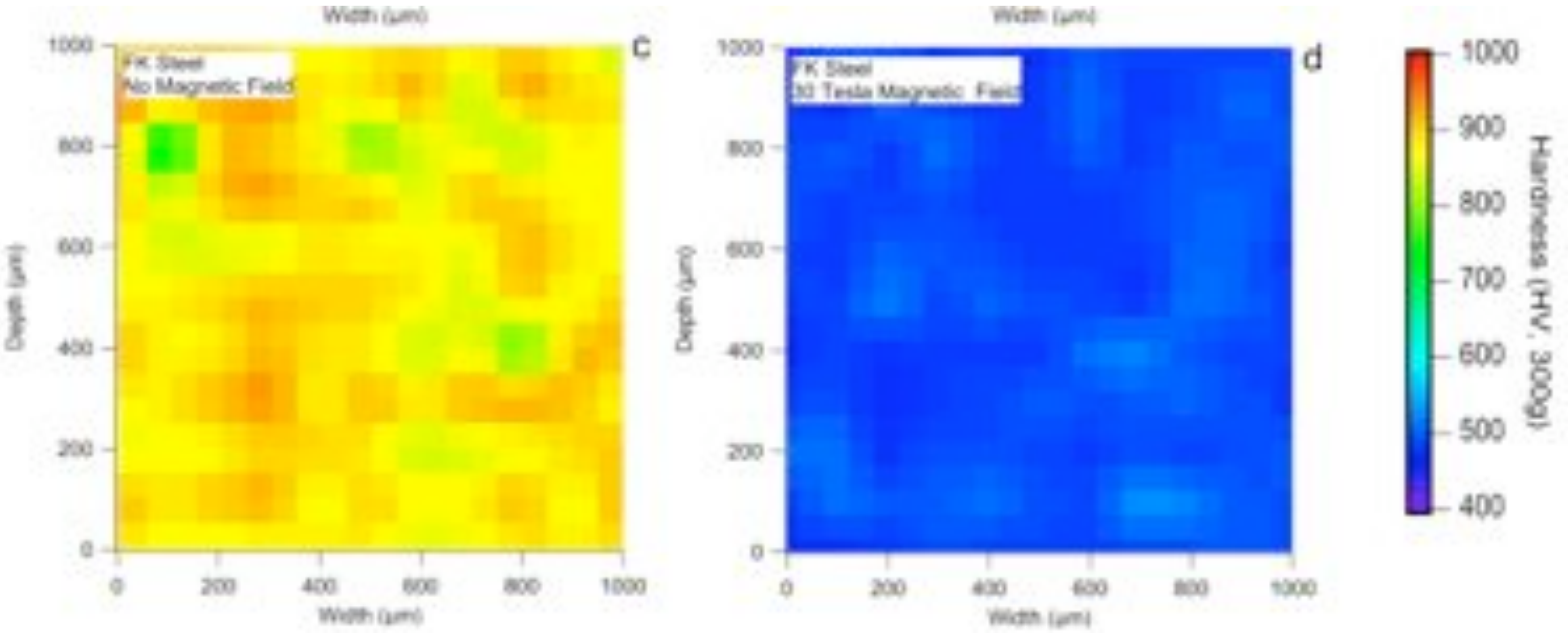
- Some dark etching regions can be seen
- Let us look at the (FK) steels with Co and Al additions

Steel (FK) with Co and Al element additions showed large differences



- What is the dark etching microstructure?
- Let us evaluate the same with hardness mapping:

Hardness maps show softer and uniform microstructure in FK steel with 30Tesla field

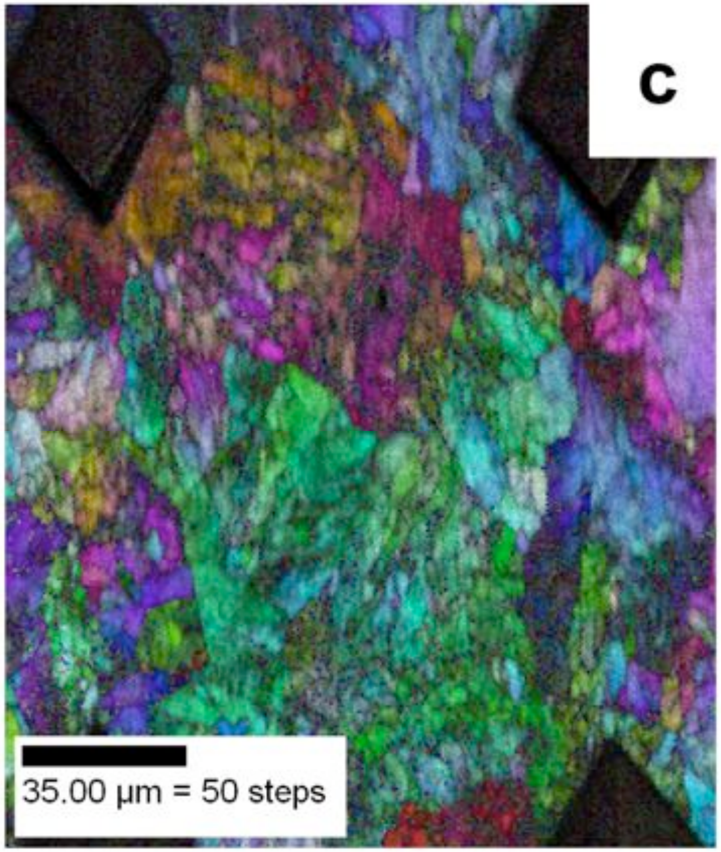


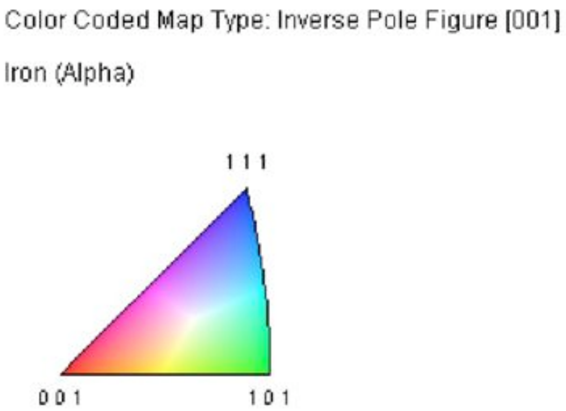
No Field - Martensite

30Tesla- Soft & Uniform microstructure

- **What is this soft microstructure?**

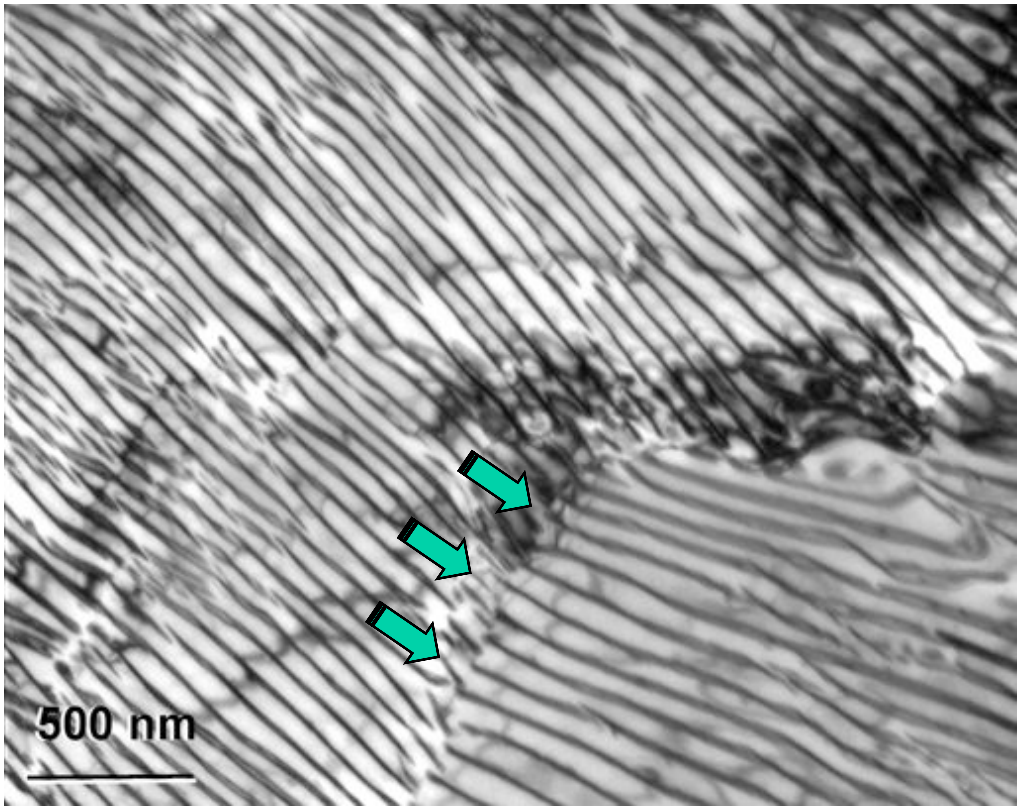
Orientation imaging microscopy shows large ferrite regions with small mis-orientations disrupted by secondary phases!





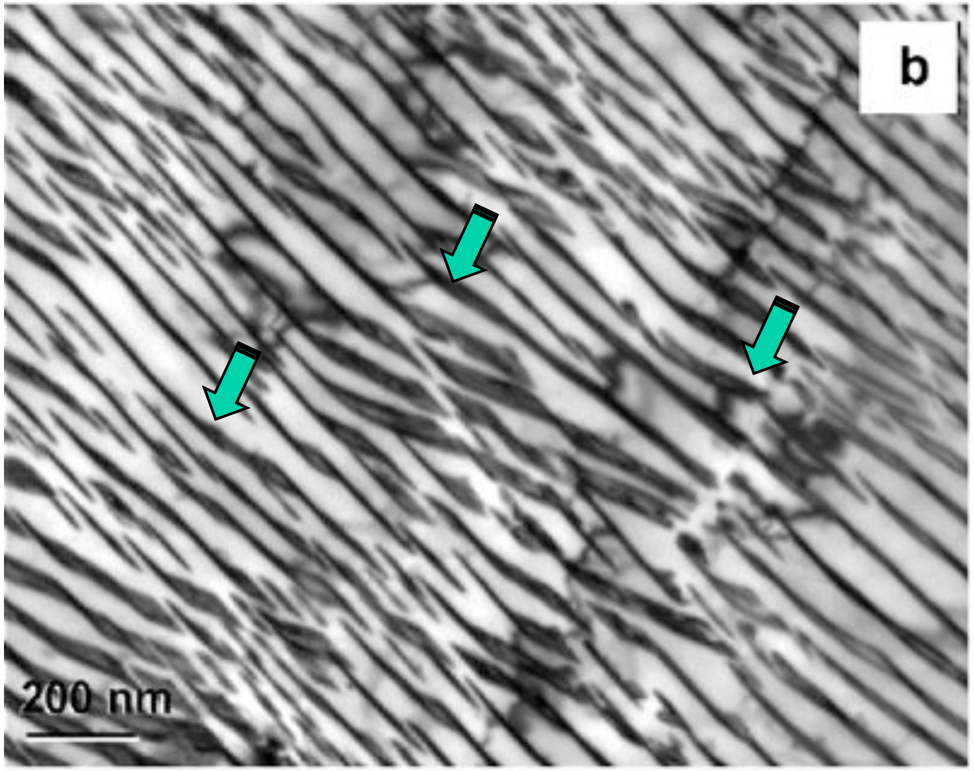
- Is it pearlite?

Transmission electron microscopy confirmed that these are indeed nanoscale pearlite



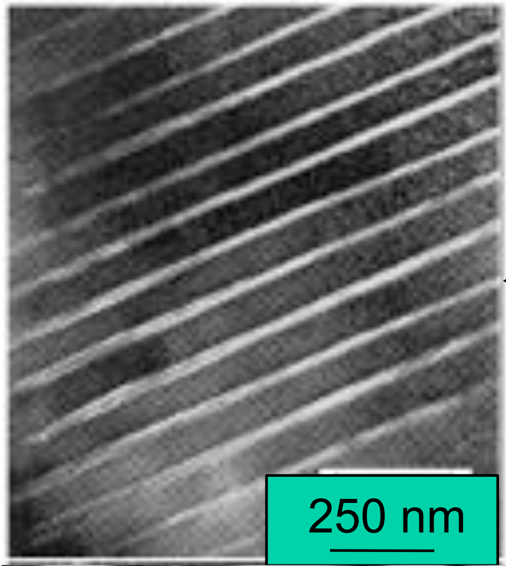
- Lamellar spacing in the order of 50 - 100 nm
- Close to Bagaryatski orientation relationships
- Arrows indicate the colony boundaries

Sub-structure of cementite show faults and changing morphology

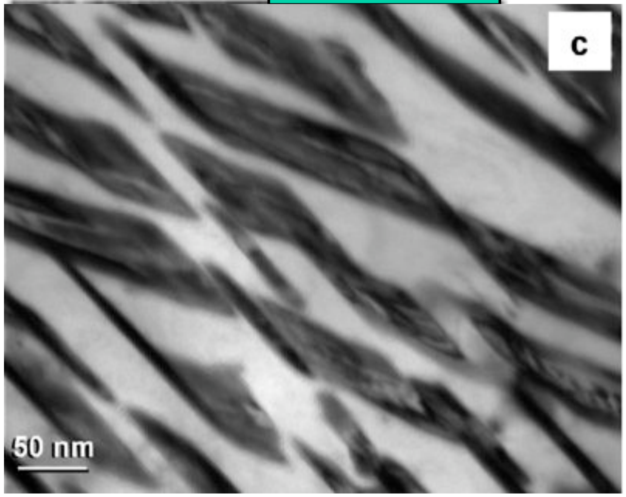


- **Frequent interruptions of the cementite morphology – indicates possible changes in the transformation front directions**

Significance of this result:



Wire Drawing



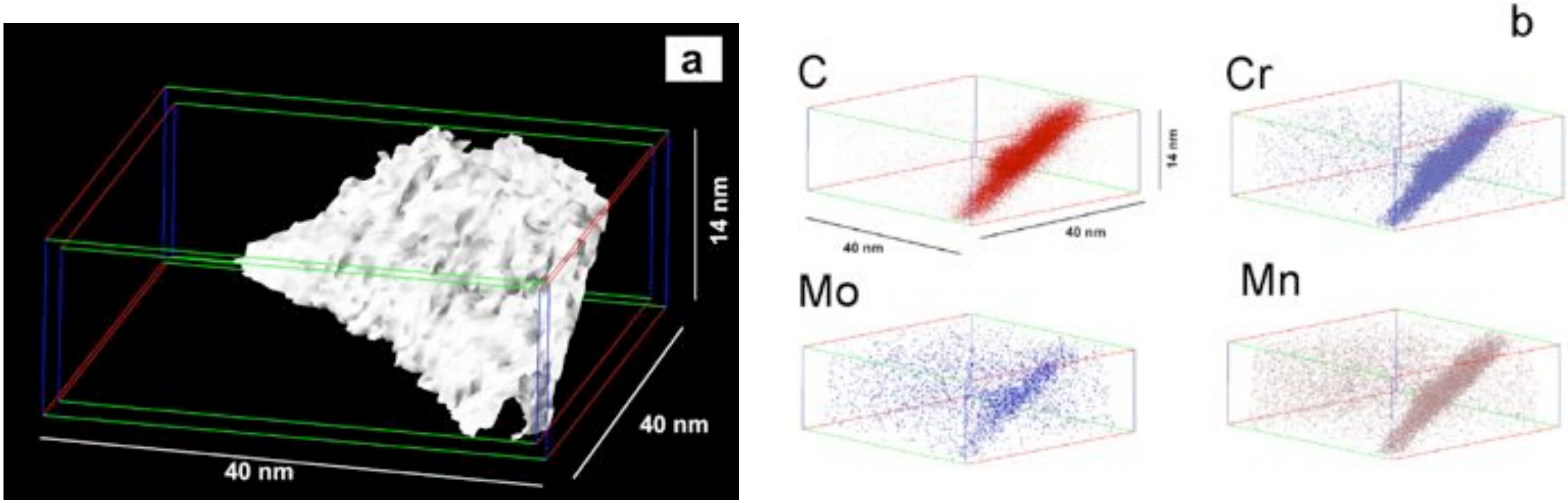
Magnetic Processing

- **The pearlite lamellar spacing is indeed closer to the pearlite after patenting process (extensive thermomechanical processing); for example:**
    - **Read and Hono (1997)**
    - **Hinchliffe and Smith (2001)**
- **We are getting finer microstructure just by continuous cooling at 1 K/s within 30 Tesla.**
- **<u>Now the question is what is the mechanism for such fine pearlite formation?</u>**

Towards understanding the Mechanisms of the transformations

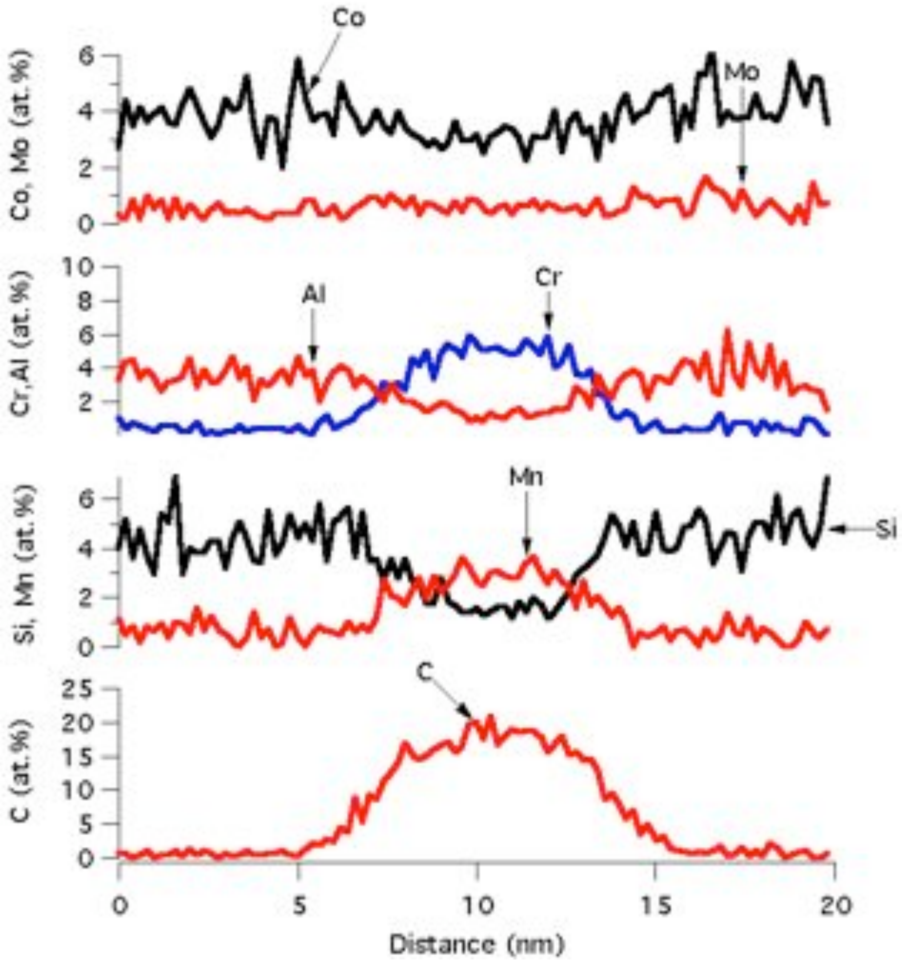
(1) Interface partitioning characteristics between ferrite and cementite

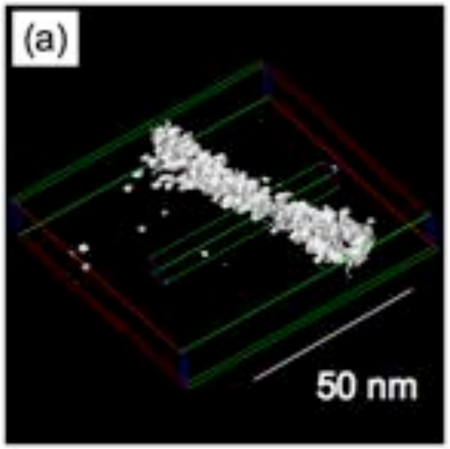
Local Electrode Atom Probe shows tendency for all elemental partitioning



- How about the magnitude of partitioning?

Preferential partitioning of Cr and Mn to cementite was observed



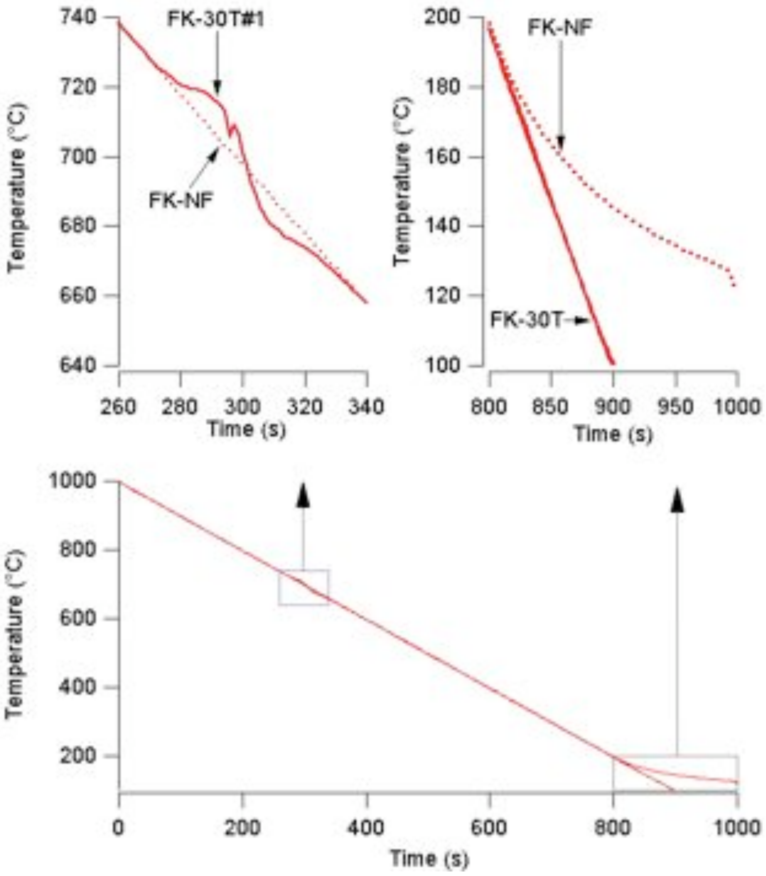


- Si, Al and Co partitions to ferrite.
- Is this partitioning close to thermodynamic equilibrium?
- We need to know the temperature of formation.

Towards understanding the Mechanisms of the transformations

(2) When does the austenite to pearlite transformation occur?

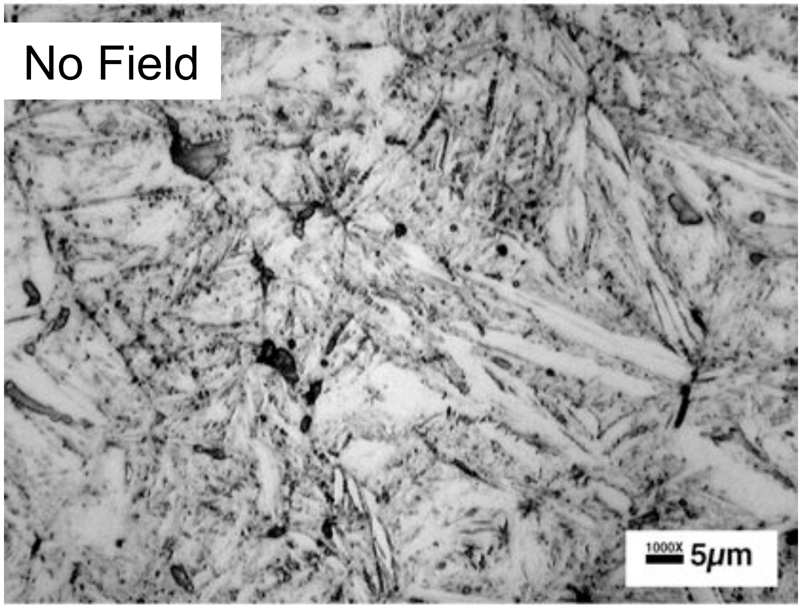
Temperature measurements showed indication for release of latent heat of transformation



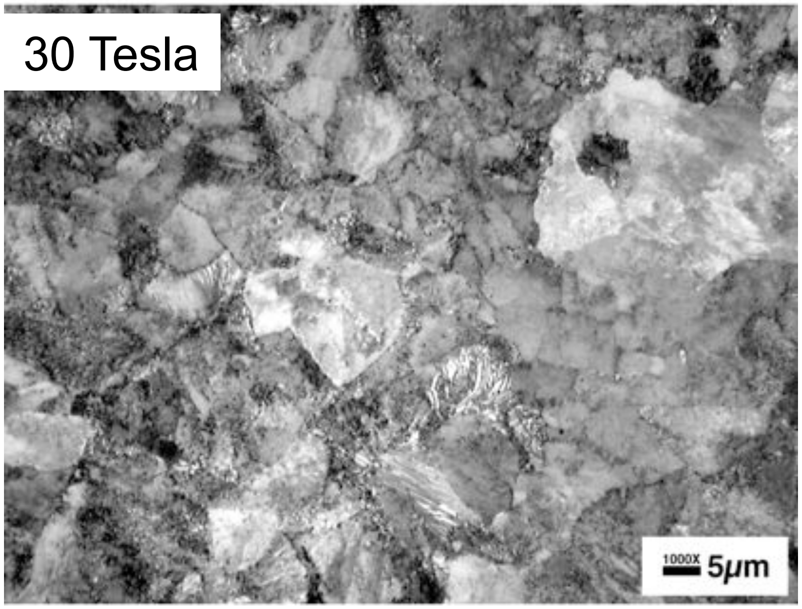
- Start of pearlite transformation ~ 720 to 740°C
- Similar heat release can be observed at 200°C in samples without magnetic field due to martensite formation
- We need to confirm these observations:

Isothermal transformations at 740°C for 5 minutes confirmed the pearlite formation temperature

FK/iso740/5/NF



FK/iso740/5/30T

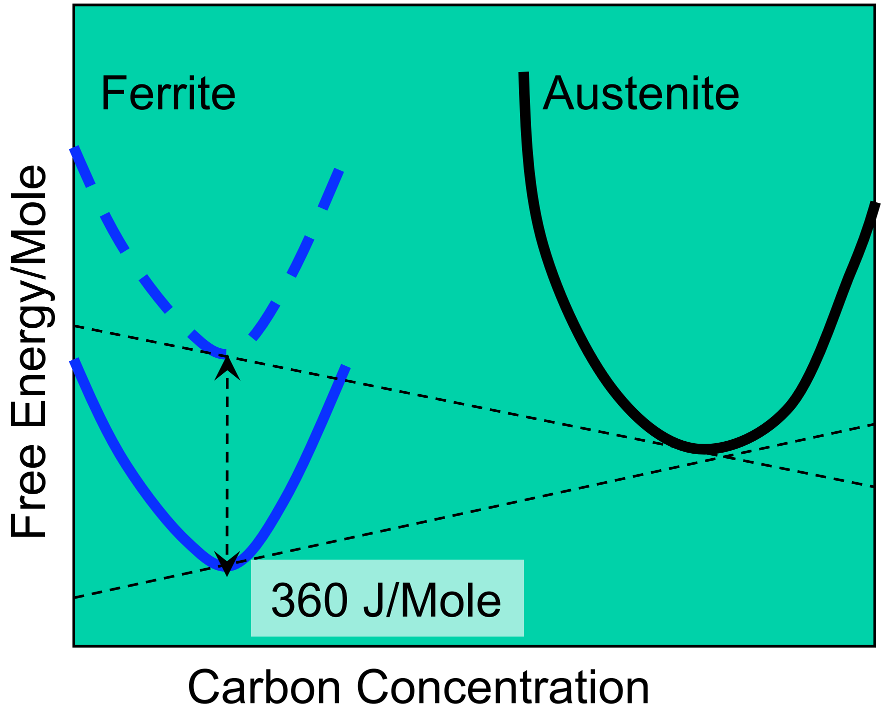


- **Now how do we model the transformation kinetics with magnetic field?**

Towards understanding the Mechanisms of the transformations

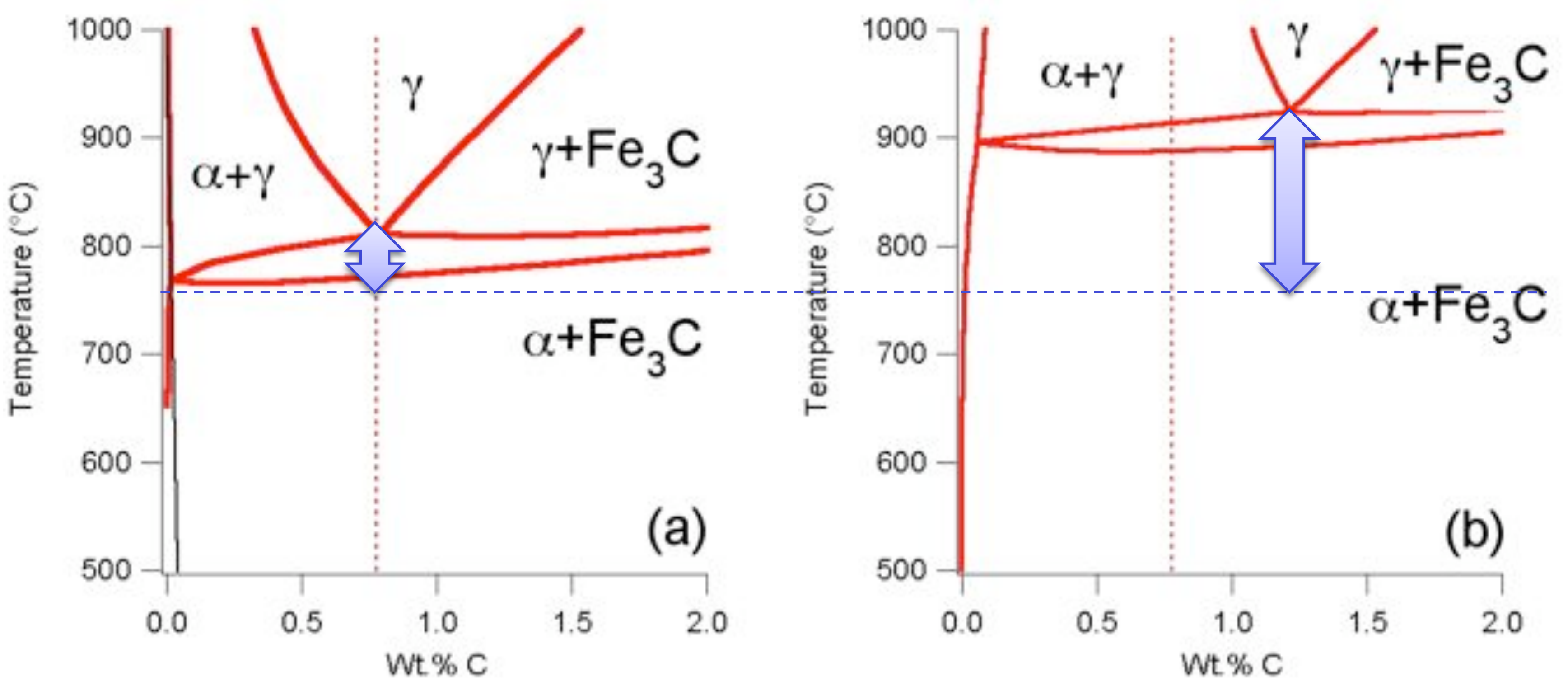
(3) Magnetic Stabilization of Ferrite and Computational thermodynamics:

Reduction of ferrite free energy as a function of magnetic field strength can be estimated



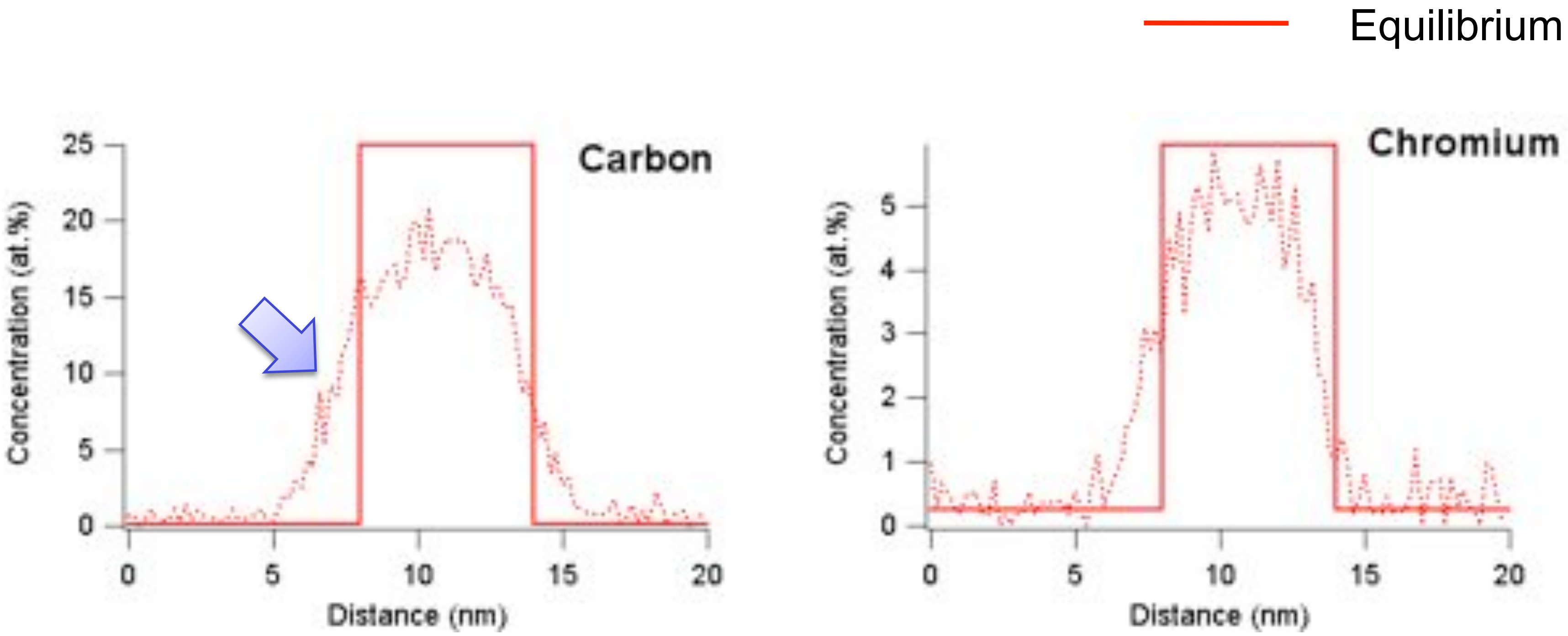
- Many publications exist in this topic
- Assumptions here
  - Reduction in free energy to be 12 J/mole/Tesla
  - For 30 Tesla = 360 J/Mole
  - No effect on cementite (may not be valid)
- Used this value in ThermoCalc® software

With magnetic field the undercooling for pearlite transformation increases



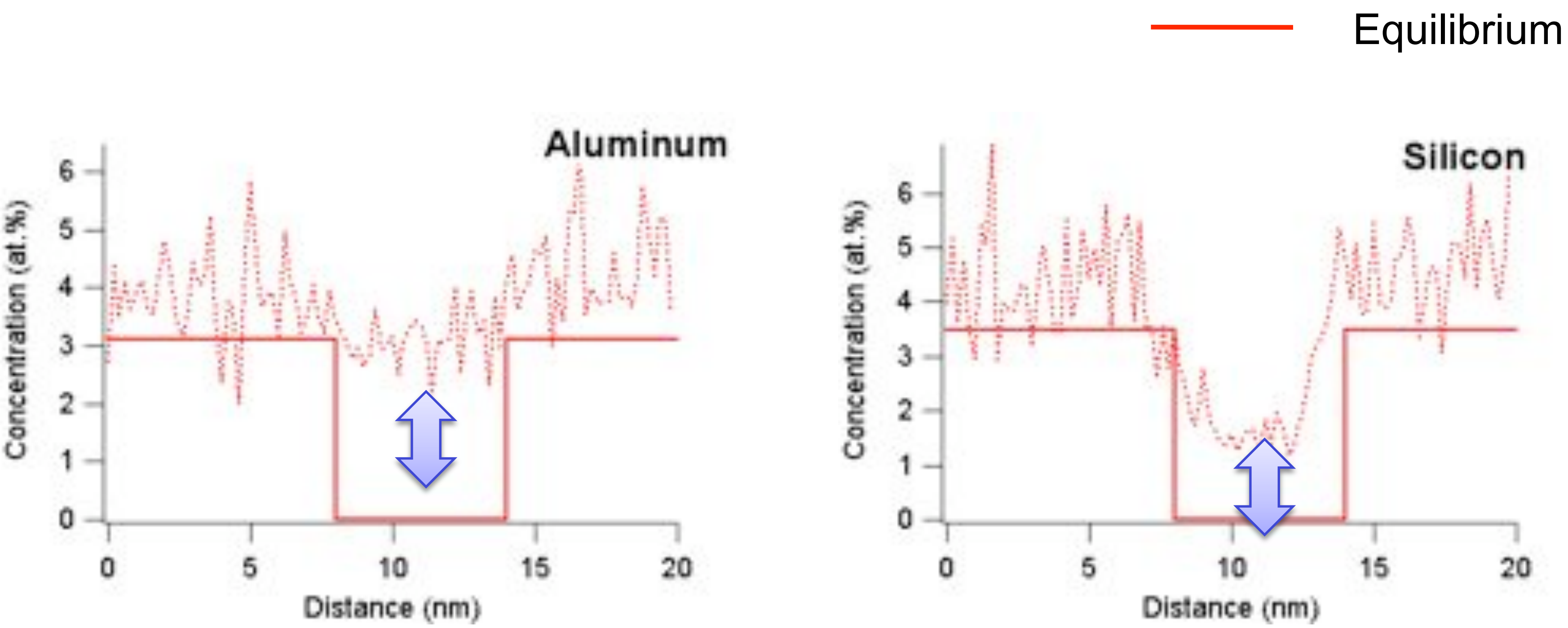
- **How does the measured elemental partition compare with thermodynamic estimations?**

Carbon and Chromium



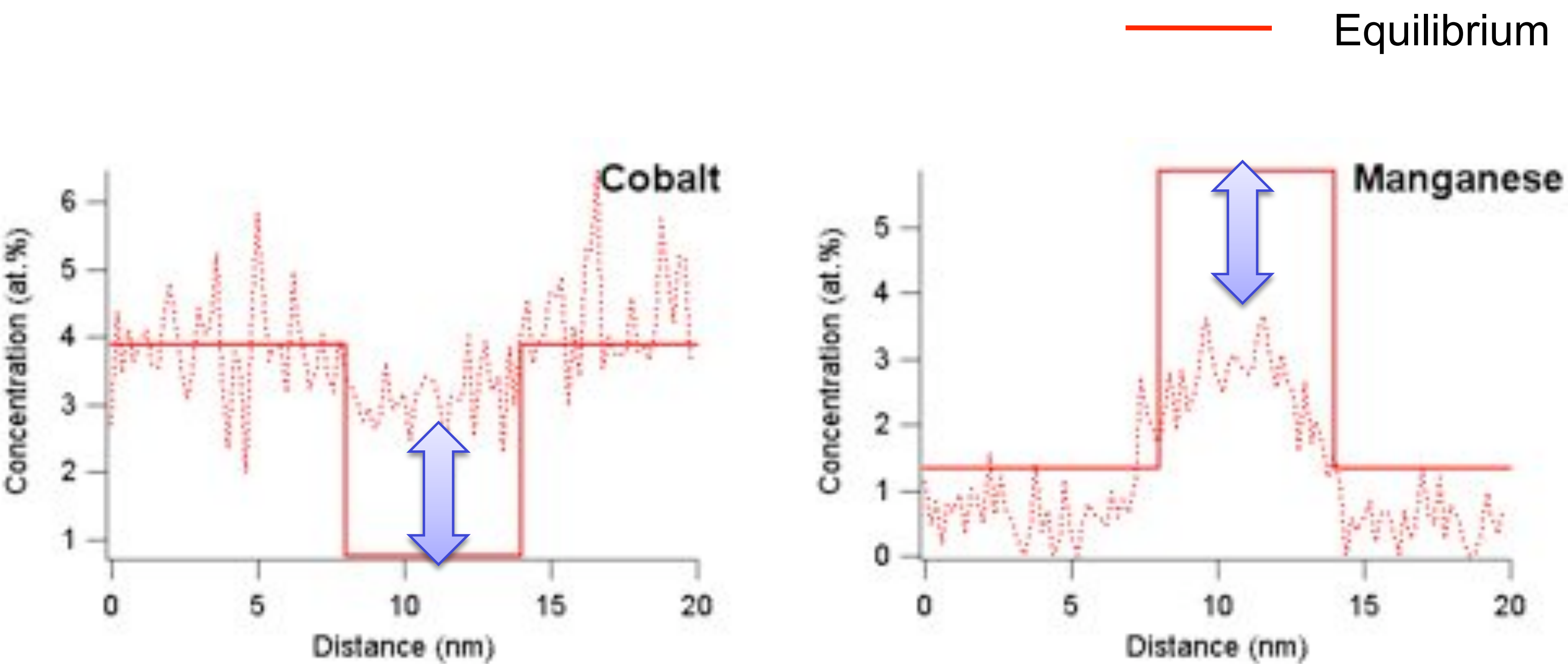
- **Supersaturation of C in ferrite!**
- **Cr is very close to equilibrium**

Aluminum and Silicon



- **Expected silicon & aluminum concentration in cementite is zero!**

Manganese and Cobalt



- **What is the significance?**

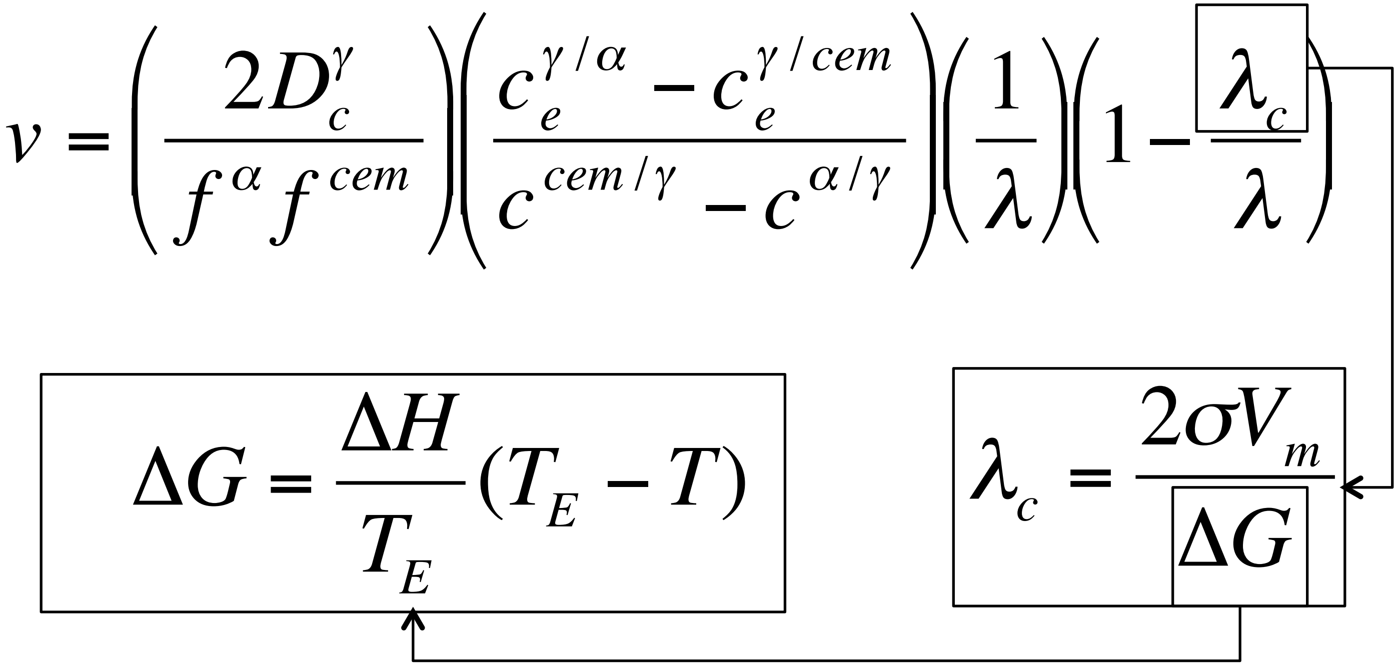
Significance of the measured elemental partitioning

- Agreement with calculated trends
- Magnitudes indicate non-equilibrium
- Trapping of silicon and aluminum shows that the transformation is occurring under large departure from local equilibrium conditions!
- No solute spikes near the interface

Towards understanding the Mechanisms of the transformations

(4) How about leveraging existing pearlite growth theories?

Zener-Hillert Growth Model was used



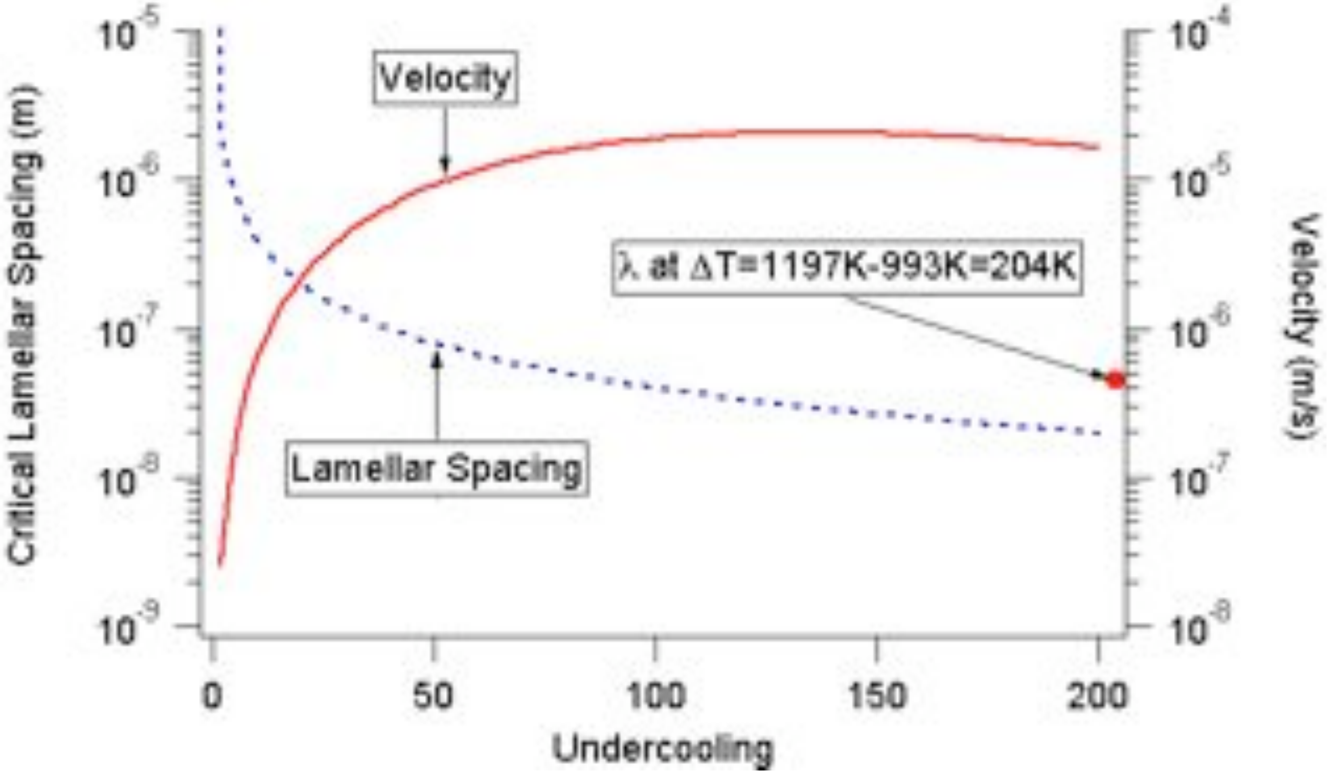
$$v = \left(\frac{2D_c^{\gamma}}{f^{\alpha} f^{cem}}\right)\left(\frac{c_e^{\gamma/\alpha} - c_e^{\gamma/cem}}{c^{cem/\gamma} - c^{\alpha/\gamma}}\right)\left(\frac{1}{\lambda}\right)\left(1 - \frac{\boxed{\lambda_c}}{\lambda}\right)$$

$$\boxed{\Delta G = \frac{\Delta H}{T_E}(T_E - T)}$$

$$\boxed{\lambda_c = \frac{2\sigma V_m}{\boxed{\Delta G}}}$$

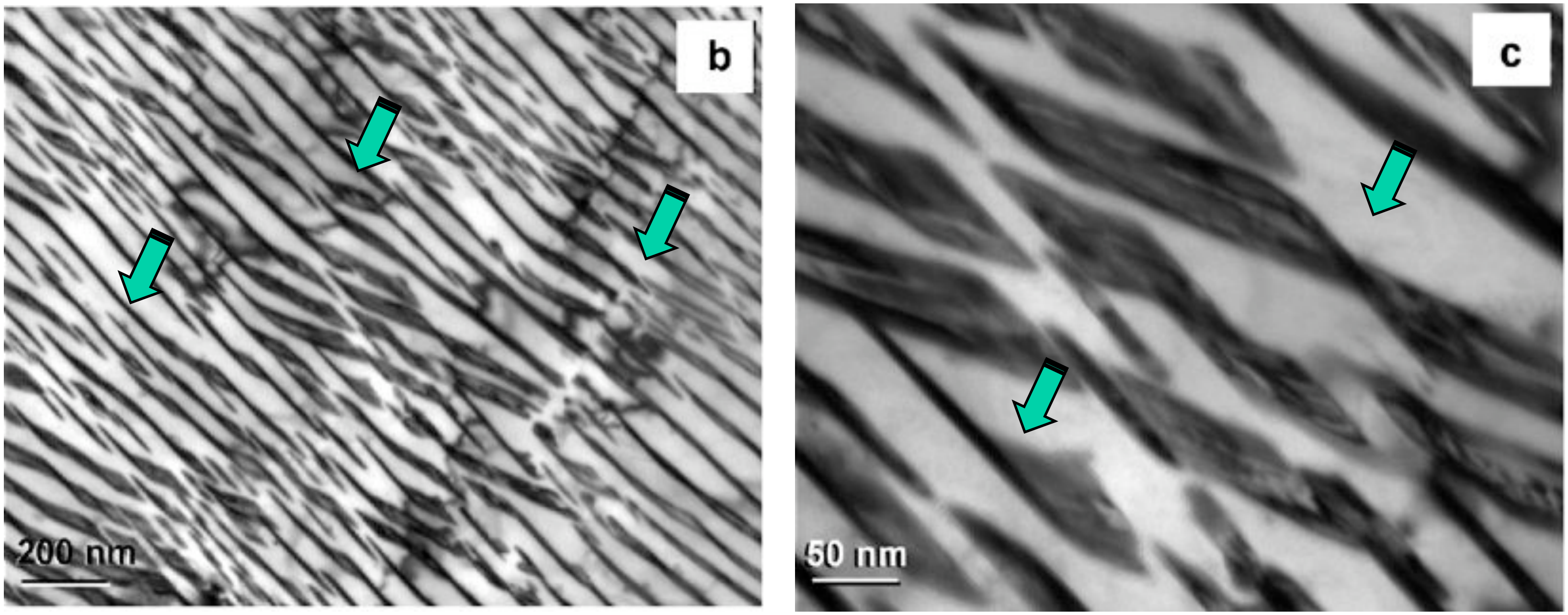
- **Using the values given in Offerman et al (2003) & by extrapolating the phase boundaries we can estimate the lamellar spacing & velocity**

Consistent with our experimental measurements of lamellar spacing



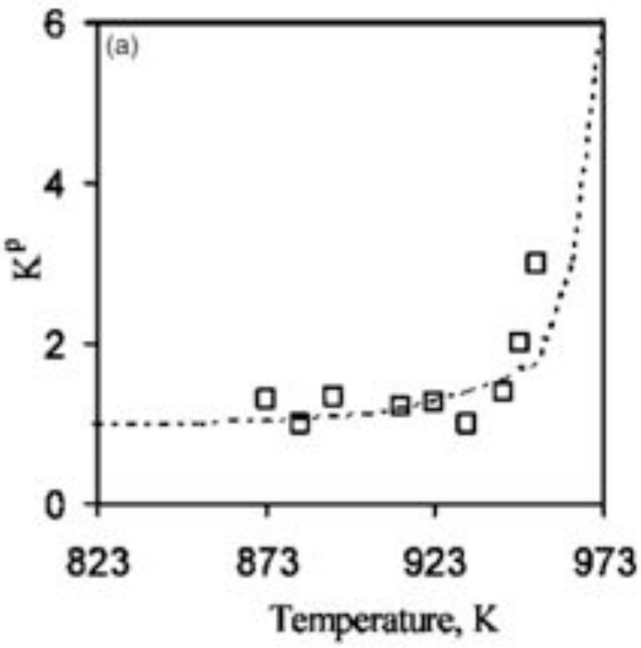
- Sensitive to interfacial energy: here it is 1 J/m^2
- Is this high velocity possible?

Sudden changes in cementite morphology and faults qualitatively support high velocity



- How about non-equilibrium partitioning?

Increase in velocity often leads to non-equilibrium partitioning/trapping at interface



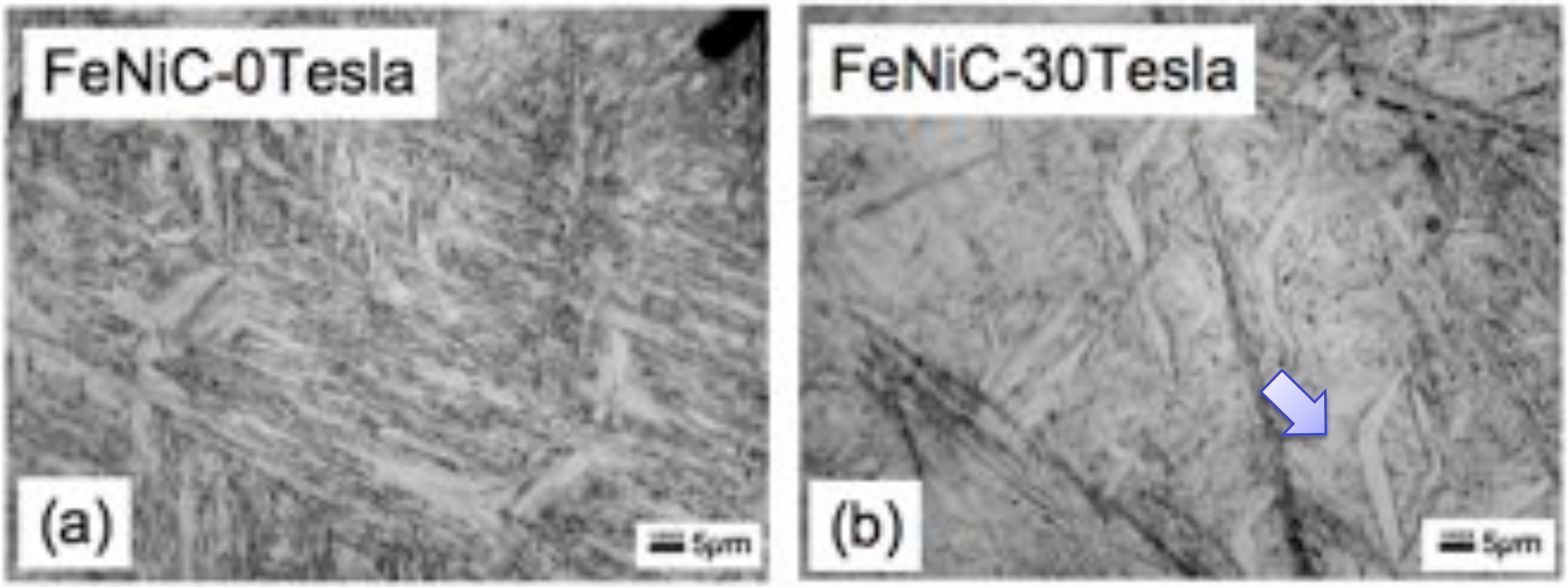
$$k_p = \frac{1 + \beta k_e}{1 + \beta}; \beta = \frac{D}{v x_c}$$

- Capdevilla et al (2002) for Fe-0.7C-1.0Mn wt.5 steel;
- However, we see different extents for different elements.
- We need better models for transition from equilibrium to non-equilibrium partitioning

Effect of Magnetic Field on Displacive Transformation under High Undercooling

Fe-Ni-C Steel experiments

Only small differences in the plate morphology.

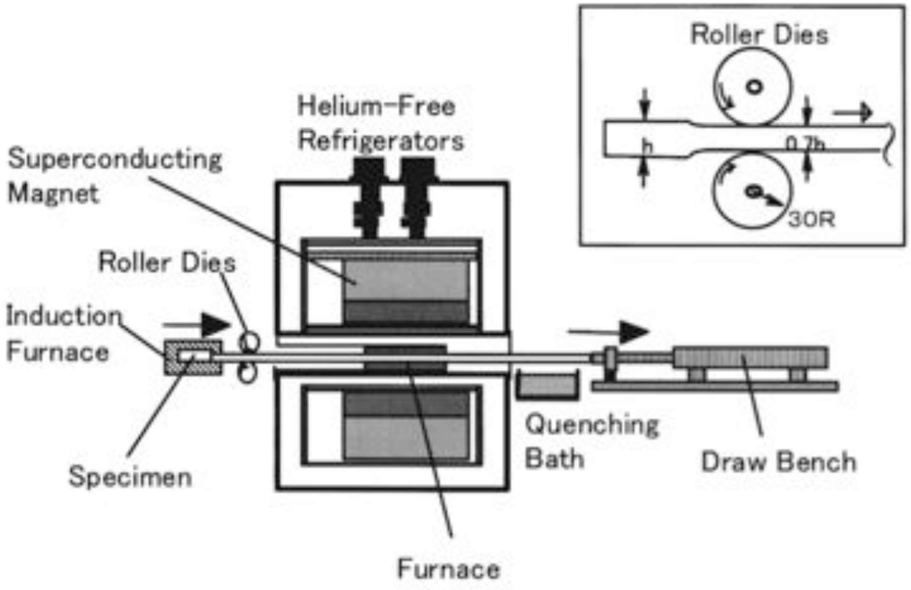


- **Chemical driving force nucleation must be overwhelming compared to magnetic field effects**
- **Also we need to measure the transformation kinetics in-situ!**

Challenges and Path forward

Three of the many are highlighted

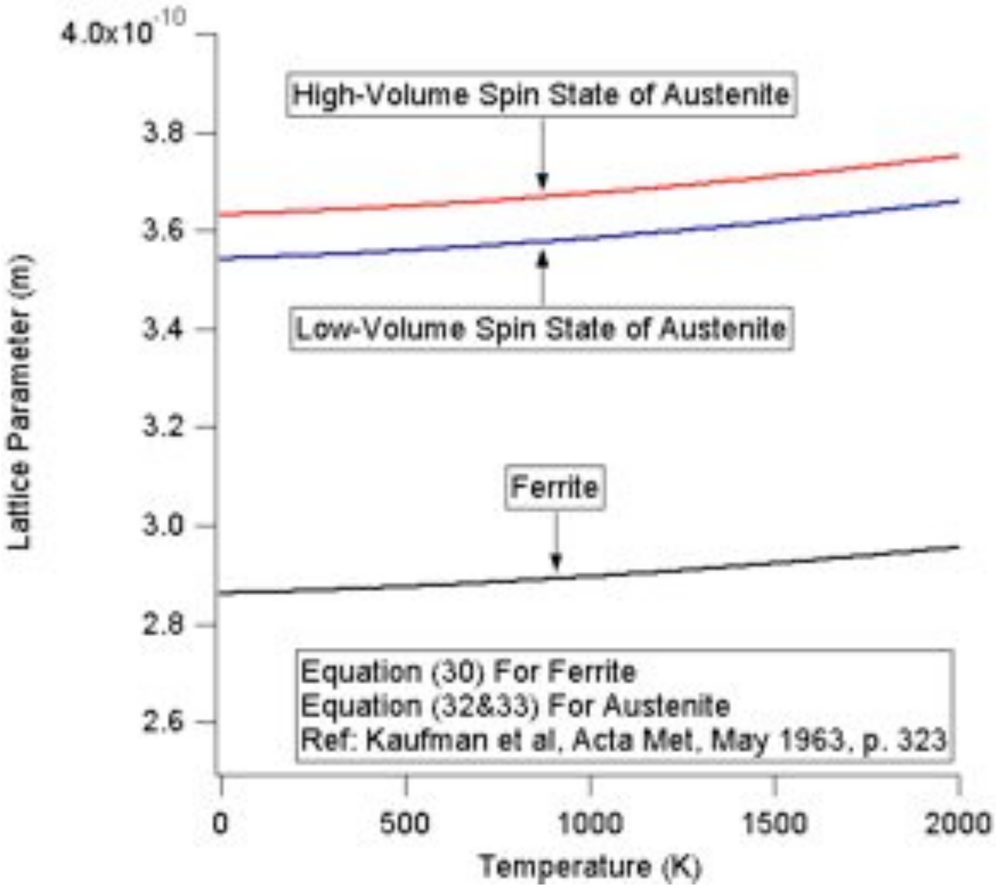
(1) Deploying thermo-mechanical-magnetic processing of steel industry



Shimotomai et al, Acta Mat (2003)

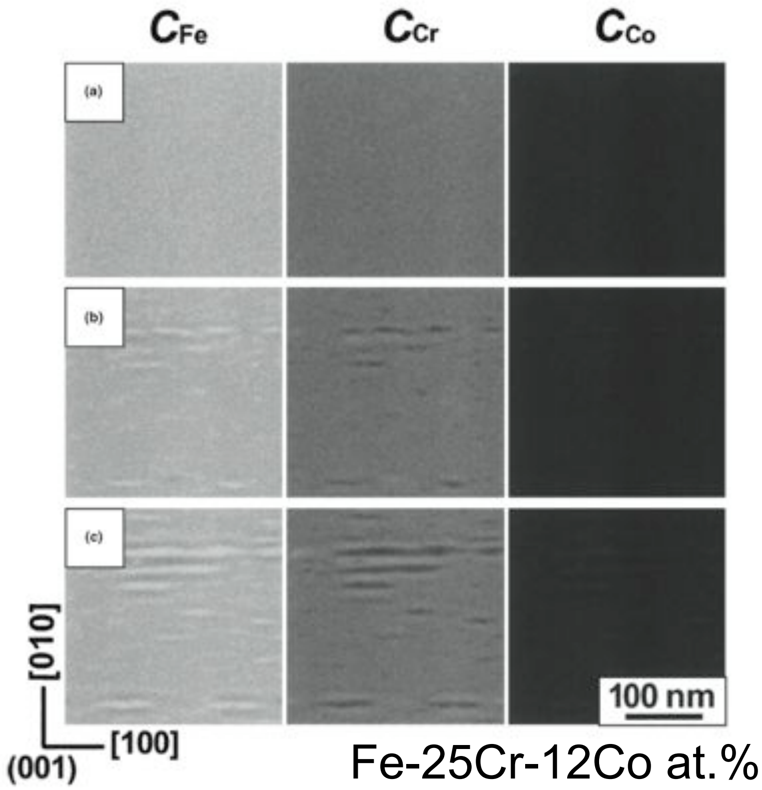
- **Magnet designs and steel processing designers have to collaborate**

(2) Fundamental understanding of interaction between magnetic field and two magnetic states of austenite



- How will this affect expansion coefficient of austenite?
- How will it change the magnetic properties?
- We need in-situ diffraction measurements

(3) We must be able to predict the three dimensional morphology as well as non-equilibrium partitioning seamlessly



Fe-25Cr-12Co at.%

Koyama and Onodera (2006)

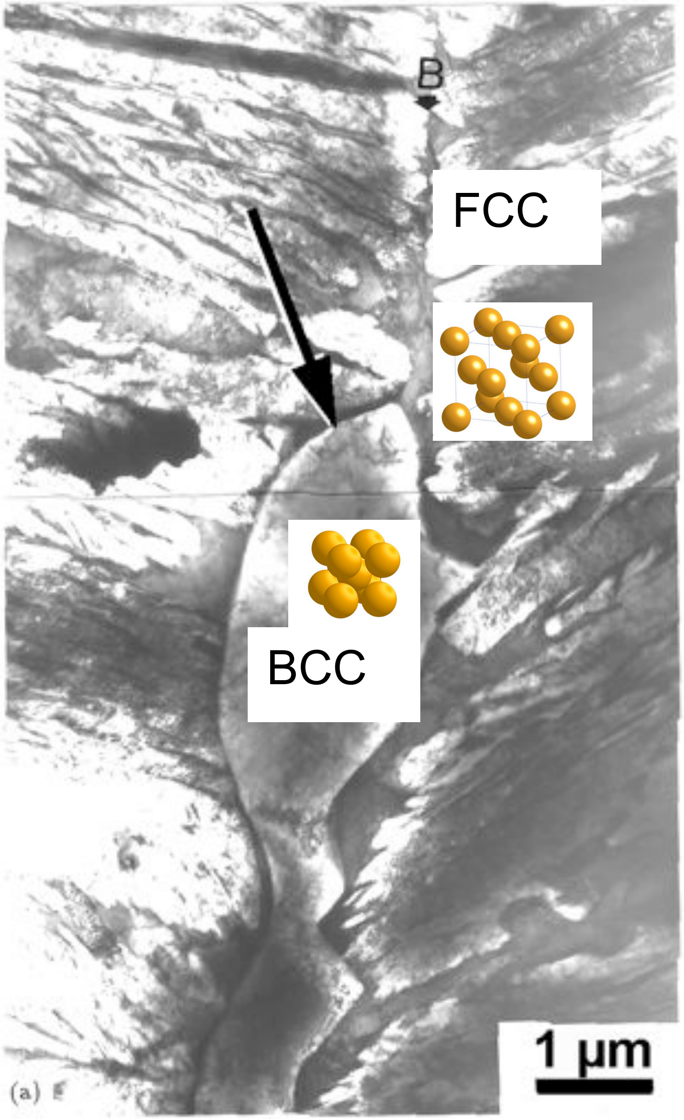
- **Phase field may offer a path way to do this with experimental feed back**
- **Need more coordinated theoretical and experimental research**

Summary

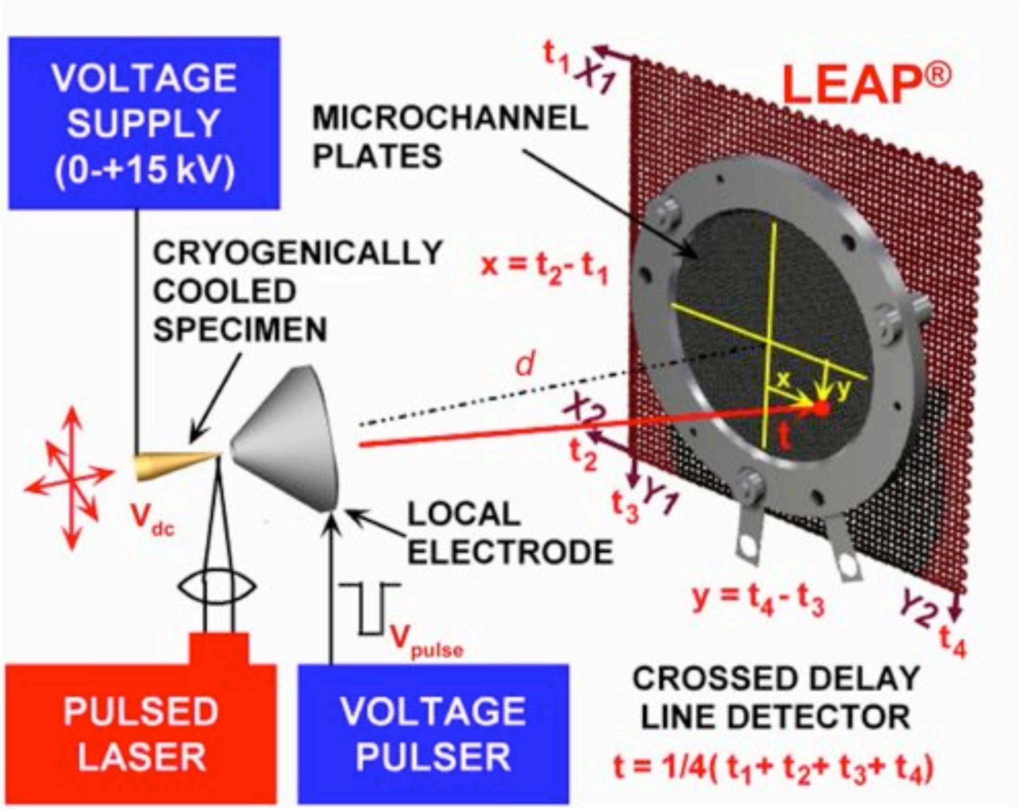
- Need for seamless equilibrium to nonequilibrium phase transformation models to describe advanced processing of metals and alloys was stressed
- Effect of 30 Tesla magnetic field on producing nanoscale pearlite was discussed
- Small lamellar spacing and non-equilibrium partitioning is attributed to high transformation interface velocity
- Challenges in deploying thermo-mechanical-magnetic processing of steels to industries are presented
- Need for coordinated theoretical-experimental research is suggested as pathway

What do we need?

- Models to describe the non-equilibrium transformations without a-priori assumptions
  - Equilibrium to Non-Equilibrium Solidification
  - Local Equilibrium (PE, NPLE)
  - Para Equilibrium
  - Displacive
- Overall Motivation:
- Seamlessly describe these conditions as a function of composition and external driving forces
- Evaluate assumptions with detailed characterization of transformation interfaces



Local electrode atom probe principles



Potential Energy ⟶ Kinetic Energy

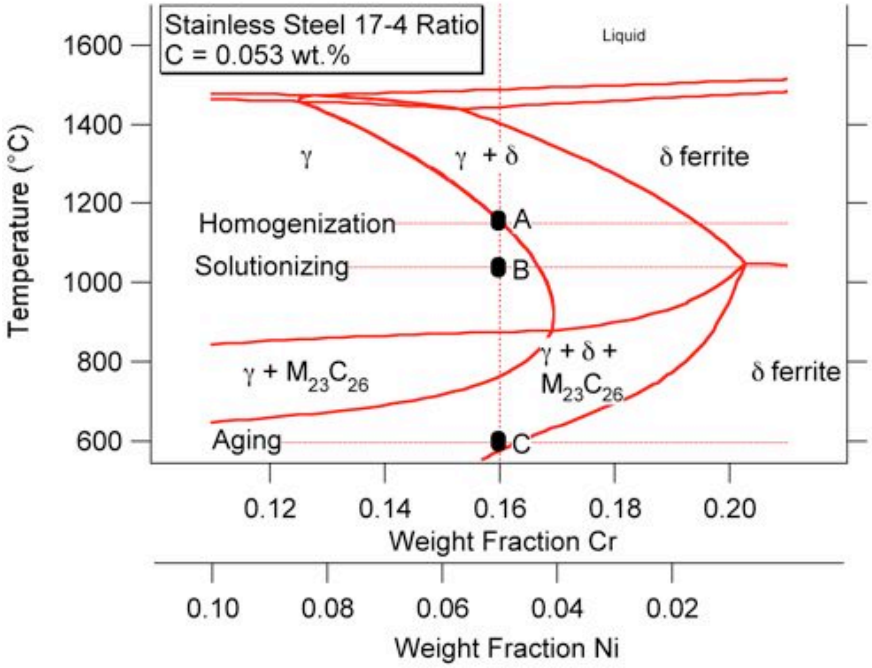
$$neE = \frac{1}{2}mv^2$$

- Uses high-voltage pulses (200 kHz) to field evaporate atoms sequentially
- Evaporated atoms are detected by time of flight spectrometer
- The detected atoms are reconstructed in a computer to evaluate the morphology and nanoscale concentration distributions

Methodologies to describe multi-component phase stability exists

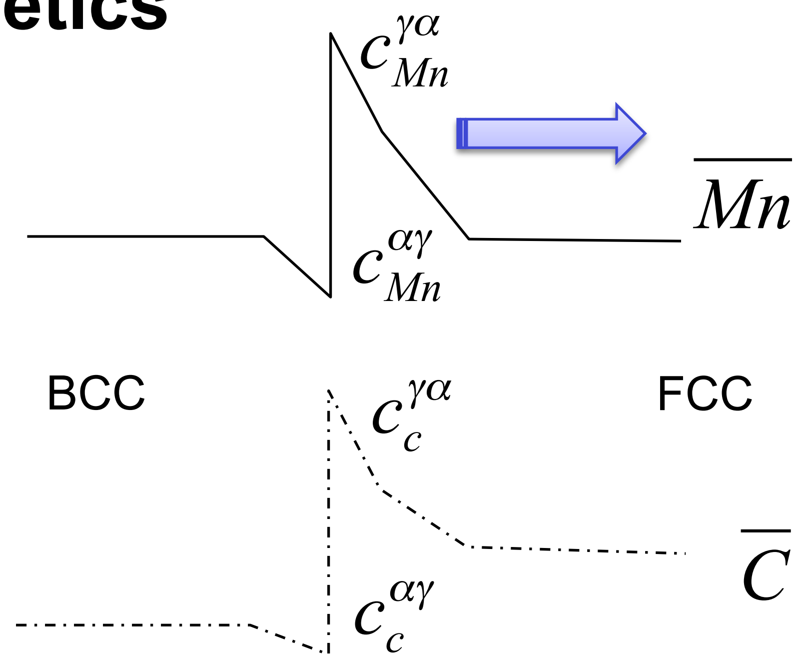
- **Software Tools**
  - **ThermoCalc®, JMatPro®, PANDAT®, MTDATA®..**
  - **Databases, TC-Fe…etc**
  - **Extension to constrained equilibrium like para-equilibrium**
- **Basis: Gibbs free energy minimization as a function of temperature and composition and phases**
- **focus 1: Better Databases**
- **focus 2: Link to 1st principles**

$$G = \sum x_i G_i^0 + RT\sum x_i \log_e x_i + \sum_i \sum_{j>i} x_i x_j \sum_v \Omega_{ij}^v (x_i - x_j)^v$$



It is possible to predict multi-component diffusion controlled kinetics

- **Software Tools**
  - **DicTra®, PrecipiCalc®,..**
  - **Databases, Mobility, Extension to Para-equilibrium, Link to phase field simulations**
  - **Phenomenological: JMatPro®**
  - **Public Domain: Materials Algorithm Project**
- **Basis: Local equilibrium with bulk and boundary diffusion**
- **focus 1: Non-equilibrium conditions**
- **focus 2: Interface & Diffusion controlled (mixed)**

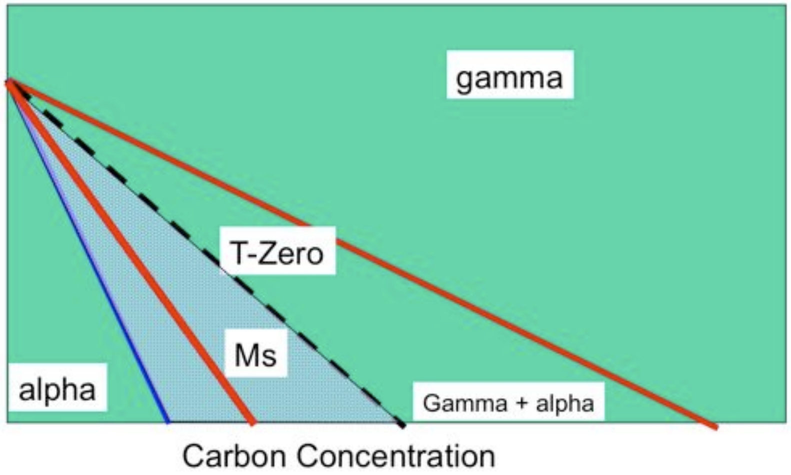


$$(c^{\alpha\gamma}_{Mn} - c^{\gamma\alpha}_{Mn})v_{\text{interface}} = -D^{BCC}_{MN}\frac{dc_{Mn}}{dx}\bigg|^{ferrite}_{\text{interface}} - D^{FCC}_{MN}\frac{dc_{Mn}}{dx}\bigg|^{austenite}_{\text{interface}}$$

$$(c^{\alpha\gamma}_{C} - c^{\gamma\alpha}_{C})v_{\text{interface}} = -D^{BCC}_{C}\frac{dc_{C}}{dx}\bigg|^{ferrite}_{\text{interface}} - D^{FCC}_{C}\frac{dc_{C}}{dx}\bigg|^{austenite}_{\text{interface}}$$

We can also describe displacive transformations in multi-component systems

- Software Tools
  - JMatPro®
  - QuesTek - Software
  - Databases, Strain Energy
  - Phenomenological: JMatPro
- Basis: Free energy change at T-zero and below should allow for strain energy
- Current focus 1: Generic models for wide range of alloy systems
- Current focus 2: Using these models to develop new steels



Formation of Martensite as well as Tempering Can be Predicted