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Magnetic interactions and interface phenomena on nanogranular thin films

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NanoSpain
Conf  2012

FEBRUARY 27 - MARCH 01, 2012
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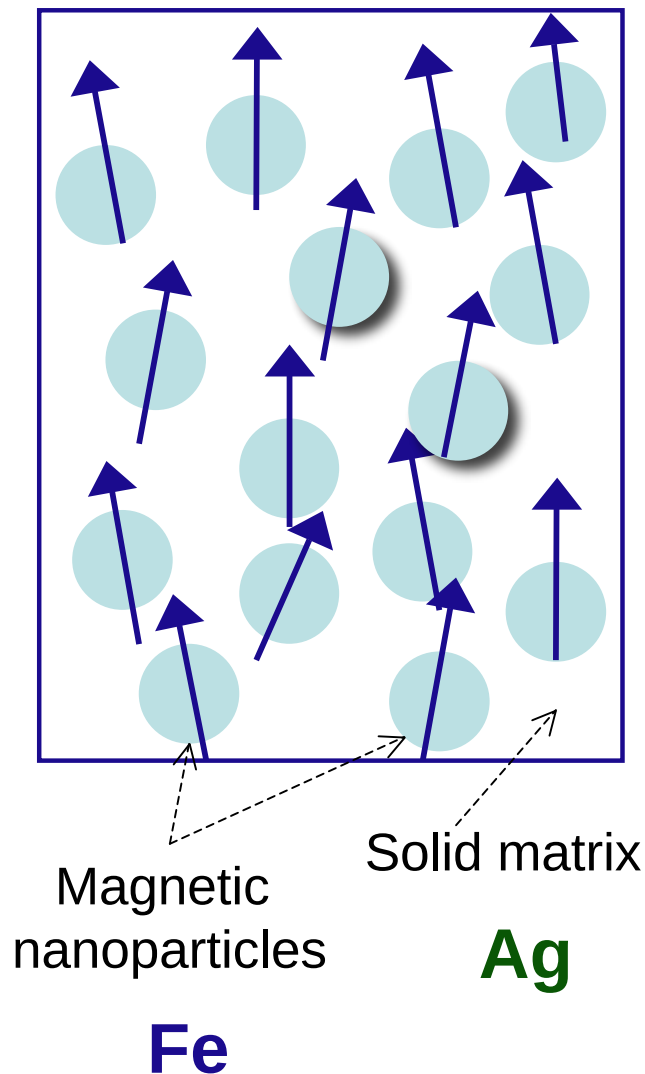


T. Neisius

Outline

- Introduction
- Motivation and aims
- Sample preparation
- Structural characterization
- Magnetic behaviour
- Conclusions

Nanogranular thin films



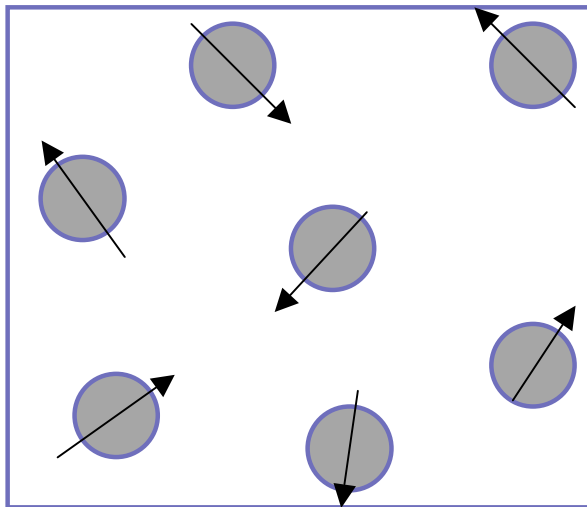
- Different preparation techniques (PLD, sputtering ...)
- **Magnetic interactions** are very important
 - Rich variety of magnetic behaviours
 - Crucial role in practical applications

Low concentrations

- **Non-interacting** monodomain uniaxial magnetic nanoparticles:

$k_B T < KV$ Magnetic moments are blocked along their easy axes

$k_B T > KV$ Magnetic moments spontaneously reverse



(i) superparamagnetism

(ii) Interacting superparamagnetism

- Superparamagnetism:

$$M = n\mu L(a) = n\mu \left[\coth(a) - 1/a \right]$$

$$a = \frac{\mu_0 \mu H}{k_B T}$$

Volume density of nanoparticles

Increasing concentrations

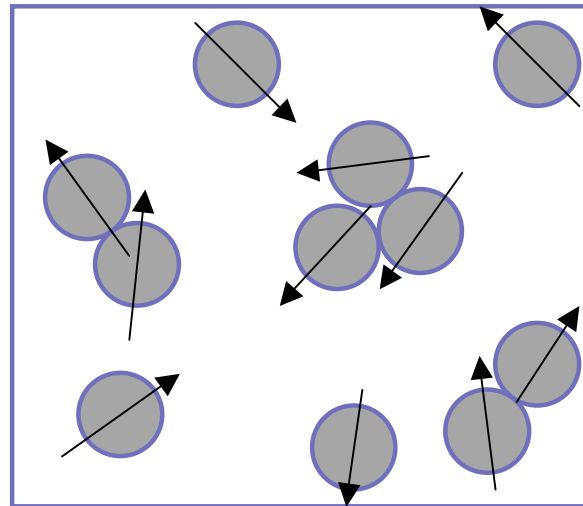
- Long range interactions of intermediate strength (dipolar, RKKY)

➡ **collective magnetic freezing.**

- **Spin glass** like behaviour: **Superspin Glass**

- ☐ **Randomness**

- ☐ **Frustration**



(iii) Spin-glass like
(iv) Superspin glass

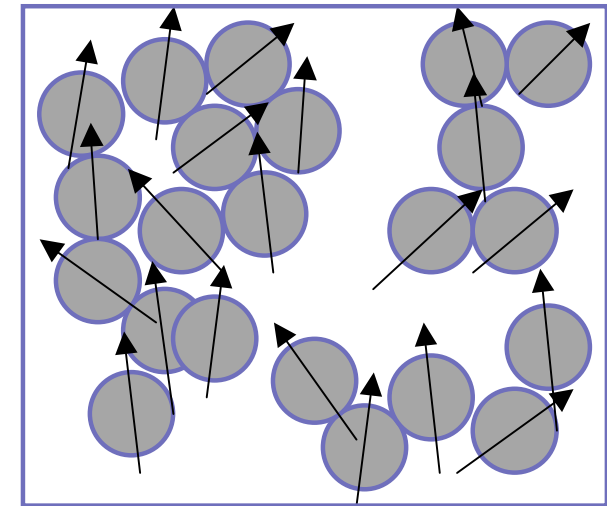
- J.L. Dormann et al, Adv. Chem. Phys. 98 (1997) 283
- S. Sahoo et al; Phys. Rev. B 63 (2002) 134406
- F. Jimenez-Villacorta et al; Phys. Rev. B 82 (2010) 134413

Volume density of nanoparticles

Higher concentrations

- Near or above the percolation.
- Long range ferromagnetic correlations.
- Direct exchange interactions, superexchange.
- The term **superferromagnetism** was introduced by Mørup *et al.* In *J. Magn. Magn. Mater.* **40**, 163 (1983).

- Sankar et al; J. Magn. Magn. Mat. 1 (2000) 221
- W. Kleemann et al; Phys. Rev. B 63 (2001) 134423
- Bedanta et al; J. Phys. D: Appl. Phys. 42 (2009) 013001

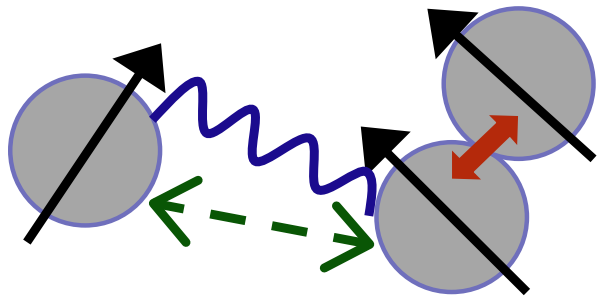


(v) Superferromagnetism

Volume density of nanoparticles

Aim of the work

1- Role of Magnetic Interactions



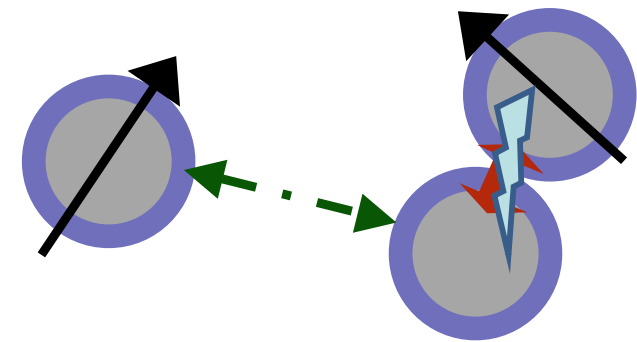
- RKKY interaction
- **Dipolar interaction**
- **Direct exchange interaction**

SPUTTERING TECHNIQUE: $\text{Fe}_x\text{Ag}_{100-x}$ ($15 \leq x \leq 55$)

2 . Interface phenomena

**Direct
exchange
interactions**

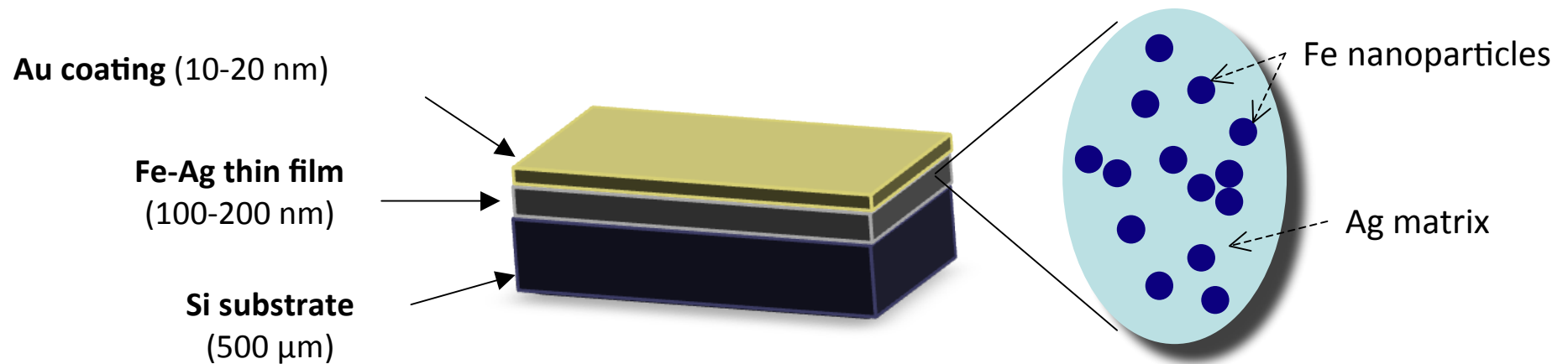
- Greatly depend on the nature of the interface



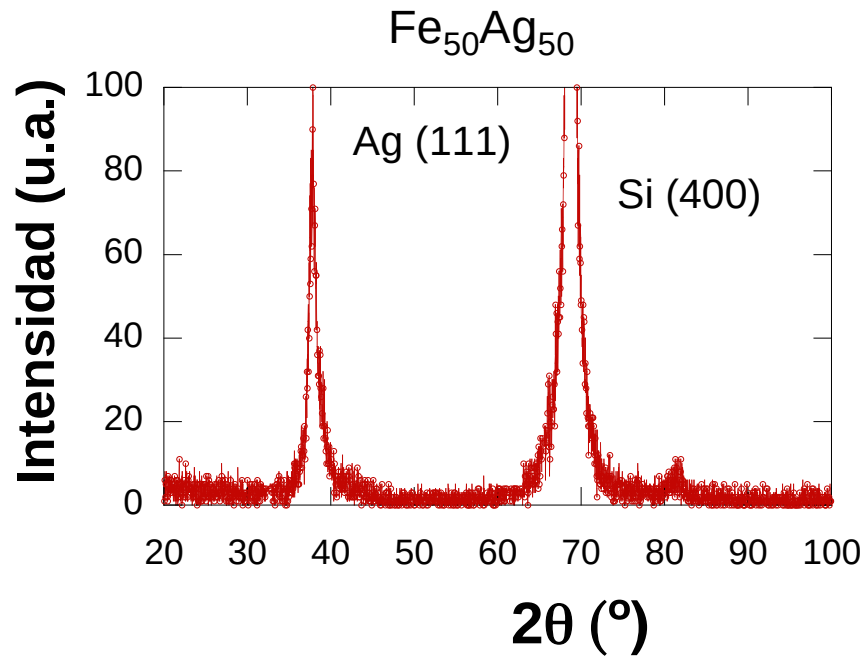
PULSED LASER DEPOSITION: $\text{Fe}_{50}\text{Ag}_{50}$

$\text{Fe}_x\text{Ag}_{100-x}$ granular thin films: $15 \leq x \leq 55$

- DC magnetron sputtering: increasing Fe content $\longrightarrow$
increases nanoparticle concentration



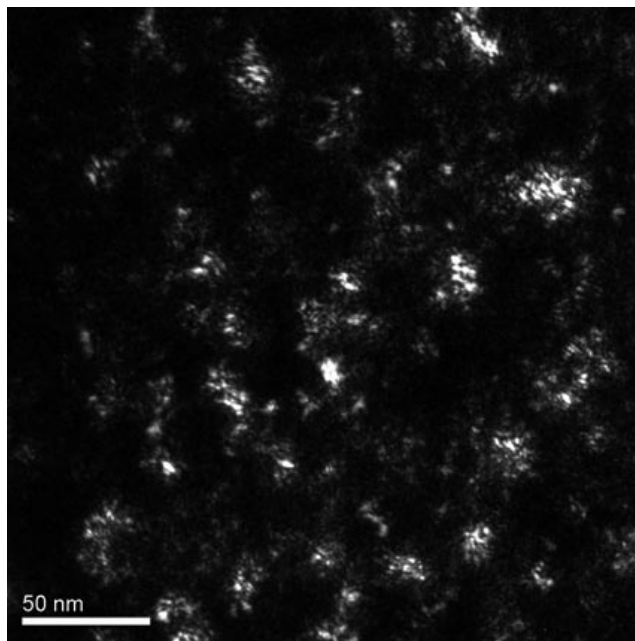
- C. Binns et al; Phys. Rev. B 66 (2002) 184413
- J. Q. Wang and G. Xiao; Phys. Rev. B 49 (1994) 3982
- P. Allia et al; Phys. Rev. B 73 (2006) 05 44 099



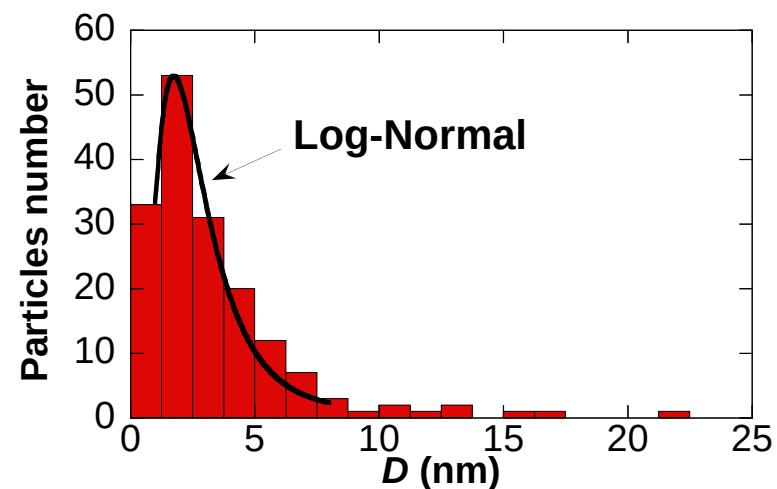
XRD

- Nanogranular Ag matrix ($\langle D \rangle = 8-12$ nm) textured in the fcc (111) direction.

Dark field-TEM

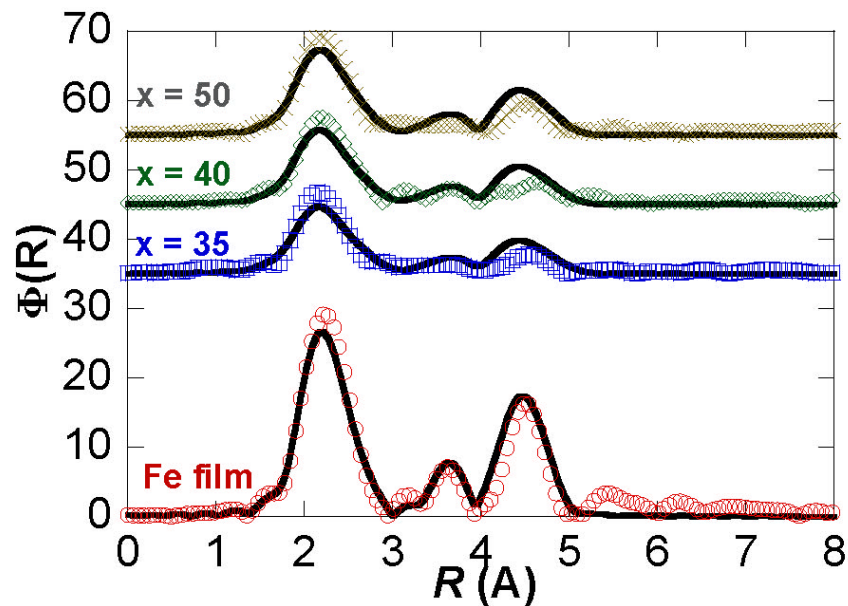
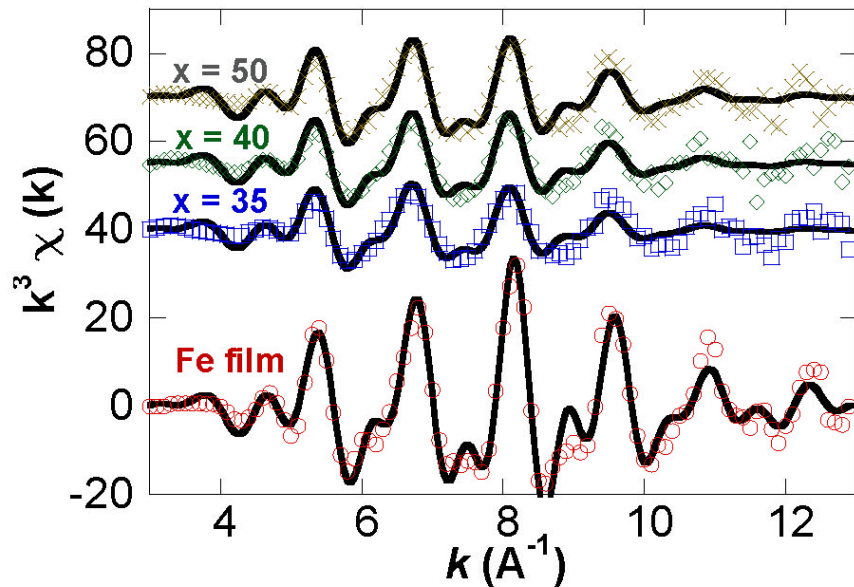


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- Fe nanoparticles present a **Log-Normal** size distribution with $\langle D \rangle = 2.5$ nm ($x = 20$)

Fe K-edge



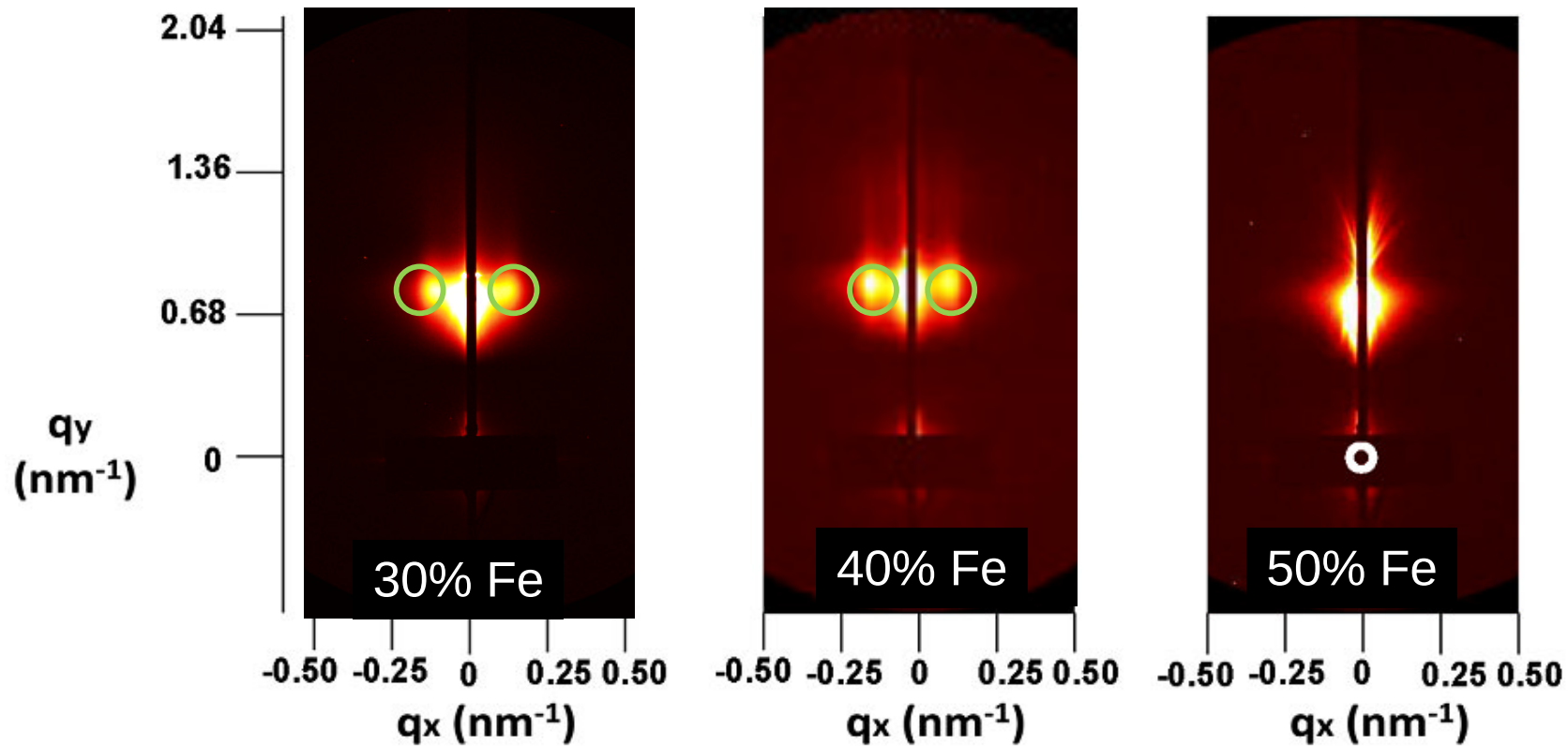
EXAFS SPECTROSCOPY

Fe bcc

High disorder due to surface effect

BM29 ESRF

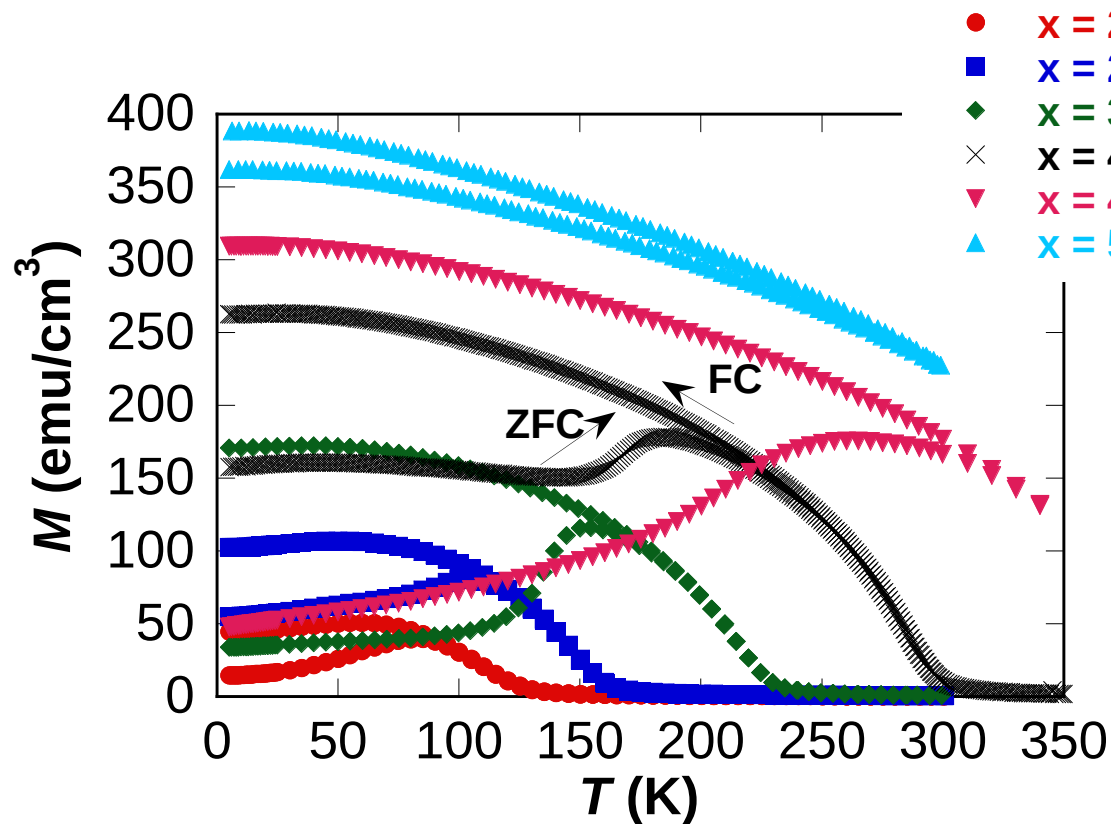
GISAXS



BM16 ESRF

Granular state up to 50%

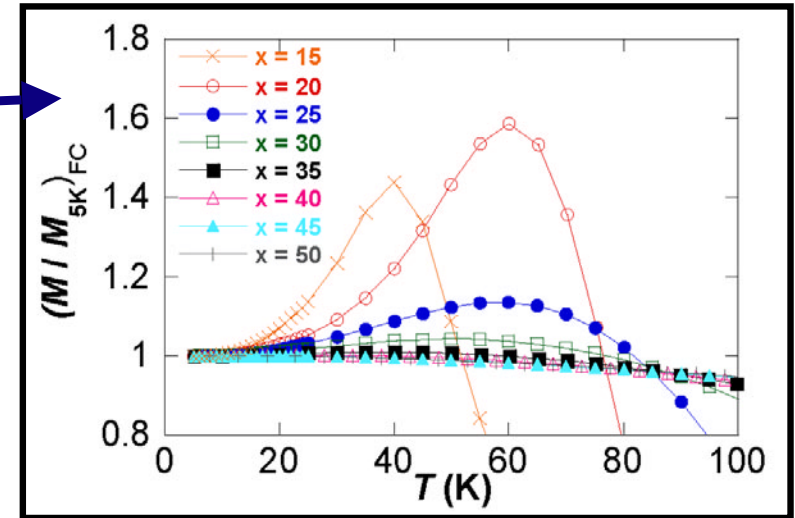
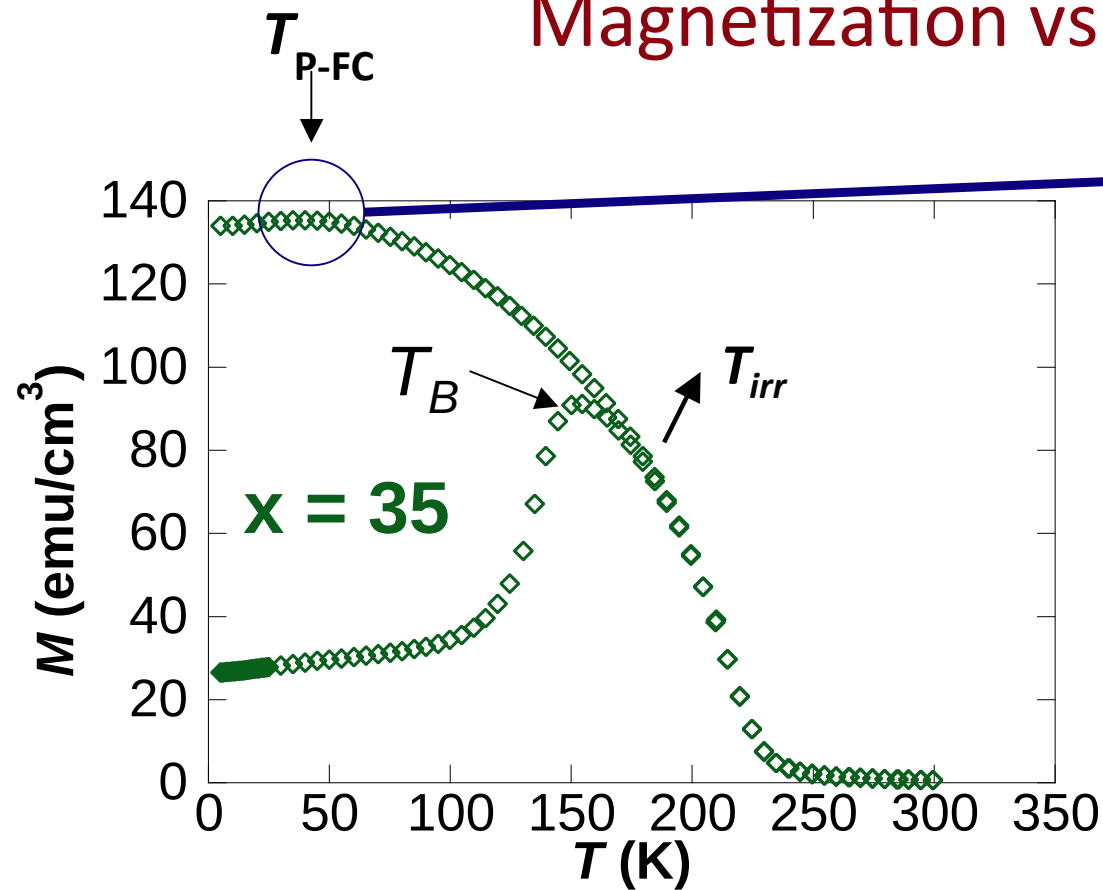
Magnetization vs temperature



$H = 50$ Oe

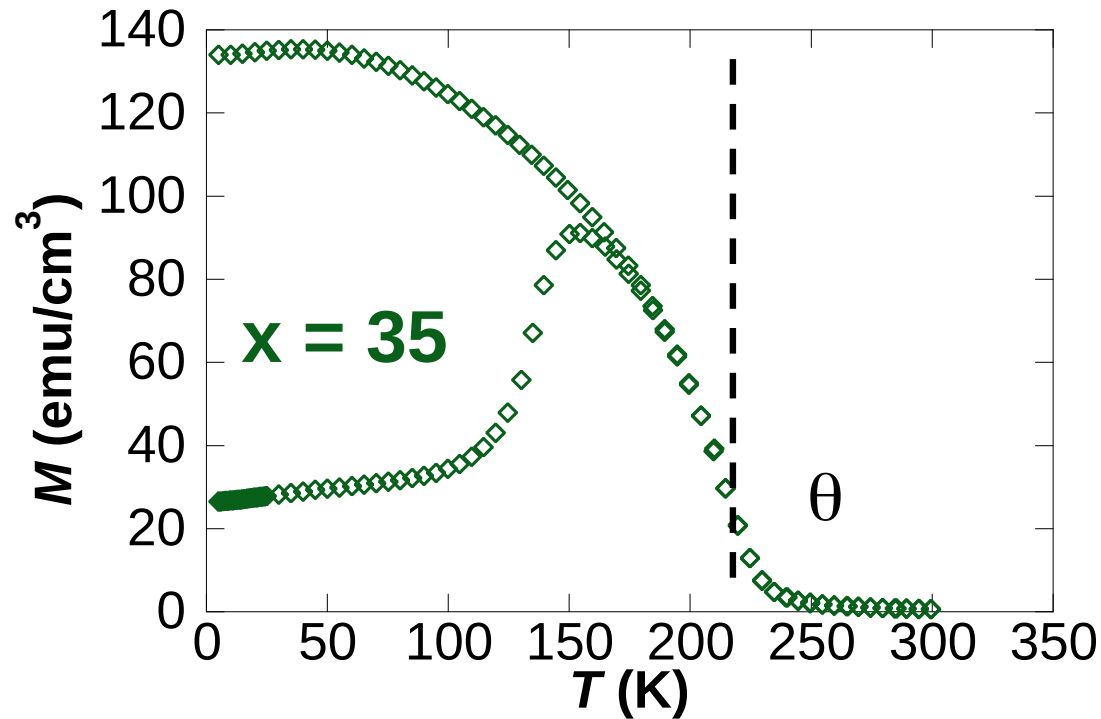
- Maximum in the ZFC curves **blocking/freezing** of the magnetic moments.
- Maximum of ZFC **displace to higher T** with increasing Fe content
- Above the maximum **Curie-Weiss transition**

Magnetization vs temperature

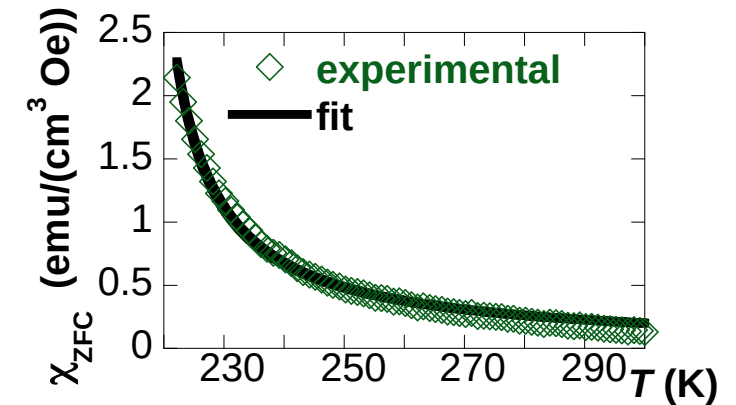


■ Maximum in the FC curves → **spin glass like** behaviour
strong dipolar interaction.

Magnetization vs temperature



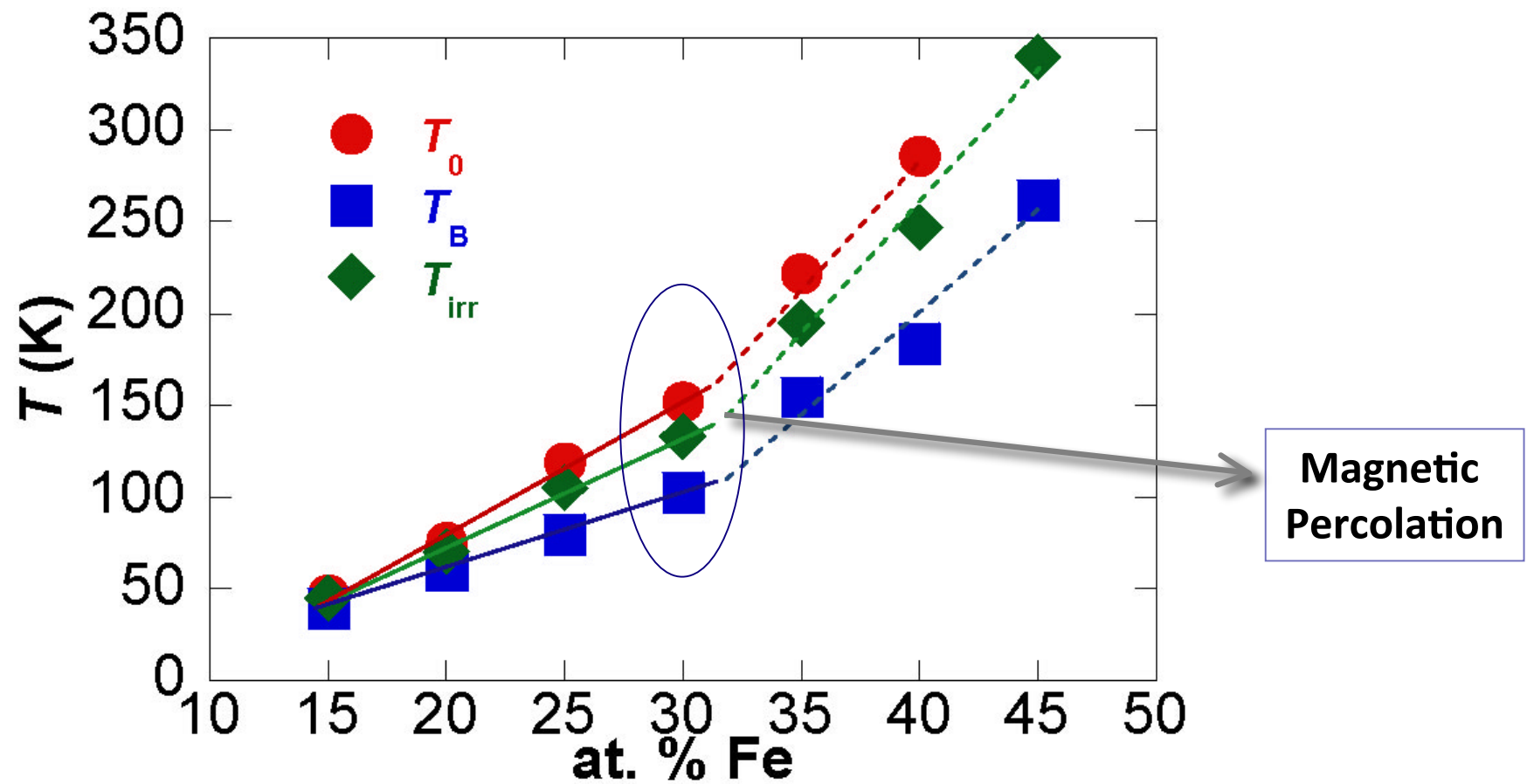
$$\chi = \frac{C}{T - \theta}$$



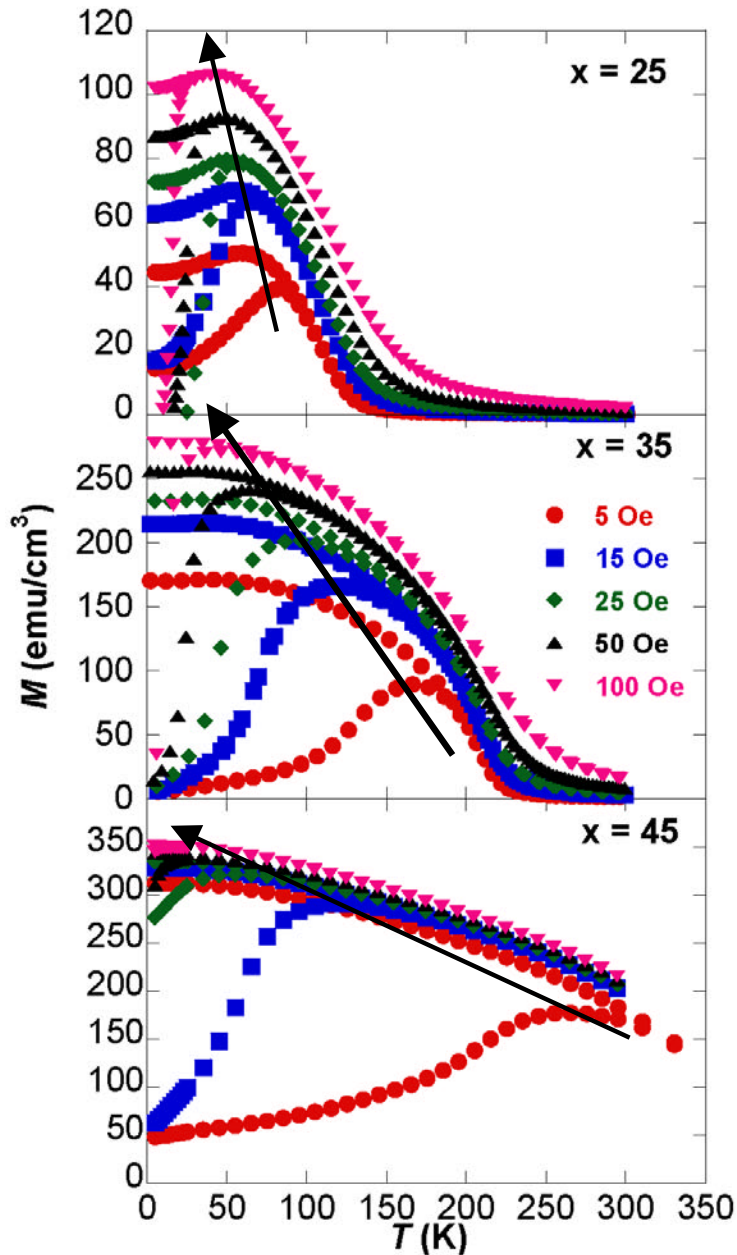
$$C = \frac{x' V M_s^2 \mu_0}{3 k_B}$$

	x=15	x=20	x=25	x=30	x = 35	x=40
θ (K)	47(2)	75(2)	119.4(2)	152(2)	222(2)	287(1)
$\langle D \rangle$ (nm)	2.7(5)	2.7(5)	2.74(5)	2.95(5)	2.56(5)	2.52(5)

Characteristic temperatures



Evolution of the ZFC-FC curves with H



■ With increasing H :

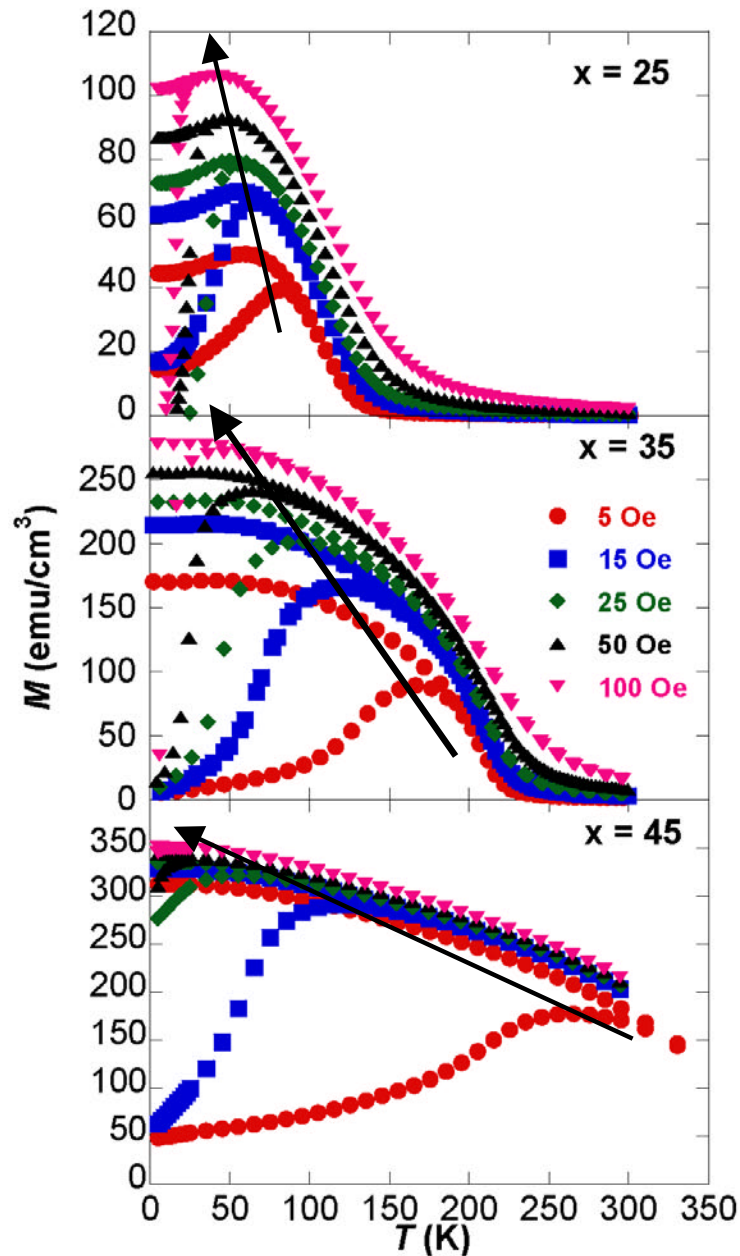
ZFC-FC curves tend to overlap.

Faster response for $x \geq 35$



longer range **ferromagnetic order**

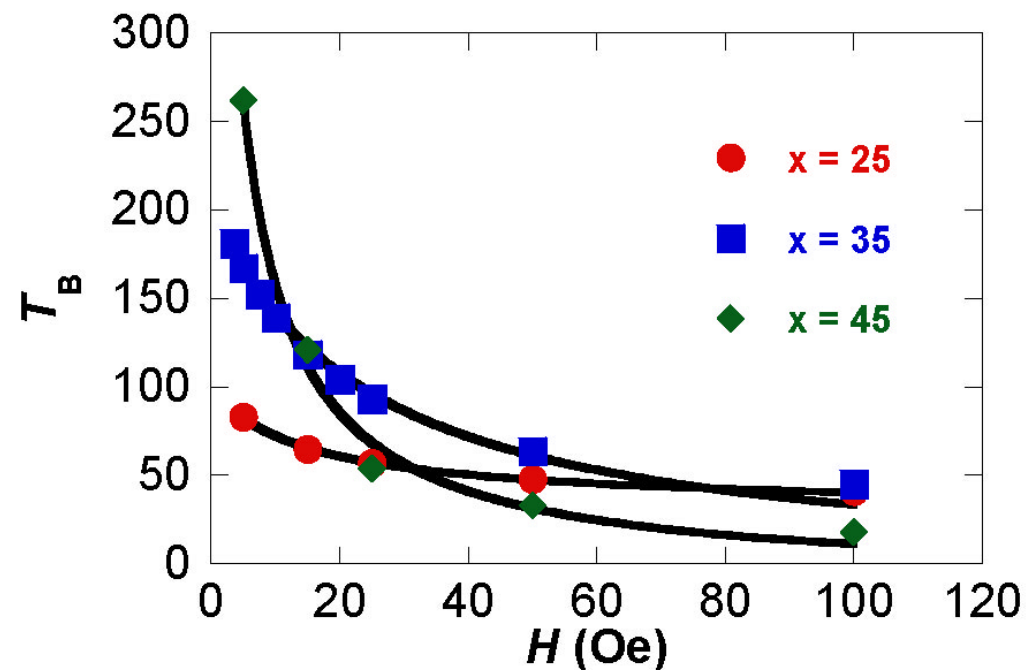
Evolution of the ZFC-FC curves with H



■ Modified random anisotropy model:

$$T_{P-ZFC}(H) = \frac{K\pi [D^3 + \varepsilon(L_H^3 - D^3)]}{6k_B \ln(\tau_m / \tau_0) [1 + x(L_H^3 - D^3) / D^3]^{1/2}} \times$$

$$\left[1 - \frac{HM_S [1 + \varepsilon(L_H^3 - D^3) / D^3]^{1/2}}{2K} \right]^{1.5}$$



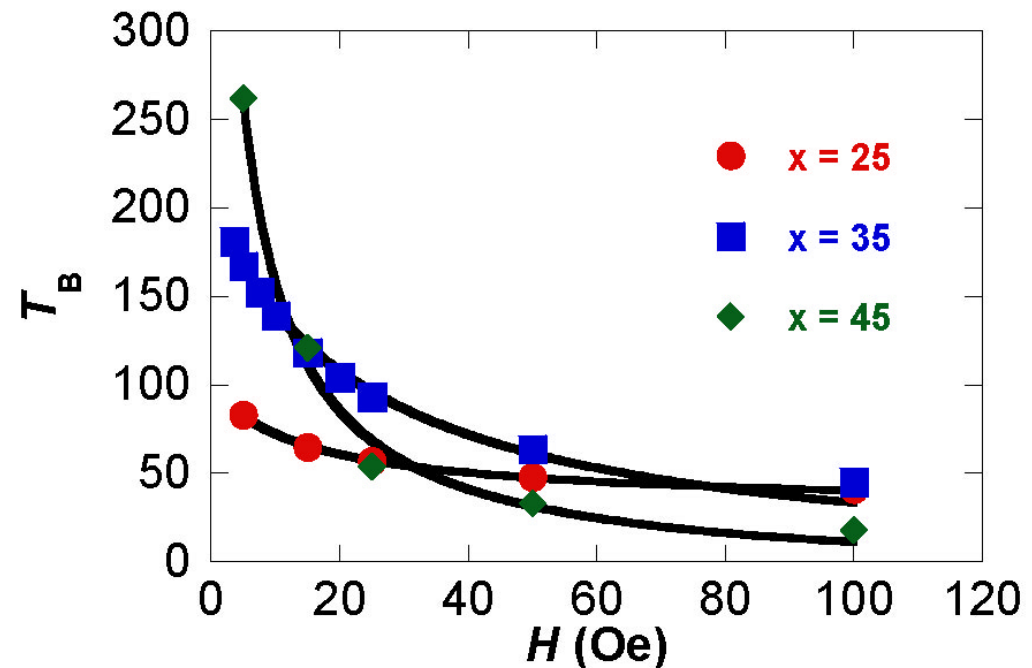
Evolution of the ZFC-FC curves with H

■ Modified random anisotropy model:

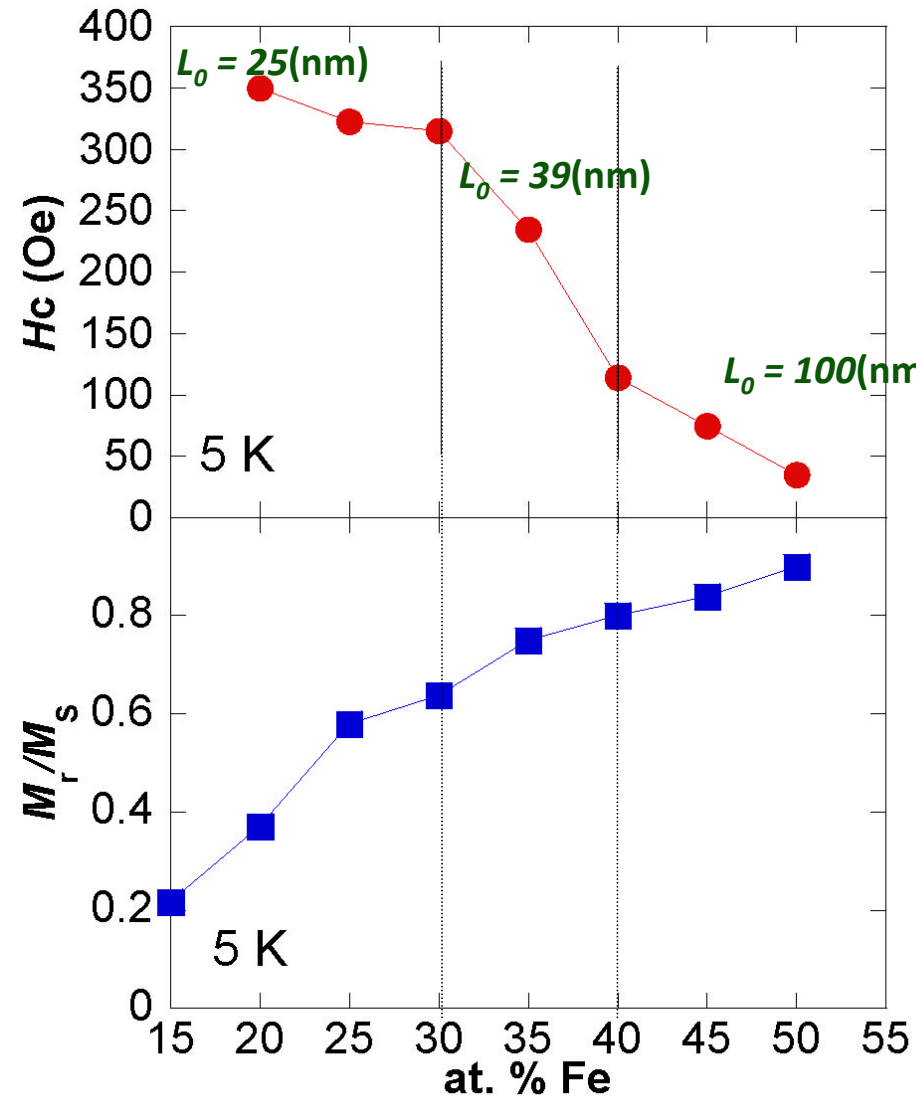
$$T_{P-ZFC}(H) = \frac{K\pi [D^3 + \varepsilon(L_H^3 - D^3)]}{6k_B \ln(\tau_m / \tau_0) [1 + \chi(L_H^3 - D^3) / D^3]^{1/2}} \times$$

$$\left[1 - \frac{HM_S [1 + \varepsilon(L_H^3 - D^3) / D^3]^{1/2}}{2K} \right]^{1.5}$$

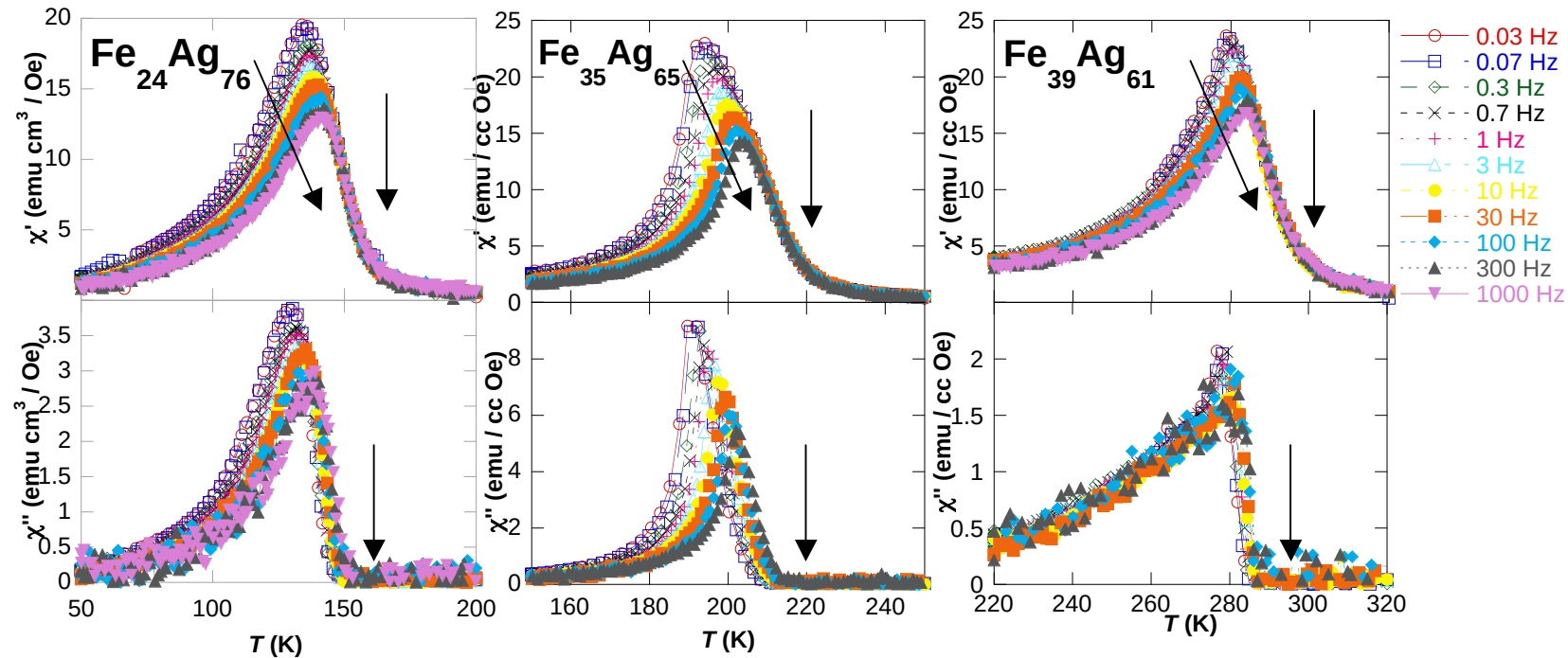
	K (J/m ³)	L_o (nm)
$x = 20$	$3.0(8) 10^5$	24(3)
$x = 35$	$2.6(8) 10^5$	39(3)
$x = 45$	$1.8 (1)10^5$	100(17)



Magnetization vs magnetic field

 $T = 5\text{K}$ 

Spin dynamic-AC susceptibility



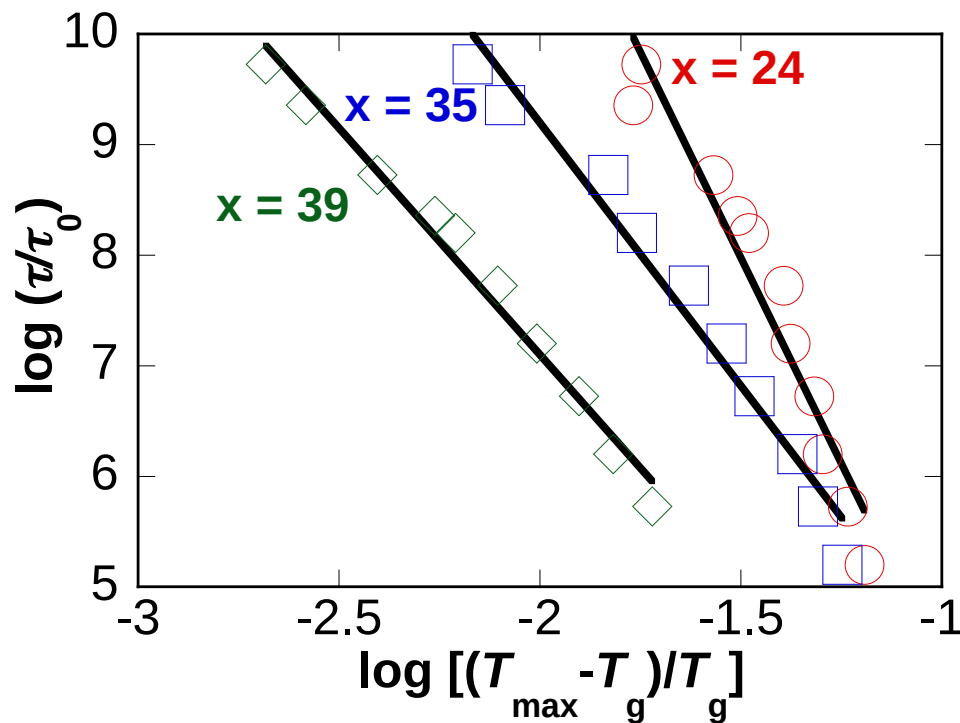
- Maximum in χ' → freezing of the magnetic moment
- Maximum displace to higher T with increasing ν
- Above the maximum, χ' decays in a dispersionless Curie-Weiss way, and χ'' becomes null → magnetic transition mediated by interactions.

AC susceptibility

- Evolution of the maximum $T_{\max}$ in χ' with frequency:

Dynamical critical exponent activation law:

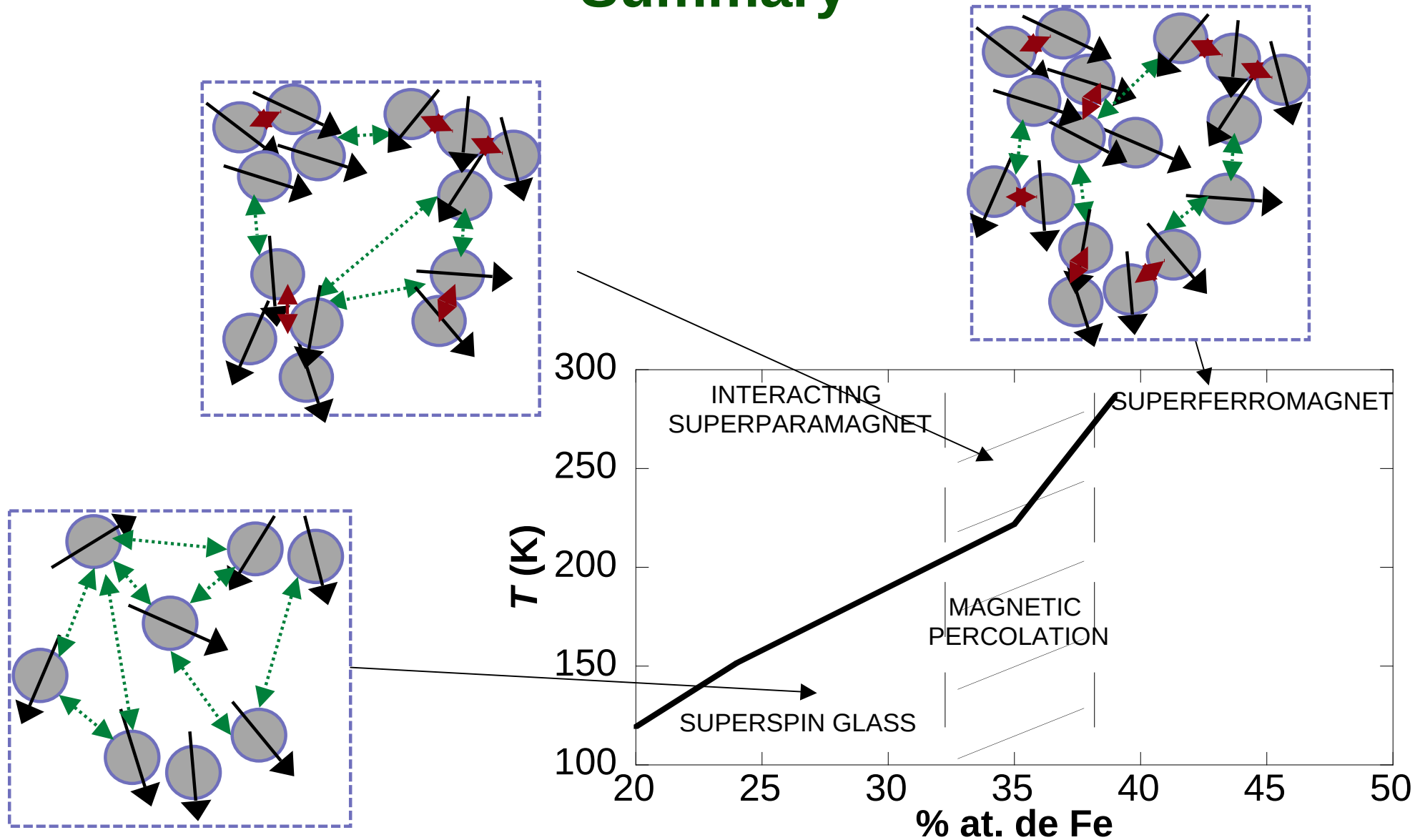
$$\tau = \tau_0 \left(\frac{T_{\max} - T_g}{T_g} \right)^{-zv}$$



	<i>Critical exponent</i>		} SSG ↓ SFM
$x = 24$	$zv = 8.3(6)$	$T_g = 132.5(3) \text{ K}$	
$x = 35$	$zv = 4.7(4)$	$T_g = 192.7(1) \text{ K}$	
$x = 39$	$zv = 4.3(3)$	$T_g = 278.9(3) \text{ K}$	

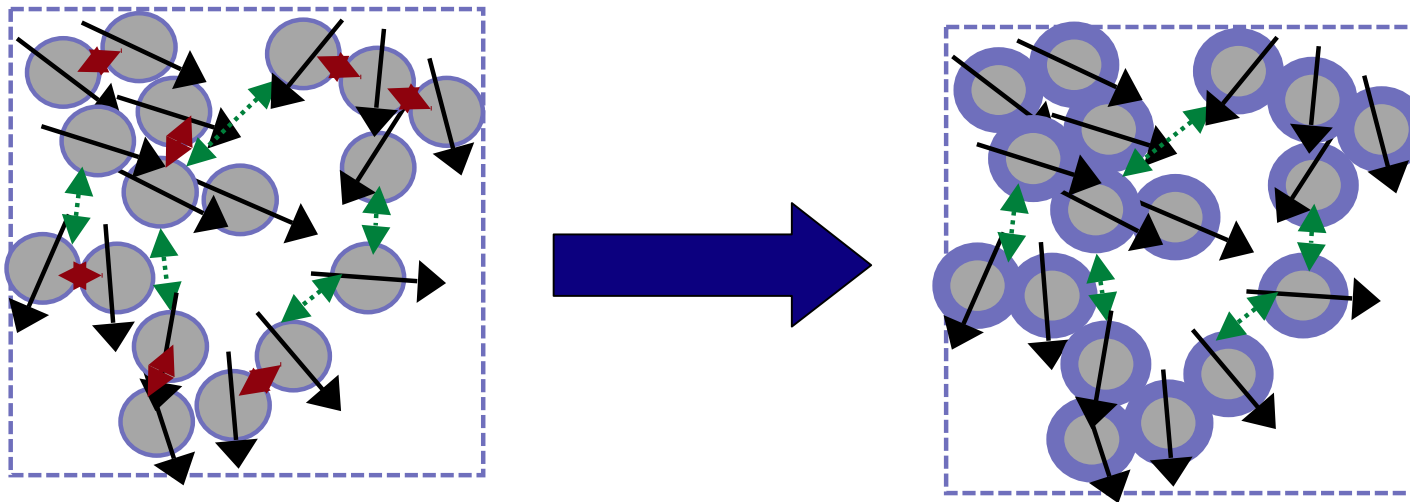
* D. Ucko et al; Phys. Rev. B 64 (2001) 104433

Summary



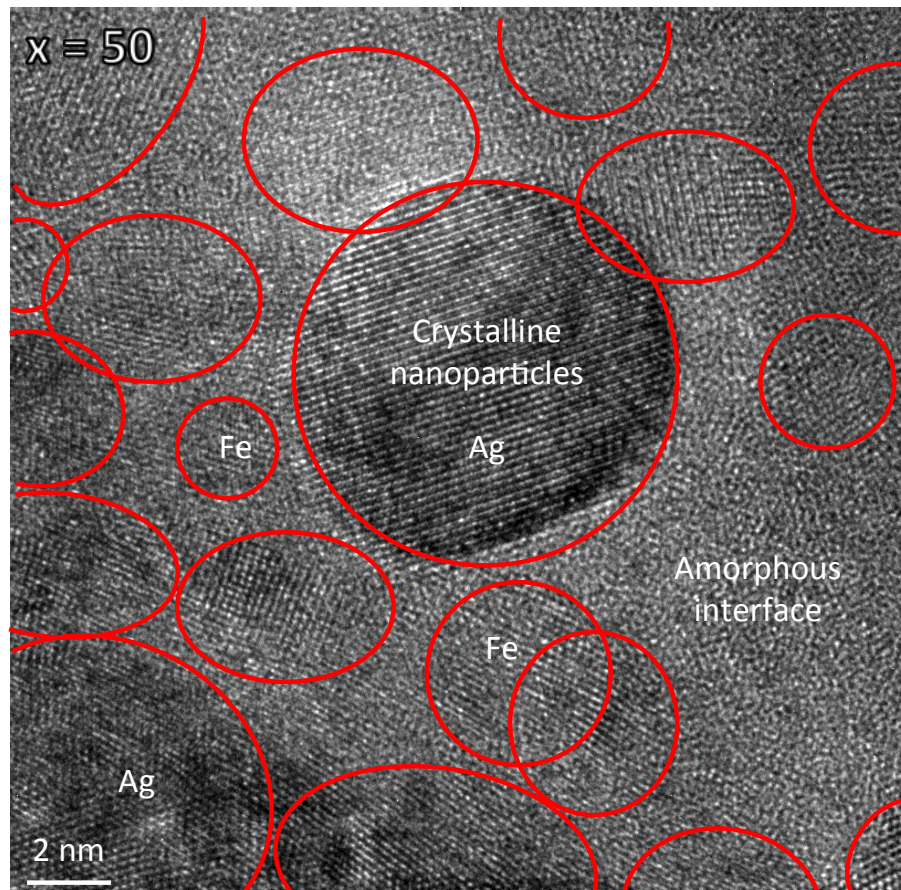
- J. Alonso, M.L. Fdez-Gubieda et al; Phys. Rev. B. 82 (2010) 054406
- M.L. Fdez-Gubieda, et al “Nanoparticles features Electromagnetic properties: From science to engineering” 2012 Research Signpost. Editors A. Chilerio and P. Allia.

Interface Phenomena



Pulsed Laser Deposition (PLD): $\text{Fe}_{50}\text{Ag}_{50}$

- **Pulsed Laser Deposition (PLD):** High energies reached during deposition allow some Fe-Ag miscibility → **modify the Fe/Ag interface**



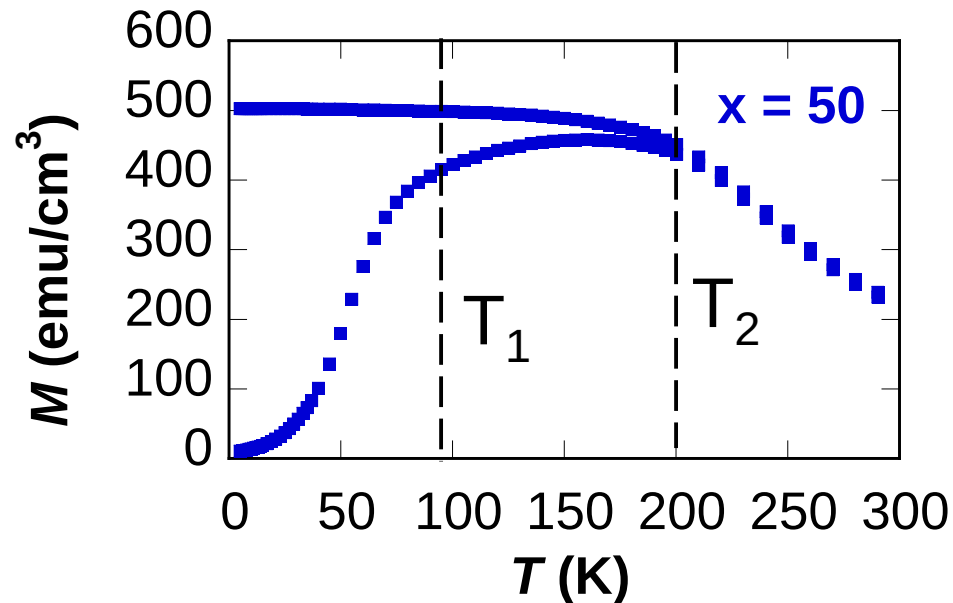
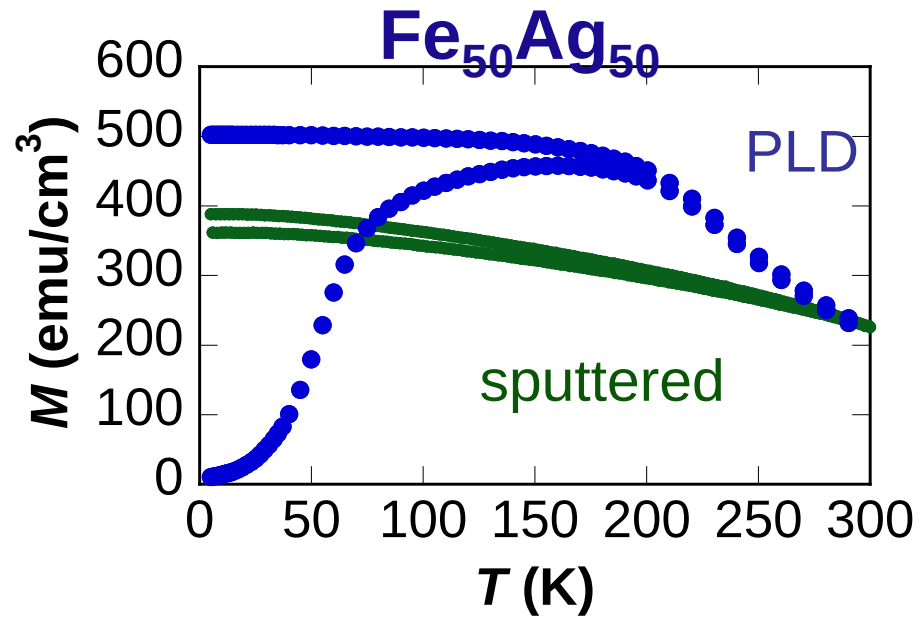
- **Crystalline nanoparticles:**

- Small bcc Fe nanoparticles (2-4 nm).
- Bigger fcc Ag nanoparticles (10-12 nm).

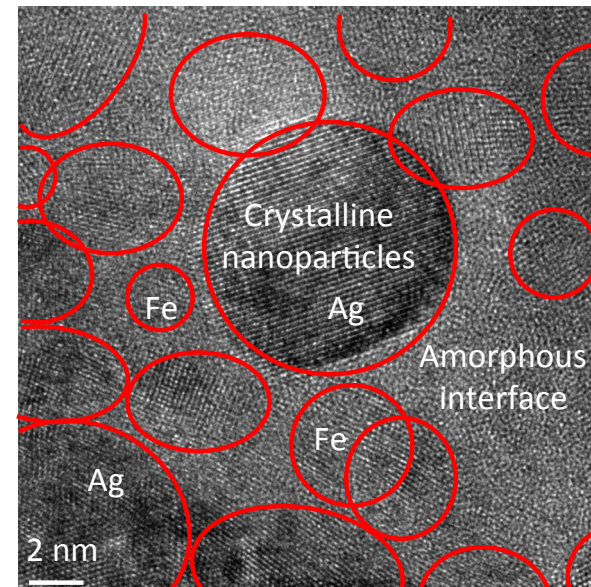
- **Amorphous interface:**

- Occupies around a 20 %.
- Variable thickness of a few nm.

Magnetization vs temperature



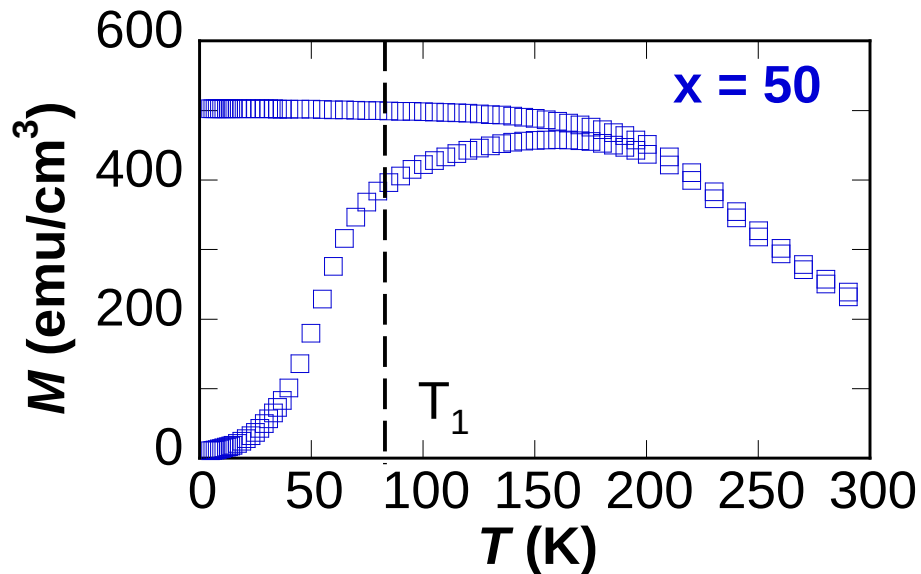
■ The interface modify the magnetic behaviour



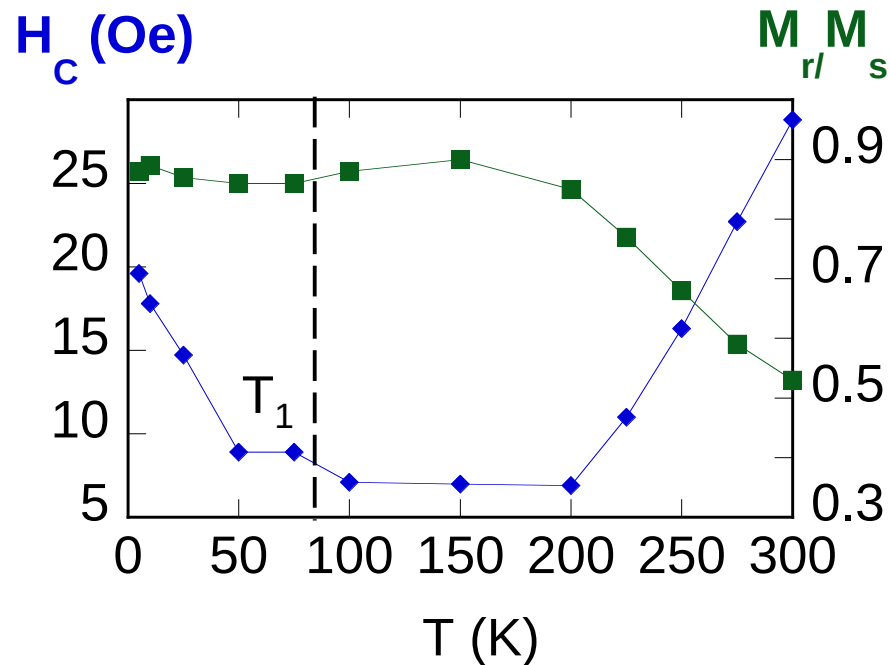
■ Two anomalous magnetic transitions

Magnetic behaviour

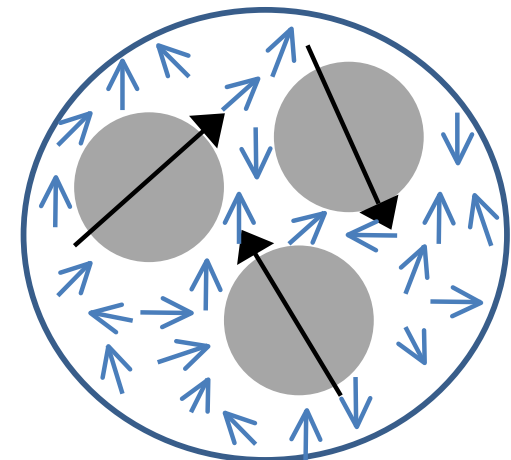
$$T < T_1$$



- High **coercivity** → low correlation length.

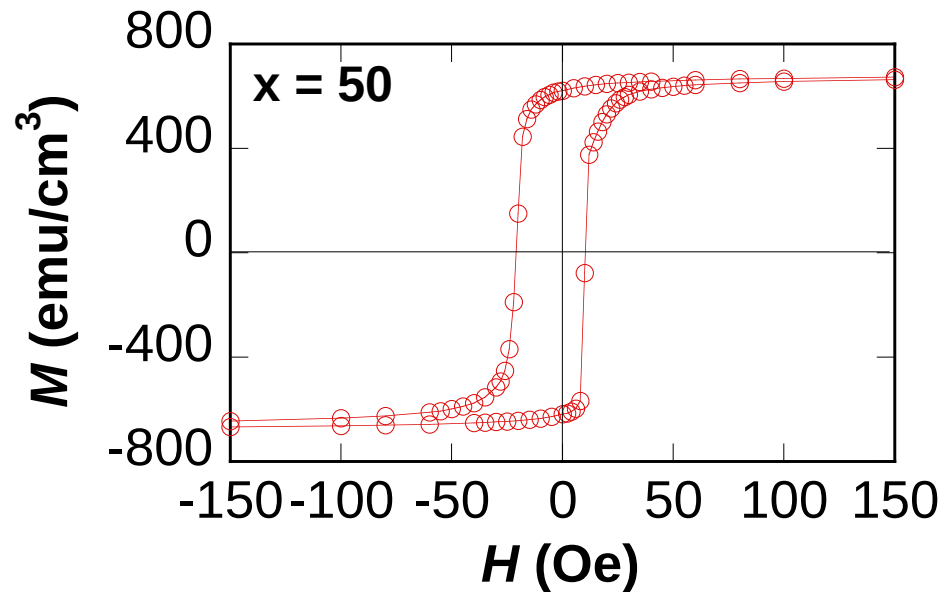


- **Magnetically disordered interface** disables coupling between Fe nanoparticles.



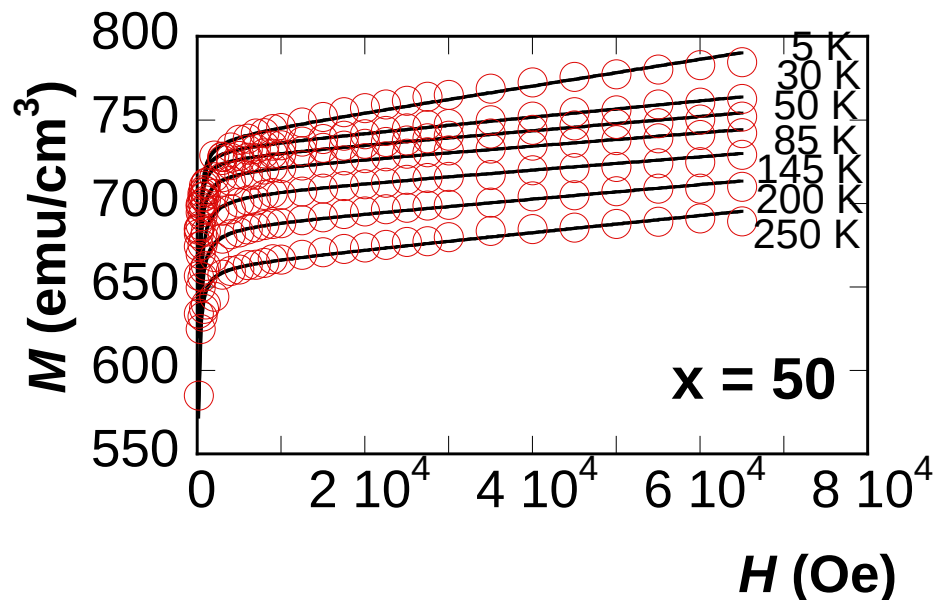
Magnetic behaviour

$T < T_1$

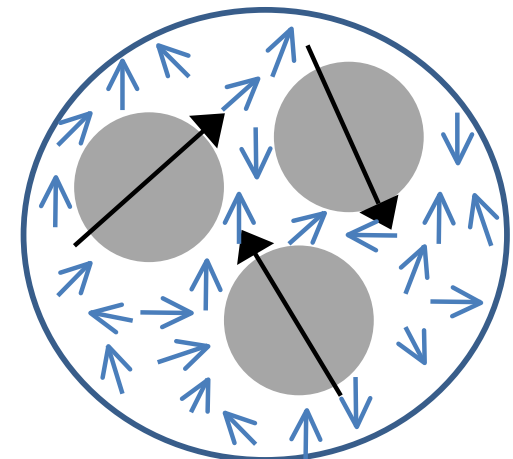


$T = 5\text{K}$

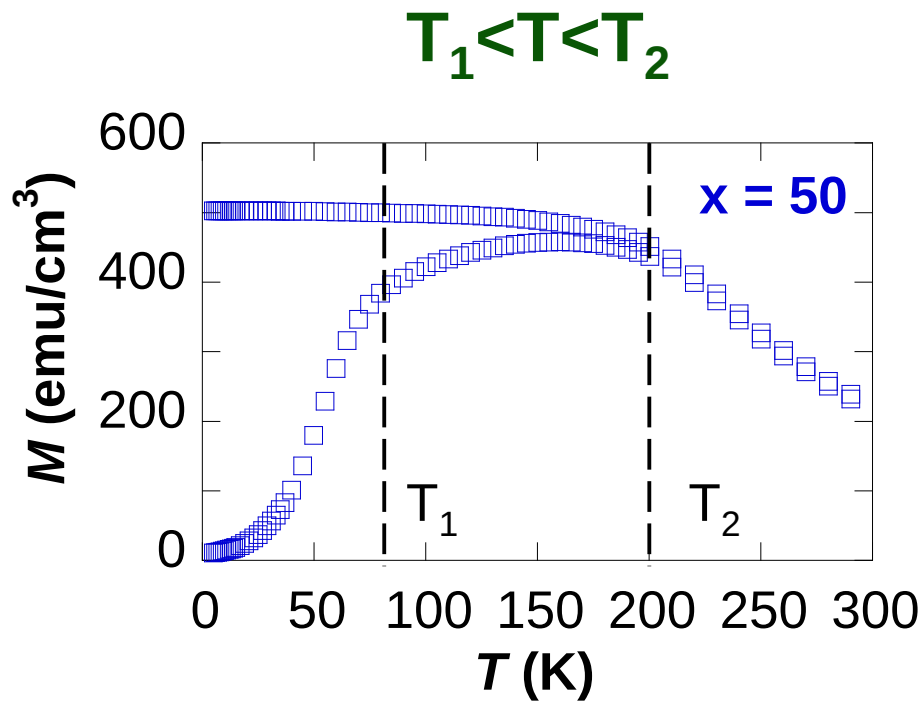
- **Exchange bias** after cooling at remanence: $H_{\text{ex}} = -4.3\text{ Oe}$
- **Unsaturated** magnetic behaviour.



Spin glass amorphous interface



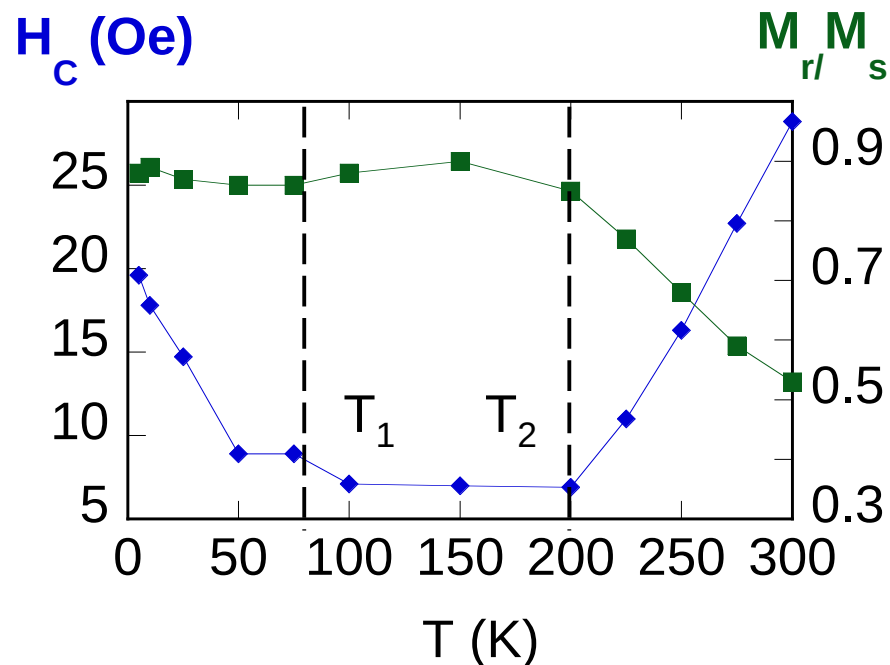
Magnetic behaviour



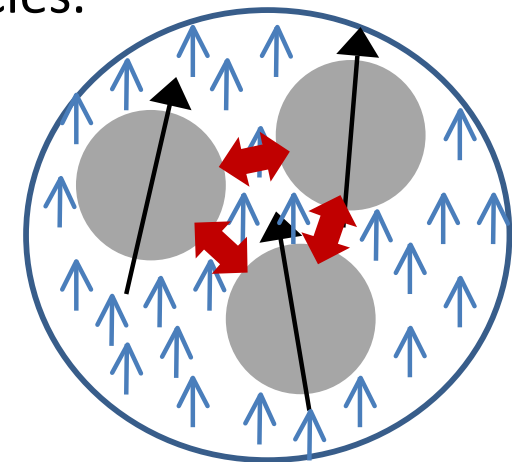
Ferromagnetic behaviour:

Smooth variation of the $M(T)$

- **Low coercitivity** and **high remanence**
 $M_r/M_s = 0.9 \rightarrow$ high correlation length.

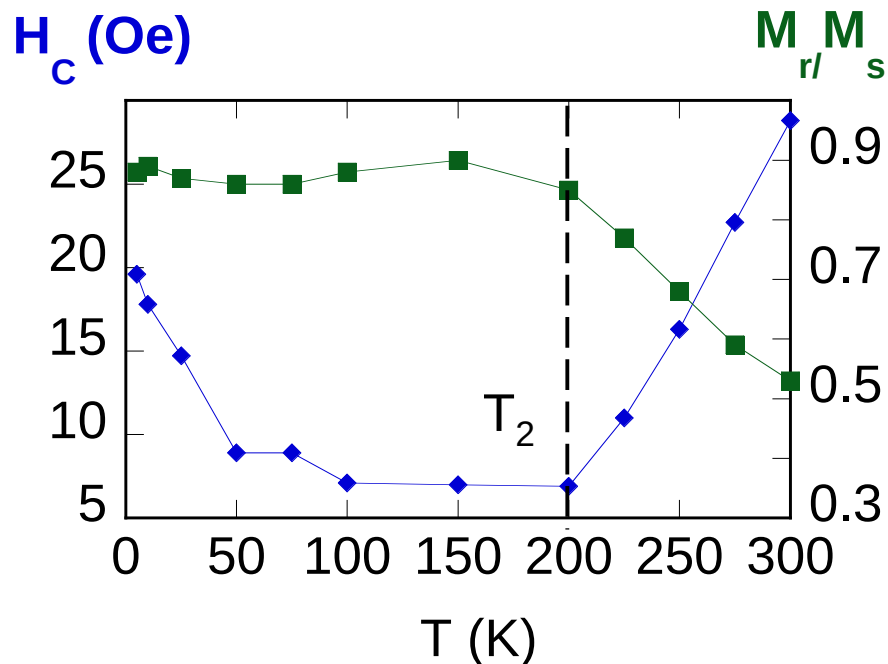
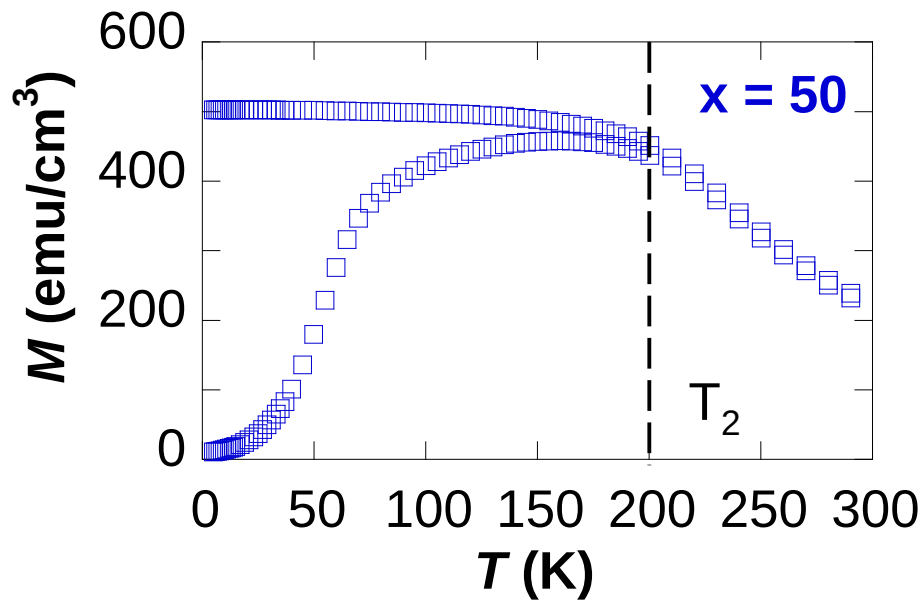


- **Ferromagnetic interface** enables coupling between Fe nanoparticles.



Magnetic behaviour

$T > T_2$



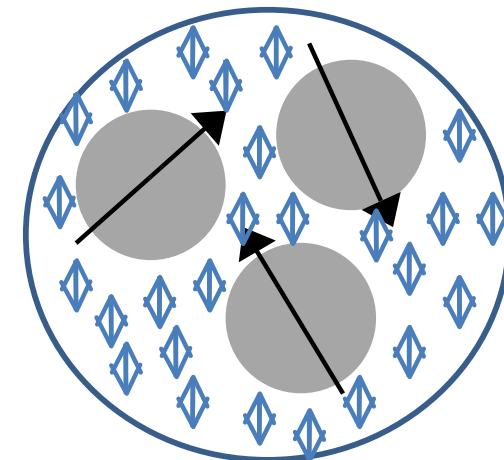
- **Coercivity** increases and **remanence** decreases → low correlation length.



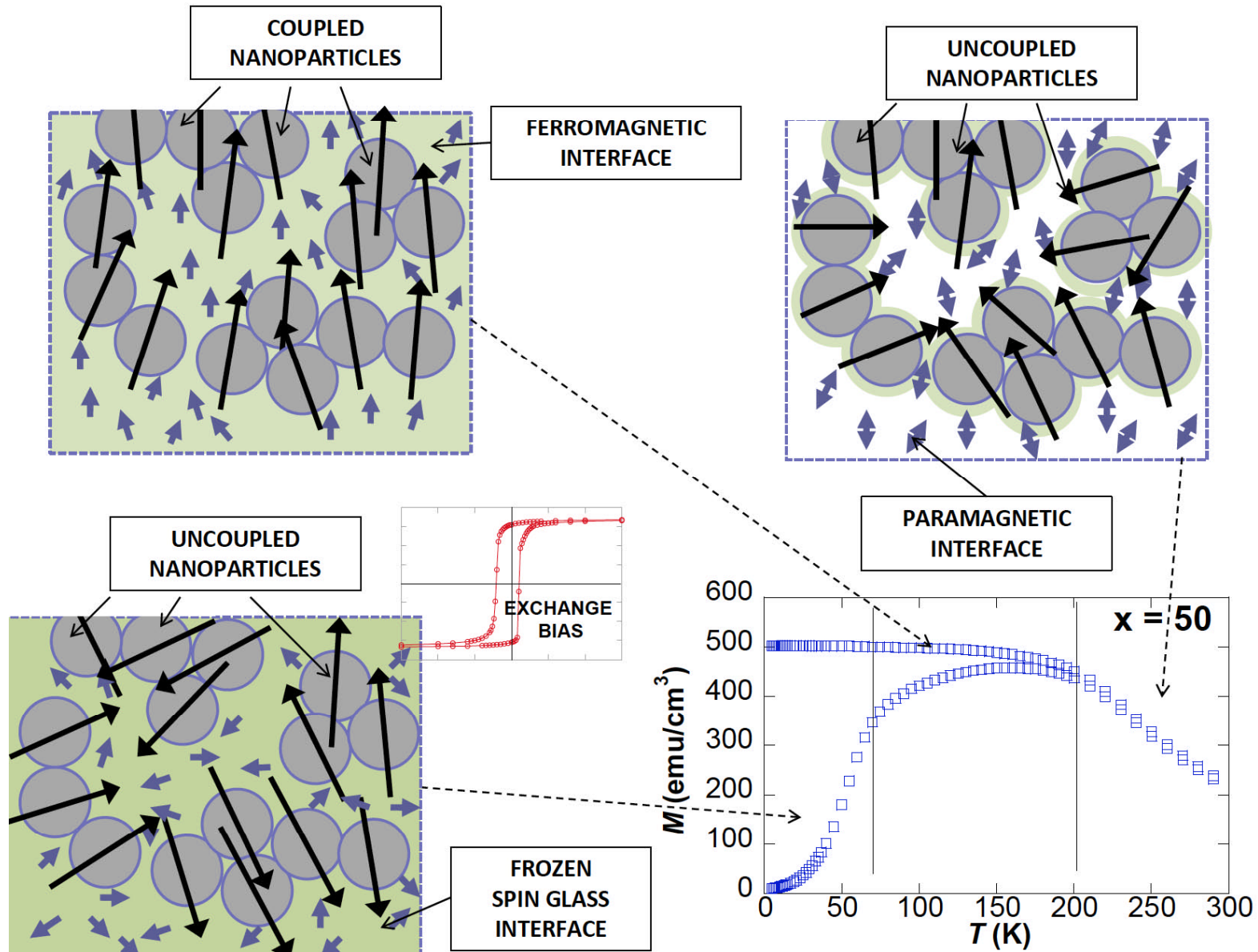
- **Paramagnetic interface** disables coupling between Fe nanoparticles.



- **Ms** decreases 15%.



Interface Phenomena



- $\text{Fe}_x\text{Ag}_{100-x}$ granular thin film composed of magnetic nanoparticles (2-3nm) inside a Ag matrix have been prepared by sputtering technique.
- Crossover from a SSG to a SFM state around 35 at. %
- More information:
 1. J. Alonso, M.L. Fdez-Gubieda et al; Phys. Rev. B. 82 (2010) 054406
 2. M.L. Fdez-Gubieda, et al “Nanoparticles features Electromagnetic properties: From science to engineering” 2012 Research Signpost. Editors A. Chilerio and P. Allia.
- Pulsed Laser Deposition Technique allow us to modify the FeAg interface
- The magnetic ordering/disordering of the interface acts as a switch in the coupling/decoupling of the magnetic nanoparticles
- More information:
 1. J. Alonso, M.L. Fdez-Gubieda et al., Nanotechnology 23 (2012) 025705



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