



WHAT IS THE DIFFERENCE IN MAGNETISM OF SIZE CONTROLLED AND OF BULK MATERIAL?

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Today it is well known fact that the reduction of dimensionality of solid materials imposes extraordinary new features.

Discovery and understanding of the properties of nanostructures, quantum dots, nanowires and other low-dimensional interfaces, have lead to numerous technological applications.

Prominent examples are applications in information processing and information-storage technologies, new light sources, etc.

www.physik.fu-berlin.de/~bab



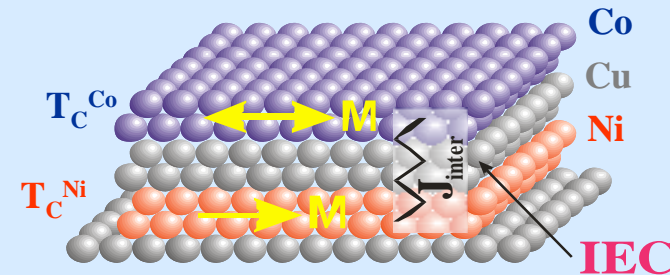
20 years ago

Part I: Fundamentals

- Curie temperature T_C ,
- Orbital- and spin- magnetic moments,
- Magnetic Anisotropy Energy (MAE)

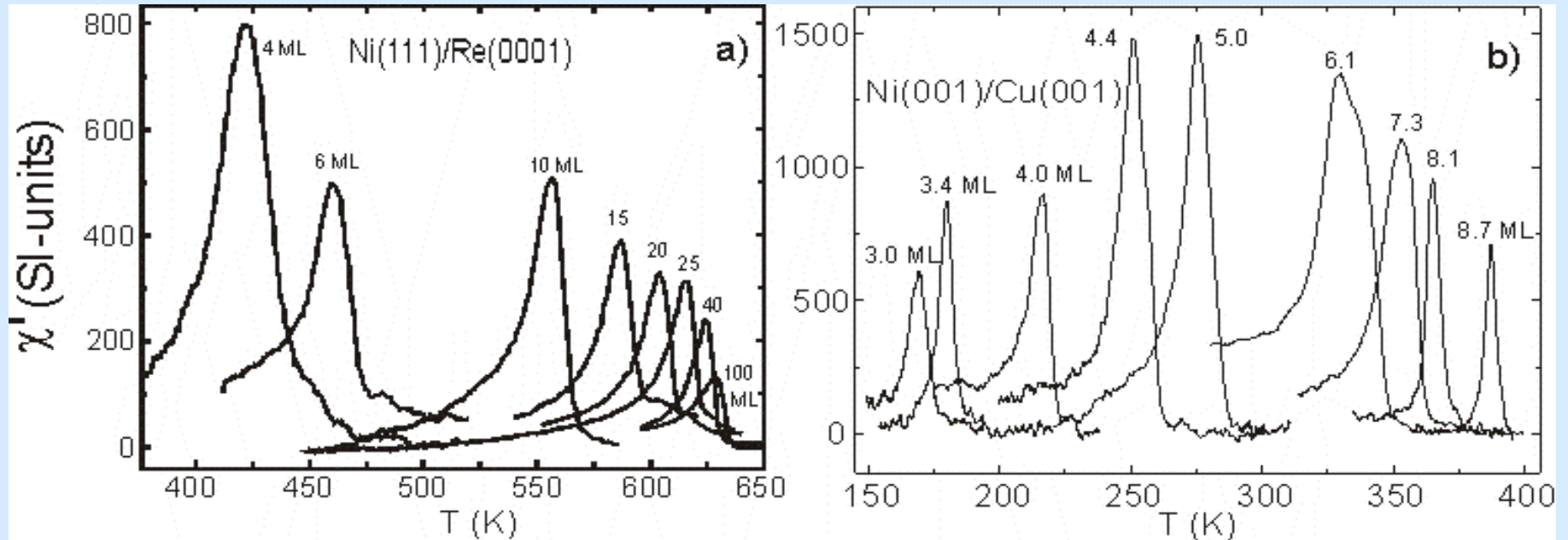
Part II: Spin dynamics in nano-magnets:

- Element specific magnetizations and T_C 's in trilayers.
- Interlayer exchange coupling and its T-dependence.
- Gilbert damping versus magnon-magnon scattering.



UHV – ac susceptibility

film prepared and measured *in-situ*



“Magnetism in thin films”

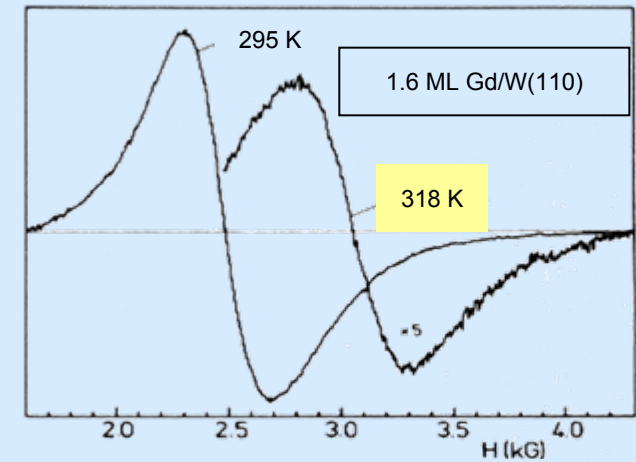
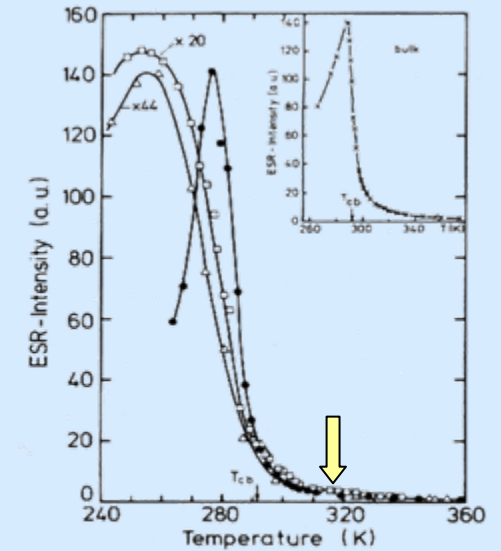
P. Pouloupoulos, K. B., J. Phys. Condens. Matter. **11**, 9495 (1999)

EPR / FMR in UHV

In situ UHV-FMR set up 1,4,9 GHz

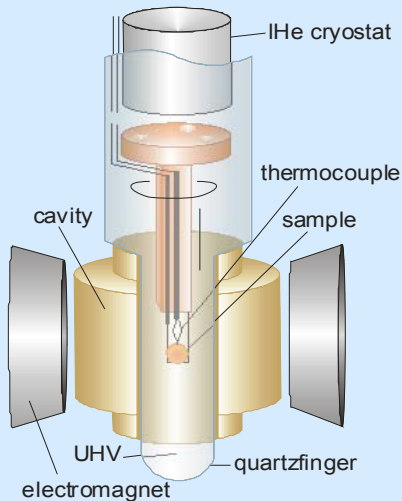
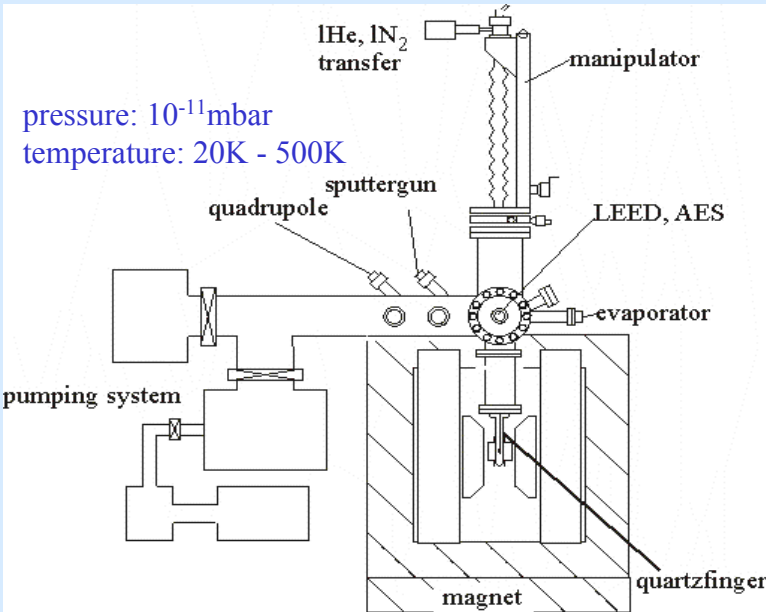
Gd/W(110)

M. Farle, K.B. PRL **58**, 511 (1987)

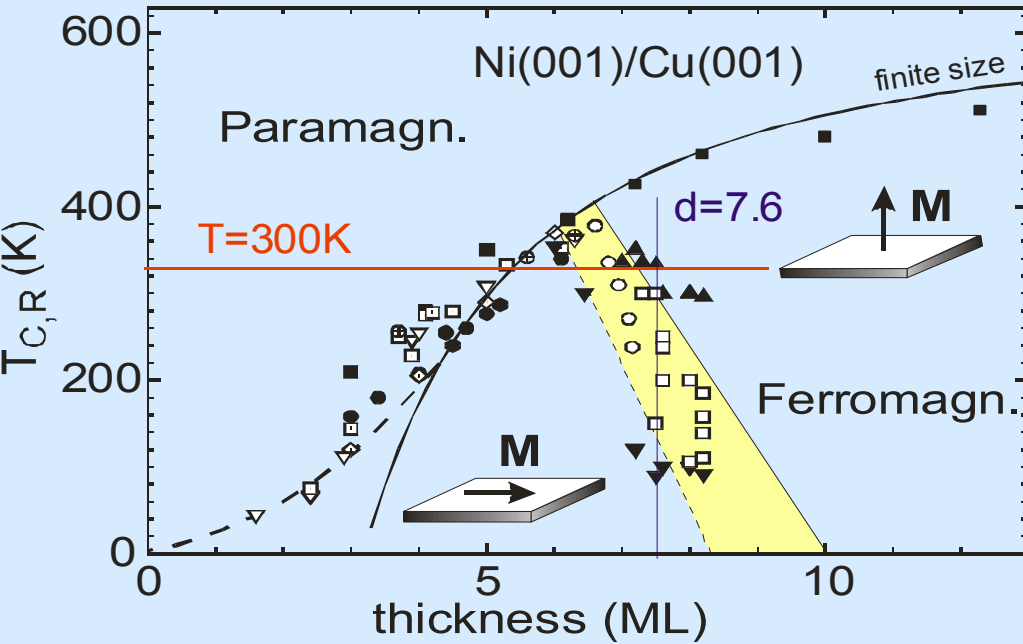


For details see:

K. B. *Handbook of Magnetism and Advanced Magnetic Materials*, Vol. 3
Ed. Kronmüller and Parkin, 2007 John Wiley & Sons, Ltd.



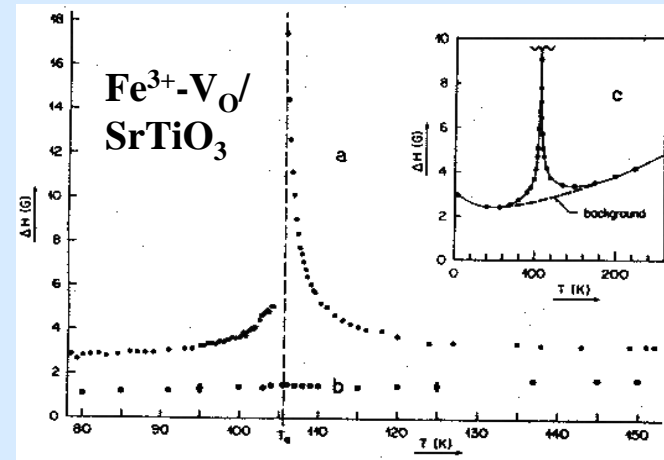
For thin films the Curie temperature can be manipulated



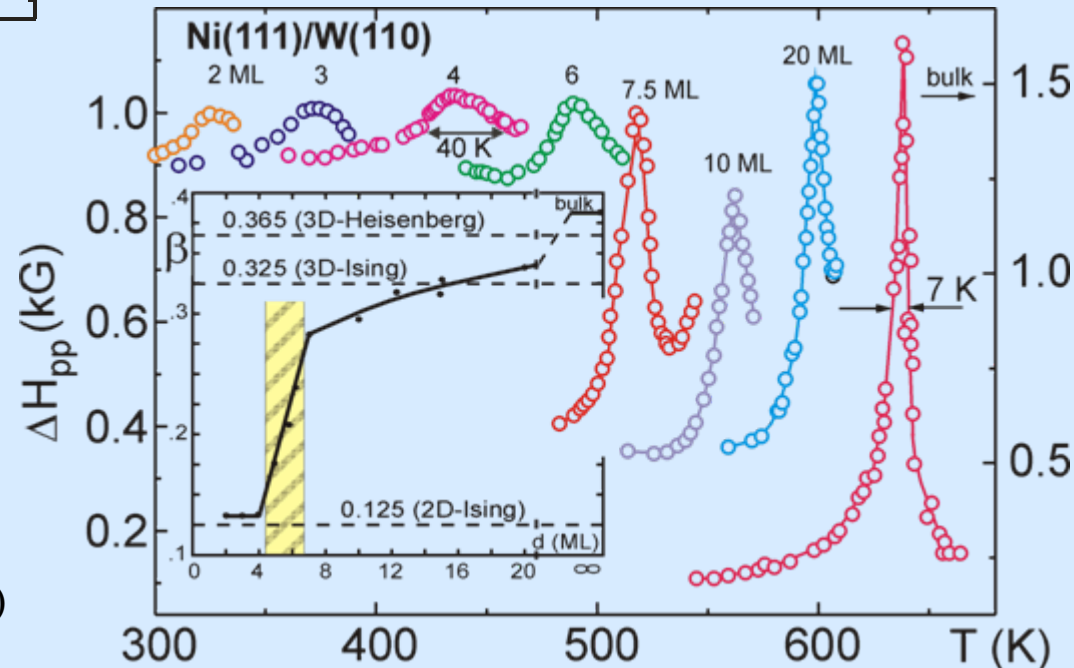
P. Pouloupoulos and K. B.
 J. Phys.: Condens. Matter **11**, 9495 (1999)

$$\frac{T_C(\infty) - T_C(d)}{T_C(\infty)} = cd^{-1/\nu}$$

Yi Li, K. B., PRL **68**, 1208 (1992)

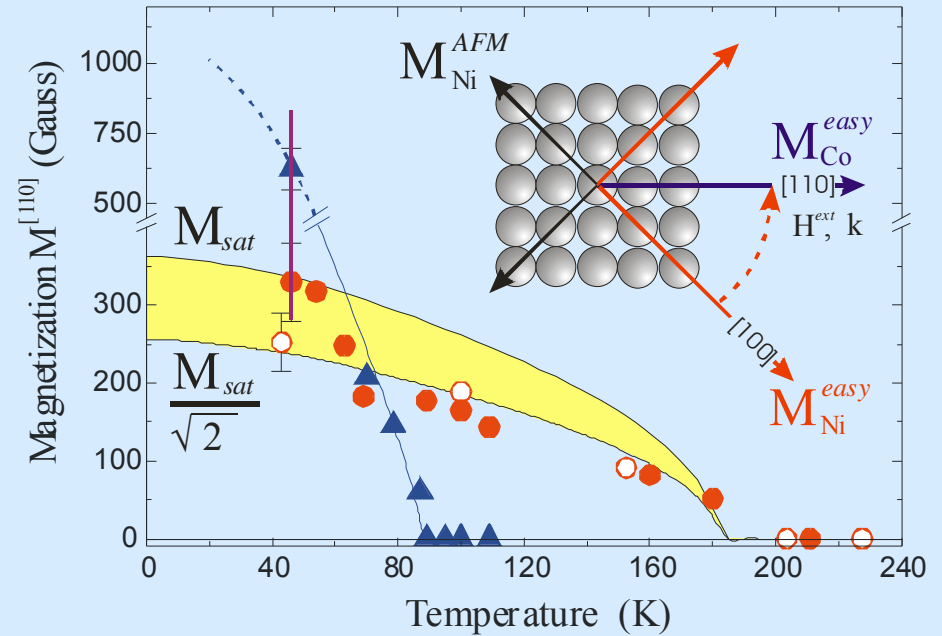
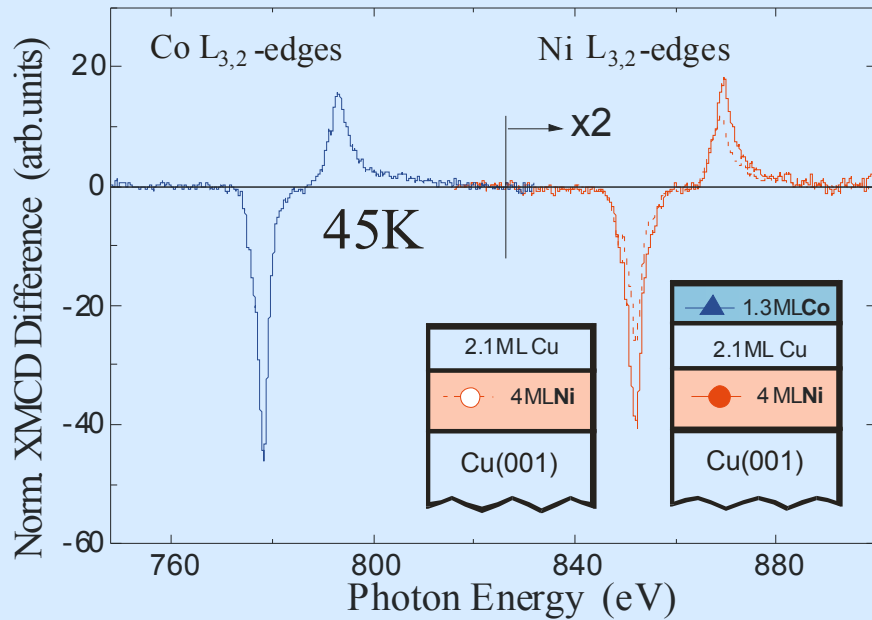


Th.v. Waldkirch, K.A. Müller, W. Berlinger, PRB (1973)



Crossover of $M_{Co}(T)$ and $M_{Ni}(T)$

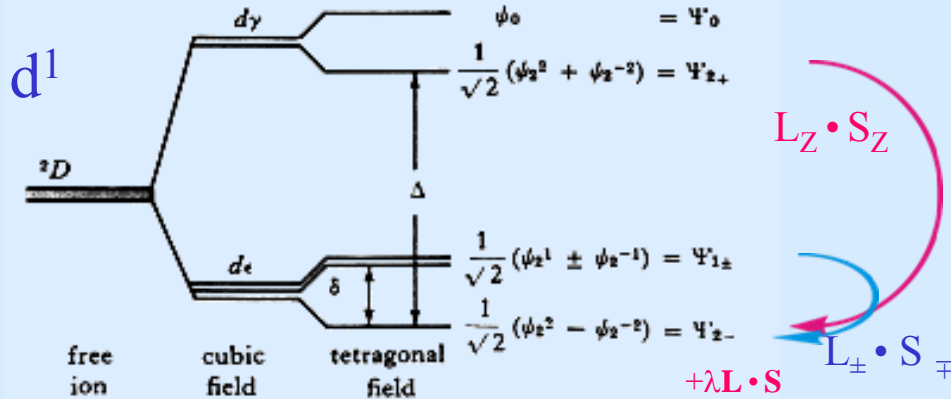
measured with X-ray magnetic circular dichroism



Two order parameter of T_C^{Ni} and T_C^{Co}
 A further reduction in symmetry happens at T_C^{low}

Orbital magnetism in second order perturbation theory

$$\mathcal{H}' = \mu_B \mathbf{H} \cdot \mathbf{L} + \lambda \mathbf{L} \cdot \mathbf{S}$$



Splitting of the 2D term by a tetragonally distorted cubic field.

$$\psi_{2-} \equiv (2)^{-1/2} \{ |2\rangle - |-2\rangle \} \equiv |2-\rangle$$

The orbital moment is quenched in cubic symmetry

$$\langle 2- | L_z | 2- \rangle = 0,$$

but not for tetragonal symmetry

$$\mathcal{H} = \sum_{i,j=1}^3 [\overbrace{\beta g_e(\delta_{ij} - 2\lambda\Lambda_{ij})}^{g_{\text{exp}}}] S_i H_j - \underbrace{\lambda^2 \Lambda_{ij}}_{B_2^0 \rightarrow K_2^0} S_i S_j + \text{diamagnetic terms in } H_i H_j \quad (3-23)$$

where Λ_{ij} is defined in relation to states ($n > 0$) as

$$\Lambda_{ij} = \sum_{n \neq 0} \frac{\langle 0 | L_i | n \rangle \langle n | L_j | 0 \rangle}{E_n - E_0} \quad (3-24)$$

$\langle 0 | \mu_B \mathbf{H} \cdot \mathbf{L} | n \rangle \quad \langle n | \lambda \mathbf{L} \cdot \mathbf{S} | 0 \rangle \quad \langle 0 | \lambda \mathbf{L} \cdot \mathbf{S} | n \rangle \quad \langle n | \lambda \mathbf{L} \cdot \mathbf{S} | 0 \rangle$

In the principal axis system of a crystal with axial symmetry, the $\underline{\Lambda}$ tensor is diagonal with $\Lambda_{zz} = \Lambda_{\parallel}$ and $\Lambda_{xx} = \Lambda_{yy} = \Lambda_{\perp}$. Under these conditions, \mathcal{H} of (3-23) can be simplified, since

$$S_x^2 + S_y^2 = S(S+1) - S_z^2$$

to give

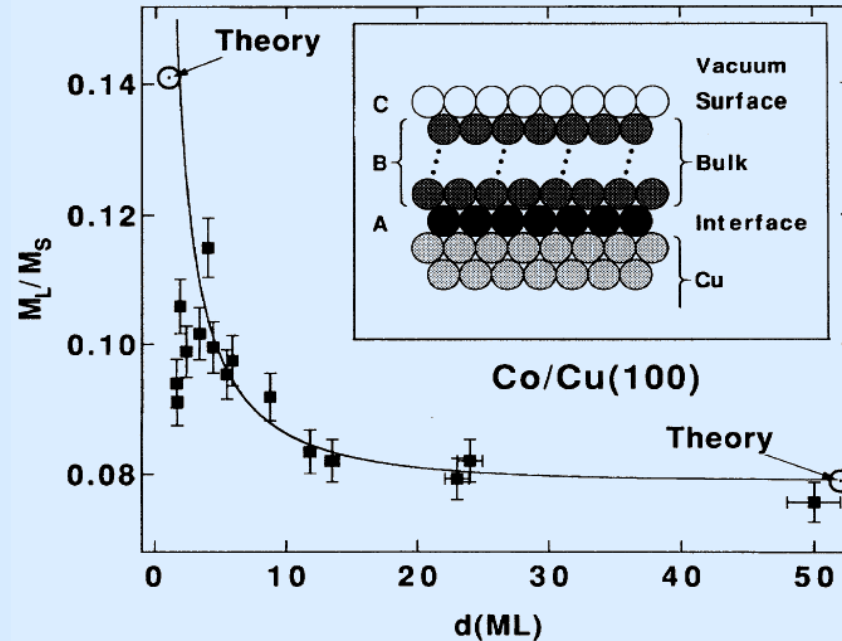
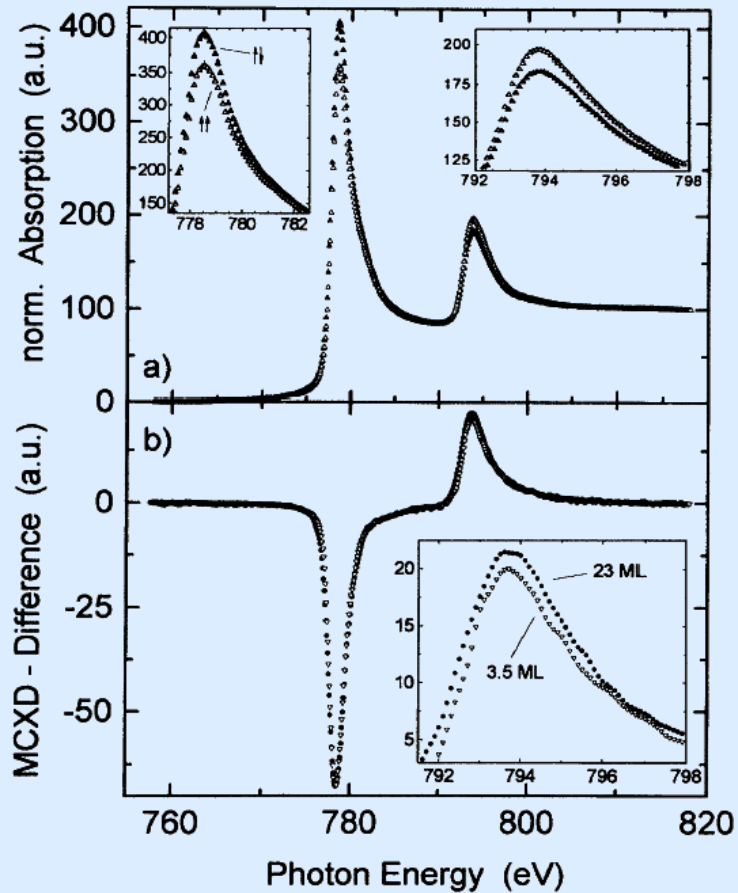
$$\mathcal{H} = g_{\parallel} \beta H_z S_z + g_{\perp} \beta (H_x S_x + H_y S_y) + D [S_z^2 - \frac{1}{3} S(S+1)] \quad (3-25)$$

where

$$\begin{aligned} g_{\parallel} &= g_e (1 - \lambda \Lambda_{\parallel}) \\ g_{\perp} &= g_e (1 - \lambda \Lambda_{\perp}) \\ D &= \lambda^2 (\Lambda_{\perp} - \Lambda_{\parallel}) \end{aligned} \quad (3-26)$$

GE. Pake, p.66

Enhancement of Orbital Magnetism at Surfaces: Co on Cu(100)

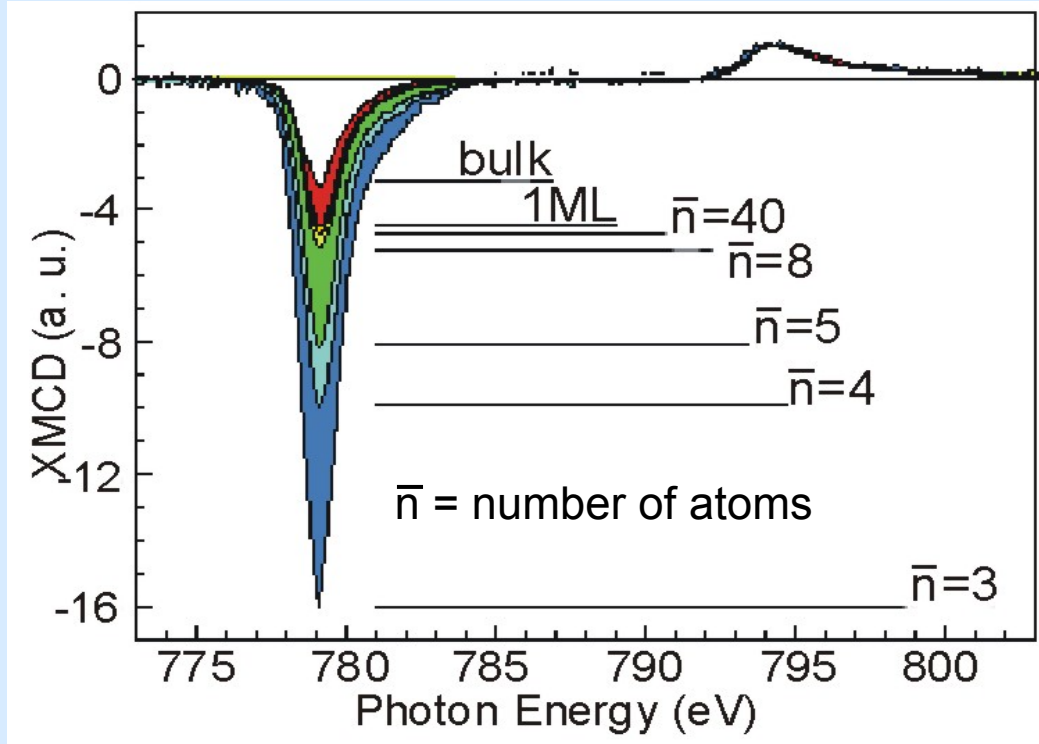


$$\left(\frac{M_L}{M_S} \right)_{\text{exp}} = \frac{Ae^{-D(d-1)/\lambda} + B\sum_{n=3}^d e^{-D(n-2)/\lambda} + C}{\sum_{n=0}^{d-1} e^{-nD/\lambda}}$$

M. Tischer et al., Phys. Rev. Lett. **75**, 1602 (1995)

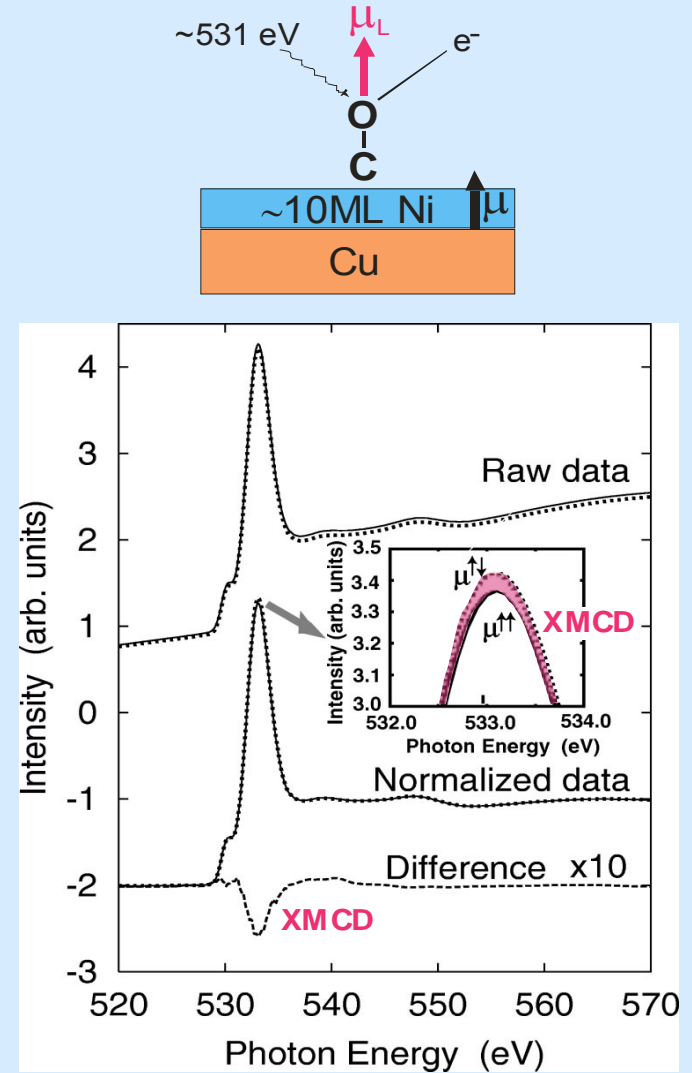
Giant Magn. Anisotropy of Single Co Atoms and Nanoparticles

P. Gambardella et al., Science **300**, 1130 (2003)



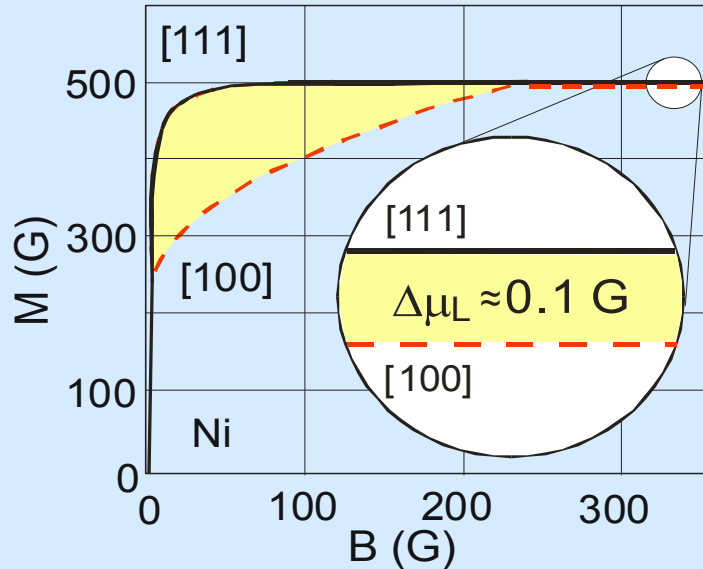
Induced magnetism in molecules

T. Yokoyama et al., PRB **62**, 14191 (2000)



Magnetic Anisotropy Energy (MAE) and anisotropic μ_L

1. Magnetic anisotropy energy = f(T)
2. Anisotropic magnetic moment \neq f(T)



$$MAE = \int \mathbf{M} \cdot d\mathbf{B} \approx \frac{1}{2} \Delta M \cdot \Delta B \approx \frac{1}{2} 200 \cdot 200 \text{ G}^2$$

$$MAE \approx 2 \cdot 10^4 \text{ erg} / \text{cm}^3 \approx 0.2 \text{ } \mu\text{eV} / \text{atom}$$

$\approx 1 \mu\text{eV}/\text{atom}$ is very small compared to
 $\approx 10 \text{ eV}/\text{atom}$ total energy **but all important**

$$g_{\parallel} - g_{\perp} = g_e \lambda (\Lambda_{\perp} - \Lambda_{\parallel})$$

anisotropic $\mu_L \leftrightarrow$ MAE

$$D = \frac{\lambda}{g_e} \Delta g$$



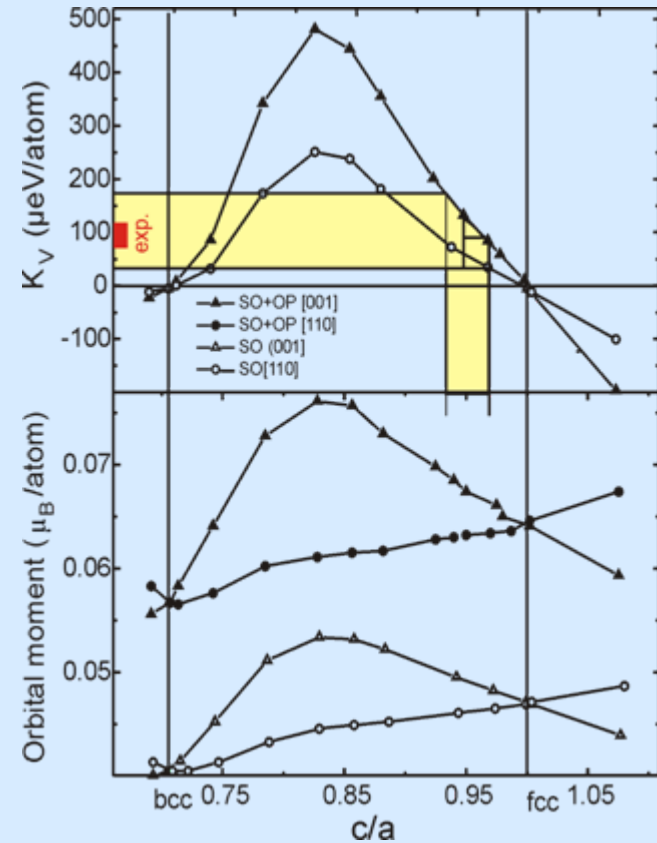
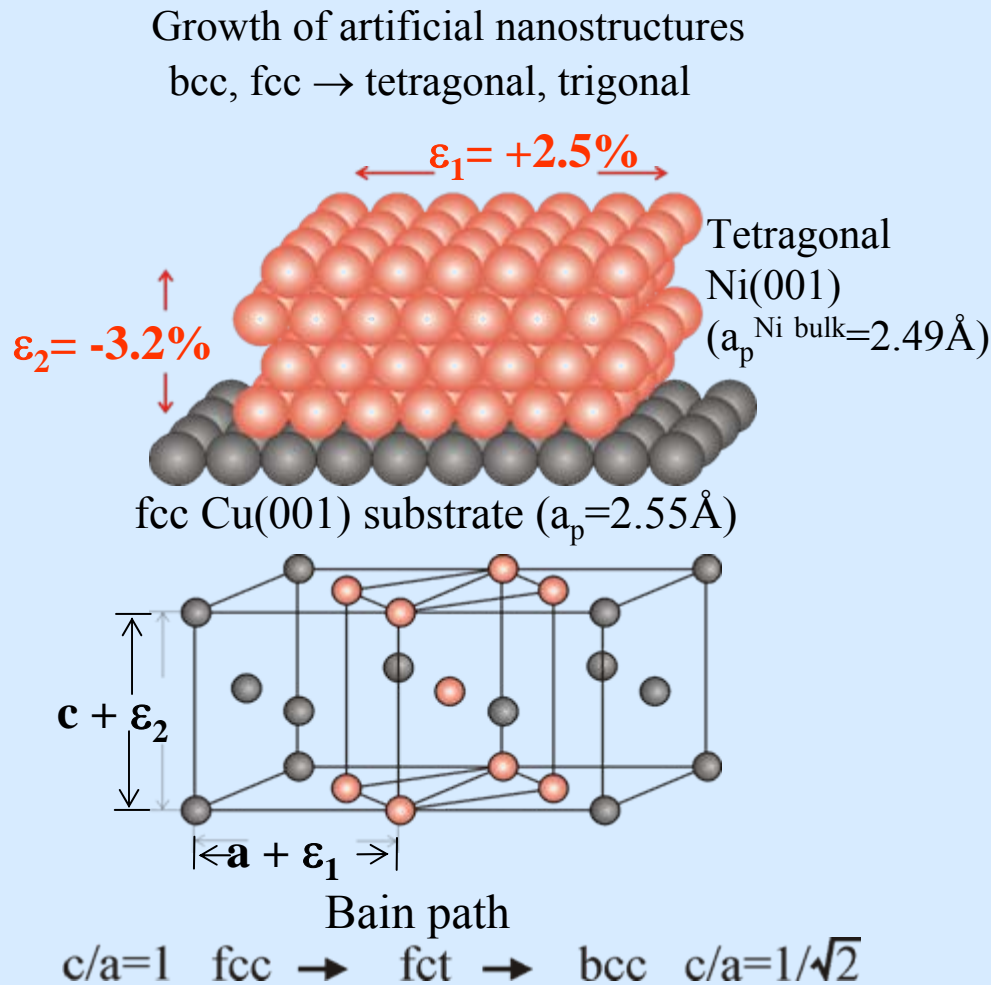
$$MAE \propto \frac{\xi_{LS}}{4\mu_B} \Delta\mu_L \quad \text{Bruno ('89)}$$

Characteristic energies of metallic ferromagnets

binding energy	1 - 10 eV/atom
exchange energy	10 - 10 ³ meV/atom
cubic MAE (Ni)	0.2 $\mu\text{eV}/\text{atom}$
uniaxial MAE (Co)	70 $\mu\text{eV}/\text{atom}$

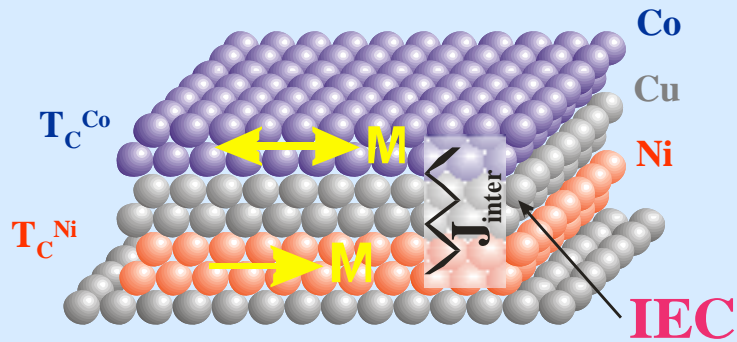
K. Baberschke, Lecture Notes in Physics, Springer **580**, 27 (2001)

There are only 2 origins for MAE: 1) dipol-dipol interaction $\sim (\underline{\mu}_1 \cdot \underline{r})(\underline{\mu}_2 \cdot \underline{r}) / r^3$ and
 2) spin-orbit coupling $\lambda \mathbf{L} \cdot \mathbf{S}$ (intrinsic K or ΔE_{band})



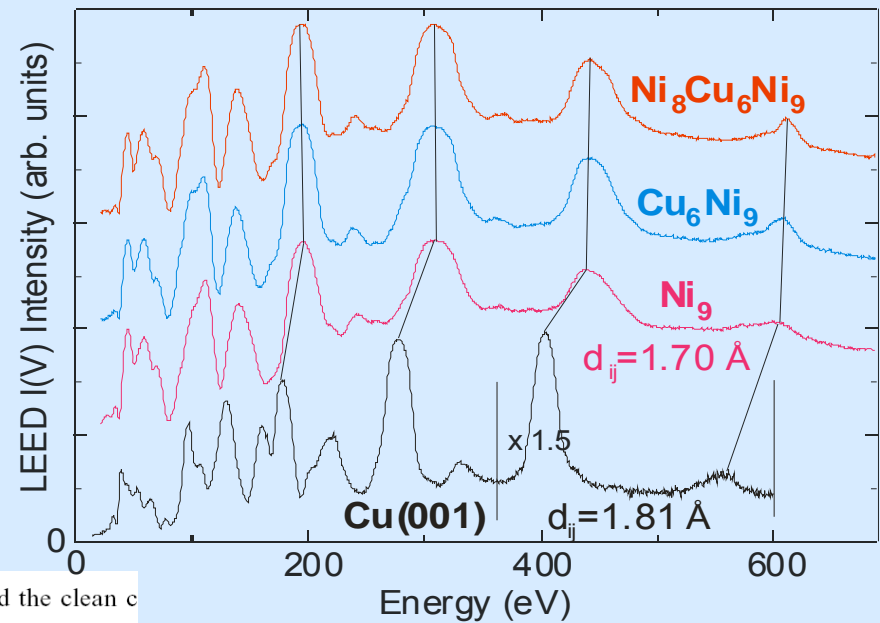
Structural changes by $\approx 0.05 \text{ \AA}$ increase MAE by 2-3 orders of magnitude ($\sim 0.2 \rightarrow 100 \mu\text{eV/atom}$)

O. Hjortstam, K. B. et al. PRB **55**, 15026 ('97)
 R. Wu et al. JMMM **170**, 103 ('97)



W. Platow et al. PRB **59**, 12641 (1999)

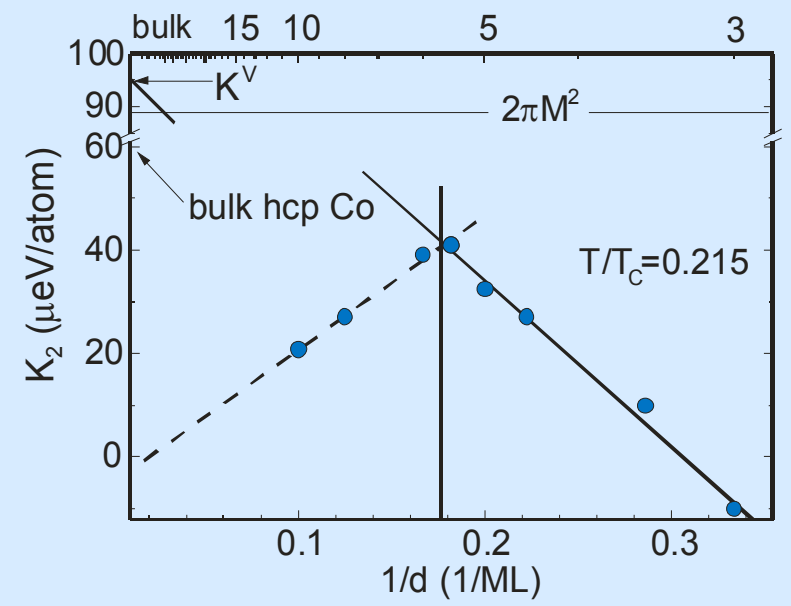
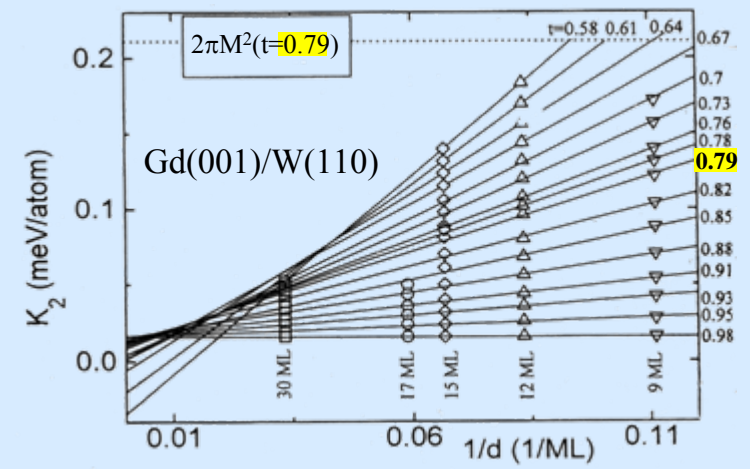
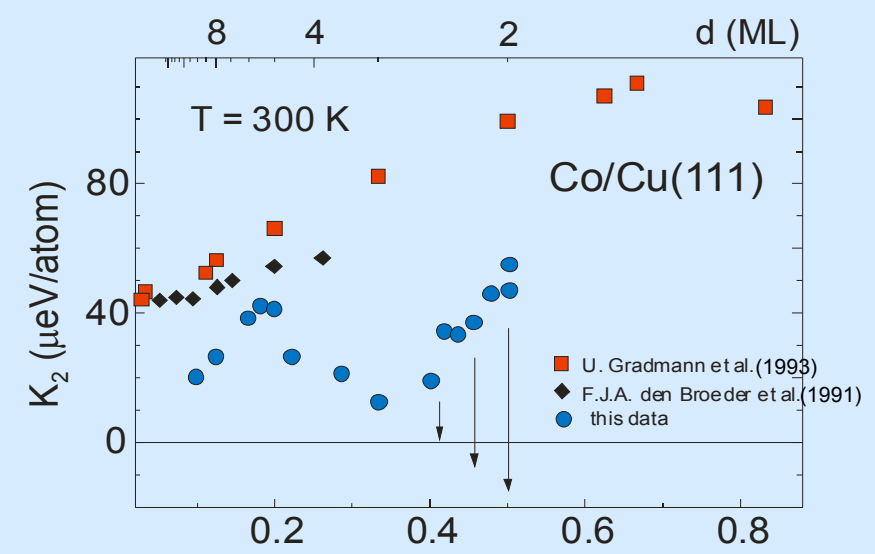
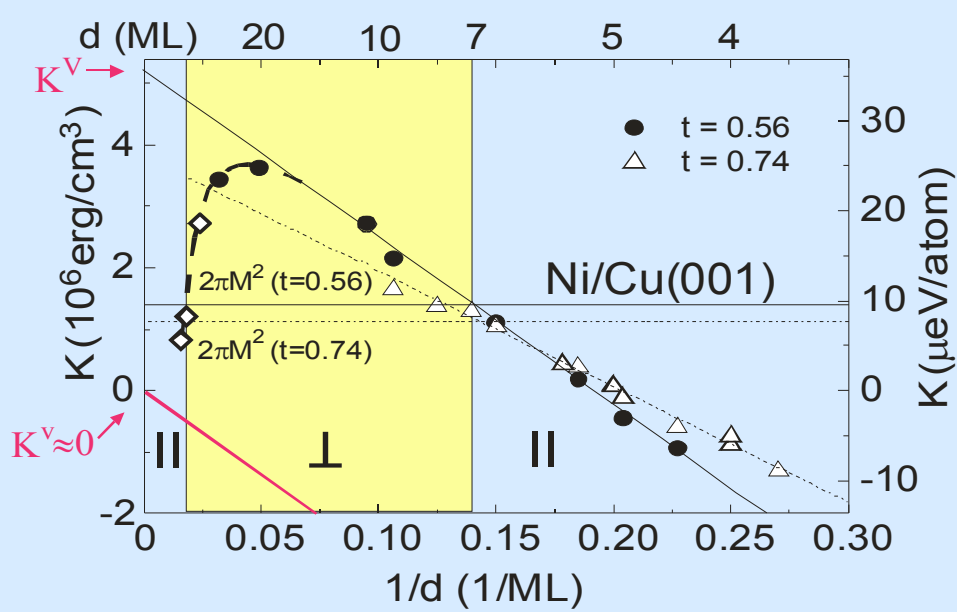
full trilayer grows in fct structure



R. Nünthel, PhD Thesis FUB 2003

TABLE I. Best-fit structural data for the nickel films of different thickness and the clean c

Parameter	0 ML	1 ML	2 ML	3 ML	4 ML	5 ML
d_{12} (Å)	$1.755^{+0.011}_{-0.007}$	$1.720^{+0.014}_{-0.018}$	$1.715^{+0.015}_{-0.015}$	$1.725^{+0.022}_{-0.016}$	$1.705^{+0.015}_{-0.011}$	$1.675^{+0.012}_{-0.014}$
d_{23} (Å)	$1.805^{+0.006}_{-0.011}$	$1.770^{+0.012}_{-0.014}$	$1.720^{+0.011}_{-0.011}$	$1.710^{+0.012}_{-0.009}$	$1.705^{+0.011}_{-0.013}$	$1.710^{+0.010}_{-0.014}$
d_{34} (Å)	1.800 ± 0.010	$1.795^{+0.012}_{-0.012}$	$1.775^{+0.014}_{-0.021}$	$1.715^{+0.024}_{-0.017}$	$1.71^{+0.014}_{-0.016}$	$1.700^{+0.014}_{-0.014}$
d_{45} (Å)	1.790 ± 0.013	$1.800^{+0.017}_{-0.014}$	$1.790^{+0.028}_{-0.015}$	$1.760^{+0.028}_{-0.017}$	$1.72^{+0.024}_{-0.017}$	$1.715^{+0.014}_{-0.014}$
d_{56} (Å)	$1.800^{+0.010}_{-0.009}$	$1.790^{+0.020}_{-0.017}$	$1.800^{+0.028}_{-0.028}$	$1.790^{+0.021}_{-0.022}$	$1.76^{+0.033}_{-0.022}$	$1.730^{+0.018}_{-0.025}$
d_b (Å)	1.790	1.79	1.79	1.79	1.77	1.70
ΔE (eV)	2270	2070	2220	2090	1450	2120
R_p	0.085	0.093	0.170	0.138	0.096	0.111



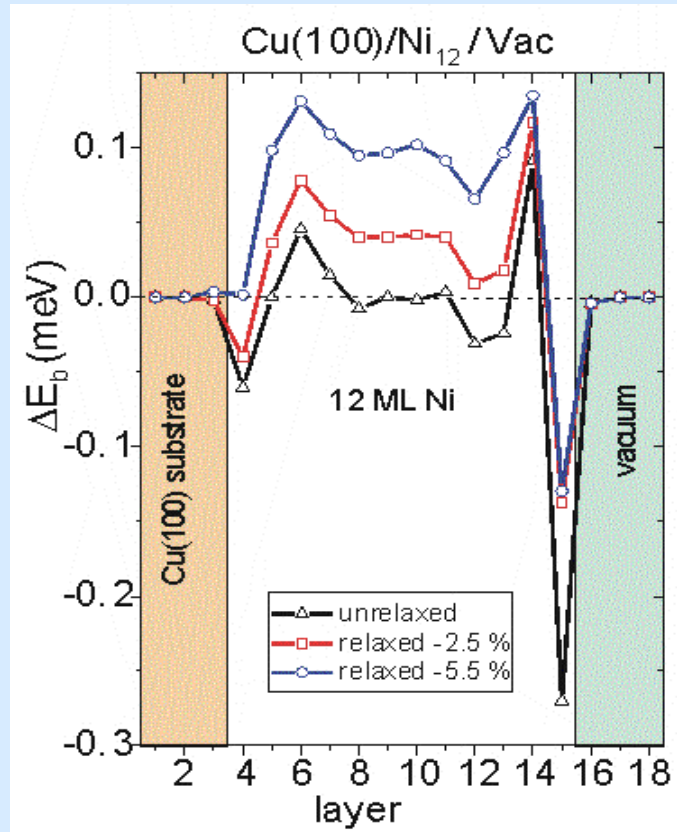
G. André et al., Surface Science **326**, 275 (1995)
 K. B. and M. Farle, J. Appl. Phys. **81**, 5038 (1997)

M. Farle et al., Surf. Sci. **439**, 146 (1999)

In a proper analysis, taking $T/T_C(d)$ in consideration, we always find a linear $K=K_V+2K_S/d$ dependence.
 A departure from this “Néel argument” indicates changes in the x-tal structure

SP-KKR calculation for rigid fcc and relaxed fct structures

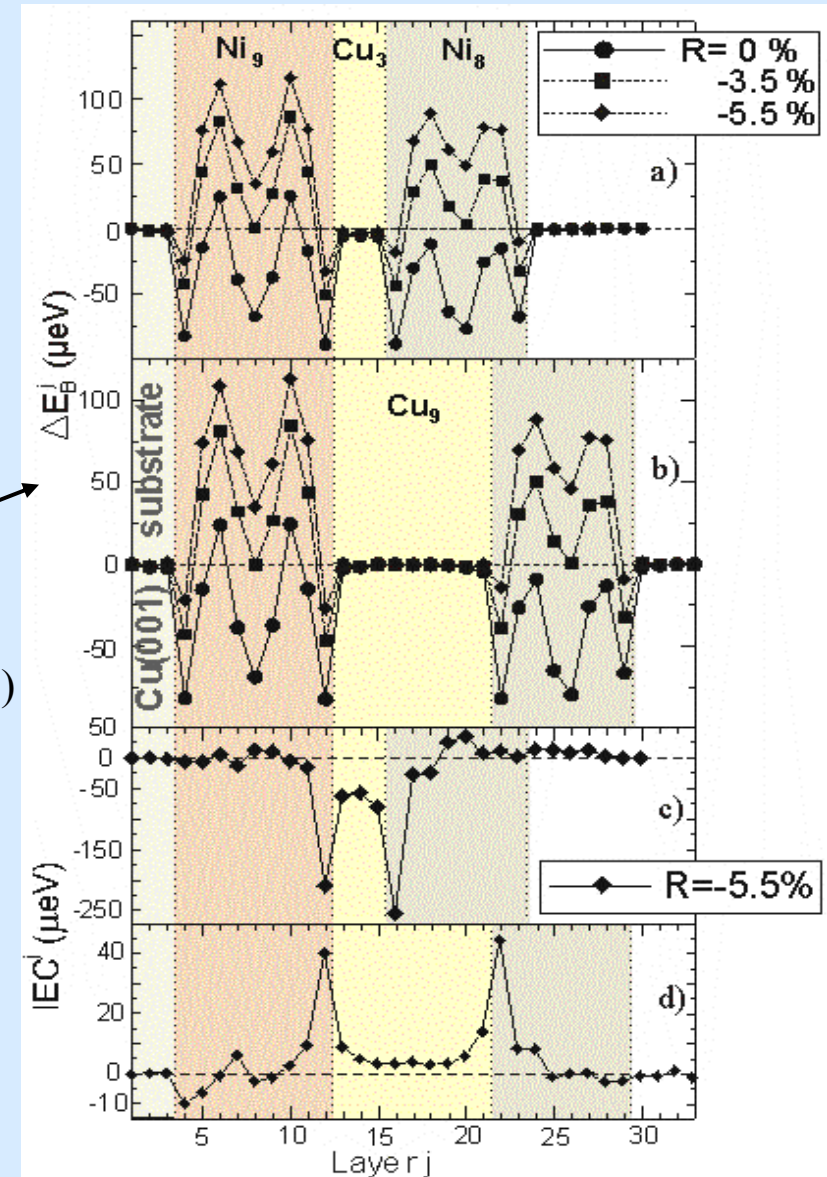
layer resolved $\Delta E_b = \Sigma K_i$ at T=0



C. Uiberacker et al.,
PRL **82**, 1289 (1999)

R. Hammerling et al.,
PRB **68**, 092406 (2003)

The surface and interface MAE are certainly large (L. Néel, 1954) but count only for one layer each. The inner part (volume) of a nanostructure will overcome this, because they count for n-2 layers.

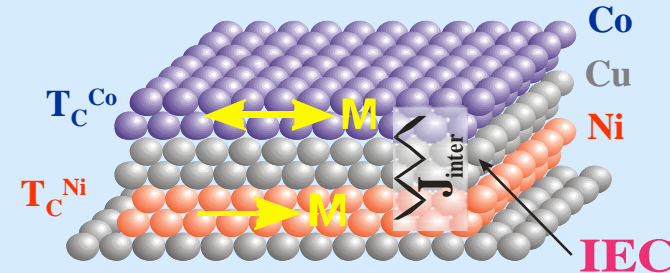


Part I: Fundamentals

- Curie temperature T_C ,
- Orbital- and spin- magnetic moments,
- Magnetic Anisotropy Energy (MAE)

Part II: Spin dynamics in nano-magnets:

- Element specific magnetizations and T_C 's in trilayers.
- Interlayer exchange coupling and its T-dependence.
- Gilbert damping versus magnon-magnon scattering.



A whole variety of experiments on nanoscale magnets are available nowadays. Unfortunately many of the data are analyzed using theoretical *static mean field (MF) model*, e. g. by assuming only magnetostatic interactions of multilayers, static exchange interaction, or static interlayer exchange coupling (IEC), etc. We will show that such a mean field ansatz is insufficient for nanoscale magnetism, 3 cases will be discussed to demonstrate the importance of *higher order spin-spin correlations* in low dimensional magnets.

$$\text{Spin-Spin correlation function } \frac{\partial}{\partial t} \langle\langle S_i^+ S_j^- \rangle\rangle \rightarrow$$

$$S_i^z S_j^+ \approx \underbrace{\langle S_i^z \rangle}_{\leftarrow} S_j^+ - \langle S_i^- S_i^+ \rangle S_j^+ - \langle S_i^- S_j^+ \rangle S_i^+ + \dots$$

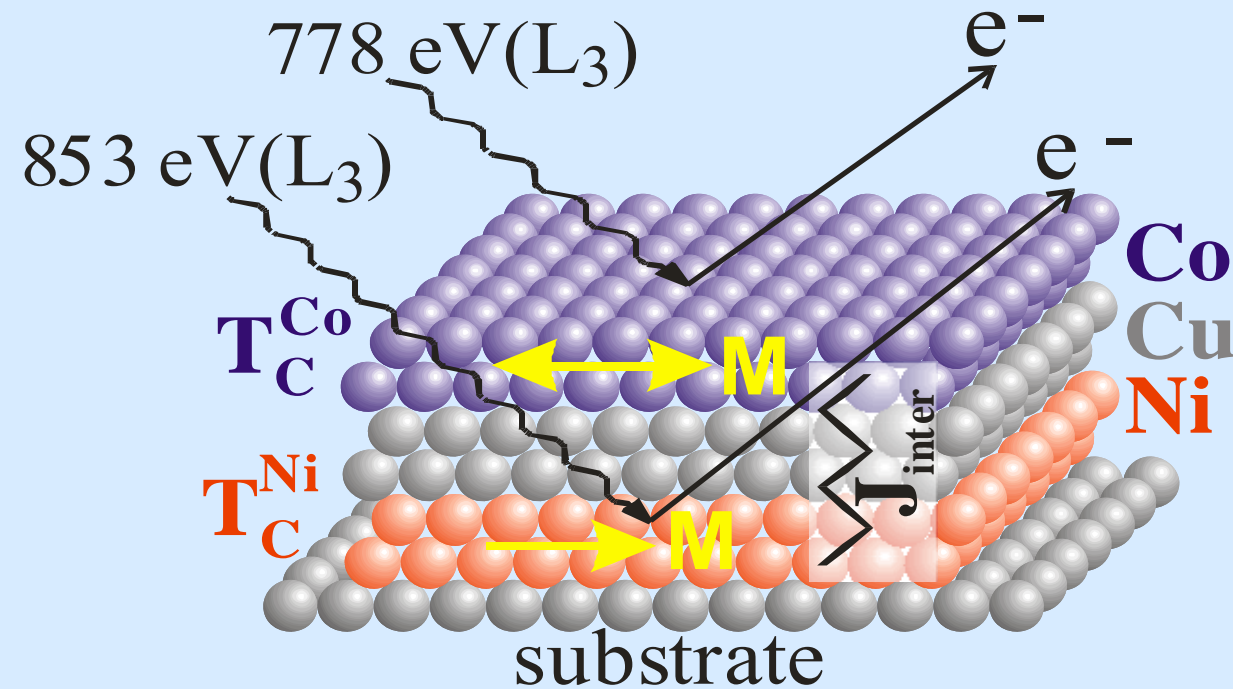
RPA \longrightarrow

The damping of spin motions in ultrathin films: Is the Landau–Lifschitz–Gilbert phenomenology applicable? [☆]

D.L. Mills^{a,*}, Rodrigo Arias^b

Physica B **384**, 147 (2006)

1. Element specific magnetizations and T_C 's in trilayers.

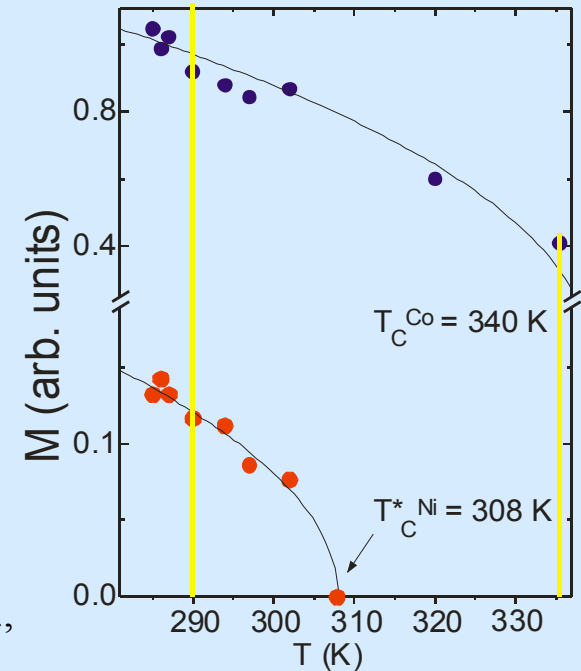
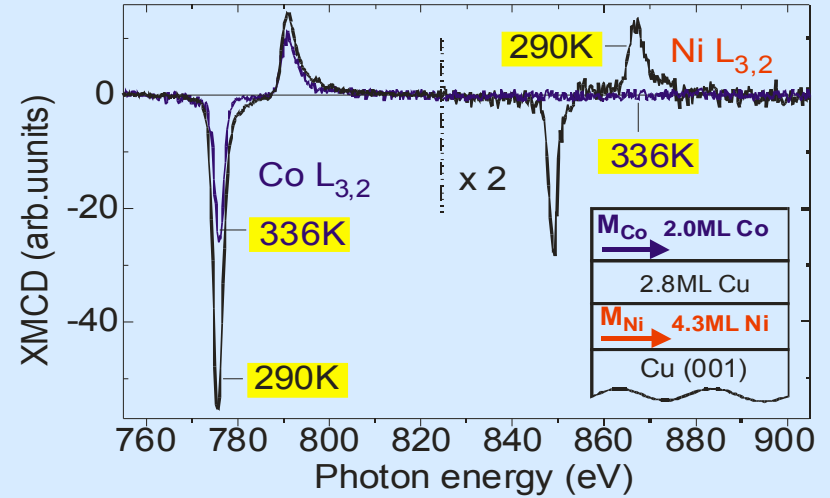
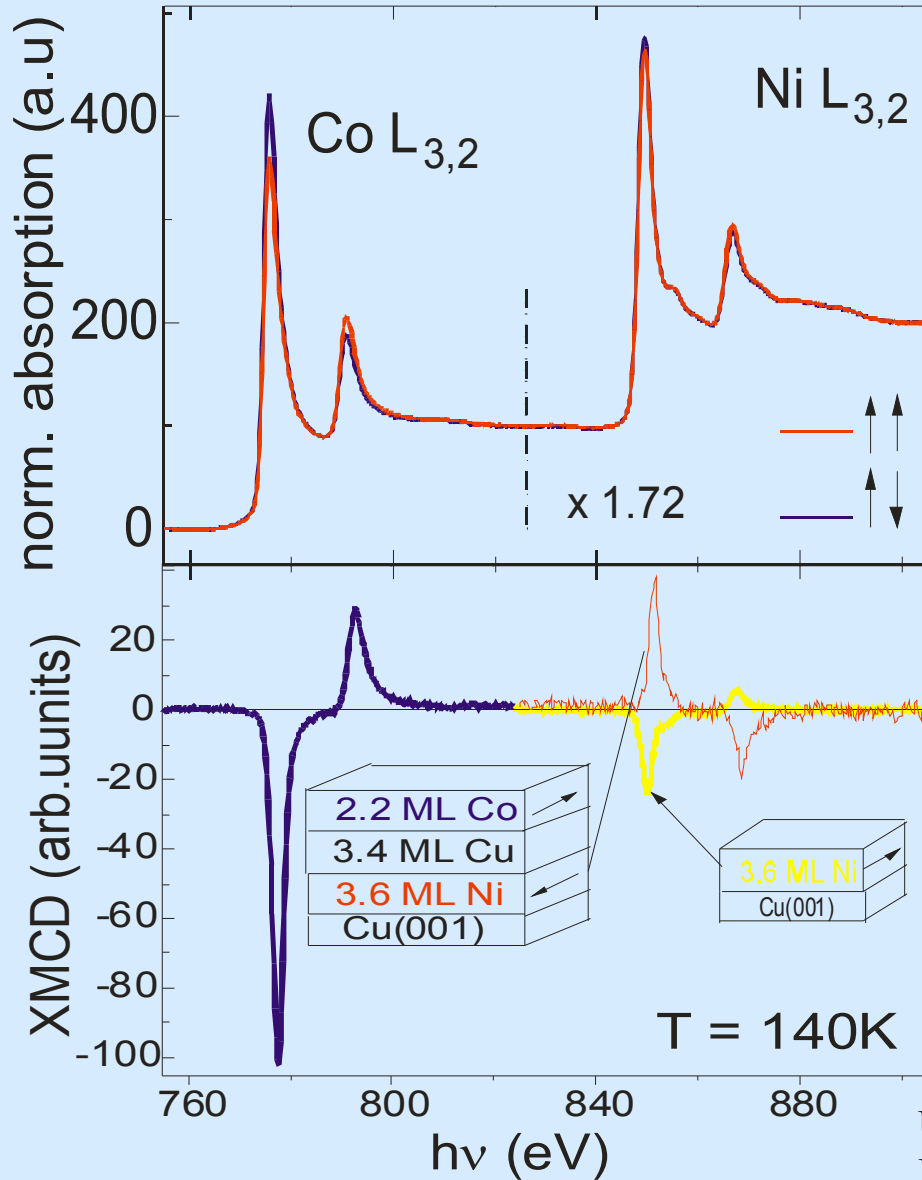


A trilayer is a prototype to study magnetic coupling in multilayers.

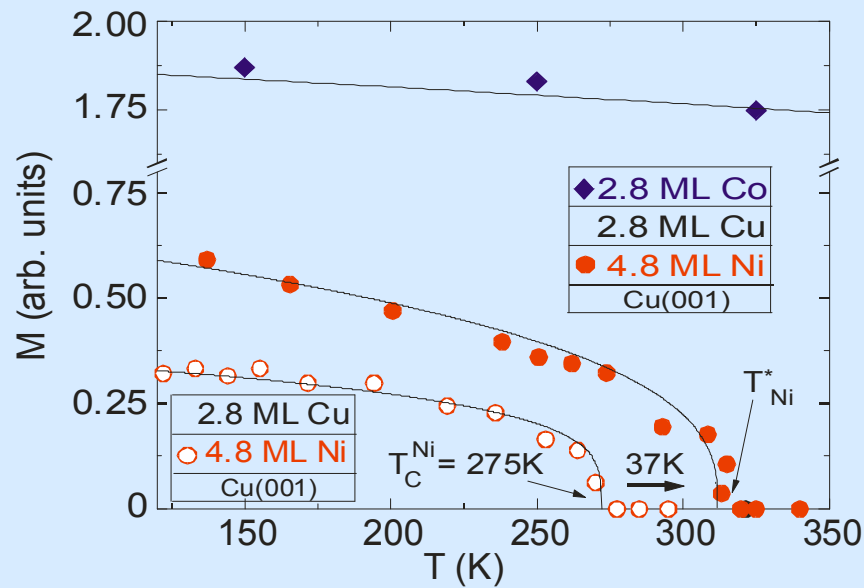
What about element specific Curie-temperatures ?

- Two trivial limits: (i) $d_{Cu} = 0 \Rightarrow$ direct coupling like a Ni-Co alloy
(ii) $d_{Cu} = \text{large} \Rightarrow$ no coupling, like a mixed Ni/Co powder
- BUT** $d_{Cu} \approx 2 \text{ ML} \Rightarrow ?$

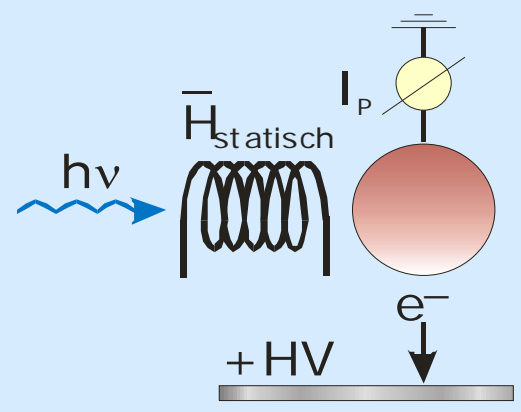
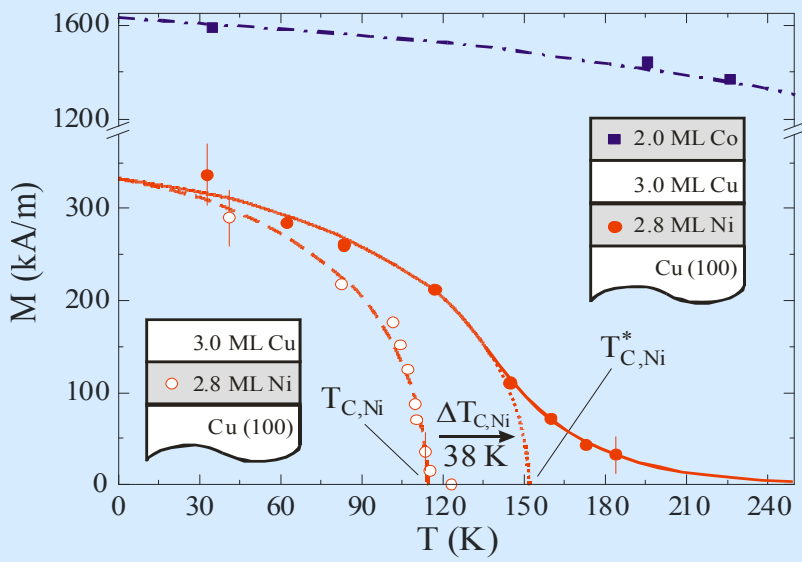
Ferromagnetic trilayers



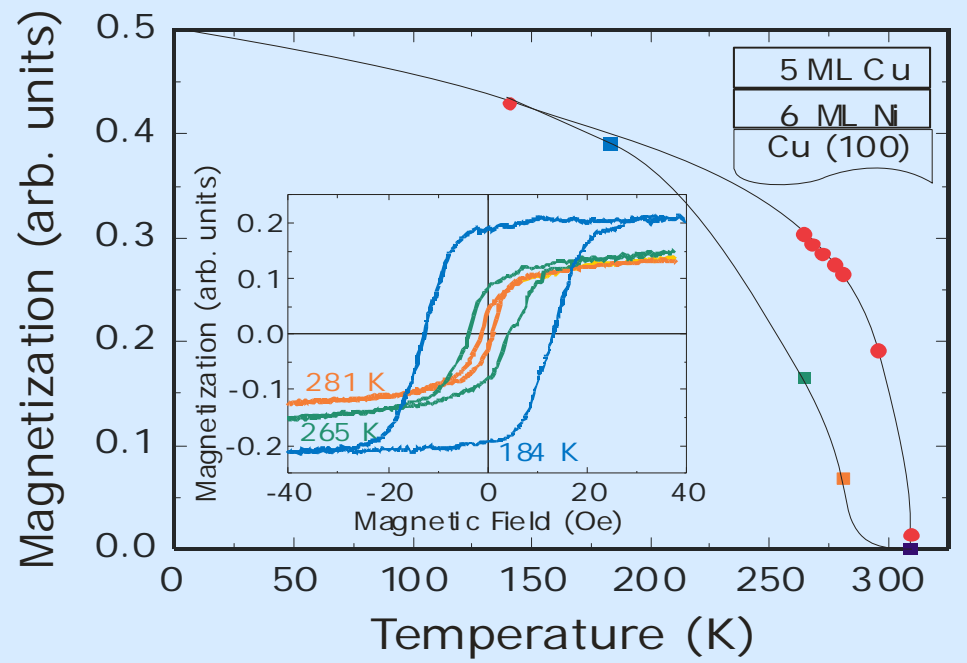
U. Bovensiepen et al.,
PRL **81**, 2368 (1998)



P. Pouloupoulos, K. B., Lecture Notes in Physics **580**, 283 (2001)

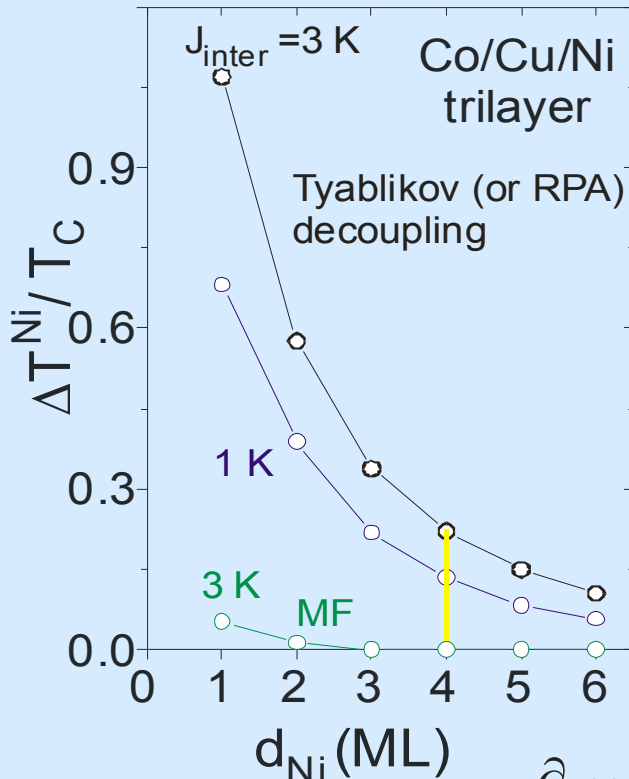


C. Sorg et al.,
 XAFS XII, June 2003
 Physica Scripta
T115, 49 (2005)



Enhanced spin fluctuations in 2D (theory)

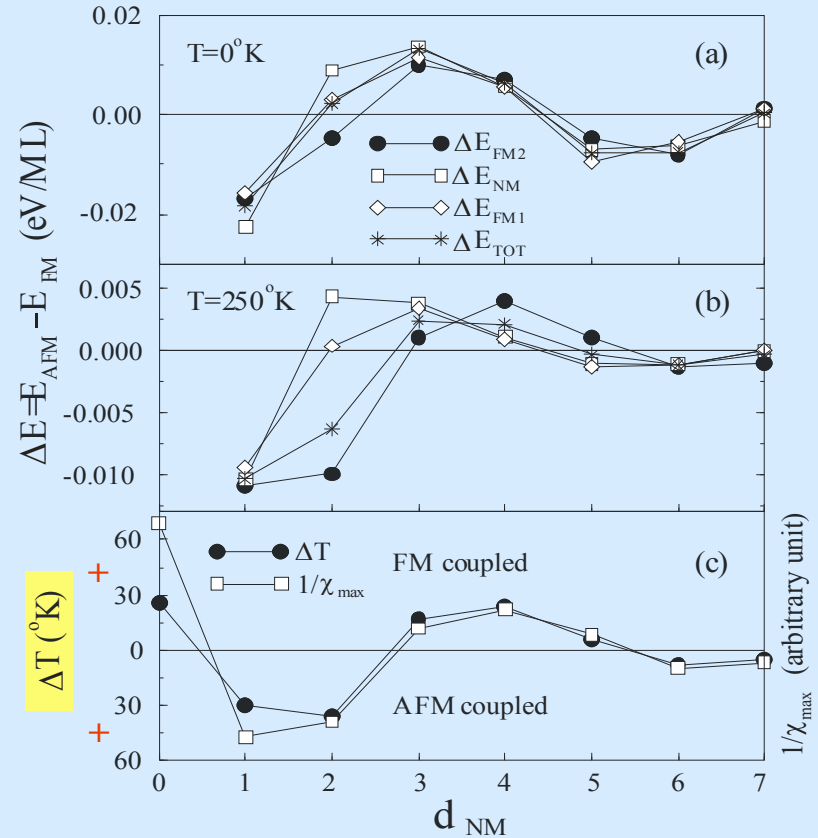
P. Jensen et al. PRB **60**, R14994 (1999)



Spin-Spin correlation function $\frac{\partial}{\partial t} \langle \langle S_i^+ S_j^- \rangle \rangle \rightarrow$
 $S_i^z S_j^+ \approx \langle S_i^z \rangle S_j^+ - \langle S_i^- S_i^+ \rangle S_j^+ - \langle S_i^- S_j^+ \rangle S_i^+ + \dots$
 RPA

$\langle S_i^z \rangle S_j^+$, mean field ansatz (Stoner model) is insufficient to describe spin dynamics at interfaces of nanostructures

J.H. Wu et al. J. Phys.: Condens. Matter **12** (2000) 2847



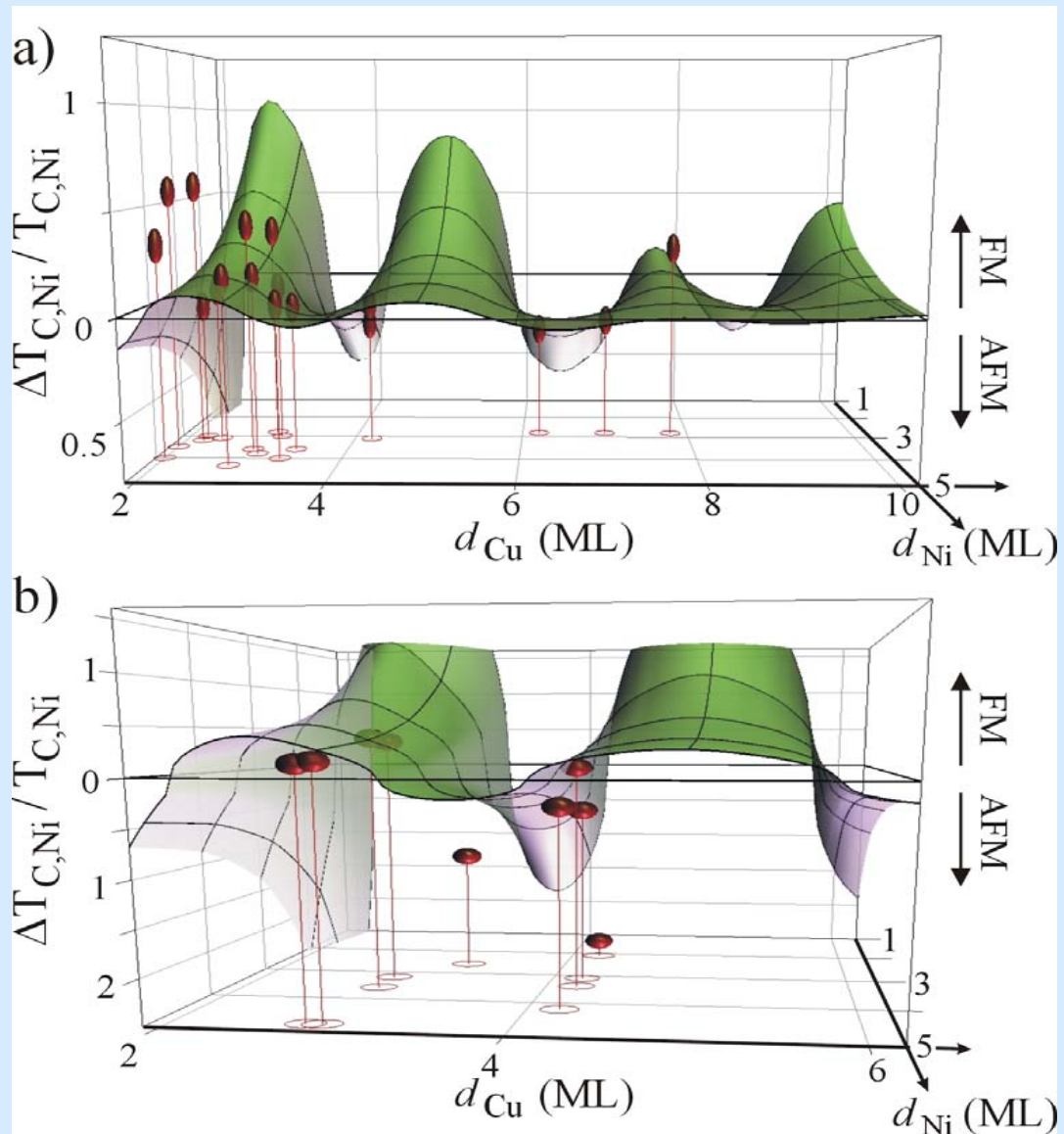
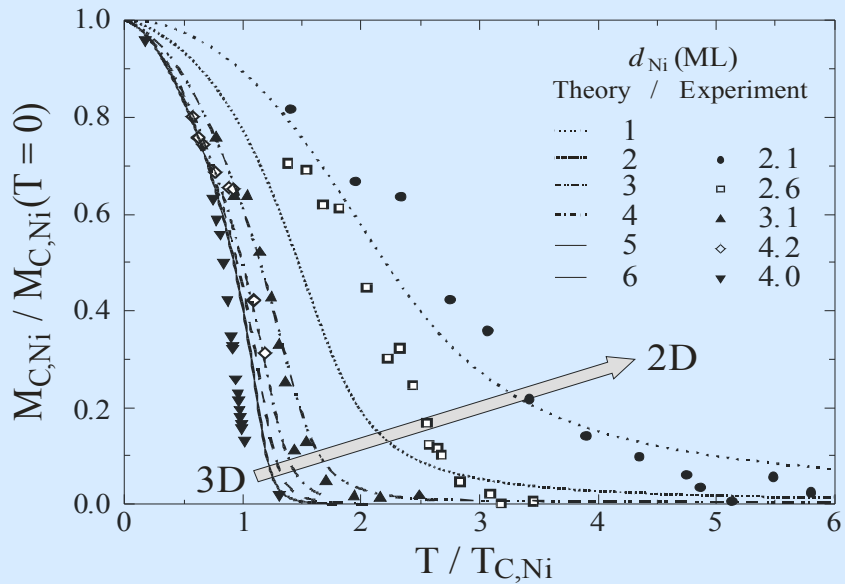
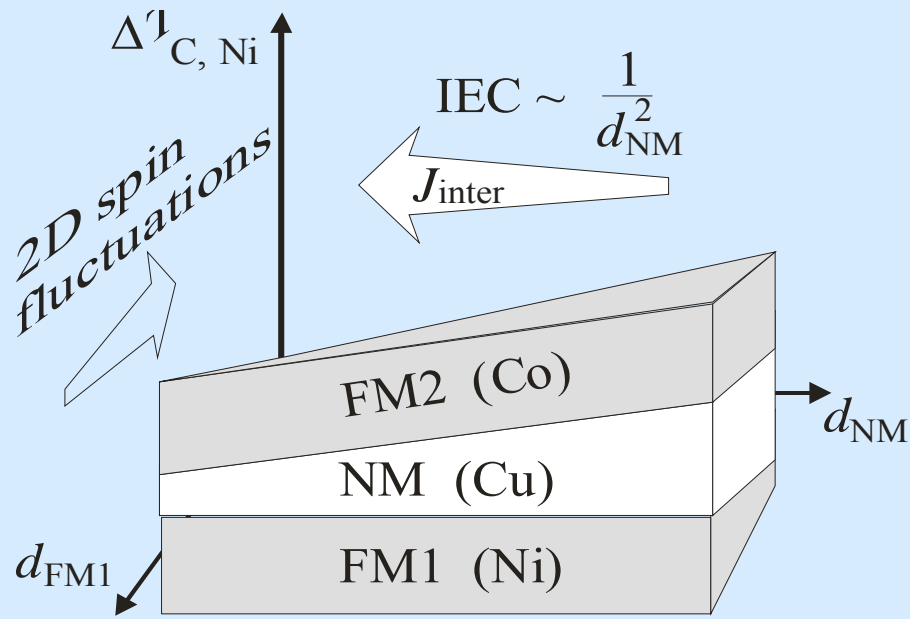
Single band Hubbard model:

Simple Hartree-Fock (Stoner) ansatz is insufficient

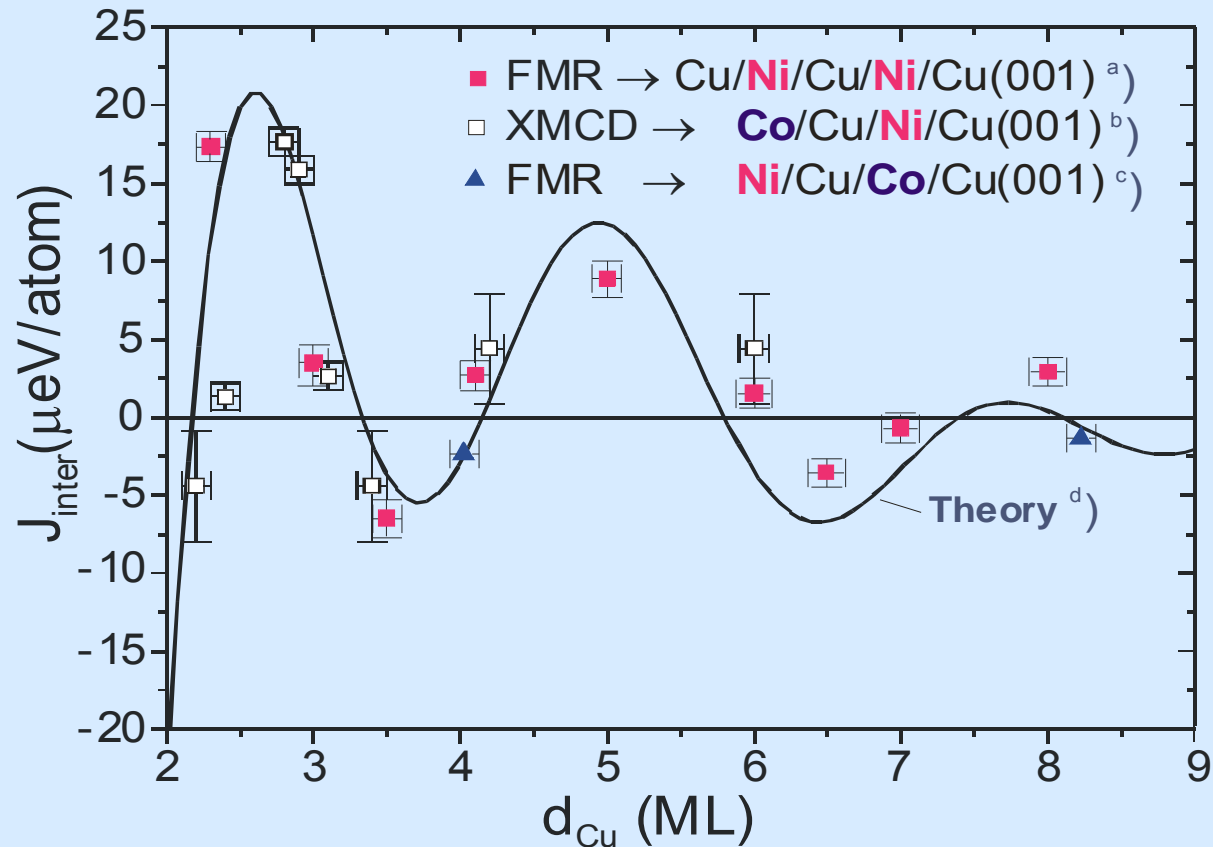
Higher order correlations are needed to explain T_C -shift

Evidence for giant spin fluctuations

[A. Scherz et al. PRB, 73 54447 (2005)]



2. Interlayer exchange coupling and its T-dependence.



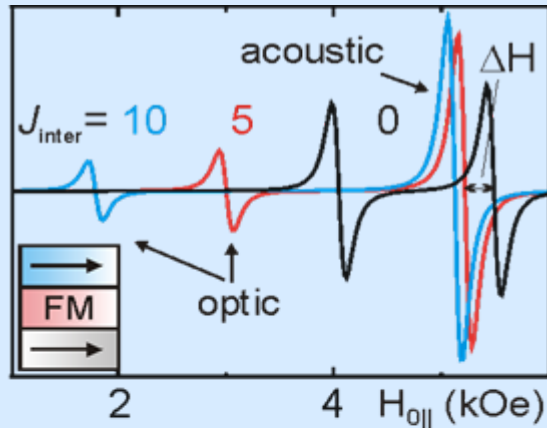
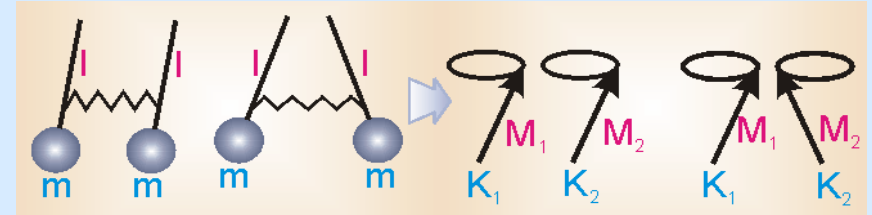
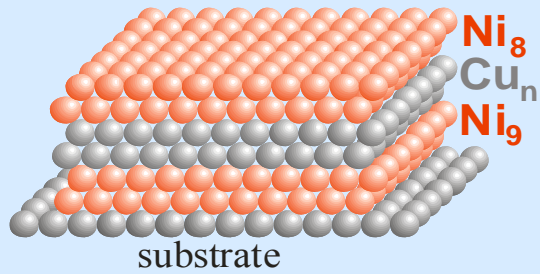
a) J. Lindner, K. B., J. Phys. Condens. Matter **15**, S465 (2003)

b) A. Ney et al., Phys. Rev. B **59**, R3938 (1999)

c) J. Lindner et al., Phys. Rev. B **63**, 094413 (2001)

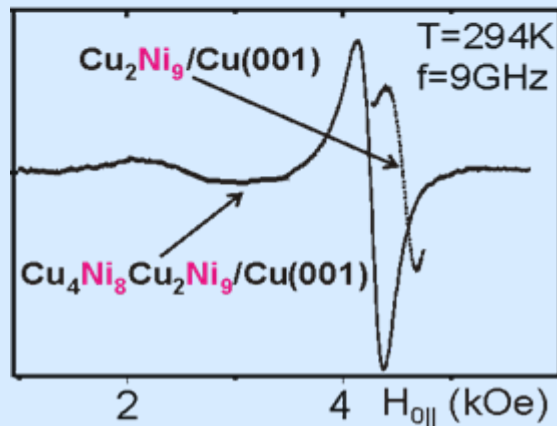
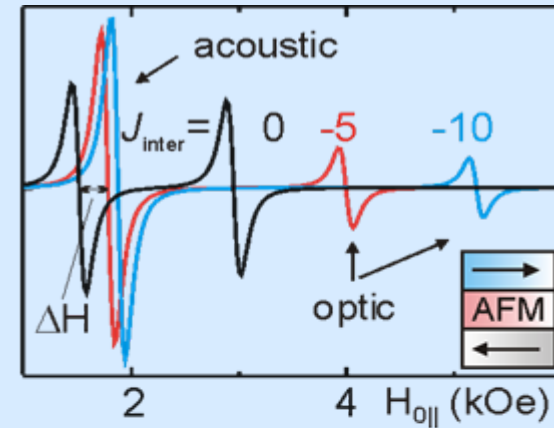
d) P. Bruno, Phys. Rev. B **52**, 441 (1995)

in-situ FMR in coupled films

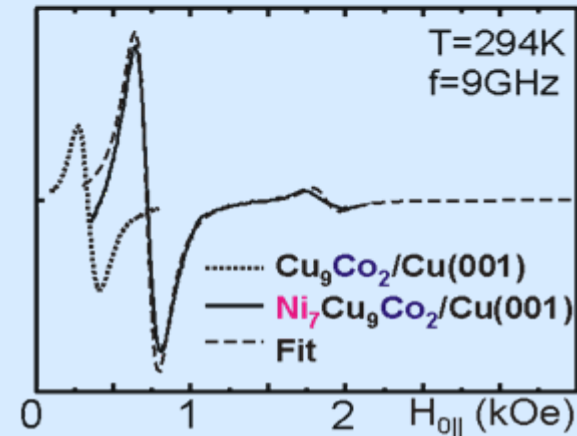


theory

FMR



in-situ
UHV-experiment



J. Lindner, K. B. Topical Rev., J. Phys. Condens. Matter **15**, R193-R232 (2003)

Temperature dependence of $J_{\text{inter}} \Leftrightarrow \Delta$ free energy

P. Bruno, PRB **52**, 411 (1995)

$$J_{\text{inter}} = J_{\text{inter},0} \left[\frac{T/T_0}{\sinh(T/T_0)} \right] \quad T_0 = \hbar v_F / 2\pi k_B d$$

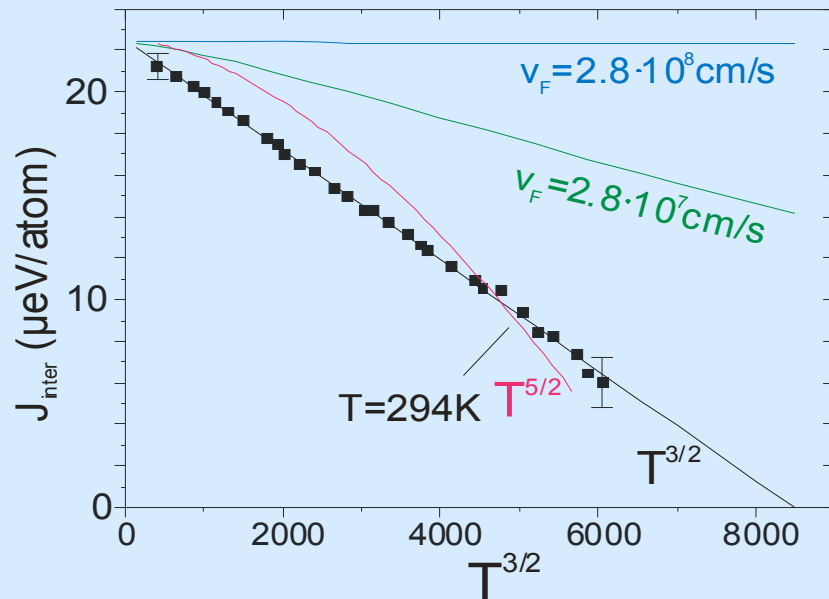
N.S. Almeida et al. PRL **75**, 733 (1995)

$$J_{\text{inter}} = J_{\text{inter},0} [1 - (T/T_C)^{3/2}]$$

Ni₇Cu₉Co₂/Cu(001)

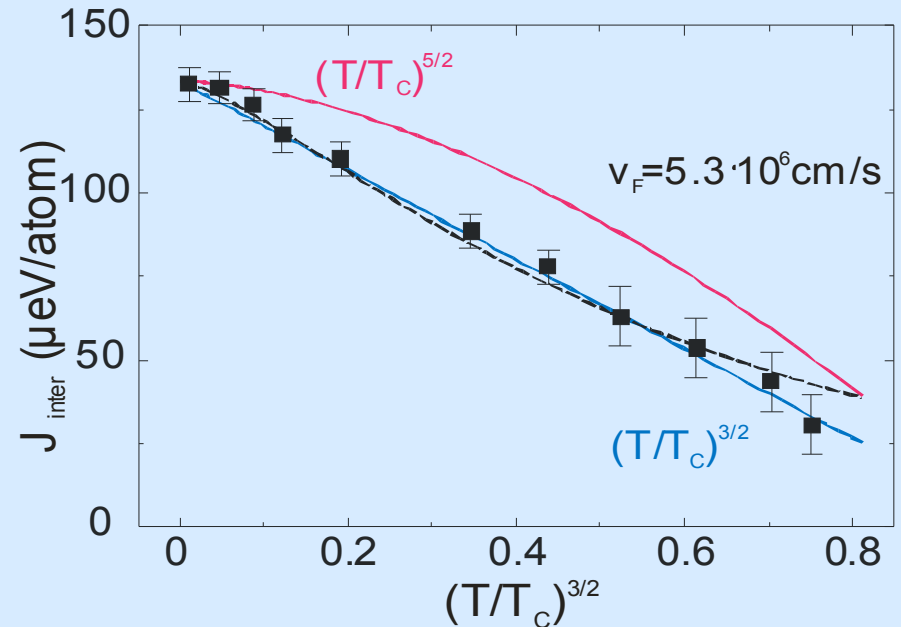
J. Lindner et al.
PRL **88**, 167206 (2002)

T=55K - 332K



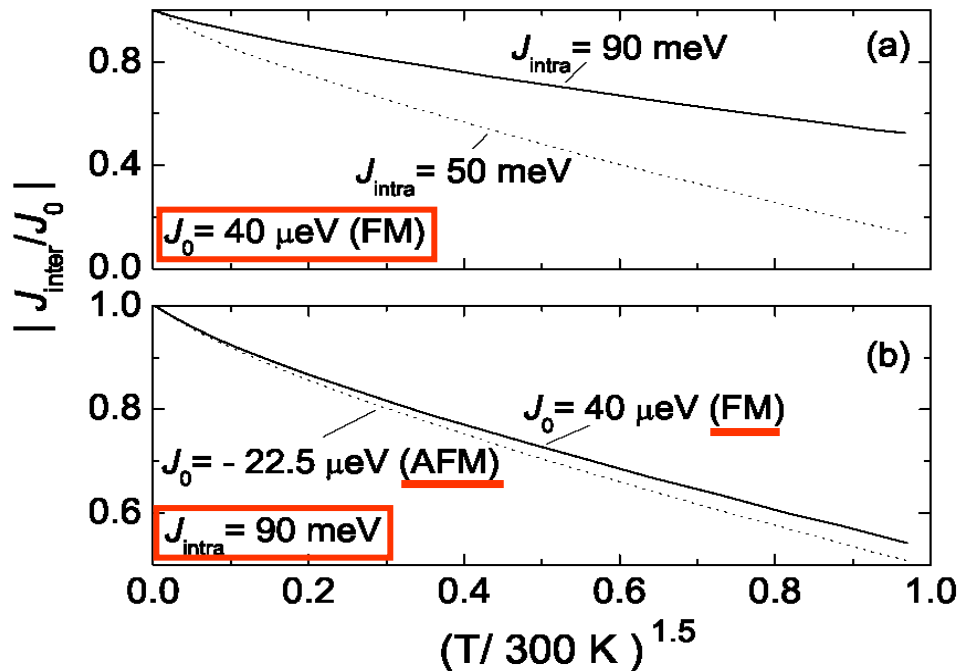
(Fe₂V₅)₅₀

T=15K - 252K, T_C=305K



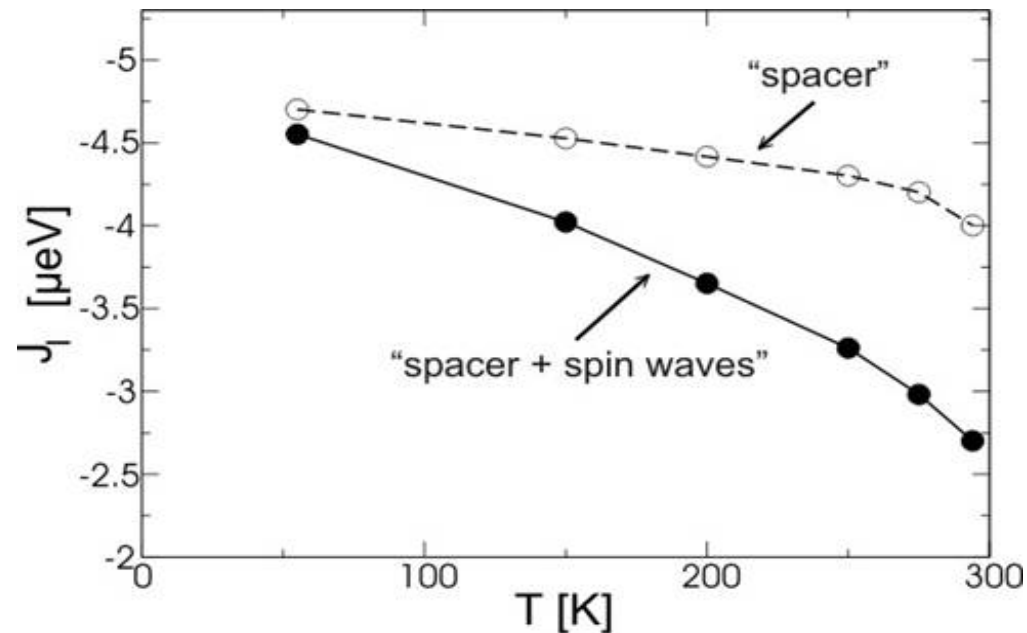
All contributions due to the spacer, interface and magnetic layers, nevertheless give an effective power law dependence on the temperature:

$$J(T) \approx 1 - AT^n, \quad n \approx 1.5 \quad (1)$$



T dependence of IEC

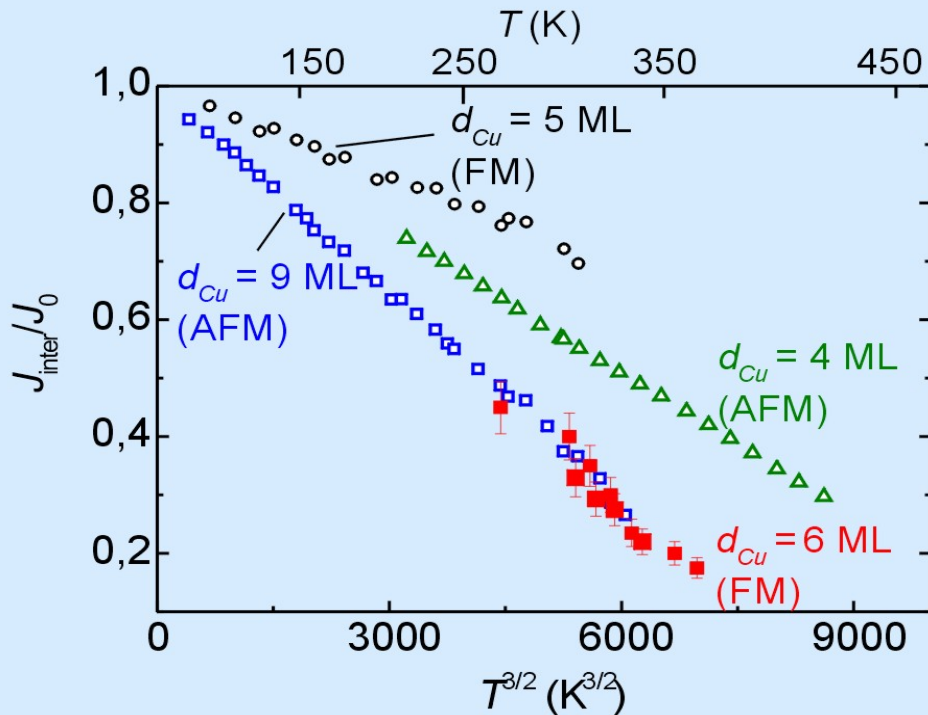
S. Schwieger et al., PRL **98**, 57205 (2007)



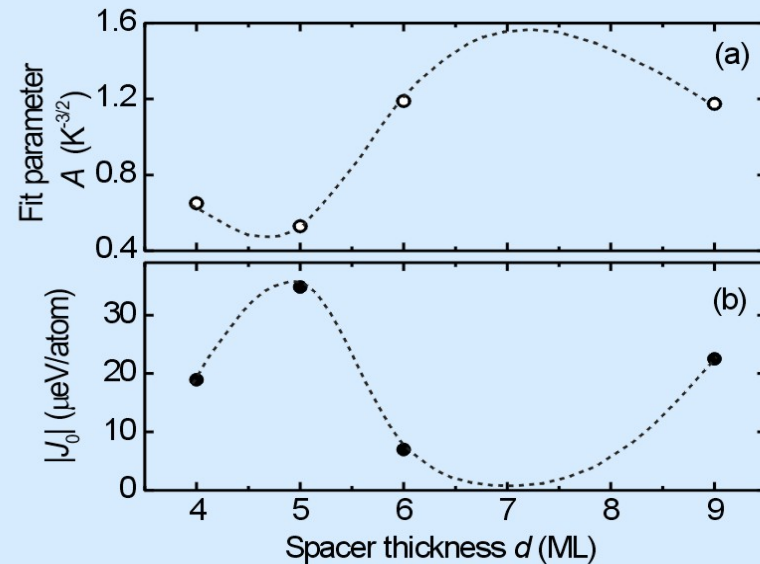
The dominant role of thermal magnon excitation in the temperature dependence of the interlayer exchange coupling: experimental verification

S. S. Kalarickal,* X. Y. Xu,† K. Lenz, W. Kuch, and K. Baberschke‡
Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

PRB 75, 224429 (2007)



$$J(T) \approx 1 - A(d)T^n, \text{ with } n \approx 1.5$$



- $A(d) \neq \text{const.}$
- $A(d) \neq \text{linear function}$
- $A(d) \approx \text{osc. function}$

- (interface)
- (electronic bandstructure)
- (spin wave excitation)

3. Gilbert damping versus magnon-magnon scattering.

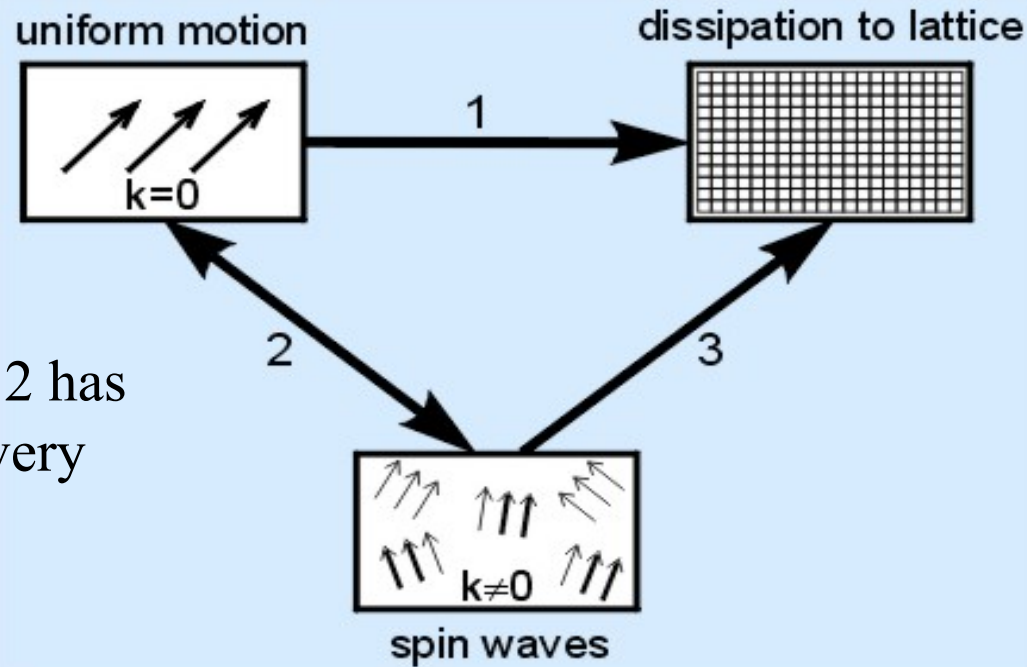
1834

IEEE TRANSACTIONS ON MAGNETICS, VOL. 34, NO. 4, JULY 1998

THEORY OF THE MAGNETIC DAMPING CONSTANT

Harry Suhl

Department of Physics, and Center for Magnetic Recording Research, Mail Code 0319,
University of California-San Diego, La Jolla, CA 92093-0319.



In nanoscale magnetism path 2 has been discussed very very little.

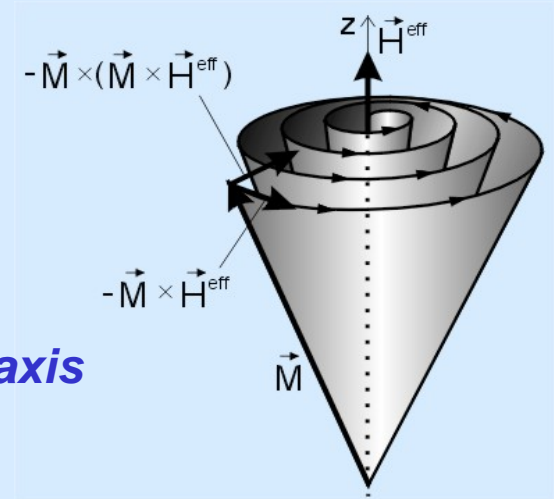
Mostly an effective damping (path 1) was modeled/fitted.

Landau-Lifshitz-Gilbert equation(1935)

$$\frac{d\mathbf{m}}{dt} = -\gamma \mathbf{m} \times \mathbf{H}_{\text{eff}} + \alpha \mathbf{m} \times \frac{d\mathbf{m}}{dt}$$

Gilbert damping

*$|M| = \text{const.}$
 M spirals on
a sphere into z -axis*



Bloch-Bloembergen Equation (1956)

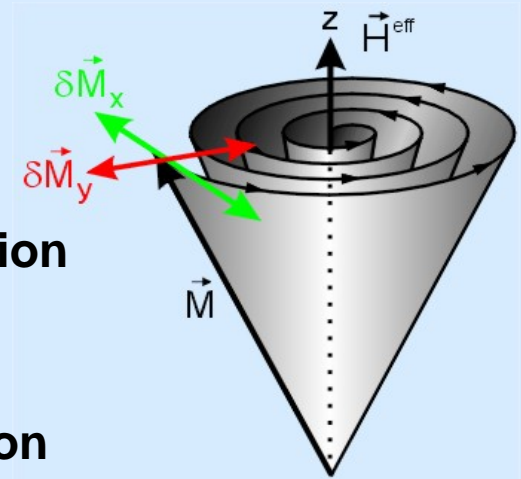
$$\frac{dm_z}{dt} = -\gamma (\mathbf{m} \times \mathbf{H}_{\text{eff}})_z - \frac{m_z - M_S}{T_1}$$

$$\frac{dm_{x,y}}{dt} = -\gamma (\mathbf{m} \times \mathbf{H}_{\text{eff}})_{x,y} - \frac{m_{x,y}}{T_2}$$

**spin-lattice relaxation
(longitudinal)**

**spin-spin relaxation
(transverse)**

$M_z = \text{const.}$



FMR Linewidth - Damping

Landau-Lifshitz-Gilbert-Equation

$$\frac{1}{\gamma} \frac{\partial \mathbf{M}}{\partial t} = -(\mathbf{M} \times \mathbf{H}_{\text{eff}}) + \frac{G}{\gamma M_S^2} \left(\mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t} \right)$$

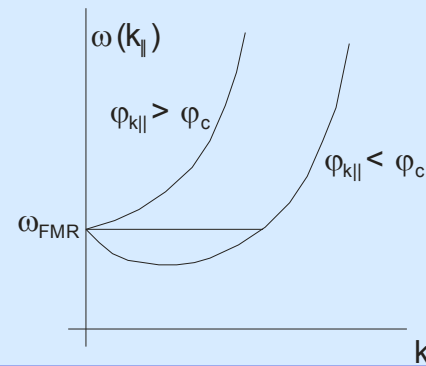
viscous damping,
energy dissipation

Gilbert-damping $\sim \omega$

$$\Delta H^{\text{Gil}}(\omega) = \frac{G}{\gamma^2 M_S} \omega$$

2-magnon-scattering

R. Arias, and D.L. Mills, *Phys. Rev. B* **60**, 7395 (1999);
D.L. Mills and S.M. Rezende in
'Spin Dynamics in Confined Magnetic Structures',
edt. by B. Hillebrands and K. Ounadjela, Springer Verlag



$$\Delta H^{2\text{Mag}}(\omega) = \Gamma \arcsin \sqrt{\frac{[\omega^2 + (\omega_0/2)^2]^{1/2} - \omega_0/2}{[\omega^2 + (\omega_0/2)^2]^{1/2} + \omega_0/2}}$$

$\omega_0 = \gamma(2K_{2\perp} - 4\pi M_S)$, $\gamma = (\mu_B/h)g$
 $K_{2\perp}$ - uniaxial anisotropy constant
 M_S - saturation magnetization

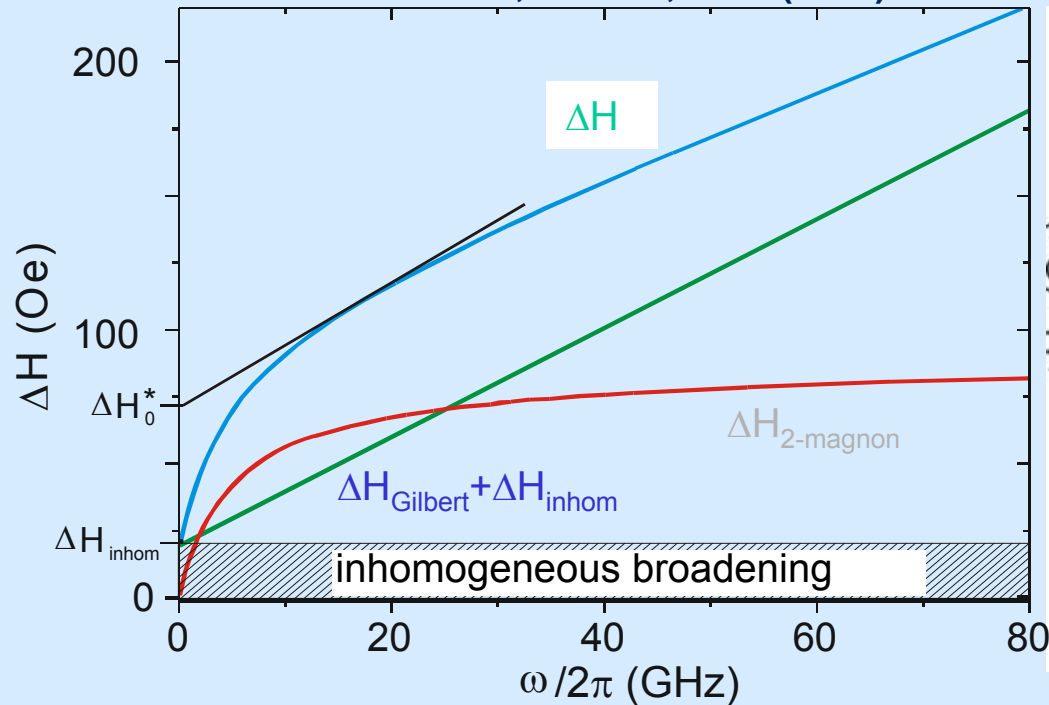
Which FMR-publication has checked (disproved) quantitatively this analytical function?

- Gilbert damping contribution:
- linear in frequency
- two-magnon excitations (thin films):
non-linear frequency dependence

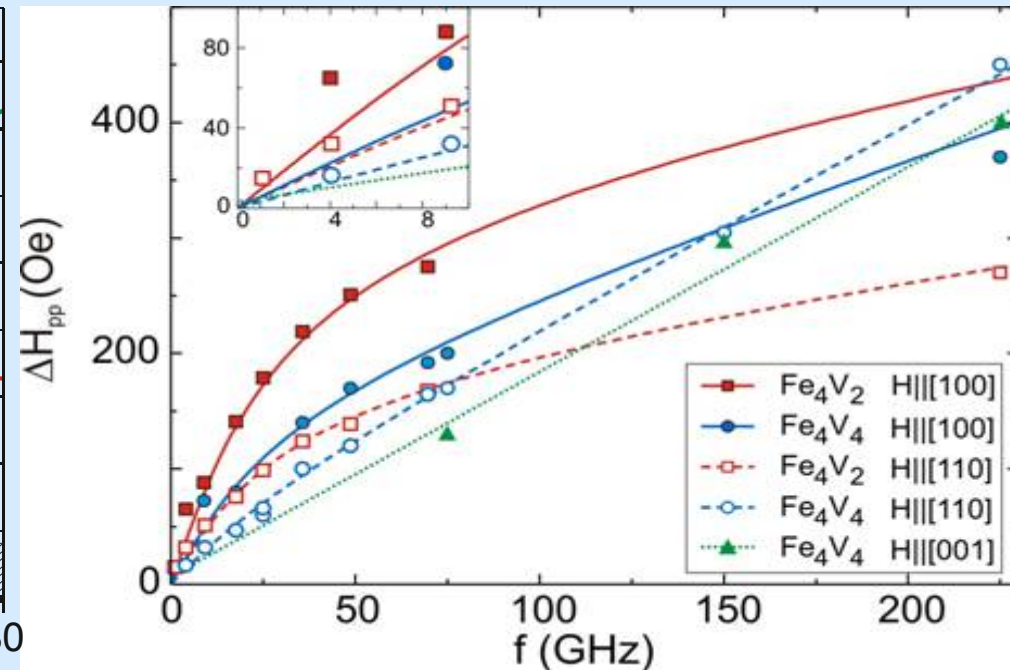
$$\Delta H_{2\text{-magnon}}(\omega) = \Gamma \arcsin \sqrt{\frac{\sqrt{\omega^2 + (\omega_0/2)^2} - \omega_0/2}{\sqrt{\omega^2 + (\omega_0/2)^2} + \omega_0/2}}$$

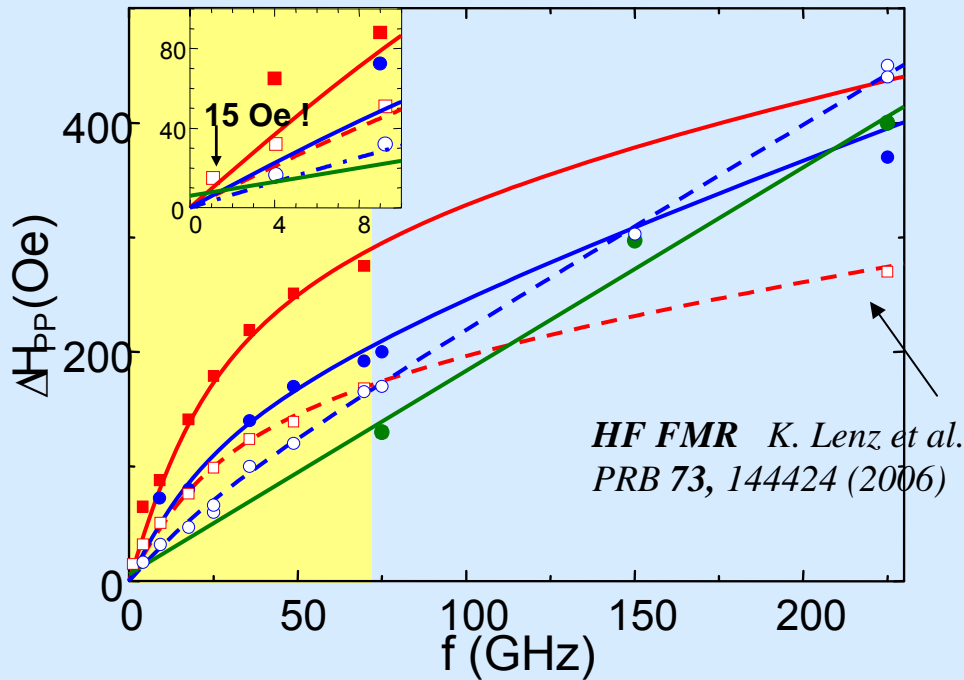
with $\omega_0 = \gamma M_{\text{eff}}$

R. Arias et al., PRB 60, 7395 (1999)



K. Lenz et al., PRB 73, 144424 (2006)





- **two-magnon scattering observed in Fe/V superlattices –**

J. Lindner et al., PRB 68, 060102(R) (2003)

real relaxation – no inhomogeneous broadening
two-magnon damping dominates Gilbert damping
by two orders of magnitude:

$1/T_2 \sim 10^9 \text{ s}^{-1}$ vs. $1/T_1 \sim 10^7 \text{ s}^{-1}$

- **recent publications with similar results:**
 - Pd/Fe on GaAs(001) – network of misfit dislocations
G. Woltersdorf et al. PRB 69, 184417 (2004)
 - NiMnSb films on InGaAs/InP
B. Heinrich et al. JAP 95, 7462 (2004)

	Γ (kOe)	$\gamma \cdot \Gamma$ (10^8 s^{-1})	G (10^8 s^{-1})	α (10^{-3})	ΔH_0 (Oe)
■ Fe_4V_2 ; H [100]	0.270	50.0	0.26	1.26	0
● Fe_4V_4 ; H [100]	0.139	26.1	0.45	2.59	0
□ Fe_4V_2 ; H [110]	0.150	27.9	0.22	1.06	0
○ Fe_4V_4 ; H [110]	0.045	8.4	0.77	4.44	0
● Fe_4V_4 ; H [001]	0	0	0.76	4.38	5.8

Angular- and frequency-dependent FMR

on

Fe₃Si binary Heusler structures
epitaxially grown on MgO(001)

d = 40nm

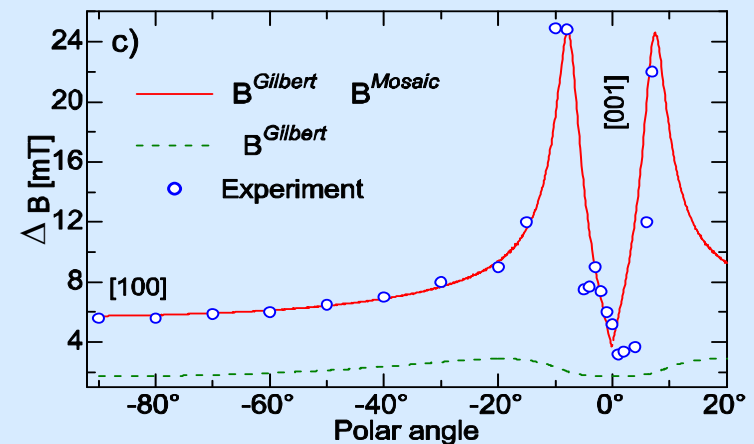
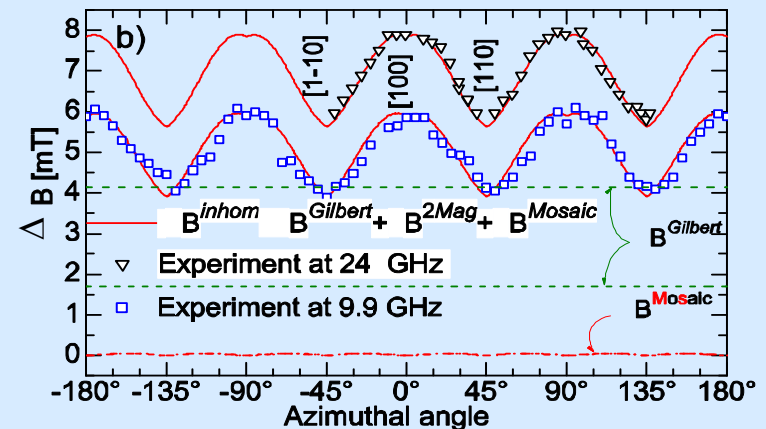
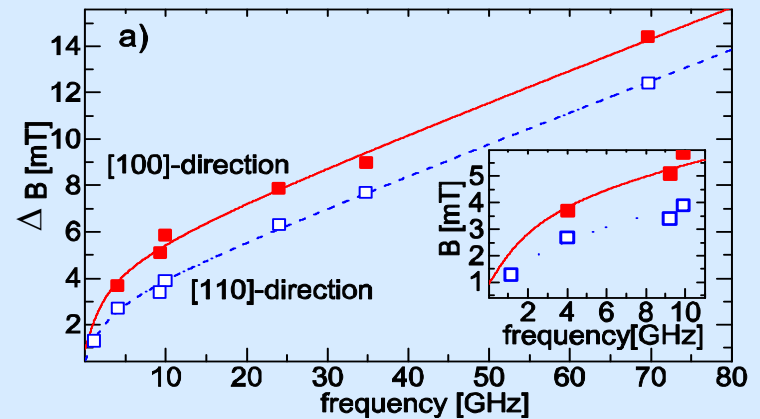
Kh. Zakeri et al.

PRB 76,104416 (2007)

Angular dependence at 9 and 24 GHz

$\gamma\Gamma \approx (26 - 53) \cdot 10^7 \text{ sec}^{-1}$, anisotropic

$G \approx 5 \cdot 10^7 \text{ sec}^{-1}$, isotropic



Conclusion

For nanoscale ferromagnets :

- use the reduced temperature $t = T/T_C$
- the orbital magnetic moment is **NOT** quenched
- the MAE may be larger by orders of magnitude

Higher order spin-spin correlations are important to explain the magnetism of nanostructures.

In most cases a *mean field model* is insufficient.

A phenomenological effective *Gilbert damping parameter* gives very little insight into the microscopic relaxation mechanism. It seems to be more instructive to separate scattering mechanisms within the magnetic subsystem from the dissipative scattering into the thermal bath

K. B. *Handbook of Magnetism and Advanced Magnetic Materials*, Vol. 3
Ed. Kronmüller and Parkin, 2007 John Wiley & Sons, Ltd.

www.physik.fu-berlin.de/~bab

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R. Wu, D.L. Mills, UCI; P. Jensen + K.H. Bennemann, FUB; W. Nolting, HUB



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