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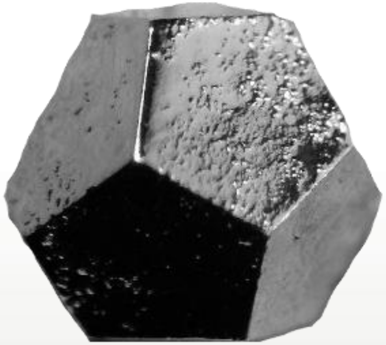

Effect of Magnetism on Lattice Dynamics:
Mössbauer Spectroscopic View

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INTRODUCTION (1)

♦ Figure of merit, adiabatic parameter (standard EPI theory):

$$\gamma = h\nu_D/E_F \sim (m/M)^{1/2} \ll 1 \qquad (1)$$

(ν_D is the Debye frequency, E_F is the Fermi energy, m and M are the electron and ion masses, respectively).

(1) $\rightarrow$ Effect of magnetism on lattice vibrations is neglegible.

♦ Kim's theory [1]:
For itinerant ferromagnetmagnets the effect can be enhanced by a factor of up to $\sim 10^2$.

[1] D. J. Kim, Phys. Rep., 171 (1988) 129

INTRODUCTION (2)

Controversies over the standard EPI theory (examples):

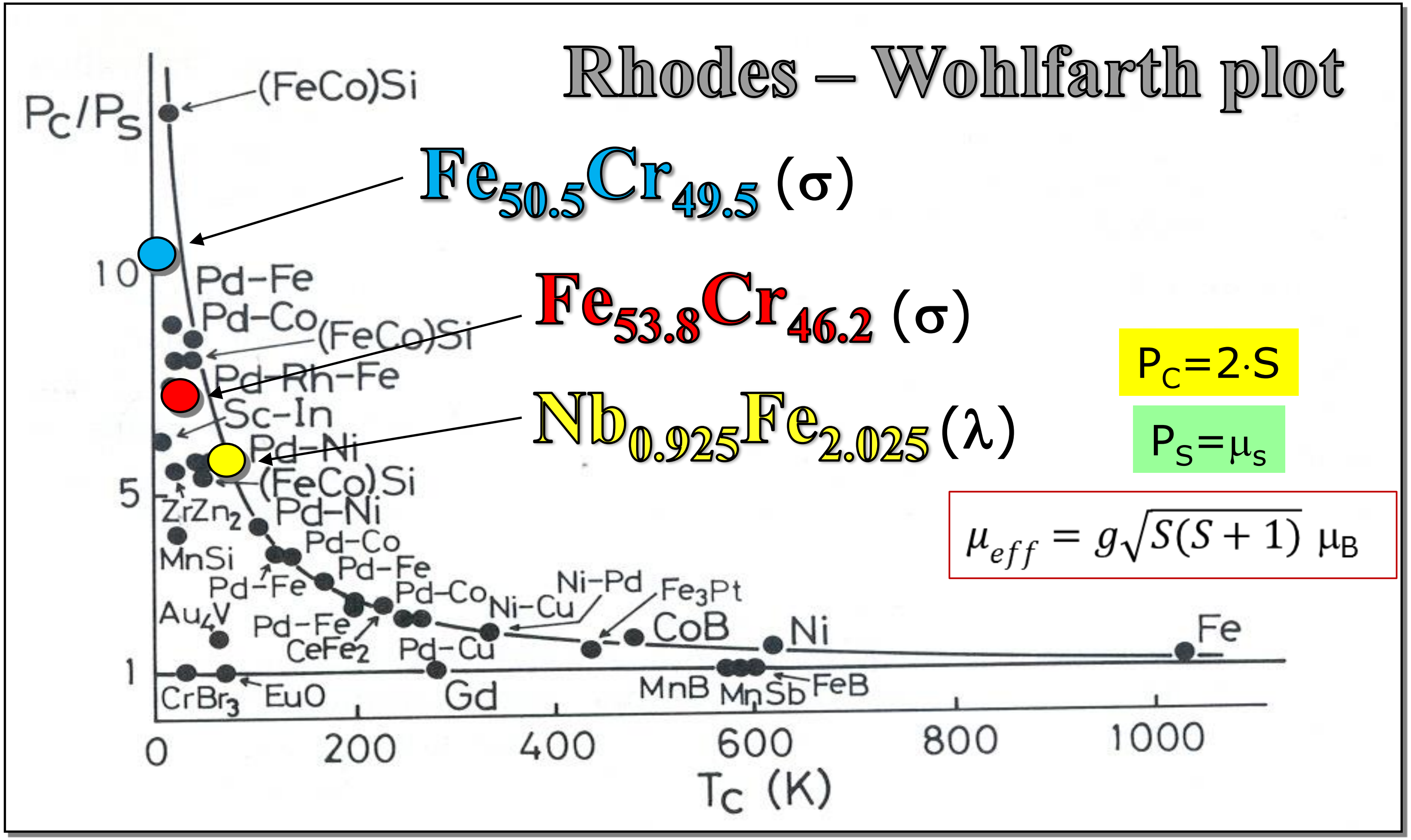
♦ *This theory neglects the effect of magnetism on lattice dynamics and fails to explain enhancement of the critical temperature in phonon-mediated superconductors* (I. S. Tupitsyn et al., PRB, 94 (2016) 155145).

♦ *Lattice vibrations strongly affect the distribution of local magnetic moment in paramagnetic Fe viz. they weaken their mean values* (B. Alling et al., PRB, 93 (2016) 224411).

♦ *The PDOS of bcc Fe-V alloys across the full composition range were studied by inelastic neutron scattering, nuclear resonant inelastic x-ray scattering, and ab initio calculations. Changes in the PDOS were revealed at crossing the Curie temperature* (M. S. Lucas et al., PRB, 82, (2010) 144306).

RHODES-WOHLFARTH CRITERION

- Ferromagnet is itinerant if $P_C / P_S > 1$



R. Rhodes and E. P. Wohlfarth, Proc. R. Soc. London, 273, 247 (1963)

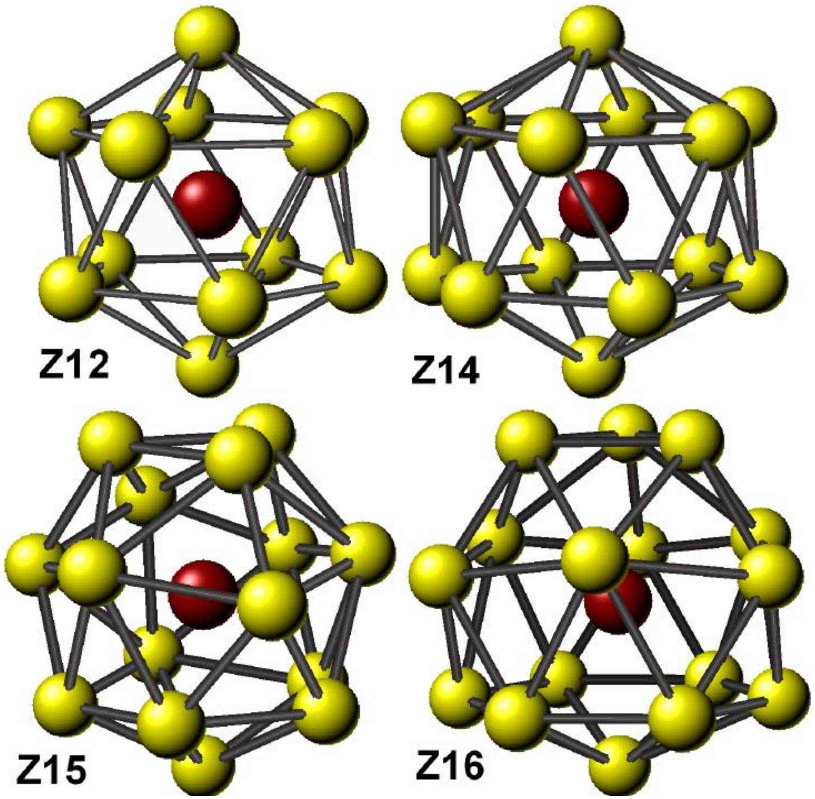
EXAMPLES TO BE PRESENTED

- ♦ Frank-Kasper or Topologically Close-Packed (TCP) Phases:
  - σ-phase: Fe-Cr and Fe-V
  - λ-phase (Laves): $NbFe_2$

- ♦ Chromium:
  - S.C. Cr(^{119}Sn)-ISDW and Cr3%Mn(^{119}Sn)-CSDW (AF)
  - Polycrystalline Cr (^{57}Fe)

FRANK-KASPER PHASES

- Intermetallic compounds with high coordination numbers (CN): $12 \le CN \le 16$.
- Most popular F-K: A15, λ (Laves), σ, μ, χ, M, P, and R.
- Complex unit cell with ≥ 2 lattice sites and many atoms e.g. λ(3), μ(13), σ(30), M(52), R(53), P(56).
- Different physical properties depending on a compound's constitution and composition: A15 (Nb_3Sn) superconductive.



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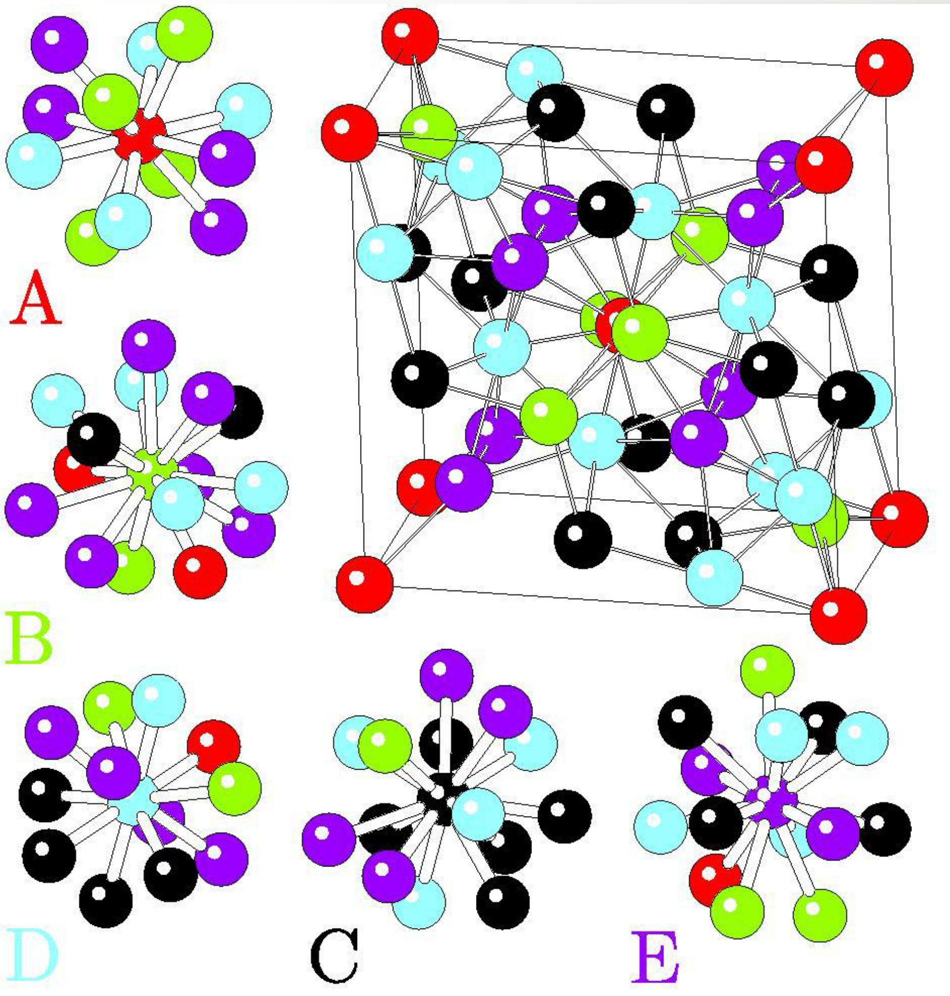
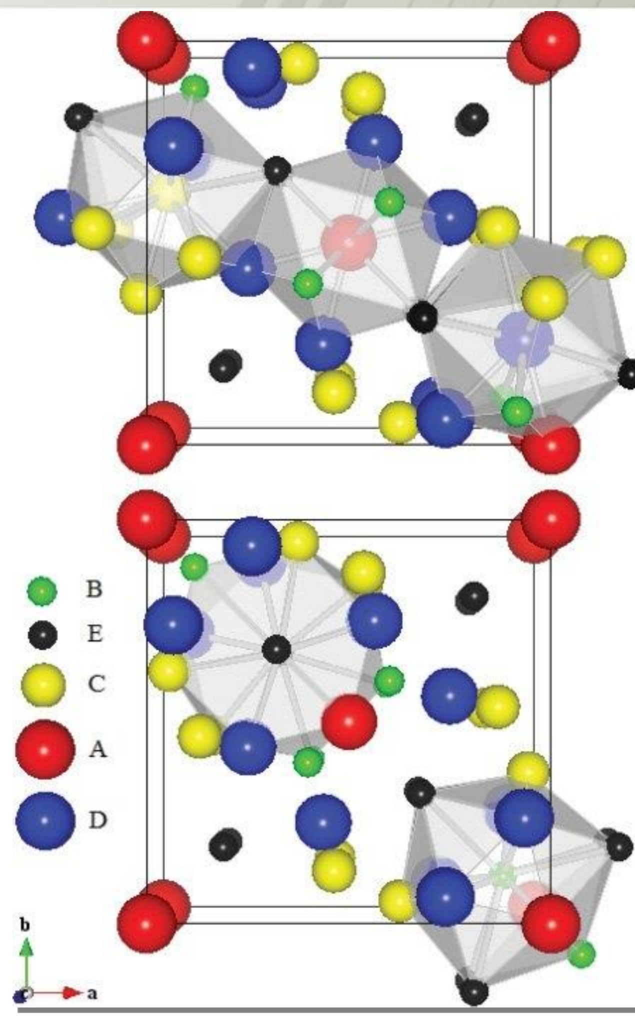
UNIT CELL

σ



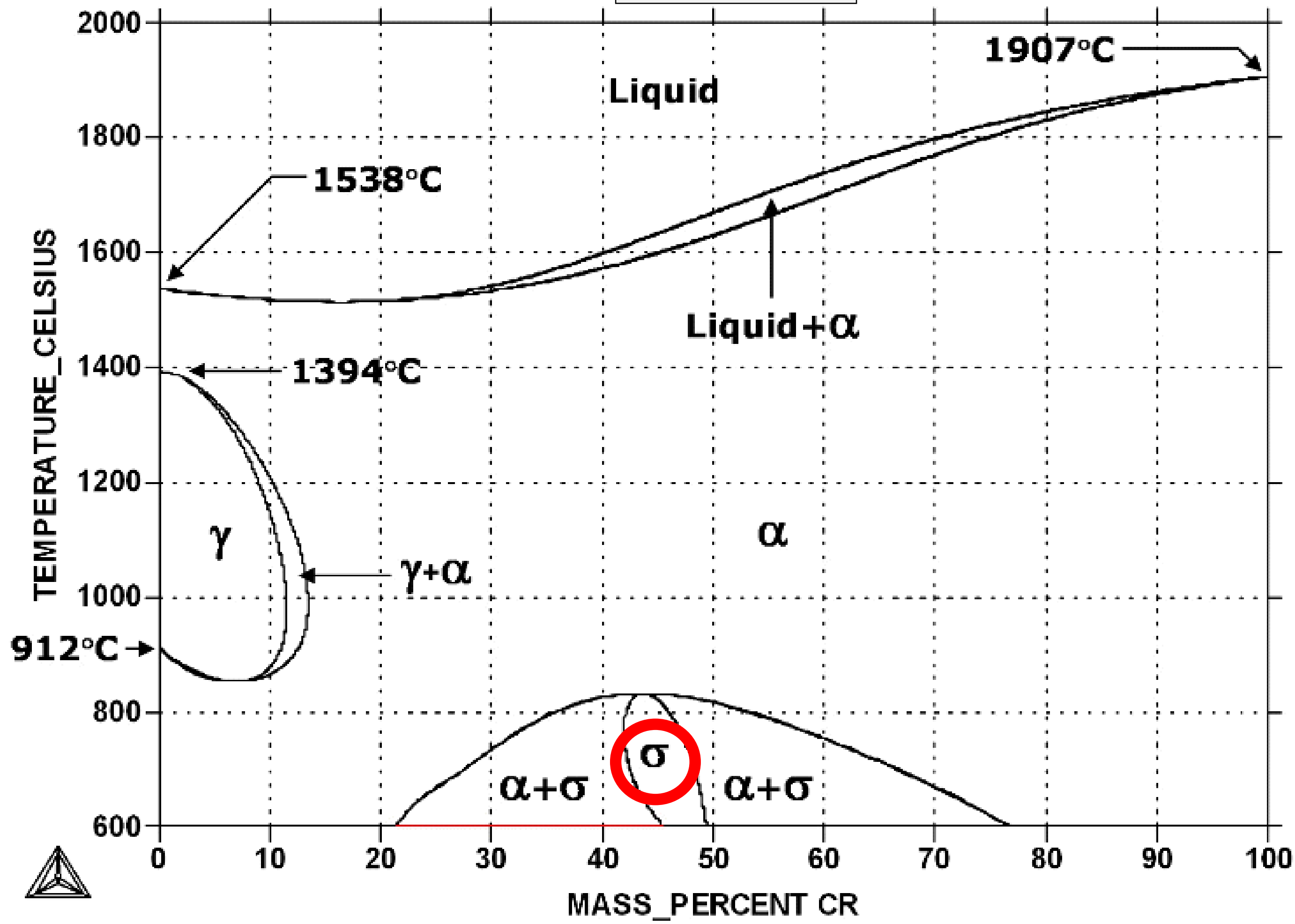
J. Cieslak et al., PRB (2010)



P. Blaha et al., PRB (2011)

CRYSTALLOGRAPHIC PHASE DIAGRAM

Fe-Cr



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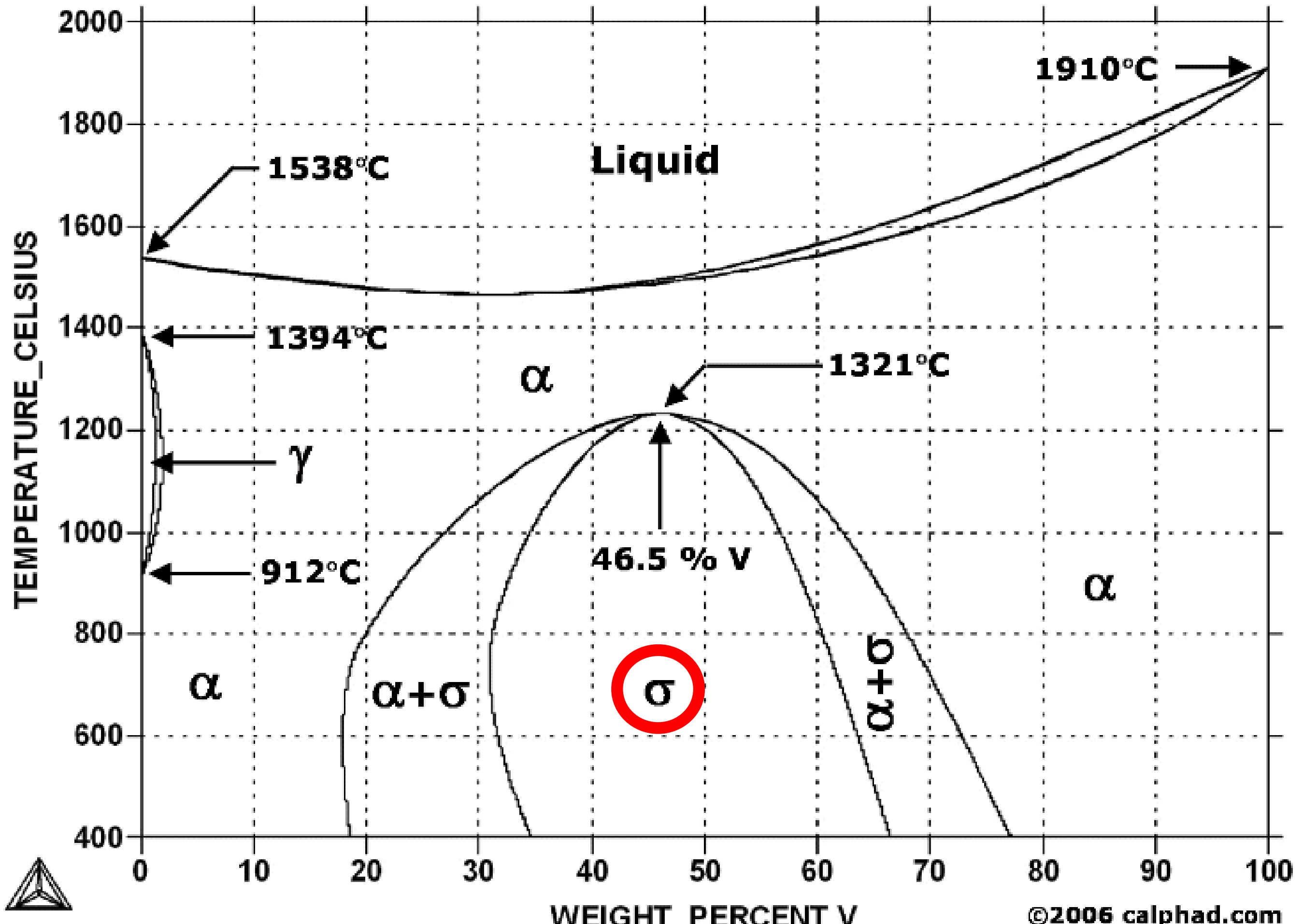
CRYSTALLOGRAPHIC PHASE DIAGRAM

Fe-V



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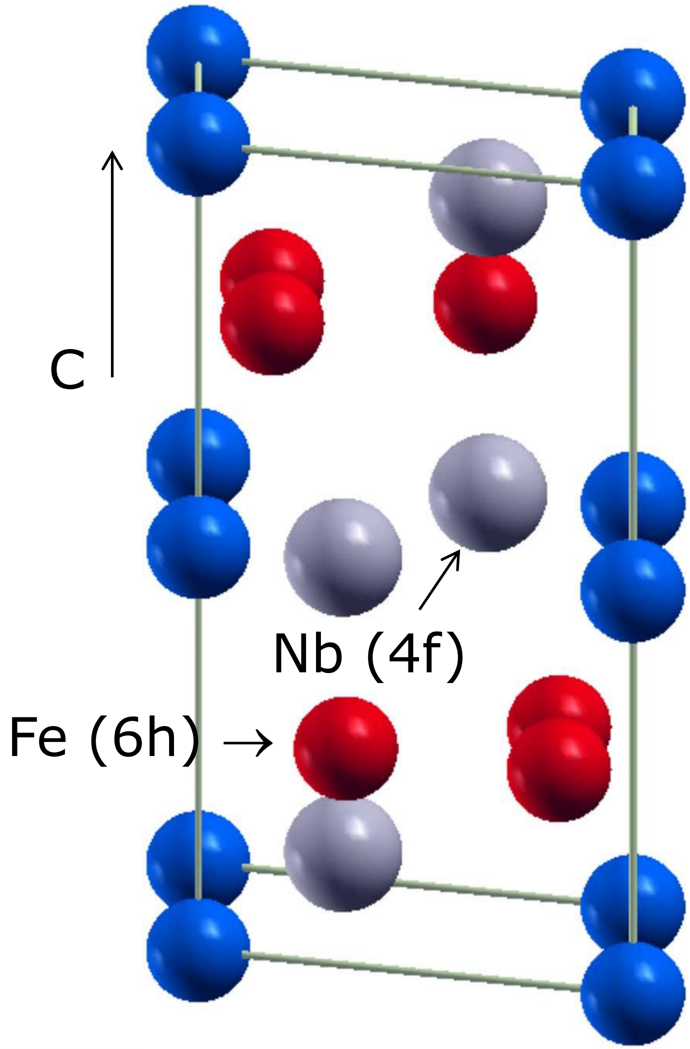
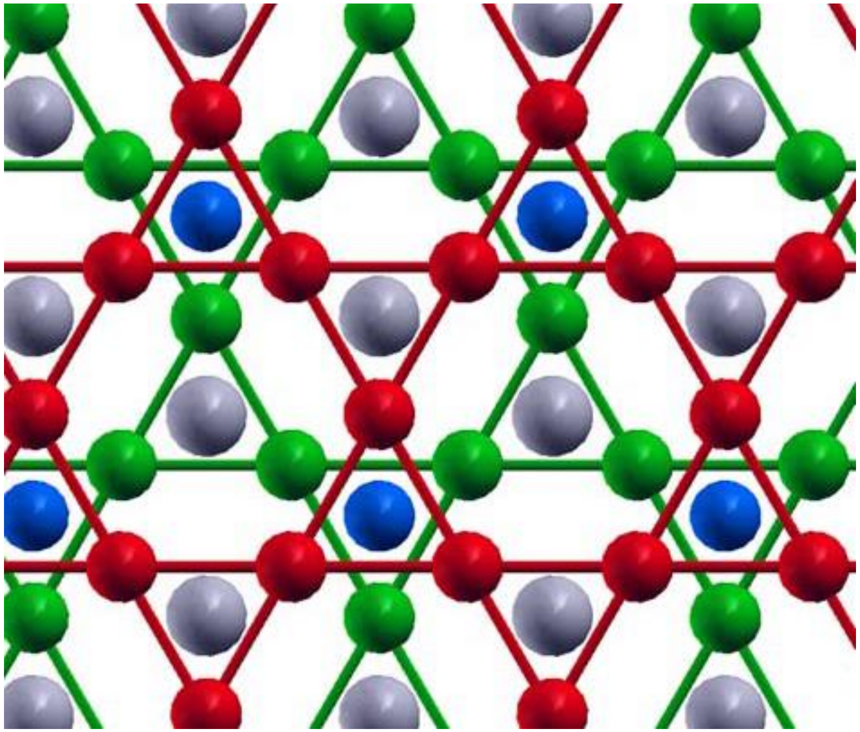
UNIT CELL
λ (C14)



2a:6h = 1:3

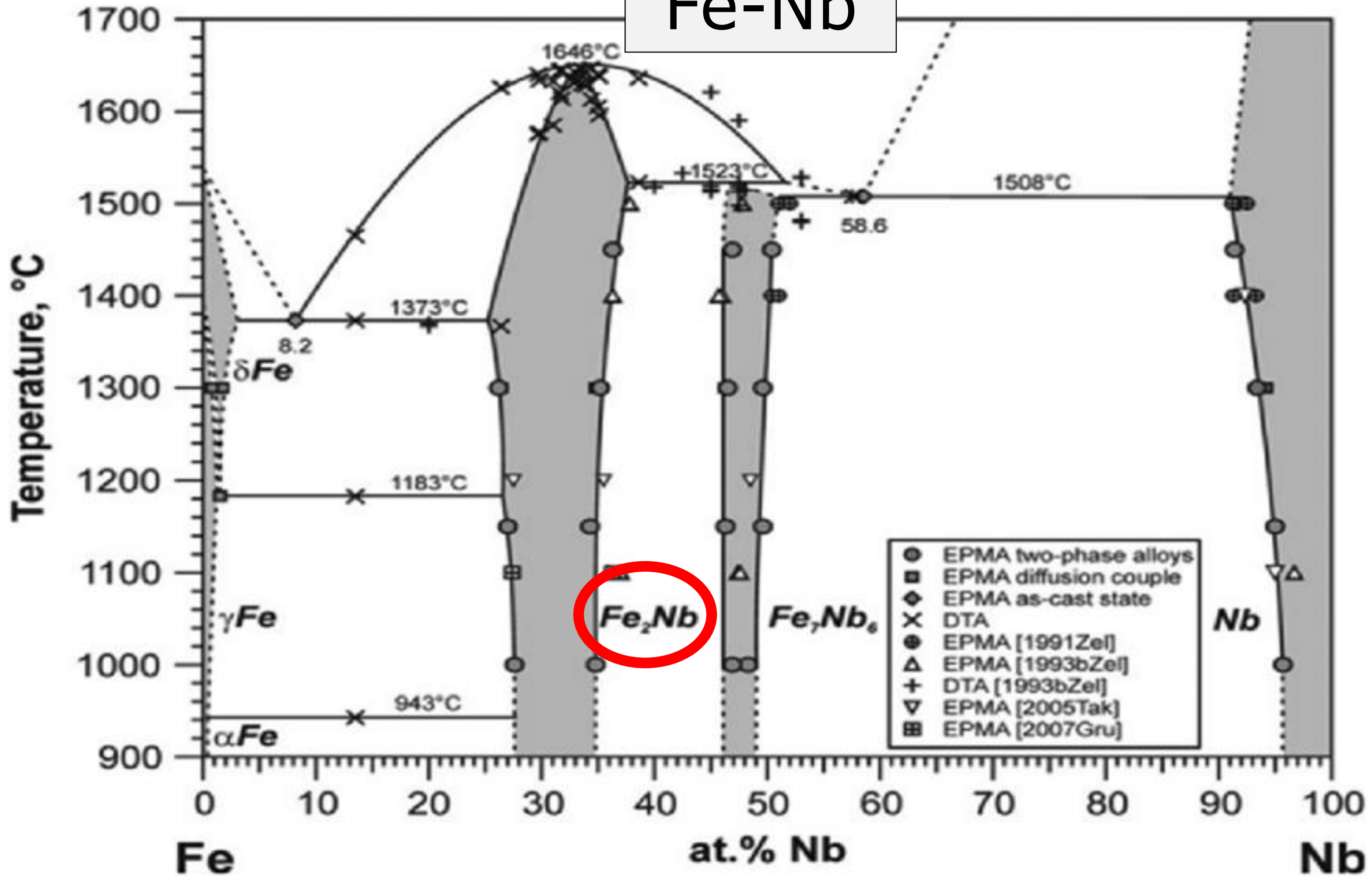


View ⊥ c

D. A. Tompsett et al., PRB, 82, 155137 (2010)

CRYSTALLOGRAPHIC PHASE DIAGRAM

Fe-Nb



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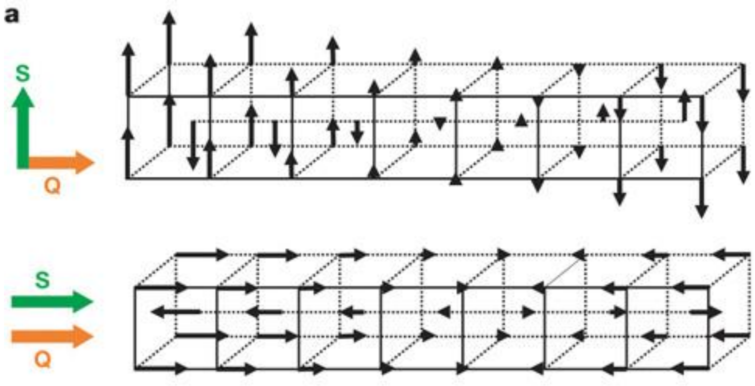
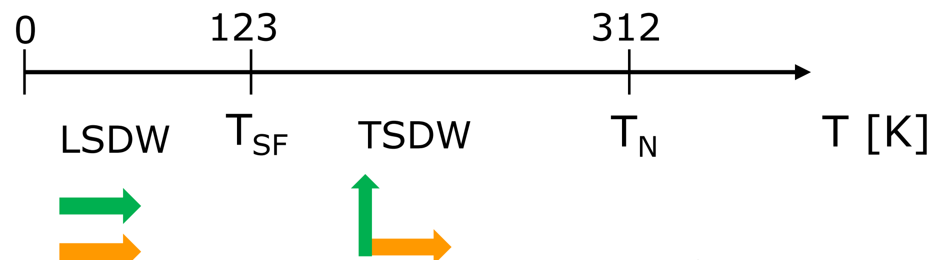
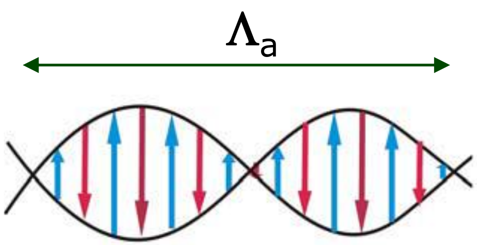
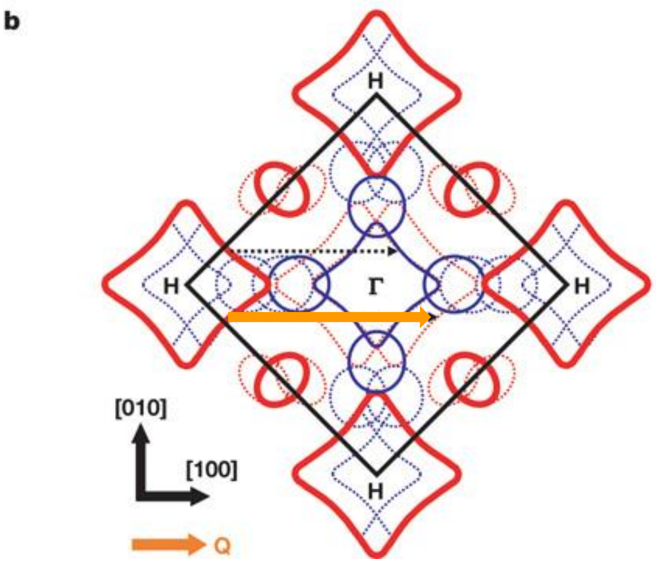
CHROMIUM - ISDW







$$\mu = \mu_1 \sin Qr$$



$$Q = \frac{2\pi(1-\delta)}{a}$$

$$\Lambda = \frac{2\pi}{Q} = \frac{a}{1-\delta}$$

$$\Lambda_a = \frac{a}{\delta}$$

T[K]	Λ_a [a]	Λ_a [Å]
4	~20	~57
312	~27	~78

EXPERIMENTAL TOOLS

- Mössbauer spectroscopy:

$$SOD = -\frac{E_\gamma}{2c^2}\left\langle v^2 \right\rangle$$

⇒ $E_k=0.5m<v^2>$

$$f = e^{-k^2<x^2>}$$

⇒ $E_p=0.5K^2<x^2>$

EXPERIMENTAL TOOLS

- Mössbauer spectroscopy (Debye model):

1. CS(T) =IS(0) + SOD(T)

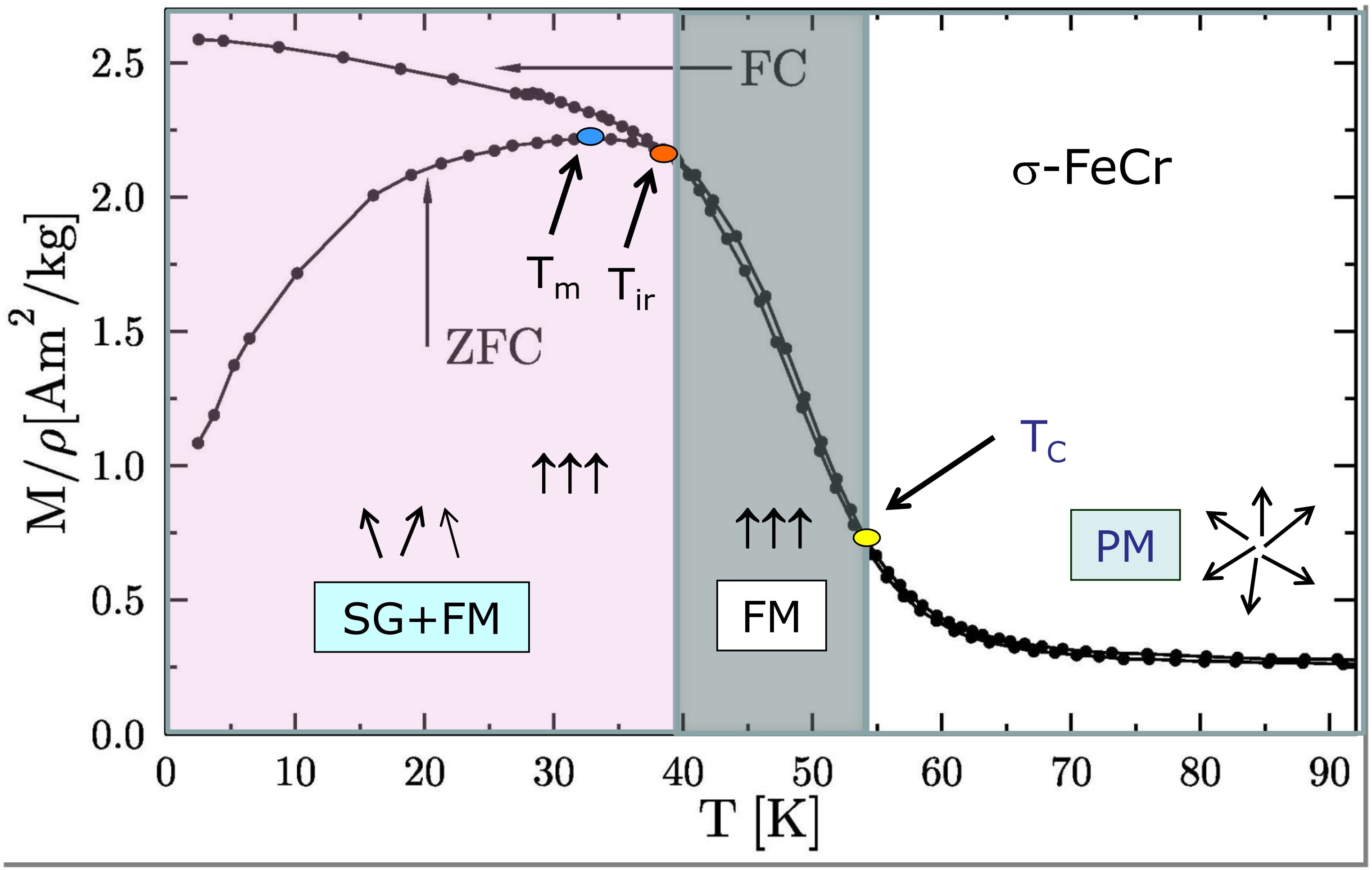
$$SOD(T) = \frac{3k_B T}{2mc}\left(\frac{3T_D}{8T} - 3\left(\frac{T}{T_D}\right)^3 \int_0^{T_D/T} \frac{x^3}{e^x - 1}dx\right)$$

2. f-factor (spectral area)

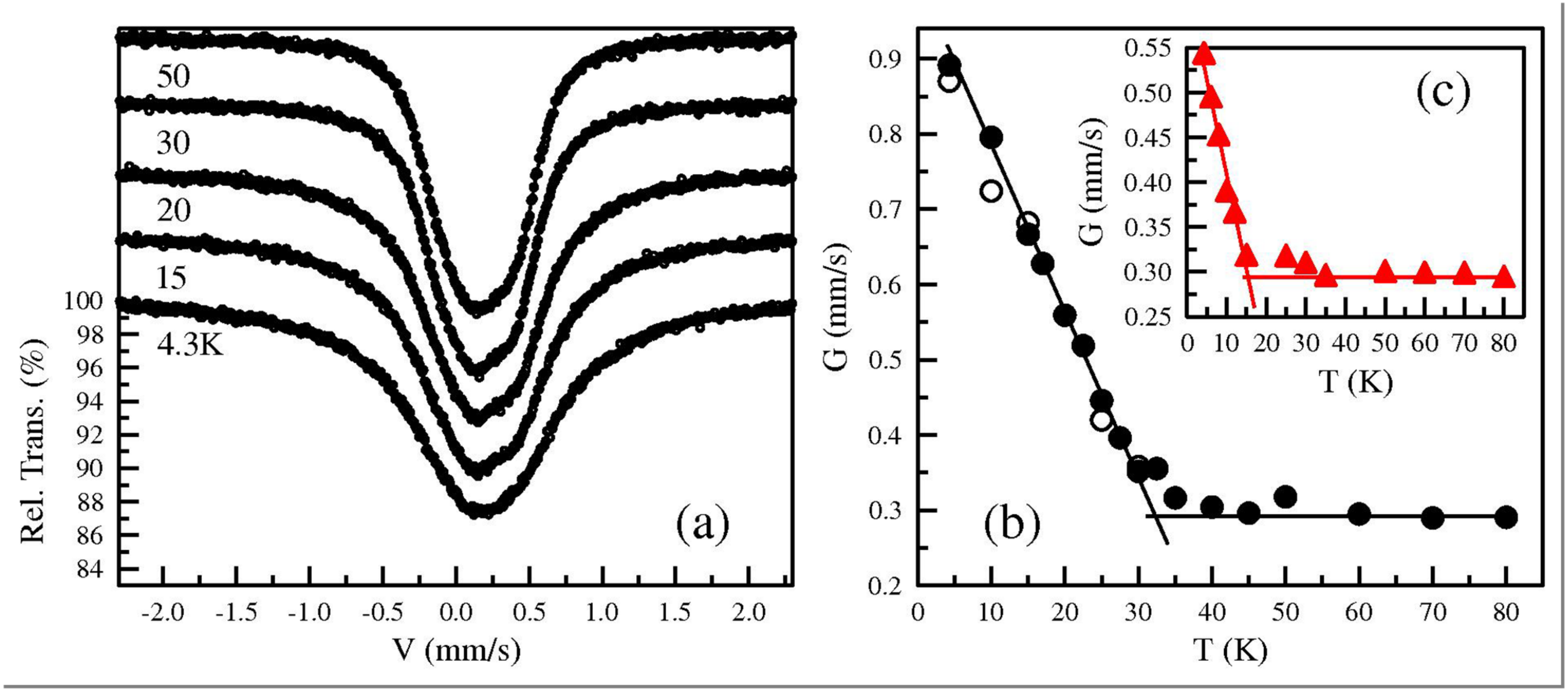
$$f = \exp[-\frac{6E_R}{k_B T_D}\{\frac{1}{4} + (\frac{T}{T_D})^2 \int_0^{T_D/T} \frac{x}{e^x - 1}dx\}]$$

RESULTS: σ-FeCr

- ***Re-entrant*** **type (PM→FM→SG+FM)**



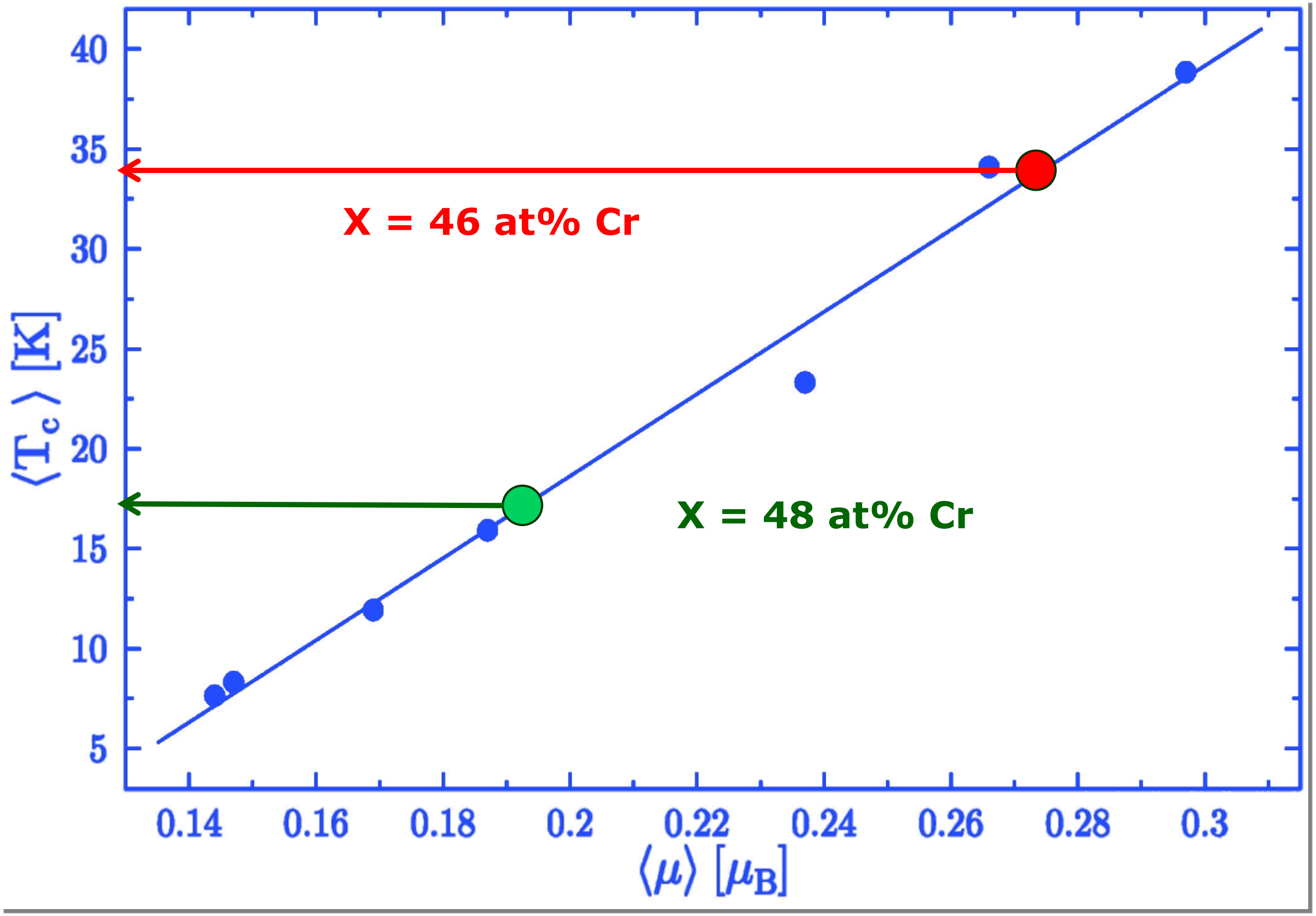
RESULTS: σ-FeCr



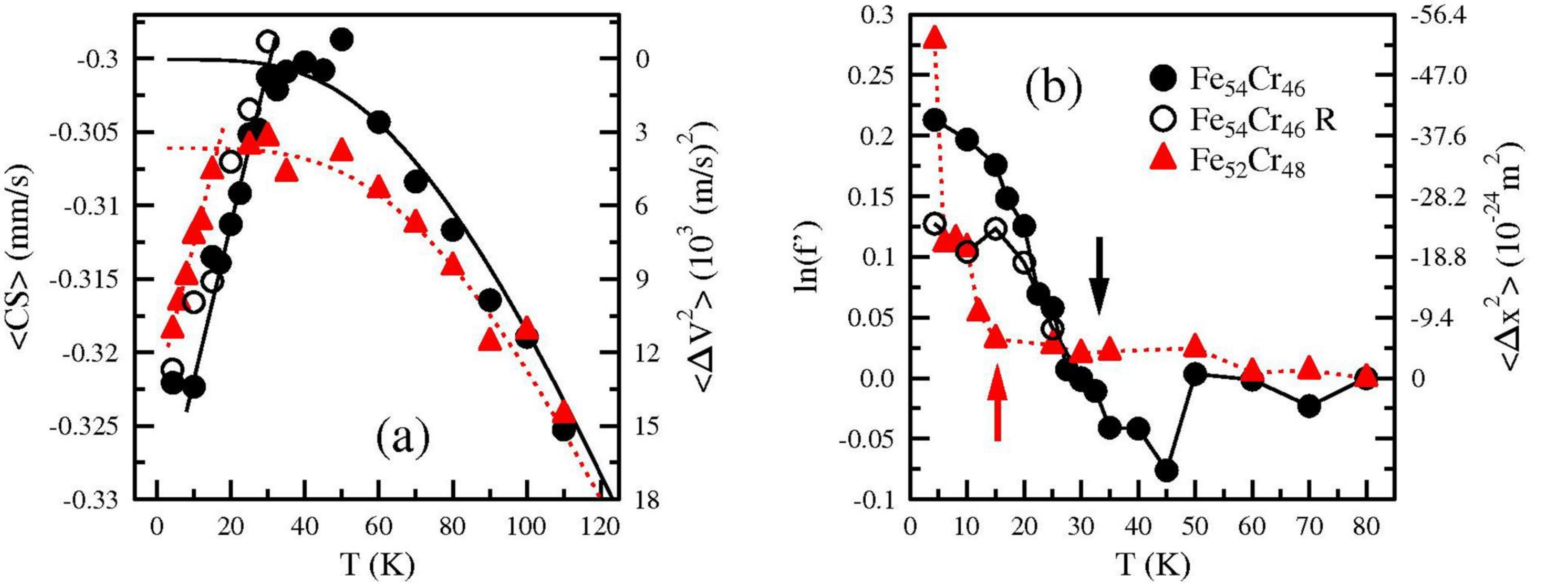
T_C: ~32 K for x =46 at%Cr; ~15 K for x=48 at% Cr

S. M. Dubiel et al., EPL, 101 (2013) 16008

RESULTS: σ-FeCr



RESULTS: σ-FeCr

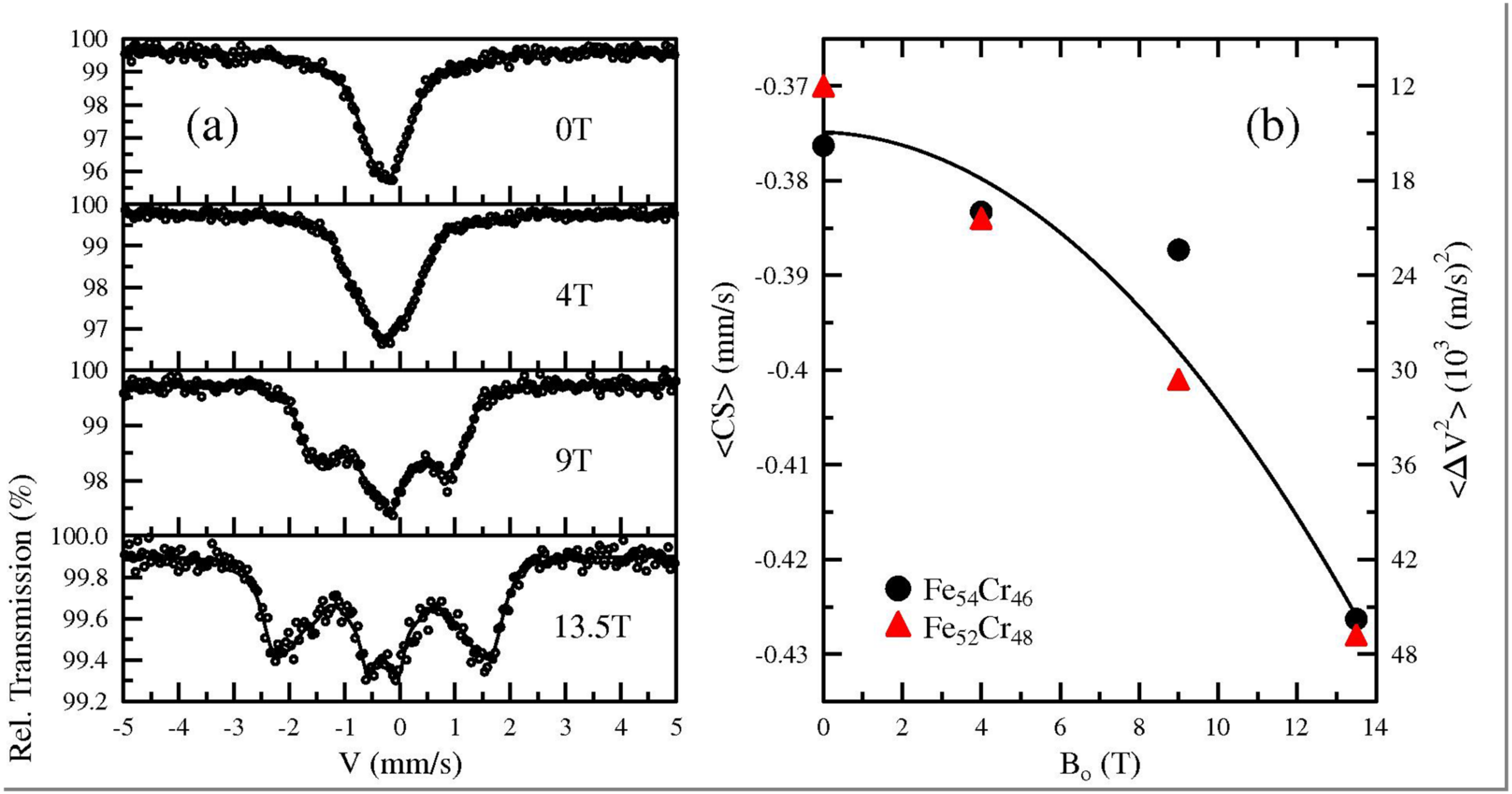


$<CS>=-0.5E_{\gamma}<v^2>/c^2$ $f=exp(-k^2<x^2>)$

There are anomalies both in <CS> and in *f* at $T \approx T_C$

S. M. Dubiel et al., EPL, 101 (2013) 16008

RESULTS: σ-FeCr



S. M. Dubiel et al., EPL, 101 (2013) 16008

<CS> follows the trend found from zero-field measurements

RESULTS: σ-FeCr

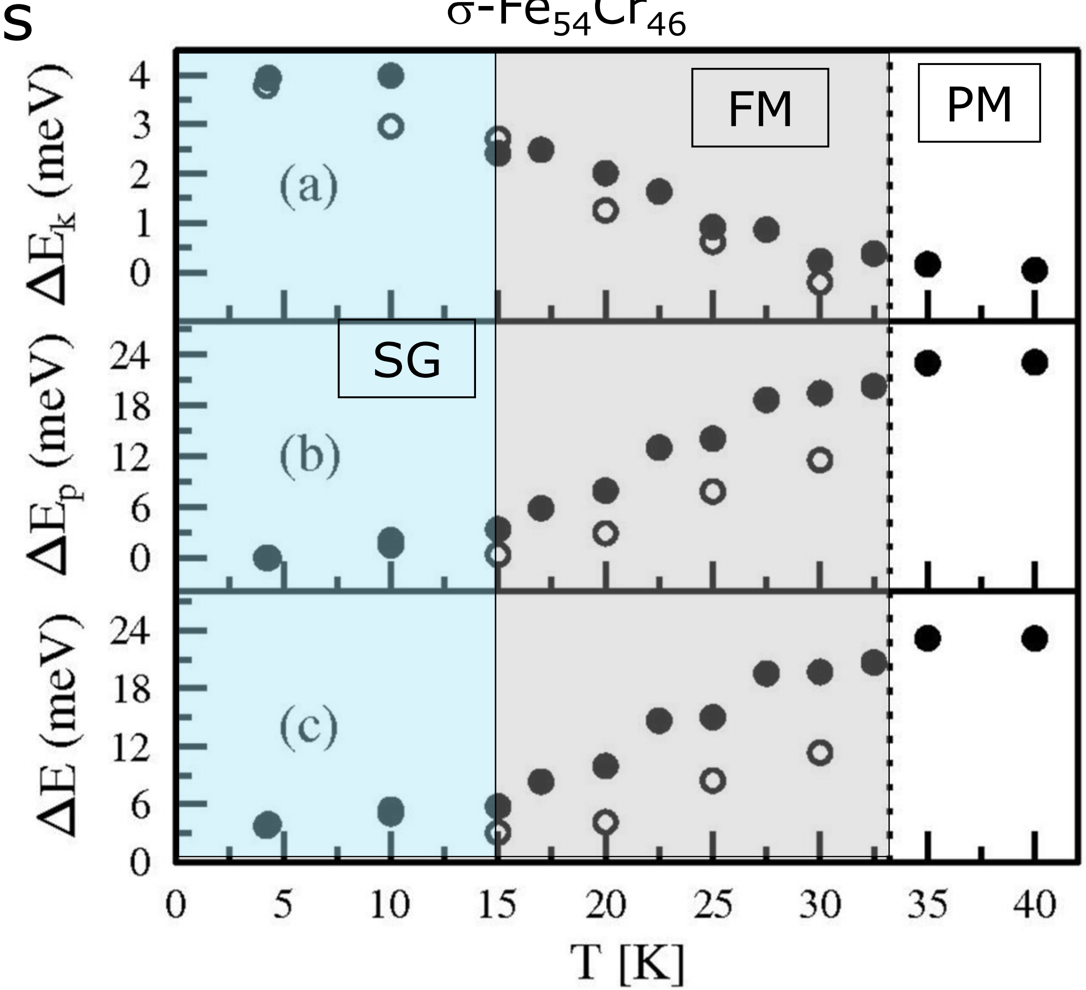
- Energetics of vibrations
  - Harmonic approximation: $E = E_k + E_p$

$$E_k = \frac{m < v^2 >}{2}$$

$$E_p = \frac{K < x^2 >}{2}$$

$$SOD = -\frac{E_\gamma}{2c^2}\langle v^2 \rangle$$ → $<v^2>$

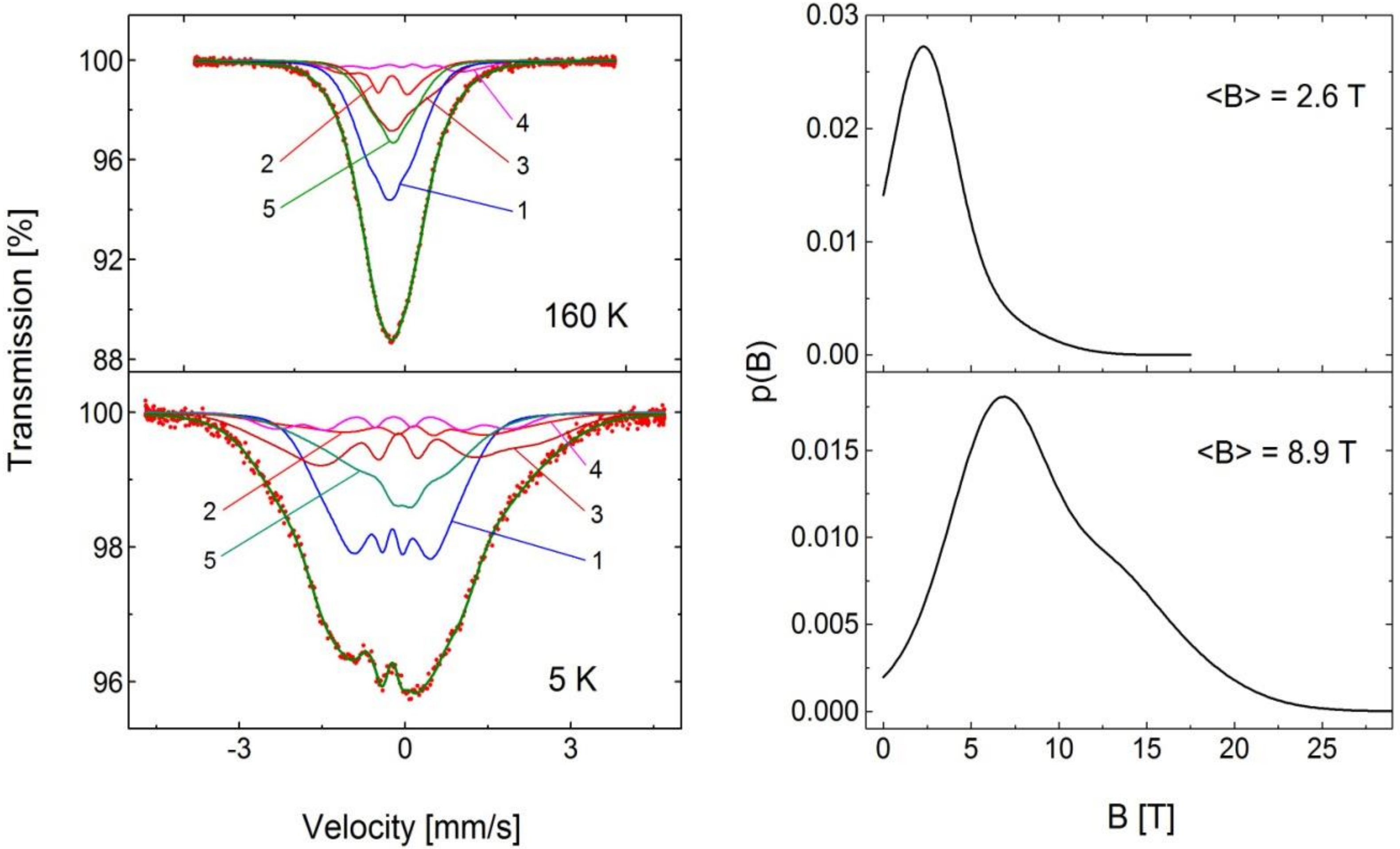
$$f = \exp\left(-k^2\langle x^2 \rangle\right)$$ → $<x^2>$



S. M. Dubiel et al., EPL, 101 (2013) 16008

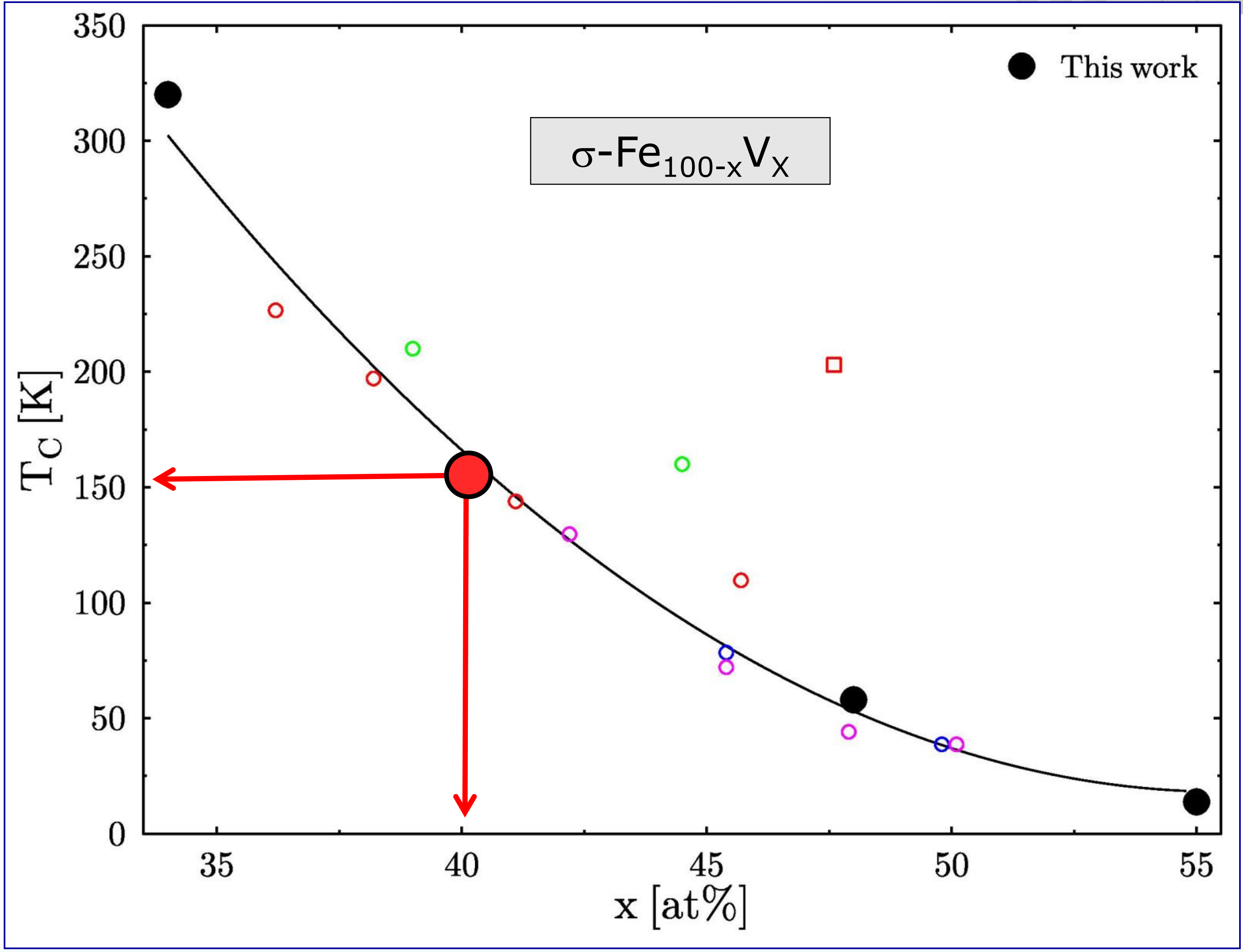
RESULTS: σ-FeV

- Mössbauer spectra: x_V=40 at% ($T_C \approx 160^oC$)



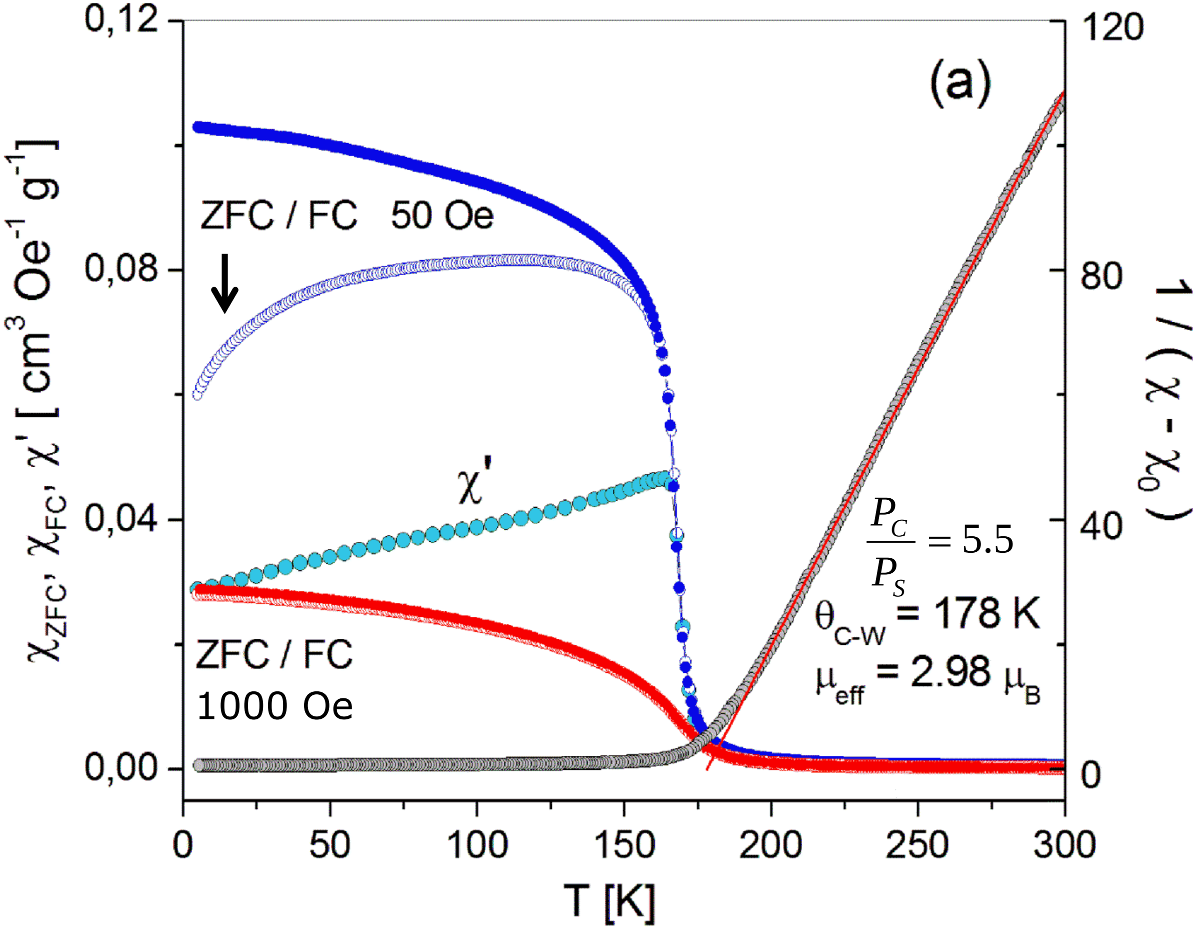
S. M. Dubiel, J. Zukrowski, JMMM, 441 (2017) 557–561

RESULTS: σ-FeV



RESULTS: σ-FeV

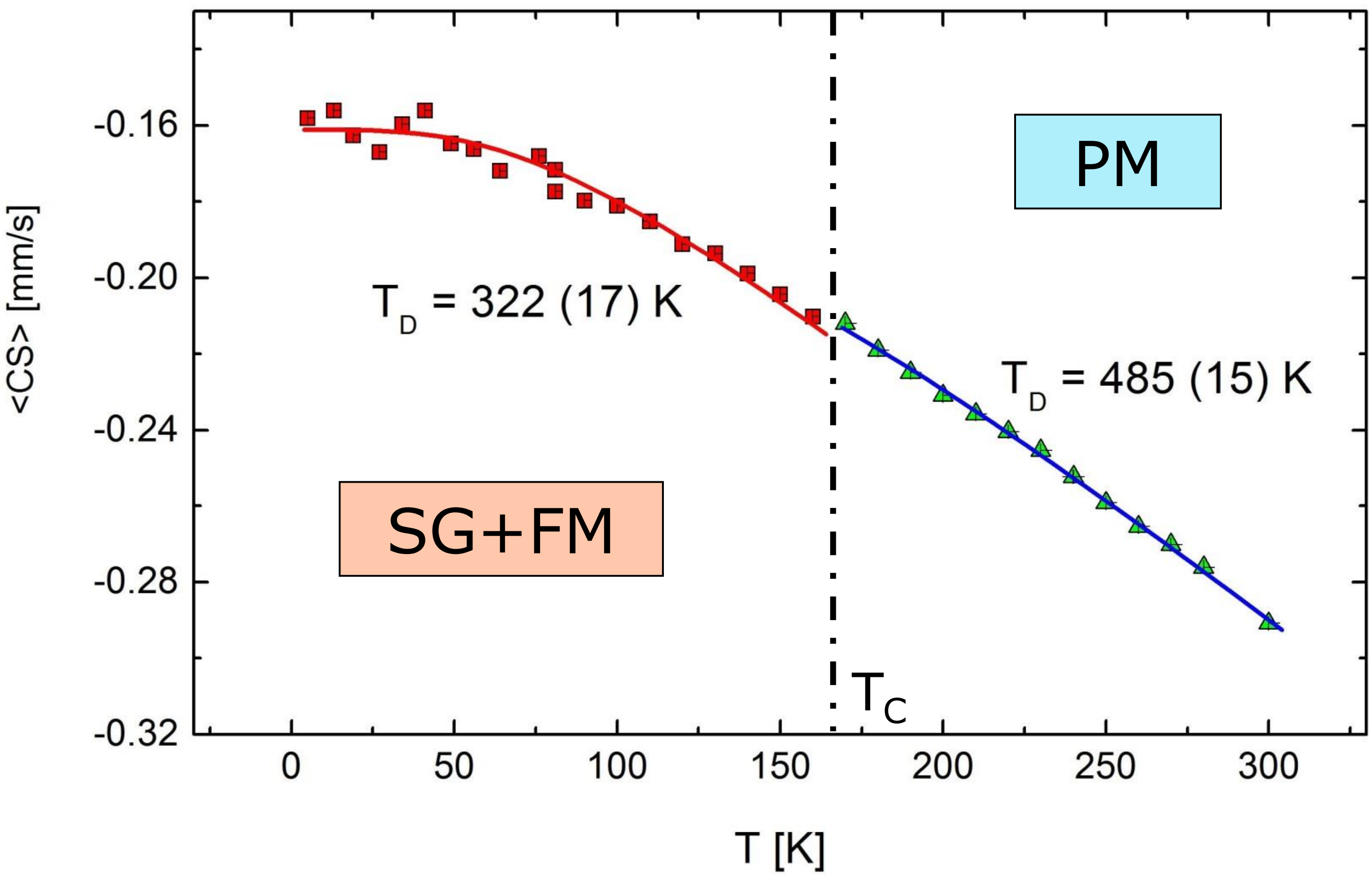
x_V=40 at%



M. Balanda et al., JMMM, 432 (2017) 430–436

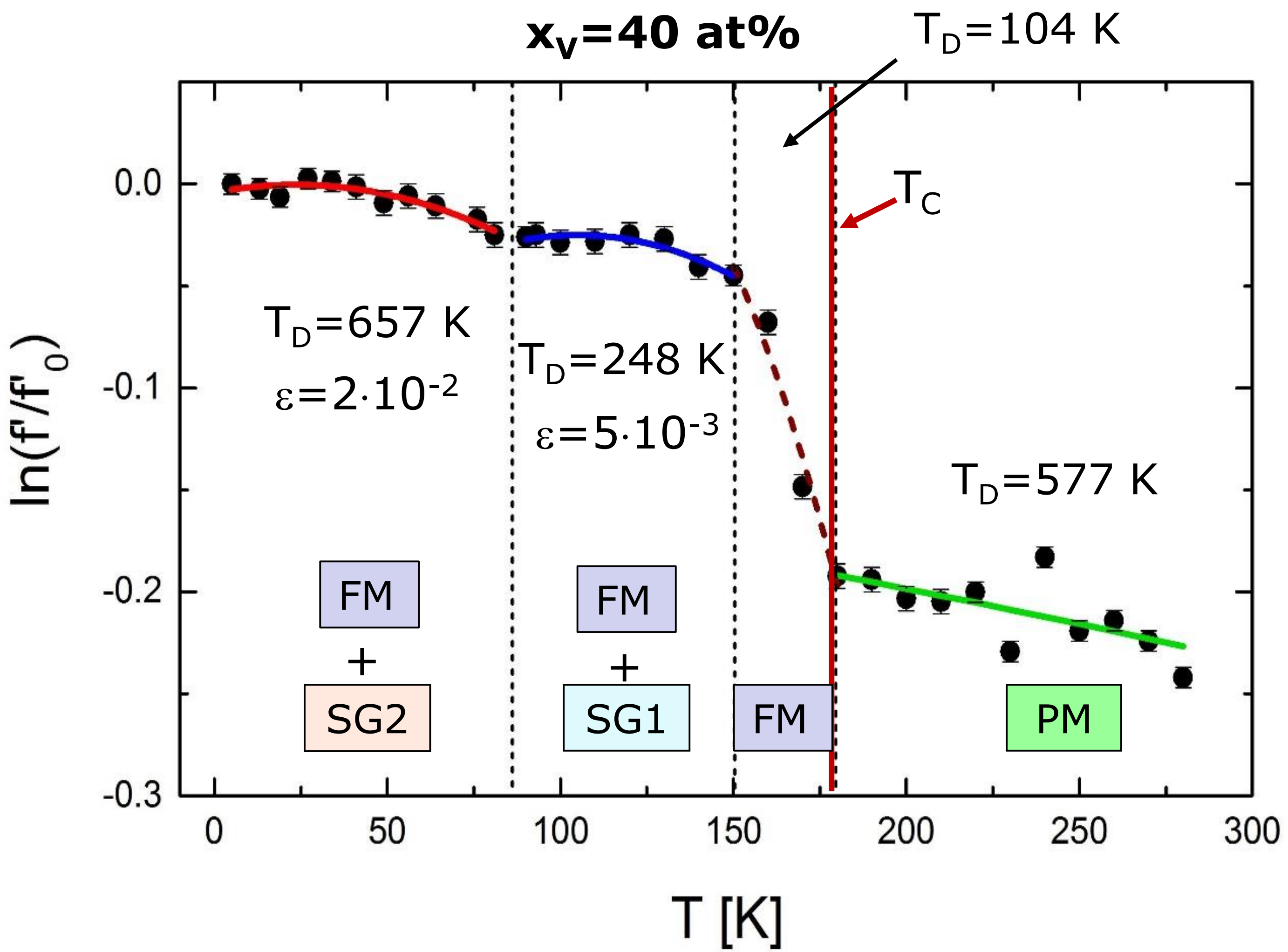
RESULTS: σ-FeV

- Debye temperature, T_D



S. M. Dubiel, J. Zukrowski, JMMM, 441 (2017) 557–561

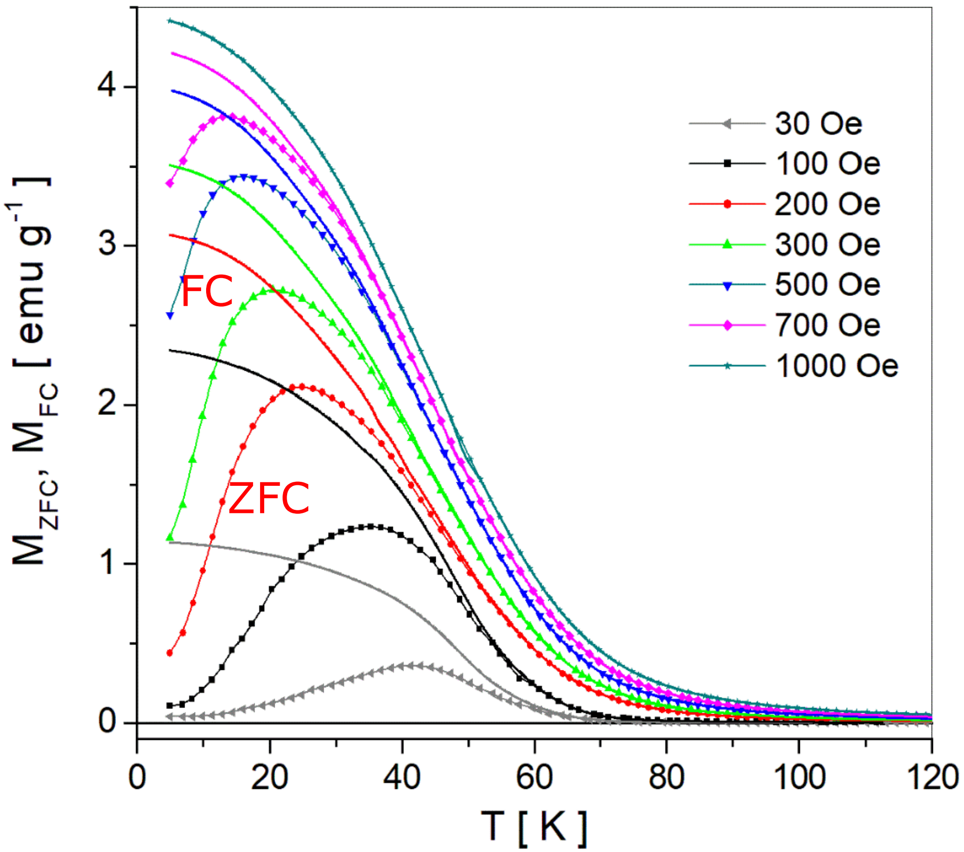
RESULTS: σ-FeV

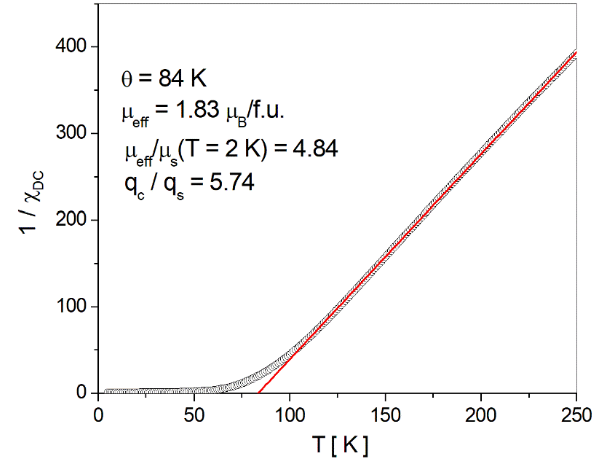


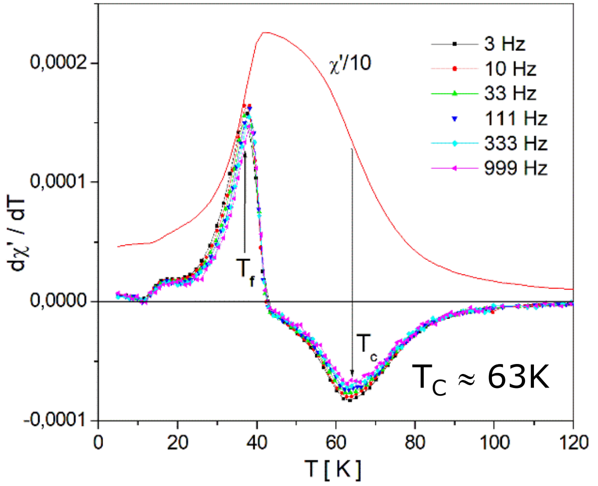
S. M. Dubiel, J. Zukrowski, JMMM, 441 (2017) 557–561

RESULTS: λ-$Nb_{0.975}$ $Fe_{2.025}$

- DC and AC magnetic susceptibilities



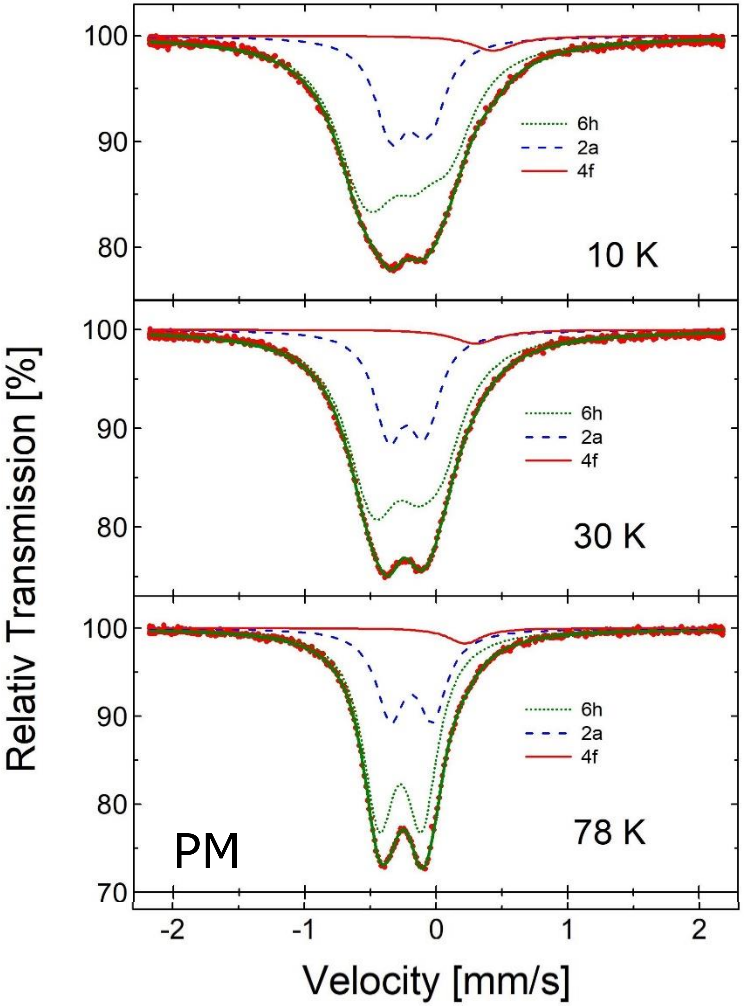


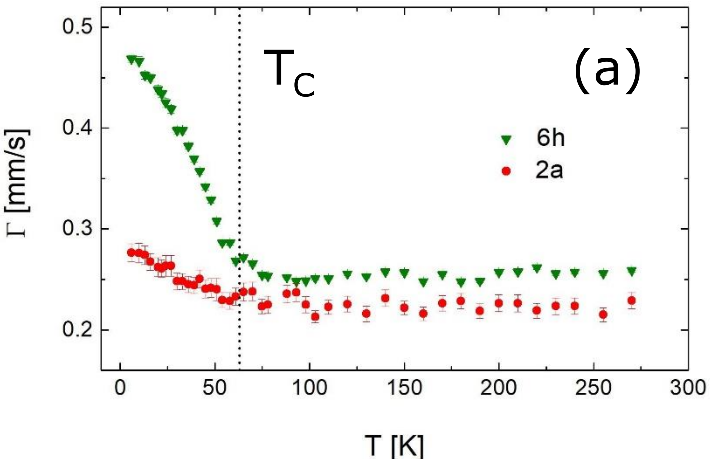


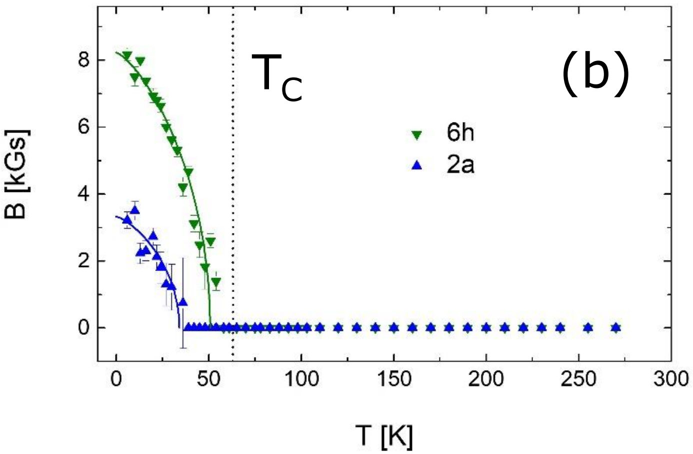
M. Balanda, S. M. Dubiel, JMMM, 454 (2018) 386

RESULTS: λ-$Nb_{0.975}$ $Fe_{2.025}$

♦ Mössbauer measurements



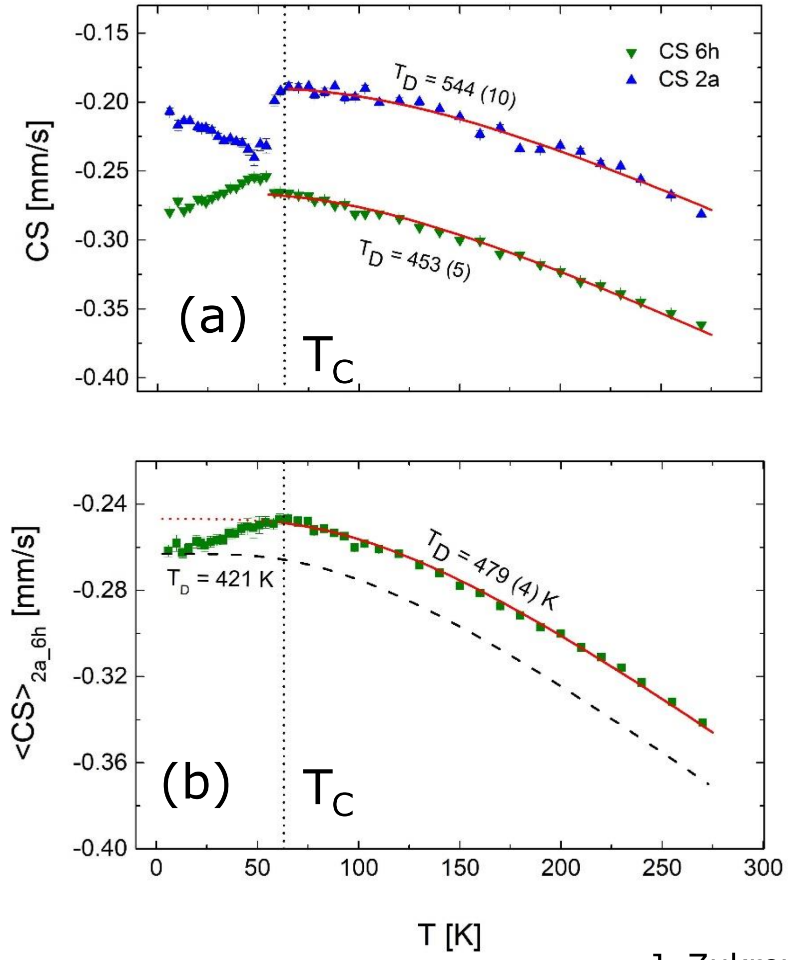


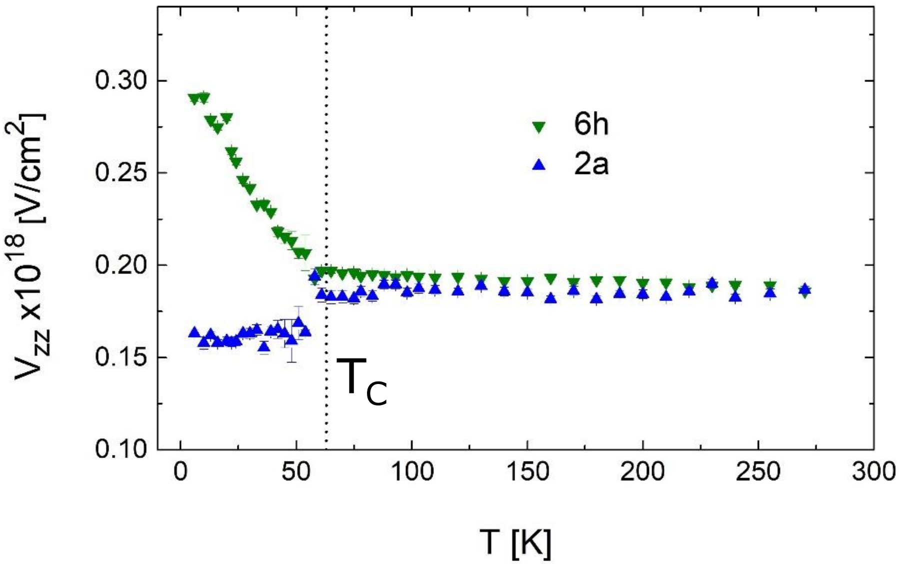


J. Zukrowski, S. M. Dubiel, JAP , 123 (2018) 223902

RESULTS: λ-$Nb_{0.975}$ $Fe_{2.025}$

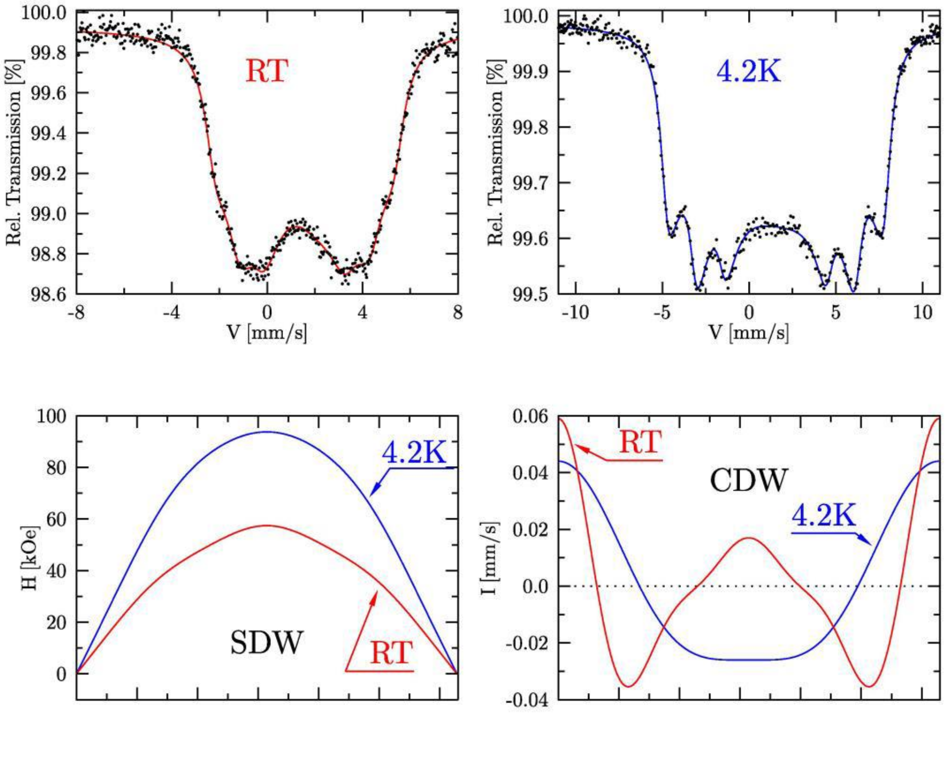
- Mössbauer measurements

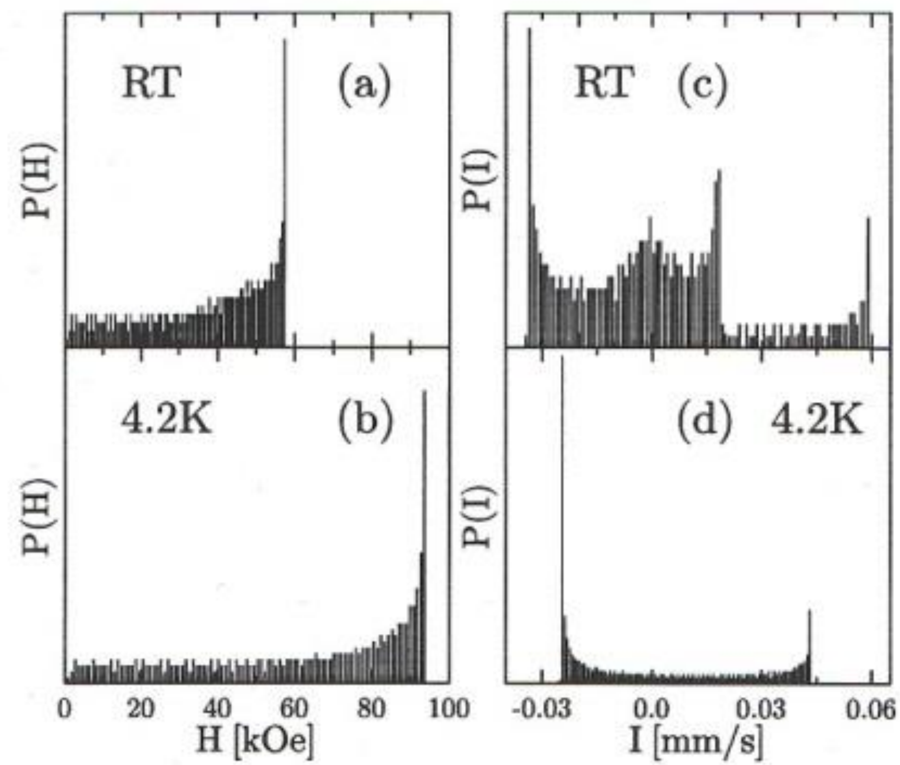




J. Zukrowski, S. M. Dubiel, JAP, 123 (2018) 223902

S.C.-CHROMIUM DOPED WITH ^{119}Sn



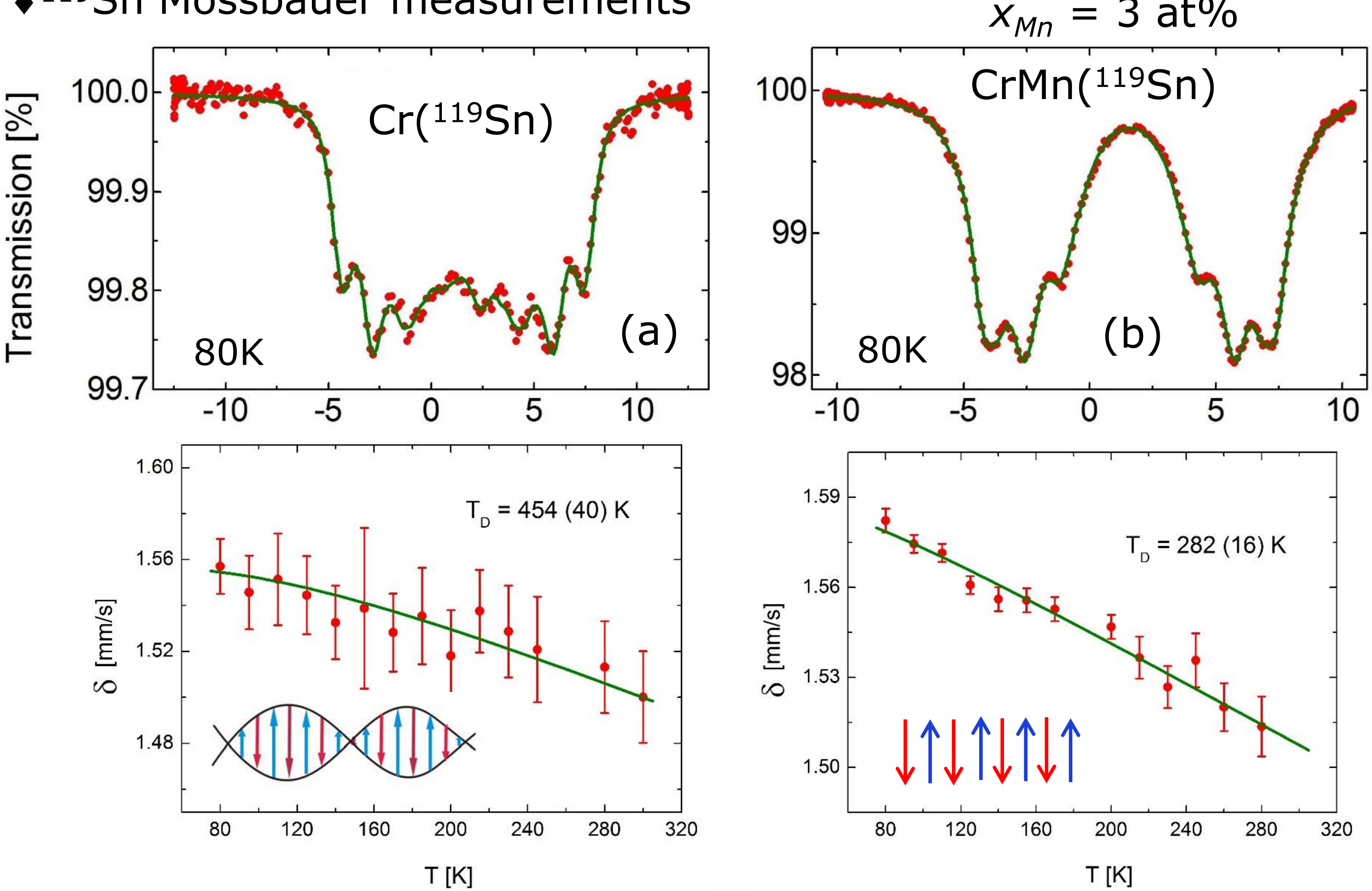


- Determination of H_3: 2.6% at 4.2K and 1.4% at 295 K (2.1% from ND at 144K – Tsunoda 1985)

S. M. Dubiel, G. LeCaer, Europhys. Lett., 4 (1987) 487; S. M. Dubiel et al., Phys. Rev. B, 53 (1996) 268

RESULTS: CHROMIUM

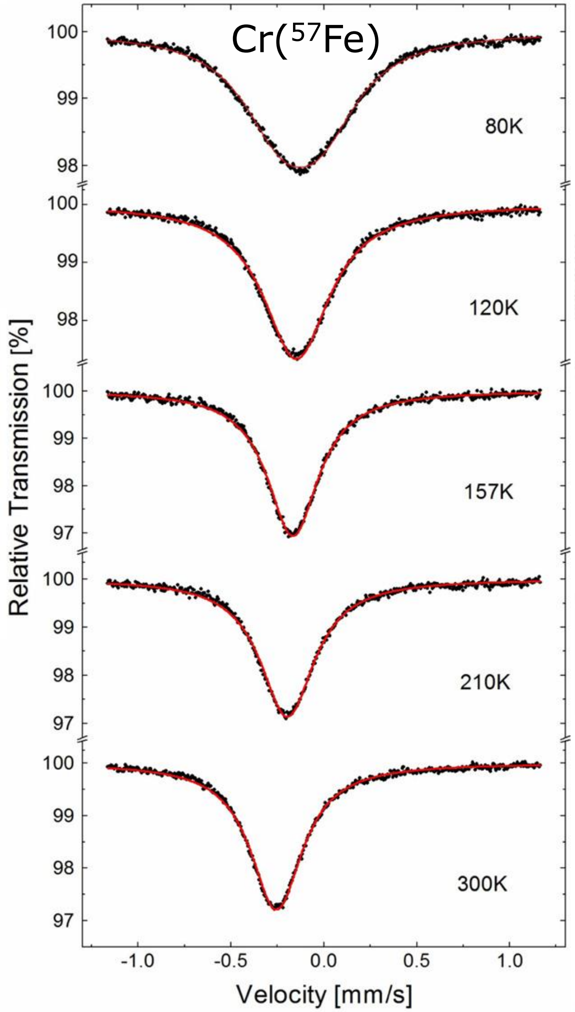
- ^{119}Sn Mössbauer measurements

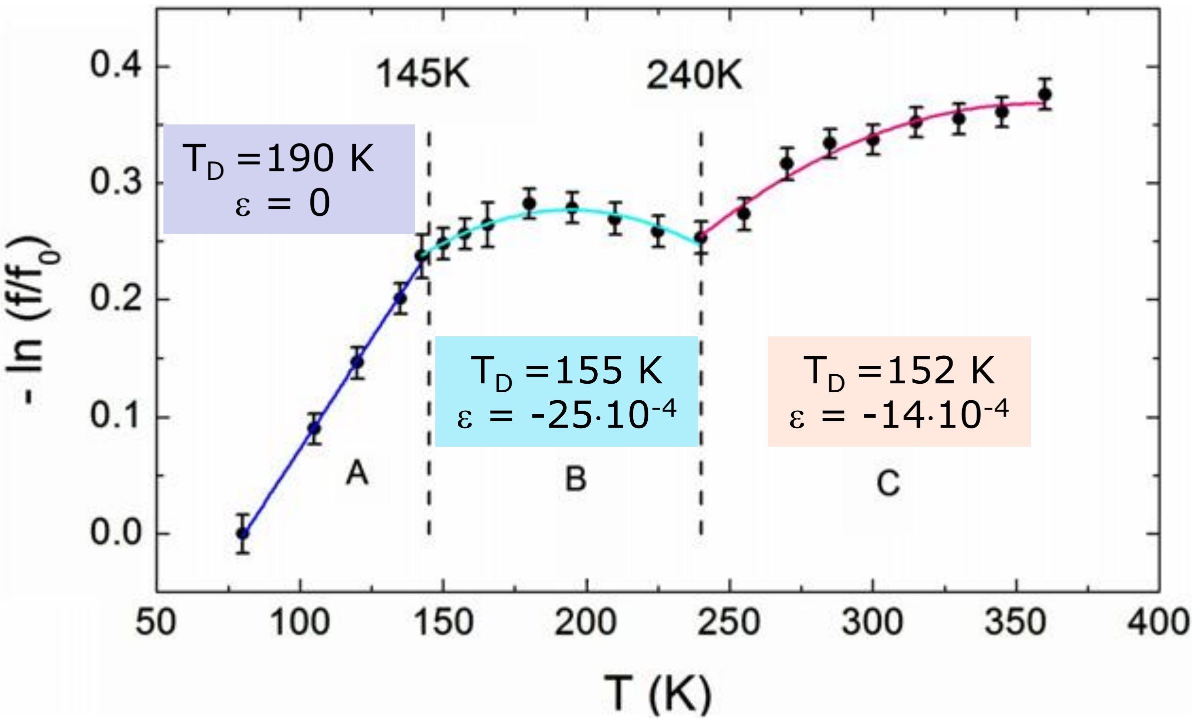


J. Zukrowski, S. M. Dubiel, EPL, 127 (2019) 26002

RESULTS: CHROMIUM

♦ ^{57}Fe Mössbauer measurements



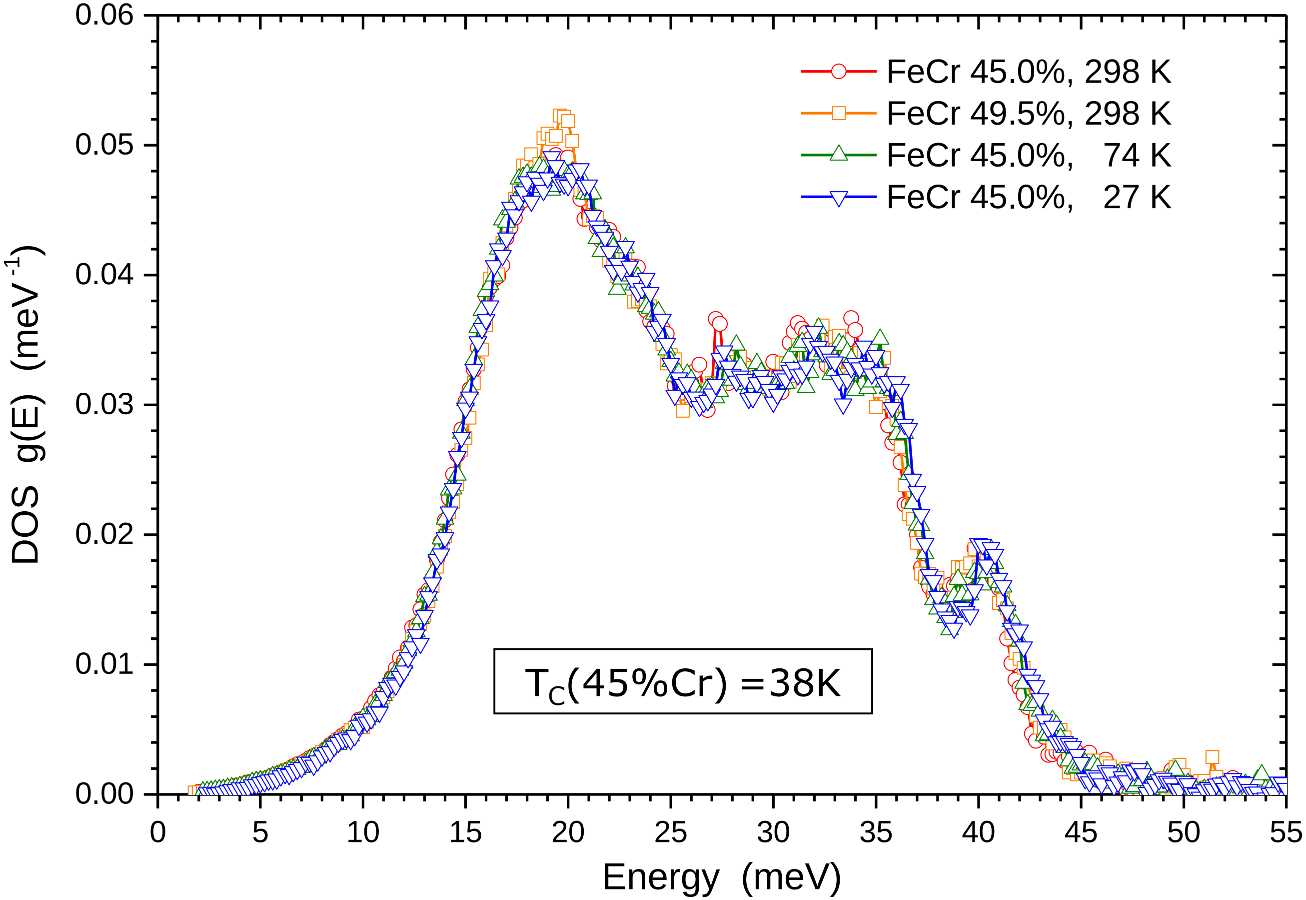


$$-\ln f / f_o = \ln f_o - \frac{6E_R T}{T_D^2}(1+\varepsilon T)$$

S. M. Dubiel et al., J. Phys.: Condens. Matter **22** (2010) 435403

NIS RESULTS: σ-FeCr

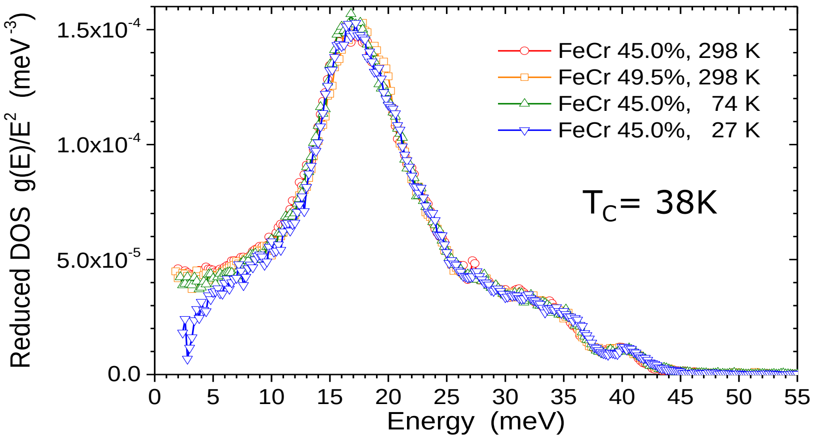
- Fe-PDOS for σ-FeCr



S.M. Dubiel, A. I. Chumakov. EPL, 117 (2017) 56001

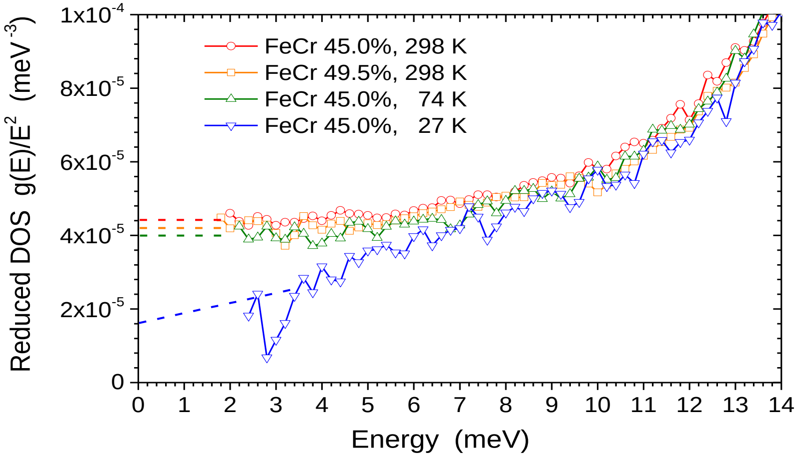
NIS RESULTS: σ-FeCr

- Effect of magnetism on sound velocity, <v>



a

$$\lim_{E\to 0}\frac{g(E)}{E^2}=\frac{m_R}{\langle m\rangle}\frac{1}{2\,\pi^2\,\hbar^3\,n\,\langle v\rangle^3}$$



b

x [at. % Cr]	T [K]	g(E)/E² [meV⁻³]	<v> [km/s]
45	298(1)	$4.4(2)\times10^{-5}$	3.68(7)
45	74.0(5)	$4.0(2)\times10^{-5}$	3.79(7)
45	27.0(8)	$1.6(4)\times10^{-5}$	5.1(4)
49.5	298(1)	$4.2(2)\times10^{-5}$	3.75(7)

Δ<v>/v=36% (!)

S.M. Dubiel, A. I. Chumakov. EPL, 117 (2017) 56001

CONCLUSIONS

Itinerant magnetism and externel magnetic field strongly affect lattice dynamics:

(1) Deviations from the Debye model are observed below T_C in σ and λ Frank-Kasper phases viz.:

(a) Average squared velocity of vibrations (kinetic energy) increases.

(b) Average squared amplitude of vibrations decreases.

(c) Strong unharmonicity exists in the SG state.

(2) Significant differences revealed by ^{119}Sn and ^{57}Fe probe atoms in Cr viz.:

(a) T_D-values revealed by ^{119}Sn are ~2-fold higher (Fe atoms are weakly coupled to SDWs).

(b) T_D-values obtained from ^{119}Sn spectra are significantly larger for ISDW than for CSDW (AF).

(3) Strong (~36%) increase in the average sound velocity was revealed for the σ-$Fe_{55}Cr_{45}$ compound in the magnetic state.

COLLABORATORS

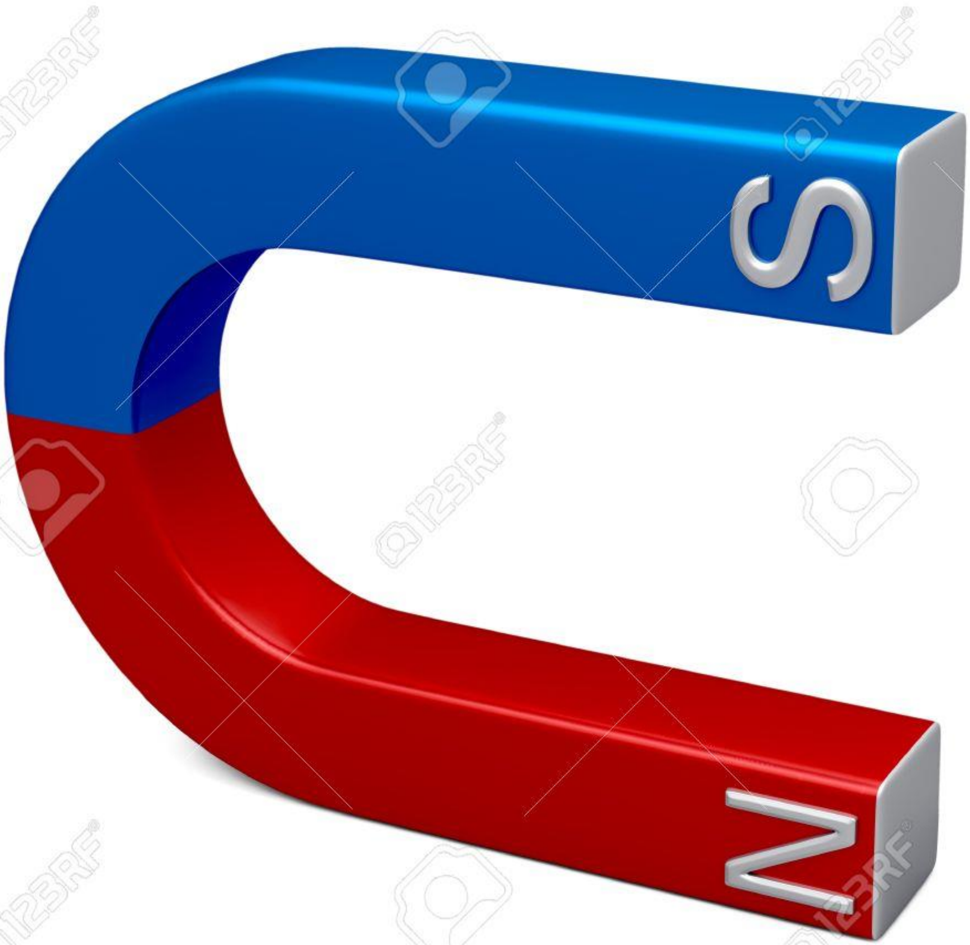
- Dr. hab. Jakub Cieślak, Krakow, PL
- Dr. Jan Żukrowski, Krakow, PL

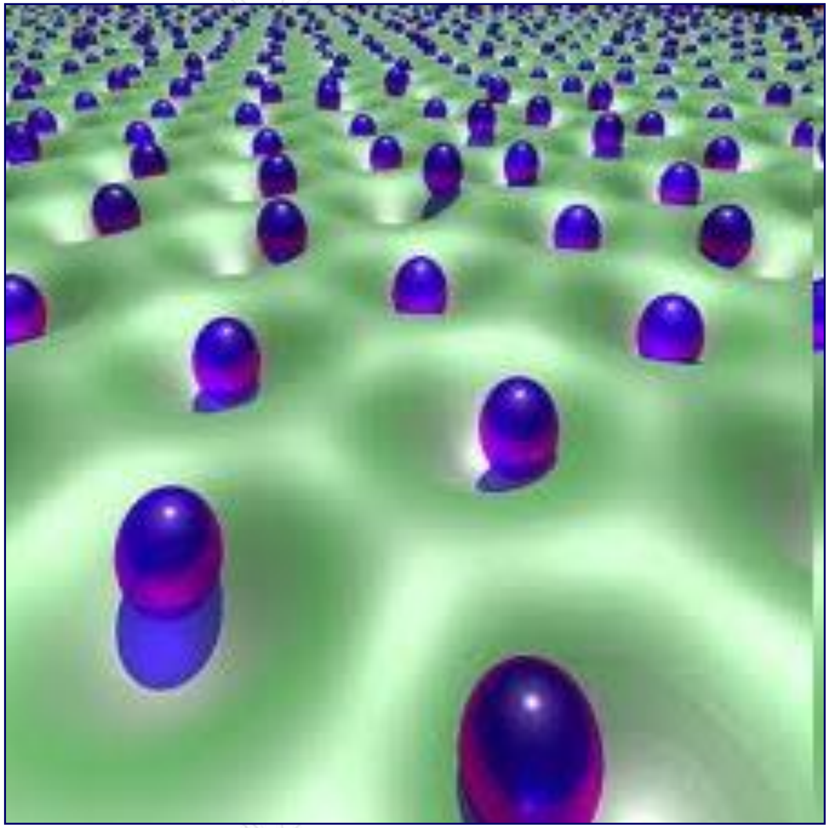
- Dr. Sasha Chumakov, Grenoble, F

- Prof. Maria Bałanda, Krakow, PL

- Prof. Michael Reissner, Vienna, A

THANK YOU!





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