Phase transitions in ferromagnetic monolayers: A playground to study fundamentals

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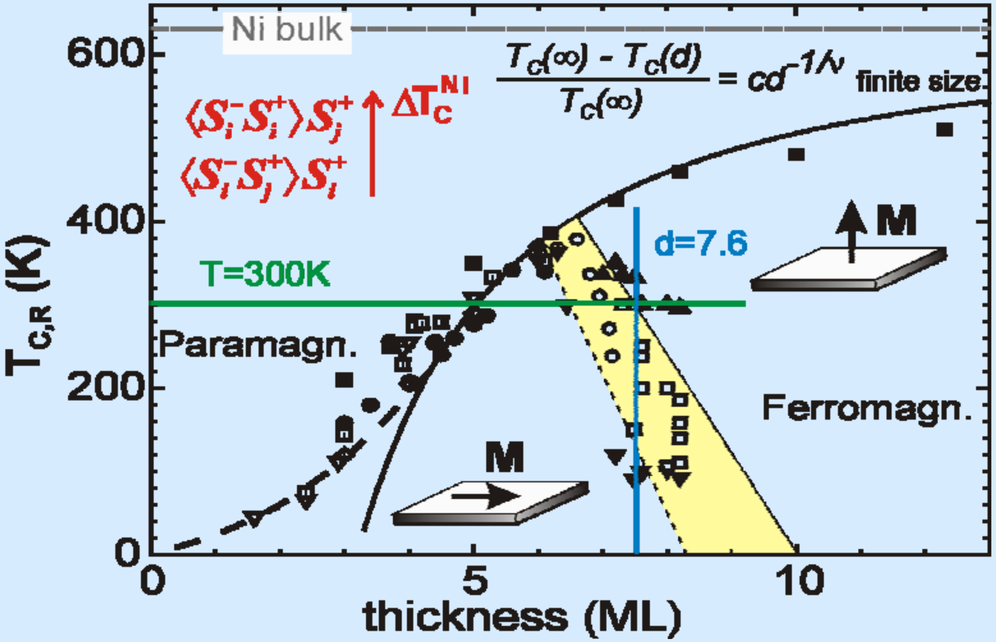
- T_{crit} , Phase Transitions
- Spin Reorientation Transition
  Magnetic Anisotropy Energy
- Manipulation of the SRT
- Summary, Take Home



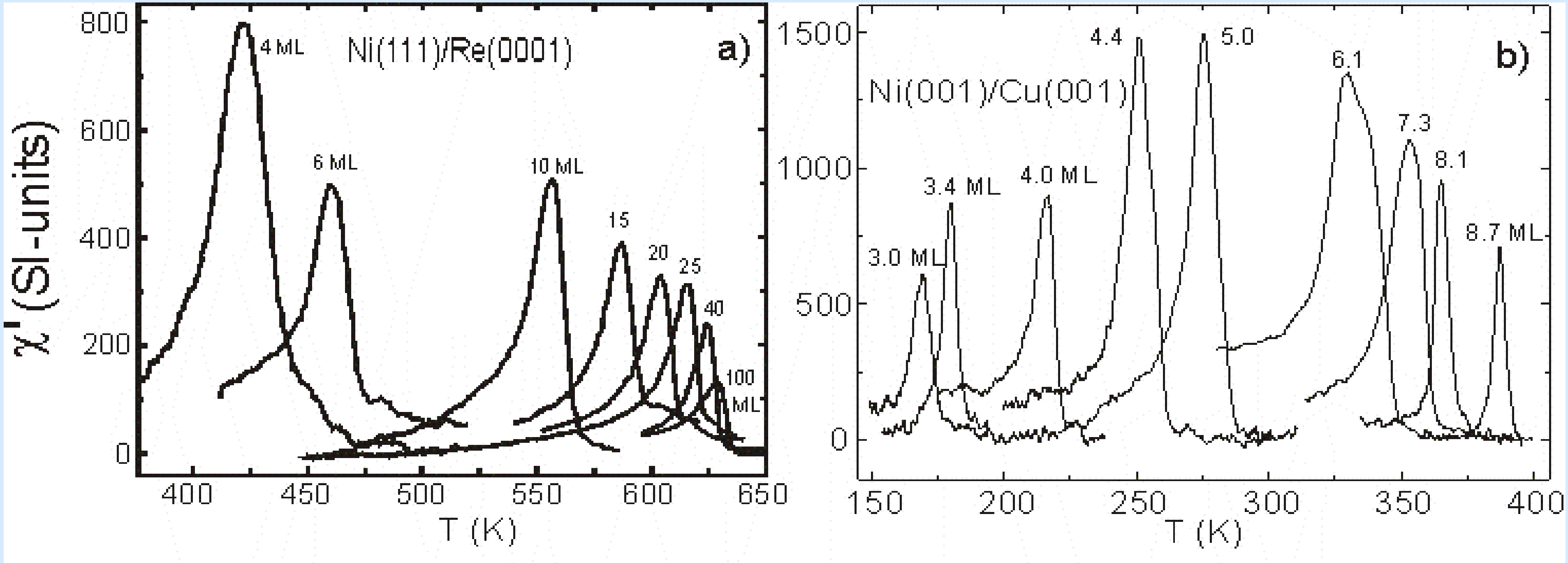
Para- to ferromagnetic phase transition, Curie temperature,

Bulk Fe, Co, Ni have only one T_C, each (few % changes are possible).

For ultrathin films T_C can be manipulated from zero to T_C^{bulk}.

New and Great !

How do we measure T_C? χ_{ac} increases at T_C ; dc-MOKE vanishes!



"Magnetism in thin films"

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

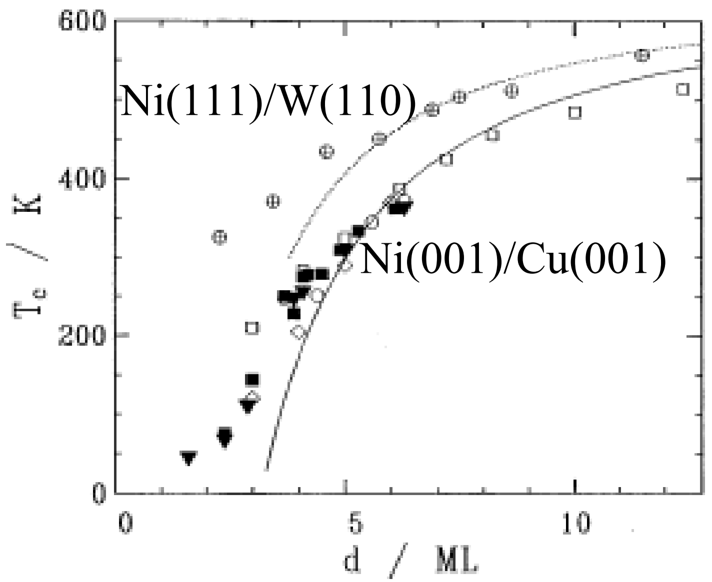
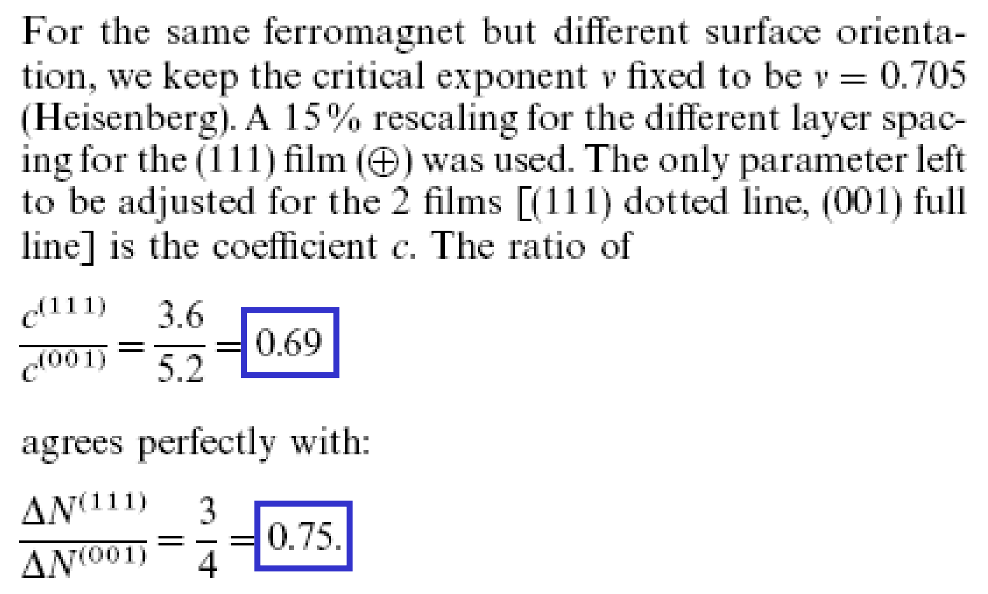


Fig. 7. Critical temperature T_c for: Ni (111)/W (110) ⊕ [6], and Ni (001)/Cu (001) dc-MCXD (▼), ac-MCXD ■ [15], and □ [14], ○ [26], ◇ [25]. The *solid* and *dotted lines* are fits to (8) with values for c given in the text

Different T_C for Ni(111) and Ni(001). How can this be explained ?



For the same ferromagnet but different surface orientation, we keep the critical exponent ν fixed to be $\nu = 0.705$ (Heisenberg). A 15% rescaling for the different layer spacing for the (111) film (⊕) was used. The only parameter left to be adjusted for the 2 films [(111) dotted line, (001) full line] is the coefficient c. The ratio of

$$\frac{c^{(111)}}{c^{(001)}} = \frac{3.6}{5.2} = \boxed{0.69}$$

agrees perfectly with:

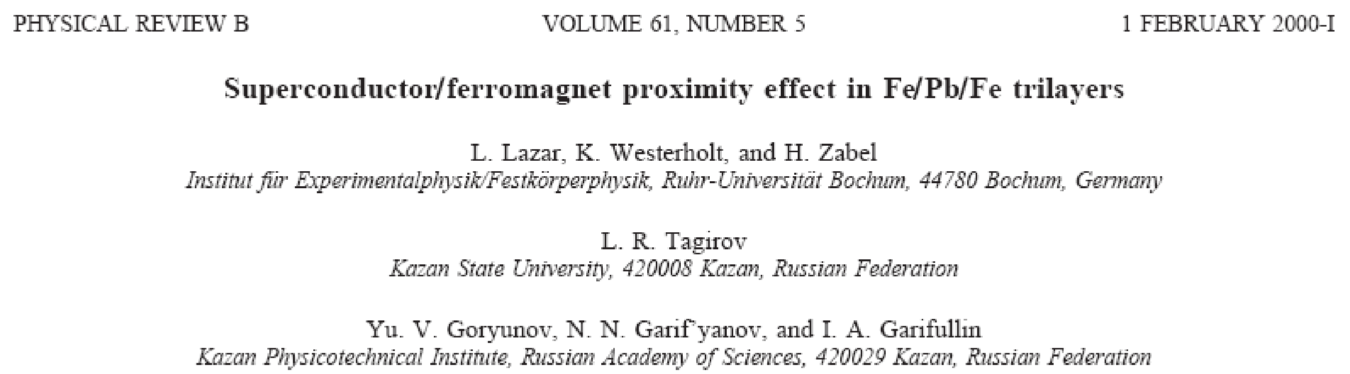
$$\frac{\Delta N^{(111)}}{\Delta N^{(001)}} = \frac{3}{4} = \boxed{0.75.}$$

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

finite size scaling,

K. B. Appl. Phys. A **62**, 417 (1996) #171

If T_C changes the reduced temperature $T/T_C = t$ is important.



PHYSICAL REVIEW B VOLUME 61, NUMBER 5 1 FEBRUARY 2000-I

Superconductor/ferromagnet proximity effect in Fe/Pb/Fe trilayers

L. Lazar, K. Westerholt, and H. Zabel
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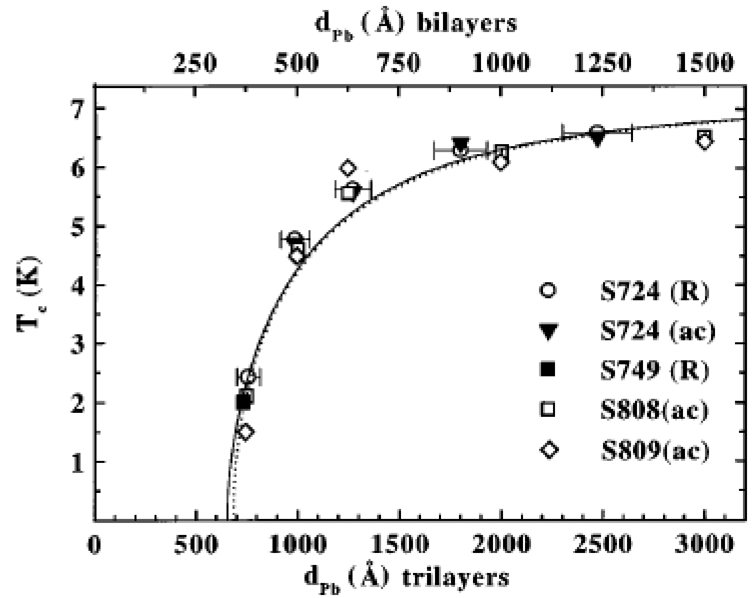


FIG. 7. Dependence of the superconducting transition temperature on the thickness of the Pb-layer for four sample series listed in Table I. The dotted and solid lines are the best fits using the theory by Radović *et al.* and the theory by Tagirov, respectively, with parameters given in the figure subscripts of Figs. 10 and 11.

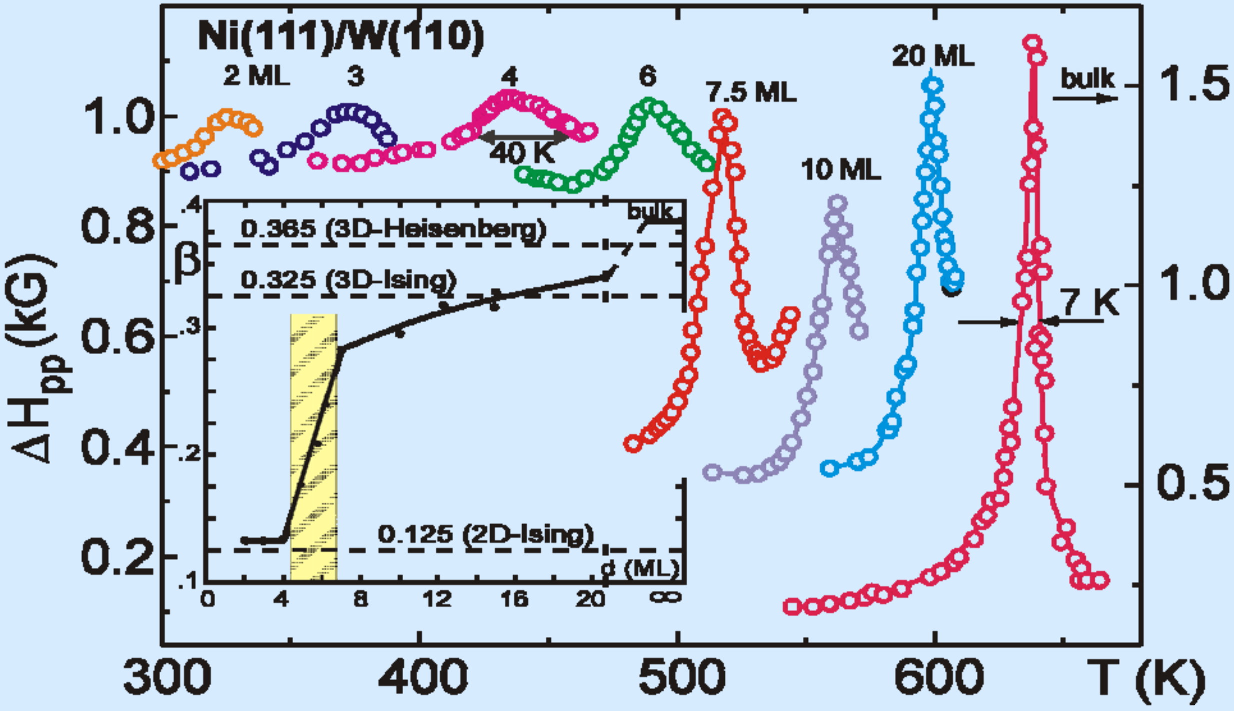
Finite size scaling is a general phenomenon and not specific for FM.

The difference lies in the different correlation lengths ξ.

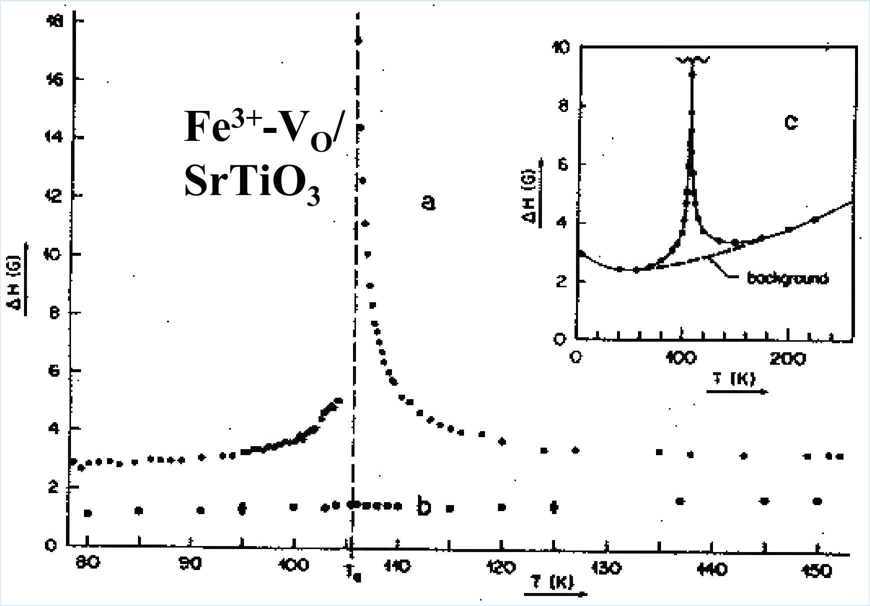
$$\xi = \xi_0 (1-t)^{-\nu}$$

Close to T_C fluctuations (Gaussian or critical) increase.

This increases the linewidth in magnetic resonance spectroscopy.

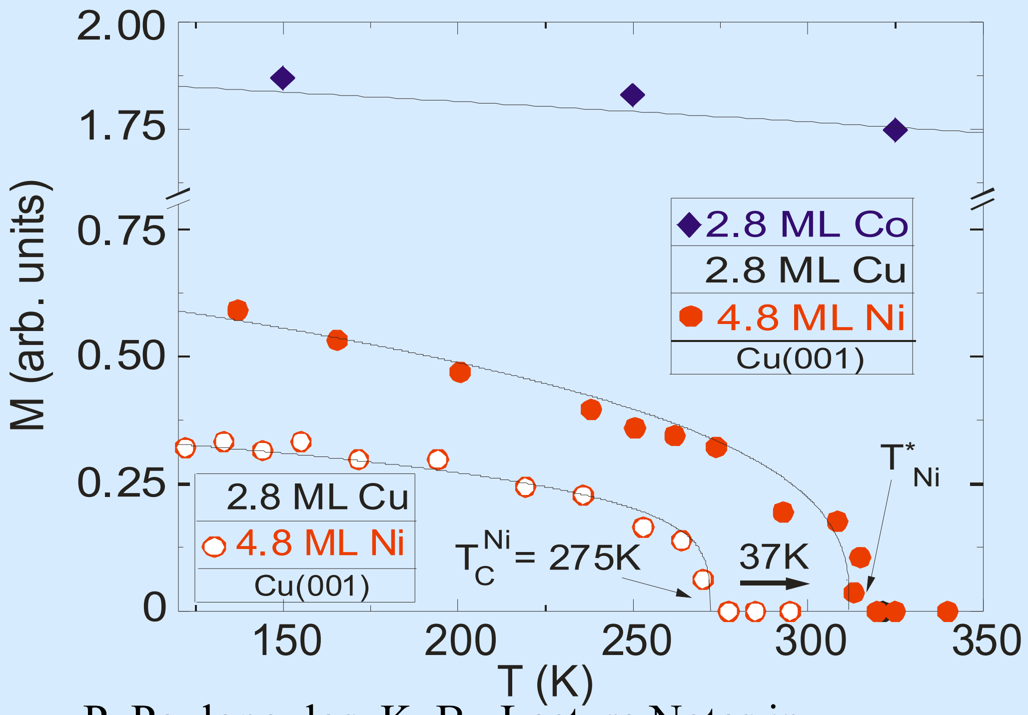


Yi Li, K. B., PRL **68**, 1208 (1992) #126

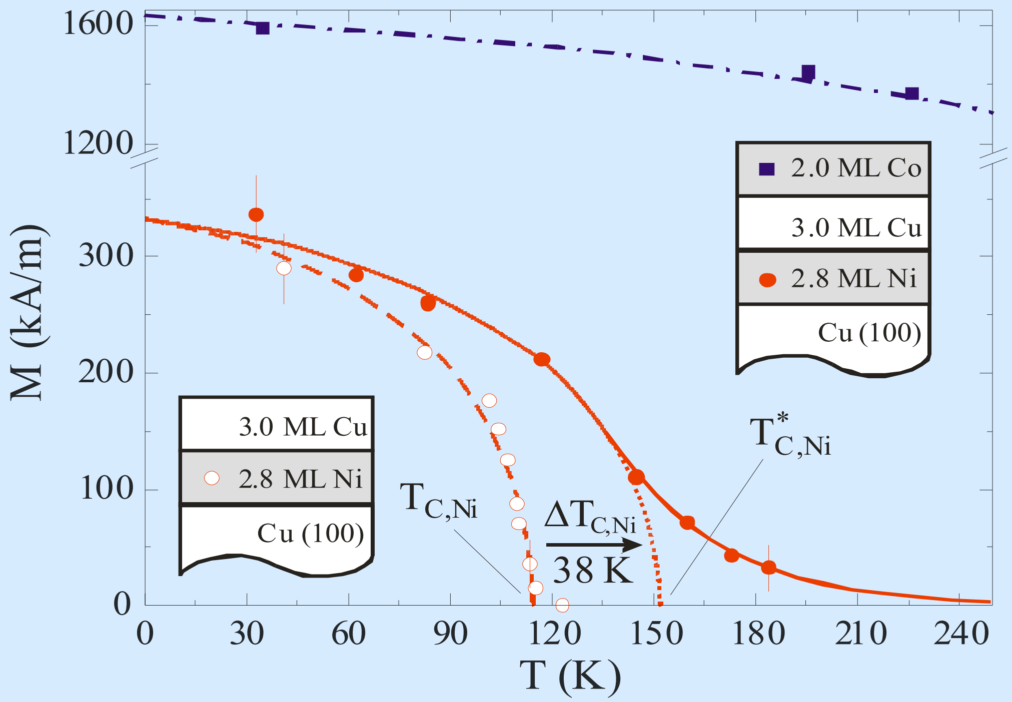


Th.v. Waldkirch, K.A. Müller,
W. Berlinger, PRB (1973)

Element specific UHV-XMCD measurements



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

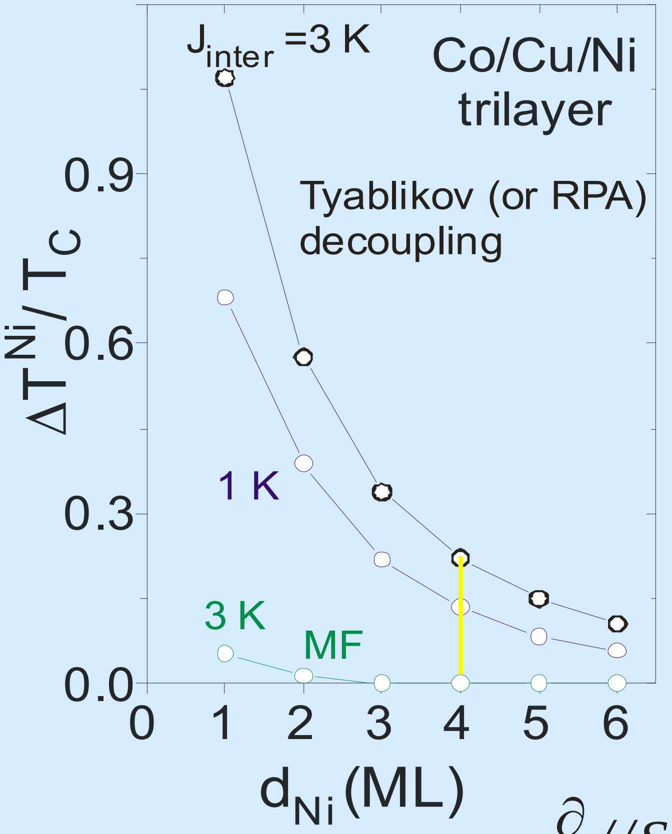


A. Scherz et al. PRB **65**, 24411 (2005)

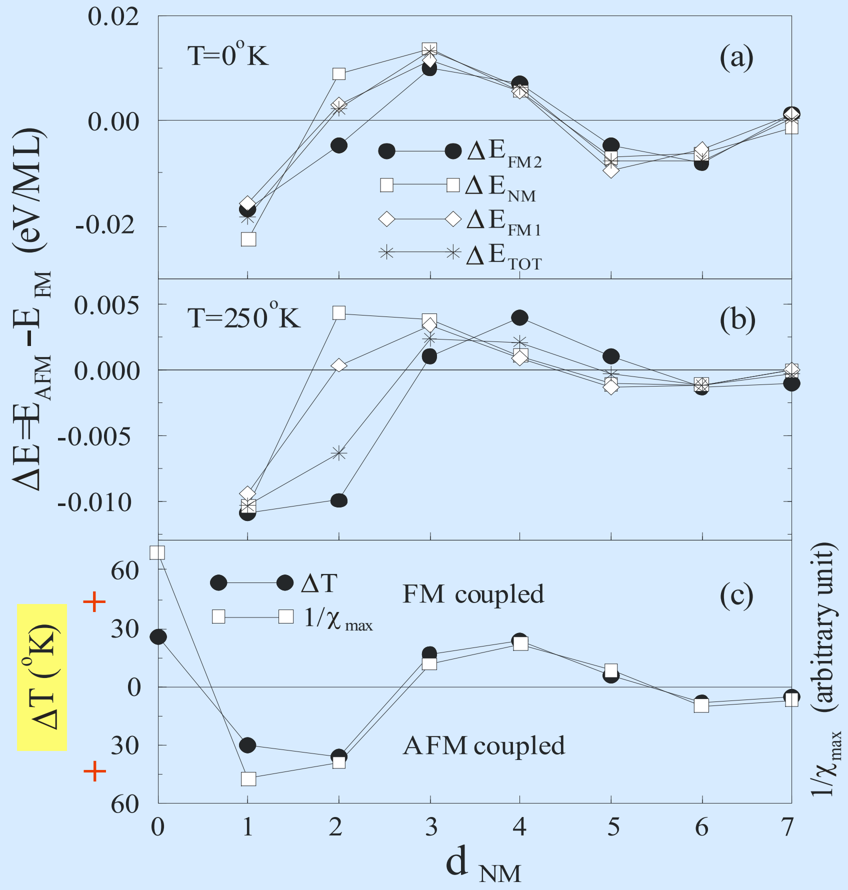
The large shift of T_C^{Ni} can **NOT** be explained by the static exchange field of Co.

Enhanced spin fluctuations in 2D (theory)

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



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



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

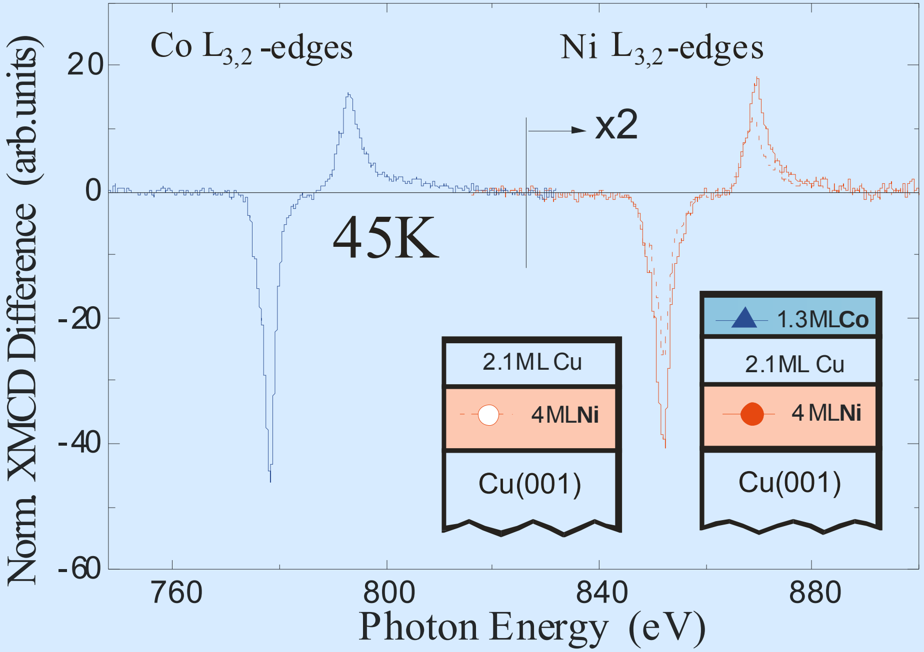
$$S_i^z S_j^+ \approx \langle S_i^z \rangle S_j^+ - \langle S_i^- S_i^+ \rangle S_j^+ - \langle S_i^- S_j^+ \rangle S_i^+ + \cdots$$

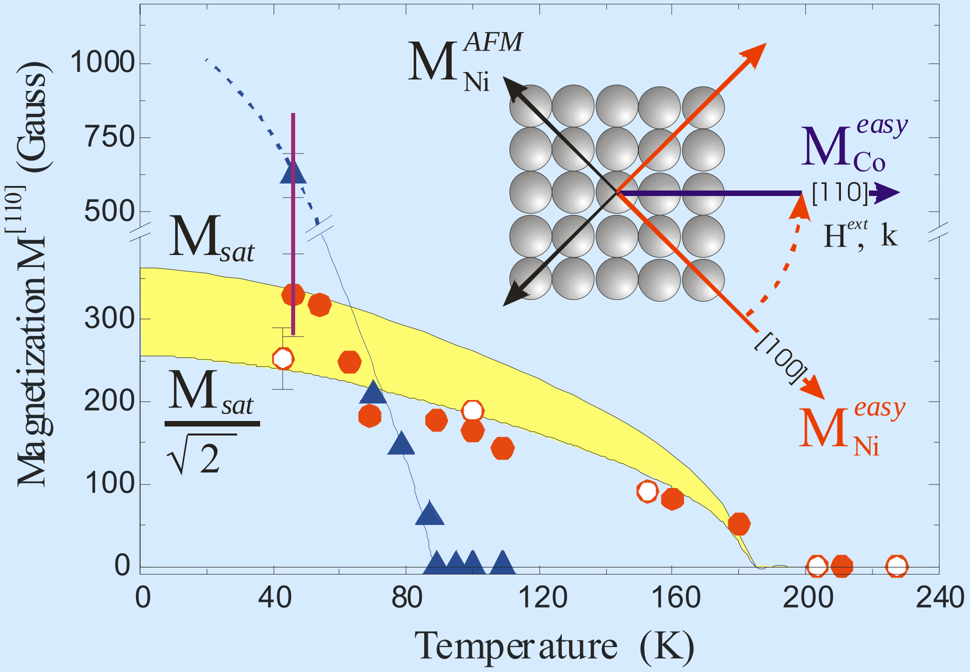
⟵ RPA ⟶

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

Single band Hubbard model:
Simple Hartree-Fock ansatz is insufficient
Higher order correlations are needed to explain T_C-shift

Two phase transitions at T_C^{Ni} and T_C^{Co} ?





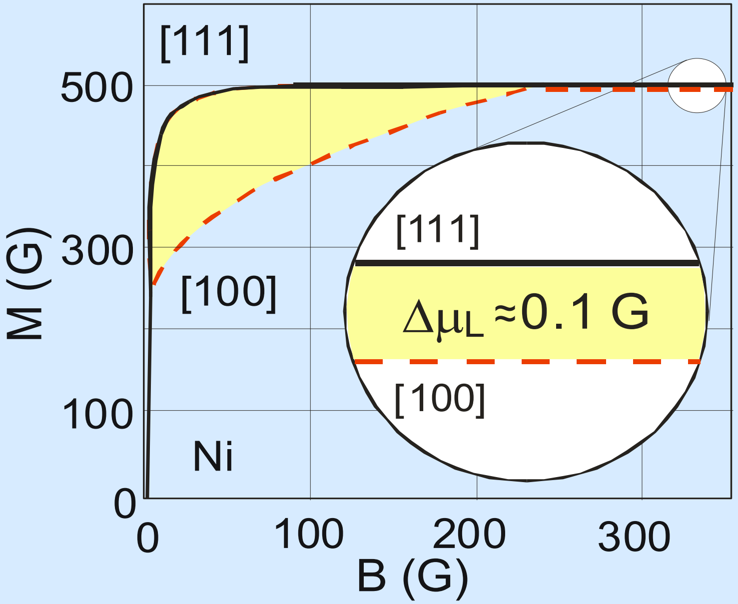
Crossover of $M_{Co}(T)$ and $M_{Ni}(T)$

A further reduction in symmetry happens at T_c^{low}

A. Scherz et al. J. Synchrotron Rad. **8**, 472 (2001) #248, 245

Magnetic Anisotropy Energy (MAE)

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



Characteristic energies of metallic ferromagnets

binding energy	1 - 10 eV/atom
exchange energy	10 - 10^3 meV/atom
cubic MAE (Ni)	0.2 µeV/atom
uniaxial MAE (Co)	70 µeV/atom

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

$$\text{MAE} = \int M\cdot dB \approx \tfrac{1}{2}\,\Delta M\cdot\Delta B \approx \tfrac{1}{2}\,200\cdot 200\ \text{G}^2$$

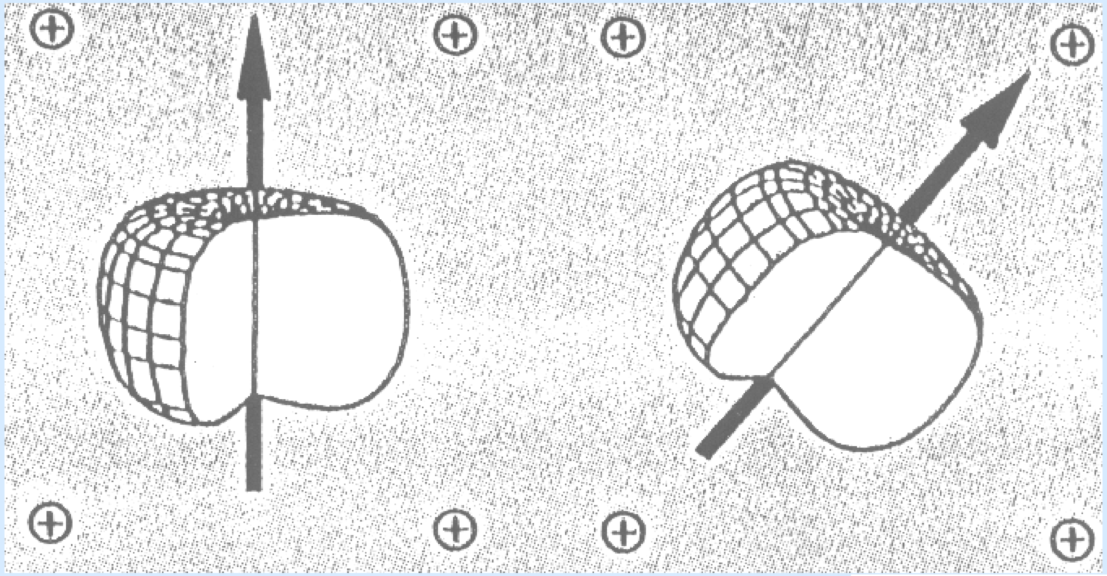
$$\text{MAE} \approx 2\cdot 10^4\ \text{erg}/\text{cm}^3 \approx 0.2\ \mu\text{eV}/\text{atom}$$

≈ 1µeV/atom is very small compared to
≈ 10 eV/atom total energy but all important

The origin of MAE

There are <u>only 2 origins</u> for MAE: 1) dipol-dipol interaction ~ $(\bar{\mu}_1 \bullet \bar{r})(\bar{\mu}_2 \bullet \bar{r})$ and

2) spin-orbit coupling $\lambda \bar{L}\,\bar{S}$ (intrinsic K or ΔE_{band})



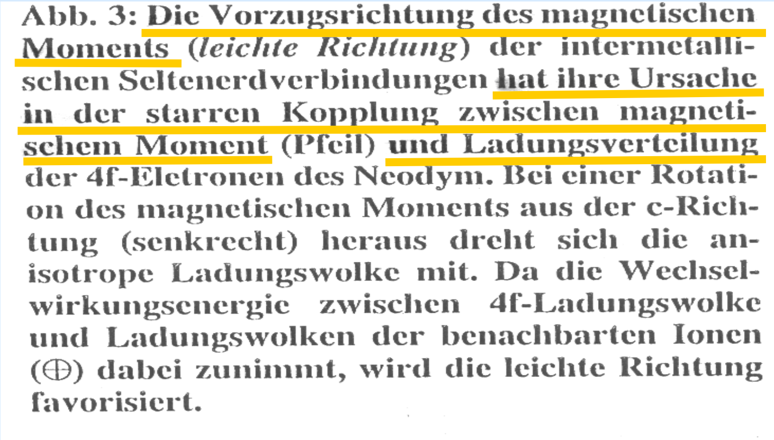
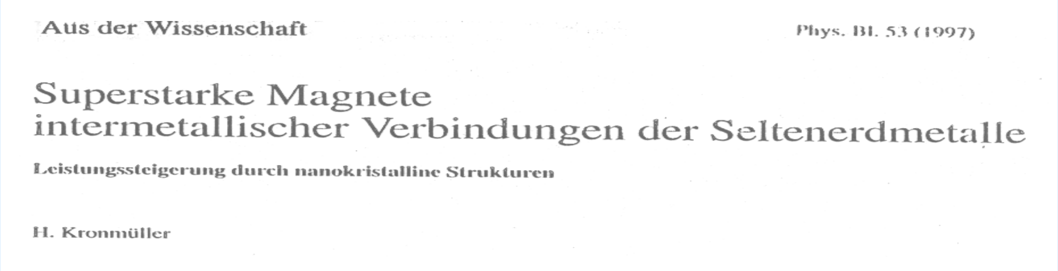


Abb. 3: Die Vorzugsrichtung des magnetischen Moments (*leichte Richtung*) der intermetallischen Seltenerdverbindungen hat ihre Ursache in der starren Kopplung zwischen magnetischem Moment (Pfeil) und Ladungsverteilung der 4f-Eletronen des Neodym. Bei einer Rotation des magnetischen Moments aus der c-Richtung (senkrecht) heraus dreht sich die anisotrope Ladungswolke mit. Da die Wechselwirkungsenergie zwischen 4f-Ladungswolke und Ladungswolken der benachbarten Ionen (⊕) dabei zunimmt, wird die leichte Richtung favorisiert.



Aus der Wissenschaft Phys. Bl. 53 (1997)

Superstarke Magnete intermetallischer Verbindungen der Seltenerdmetalle

Leistungssteigerung durch nanokristalline Strukturen

H. Kronmüller

Free energy density of MAE, K

(intrinsic, after substraction of the shape anisotropy $2\pi M^2$)

tetragonal [e.g. Ni, Co, Fe (001) / Cu (001)]:

$$E_{tetr} = - K_2 \cos^2\theta \quad - {}^1/_2\, K_{4\perp} \cos^4\theta - {}^1/_2\, K_{4||}\, {}^1/_4\, (3+\cos 4\varphi) \sin^4\theta + \ldots$$ *(Bab et al.)*

$$= (K_2 + K_{4\perp}) \sin^2\theta \quad - {}^1/_2\, (K_{4\perp} + {}^3/_4\, K_{4||}) \sin^4\theta - {}^1/_8\, K_{4||} \cos 4\varphi \sin^4\theta + \ldots$$

$$= K_2' \sin^2\theta \quad + K_{4\perp} \sin^4\theta + K_{4||} \cos 4\varphi \sin^4\theta + \ldots$$ *(traditional)*

hexagonal [e.g. Ni (111), Gd (0001) / W (110)]:

$$E_{hex} = k_2 \sin^2\theta + {}^1/_2 k_{2||} \cos 2\varphi \sin^2\theta + k_4 \sin^4\theta + k_{6\perp} \sin^6\theta + k_{6||} \cos 6\varphi \sin^6\theta + \ldots$$

each K_i has a „volume“ and „surface“ contribution

$$K_i = K_i^v + 2K_i^s/d$$

M. Farle Rep. Prog. Phys. **61**,755 (1998)
K. B. *Handb. Magn. Magn. Mat.* **3,** 1617 (2007) # see web

Spin reorientation in bulk ferromagnets

SRT for hcp Co
$\sin\theta = (K_2/2K_4)^{1/2}$

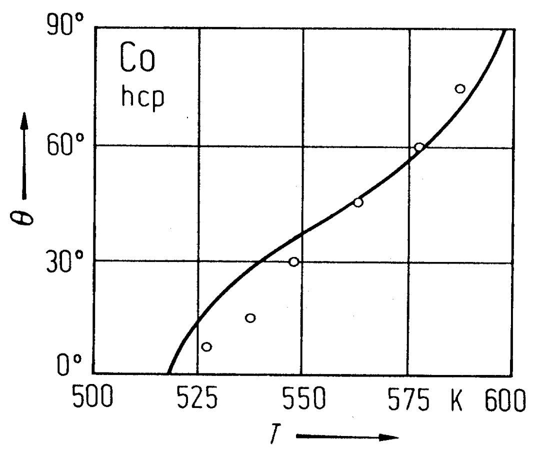


Fig. 5. Temperature dependence of the angle θ between the direction of spontaneous magnetization and the c axis of a single crystal of hcp Co [61 B 5]. Points: data. Curve: calculated from $\sin\theta=(-K_1/2K_2)^{1/2}$.

LB III, 19a, p.45

NIGH, LEGVOLD, AND SPEDDING PR **132**, 1092 (63)

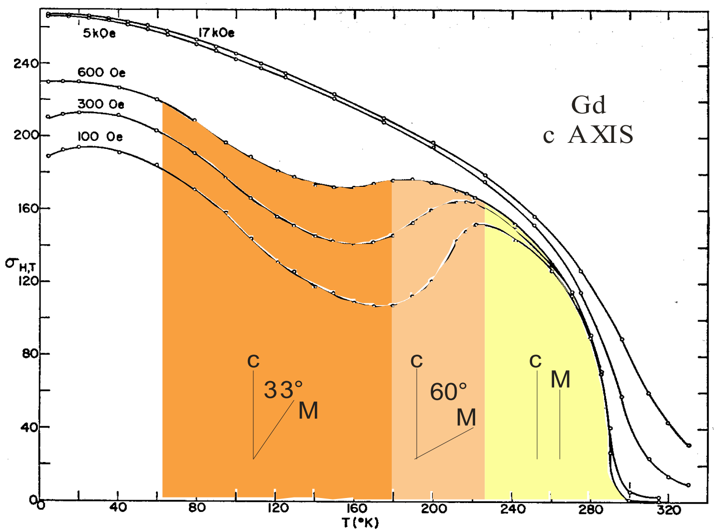


FIG. 4. Magnetic moment per gram versus temperature for the c-axis crystal. Internal magnetic fields are indicated.

Gd ist not isotropic, it has K_2, K_4, $K_6 \neq 0$

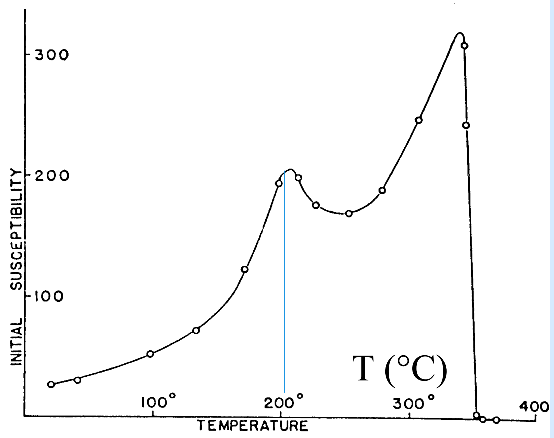
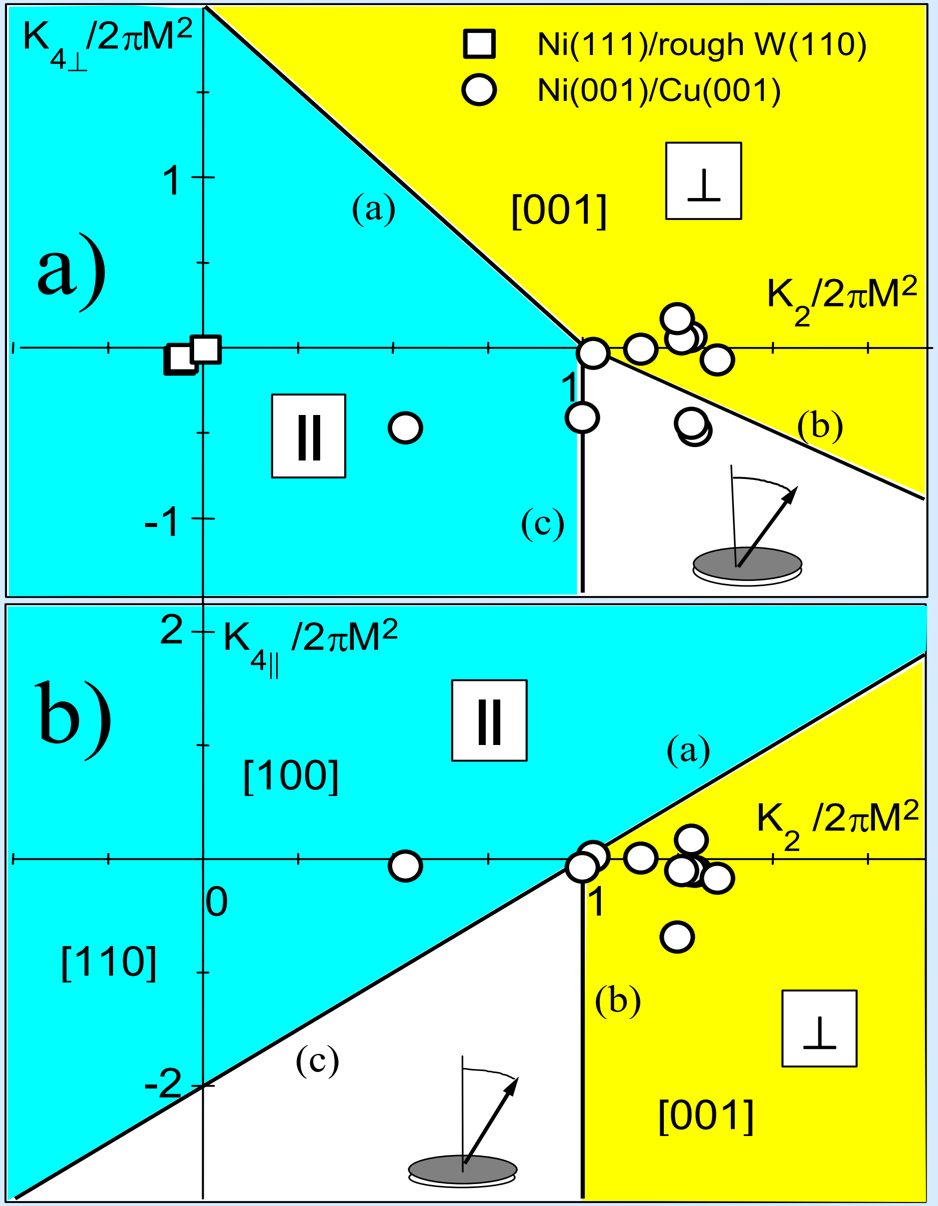


FIG. 6. The variation of the initial susceptibility with temperature in nickel.

At the extremal value of K_2 a reorientation and second maximum in χ appears

SRT in ultrathin Ni films



A. Berghaus, M. Farle, Yi Li, K. B.
Second Intern. Workshop on the Magnetic Properties of Low-Dimensional Systems.
Proc. in Physics **50**, 61 (1989) #108

M. Farle et al., PRB **55**, 3708 (1997) #176

Only with $K_4 \neq 0$ a continues SRT is possible!

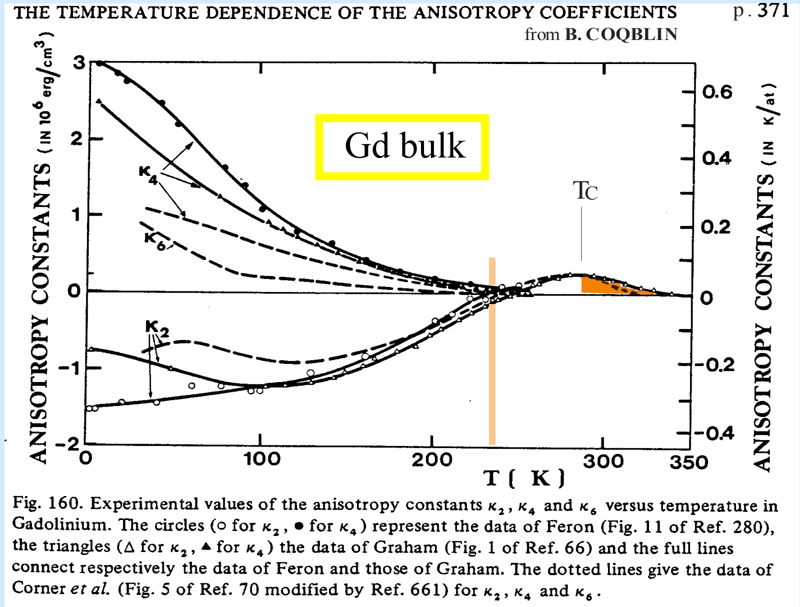
$$\sin\theta = (K_2/2K_4)^{1/2}$$

Do not use $K_{eff} = 2\pi M^2 - K_i$ …
because f(T) and g(T) are different.
Use the ratio $K_i / 2\pi M^2 \Rightarrow f(T) / g(T)$

What causes the SRT?

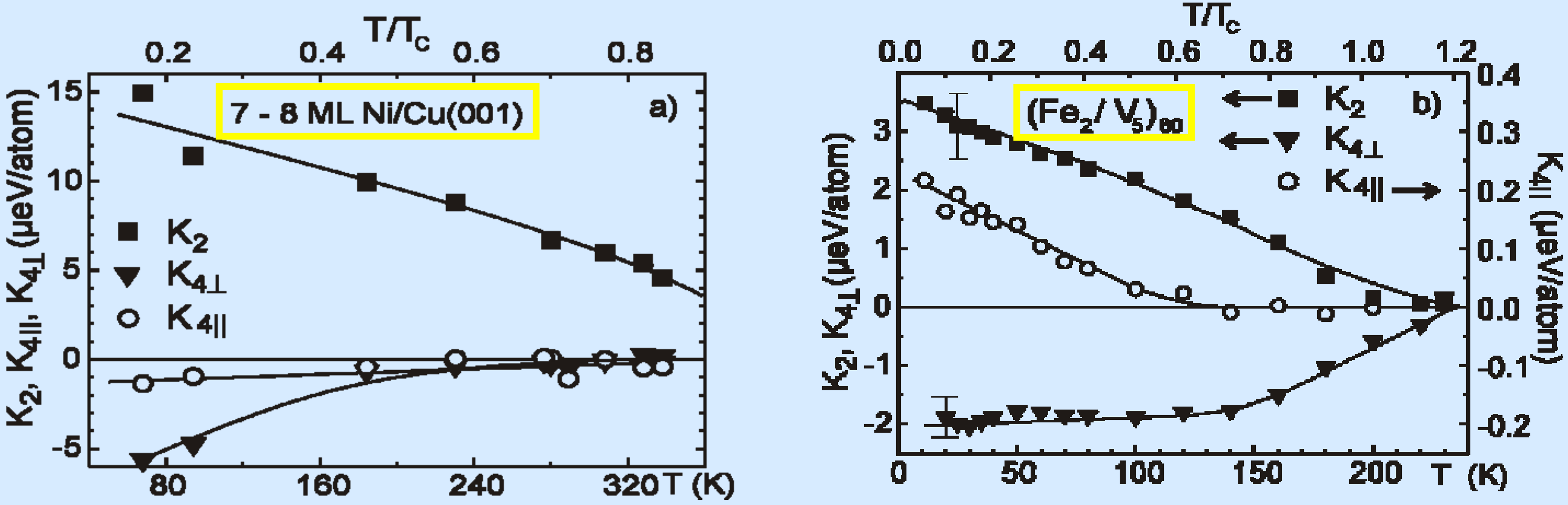
There is only **one** reason, the K_i (T,d)!

Higher order MAE K_i



THE TEMPERATURE DEPENDENCE OF THE ANISOTROPY COEFFICIENTS p. 371
from **B. COQBLIN**

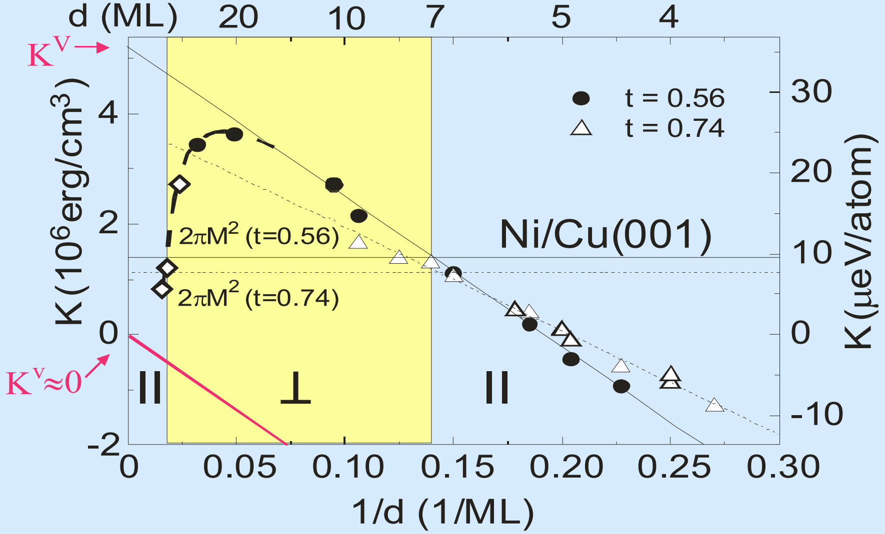
Fig. 160. Experimental values of the anisotropy constants κ_2, κ_4 and κ_6 versus temperature in Gadolinium. The circles (○ for κ_2, ● for κ_4) represent the data of Feron (Fig. 11 of Ref. 280), the triangles (△ for κ_2, ▲ for κ_4) the data of Graham (Fig. 1 of Ref. 66) and the full lines connect respectively the data of Feron and those of Graham. The dotted lines give the data of Corner *et al.* (Fig. 5 of Ref. 70 modified by Ref. 661) for κ_2, κ_4 and κ_6.



Ni is the only 3d ferromagnet for which $T_C(d)$ is measured over the full range of temperature and thickness

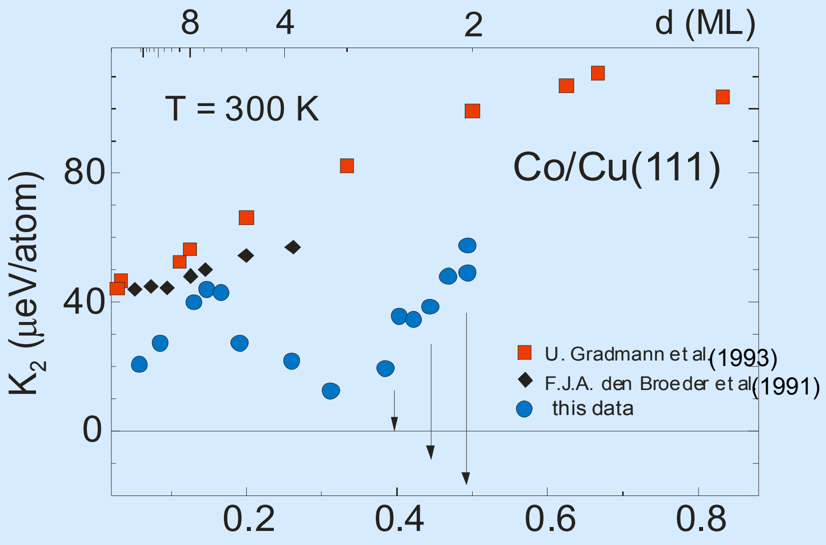
To analyze SRT in thin films is difficult and tedious because there is a T and 1/d dependence:

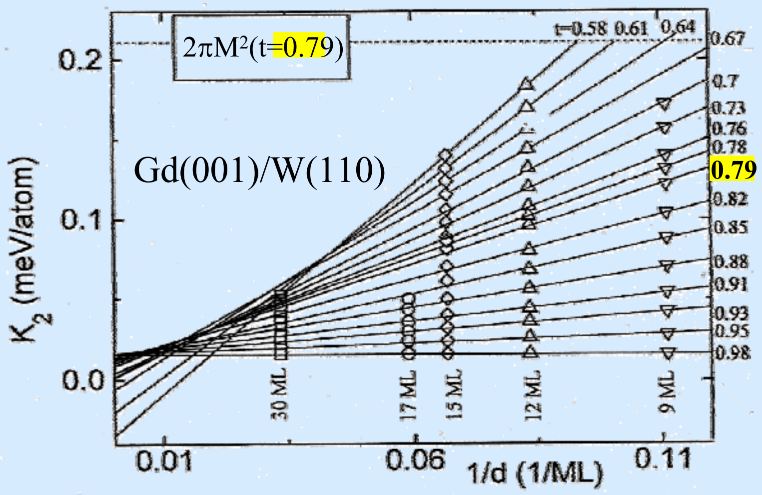
$T/T_C(d)$ and $K(T)=K_V(T)+2K_S(T)/d$



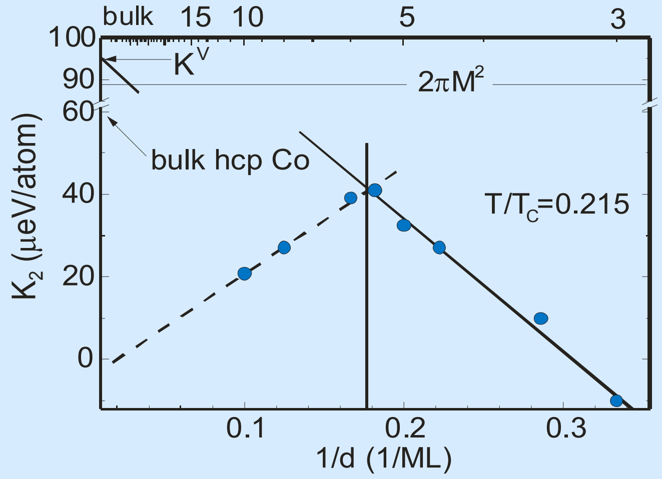
Why does Ni undergo a SRT and Co not ?

=> #330





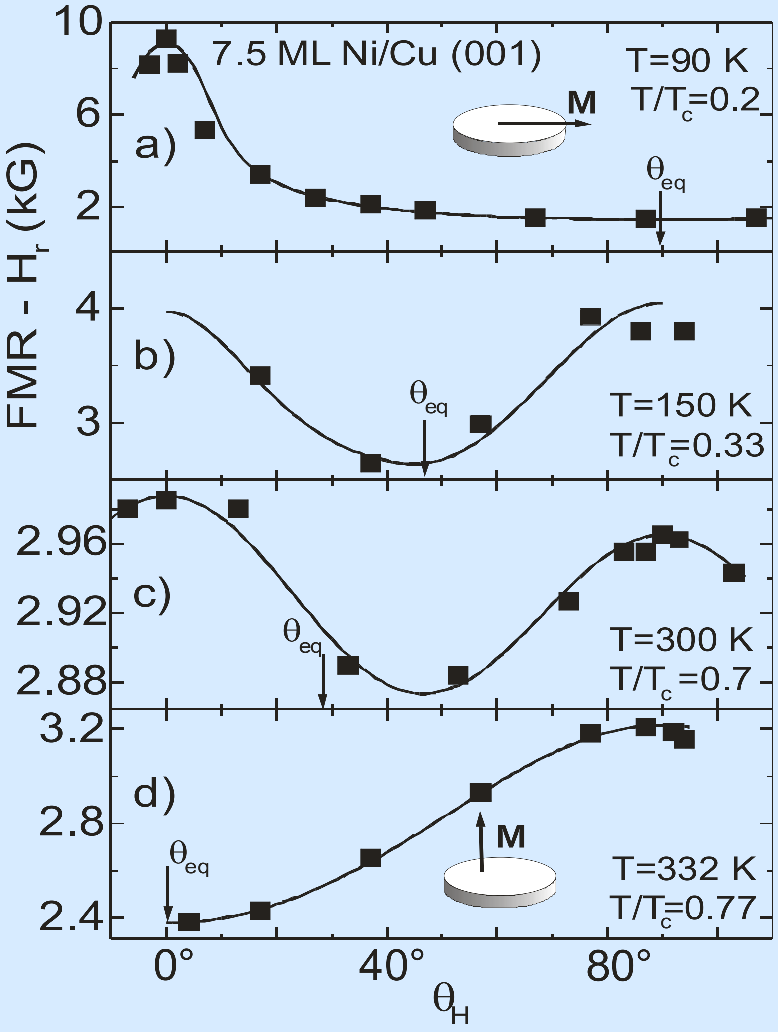
G. André et al., Surface Science **326**, 275 (1995)
K. B. and M. Farle, J. Appl. Phys. **81**, 5038 (1997)

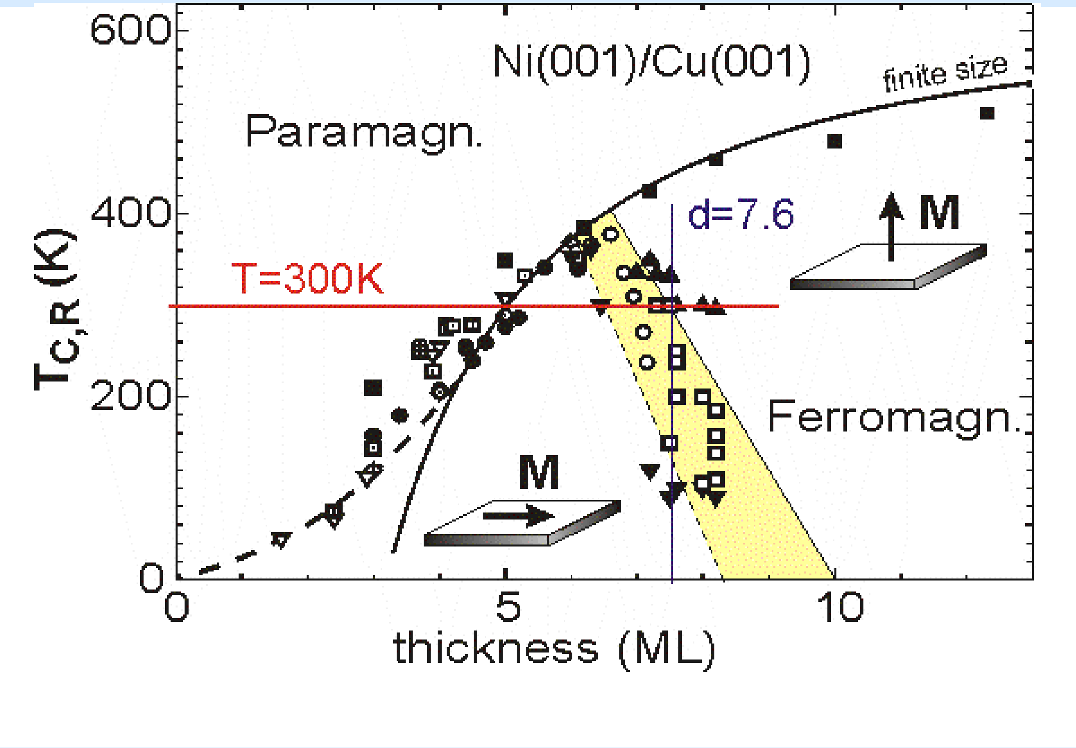


M. Farle et al., Surf. Sci. **439**, 146 (1999)

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.

Experimental FMR evidence for a continuous rotation

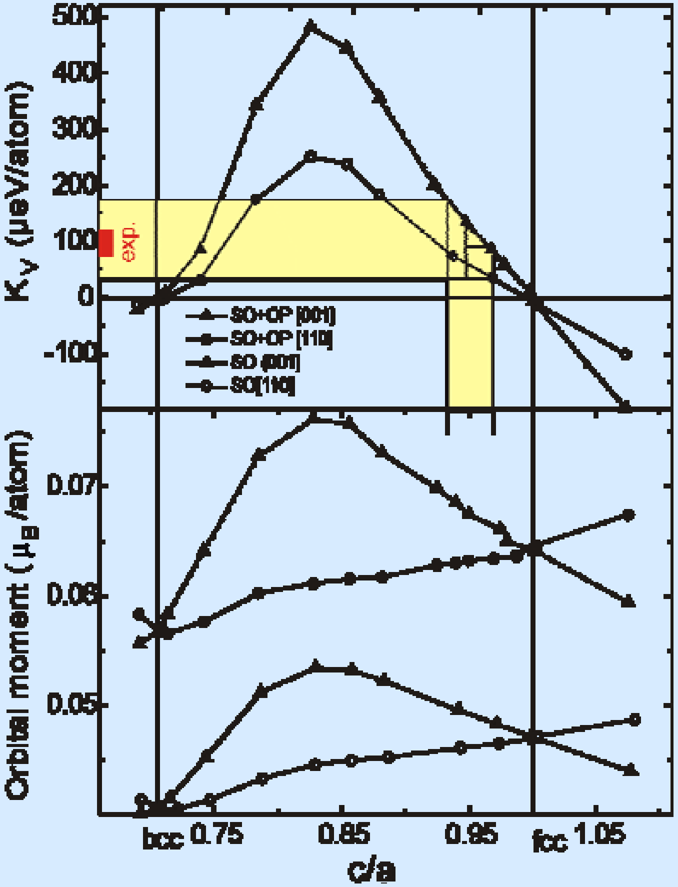




Farle et al. PRB **56**, 5100 (1997) #185

SRT is caused by the temperature dependence of K_i (T).

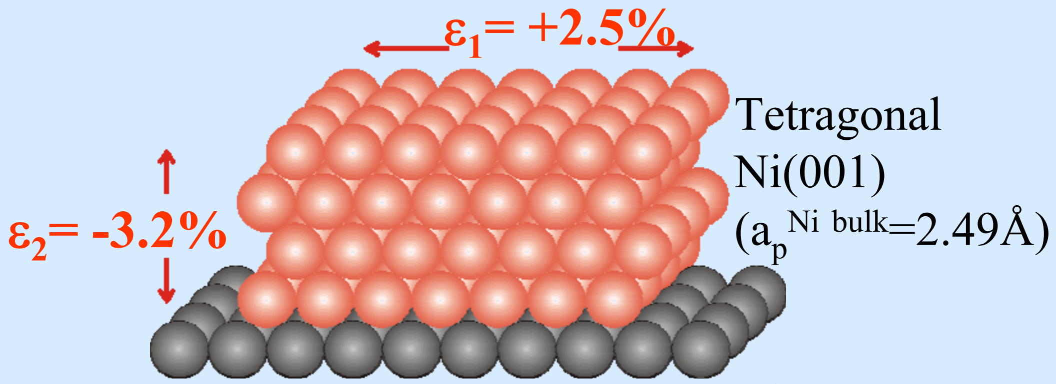
Hypothetical structure in theory



Structural changes by ≈ 0.05 Å increase MAE by 2-3 orders of magnitude (~0.2→100μeV/atom)

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

Growth of artificial nanostructures
bcc, fcc → tetragonal, trigonal

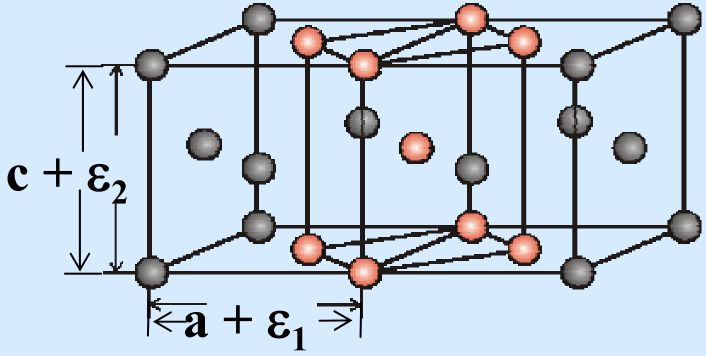


$\varepsilon_1 = +2.5\%$

$\varepsilon_2 = -3.2\%$

Tetragonal
Ni(001)
($a_p^{\text{Ni bulk}}$=2.49Å)

fcc Cu(001) substrate (a_p=2.55Å)



$c + \varepsilon_2$

$a + \varepsilon_1$

Bain path

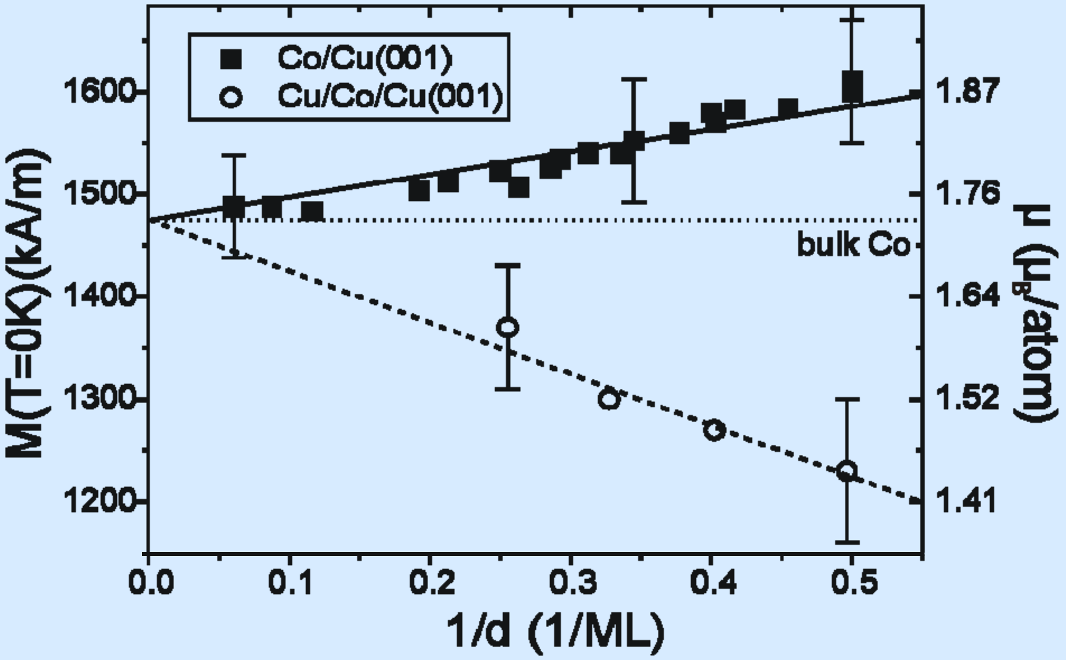
c/a=1 fcc → fct → bcc c/a=$1/\sqrt{2}$

Manipulation of SRT

Magnetic moments at surface/interface

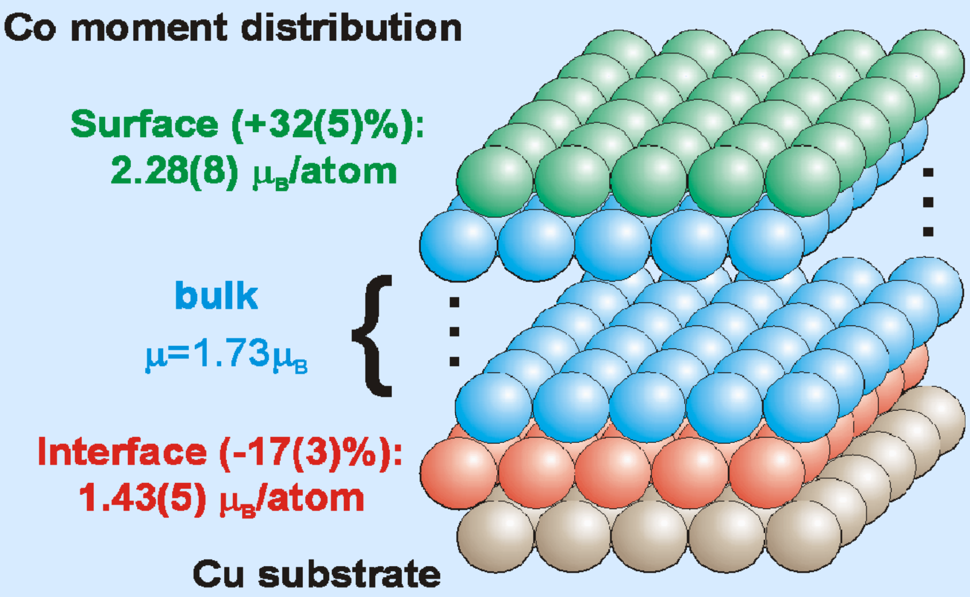
UHV-SQUID measurements

$$m_{tot} = m_{vol} + \frac{m_{surf} + m_{inter} - 2}{d}$$ (linear with 1/d)



Theory:

Hjortstam et al., PRB **53**, 9204 (1996)
Pentcheva et al., PRB **61**, 2211 (2000)



Co moment distribution

Surface (+32(5)%):
2.28(8) μ_B/atom

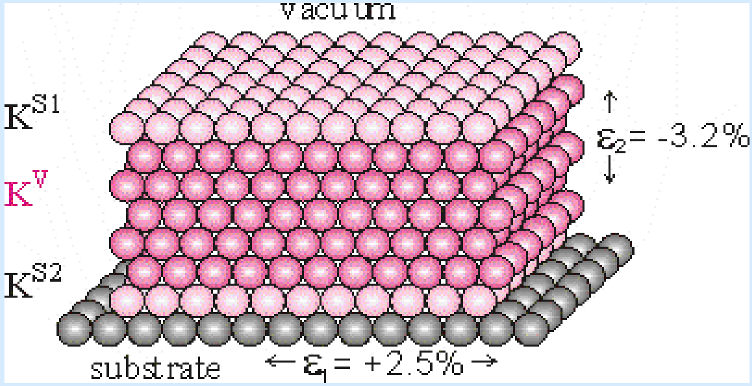
bulk
μ=1.73μ_B

Interface (-17(3)%):
1.43(5) μ_B/atom

Cu substrate

A. Ney et al.
Europhys. Lett. **54**, 820 (2001)

Ni/Cu(001)

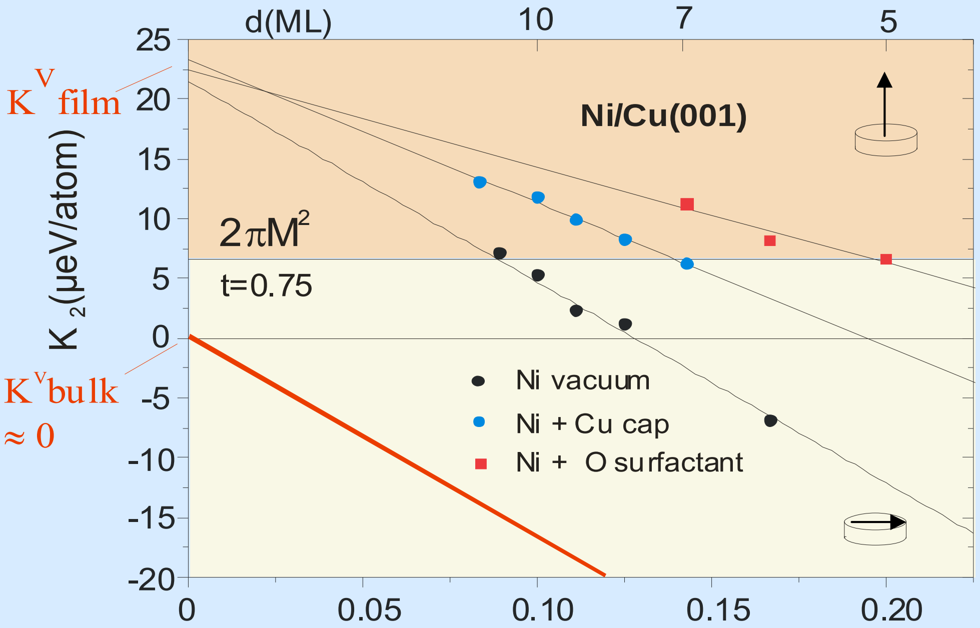


$$F \sim \left(2\pi M^2 - K_{2\perp}\right)\cos^2\theta$$

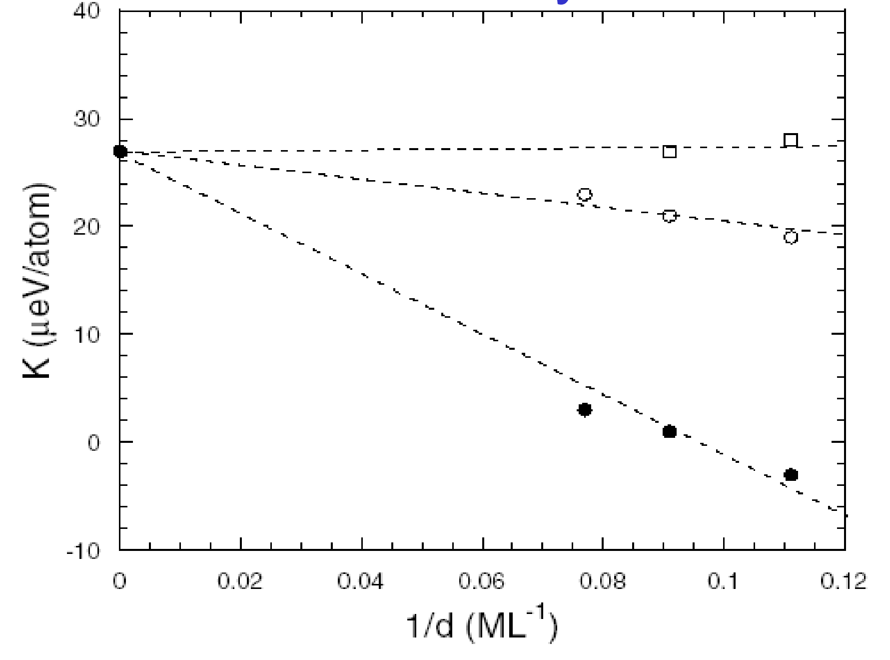
$$K = K^V + \frac{K^{S1} + K^{S2}}{d}$$

Interface	K_S (μeV/atom)	d_C (ML)
Ni/vacuum	-107	10.8
Ni/Cu	-59	7.6
Ni/CO (van Dijken et al.)	-81	7.3
Ni/H_2 (van Dijken et al.)	-70	6.8
Ni/O (surfactant)	-17	4.9

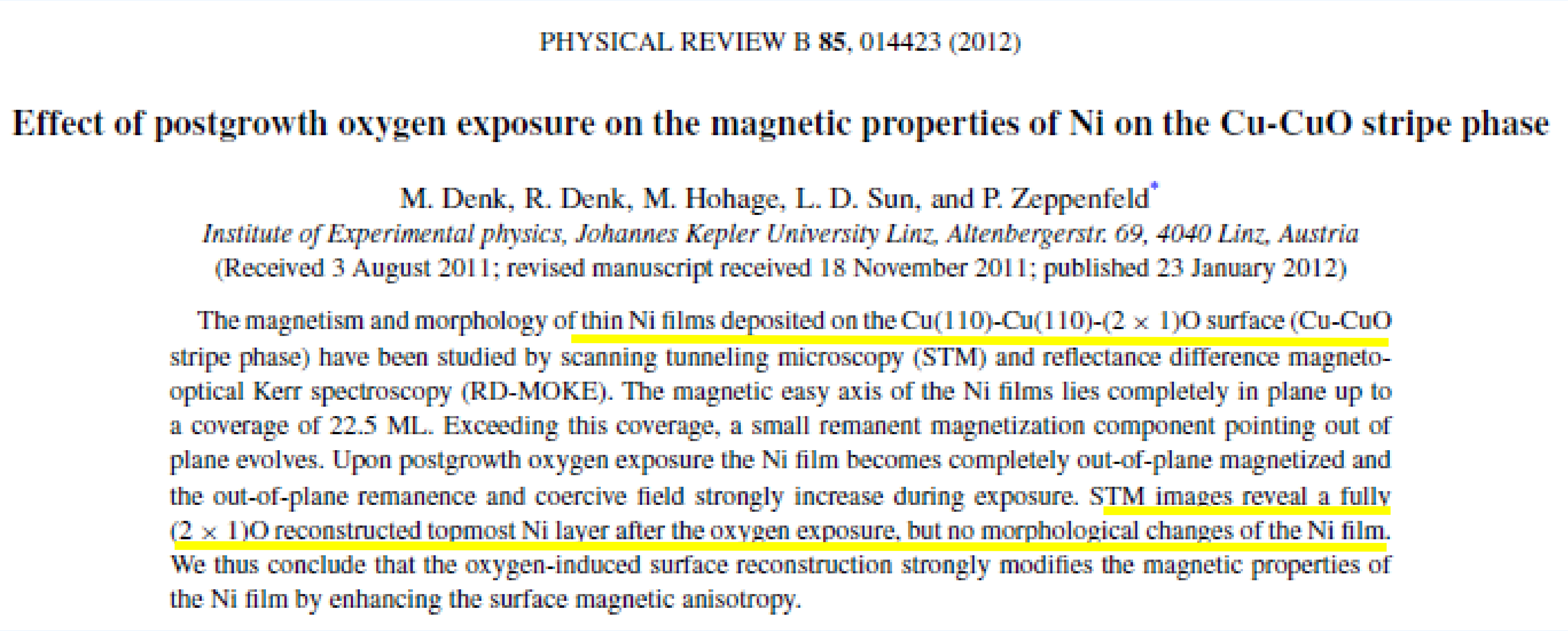
Experiment



Theory



Jisang Hong et al., *Phys. Rev. Lett.* **92**, 147202-1 (2004)



PHYSICAL REVIEW B **85**, 014423 (2012)

Effect of postgrowth oxygen exposure on the magnetic properties of Ni on the Cu-CuO stripe phase

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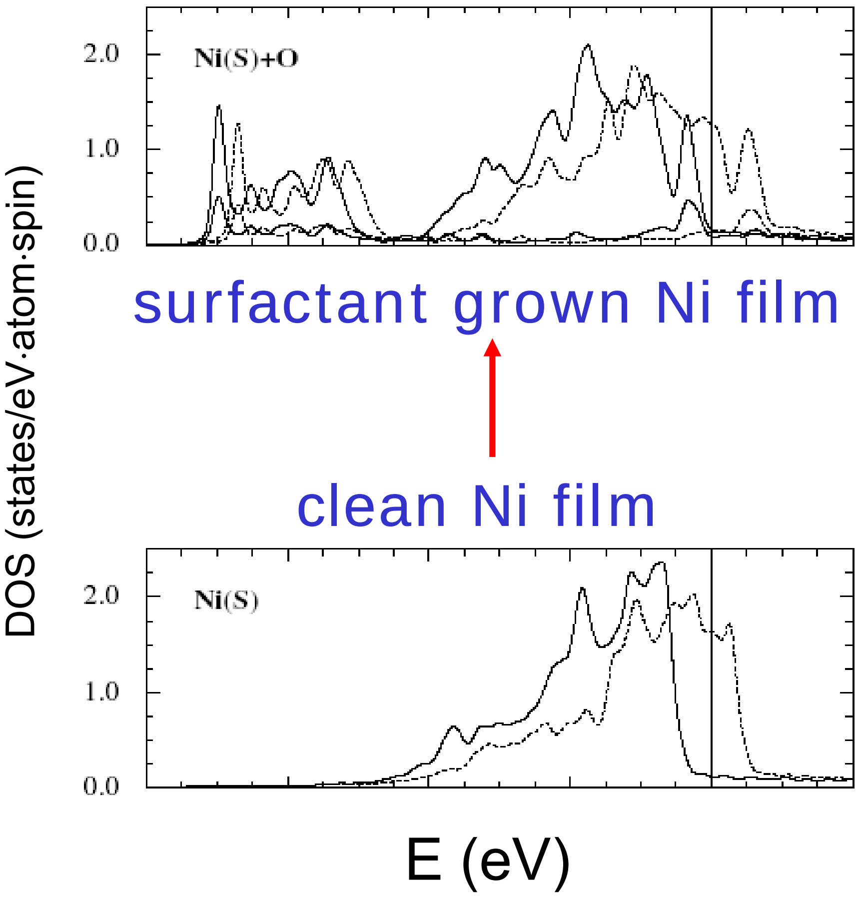
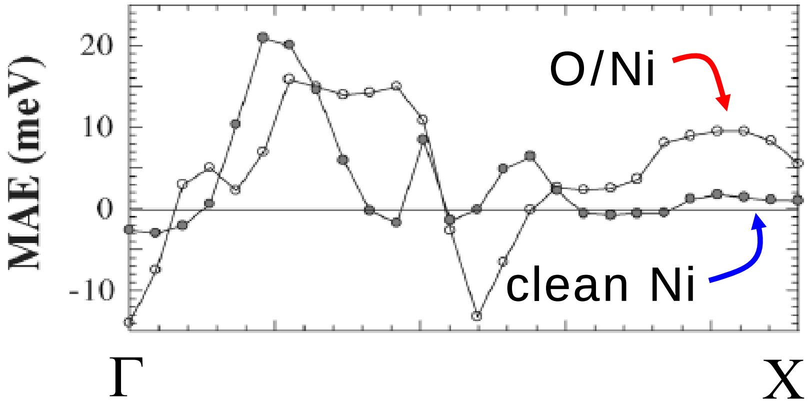
The magnetism and morphology of thin Ni films deposited on the Cu(110)-Cu(110)-(2 × 1)O surface (Cu-CuO stripe phase) have been studied by scanning tunneling microscopy (STM) and reflectance difference magneto-optical Kerr spectroscopy (RD-MOKE). The magnetic easy axis of the Ni films lies completely in plane up to a coverage of 22.5 ML. Exceeding this coverage, a small remanent magnetization component pointing out of plane evolves. Upon postgrowth oxygen exposure the Ni film becomes completely out-of-plane magnetized and the out-of-plane remanence and coercive field strongly increase during exposure. STM images reveal a fully (2 × 1)O reconstructed topmost Ni layer after the oxygen exposure, but no morphological changes of the Ni film. We thus conclude that the oxygen-induced surface reconstruction strongly modifies the magnetic properties of the Ni film by enhancing the surface magnetic anisotropy.

Results of ab initio calculations for O/Ni/Cu(001)

Density of states



MAE along $\overline{\Gamma}\overline{X}$ axis

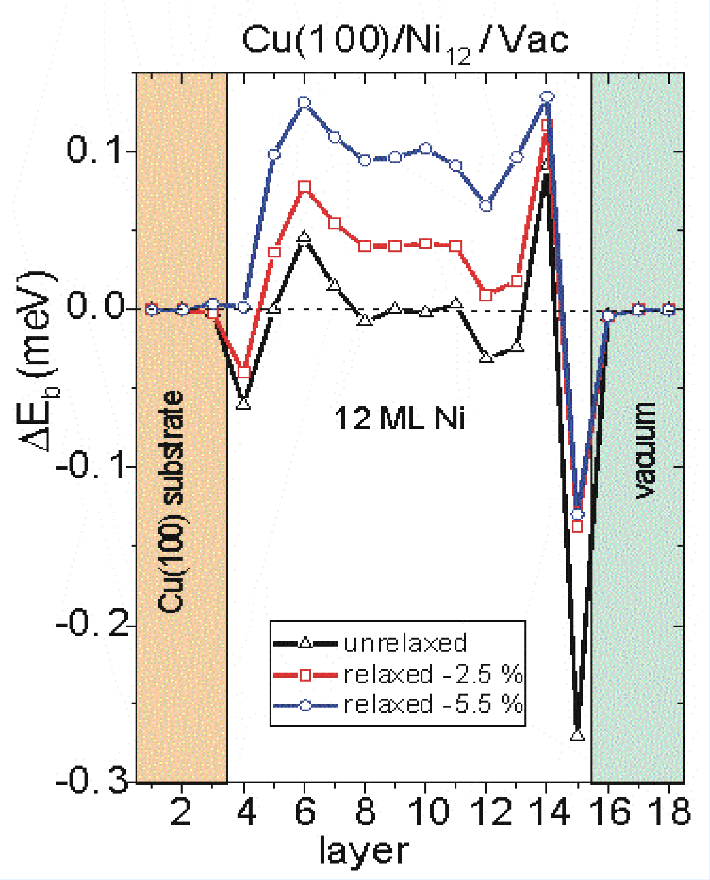


- DOS shows that topmost Ni moment is basically unchanged
- O-induced surface state seen in the vicinity of X-point is responsible for change in MAE
- theory reveals induced moment in surfactant oxygen

Jisang Hong et al., *Phys. Rev. Lett.* **92**, 147202-1 (2004)

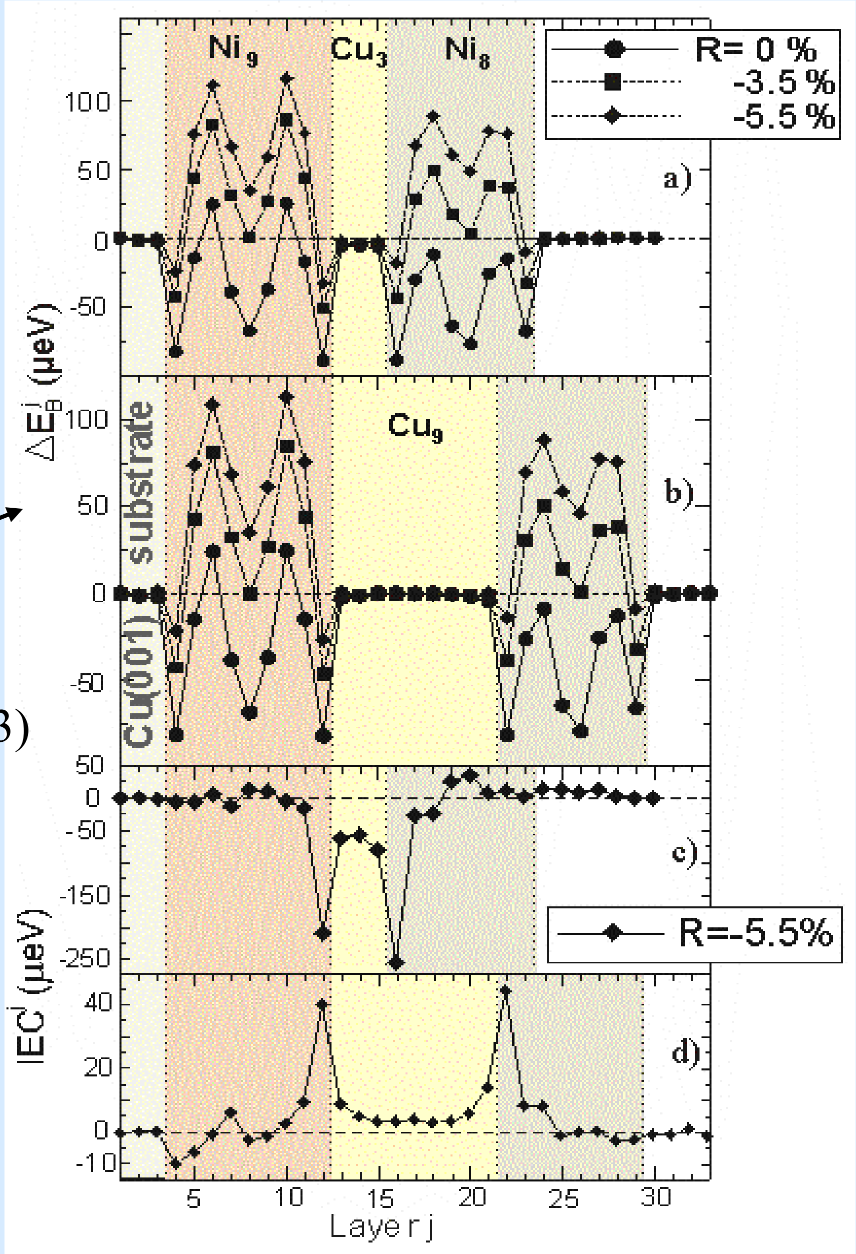
SP-KKR calculation for rigit fcc and relaxed fct structures

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 nano-structure will overcome this, because they count for n-2 layers.

Summary

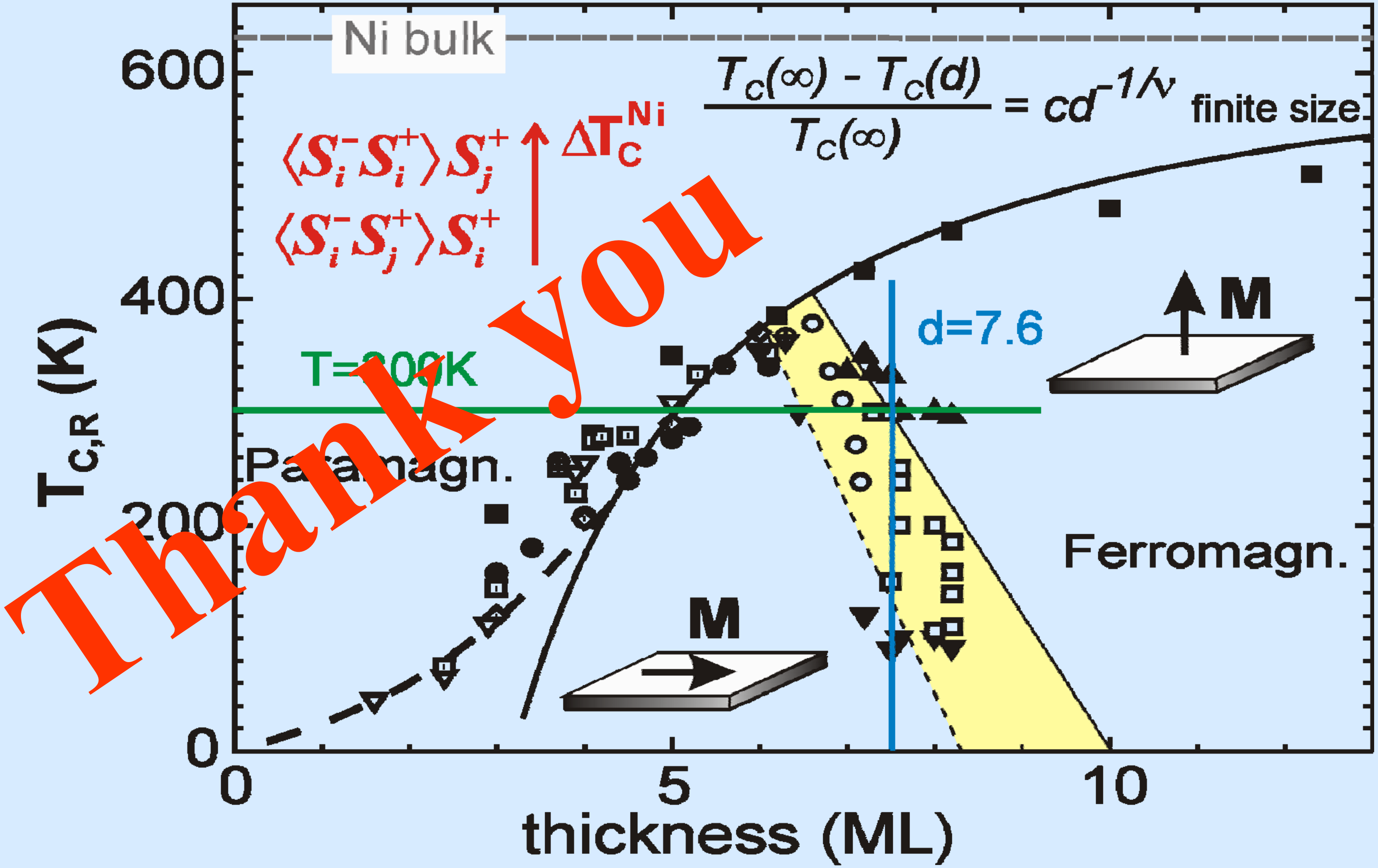
A whole variety of experiments on nanoscale magnets are available nowadays. Unfortunately many of the data are analyzed using theoretical *static mean field (MF) model*, also textbook for *Untrathin Magnetic Structures* use the bulk magnetism approach.
Such a mean field ansatz is insufficient for nanoscale magnetism, we demonstrate the importance of *higher order spin-spin correlations* in low dimensional magnets.

The spin is not a good quantum number, it ignores the orbital magnetism.
The orbital magnetic moment creates the MAE.
And the MAE manipulates the SRT.

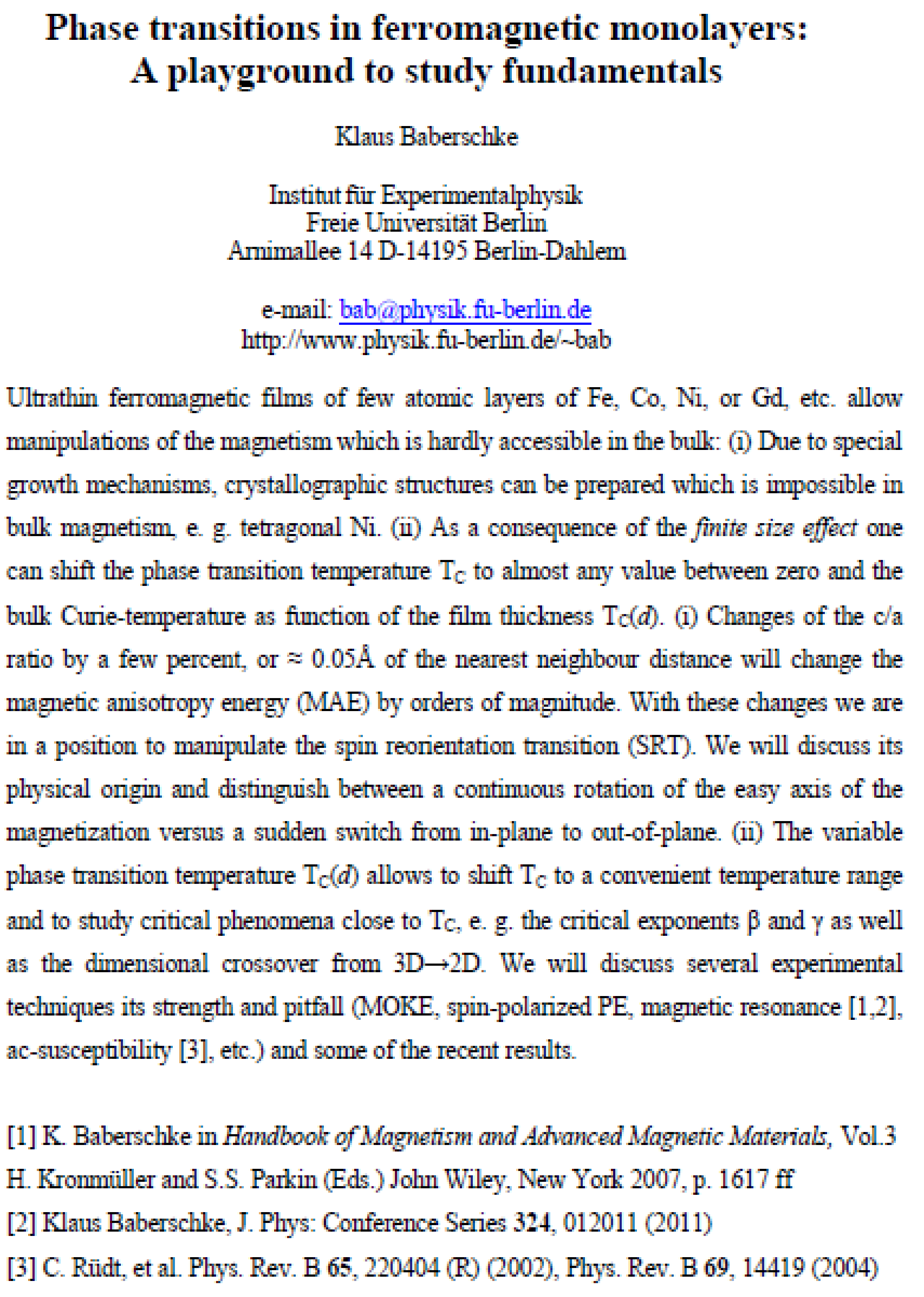
Conclusion:

The magnetism of FM monolayers is a very reach playground and a fruitful collaboration between theory and experiment.

Take Home Cartoon



http://www.physik.fu-berlin.de/~bab (#xyz see publ.list)



Phase transitions in ferromagnetic monolayers: A playground to study fundamentals

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Ultrathin ferromagnetic films of few atomic layers of Fe, Co, Ni, or Gd, etc. allow manipulations of the magnetism which is hardly accessible in the bulk: (i) Due to special growth mechanisms, crystallographic structures can be prepared which is impossible in bulk magnetism, e. g. tetragonal Ni. (ii) As a consequence of the *finite size effect* one can shift the phase transition temperature T_C to almost any value between zero and the bulk Curie-temperature as function of the film thickness $T_C(d)$. (i) Changes of the c/a ratio by a few percent, or ≈ 0.05Å of the nearest neighbour distance will change the magnetic anisotropy energy (MAE) by orders of magnitude. With these changes we are in a position to manipulate the spin reorientation transition (SRT). We will discuss its physical origin and distinguish between a continuous rotation of the easy axis of the magnetization versus a sudden switch from in-plane to out-of-plane. (ii) The variable phase transition temperature $T_C(d)$ allows to shift T_C to a convenient temperature range and to study critical phenomena close to T_C, e. g. the critical exponents β and γ as well as the dimensional crossover from 3D→2D. We will discuss several experimental techniques its strength and pitfall (MOKE, spin-polarized PE, magnetic resonance [1,2], ac-susceptibility [3], etc.) and some of the recent results.

[1] K. Baberschke in *Handbook of Magnetism and Advanced Magnetic Materials*, Vol.3 H. Kronmüller and S.S. Parkin (Eds.) John Wiley, New York 2007, p. 1617 ff

[2] Klaus Baberschke, J. Phys: Conference Series **324**, 012011 (2011)

[3] C. Rüdt, et al. Phys. Rev. B **65**, 220404 (R) (2002), Phys. Rev. B **69**, 14419 (2004)