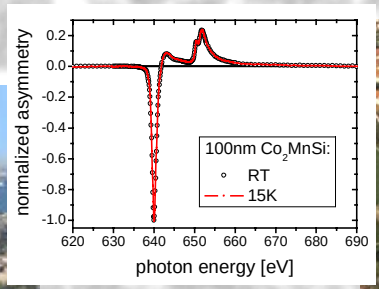
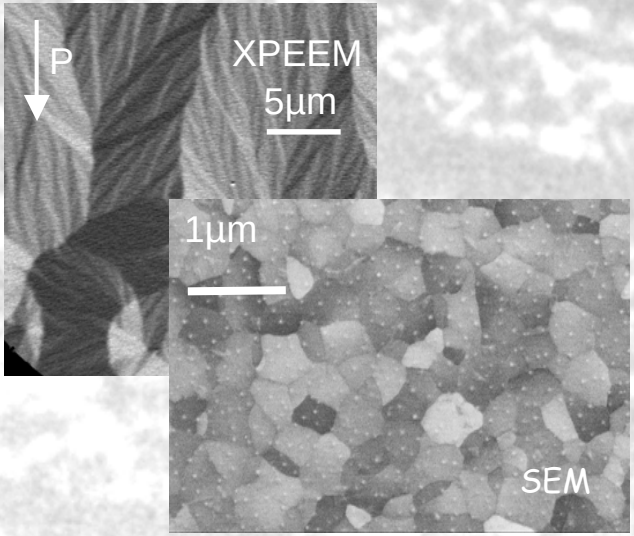
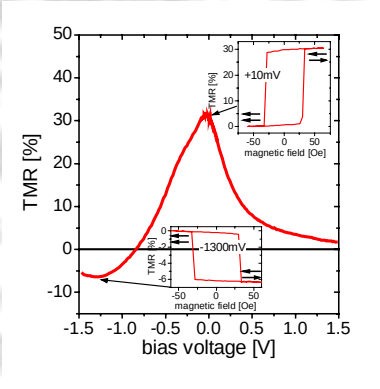


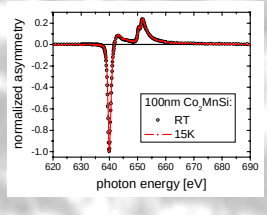
"Magnetic and chemical interface properties of magnetic tunnel junctions investigated by X-ray absorption spectroscopy"

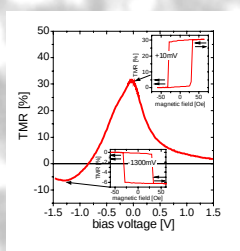


Jan Schmalhorst

Thin Films and Nanostructures
Department of Physics
Bielefeld University
Germany







Acknowledgements

...people:

A. Hütten, A. Thomas, S. Kämmerer, D. Ebke, M. Sacher, V. Höink, N.-N. Liu, G. Reiss (Bielefeld, Germany)

A. Scholl, E. Arenholz, A. Doran, T. Young (ALS)

...financial support:

Deutsche Forschungsgemeinschaft (DFG)
Bundesministerium für Bildung und Forschung (BMBF)
Europäische Gemeinschaft (EU)
Department of Energy (DOE)

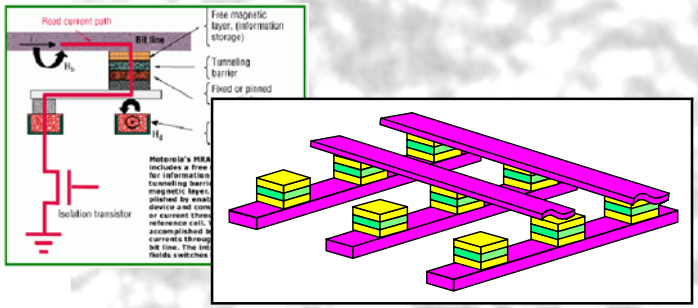
Motivation

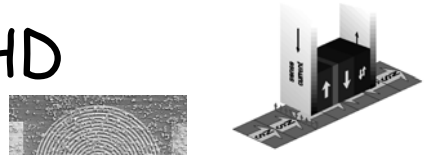
Fundamentals of spin-electronics:

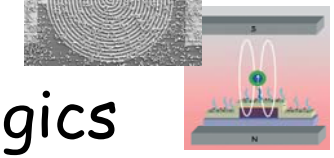
e^--charge + e^--spin = $ + fun!

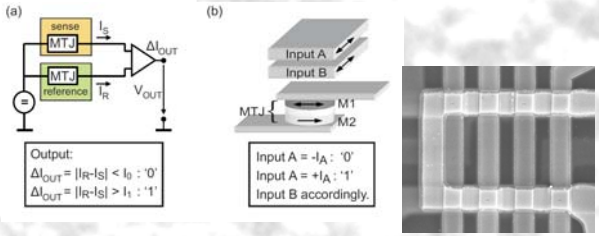
...some spin-electronic applications:

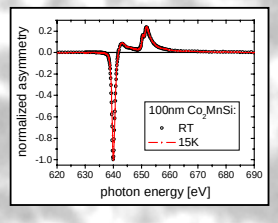
- non-volatile memory (MRAM)
- read-heads for HD
- sensors
- programmable logics
- ...

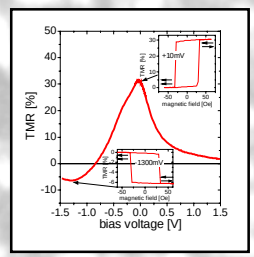












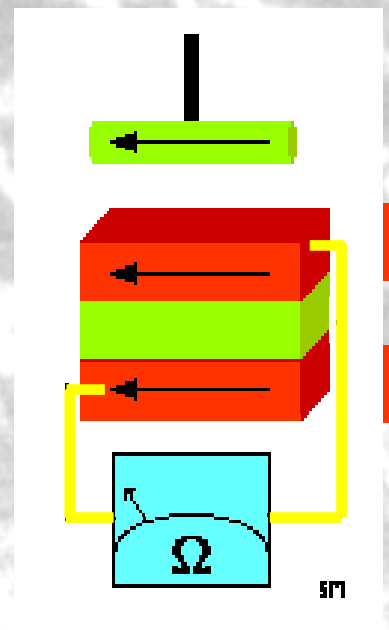
Outline

- Basics of **M**agnetic **T**unnel **J**unctions (MTJs)
- Brief history of Tunnel Magnetoresistance (the past and the future)
- Preparation and characterization techniques
- Some basic properties of the Heusler alloy Co_2MnSi
- Results:
  - bulk properties of Co_2MnSi and other Heusler thin films
  - chemical and magnetic properties at Co_2MnSi / Al-O interface
  - characteristic transport properties of Co_2MnSi based MTJs compared with Co-Fe/Ni-Fe and Co-Fe-B based junctions
  - interpretation of transport properties on the base of XMCD/XAS investigations
- Conclusions

Magnetic tunnel junction (MTJ)

... a basic device for spin-electronics...

...the MTJ stack:



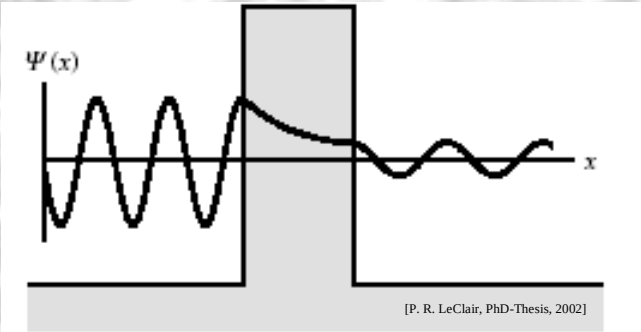
ferromagnetic metal

insulator

ferromagnetic metal

...fundamental physics:

QM tunnel effect
(spin-polarized)

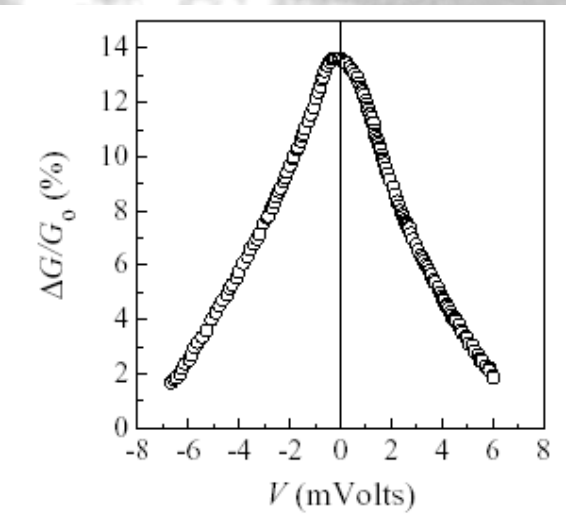


...tunnel magnetoresistance:

$TMR = (R^{AP}-R^{P}) / R^{P}$

...free particles can cross a potential barrier with finite probability!

...brief history of TMR in „conventional" MTJs:



1975:
M. Jullière
Fe/Ge/Co: 14% @ 4.2K

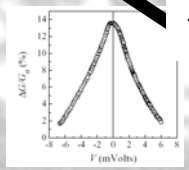
$$TMR = \frac{2P_{eff}^{FM1}P_{eff}^{FM2}}{1 - P_{eff}^{FM1}P_{eff}^{FM2}}$$

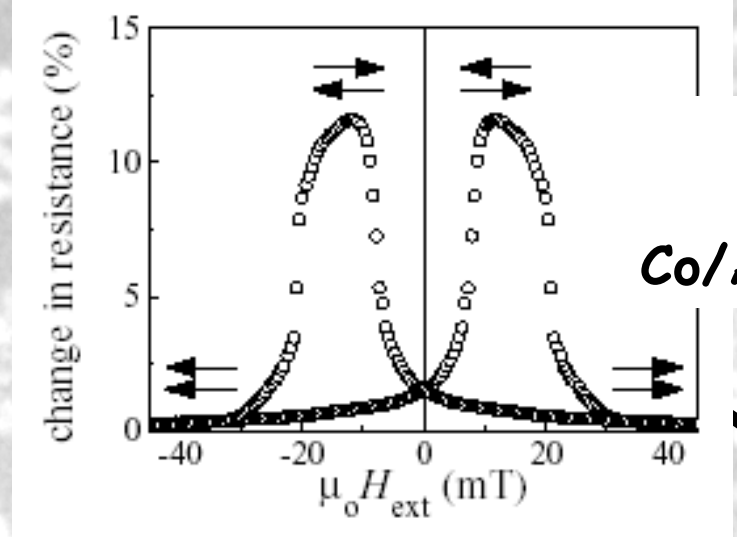
$$P_{eff} = \frac{Z^{\uparrow} - Z^{\downarrow}}{Z^{\uparrow} + Z^{\downarrow}}$$

time

...brief history of TMR in „conventional" MTJs:

1975:
14% @ 4.2K
TMR = $2P_1P_2/(1-P_1P_2)$



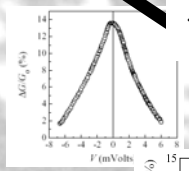


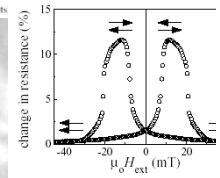
1995:
J. Moodera
Co/Al-O/Co: 12% @ RT

time

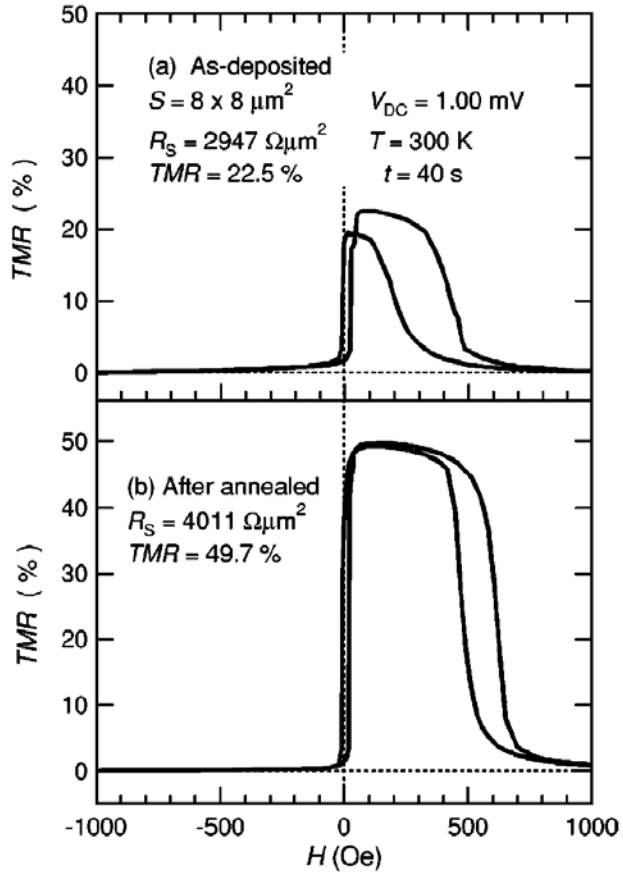
...brief history of TMR in „conventional" MTJs:

1975:
14% @ 4.2K
TMR = $2P_1P_2/(1-P_1P_2)$





1995:

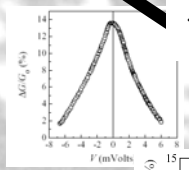


2000:
X.-F. Han et al.
Co-Fe/Al-O/Co-Fe:
50% @ RT

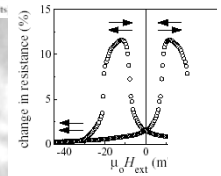
time

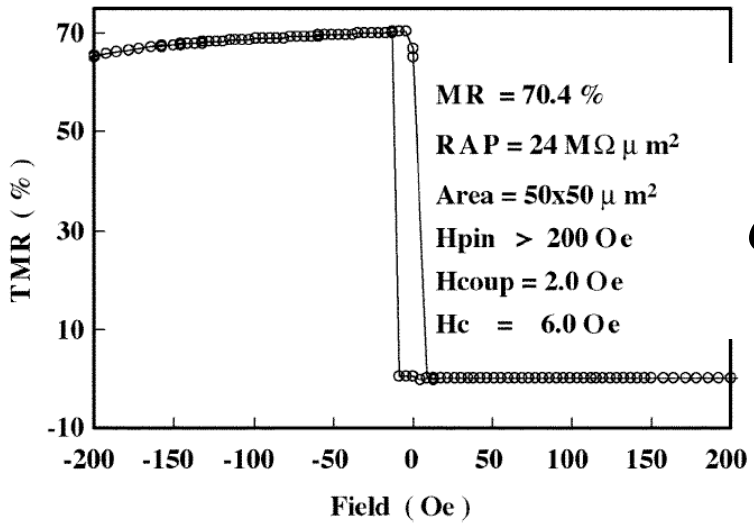
...brief history of TMR in „conventional" MTJs:

1975:
14% @ 4.2K
TMR = $2P_1P_2/(1-P_1P_2)$



1995:
12% @ RT



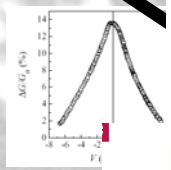


Jan. 2004:
D. Wang et al.
Co-Fe-B/Al-O/Co-Fe-B:
70% @ RT

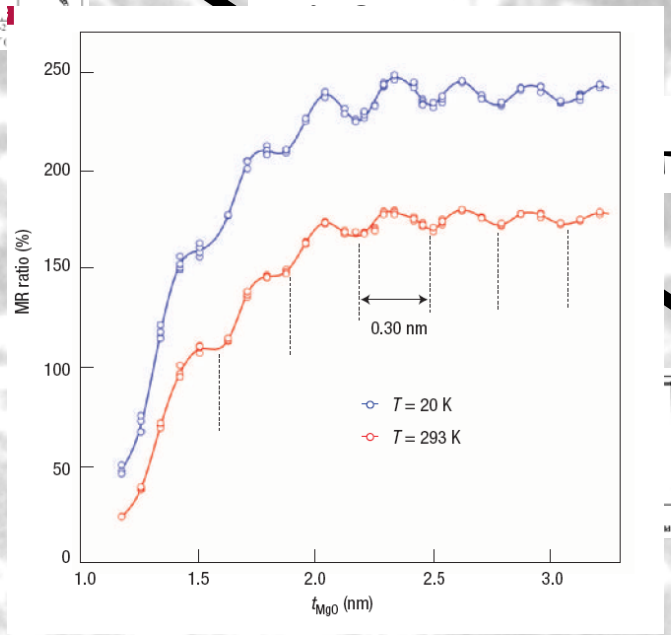
time

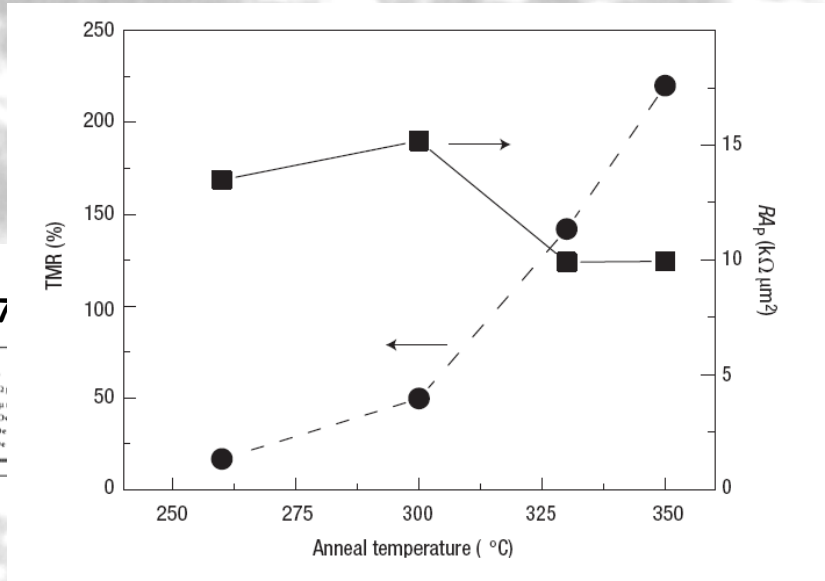
...brief history of TMR in „conventional" MTJs:

1975:
14% @ 4.2K
TMR = $2P_1P_2/(1-P_1P_2)$



1995:





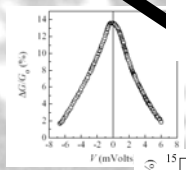
Dec. 2004:
S. Yuasa
Fe/MgO/Fe:
180% @ RT

Dec. 2004:
S. Parkin
Co-Fe/MgO/Co-Fe:
220% @ RT

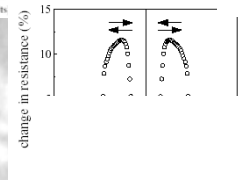
time

...brief history of TMR in „conventional" MTJs:

1975:
14% @ 4.2K
TMR = $2P_1P_2/(1-P_1P_2)$



1995:
12% @ RT



2000:

Table I

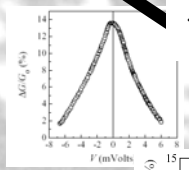
Reference layer	Free layer	T_a^{op} (°C)	TMR ratio (%)	RA ($\Omega\mu m^2$)
$Co_{40}Fe_{40}B_{20}$	$Co_{40}Fe_{40}B_{20}$	400	355	547
$Co_{40}Fe_{40}B_{20}$	$Co_{50}Fe_{50}$	400	277	1060
$Co_{40}Fe_{40}B_{20}$	$Co_{90}Fe_{10}$	350	131	714
$Co_{50}Fe_{50}$	$Co_{40}Fe_{40}B_{20}$	325	50	1042
$Co_{50}Fe_{50}$	$Co_{50}Fe_{50}$	270	12	740
$Co_{90}Fe_{10}$	$Co_{40}Fe_{40}B_{20}$	300	75	475
$Co_{90}Fe_{10}$	$Co_{90}Fe_{10}$	270	53	571
(pseudo-spin valve with 1.7 nm thick MgO-barrier)				
$Co_{40}Fe_{40}B_{20}$	$Co_{40}Fe_{40}B_{20}$	450	450	3700

Aug. 2006:
S. Ikedaa et al.
Co-Fe-B/MgO/Co-Fe-B:
450% @ RT
P=0.83

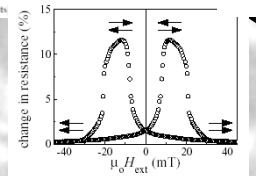
time

...brief history of TMR in „conventional" MTJs:

1975:
14% @ 4.2K
TMR = $2P_1P_2/(1-P_1P_2)$

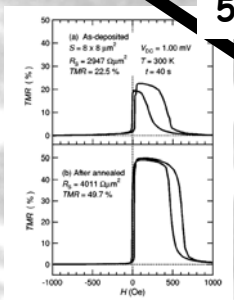


1995:
12% @ RT

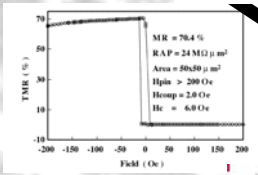


incoherent tunneling

2000:
50% @ RT

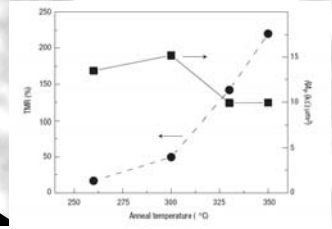


Jan. 2004:
70% @ RT



coherent tunneling (?)

Dec. 2004:
220% @ RT



Dec. 2004:
180% @ RT

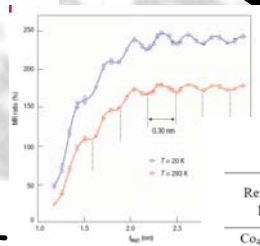


Table I

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(pseudo-spin valve with 1.7 nm thick MgO-barrier)				
$Co_{40}Fe_{40}B_{20}$	$Co_{40}Fe_{40}B_{20}$	450	450	3700

Aug. 2006:
450% @ RT
P=0.83

time

Giant TMR has been predicted for Fe/MgO/Fe(001) by Butler et al. and Mathon et al. in 2001.

...not only coherent tunneling can increase the TMR:

Take **Jullière's** formula serious: $TMR = 2P_1P_2/(1-P_1P_2)$

$\Rightarrow$ TMR = ∞ for $P_1 = P_2 = 1.0$ (half-metallic FM!)

Some materials predicted to be half-metallic:

- CrO_2
- Fe_3O_4
- manganites like $La_{2/3}Sr_{1/3}MnO_3$, $La_{2/3}Ca_{1/3}MnO_3$, ...
- Heusler alloys like Co_2MnSi, Co_2FeSi,...

Recent results on Heusler alloy based MTJs (low T, small V):

- $Co_2MnSi^{(100)\ epitax.}$/Al-O/Co-Fe: $P_{Co2MnSi} = 0.89$ [1]
- $Co_2MnSi^{(110)\ tex.}$/Al-O/Co-Fe: $P_{Co2MnSi} = 0.66$ [2]
- $\{Co_2MnSi/Co_2FeSi\}_{10x}^{(110)\ tex.}$/Al-O/Co-Fe: $P_{Co2FeSi} = 0.74$ [3]

[1] Oogane et al., J. Phys. D: Appl. Phys. **39** (2006) 834
[2] J. Schmalhorst et al., Appl. Phys. Lett. **86** (2005) 152102
[3] D. Ebke et al., Appl. Phys. Lett, accepted (2006)

...the work presented today is published in:

[A] J. Schmalhorst, S. Kämmerer, M. Sacher, G. Reiss, A. Hütten, A. Scholl: "Interface structure and magnetism of magnetic tunnel junctions with Co_2MnSi electrode", Phys. Rev. B **70** (2004) 024426

[B] J. Schmalhorst, M. D. Sacher, O. Schebaum, A. Thomas, G. Reiss, A. Hütten, E. Arenholz: "Transport properties of magnetic tunnel junctions with Co_2MnSi electrode: influence of temperature-dependent interface magnetization and electronic band structure", Phys. Rev. B, submitted

[C] J. Schmalhorst, M. D. Sacher, V. Höink, G. Reiss, A. Hütten, D. Engel, A. Ehresmann: „Magnetic tunnel junctions with Co_2MnSi electrode: magnetic and chemical properties of the bulk and of the electrode / barrier interface", J. Appl. Phys., accepted

[D] D. Ebke, J. Schmalhorst, N.-N. Liu, A. Thomas, G. Reiss, A. Hütten: "Large tunnel magnetoresistance in tunnel junctions with Co2MnSi / Co2FeSi multilayer electrode", Appl. Phys. Lett., accepted

Why is the implementation of Heusler alloys in MTJs so challenging?

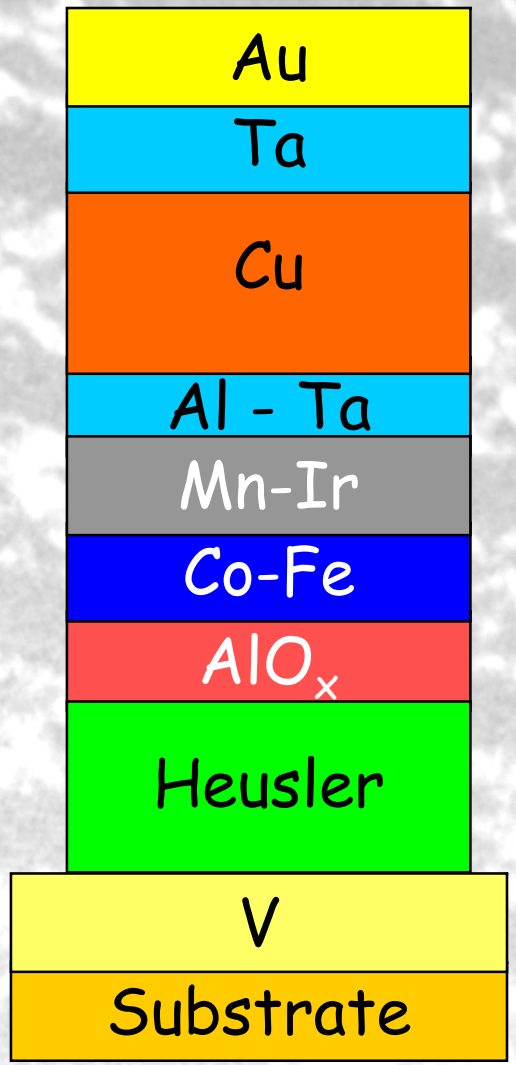
MTJ:

- very sensitive to barrier-electrode interface
- thermal stability limited to typically 350°C

sputtered Heusler alloy:

- half-metallicity usually very sensitive to atomic disorder
- thermal treatment at high T required to promote atomic order
  (e.g. ≈450°C for Co_2MnSi)

…typical layer stack of our Heusler based MTJs:



We need to know the transport properties of "full" junctions…

magnetoresistance:

$$R = R(V, T, H_{ex})$$

current-voltage-characteristic:

$$dj/dV = dj/dV\ (V,T,H_{ex})$$

…and their structural and magnetic properties:

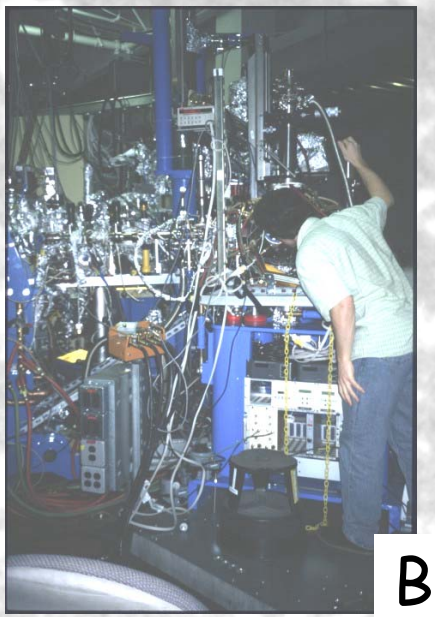
AGM, XRD, Auger depth profiling, XMCD/XAS

Why do we apply XAS and XMCD to "half" junctions?

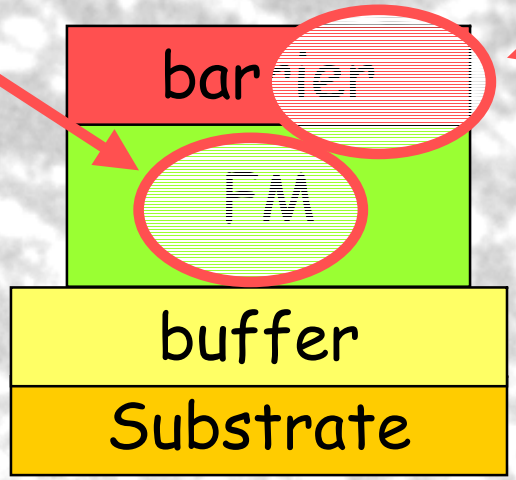
⇒ element specific chemical and magnetic properties
of Heusler / barrier interface and Heusler bulk...

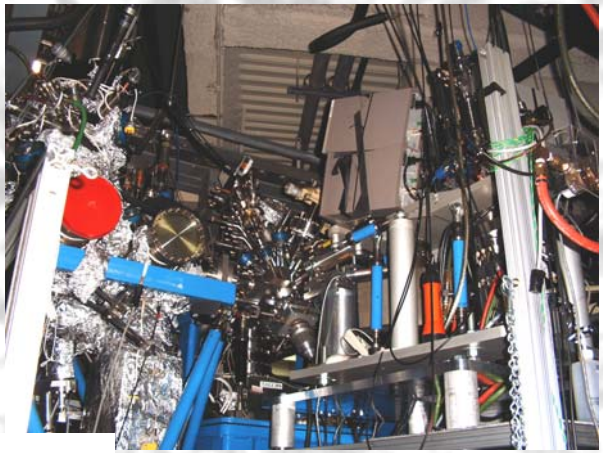
XAS / XMCD (FY)

XAS / XMCD (TEY)



BL 4.0.2

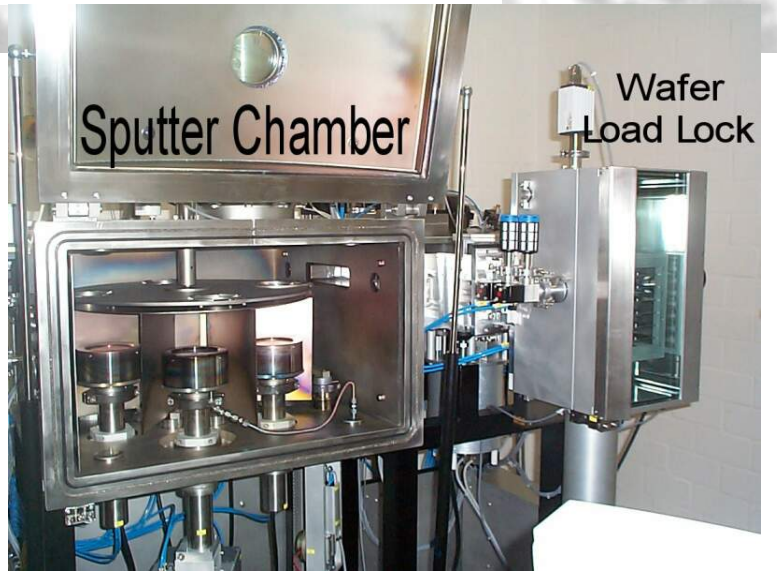




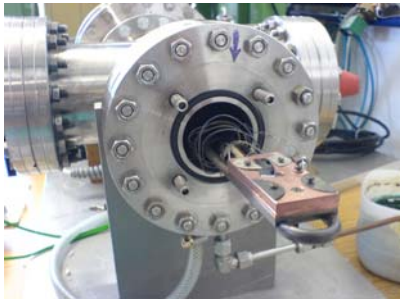
BL 7.3.1.1

How do we fabricate our MTJs?

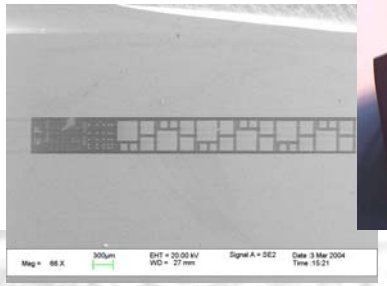
1) magnetron sputter deposition at <10^{-7} mbar (polycrystalline films)

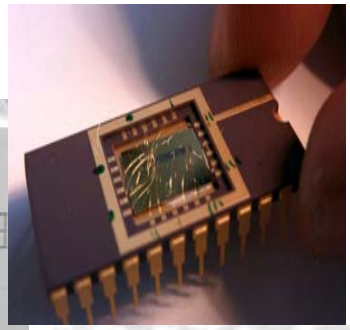


2) in-situ and/or ex-situ annealing



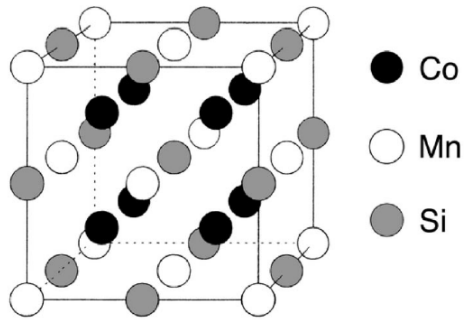
3) patterning / bonding





...some properties of the Heusler alloy Co_2MnSi:

Theory [PRB **65** (2002) 184431, I. Galanakis, private communication]:



4x fcc:
Mn: 0 0 0
Si: $\frac{1}{2}$ $\frac{1}{2}$ $\frac{1}{2}$
Co: $\frac{1}{4}$ $\frac{1}{4}$ $\frac{1}{4}$, $\frac{3}{4}$ $\frac{3}{4}$ $\frac{3}{4}$

...similar properties are found for Co_2FeSi!

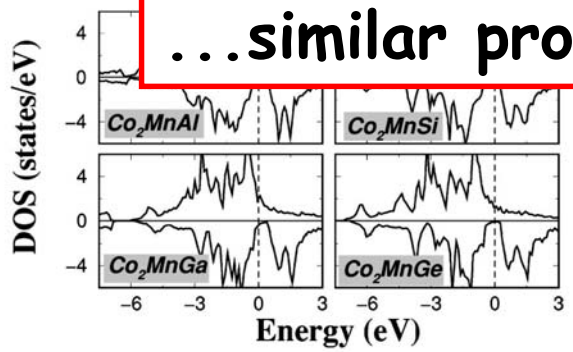


FIG. 2. Calculated spin-projected DOS for the Co_2MnZ compounds, where Z stands for Al, Ga, Si, and Ge. They all possess a finite very small spin-down DOS around the Fermi level.

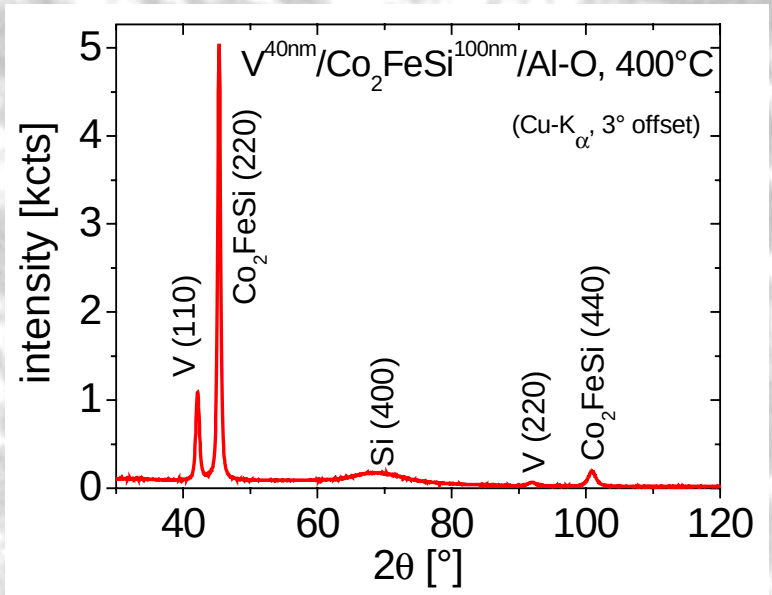
DOS$^{(\downarrow)}$ very small
$\Rightarrow$ high P_{eff} expected!

	Co	Mn	Si
$m^{spin}[\mu_B]$	1.021	2.971	-0.074
$m^{orb}[\mu_B]$	0.029	0.017	0.001

Experiment [J. Phys.: Con. Mat. **12** (2000) 1827]: $M = 4.96\mu_B$/f.u., $T_C = 985K$

…some bulk properties of our Co_2MnSi and Co_2FeSi single layers and {Co_2MnSi/Co_2FeSi} multi layers:

A) (110) –textured on V(110)-buffer



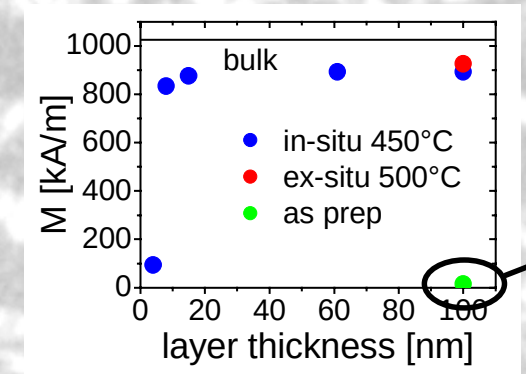
→ similar results found for Co_2MnSi single layers and {Co_2MnSi/Co_2FeSi} MLs

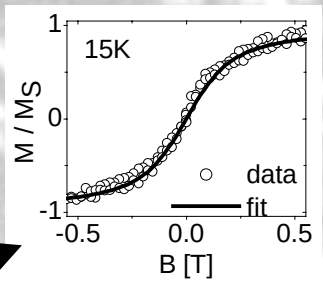
B) optimal annealed films have small coercivity at RT: H_C = 10-20Oe
→ used as "free" electrode

…some bulk properties of our Co_2MnSi and Co_2FeSi single layers and {Co_2MnSi/Co_2FeSi} multi layers:

C) annealing important to induce atomic order and increase magnetization

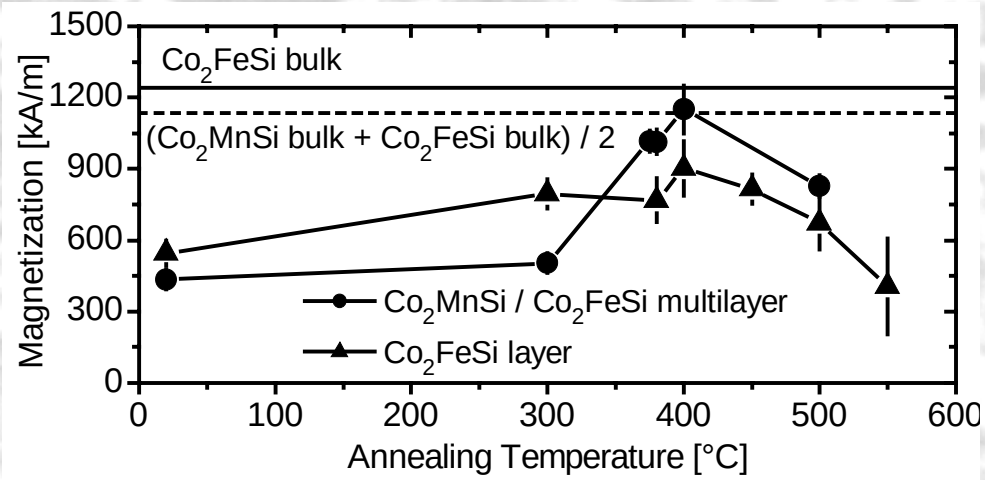
Co_2MnSi @RT:





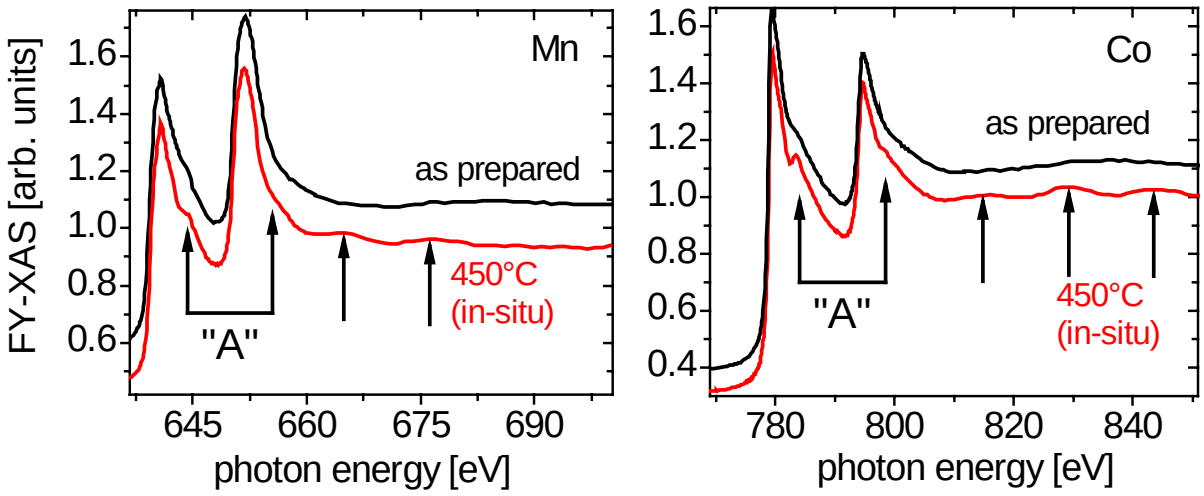
As prepared Co_2MnSi films are superparamagnetic
→ Langevin function:
$\mu \approx 300\mu_B$

100nm thick Co_2FeSi and {Co_2MnSi/Co_2FeSi} @RT:



Chemical states at Co_2MnSi-AlO_x interface compared to Co_2MnSi bulk (100nm):

Co_2MnSi bulk: XAS at Co- and Mn-$L_{3,2}$-edge: 2p → 3d (4s)

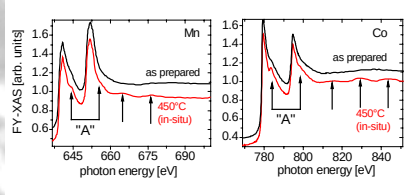


- EXAFS oscillations indicate atomic ordering
- "A": peaks in 3d-density of states 4eV above E_F (rep. by SPRKKR)

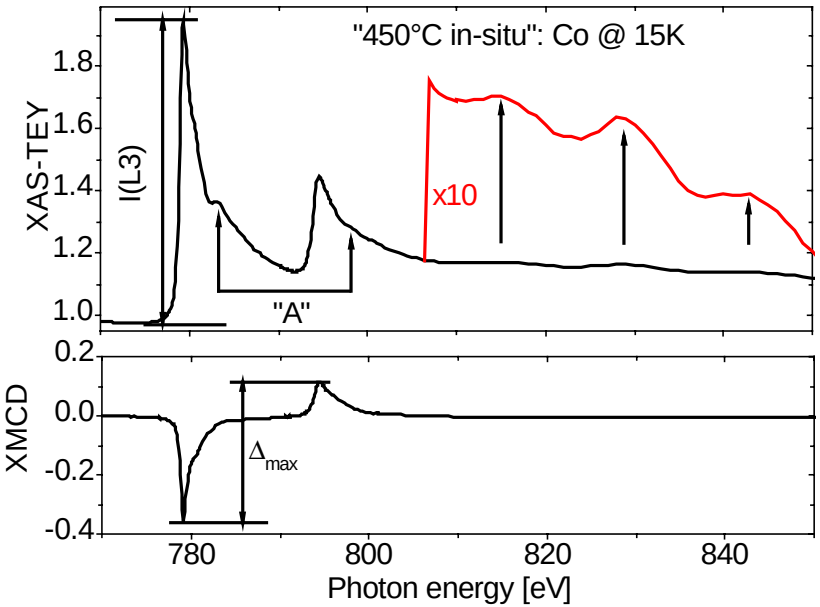
→ annealing changes bandstructure and improves atomic order!

Chemical states at Co_2MnSi-AlO_x interface compared to Co_2MnSi bulk (100nm):

Co_2MnSi bulk:

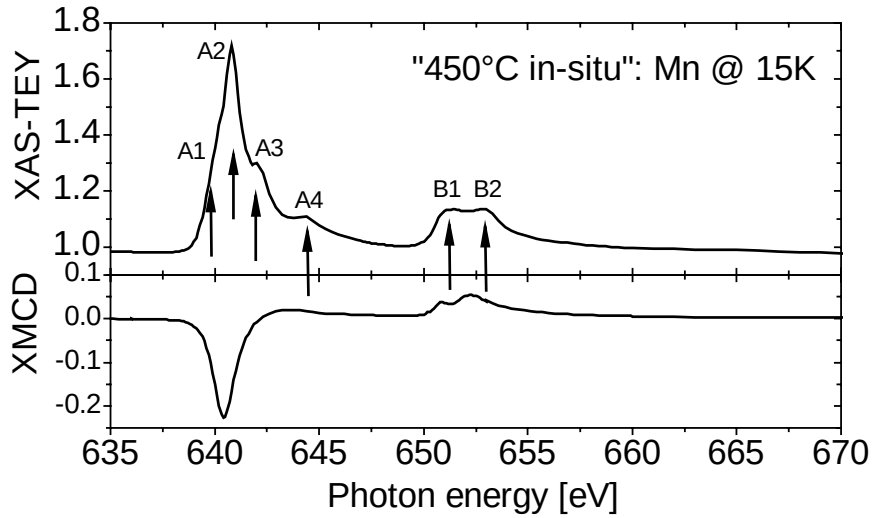


Co at Co_2MnSi / Al-O interface:



→ similar to bulk properties

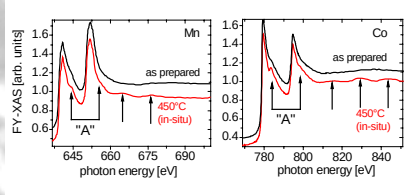
Mn at Co_2MnSi / Al-O interface:



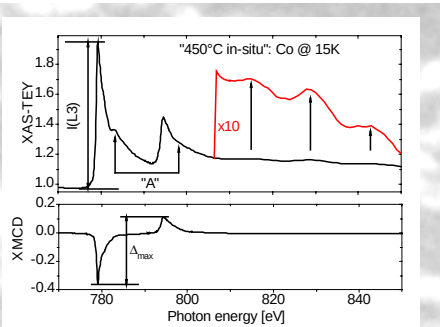
→ interfacial MnO masks finger-prints of ordering process

Magnetic moments at Co_2MnSi-AlO_x interface compared to Co_2MnSi bulk (100nm):

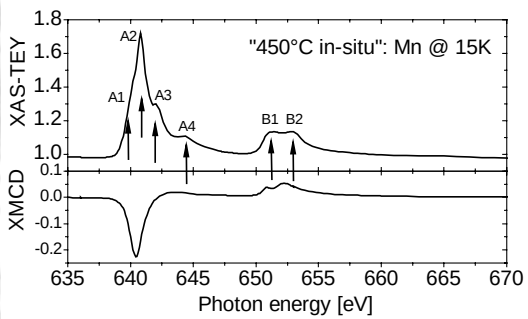
Co_2MnSi bulk:



Co at Co_2MnSi / Al-O interface:



Mn at Co_2MnSi / Al-O interface:



→ Sum rules analysis[1] of TEY („450°C in-situ):
$m^{spin}(Mn)/m^{spin}(Co) = 1.59\mu_B/1.15\mu_B$
$= 1.38 < 2.9$ (theory[2])

interfacial MnO reduces Mn moment

→ Estimation of bulk moment ratio (FY + TEY):
$m^{spin}(Mn)/m^{spin}(Co) \approx 3$

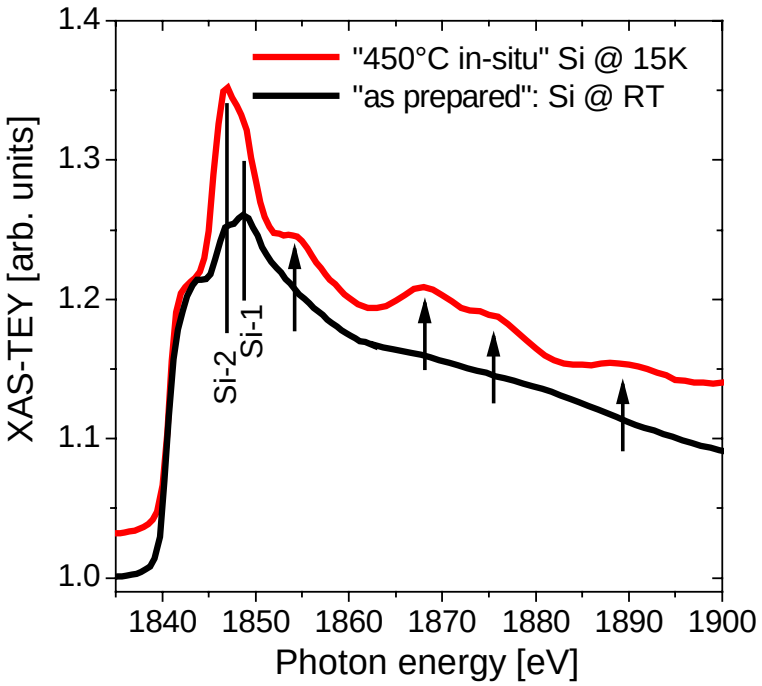
→ Temperature dependence of interfacial magnetic moments:

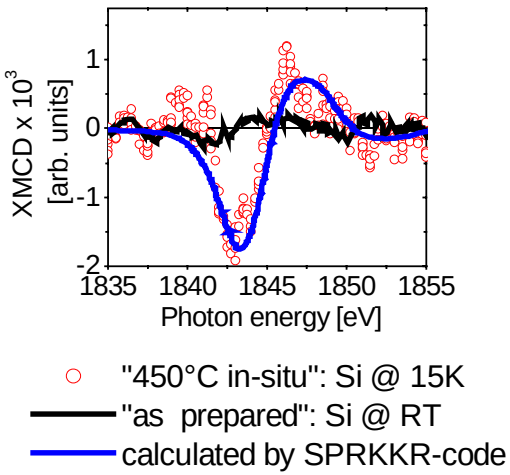
sample	m(LT) / m(300K)
Co_2MnSi / Al-O „as prepared"	Co: 14.0 Mn: 13.5 LT= 15K
Co_2MnSi / Al-O „450°C in-situ"	Co: 1.06 Mn: 1.11
Co-Fe-B/Al-O „275°C"	Co: 1.02 Fe: 1.03

Δm/m 3x larger!

[1] Chen et al., PRL **75** (1995) 152, jj-mixing neglected
[2] Galanakis et al., PRB **66** (2002) 174429

Chemical and magnetic properties of Si at Co_2MnSi-AlO_x interface:





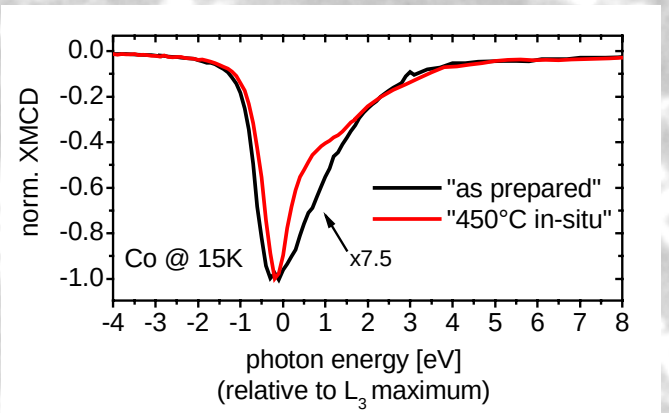
XAS at Si-K edge:
1s → 2p

- "Si-1": interfacial SiO_2 in addition to interfacial MnO
- EXAFS oscillations indicate atomic ordering
- "Si-2": peak in 2p-density of states 6eV above E_F (rep. by SPRKKR)

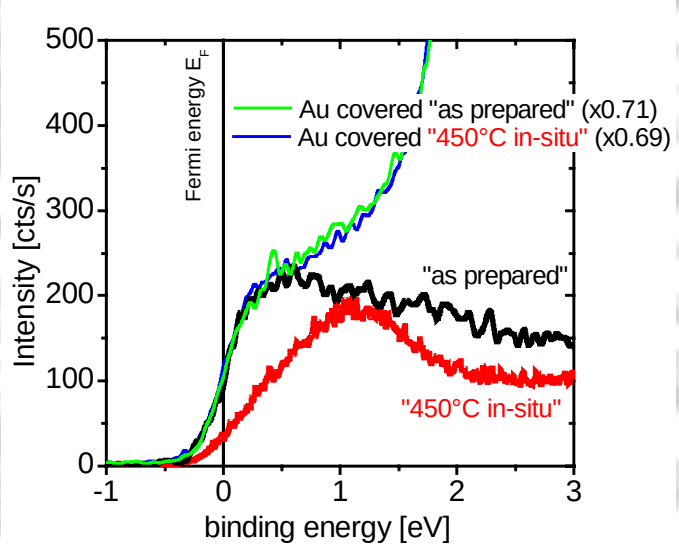
→ similar fingerprints of ordering as for Co and Mn!

...further hints to annealing induced changes of the electronic bandstructure at Co_2MnSi-AlO_x interface:

unoccupied (d-)states:



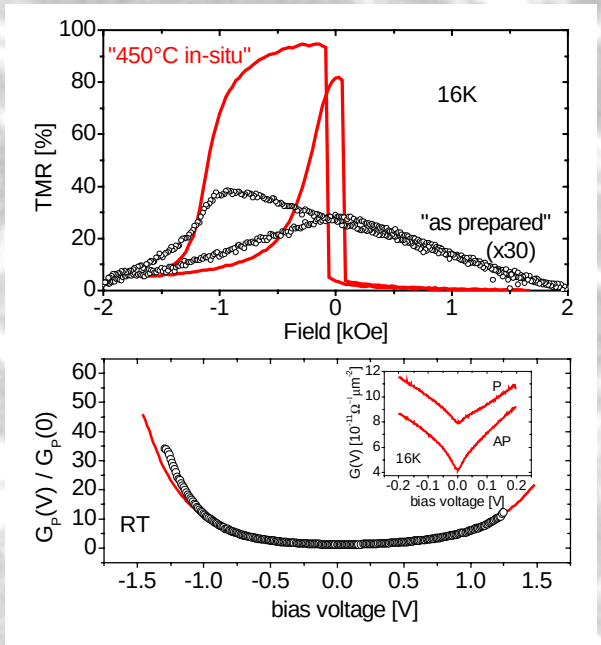
occupied states:

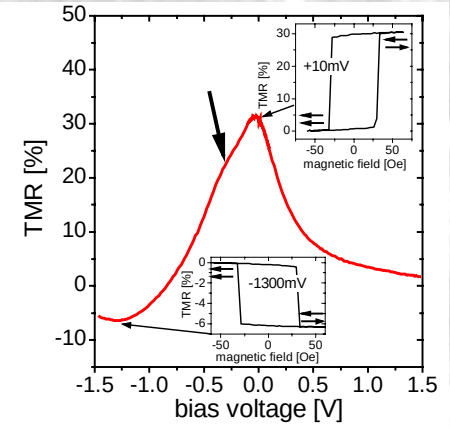


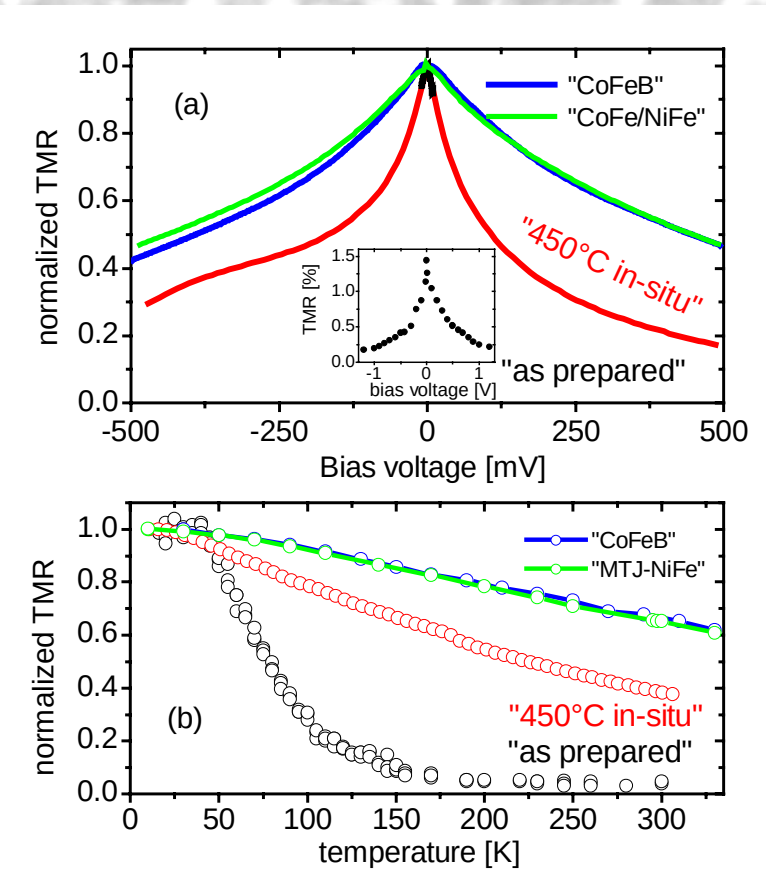
mono-XPS (hv=1486.7eV, emission angle= 20°)

→ important for interpretation of TMR vs V dependence...

Characteristic transport properties of Co_2MnSi alloy based MTJs...

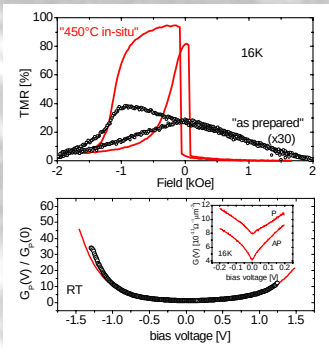


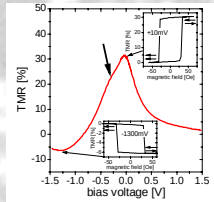


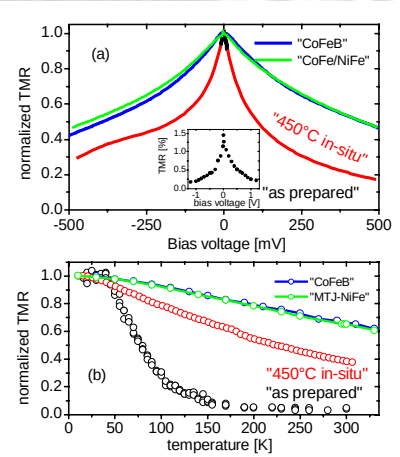


...compared with "conventional" CoFeB and NiFe/CoFe based MTJs with Al-O barrier...

Characteristic transport properties of Co_2MnSi alloy based MTJs...







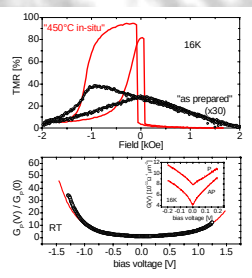
→ „as prepared" and „450°C in-situ"have similar tunnel characteristic (similar barrier shape!)

„as prepared":
→ superparamagnetic switching behavior
→ strongest temperature dependence
→ strongest bias voltage dependence
→ TMR always positive

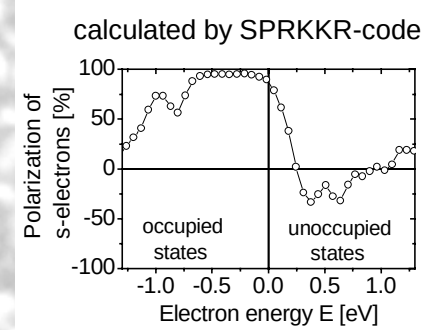
„450°C in-situ":
→ TMR(T) convex, stronger than for „conventional" MTJs (CoFeB, NiFe/CoFe)
→ TMR(V) stronger than for „convent. " MTJs
→ negative TMR for large negative bias voltage

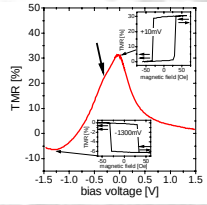
→ similar results are found for Co_2MnSi single layer and {Co_2MnSi/Co_2FeSi} ML based MTJs!

Interpreting the transport properties of MTJs with ordered Co_2MnSi electrode on the base of XAS/XMCD investigations...



TMR inversion reflects the band structure of well ordered Heusler alloy

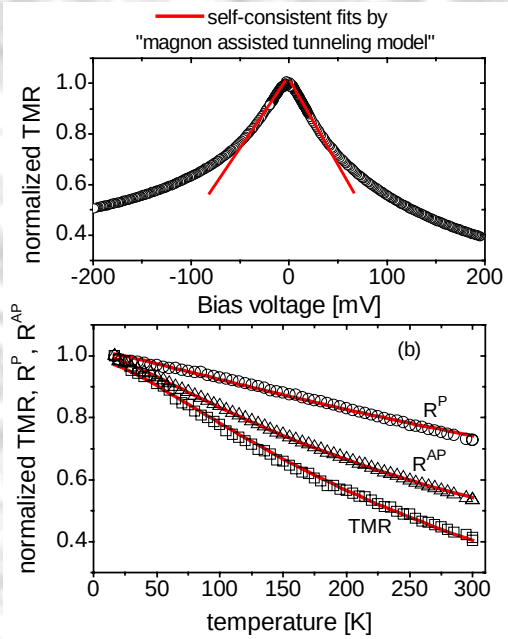


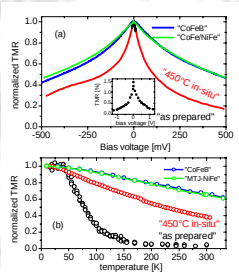


Current limitation of effective spin-polarization (66% @ 20K/1mV):

→ interfacial MnO/SiO_2 hinders perfect atomic order of Heusler

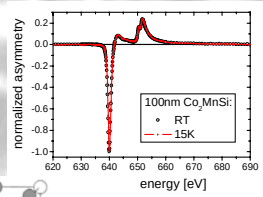
→ spin scattering on paramagnetic Mn^{2+} ions and unpolarized conductance via defect states in the barrier (quasi-elastic processes!)

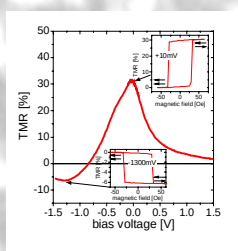


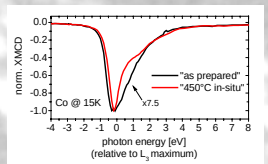


Magnon assisted tunneling[1] is suggested to be considerably stronger in "450°C in-situ" than in NiFe/Al-O/CoFe and CoFeB/Al-O/CoFeB MTJs.

[1] Han et al., PRB **63** (2001) 224404







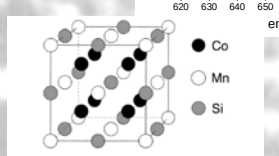
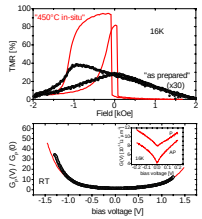
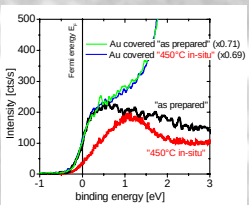


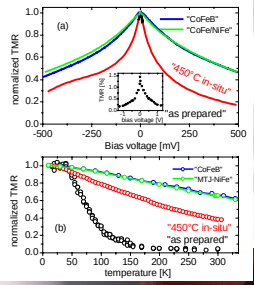
FIG. 1. The crystal structure of Heusler Co_2MnSi.

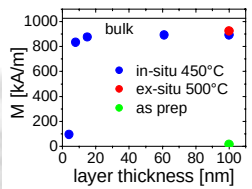


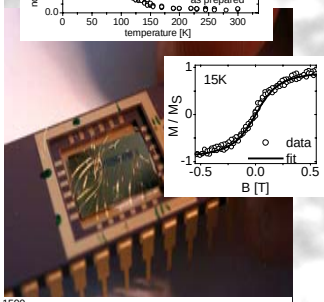


Conclusions

1. Implementation of Co_2MnSi and Co_2FeSi single layer and {Co_2MnSi / Co_2FeSi} multi layer electrodes in Al-O based MTJs is possible!
2. High spin-polarization of up to 0.74 in MTJs achieved, i.e., larger than for conventional 3d-metal alloy / AlO_x-interfaces!
3. Temperature dependent X-ray absorption spectroscopy and magnetic circular dichroism is very useful to understand the transport properties of complex MTJ systems!







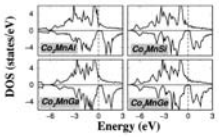
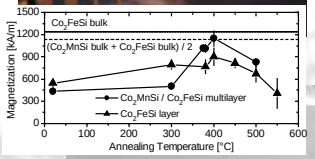


FIG. 2. Calculated spin-projected DOS for the Co_2MnZ compounds, where Z stands for Al, Ga, Si, and Ge. They all possess a finite very small spin-down DOS around the Fermi level.



Thank you for your attention!