European School on Magnetism 2009

Inhomogeneities in magnetic systems

Alberta Bonanni

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Time-table

04.09.2009	**J. Mike D. Coey** Magnetism of dilute oxides
09.09.2009	**A. Bonanni** Inhomogeneous magnetic systems: introduction and examples
	Wiktor Stefanowicz Measurements issue
	Bogdan Faina Materials issue
	Coffee break
	Samaresh Guchhait Amorphous GeC:Mn
	G. Irina Groza AntiferromagneticCoO
	Discussion
	A. Bonanni Inhomogeneous magnetic systems: control and applications
	Discussion

Before applying any model

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We must gain knowledge about the distribution of magnetic ions

Before applying any model

We must gain knowledge about the distribution of magnetic ions

\ at surfaces

\ at interfaces

\ in the bulk

Two examples

Tunneling magnetoresistance [TMR]:

magnetic ions distribution at the interface

Diluted Magnetic Semiconductors [DMS]:

magnetic ions distribution in the volume

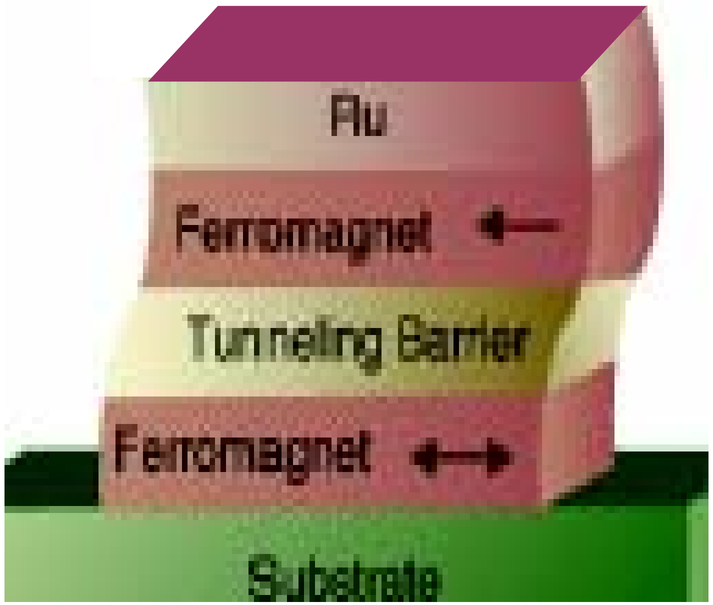
TMR

Julliere‘s model

Jullière‘s model:

\ constant tunneling matrix elements
\ electrons tunnel without spin-flip

$$\mathrm{TMR} = \frac{2P_1P_2}{1 - P_1P_2}$$



TM films → TMR ~ 2\3 ~ 67%

cfr. B. Dieny

Julliere's model – Fe/MgO/Fe

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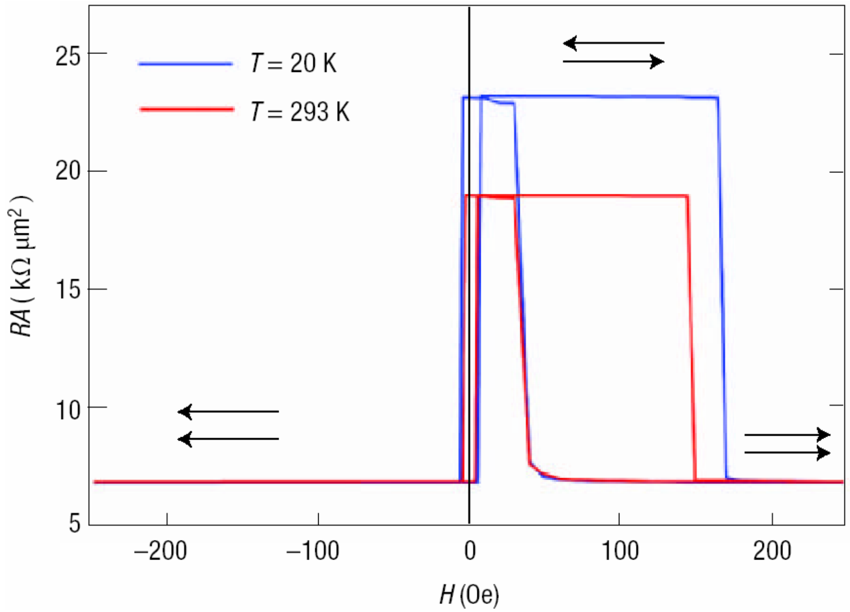

TMR > 350%

S. Yuasa et al., Nature Materials 3, 868 [2004]

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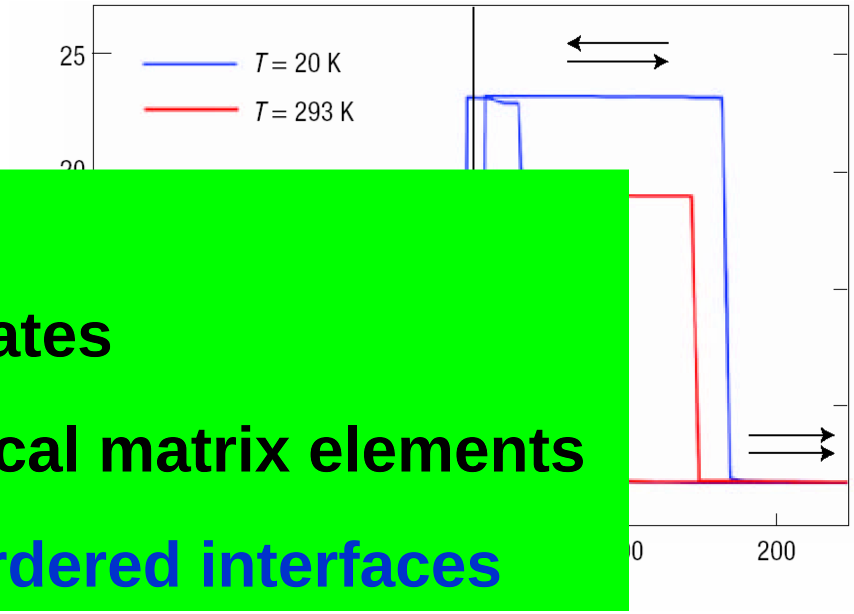


- ***k* || not conserved**
- **average density of states**
- **no quantum mechanical matrix elements**

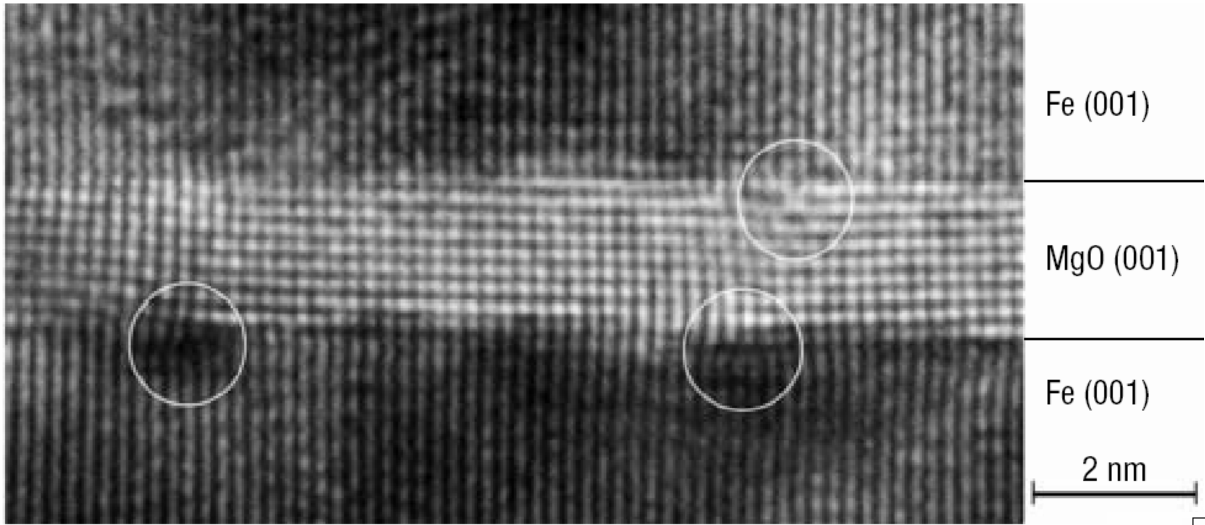
→ appropriate for disordered interfaces

TM films → TMR ~ 2\3 ~ 67%

TMR > 350%

S. Yuasa et al., Nature Materials 3, 868 [2004]

Fe/MgO/Fe



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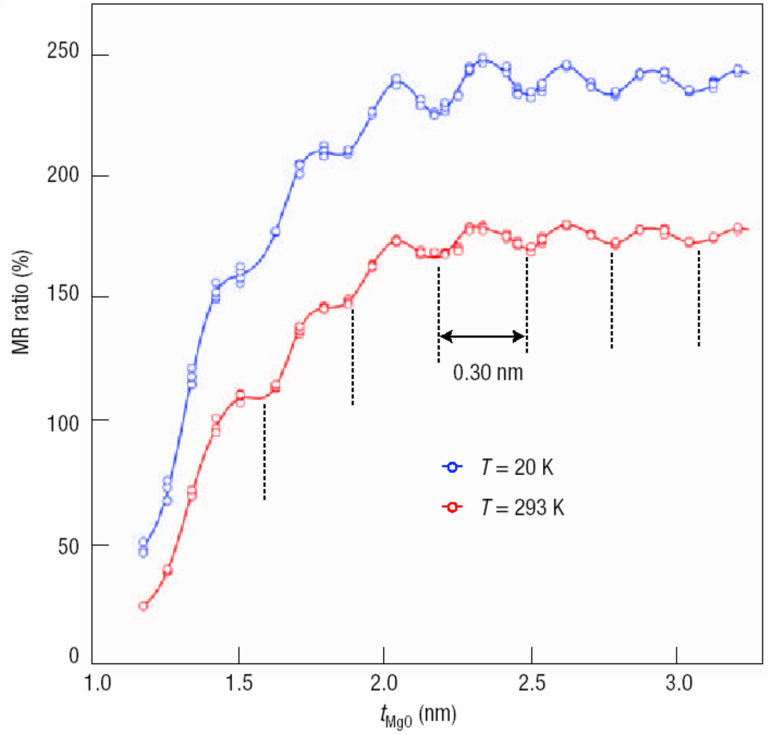
High-quality interface

Quantum effects important

Fabry-Perot interferences



Quantum mechanical model

W. H. Butler *et al*., Phys. Rev. B 63, 054416 [2001]

J. Mathon *et al*., Phys. Rev. B 63, 220403R [2001]

TMR ~ 1000% for:

\ ideal interface

\ T = 0 K

\ thickness = 20 ML

Quantum mechanical model –CoFeB/MgO/CoFeB

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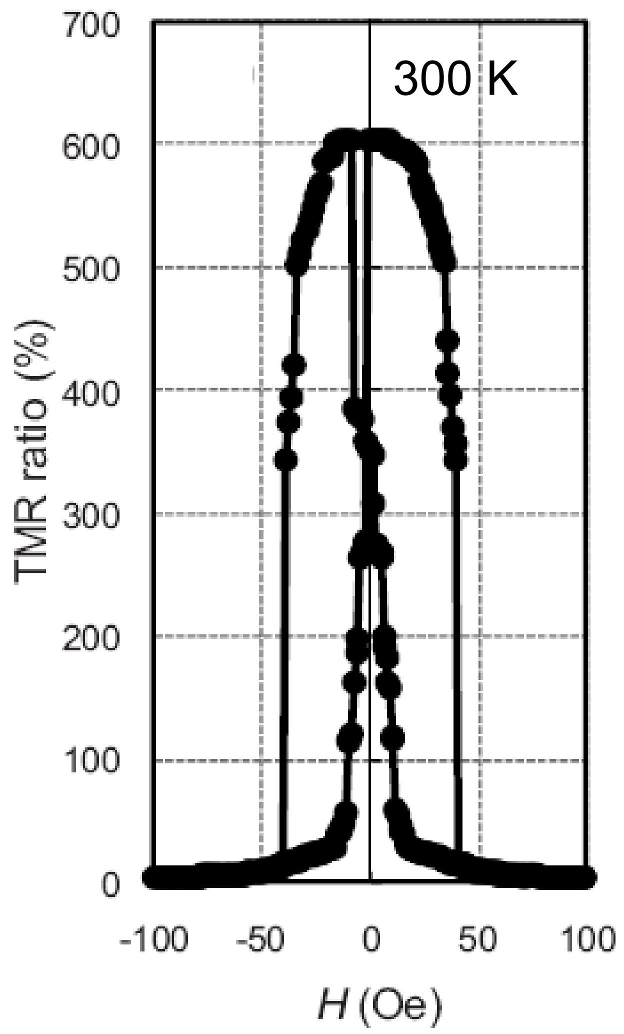

S. Ikeda *et al*., Appl. Phys. Lett 93, 082508 [2008]

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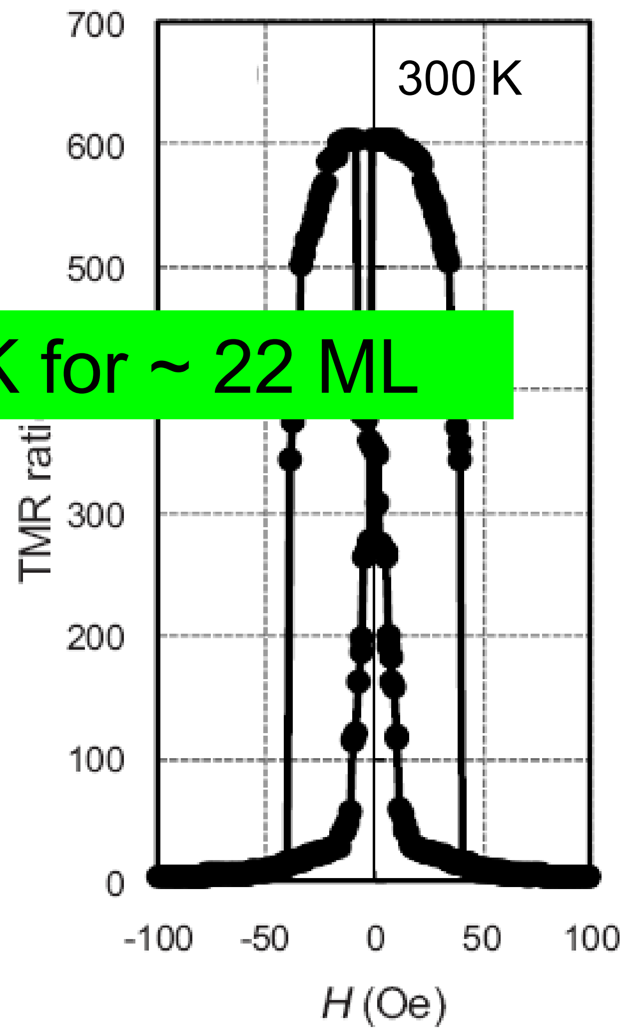
\ ideal interface

\ T = 0 K

\ thickness = 20 ML

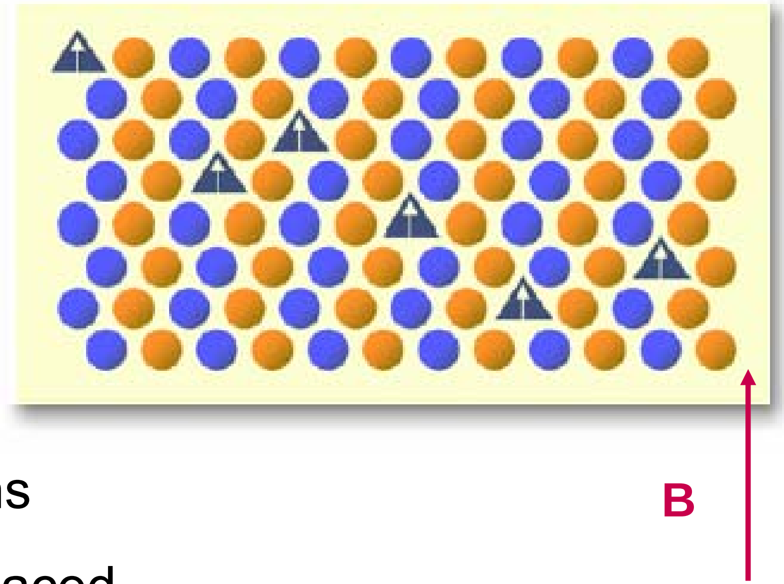
TMR = 1100% at 5 K for ~ 22 ML



S. Ikeda *et al*., Appl. Phys. Lett 93, 082508 [2008]

DMS

DMS – recalling the basics



Diluted Magnetic Semiconductors

\ semi-conducting materials

\ in which a fraction of the host cations

\ is substitutionally and randomly replaced

\ by **transition metals** or **rare earths**

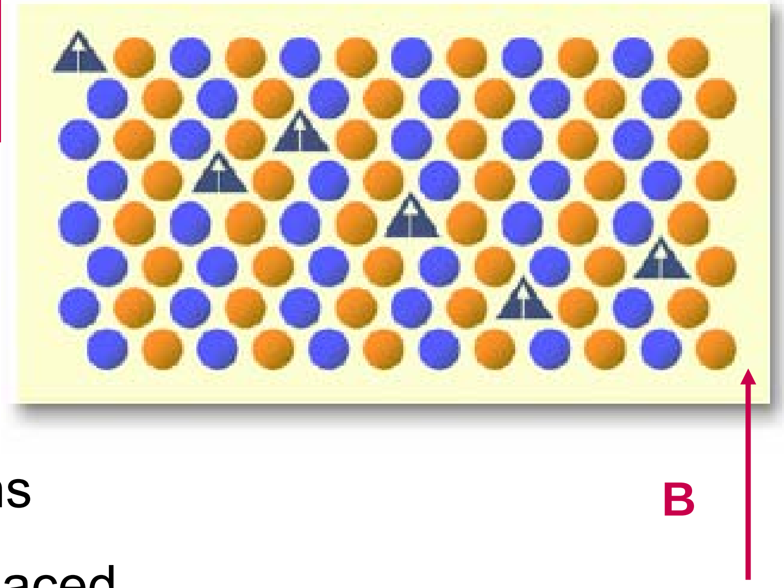
transition metals → partially filled *d*-states

rare earths → partially filled *f*-states

↓

unpaired electrons → magnetic behaviour

DMS – recalling the basics



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↓

unpaired electrons → magnetic behaviour

Challenges:

\ ferromagnetism

\ T_C above RT

Challenge

RT ferromagnetism in DMS theoretically requires:

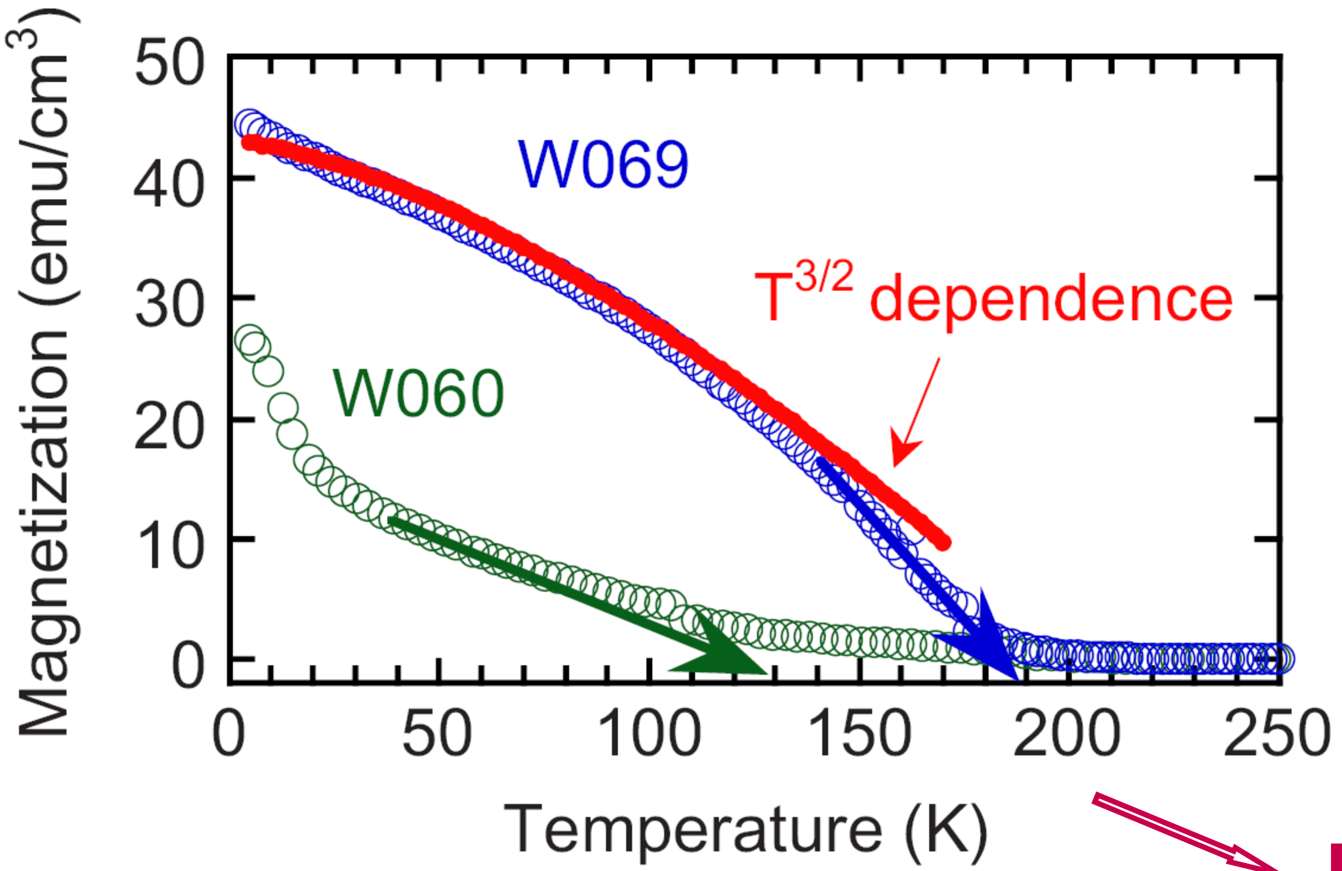
1] holes

2] magnetic ions

T.Dietl *et al*., Science **287**, 1019 [2000]

Ferromagnetic DMS: status

State-of-the-art for (Ge,Mn)Te



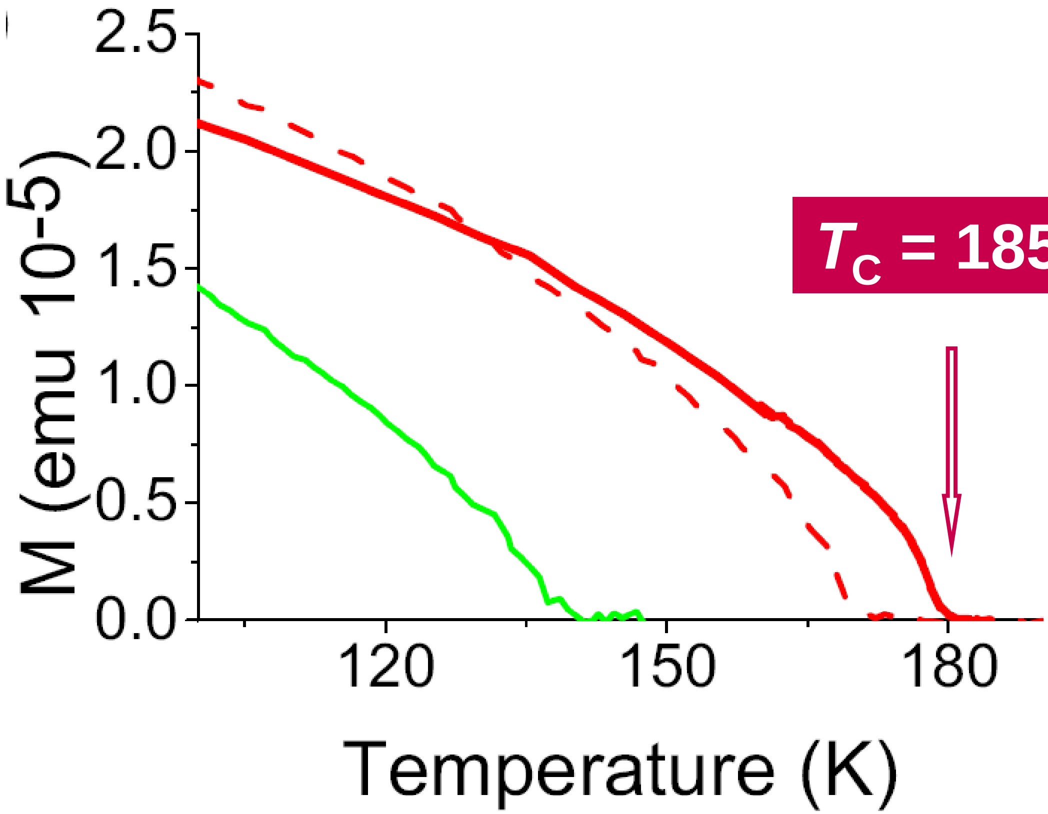
T_C = 190 K

8% (Ge,Mn)Te **1.57 x 10^{21} holes cm^{-3} :** **MBE**

Y. Fukuma *et al.* Appl.Phys.Lett. **93**, 252502 [2008]

State-of-the-art for (Ga,Mn)As

Situation 16 years (!) after the discovery of carrier-mediated mechanism in III-V



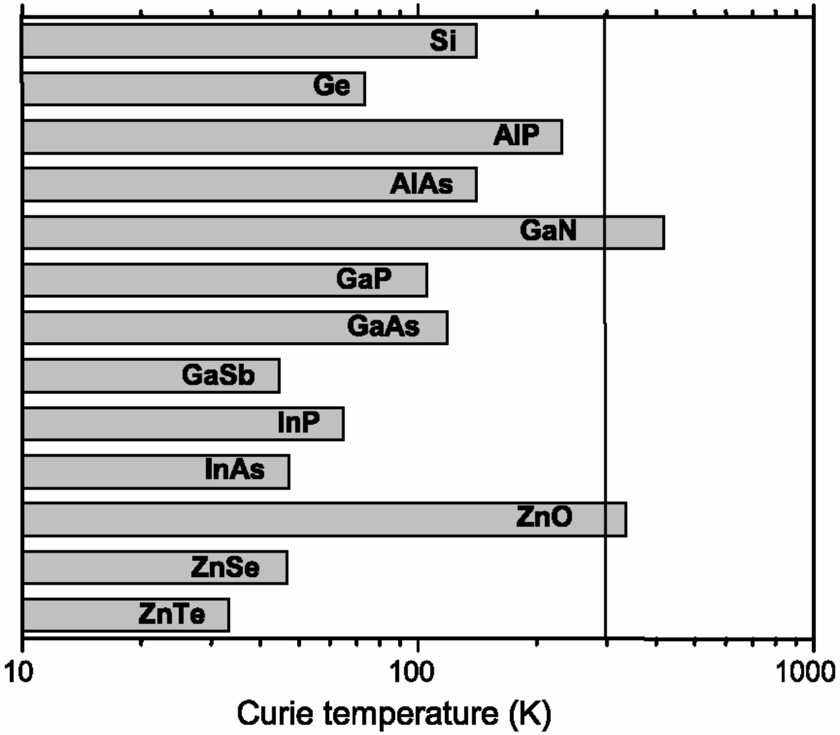
T

8% (Ga,Mn)As: annealed/etched/annealed

K.Olejník *et al.* Phys. Rev. B **78**, 054403 [2008]

M. Wang *et al.* Appl. Phys. Lett. **93**, 132103 [2008]

High T_C: most promising DMS



p-d Zener model prediction of T_C

5% Mn d^5, $p = 3.5 \times 10^{20}$ cm^{-3}

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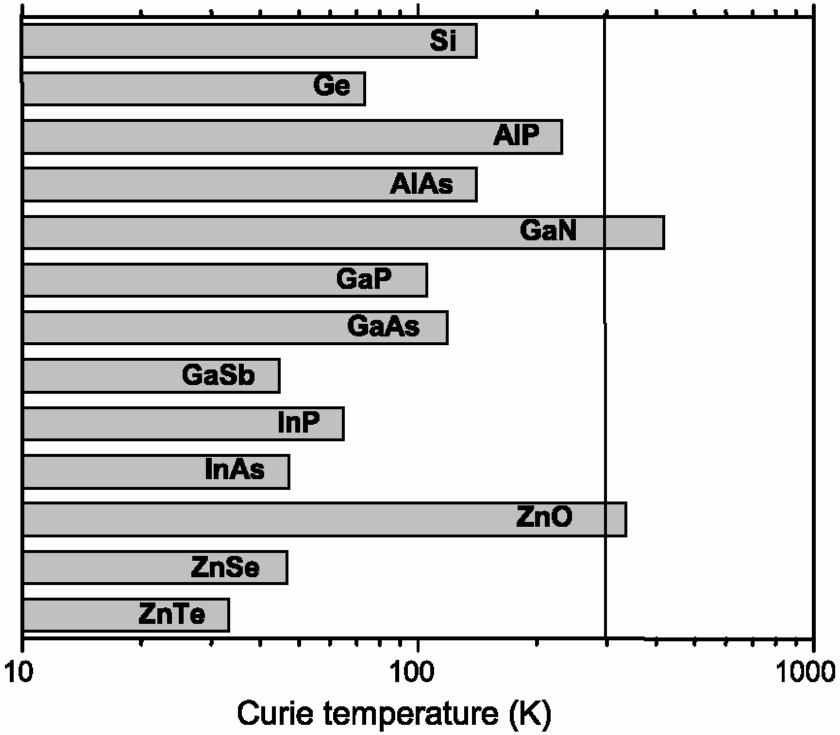

p-d Zener model prediction of T_C

5% Mn d^5, $p = 3.5 \times 10^{20}$ cm^{-3}

GaN & ZnO:

small lattice constant a_0 → strong *p-d* hybridization

\ increased $N_0\beta$

\ large T_C

T.Dietl *et al*., Science **287**, 1019 [2000]

Challenge

RT ferromagnetism in DMS theoretically requires:

1] holes

2] magnetic ions

T.Dietl *et al*., Science **287**, 1019 [2000]

Magnetization above room temperature reported even:

1] without valence band holes

2] without magnetic ions (!)

Magnetically doped materials

With spontaneous magnetization at 300 K

wz-c-**(Ga,Mn)N**, (In,Mn)N, (Ga,Cr)N, (Al,Cr)N, (Ga,Gd)N, **(Ga,Fe)N**

(Ga,Mn)As, (In,Mn)As, (Ga,Mn)Sb, (Ga,Mn)P:C

(Zn,Mn)O, (Zn,Ni)O, (Zn,Co)O, (Zn,V)O, (Zn,Fe,Cu)O

(Zn,Cr)Te

(Ti,Co)O_2, (Sn,Co)O_2, (Sn,Fe)O_2, (Hf,Co)O_2

(Cd,Ge,Mn)P_2, (Zn,Ge,Mn)P_2, (Zn,Sn,Mn)As_2

(Ge,Mn)

(La,Ca)B_6,C, C_{60}, HfO_2…

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(Ga,Mn)As, (In,Mn)As, (Ga,Mn)Sb, (Ga,Mn)P:C

Origin of ferromagnetism

“The most challenging issue in nowadays physics of magnetism“

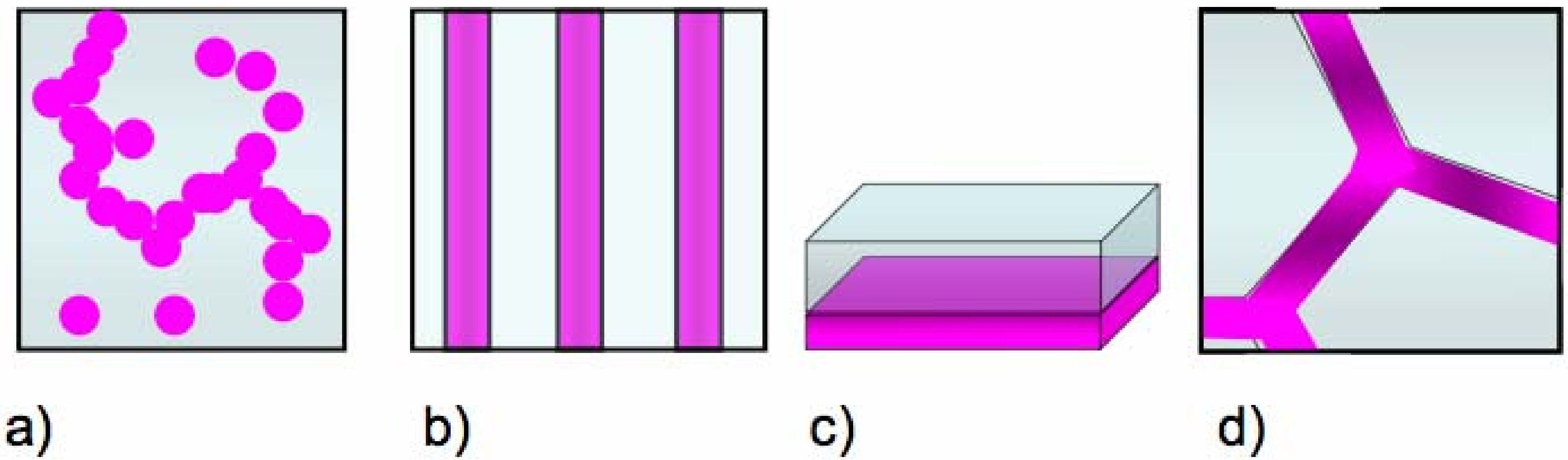
Mike Coey 04.09.2009 EMC

Phantom ferromagnetism

On account of the difficulty encountered and the sometimes ephemeral nature of the magnetic moment, we will refer to the phenomenon as phantom ferromagnetism

P.J. Grace *et al.* Adv.Mat. **21**, 71 [2009]

Inhomogeneous FM



a) random distribution

b) crystallographic or chemical phase separation

c) aggregation at surfaces or interfaces

d) aggregation in grain boundaries

To elucidate origin of ferromagnetism:

combine controlled epitaxy with comprehensive **nanocharacterization** for each material

New paradigm

Extended characterization – already online during growth

\ to elucidate correlation between fabrication conditions and

- structural [synchrotron XRD, HRTEM, EDS]
- magnetic [SQUID, EPR]
- optical [PL, magneto-optics]
- electrical [(magneto-)transport]
- chemical [EDS, SIMS]

properties

New paradigm

Extended characterization – already online during growth

\ to elucidate correlation between fabrication conditions and

- structural [synchrotron XRD, HRTEM, EDS]

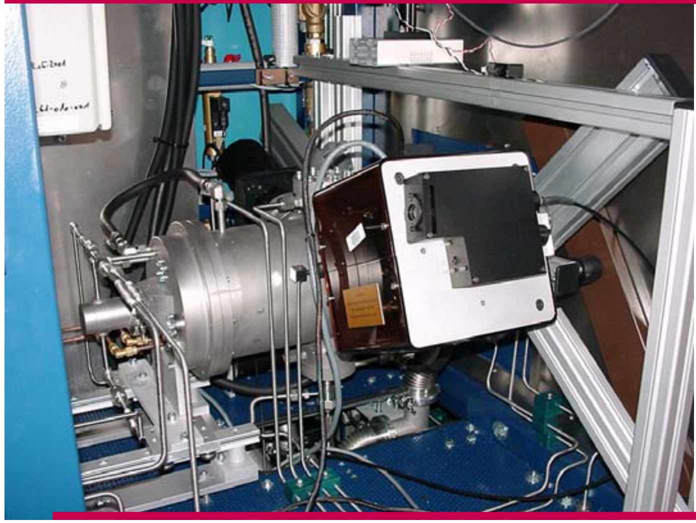
  - advanced microscopic characterization -

- magnetic [SQUID, EPR]
- optical [PL, magneto-optics]
- electrical [(magneto-)transport]
- chemical [EDS, SIMS]

} standard macroscopic characteriz.

properties

Growth – (Ga,Fe)N:Si(Mg)



- **MOVPE** [*in-situ:* ellipsometry, laser reflectometry]
  reactor: **AIXTRON 200**
- c-plane **Al_2O_3 substrates**
- **Precursors:** TMGa, NH_3, Cp_2Fe, Cp_2Mg, SiH_4, Cp_2Mn

- **Growth procedure:**

  1] substrate nitridation

  2] LT (540 °C) GaN nucl. layer

  3] annealing/recrystallisation

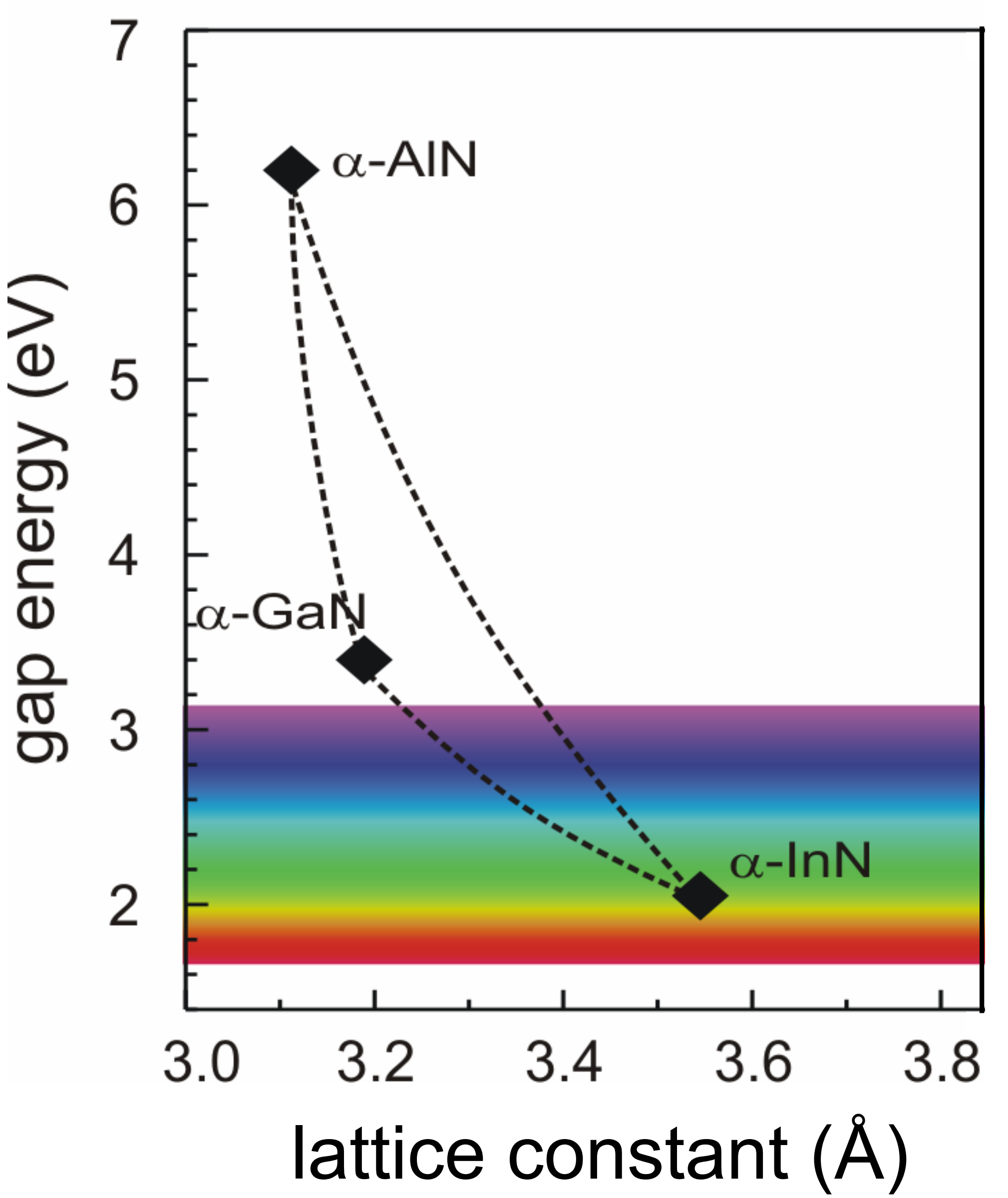
  4] 1 µm HT (1050 °C) GaN

  5] 0.5 – 1 µm (Ga,Fe)N:Si(Mg)

  a] 800 – 950 °C

  b] 50 – 400 sccm Cp_2Fe

III-Nitrides



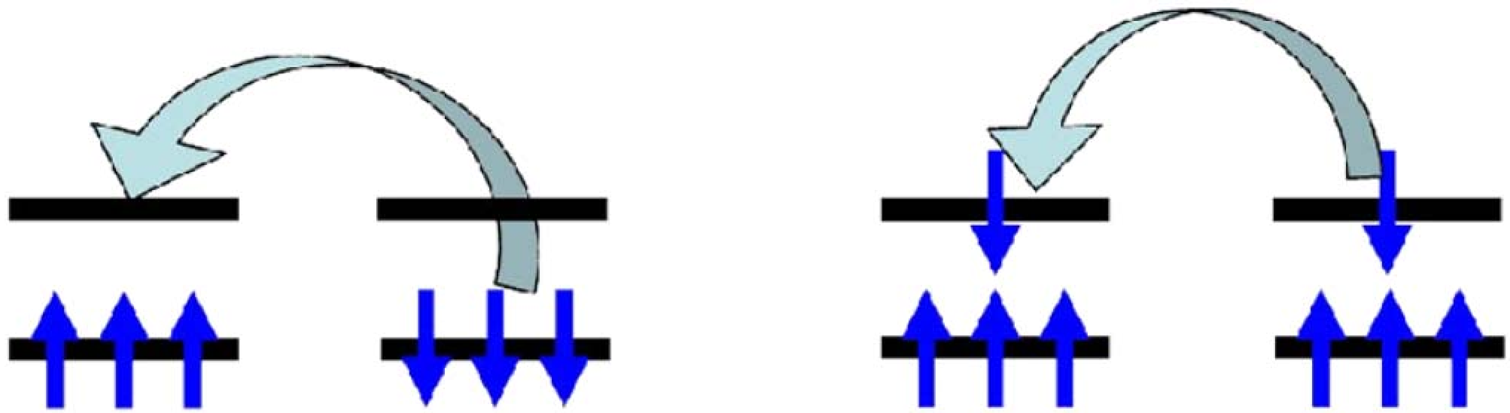
[Ferro]magnetic coupling without magnetic ions?

DFT [LSDA]:

\ cation vacancies

\ promote local magnetic moments

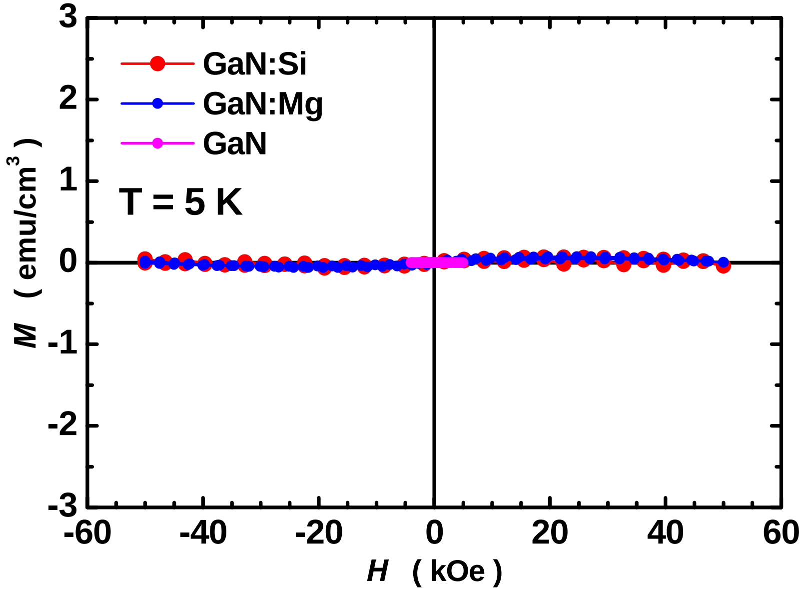
\ long-range magnetic coupling between intrinsic defects



Neutral cation Vacancies — Charged cation Vacancies

P. Dev *et al.* Phys.Rev.Lett. 100, 117204 [2008]

Not in high-quality samples - GaN



GaN(:Mg,Si) without Fe

⬇

no ferromagnetism

A. Bonanni *et al.* Phys.Rev.Lett. **101**, 135502 [2008]

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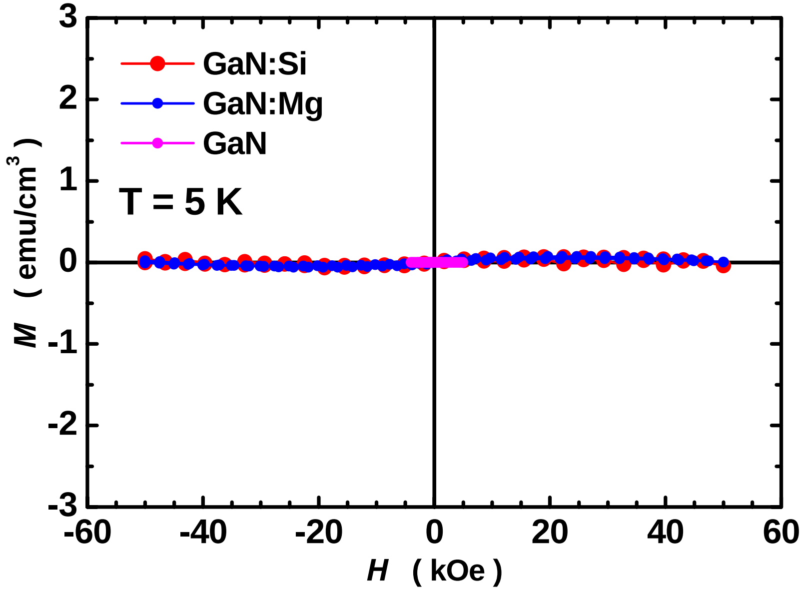

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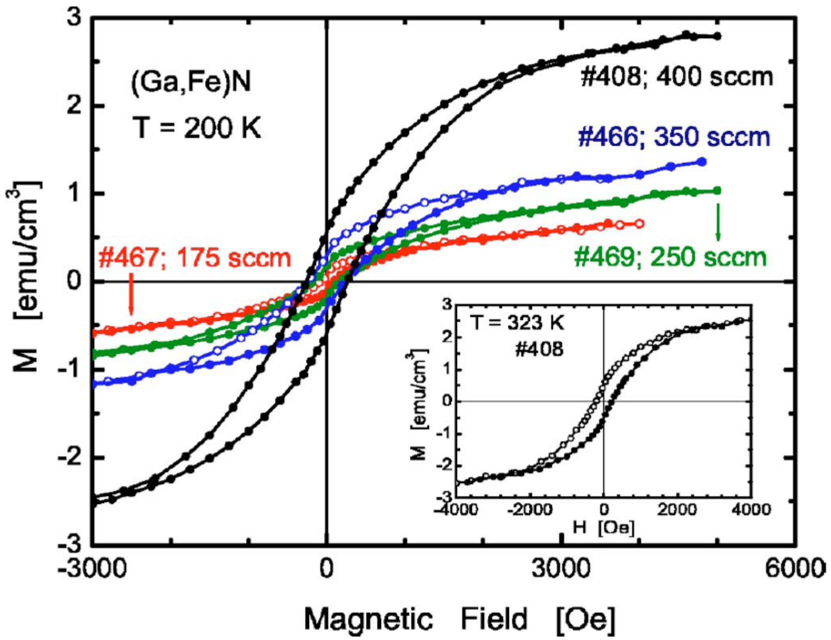


(Ga,Fe)N

↓

paramagnetism + ferromagnetism

A.Bonanni *et al.* Phys.Rev.B **75**, 125210 [2007]

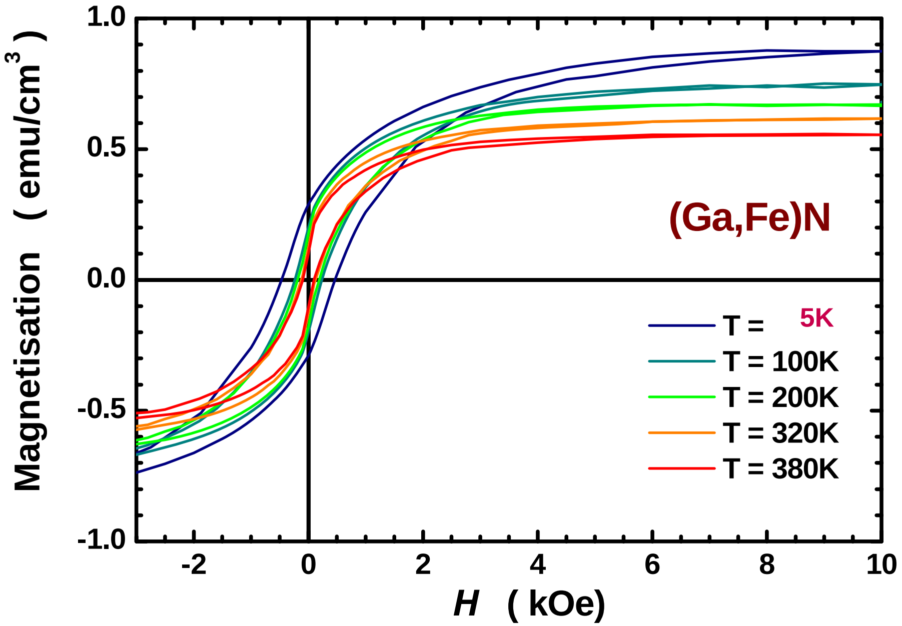
Phys. Rev.Lett. **101**, 135502 [2008]

Beyond the solubility limit – (Ga,Fe)N

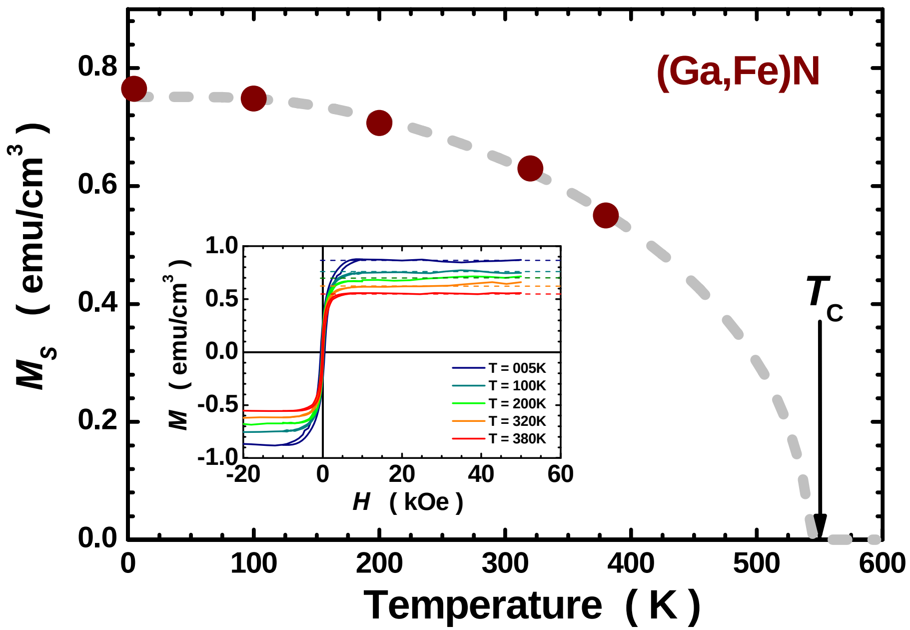
Superposition of paramagnetic and ferromagnetic response

A. Bonanni *et al.* Phys.Rev. B **75**, 125210 [2007]

Evaluation of T_C

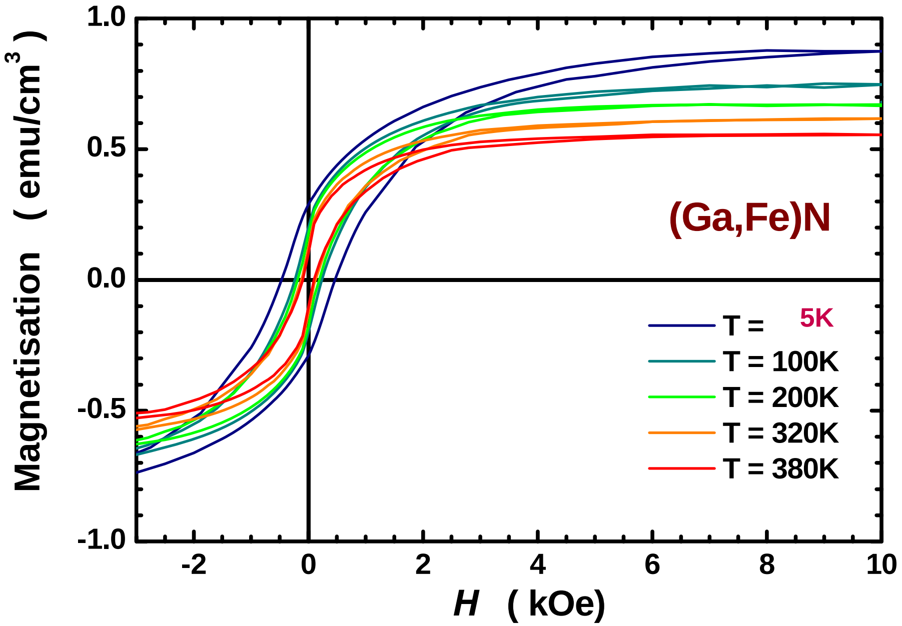


Ferromagnetic response persisting at room-temperature



- values of spontaneous magnetization M_S from high field measurements
- M_S vs T → Brillouin-like function → T_C

Evaluation of T_C



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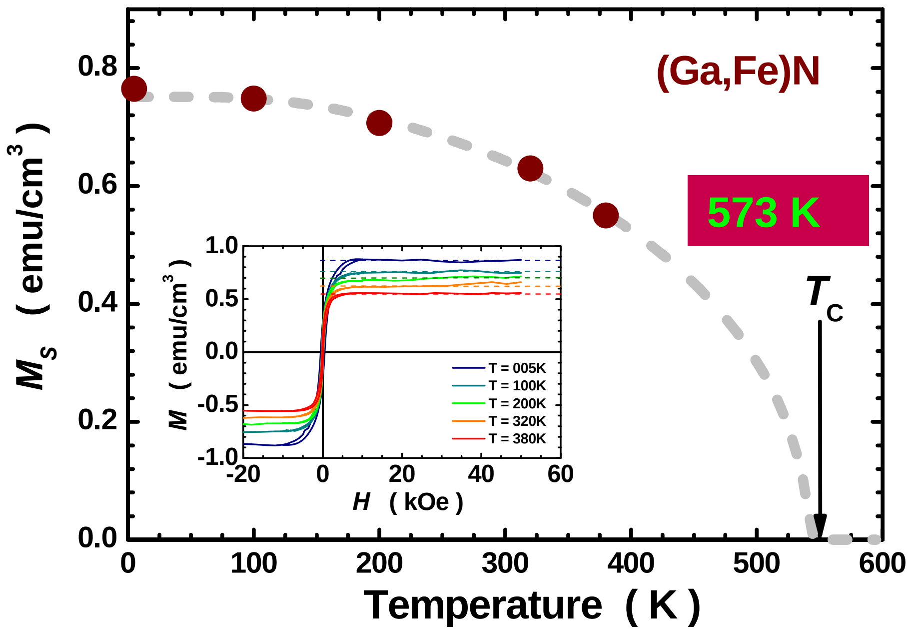

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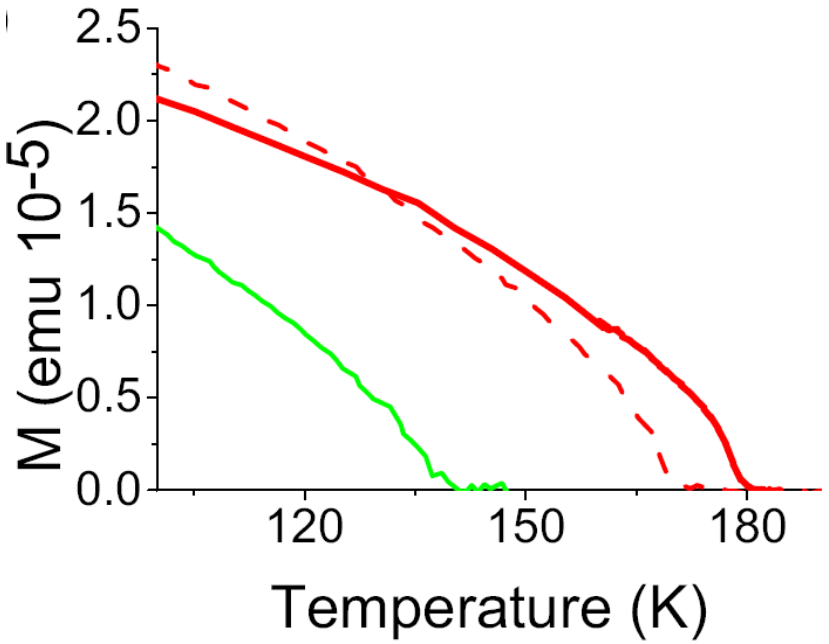
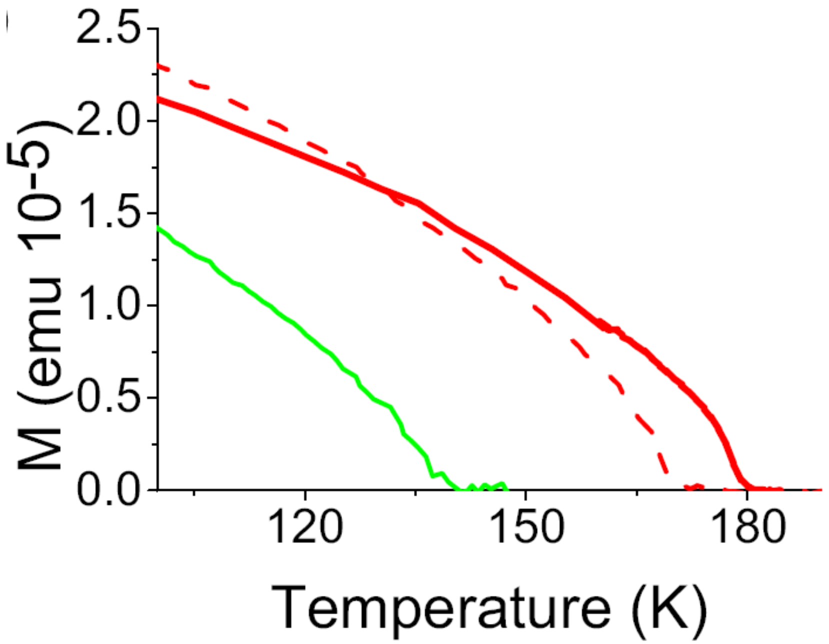


Brillouin function dependence [e.g. GaMnAs]

Evaluation of T_C



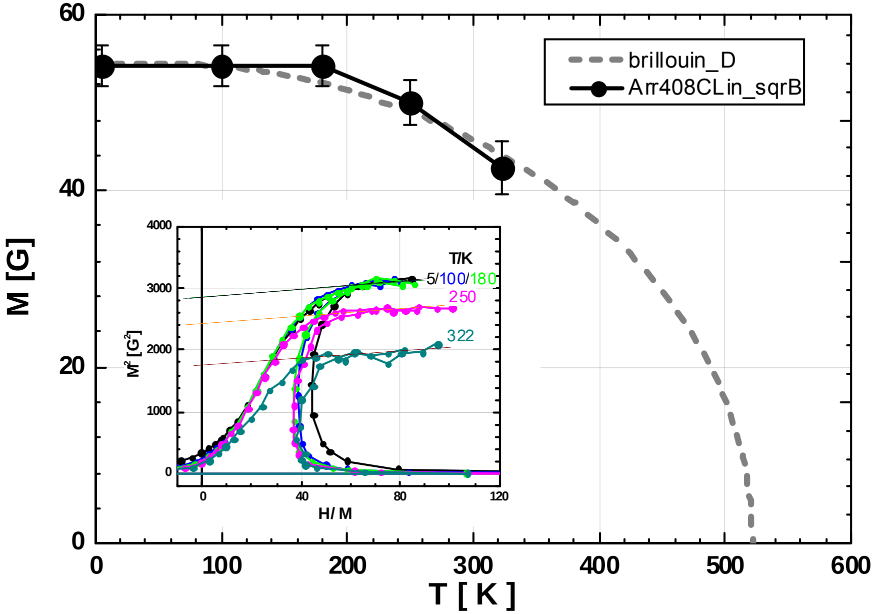
Brillouin function dependence [e.g. GaMnAs]

Otherwise:

Arrott plot

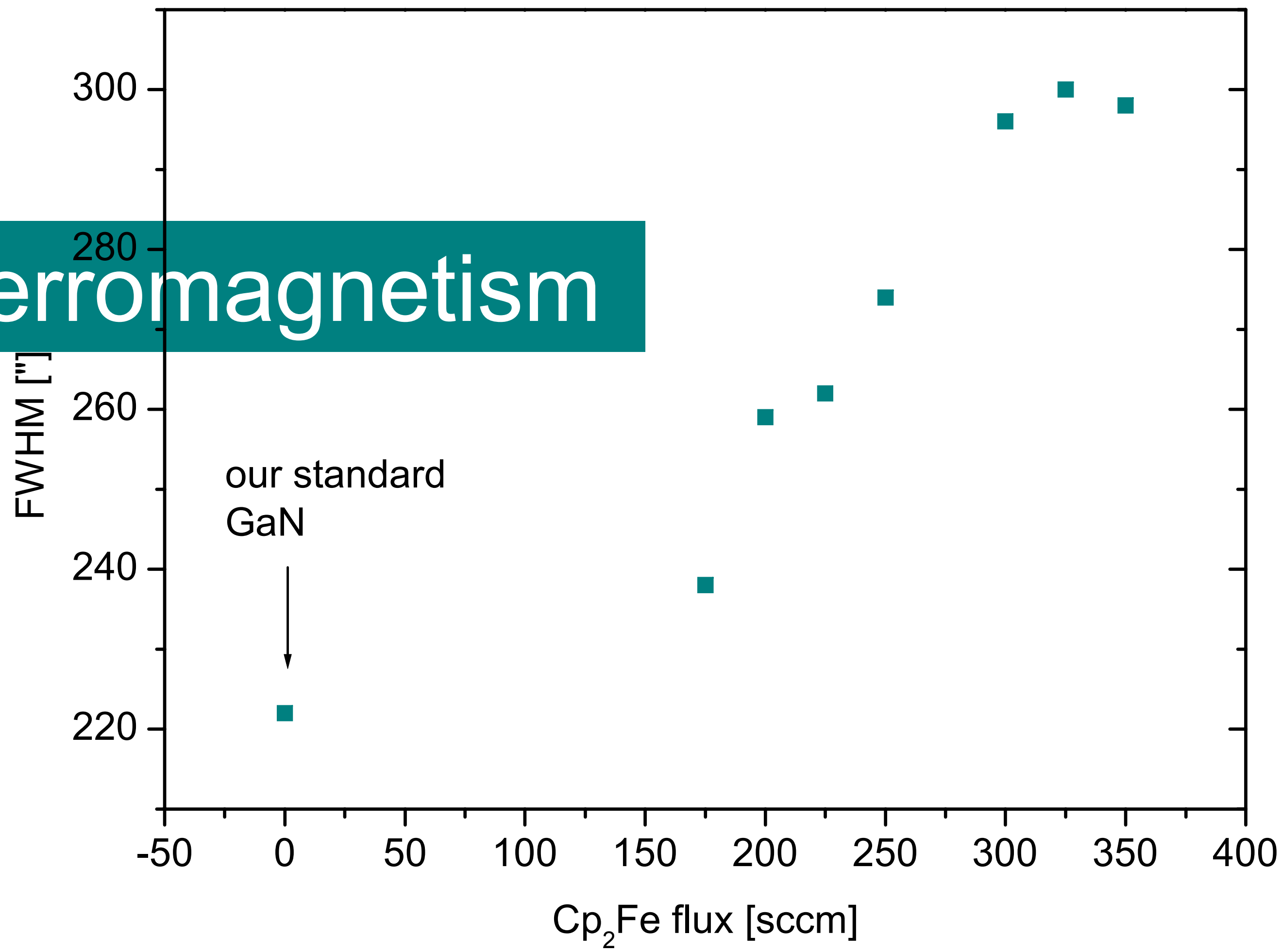
M^2 vs. H/*M*

extrapolated to 0 field



A. Arrott, Phys. Rev. **108**, 3194 [1957]

Open question



Origin of ferromagnetism

Review: A. Bonanni, Semicond.Sci.Technol. **22**, R41 [2007]

ESRF – European Synchrotron Radiation Facility

Grenoble - France

Synchrotron powder diffraction

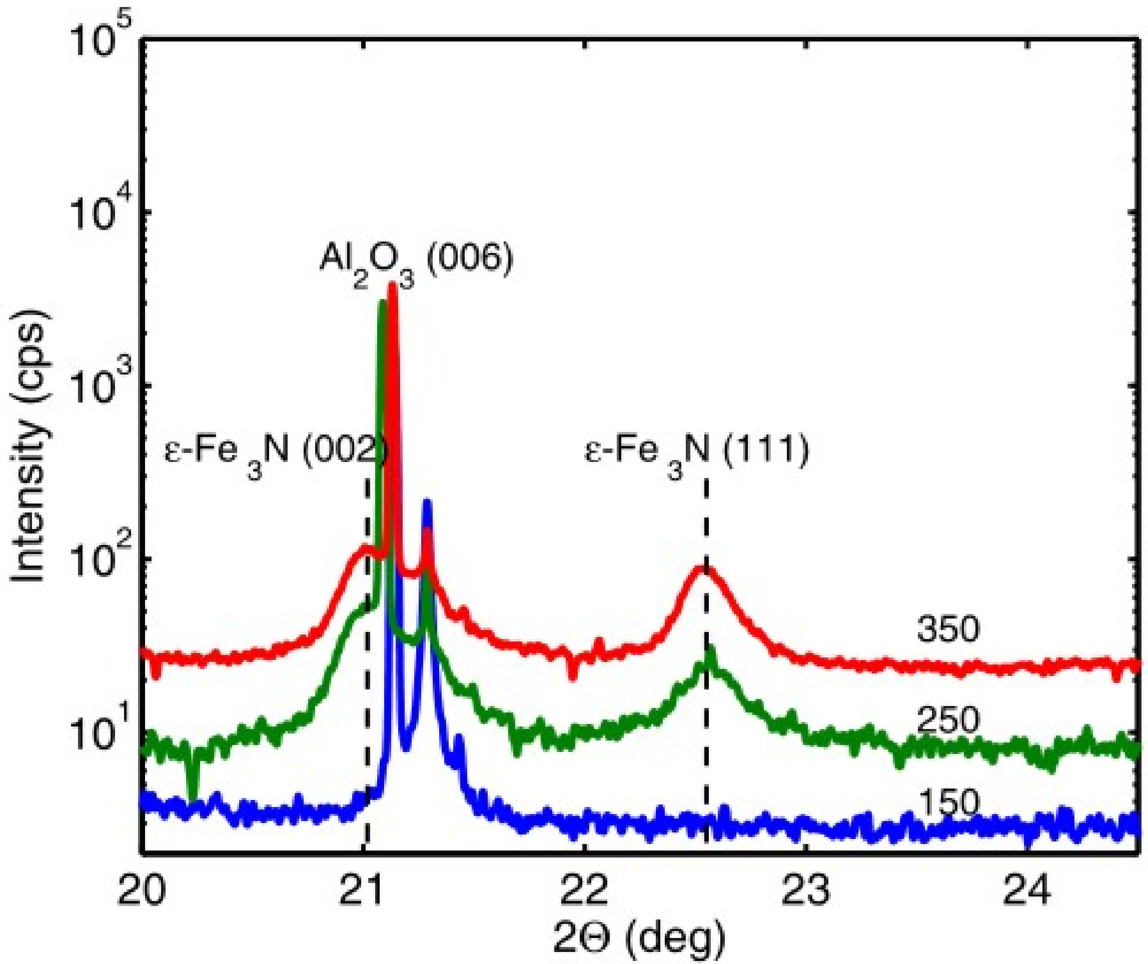
Above the solubility limit of Fe into GaN:

formation of nanocrystals confirmed by synchrotron XRD

\ ID31 beamline ESRF [Grenoble – France]

\ Powder diffraction

\ E = 15.5 keV



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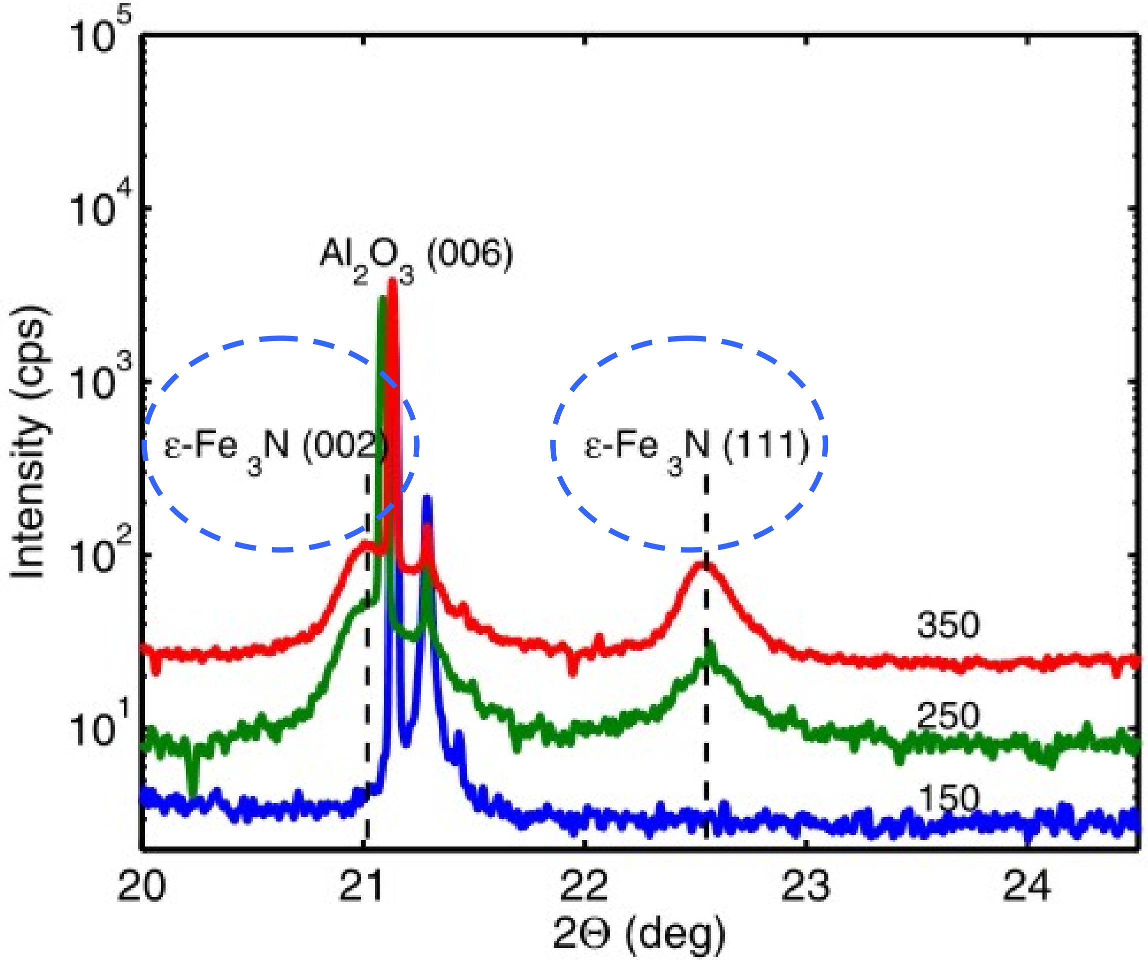
ε-Fe_3N hexagonal

T_C = 575 K



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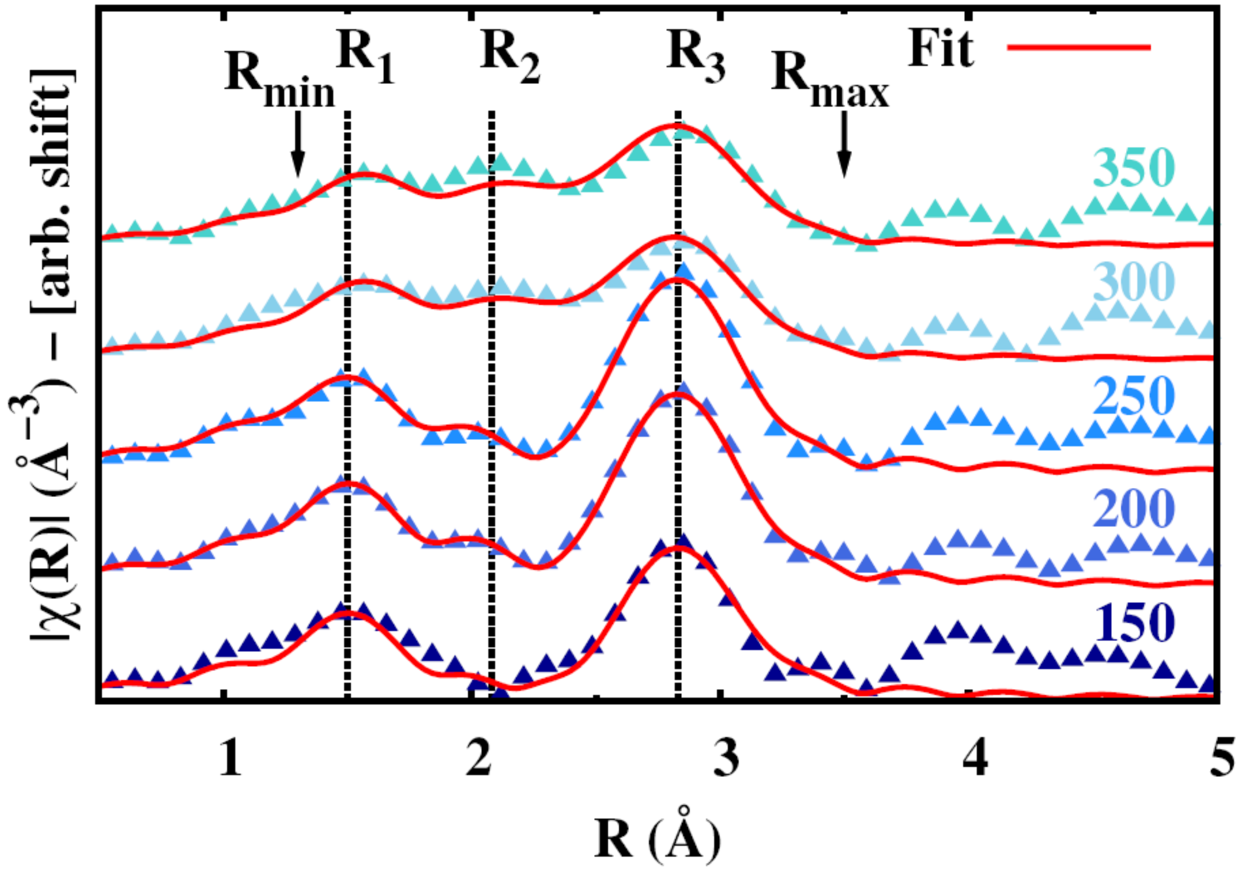

M. Rovezzi, ..AB, ...Phys.Rev. B **79**, 195209 [2009]

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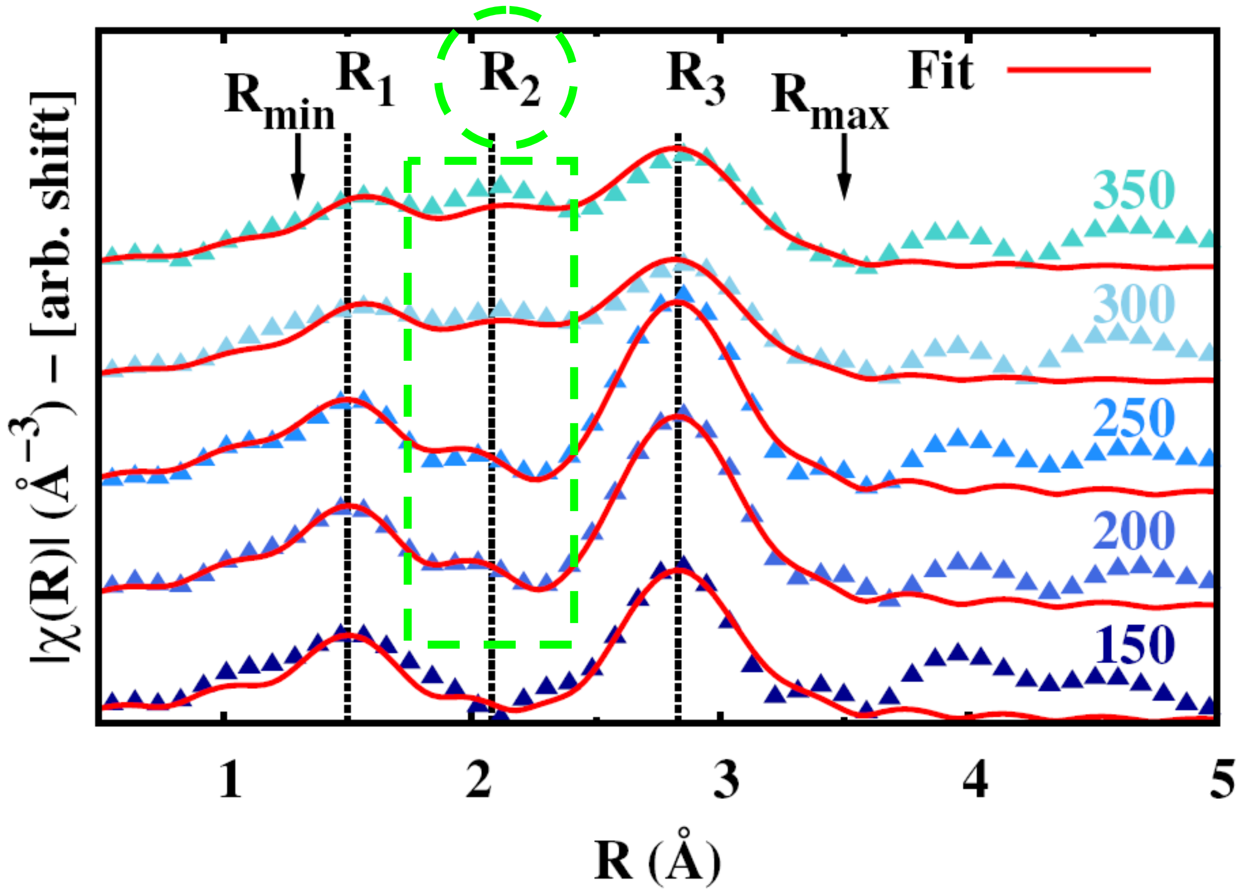
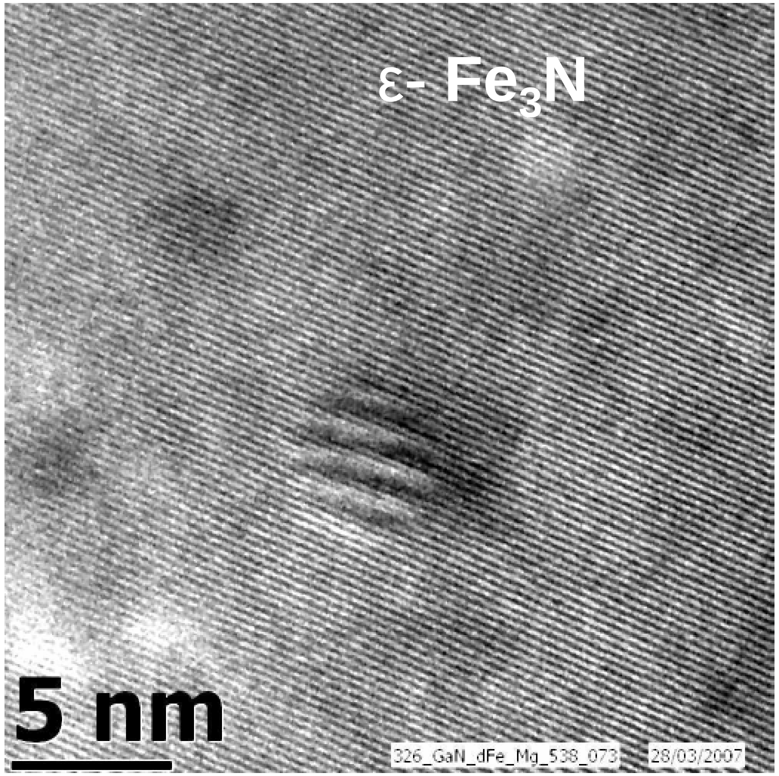


M. Rovezzi, ..AB, ...Phys.Rev. B **79**, 195209 [2009]

This is not the whole story



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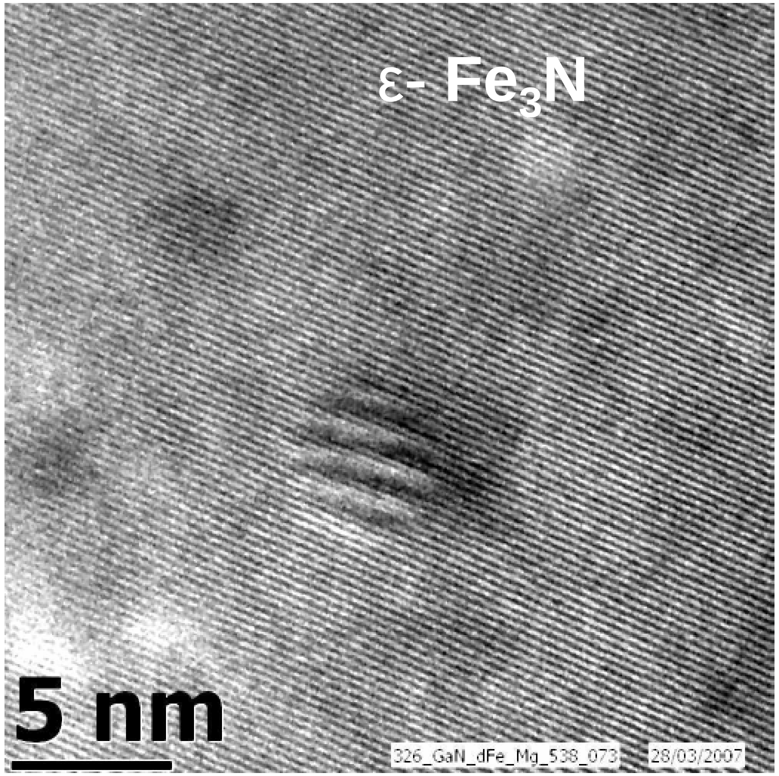

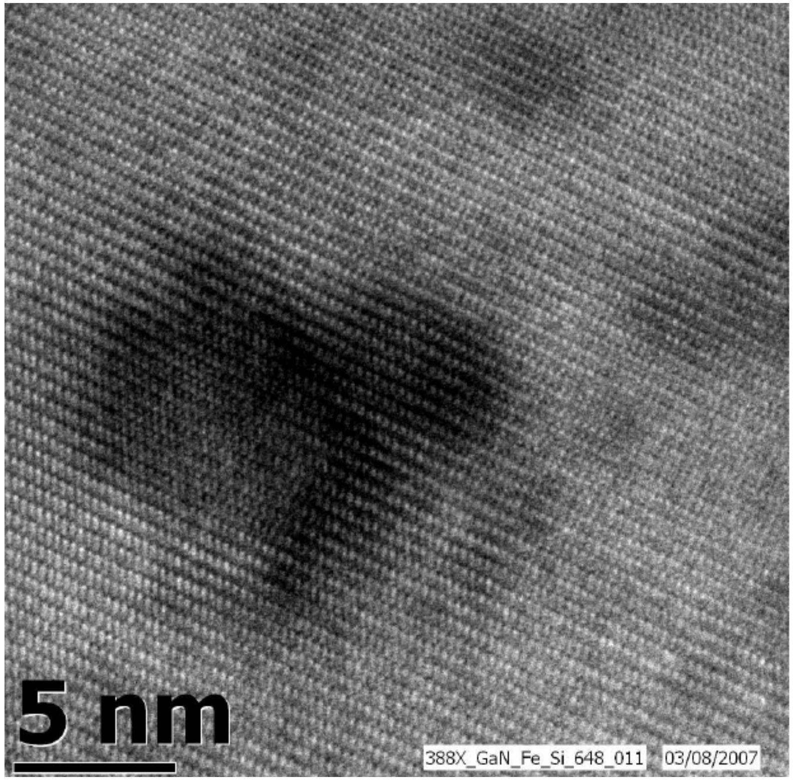

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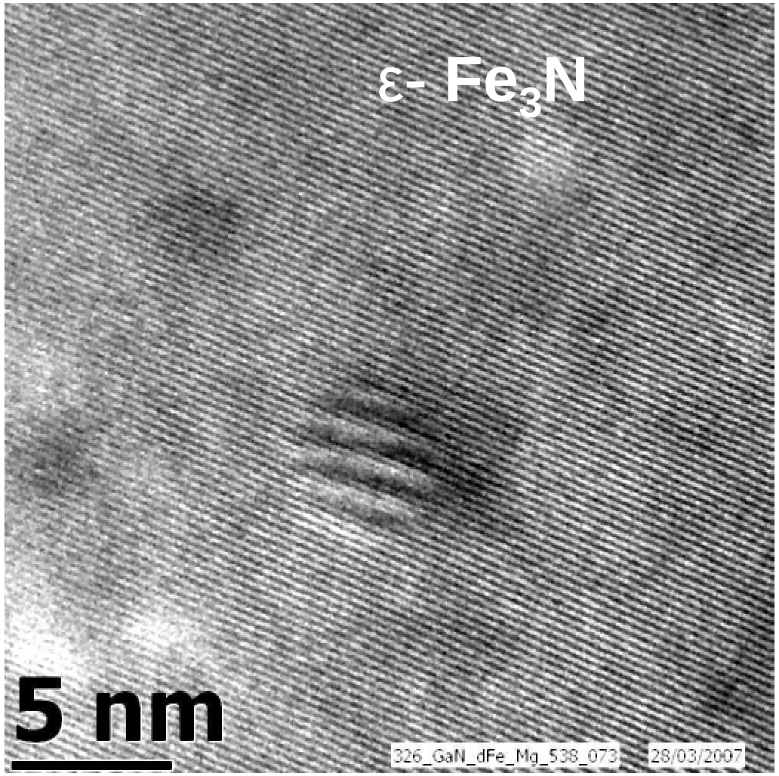

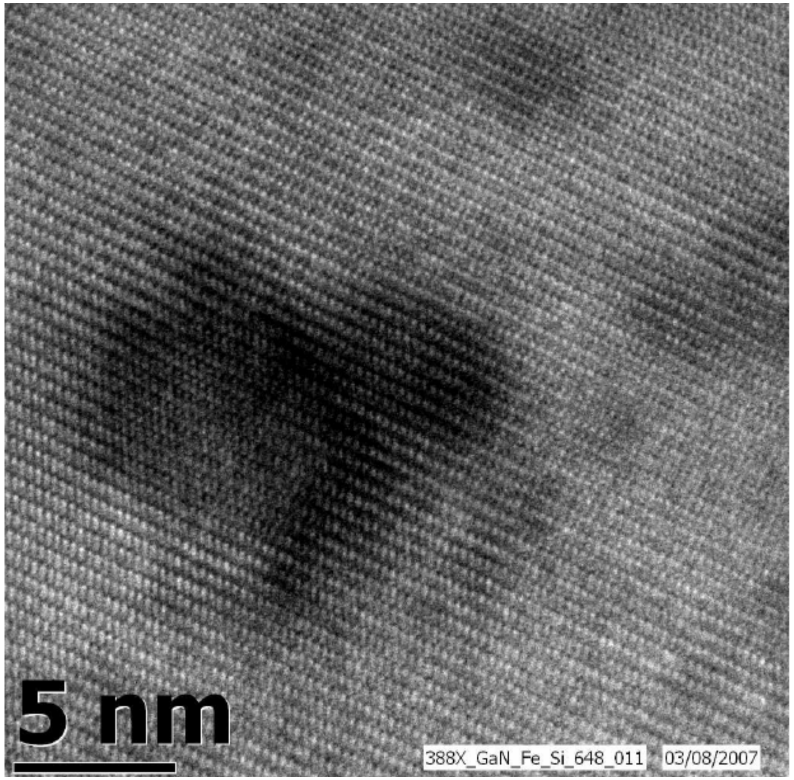

Related system → (Ga,Mn)N M. Zając et al., J.Appl.Phys **93**, 4715 [2003]

A. Martínez-Criado *et al.,* Appl.Phys.Lett. **86**, 131927 [2005]

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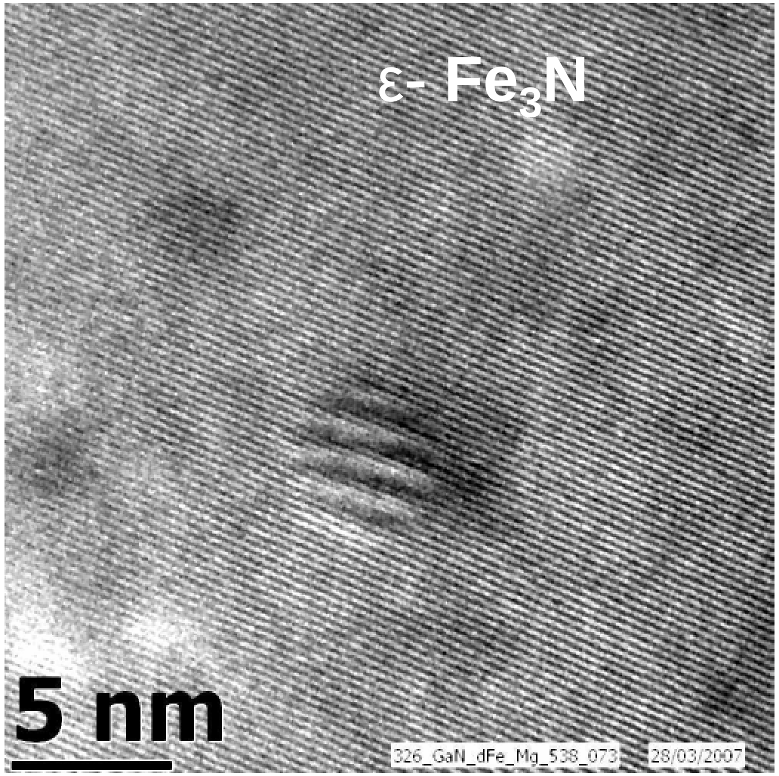

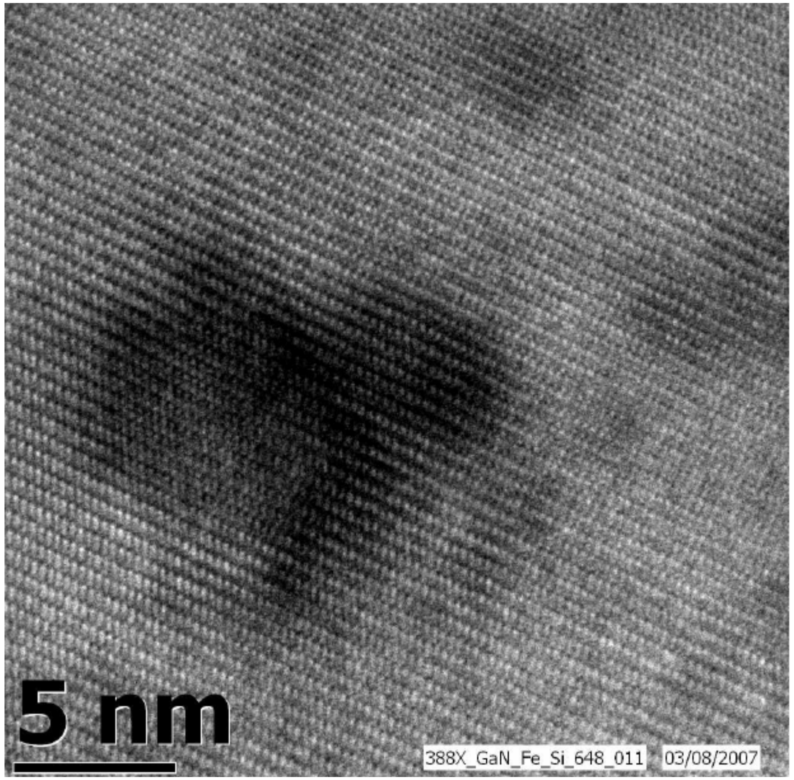

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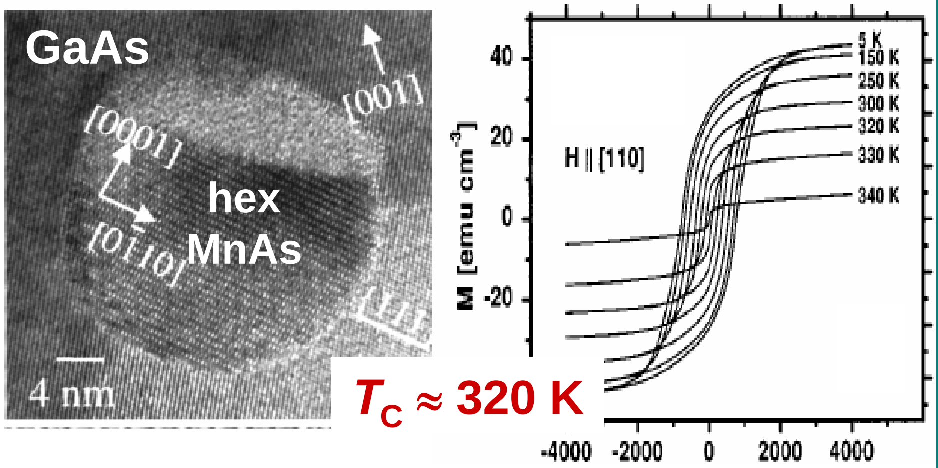
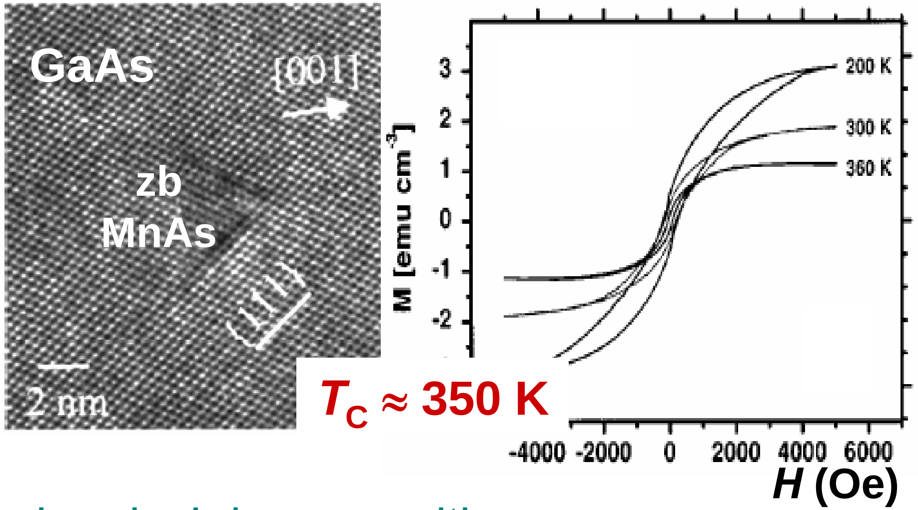
Buried condensed magnetic semiconductors [CMS]

source of high temperature ferromagnetism

Phase separation: (Ga,Mn)As



$T_C \approx 320$ K



$T_C \approx 350$ K

crystallographic decomposition

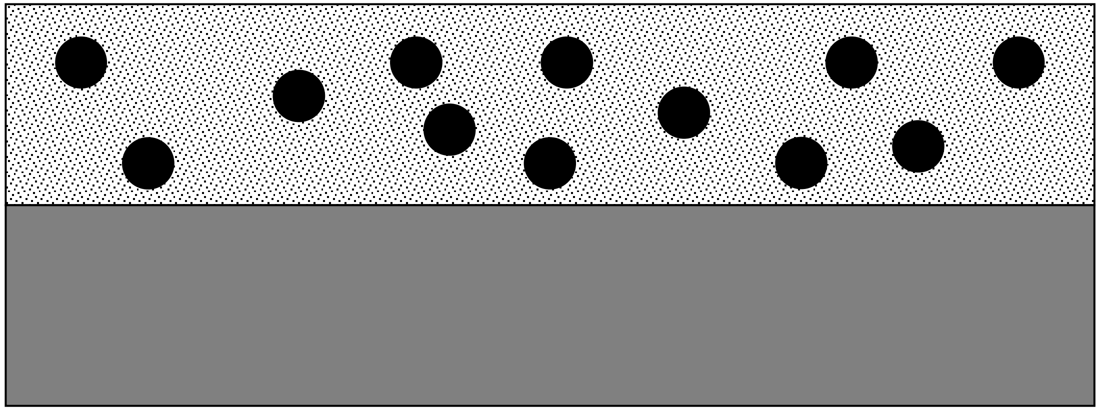
Moreno *et al.* (Berlin) *JAP* [2002]

chemical decomposition

Mn-rich regions

- control magnetic properties — De Boeck *et al.* *APL* [1996]
- enhanced magnetooptical effects [MCD] — Akinaga *et al.* *APL* [2000]; Shimizu *et al.* *APL* [2001]; Yokoyama *et al.* *JAP* [2005]
- affect conductance and Hall effect — Heimbrodt *et al.* *PRB* [2004]

From the box



Anisotropy field

$H_a = (2\,K_{eff})/(\mu_0\,M_s)$

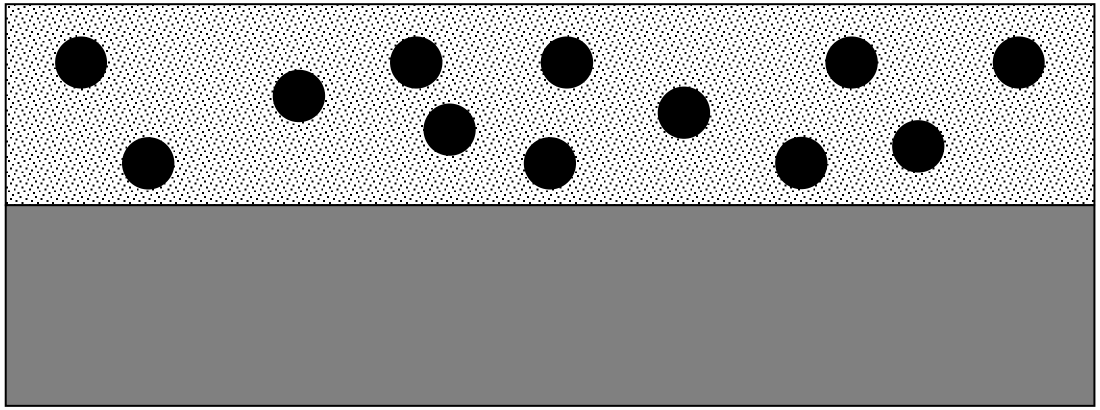
$K_{eff} = K_1 - \mu_0(N_{||} - N_{\perp})\,M_s$

SQUID → total magnetic moment

How do we setermine the saturation magnetisation M_s?

from full volume or from the volume of the particle?

From the box



Anisotropy field

$H_a = (2\,K_{eff})/(\mu_0\,M_s)$

$K_{eff} = K_1 - \mu_0(N_{||} - N_{\perp})\,M_s$

SQUID → total magnetic moment

How do we setermine the saturation magnetisation M_s?

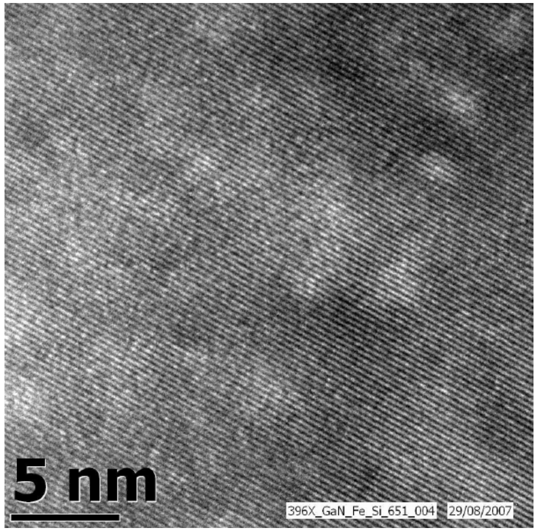
from full volume or from the volume of the particle?

volume of the ferromagnetic particle

Fe incorporation into GaN host

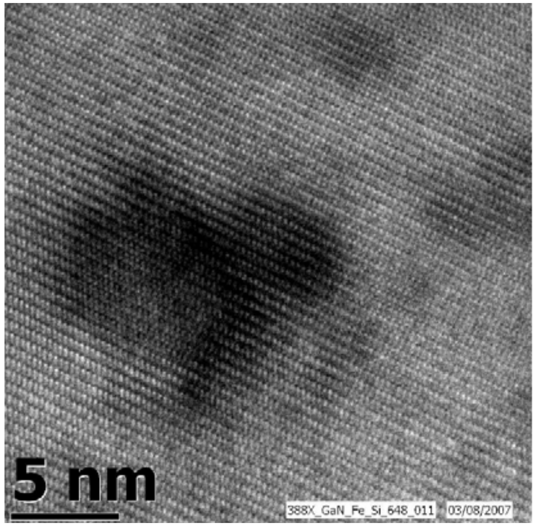
diluted material

paramagnetic behavior of Fe^{3+} ions



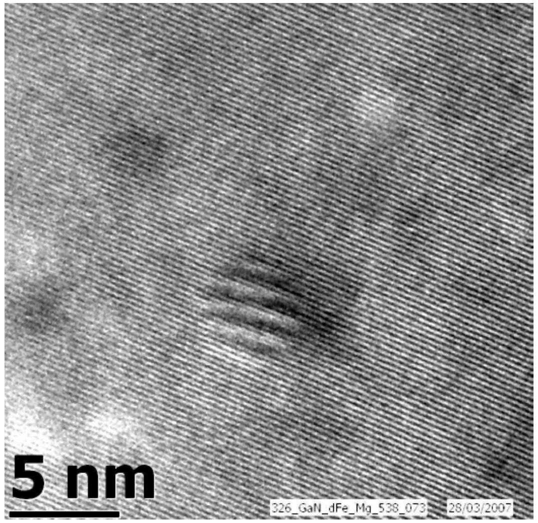
chemical decomposition

nano-scale chemical phase separation / regions with high magnetic ions concentration / novel magnetic phases stabilized



crystallographic decomposition

of known ferromagnetic, ferrimagnetic, antiferromagnetic compounds in semiconductor matrix



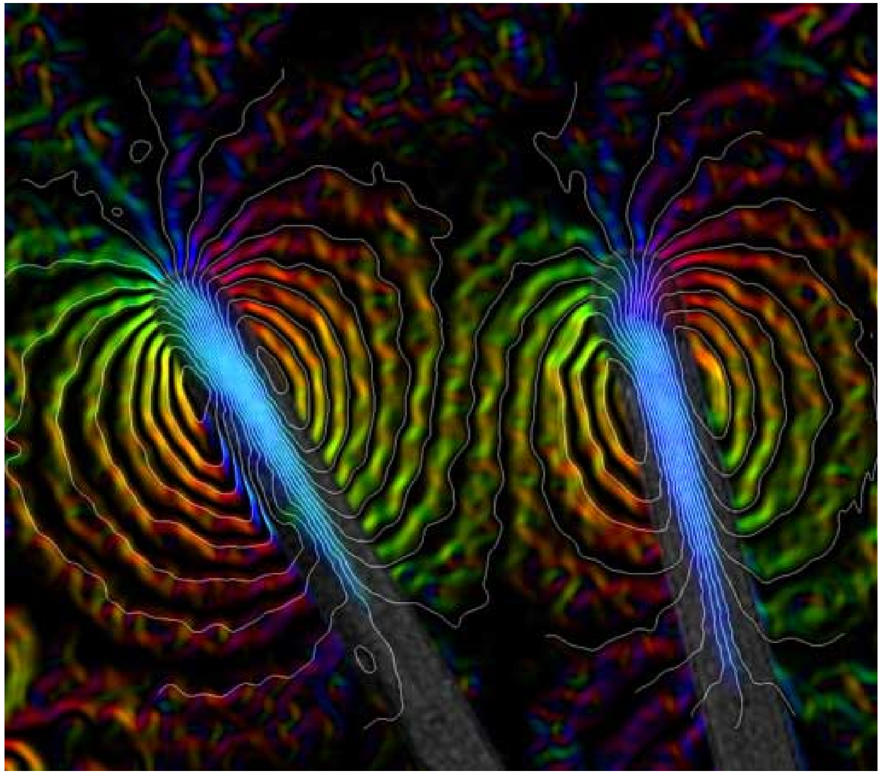
Advanced microscopy

Electron holography

\ TEM technique recording the phase of an electron wave

\ phase affected by a magnetic field

\ quantitative information on:



Field

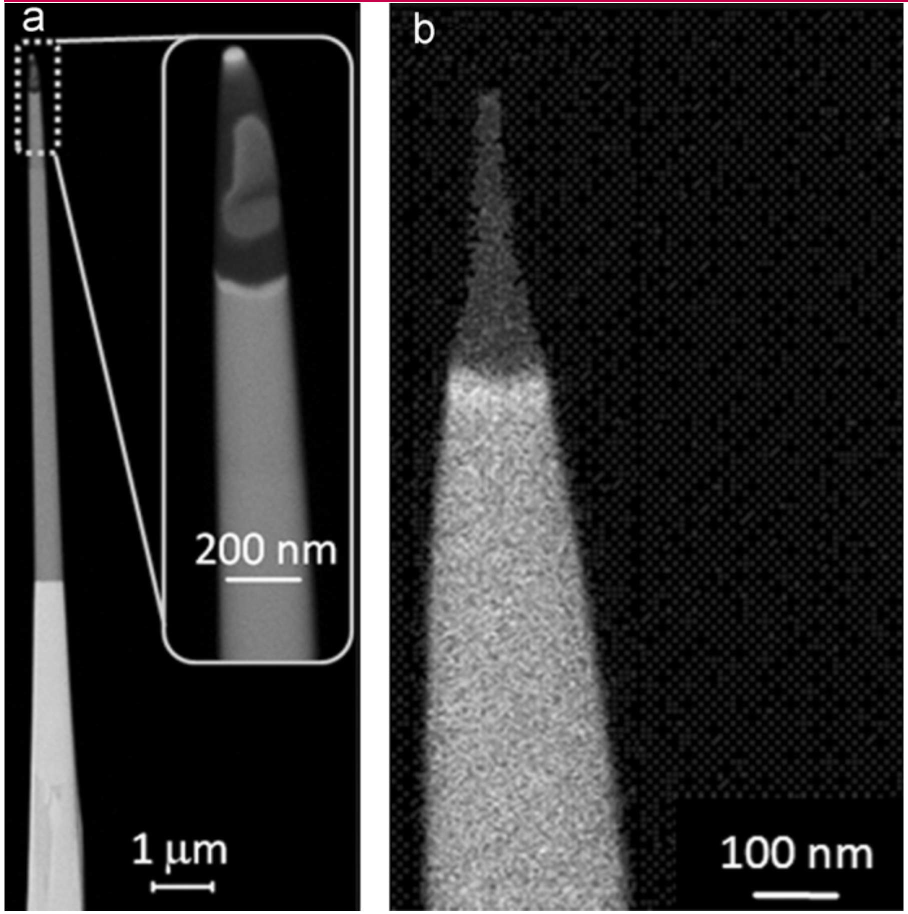
Magnetic moment

Magnetic nanotubes – courtesy of:

E. Simpson, T. Kasama, R. Dunin-Borkowski [DTU Copenhagen];
Y. Hayashi [Cambridge, Engineering Department]

R. Dunin-Borkowski *et al.* **Magnetic Microscopy of Nanostructures** Ch.5 Springer, Berlin [2006]

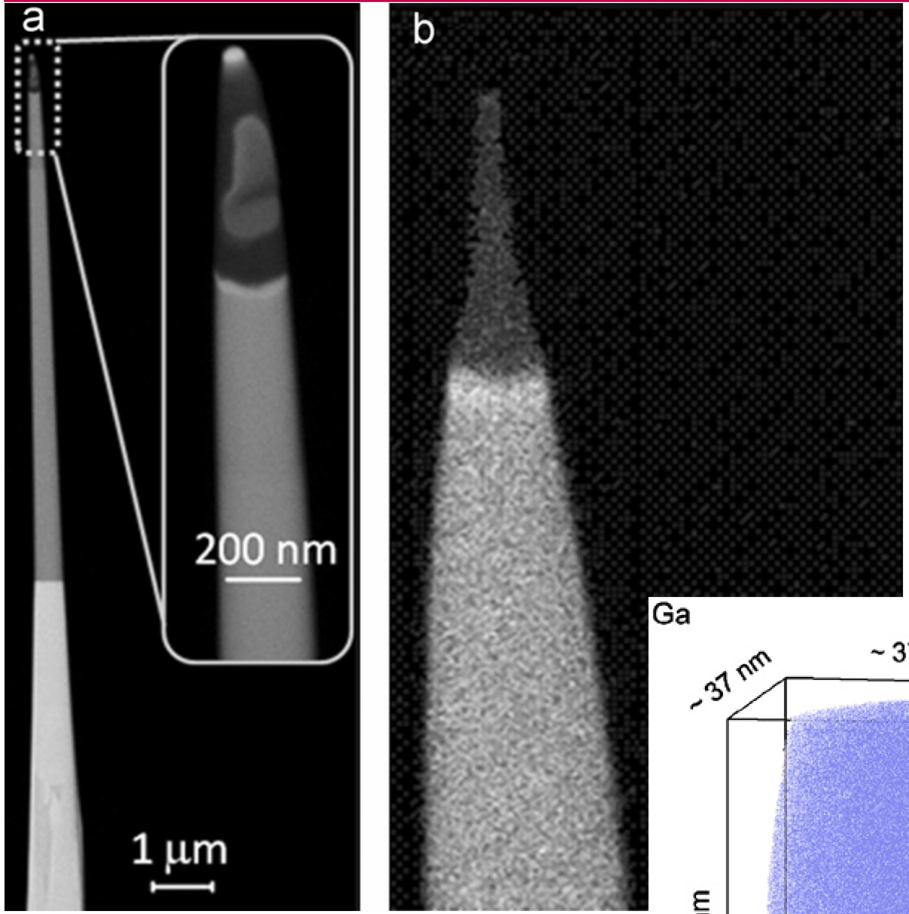
3D - atom probe



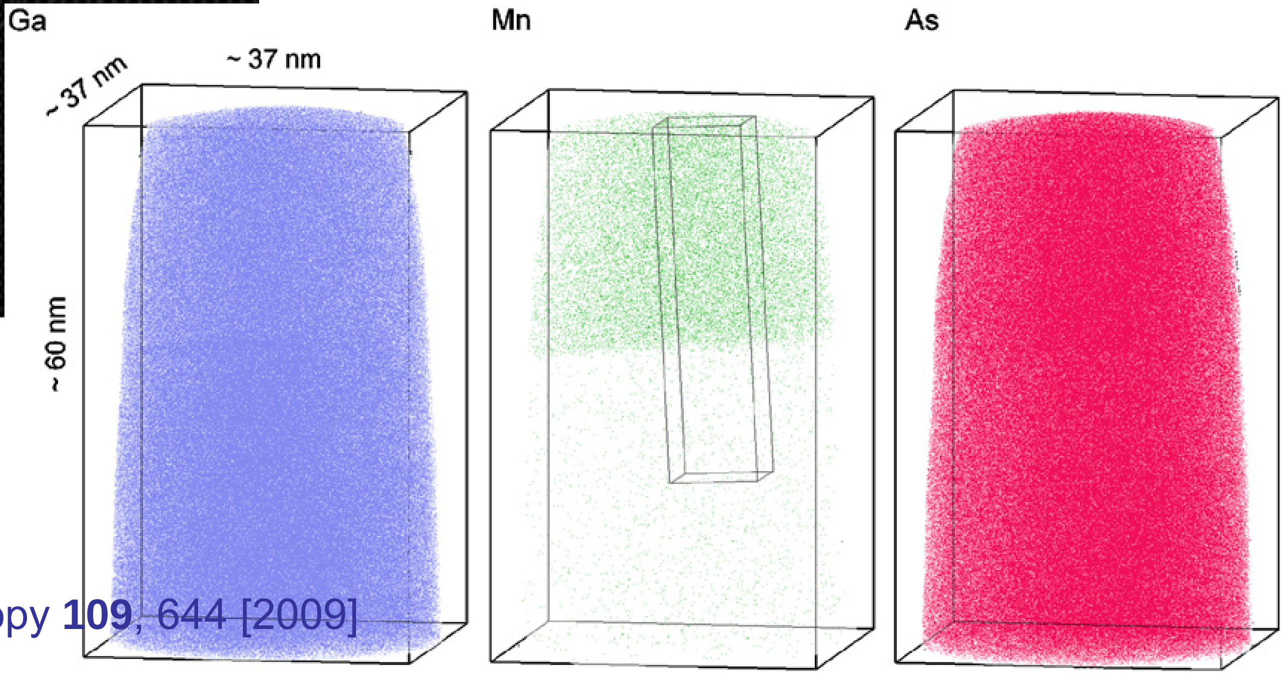
(Ga,Mn)As/GaAs

M. Kodzuka et al. Ultramicroscopy **109**, 644 [2009]

3D - atom probe



Compositional analysis at the nm-scale



(Ga,Mn)As/GaAs

M. Kodzuka et al. Ultramicroscopy **109**, 644 [2009]