PHASE TRANSITIONS IN FERROMAGNETIC MONOLAYERS?

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Lecture 1

1. Para- to ferromagnetic phase transition, Curie temperature, finite size scaling, reduced temperature $T/T_C = t$.
2. Spin reorientation transition (SRT). Is it a phase transition at all? Continuous or discrete?
3. FM trilayers, do they have 2 Curie temperatures? Importance of higher order spin-spin correlations.

Lecture 2

4. Curie - Weiss susceptibility, χ_{ac} measured by means of mutual inductance, MOKE, and XMCD.
5. How do we determine T_C and the critical exponents?
6. Non linear χ_{ac}, the interpretation of higher harmonics in χ_{ac}.

Prolog

In these two lectures we will focus on the physics of single domain magnetism. For many ultrathin films a single domain is the thermodynamic ground state (see textbooks e. g. Chikazumi), not necessarily in *status nascendi.*

For those films the concept of phase transitions, critical phenomena in two dimensions will be lectured. This is useful to be discussed, before analyzing multi-domain states and domain-wall features. Results of various experimental methods (like SQUID, χ_{ac}, FMR, MOKE, XMCD) will be used. But the focus lies on the measured observables and the concept, less on the details of the technique, itself.

Most of the examples used here, mainly from our own work, will elucidate the importance and novelty of magnetism in low dimensions. Unfortunately beautiful experiments in ultrathin films are quite oven interpreted using simple models (like MFA, etc.) which are good for bulk magnetism but insufficient for 1D or 2D.

note: citation with #xyz gives reference to our publ. list (web)

Former Group Members

BESSY-crew: H. Wende, C. Sorg, A. Scherz, F. Wilhelm

Lab. experiments: J. Lindner, K. Lenz, C. Rüdt, R. Nünthel, E. Kosubek

Support:
BMBF (BESSY), DFG (lab.)

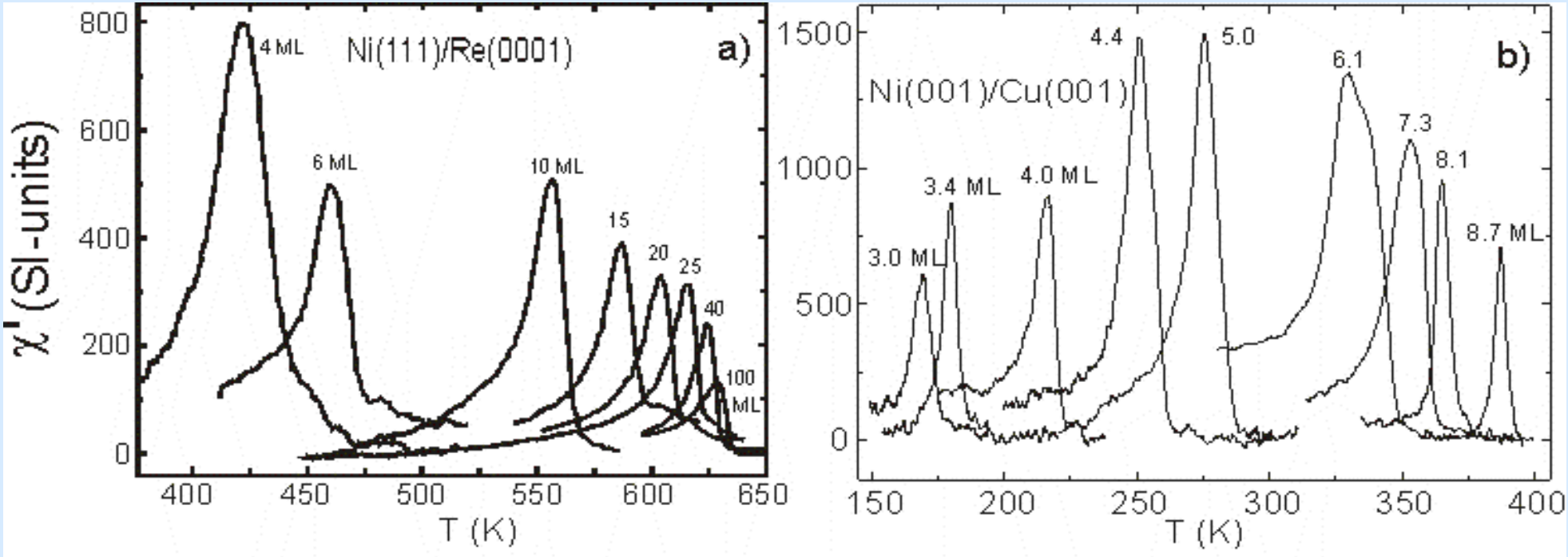
1. Para- to ferromagnetic phase transition, Curie temperature, finite size scaling, reduced temperature $T/T_C = t$.

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

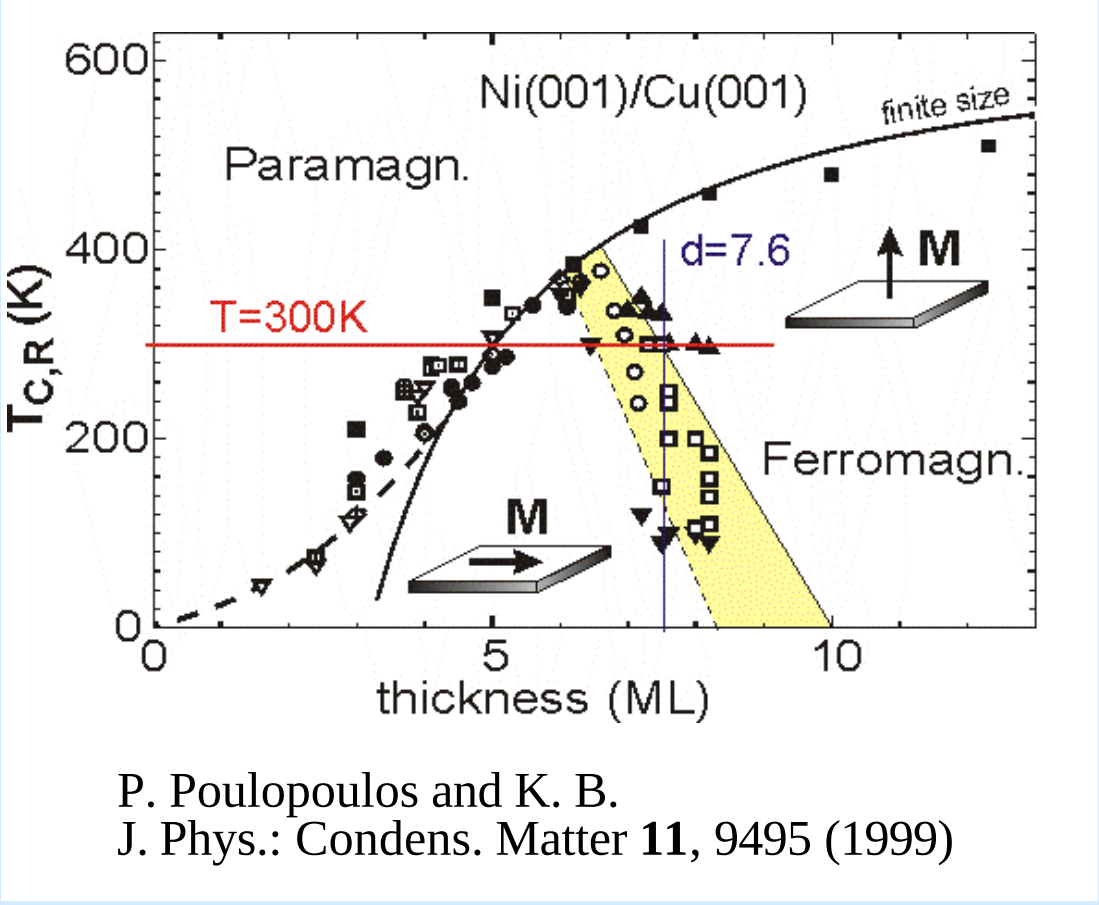
For ultrathin films T_C can be manipulated from zero to T_C^{bulk}.

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)



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

Finite size scaling

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

Reduced temperature

$$1-t = cd^{-1/n}$$

$\nu = 0.705$

critical exponent for correlation length

Finite size scaling is an asymptotic solution for d => 8 .
It does not work for d = 1,2,3, … MLs

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

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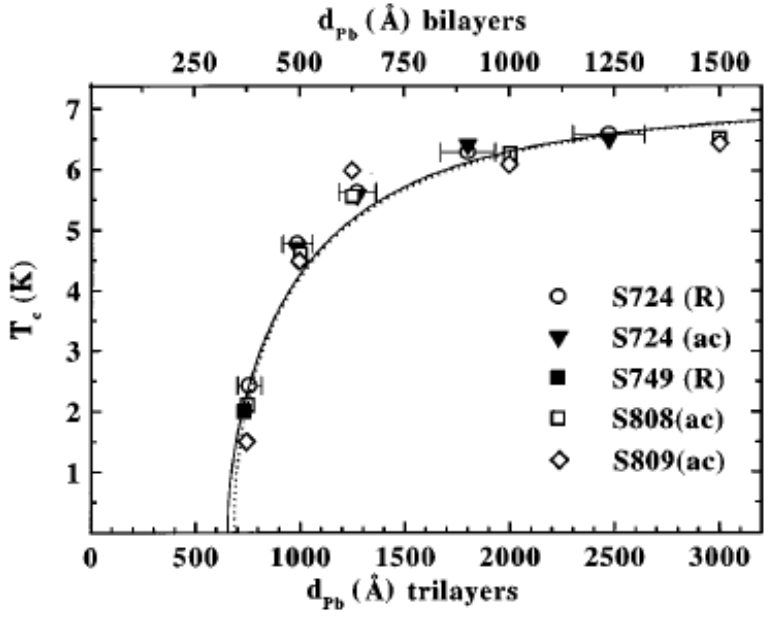


FIG. 7. Dependence of the superconducting transition temperature on the thickness of the Pb-layer for four sample series listed in Table I. The dotted and solid lines are the best fits using the theory by Radović *et al.* and the theory by Tagirov, respectively, with parameters given in the figure subscripts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

$$x = x_0 (1-t)^{-n}$$

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

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

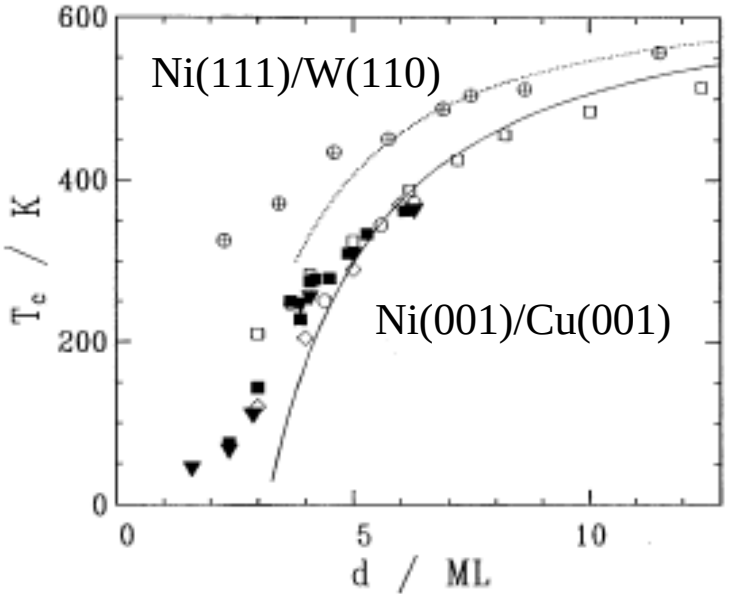


Fig. 7. Critical temperature T_c for: Ni (111)/W (110) ⊕ [6], and Ni (001)/Cu (001) dc-MCXD (▼), ac-MCXD ■ [15], and □ [14], ○ [26], ◇ [25]. The *solid* and *dotted lines* are fits to (8) with values for c given in the text

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

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

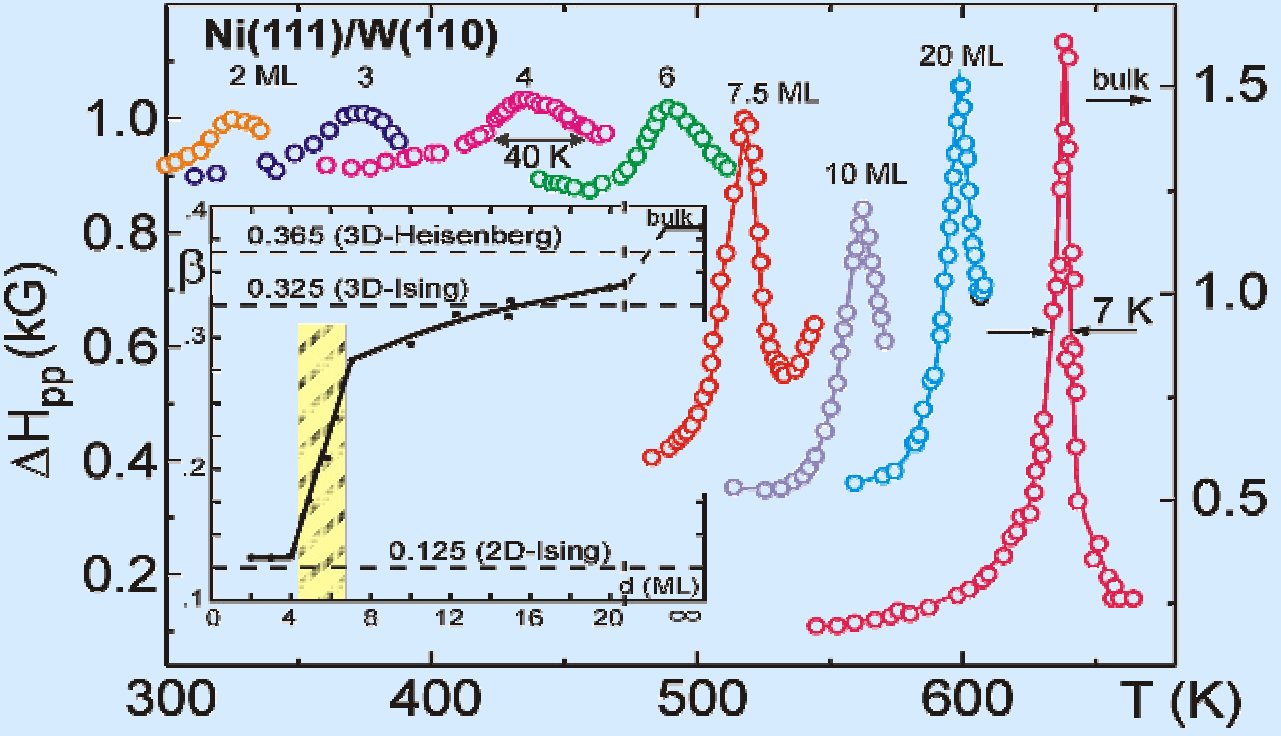
agrees perfectly with:

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

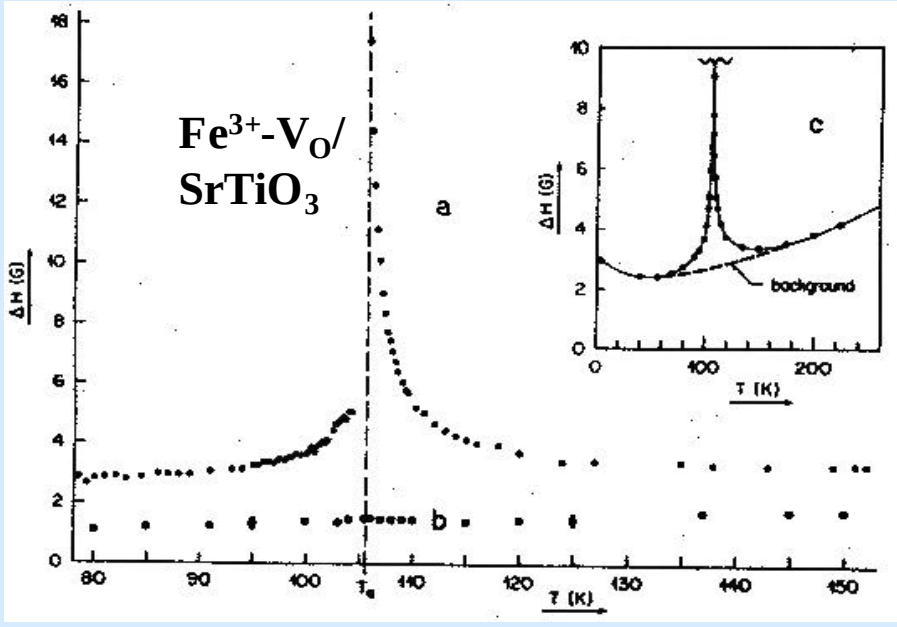
K. B. # 171

Early papers discussed different DOS. We think, it's unlikely.

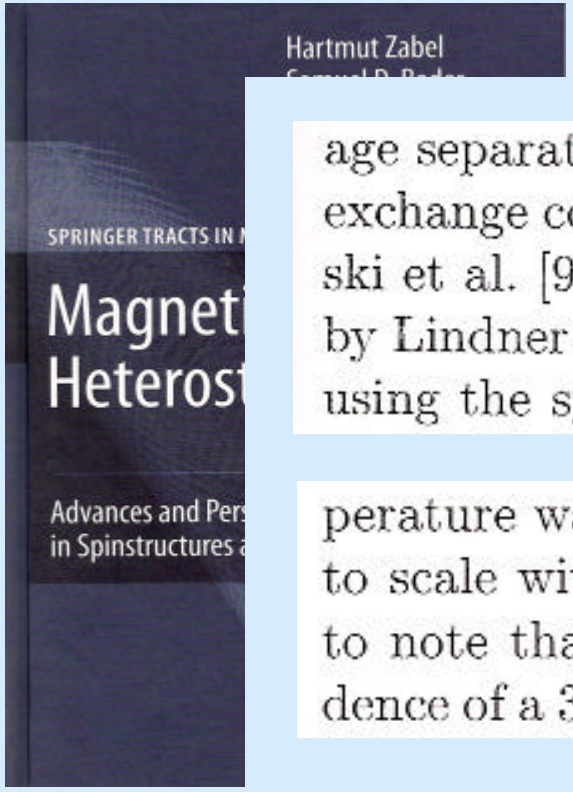
Close to T_C fluctuations increase, be it Gaussian or critical.
This increases the linewidth in magnetic resonance spectroscopy.



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



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



Importance of reduced temperature $t = T/T_C$

age separation between atomic terraces. The temperature dependence of the exchange coupling was measured over the interval from 77 to 400 K by Celinski et al. [95] using the system Ag-substrate/9Fe/10,12bccCu/16Fe(001) and by Lindner and Baberschke [35] over the temperature range from 55 to 350 K using the system fcc Cu-substrate/7Ni/5,9Cu/2Co(001). The integers repre-

perature was reduced from 350 K to 55 K; moreover, this increase was found to scale with an inverse 3/2 power law in temperature [35]. It is interesting to note that in the Ag-substrate/9Fe/10,12bccCu/16Fe(001)samples no evidence of a 3/2 power law was found. In the Cu-substrate/7Ni/5,9Cu/2Co(001)

With the highest T of 400*K* for thicker Fe films, $t = T/T_C$ will be $= 0.4$, only.

We know only about one experiment in which more or less the full range of reduced temperature was used. For 3 different multi- and trilayer systems (Ni/Cu/Co, Ni/Cu/Ni, and an Fe/V multilayer) the reduced temperature covers almost the total range of $0 < t = 0.9$. All 3 cases show an almost perfect power law behavior with a 3/2 exponent. K. B. # 330, Phil. Mag. 2008

2. Spin Reorientation Transition (SRT).

Is it a phase transition at all? Continuous or discrete?

Spin reorientation in bulk Gd

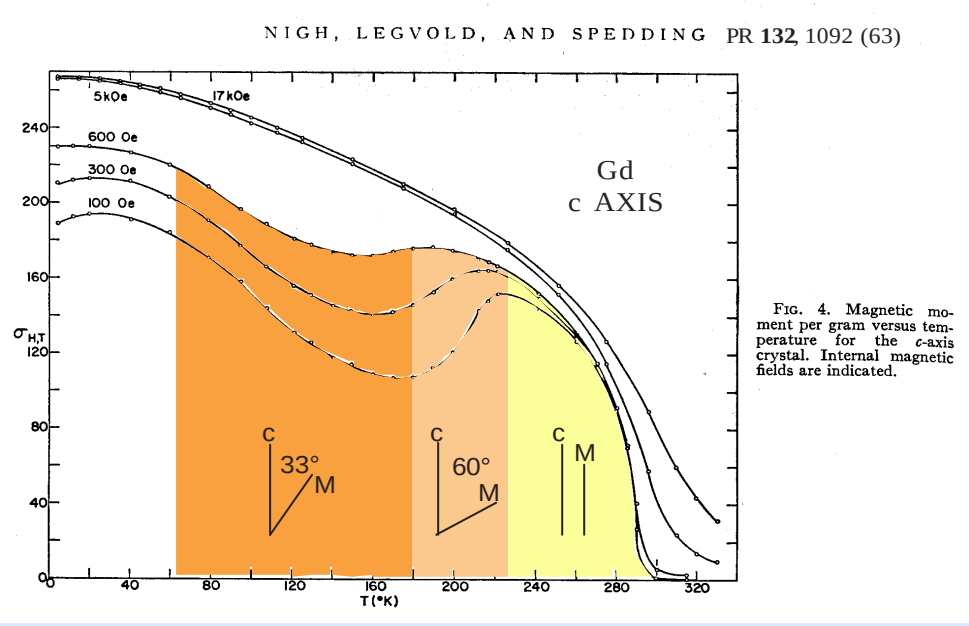


FIG. 4. Magnetic moment per gram versus temperature for the *c*-axis crystal. Internal magnetic fields are indicated.

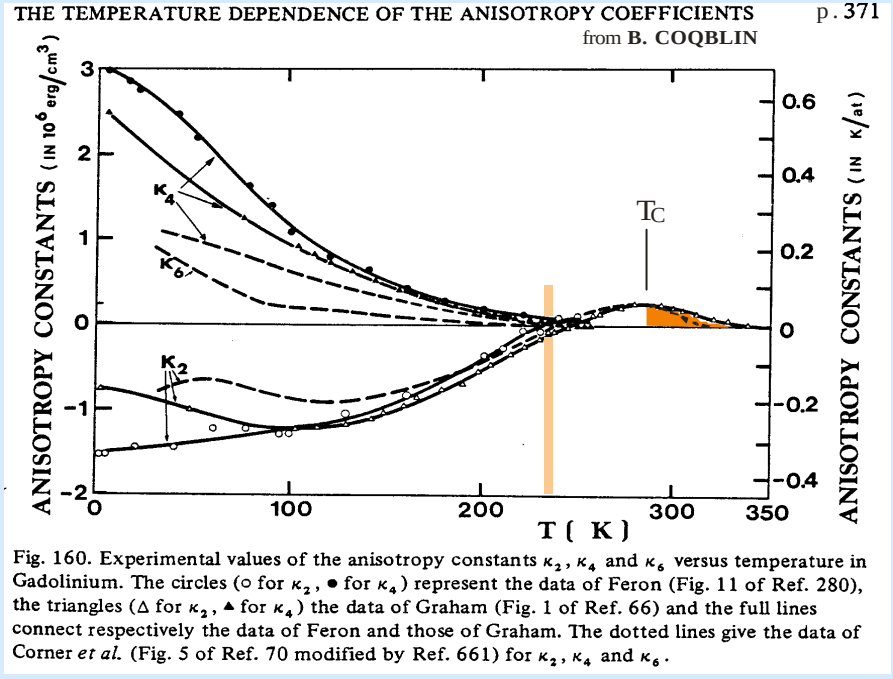


Fig. 160. Experimental values of the anisotropy constants κ_2, κ_4 and κ_6 versus temperature in Gadolinium. The circles (○ for κ_2, ● for κ_4) represent the data of Feron (Fig. 11 of Ref. 280), the triangles (Δ for κ_2, ▲ for κ_4) the data of Graham (Fig. 1 of Ref. 66) and the full lines connect respectively 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.

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

Note also finite MAE above T_C

Spin reorientation in bulk Co und Ni

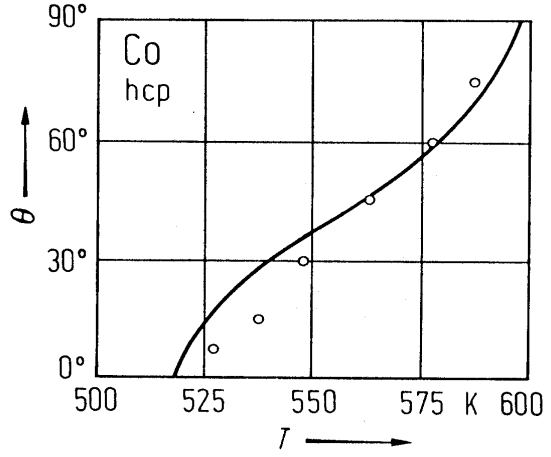


Fig. 5. Temperature dependence of the angle θ between the direction of spontaneous magnetization and the c axis of a single crystal of hcp Co [61 B 5]. Points: data. Curve: calculated from $\sin\theta = (-K_1/2K_2)^{1/2}$.

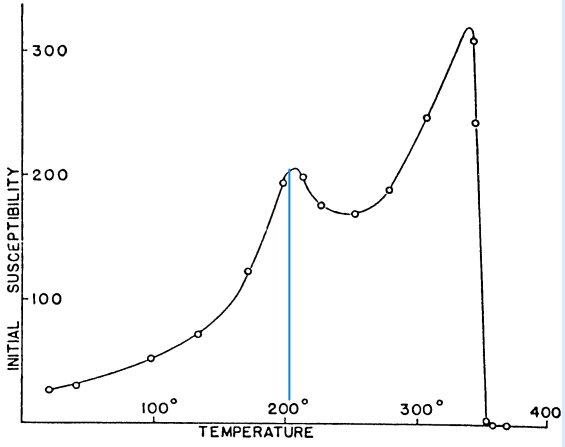


FIG. 6. The variation of the initial susceptibility with temperature in nickel.

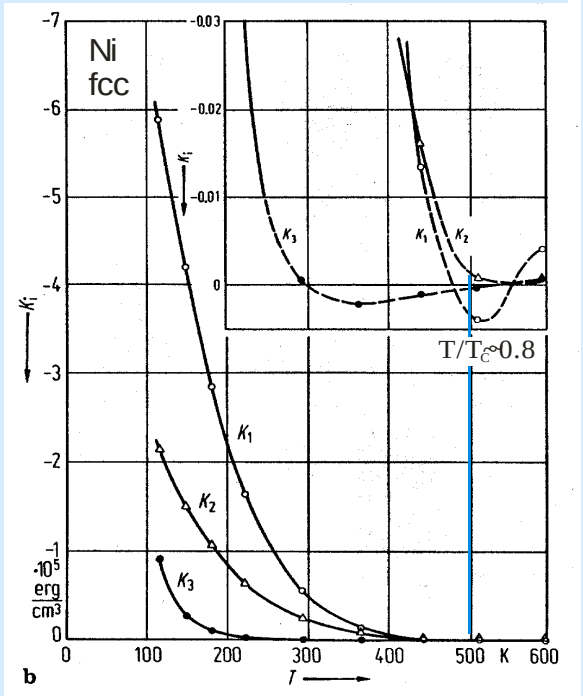


Fig. 7. Temperature dependence of magnetocrystalline anisotropy constants of Ni. (a) K_1. *1*: [68 F 1], *2*: [74 T 1], *3*: [77 B 2], *4*: [77 O 1]. Solid line: calculation [77 O 1]. (b) K_1, K_2, and K_3. Accuracy of data is considerably reduced near T_C: dashed lines in the insert [68 A 1]. (c) K_3. *1* and *2*: [76 A 1], *3*: [69 F 2], *4*: [77 B 2]. Solid line is to guide the eye through confidence limits [76 A 1].

χ_{Ni} and T in Celsius

LB III, 19a, p.45

SRT for hcp Co

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

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

Free energy density of MAE, K

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

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

$E_{tetr} = -K_2 a_z^2 - {}^1/_2 K_{4\wedge} a_z^4 - {}^1/_2 K_{4\div} (a_x^4 + a_y^4) + \ldots$ *(B.Heinrich et al.)*

$= -K_2 \cos^2\theta - {}^1/_2 K_{4\wedge} \cos^4\theta - {}^1/_2 K_{4\div} {}^1/_4 (3+\cos 4\varphi) \sin^4\theta + \ldots$ *(Bab et al.)*

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

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

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

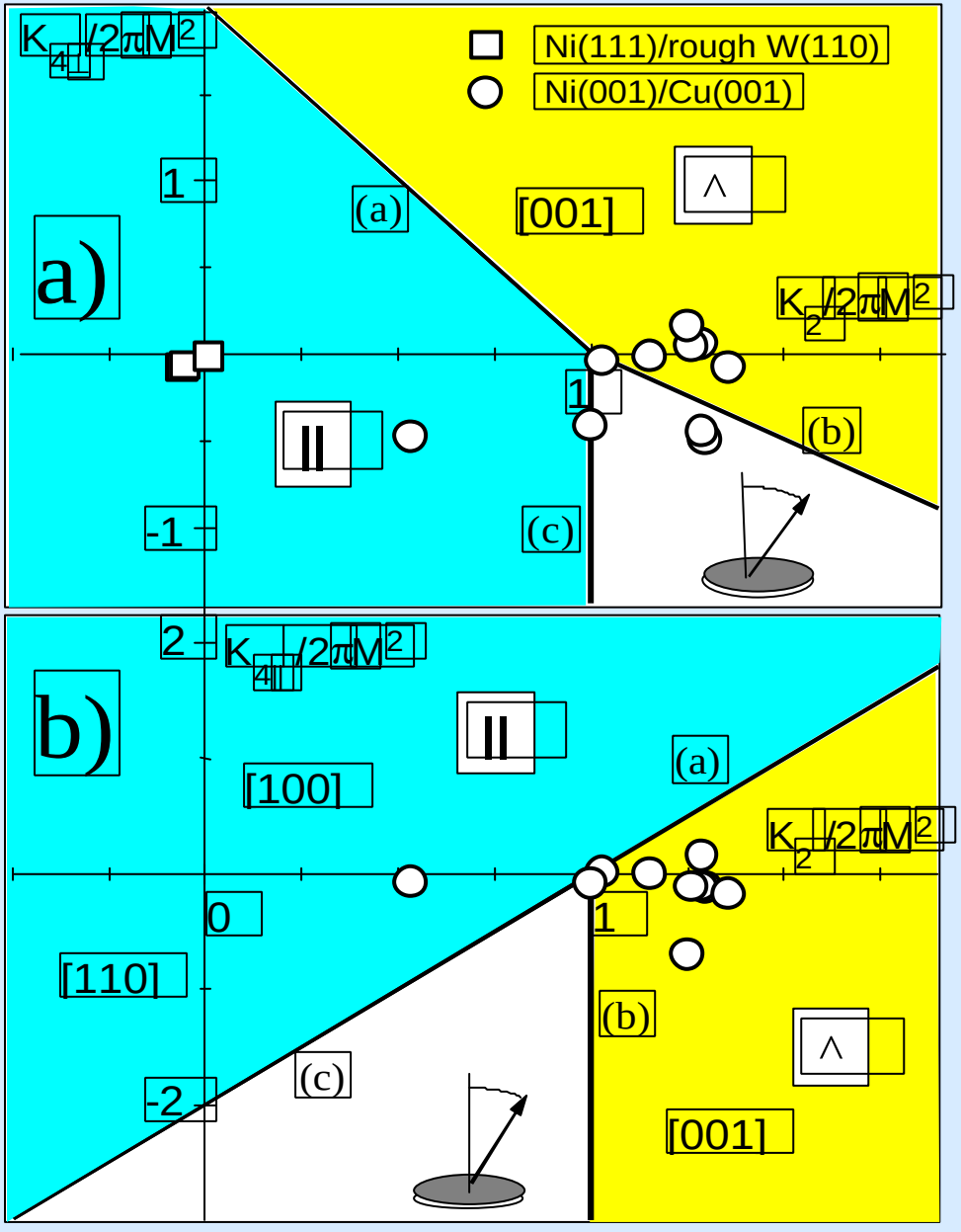
$E_{hex} = k_2 \sin^2\theta + {}^1/_2 k_{2\div} \cos 2\varphi \sin^2\theta + k_4 \sin^4\theta + k_{6\wedge} \sin^6\theta + k_{6\div} \cos 6\varphi \sin^6\theta + \ldots$

$K = k_2 Y_2^0 + k_{4m} Y_4^m + \ldots$ *Legendre polyn. (B. Coqblin)*

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) and K. B. Handb. Magn. Magn. Mat. **3,** 1617 (2007) # see web



A. Berghaus, M. Farle, Yi Li, K. B.
Absolute determ. of the mag. anisotropy of ultrathin Gd and Ni/W(110).
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

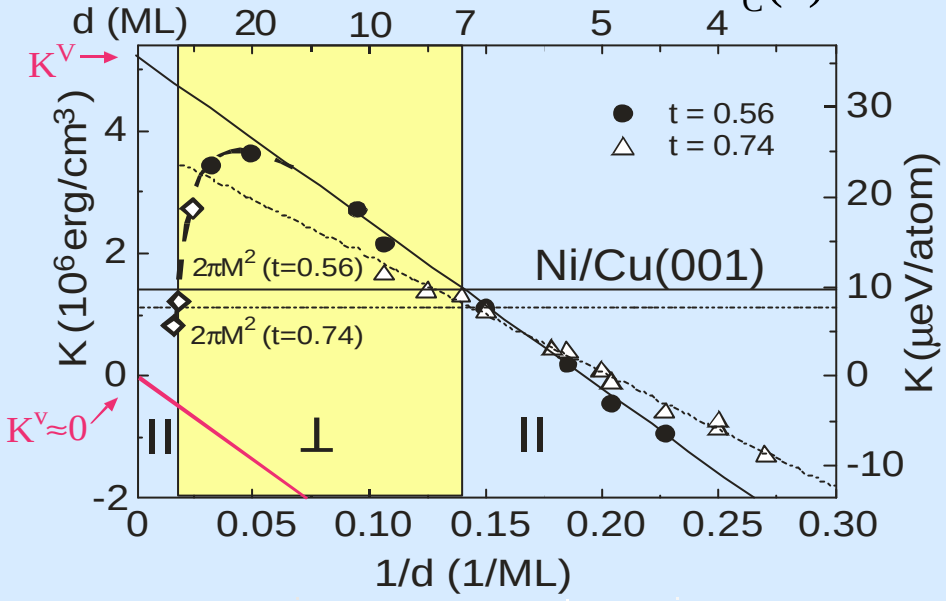
Only with $K_4 \neq 0$ a continues SRT is possible!

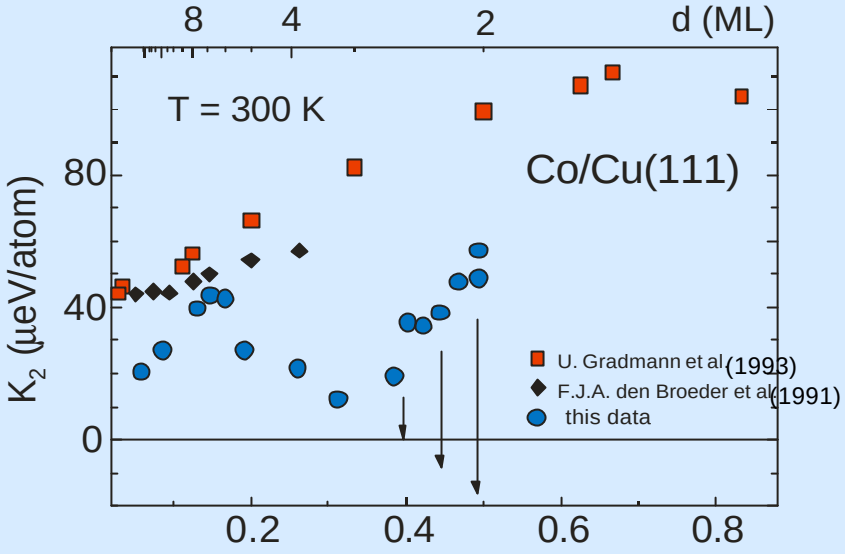
$$\sin\theta = (K_2/2K_4)^{1/2}$$

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

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

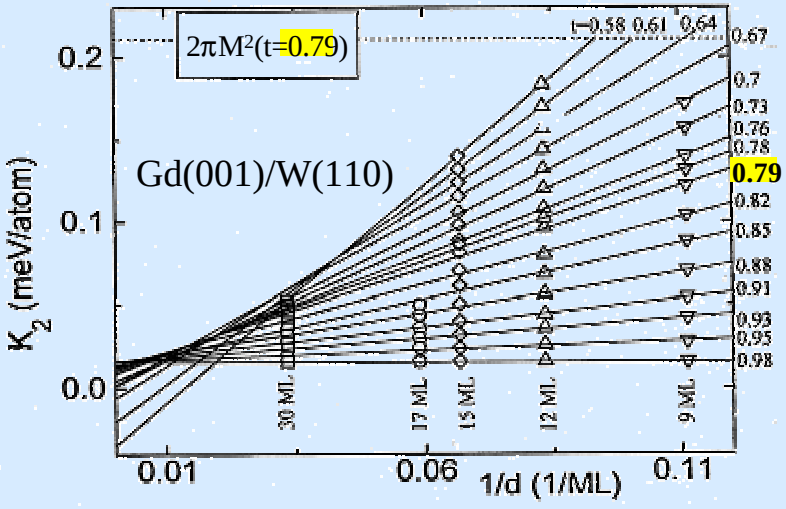
$T/T_C(d)$ and $K(T)=K_V(T)+2K_S(T)/d$



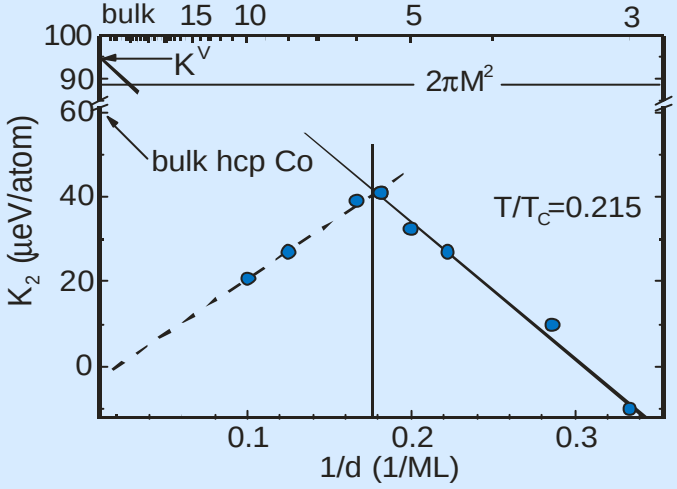


Why does Ni undergo a SRT and Co not ?

=> #330

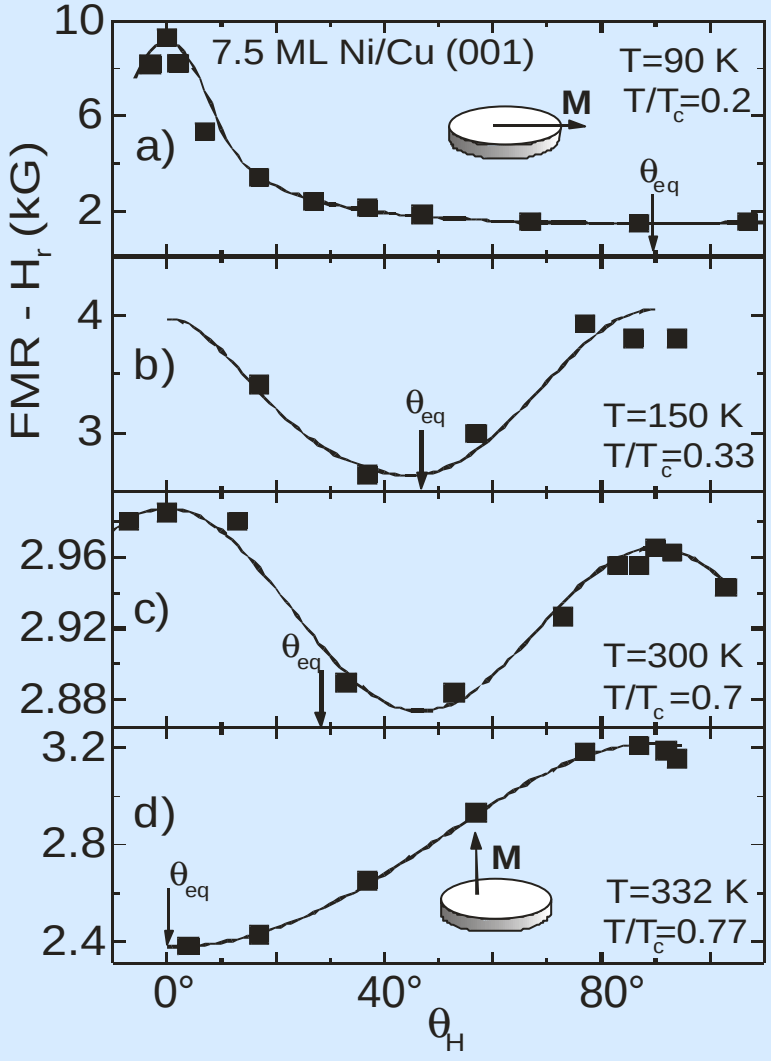


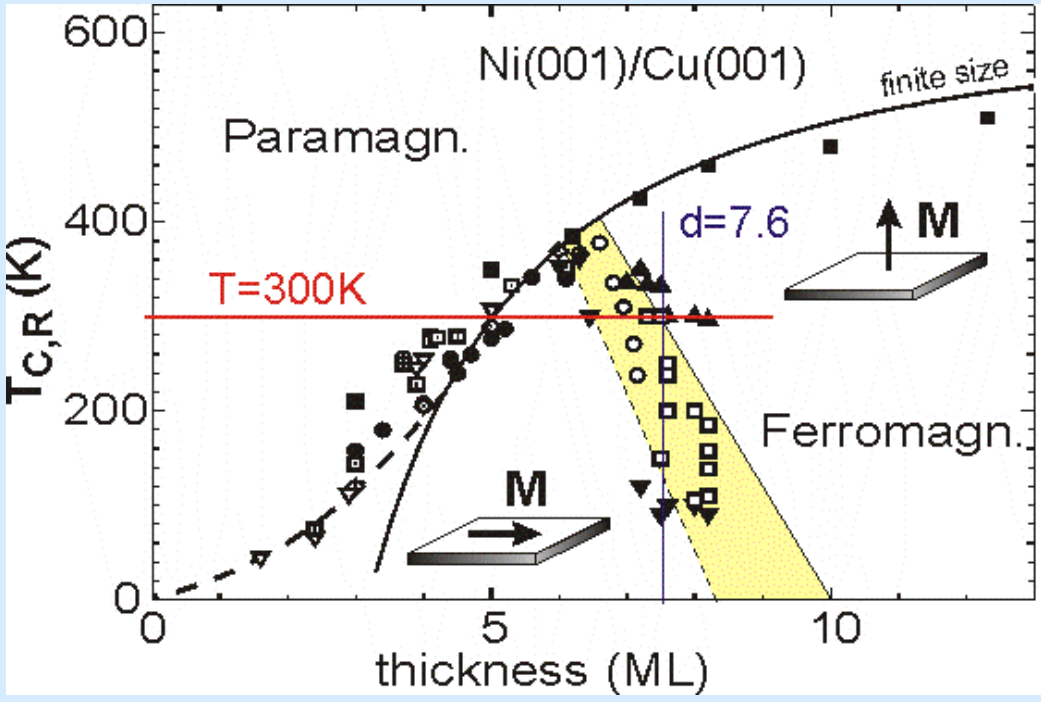
G. André et al., Surface Science **326**, 275 (1995)
K. B. and M. Farle, J. Appl. Phys. **81**, 5038 (1997)



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.



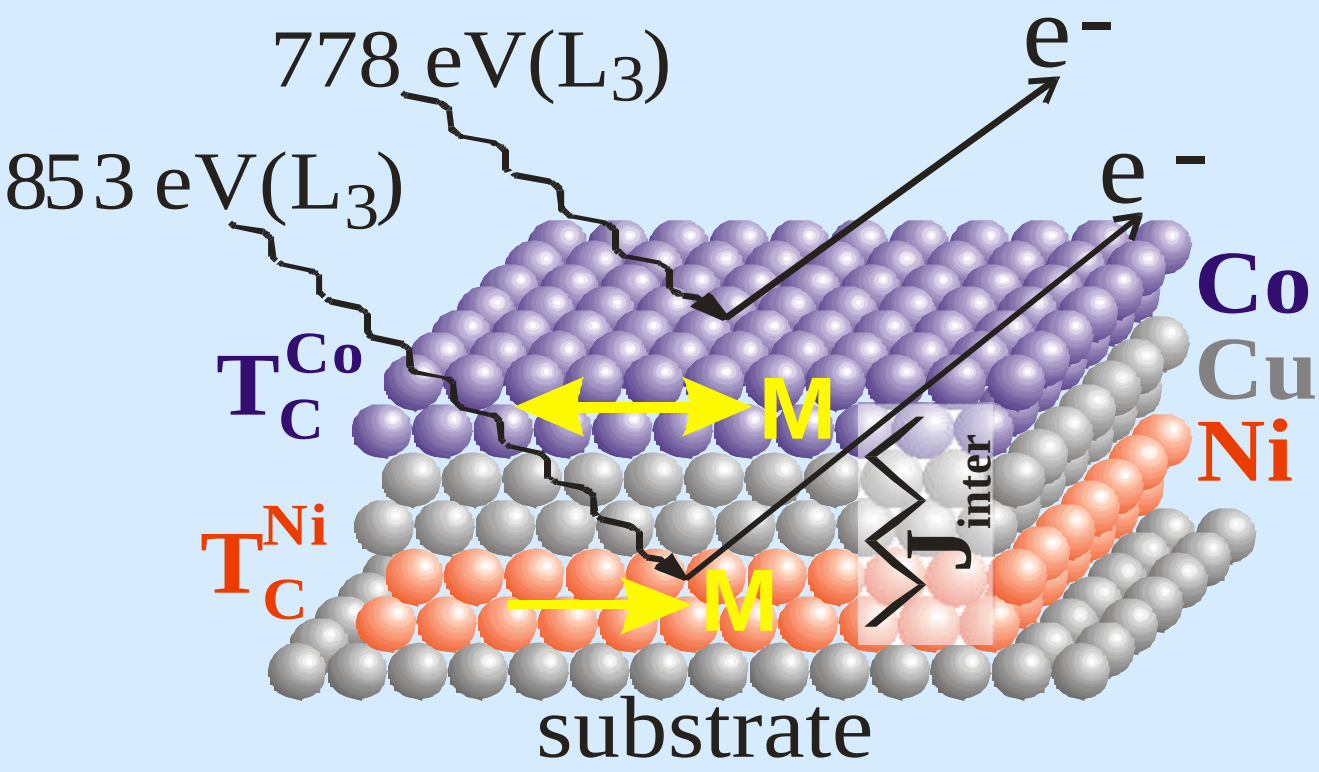


Farle et al. PRB **56**, 5100 (1997) #185

SRT can be a discrete or continuous rotation of the easy axis.
It is caused by the temperature dependence of $K_i(T)$.
I would not call it a "phase transition of first/second order".

3. FM trilayers, do they have 2 Curie temperatures?

Importance of higher order spin-spin correlations.



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

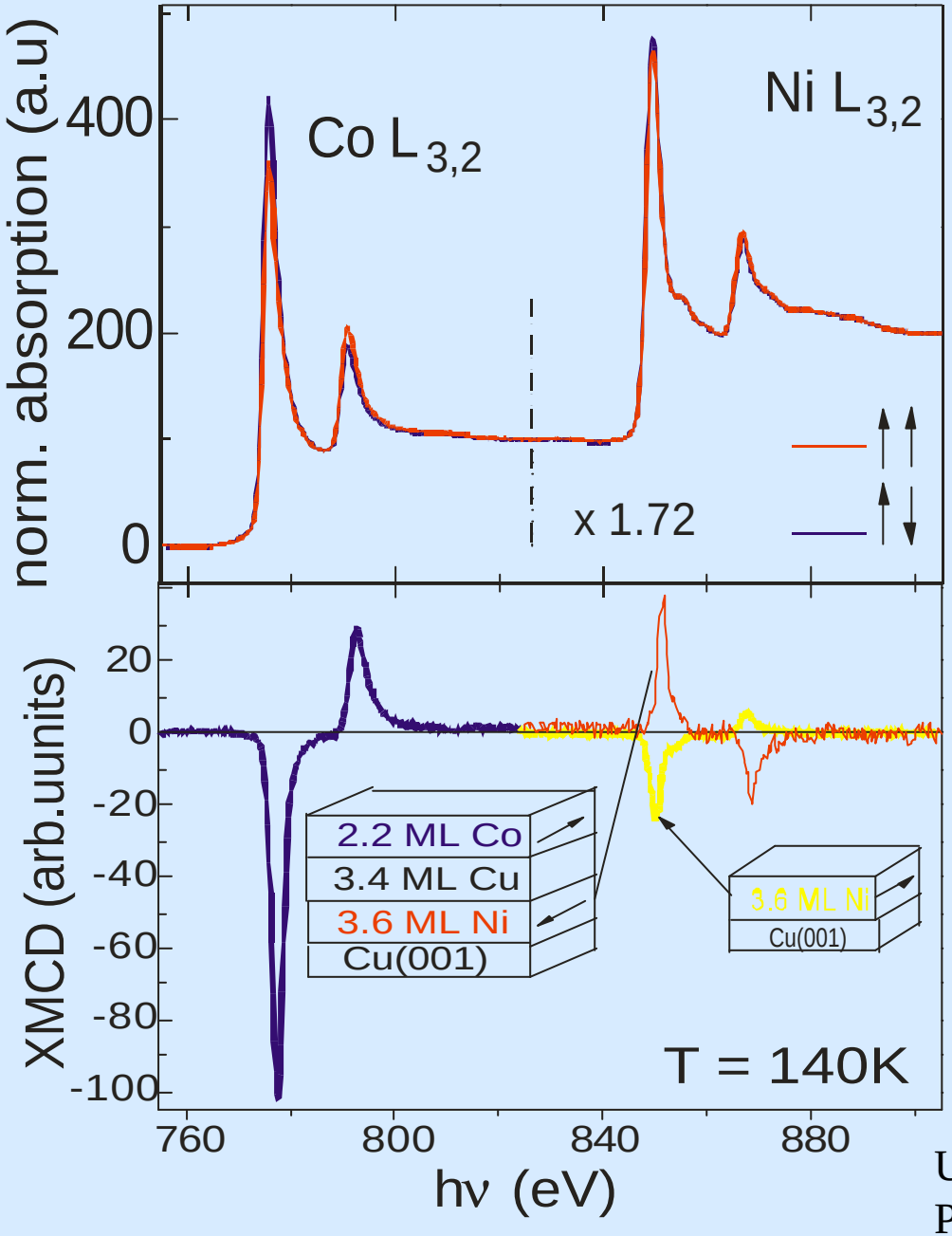
What about element specific Curie-temperatures ?

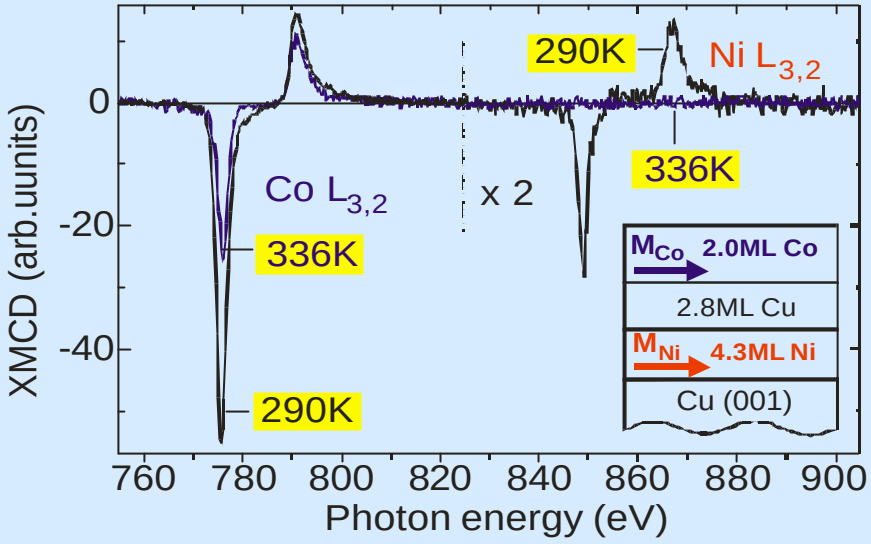
Two trivial limits: (i) $d_{Cu} = 0 \Rightarrow$ direct coupling like a Ni-Co alloy

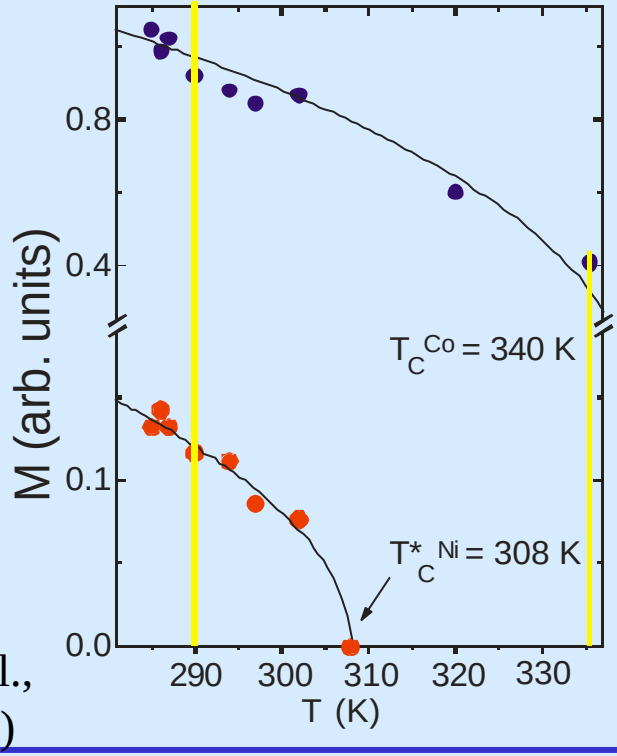
(ii) d_{Cu} = large $\Rightarrow$ no coupling, like a mixed Ni/Co powder

BUT $d_{Cu} \approx 2$ ML $\Rightarrow$ **?**

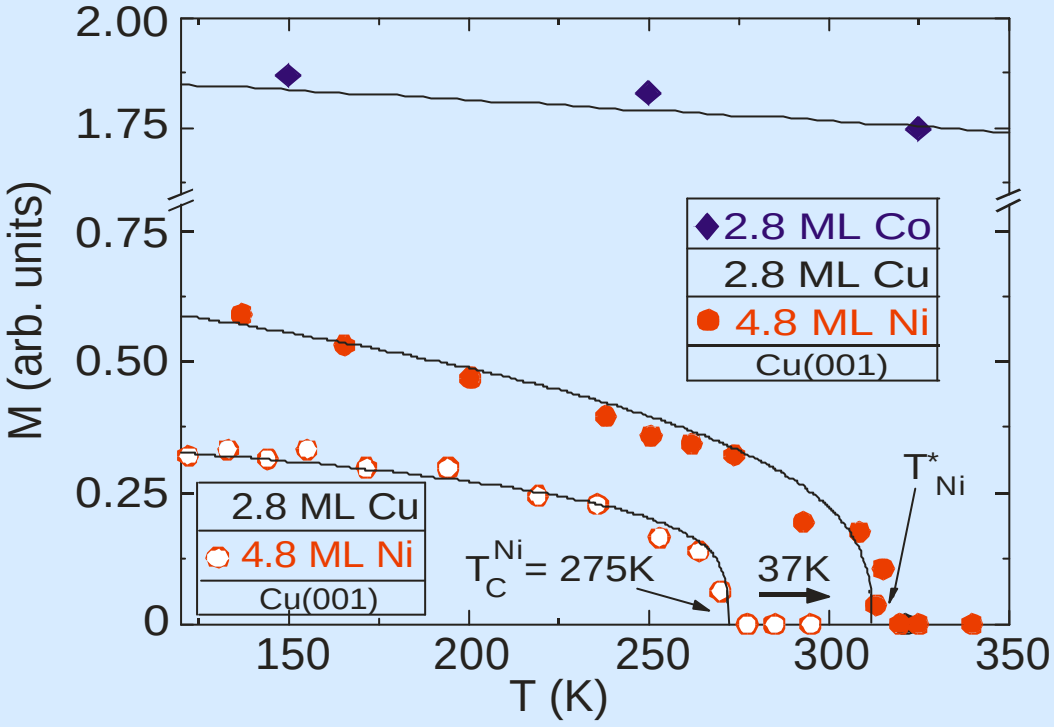
Element specific magnetizations and T_C's in trilayers.



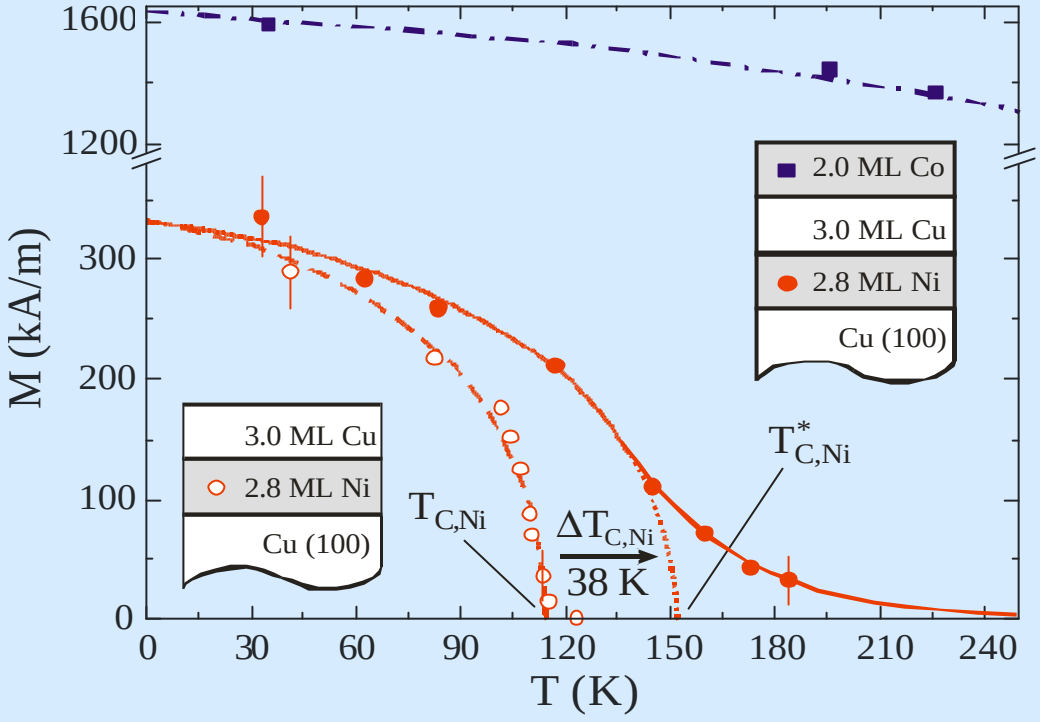




U. Bovensiepen et al.,
PRL **81**, 2368 (1998)



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.

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, and demonstrate the importance of *higher order spin-spin correlations* in low dimensional magnets.

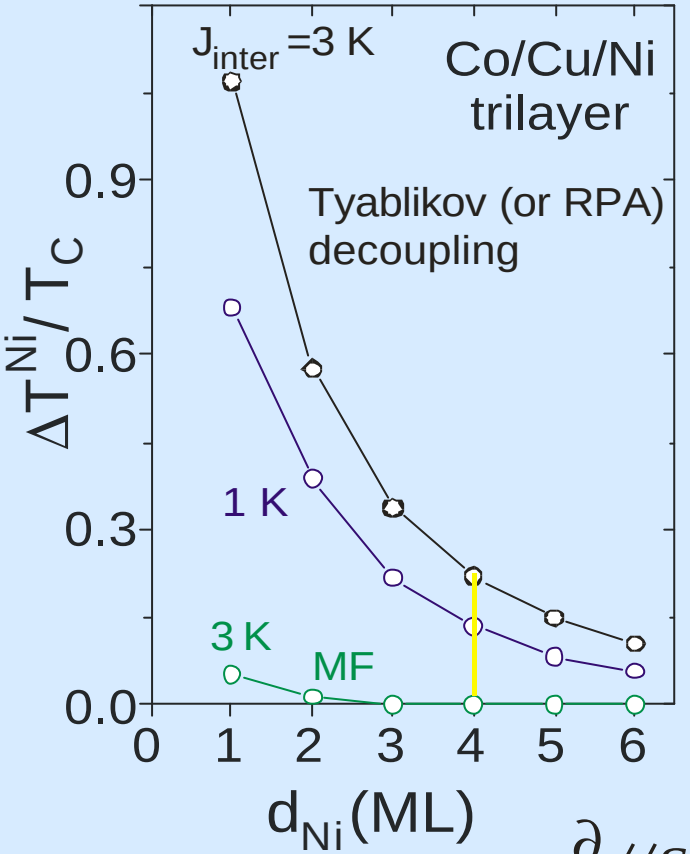
Spin-Spin correlation function $\frac{\partial}{\partial t}\langle\langle S_i^+ S_j^- \rangle\rangle \rightarrow$

$$S_i^z S_j^+ \approx \langle S_i^z \rangle S_j^+ - \langle S_i^- S_j^+ \rangle S_j^+ - \langle S_i^- S_j^+ \rangle^+ S_i^+ \quad \bullet\bullet\bullet$$

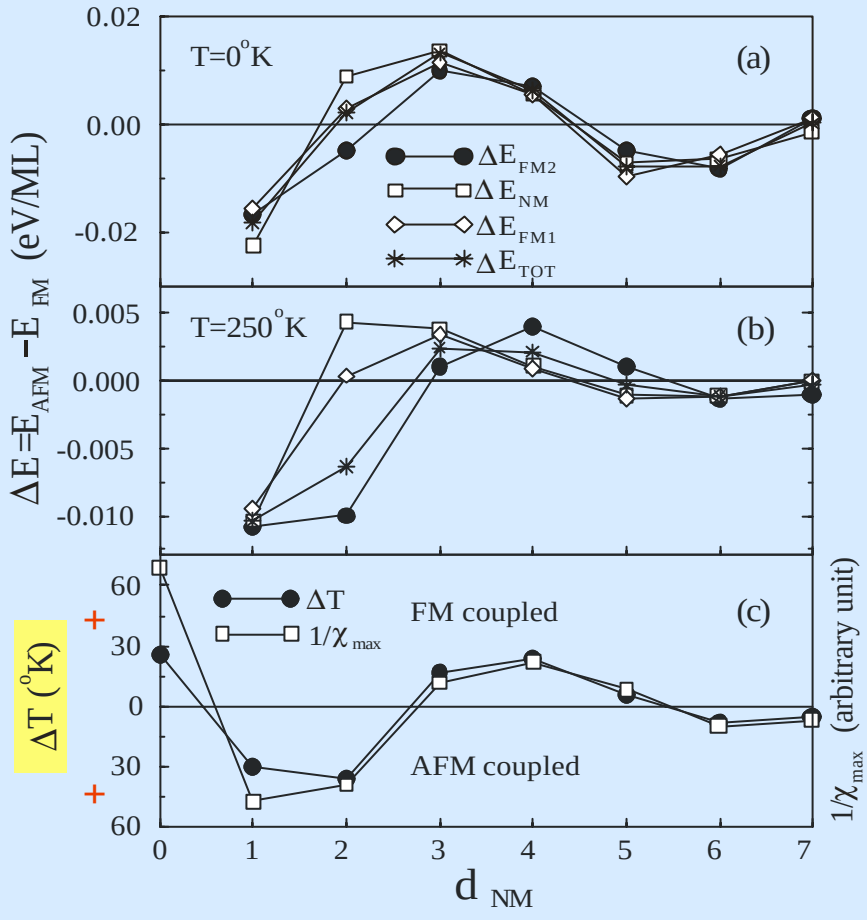
$\longleftarrow$ RPA $\longrightarrow$

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

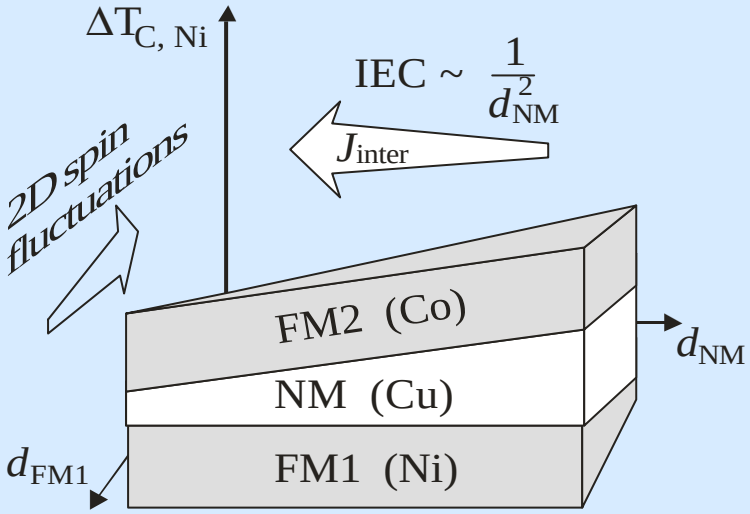
$$S_i^z S_j^+ \approx \langle S_i^z \rangle S_j^+ - \langle S_i^- S_i^+ \rangle S_j^+ - \langle S_i^- S_j^- \rangle^+ S_i^+ \quad \bullet\bullet\bullet$$

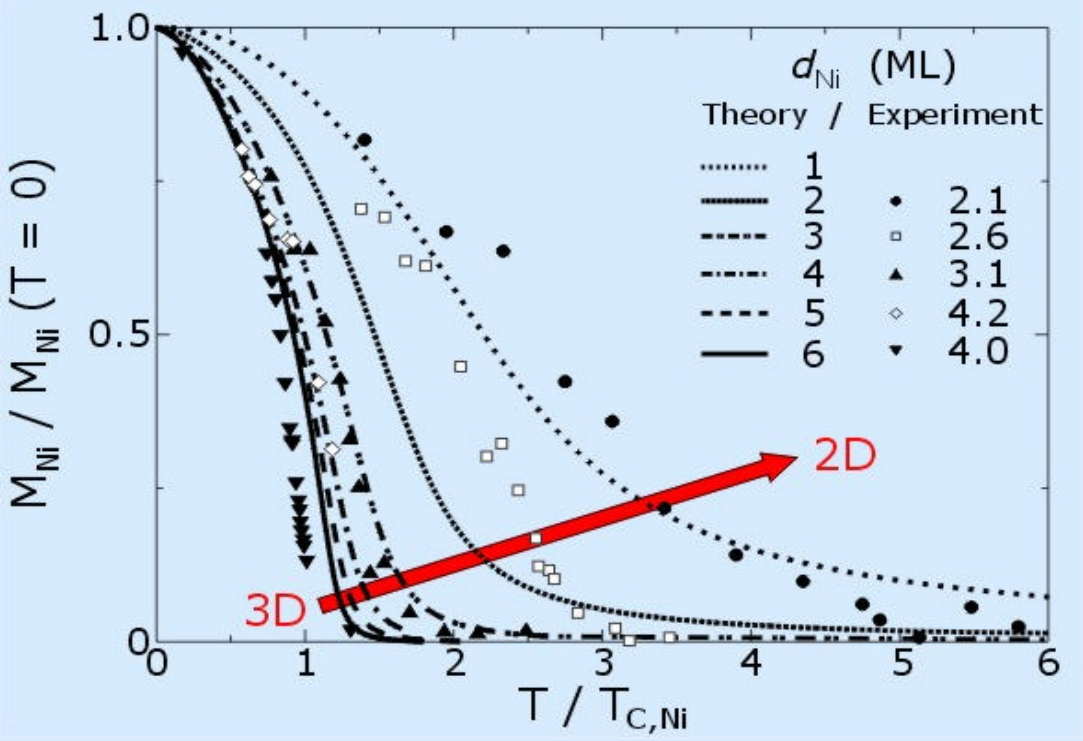
RPA

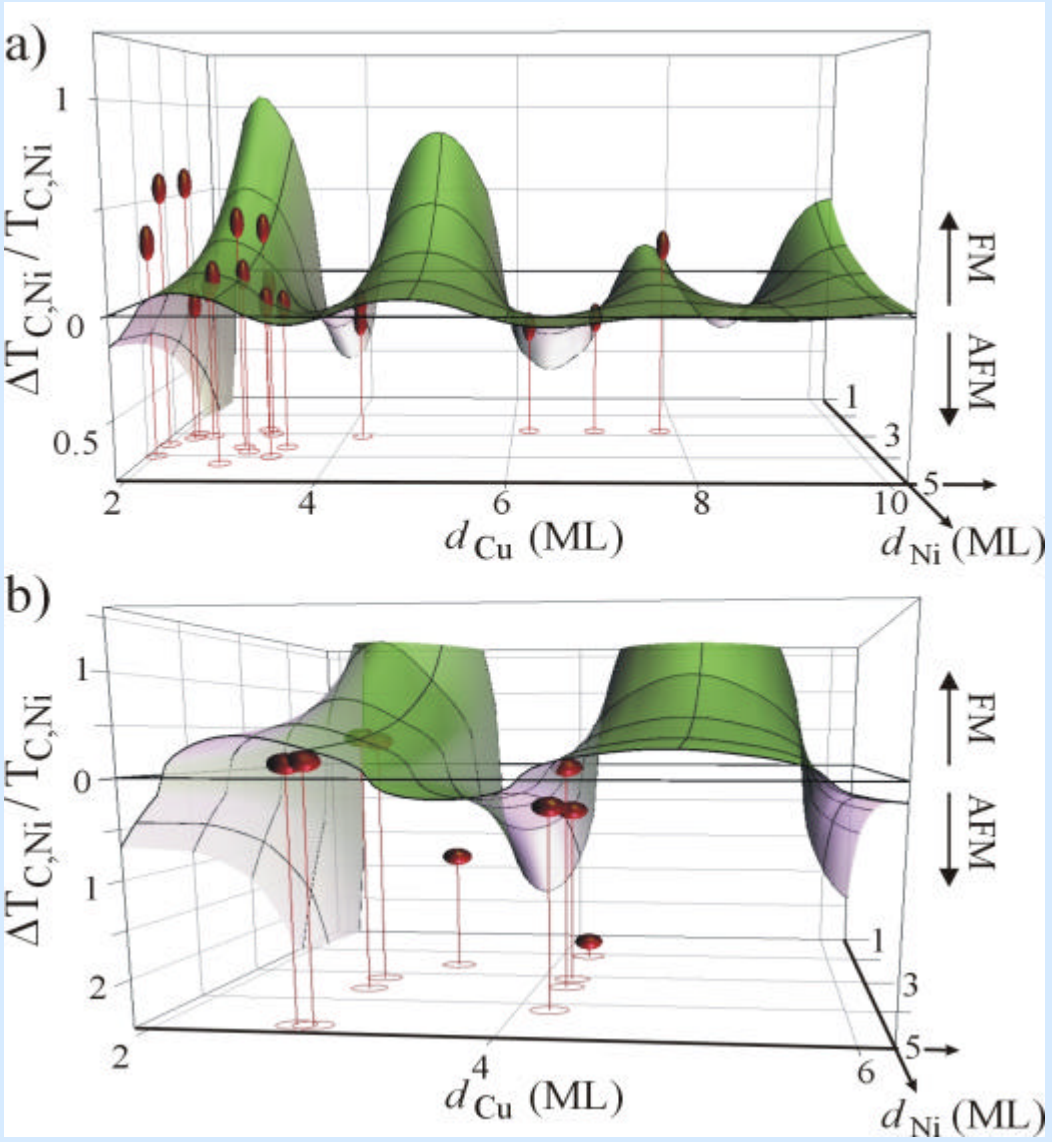
$\langle S_i^z \rangle S_j^+$ mean field ansatz (Stoner model) is insufficient to describe spin dynamics at interfaces of nanostructures

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

Evidence for giant spin fluctuations (A. Scherz, C. Sorg et al. PRB **72**, 54447 (2005))







Theory of weakly coupled two-dimensional magnets

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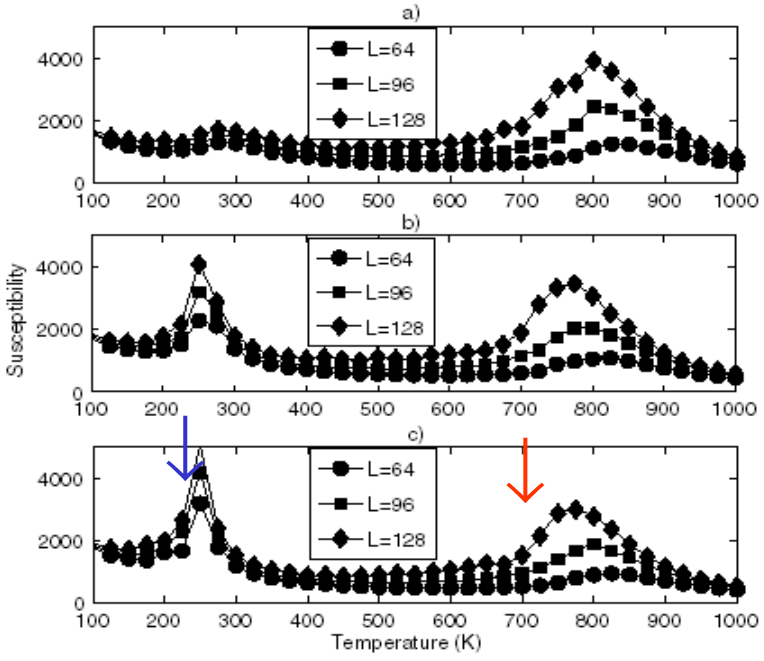


Figure 4. Susceptibility of the $Co_2/Cu_n/Ni_4$ trilayer system for (a) $n = 1$, (b) $n = 2$ and (c) $n = 3$, plotted for three different lattice sizes (number of spins in plane is $L \times L$).

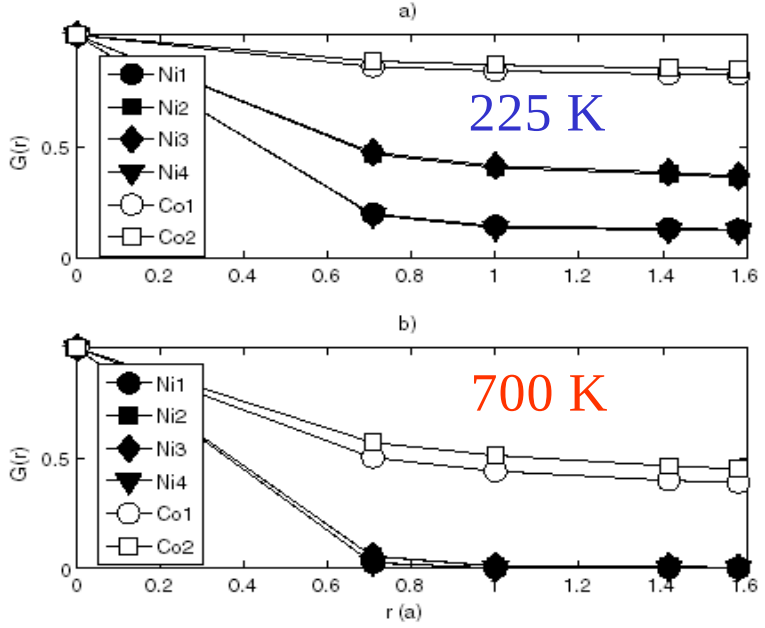
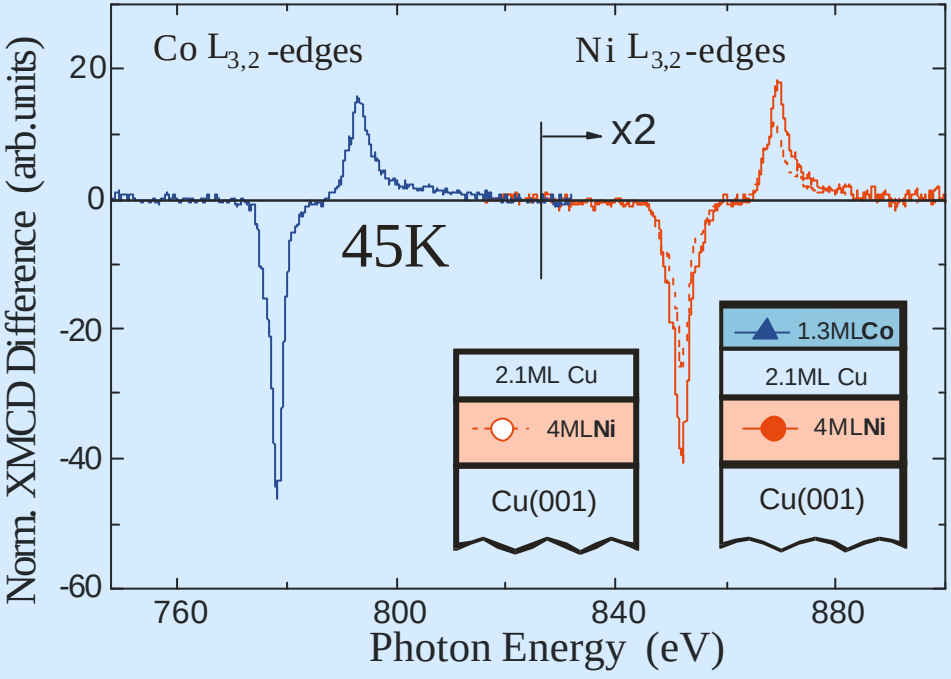


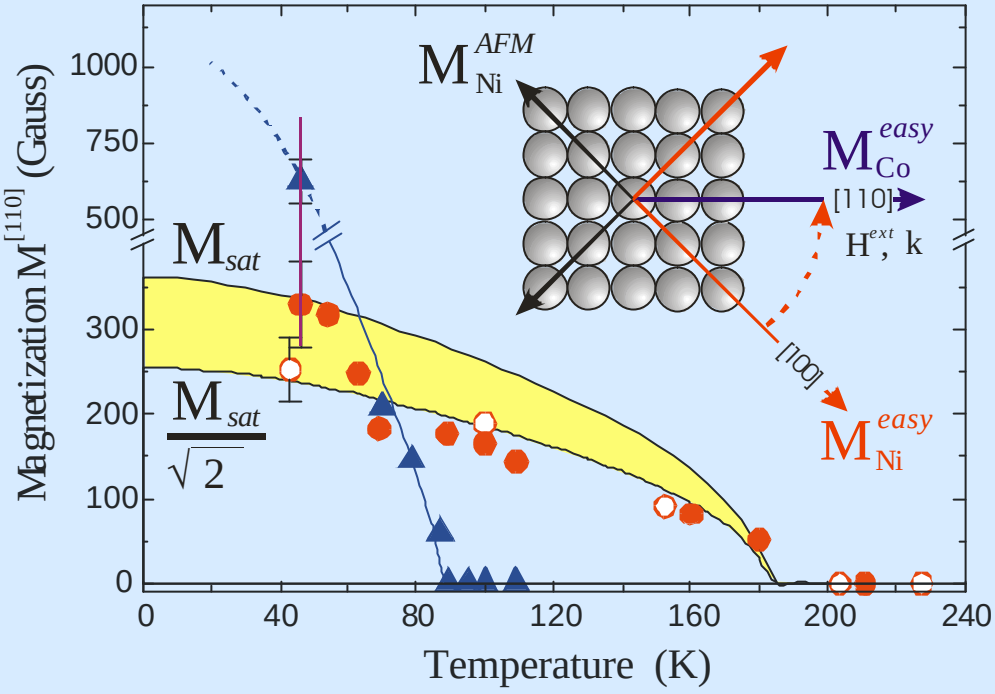
Figure 5. The spin–spin pair correlation function in-plane plotted as a function of distance (measured in lattice constants) of the $Ni_4/Cu_3/Co_2$ trilayer system for (a) $T = 225$ K and (b) $T = 700$ K. The notation of the different layers is the same as in figure 1.

"…the peak at low temperatures is associated with the disappearance of magnetism in the Ni layers … "

"…we come to the conclusion that two distinct temperatures are relevant …and a lower temperature where the Ni spin-spin correction … undergoes an abrupt modification."

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





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

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Conclusion:

The magnetism of FM monolayers is a very reach playground.