Universität Bielefeld

Fakultät für Physik

CENTER FOR SPINELECTRONIC MATERIALS AND DEVICES

HEUSLER COMPOUNDS (AND RELATED) IN MAGNETIC TUNNEL JUNCTIONS

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Collaborations:

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J. Moodera, Cambridge / Elke Arenholz, ALS Berkeley
Siemens AG / Intel Corp. / Bosch / Singulus / Sensitec

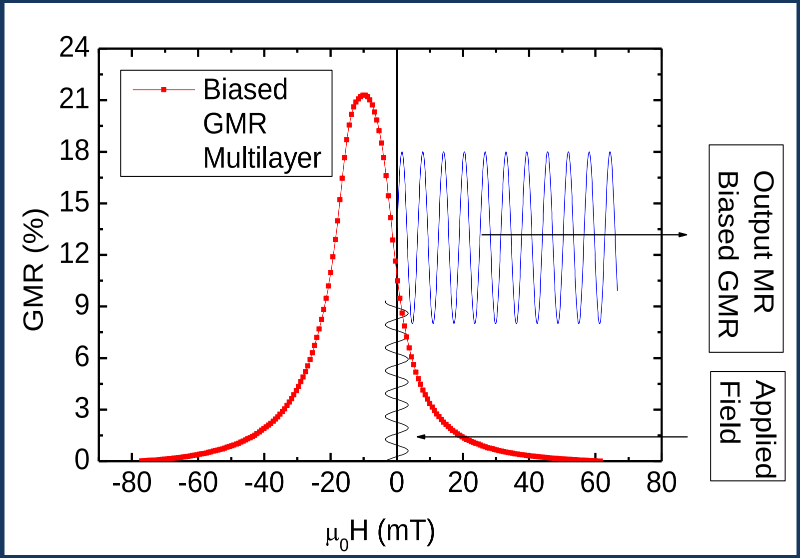
Funding

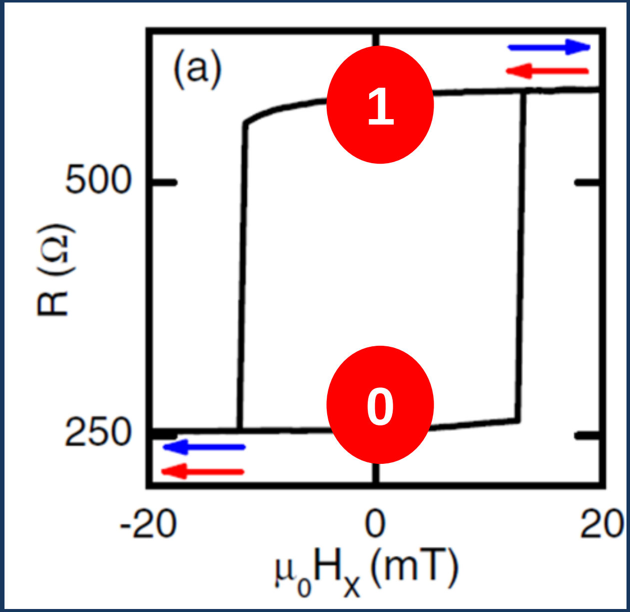
DFG
Priority
Programme
SpinCat

FAQ‘s: Where is Bielefeld ?

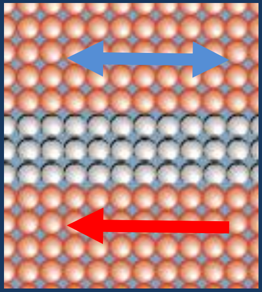
There

Enhanced MR is very general in very thin (1nm) layered structures:





Metallic spacer (Cu, Cr, ..): Giant MagnetoResistance

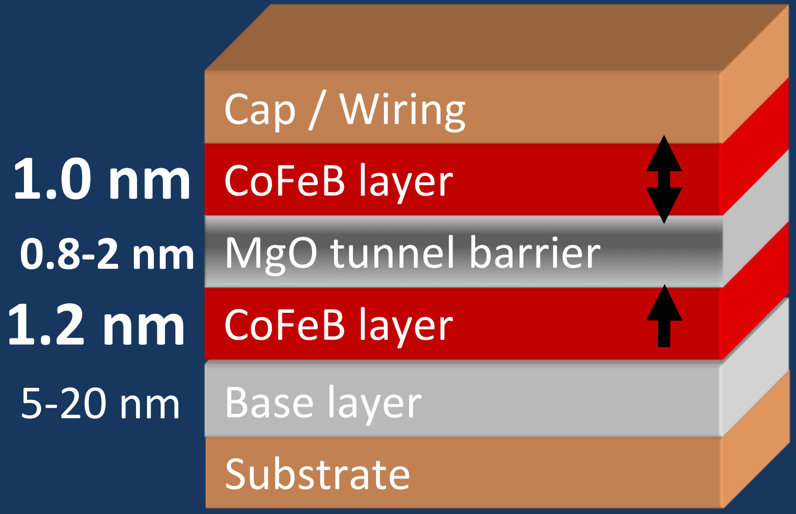


Insulating spacer (MgO, Al_2O_3, ..): Tunneling MagnetoResistance

Now, we have a RAM

The reference– ultrathin CoFeB

The samples:



MTJ stack sequence for this study:
pseudo-spinvalves with
ultrathin CoFeB and varying MgO

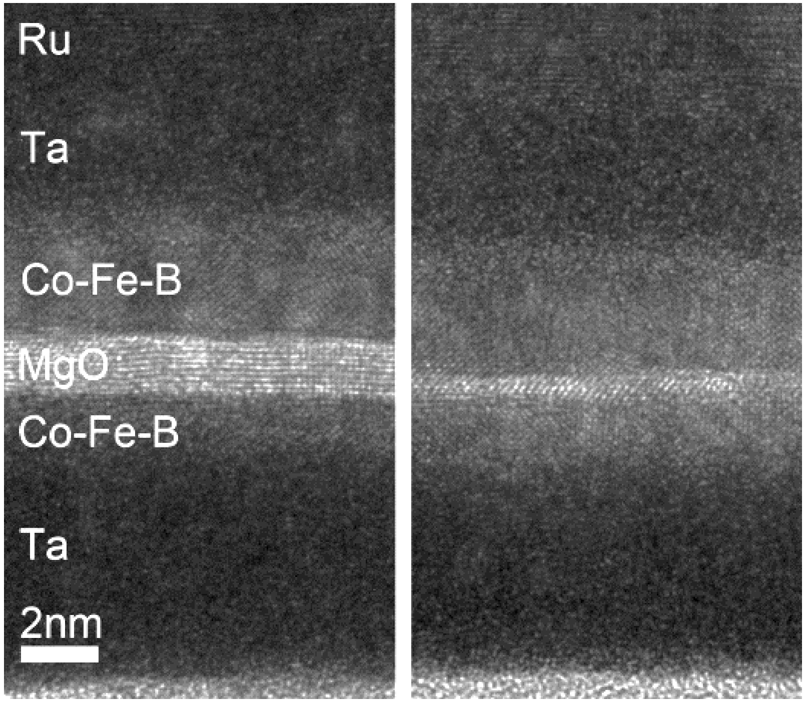


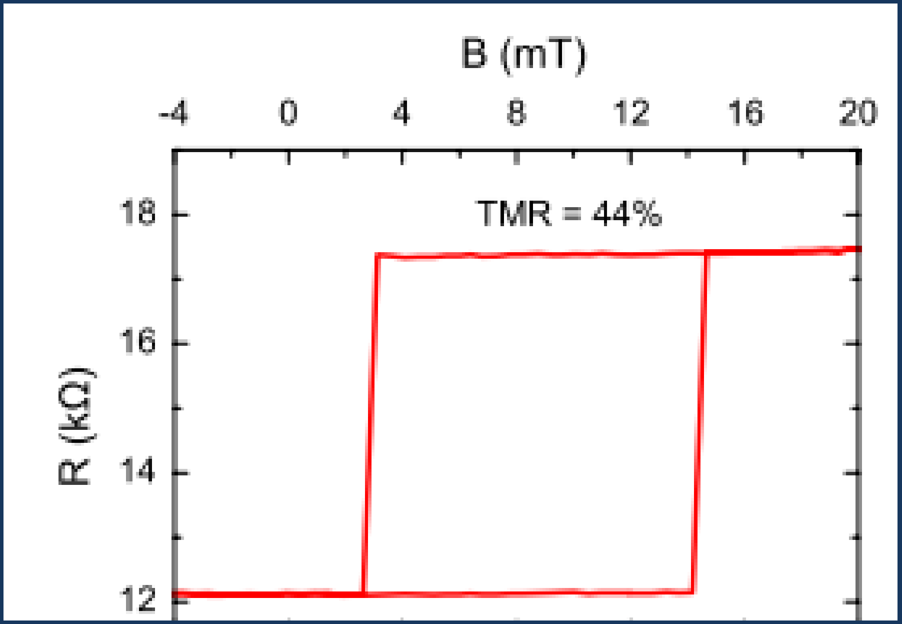
Fig. 1. HRTEM images of a thick 10 ML (left) and a heated 3 ML MgO barrier (right). The IQR values are $(5.6 \pm 1.5)^\circ$ (10 ML) and $(6.7 \pm 0.8)^\circ$ (3 ML).

Magnetic Tunnel Junctions with ultrathin CoFeB are perpendicular

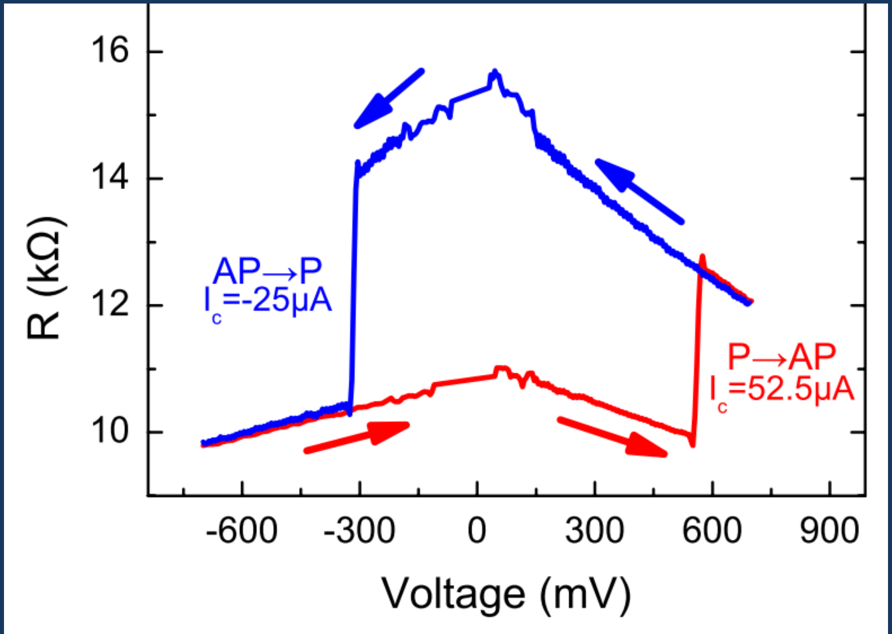
and work down to MgO barrier-thickness of 3 monolayers

The reference– ultrathin CoFeB STT switching

Results for STT-switching this ultrathin CoFeB/MgO/CoFeB system for low-RA MgO and small junctions:



Resistance vs. external magnetic field for perpendicular MTJs
1.0nm CoFeB / 4 ML MgO / 1.2nm CoFeB
gives around 40-50% TMR



RV-characteristic with an applied field of 8.6 mT

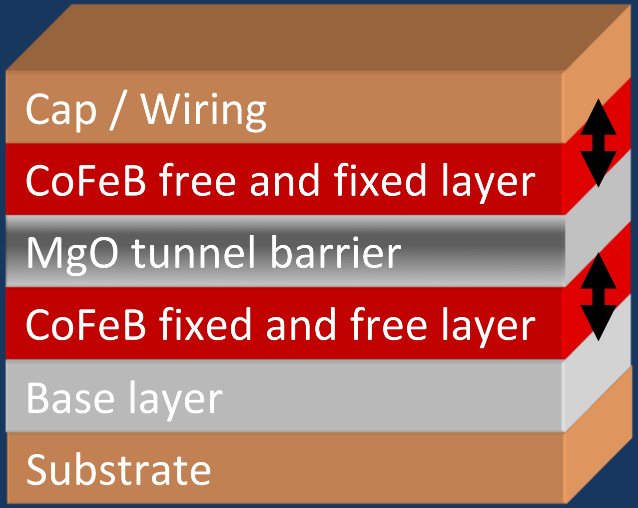
- average critical current density:

$2 \cdot 10^5$ A/cm^2 (!!)

Now we have a working and stable STT-MRAM

The reference – ultrathin CoFeB magnetic properties

Collaboration with
M. Münzenberg group



Cap / Wiring

CoFeB free and fixed layer

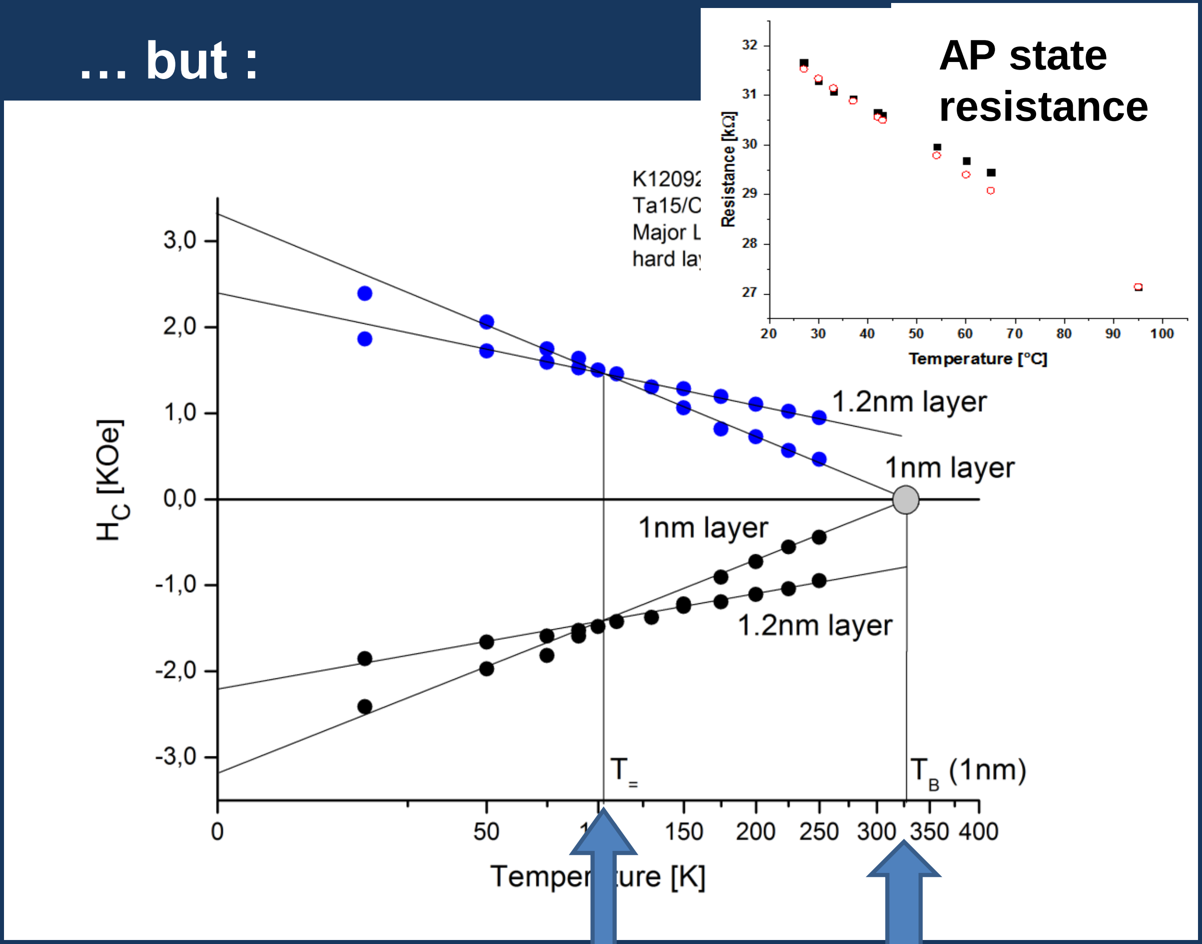
MgO tunnel barrier

CoFeB fixed and free layer

Base layer

Substrate

… but :



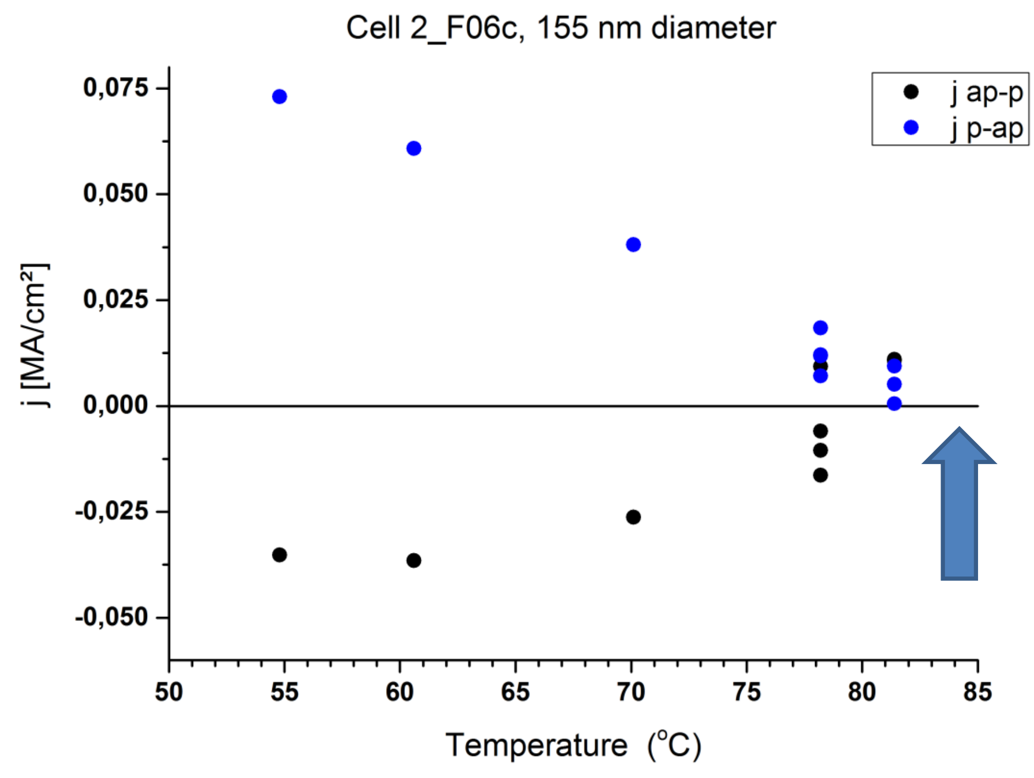
1. the layers change their role ..
low T: thick layer is the free layer
high T: thin layer is free

2. the thin layer shows $H_C^{\perp} = 0$
? anisotropy switches to ip *or*
? superparamagnetic

to be published

The reference – ultrathin CoFeB low current STT

Critical current for STT switching the MTJ as a function of temperature (collaboration with M. Münzenberg group)



Temperature for $H_C^{\perp} = 0$ from last page

– 1nm layer has „no anisotropy“

.. nearly zero critical current density possible by controlling temperature[1]

- ? good for TAS-STT-MRAM[2]

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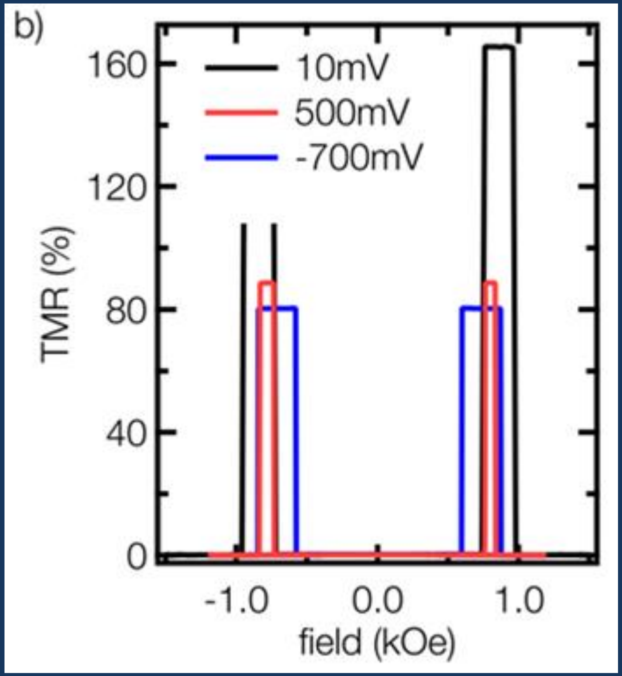
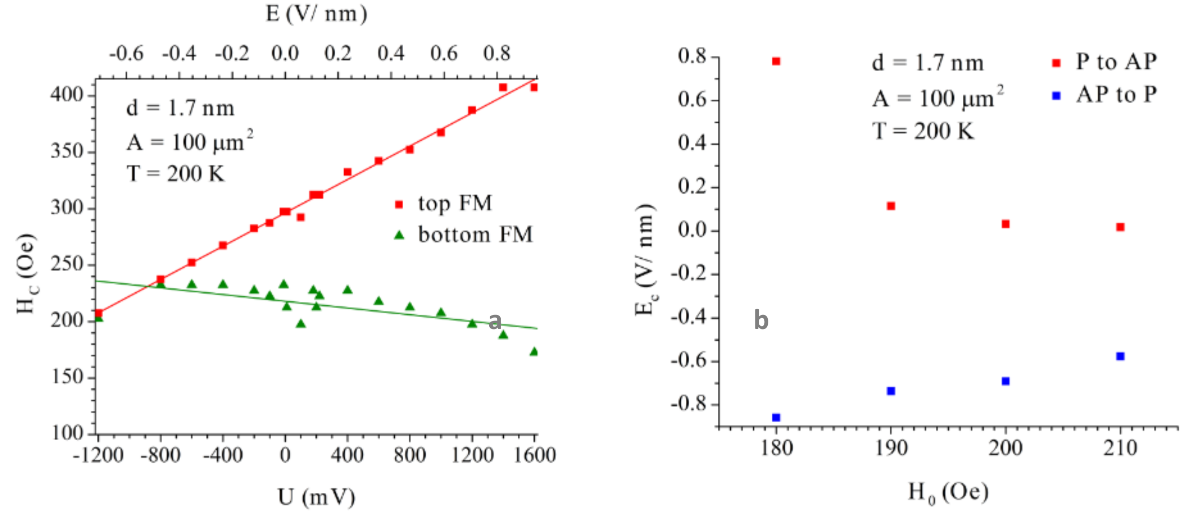
[1] to be published, [2] see I.L. Prejbeanu et.al., J. Phys. D: Appl. Phys. 46 (2013) 074002 (Spintec / Crocus)

The reference – ultrathin CoFeB E-field shift of H_c





(a) Dependence of the coercive fields of the electrodes of an MTJ on the applied voltage for the top (1nm thick) and bottom electrode (1.2nm thick) and

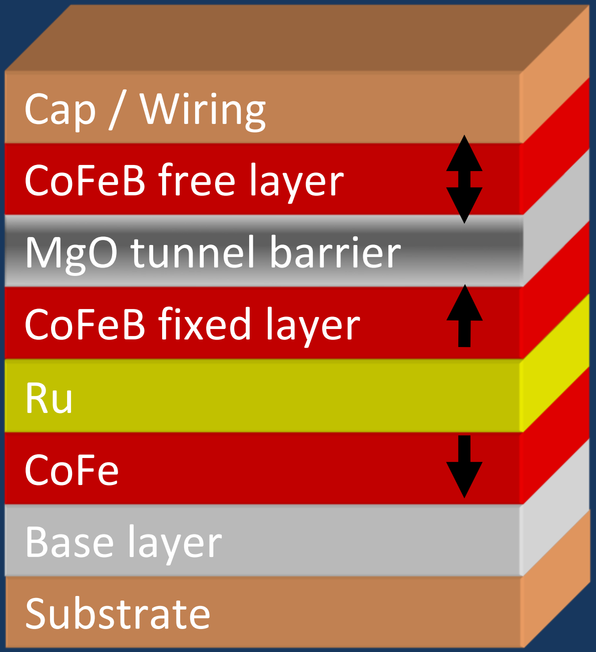
(b) Electric field leading to a magnetization switching at an applied bias field H_o (A. Gebauer, Bachelor thesis, publication in preparation)

.. H_c shift about four times larger for 1.0nm than for 1.2nm CoFeB

.. Open: temperature dependence, possible synergy with STT …

Why look for new Materials ?

But:

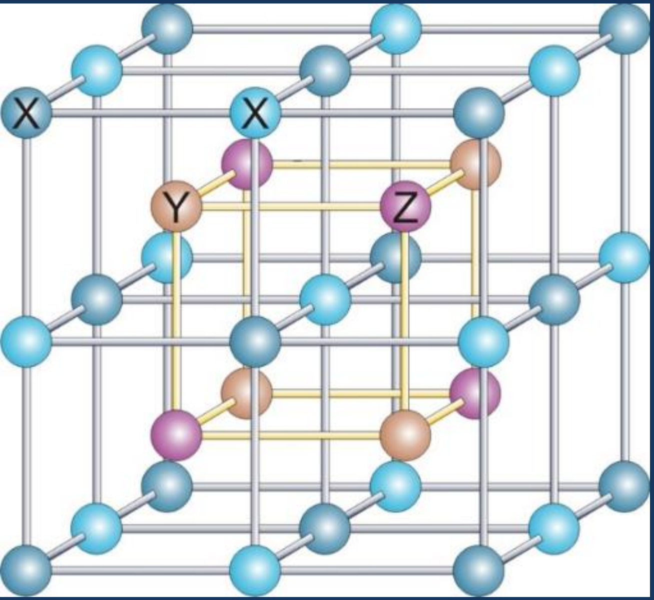


- → **„Ultrathin“ requires expensive equipment and process control**
- → **The perpendicular magnetization comes from the interfaces between MgO and CoFeB (sensitive property)**
- → **Thin CoFeB has a relatively large magnetic damping (speed issue !)**
- → **Base layer (antiferromagnet) is expensive**

→ **Narrow production window, relatively expensive, low speed**

→ **Need new materials !**

Heusler compounds (and related):



N. Tezuka et al., Appl. Phys. Lett. **94**, 162504 (2009)

P. Webster,J. Phys. Chem. Solids **32**, 1221 (1971)

Bielefeld University contributes > 20 “Heusler” publications since 2006

D. Ebke, PhD thesis (2010)

- X_2YZ composition
- crystallographic $L2_1$ structure
- high spin polarization / TMR ratios
- high Curie temperature T_C
- well known with in plane magnetization

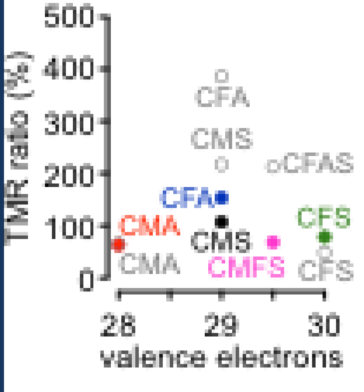
S. Kämmerer et.al., Appl. Phys. Lett. 85 (2004) 79

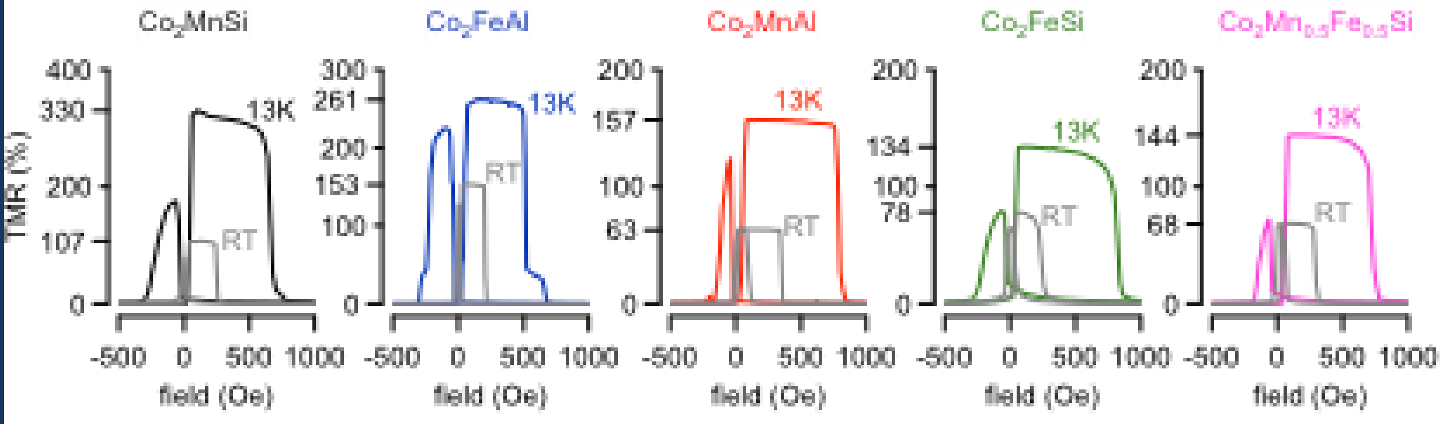
New Materials – Heusler compounds

General goals:

- thermal stability (KV > 50 – 60 k_BT)
- switching current low (0.1-1 MA/cm^2)
- TMR ratio (100 - 200% @ RT, better more)
- fast (nsec or faster)
- cheap, reliable, easy to prepare, …

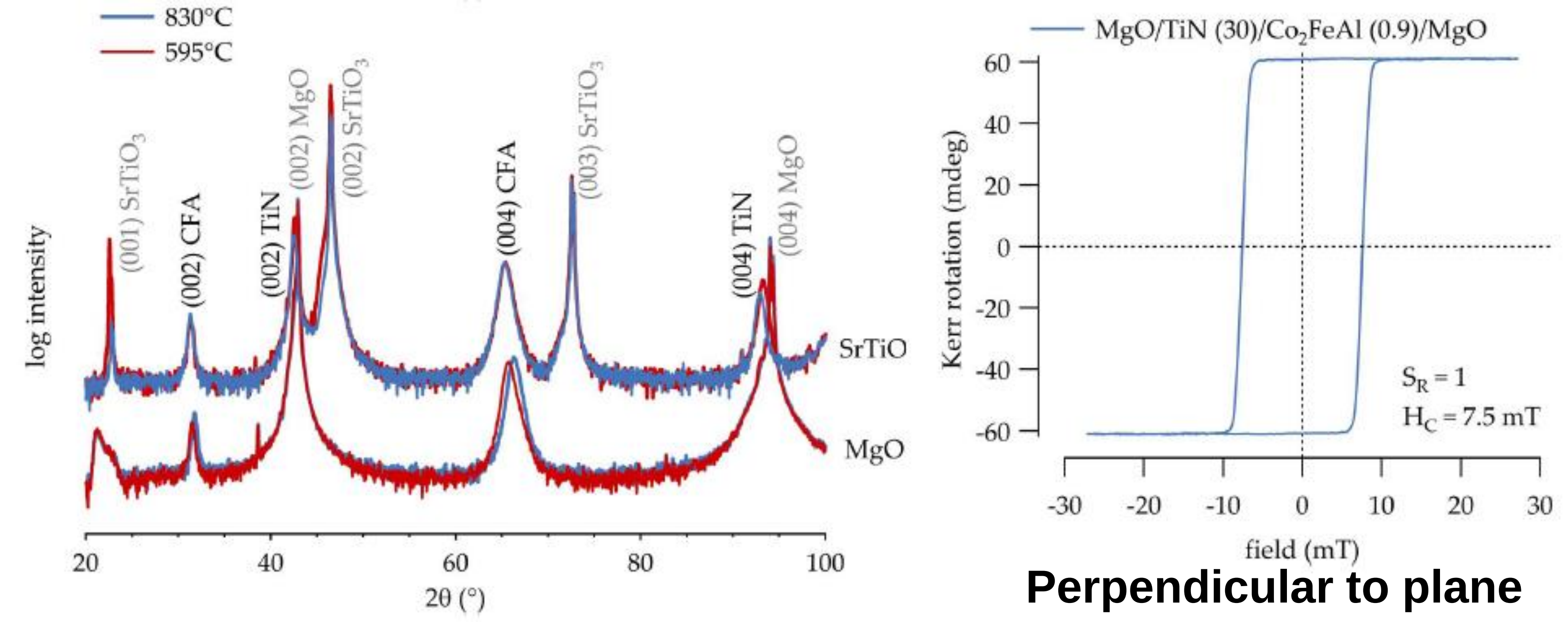
Heusler electrodes in TMR cells with in plane anisotropy





Very large TMR possible: H.X. Liu et.al., Appl. Phys. Lett. 101, 132418 (2012)
(2000% @ LT, 350% @ RT, Co_2Mn_xSi, x=1.3)

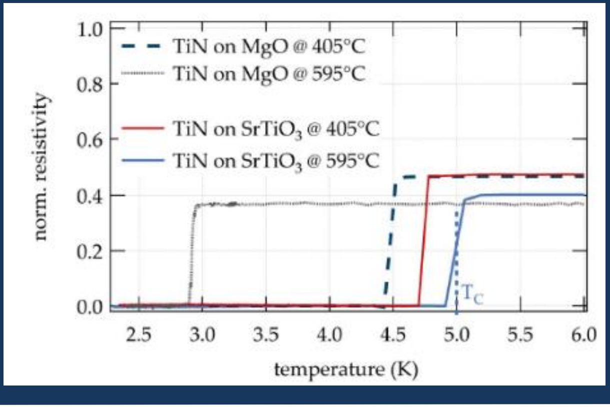
New Materials: Ultrathin Heuslers



Perpendicular to plane

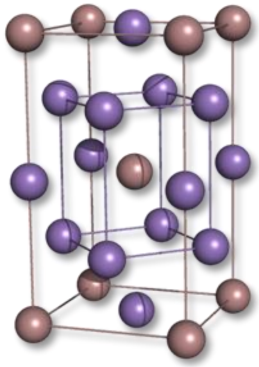
→ **Ultrathin Co_2FeAl Heusler compound on TiN gives smooth surface and perpendicular anisotropy**

→ **TiN is a good conductor, very stable (and superconducting)**

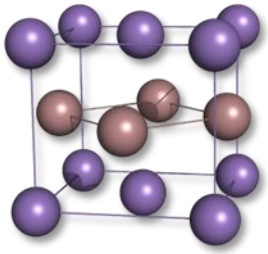


A. Niesen, in preparation

Heusler with perpendicular cryst. anisotropy: The Mn_xY family



$x = 0.1 .. 1.5$



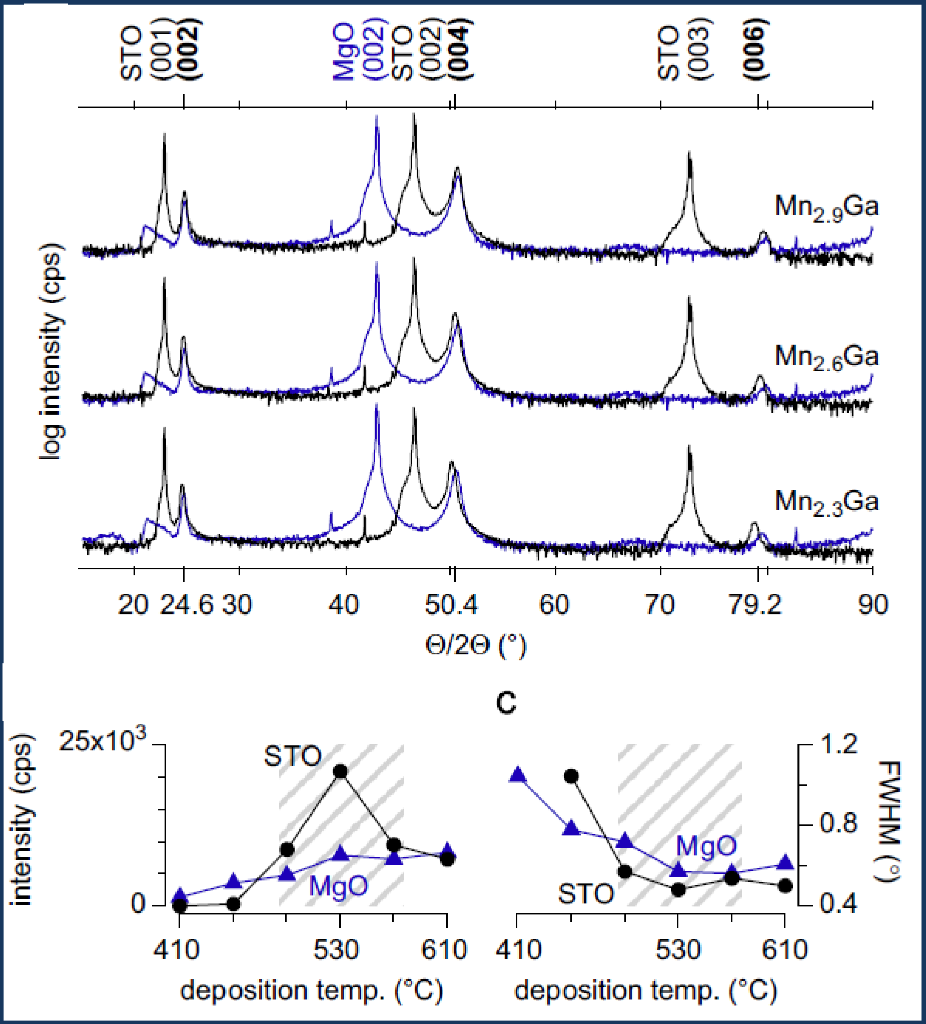
$x = 2$

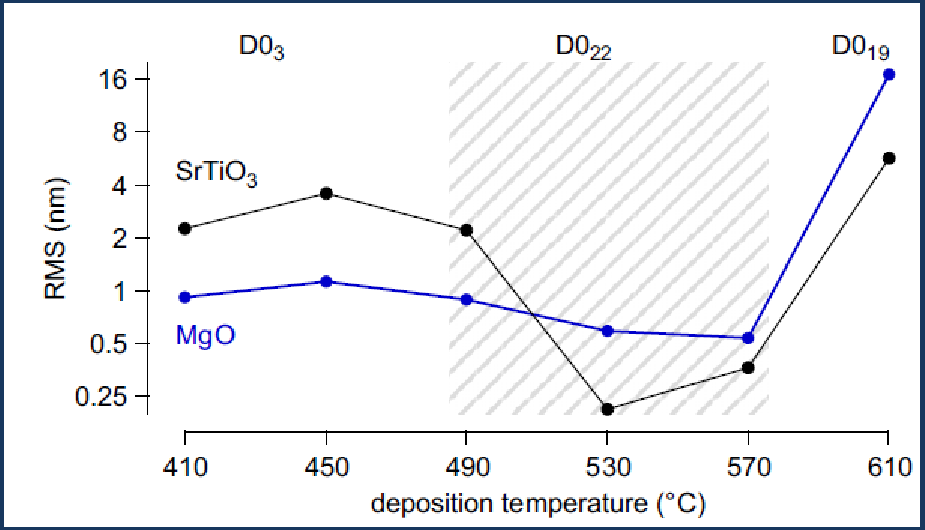
Example $Mn_{3-x}Ga$

- predicted high spin polarization (P=88%)
- perpendicular properties down to 5nm thickness
- high Curie temperature ($T_C \approx 770K$)
- low magnetic moment (about 0.26µB / f.u.)
- tunable magnetic behavior:

  H_C decreases with increasing x (leak of Mn)

New Materials: Mn_xGa



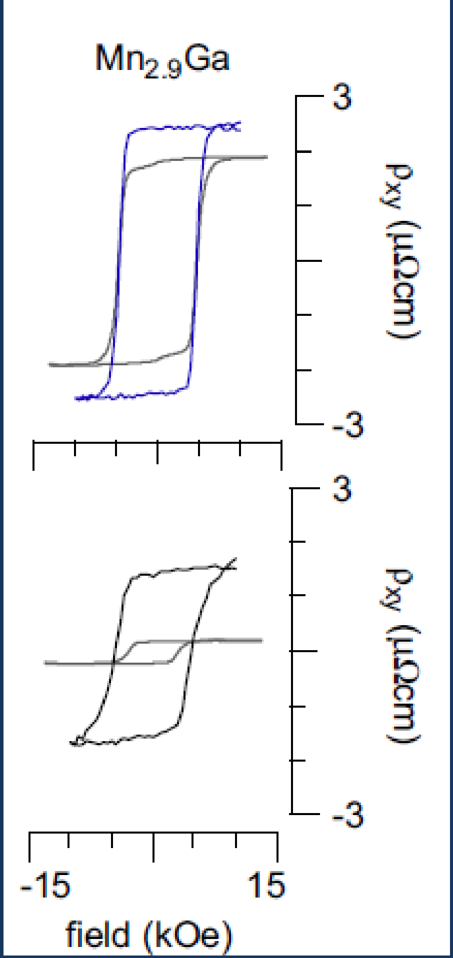


X-ray diffraction (left) of Mn_xGa

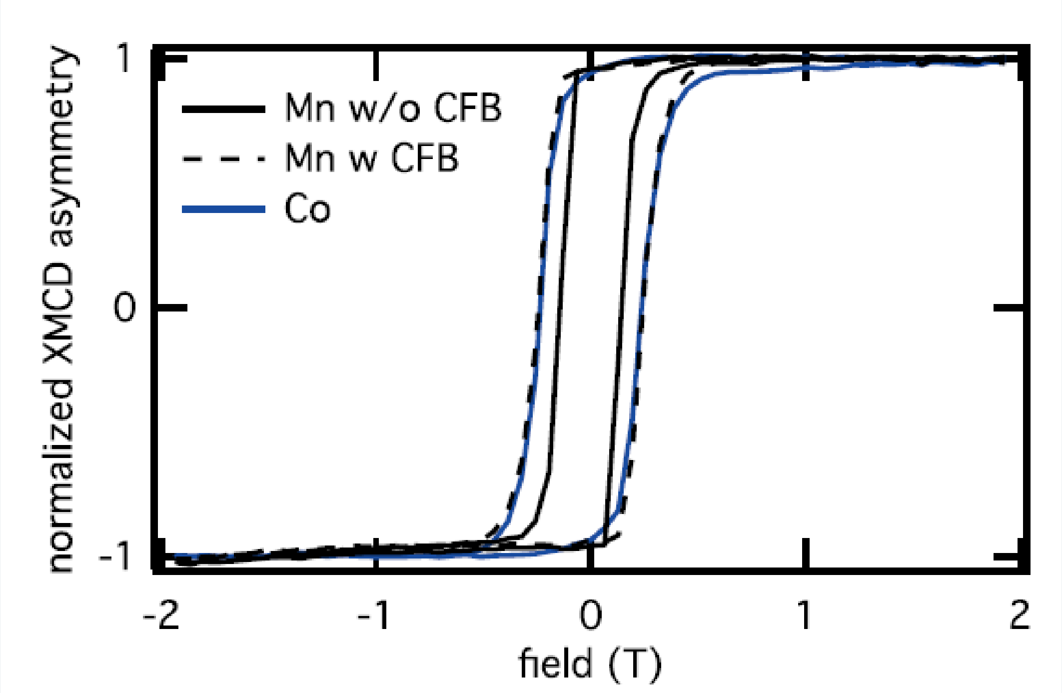
and

roughness (top) for $Mn_{2.9}Ga$

New Materials: Mn_xGa

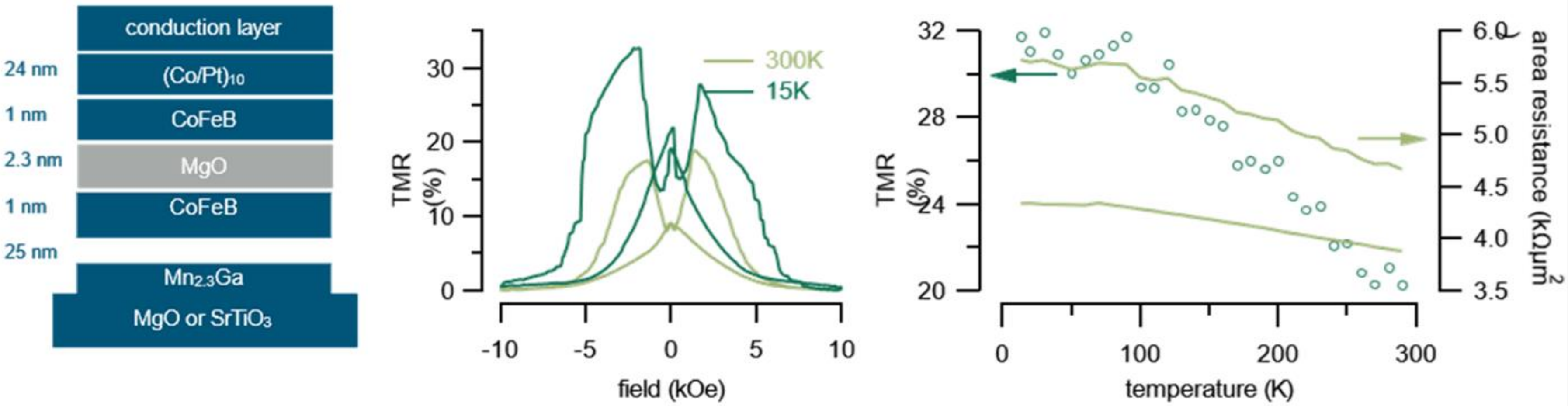


Overlay of magnetization (grey) and transversal Hall resistivity for $Mn_{2.9}Ga$ deposited on MgO (top) and STO (bottom) substrates at 530^{o}C.



Element-specific hysteresis loops.
Dashed black → normalized Mn XMCD asymmetry for a sample with (without) 1 nm CoFeB @ RT, normal incidence, out-of-plane magnetic field.
Blue curve → normalized Co XMCD asymmetry.

New Materials: Mn_xGa



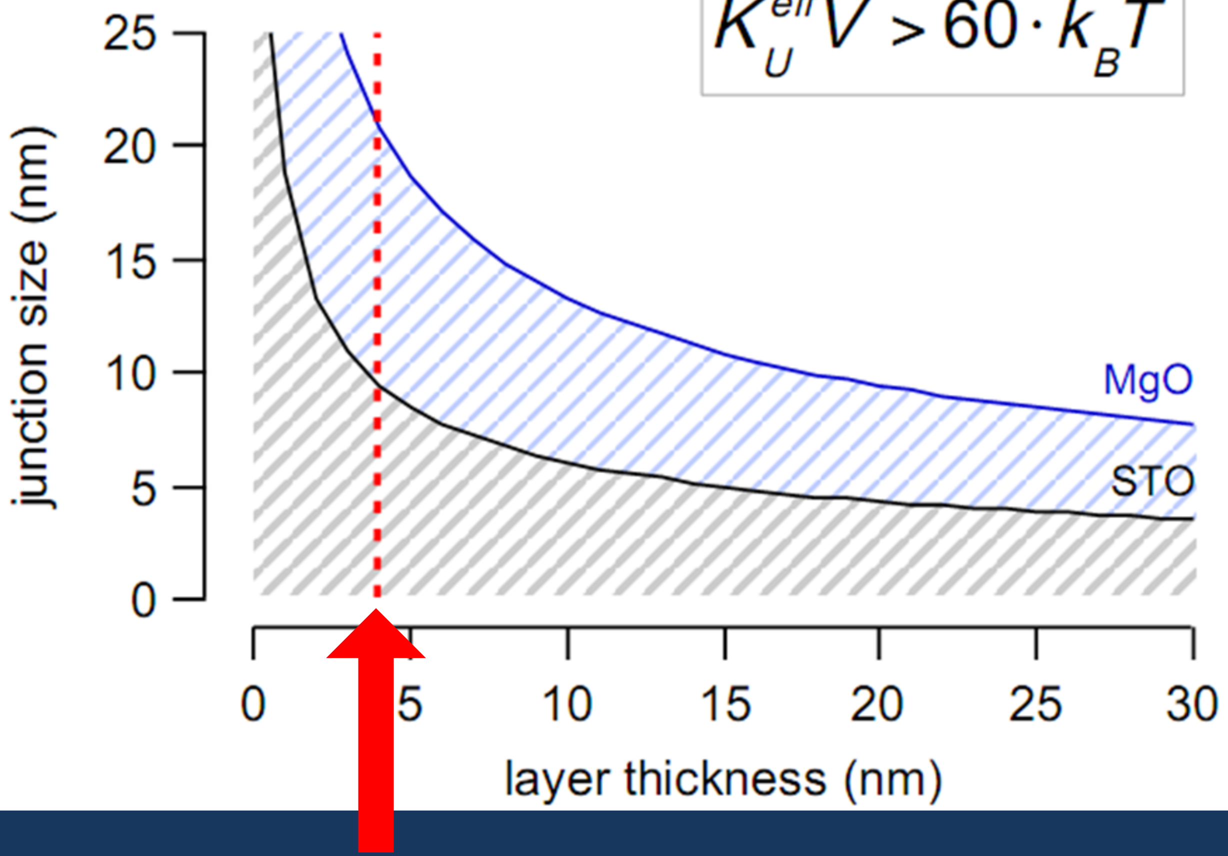
- first reported TMR effect for perpendicular Mn-Ga compound[1]
- highest TMR effect for samples with $\{Co/Pt\}_{10}$ multilayer counter electrode
- other groups reported higher TMR effects *for different* interlayer[2]
- no post-annealing process took place
- reasonable TMR ratio only for samples with ferromagnetic interlayer

[1] Glas, Integration of $Mn_{3-x}Ga$ Thin Films Into Magnetic Tunnel Junctions, Diploma Thesis, Bielefeld, 2012
[2] Ma et al., J. Appl. Phys. 114, 163913 (2013).

M. Glas, D. Ebke, I.-M. Imort, P. Thomas, GR, JMMM 333 (2013) 134

New Materials: Mn_xGa

$$K_U^{eff} V > 60 \cdot k_B T$$



assuming:

$T = 400K$

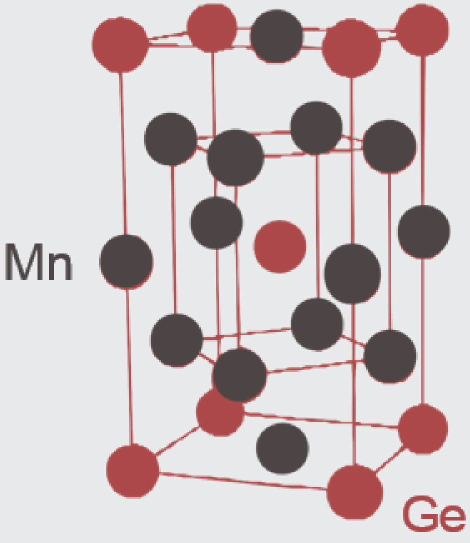
$K_u^{eff}(MgO) \approx 1.9 \cdot 10^6 \, erg/cm^3$

$K_u^{eff}(STO) \approx 9.4 \cdot 10^6 \, erg/cm^3$

Could work down to 5nm feature size !

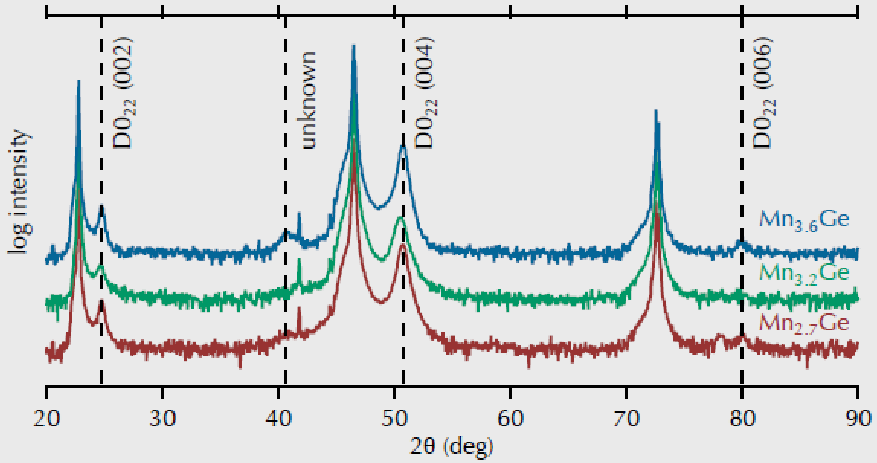
$Mn_{3\pm x}$Ge properties

- $Mn_{3\pm x}$Ge compound crystallises in $D0_{22}$ phase
- lattice constants:
  - a,b = 3.816Å
  - c = 7.261Å
- magnetic moment:
  - $0.4\mu_B$/f.u. corresponds to $175 kAm^{-1}$
- Anisotropy constant between 0.9 and $1.2 MJm^{-3}$ was reported
- spin polarisation of 46% via point contact Andreev reflection
- no single crystal phase was obtained by other groups



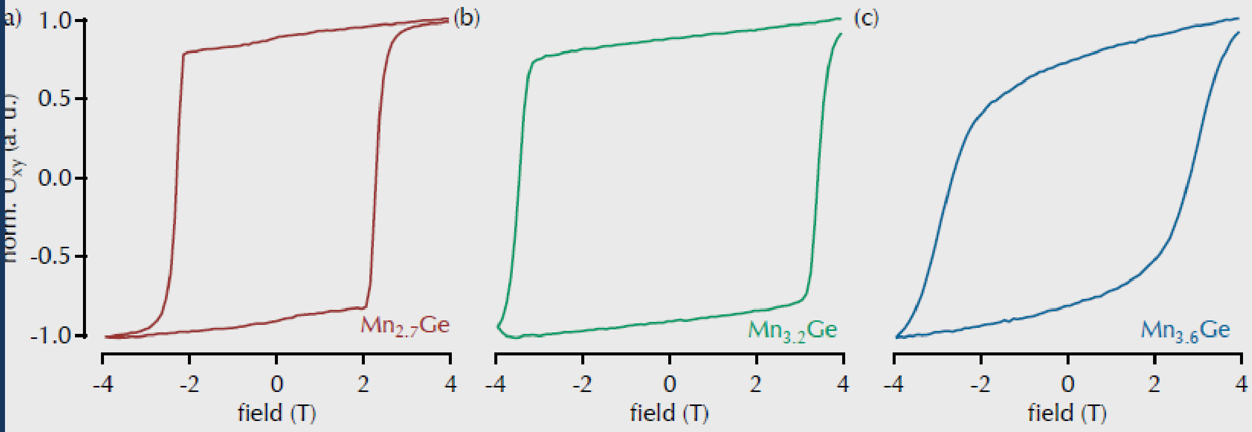
[3] Kurt et al., Appl. Phys. Lett. 101, 132410 (2012)

New Materials: Mn_xGe

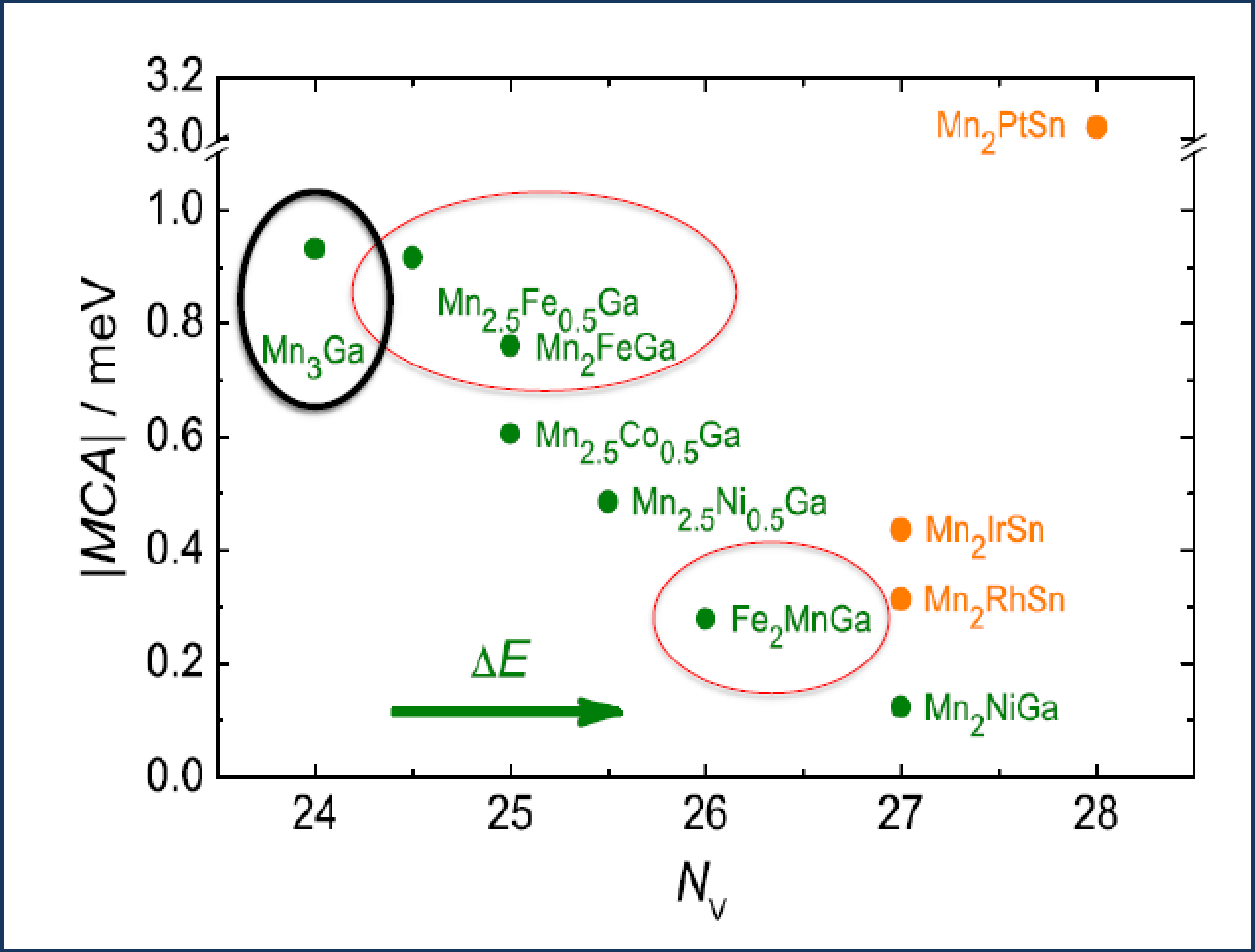


- $D0_{22}$ structure was achieved for different compositions
- Higher crystallinity for Mn rich samples
- Unknown reflex for the $Mn_{3.6}Ge$ sample

- Highest H_c of 3.25T for $Mn_{3.2}Ge$
- upcoming in-plane component for $Mn_{3.6}Ge$
- critical Mn content achieved



New Materials: MnFeGa



New Materials: MnFeGa

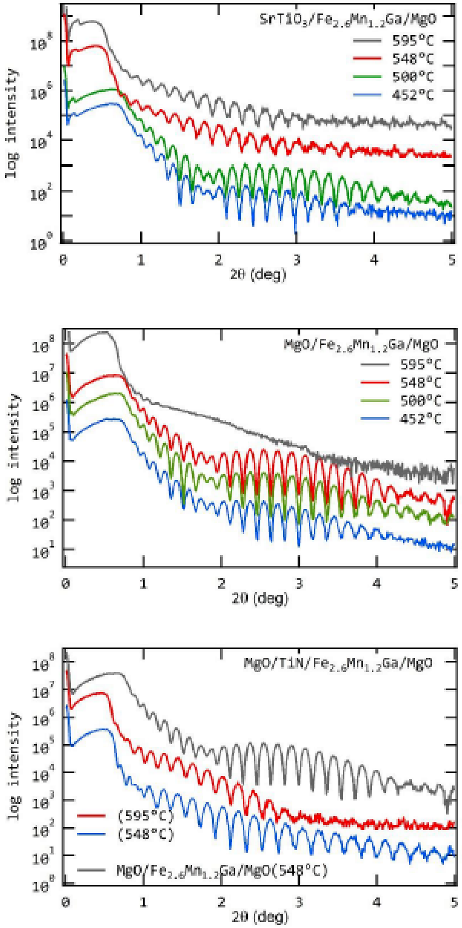
CRYSTALLINE PROPERTIES

- deposition time = 220 sec
- thickness: d ≈ 46 ± 0.4 *nm*
- roughness (measured via XRR):

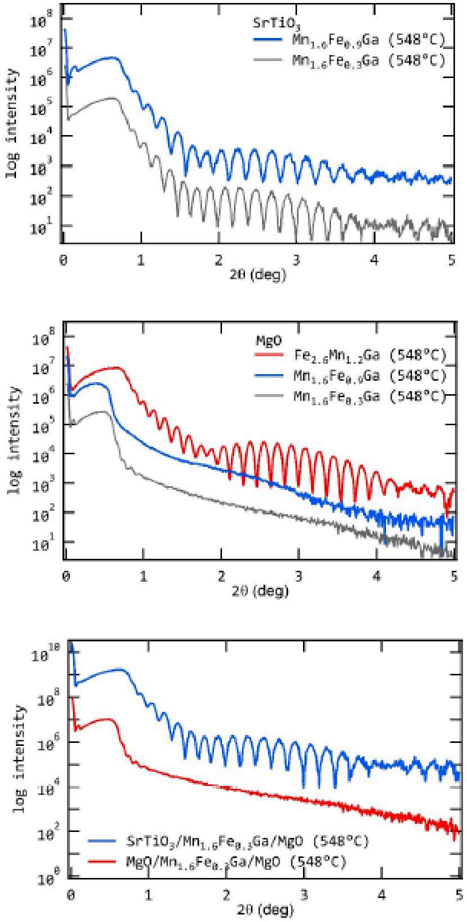
  ≈ 0.5 ± 0.05 *nm*

  (high Fe amount)
- low Fe amount leads to high roughness (measured by AFM)

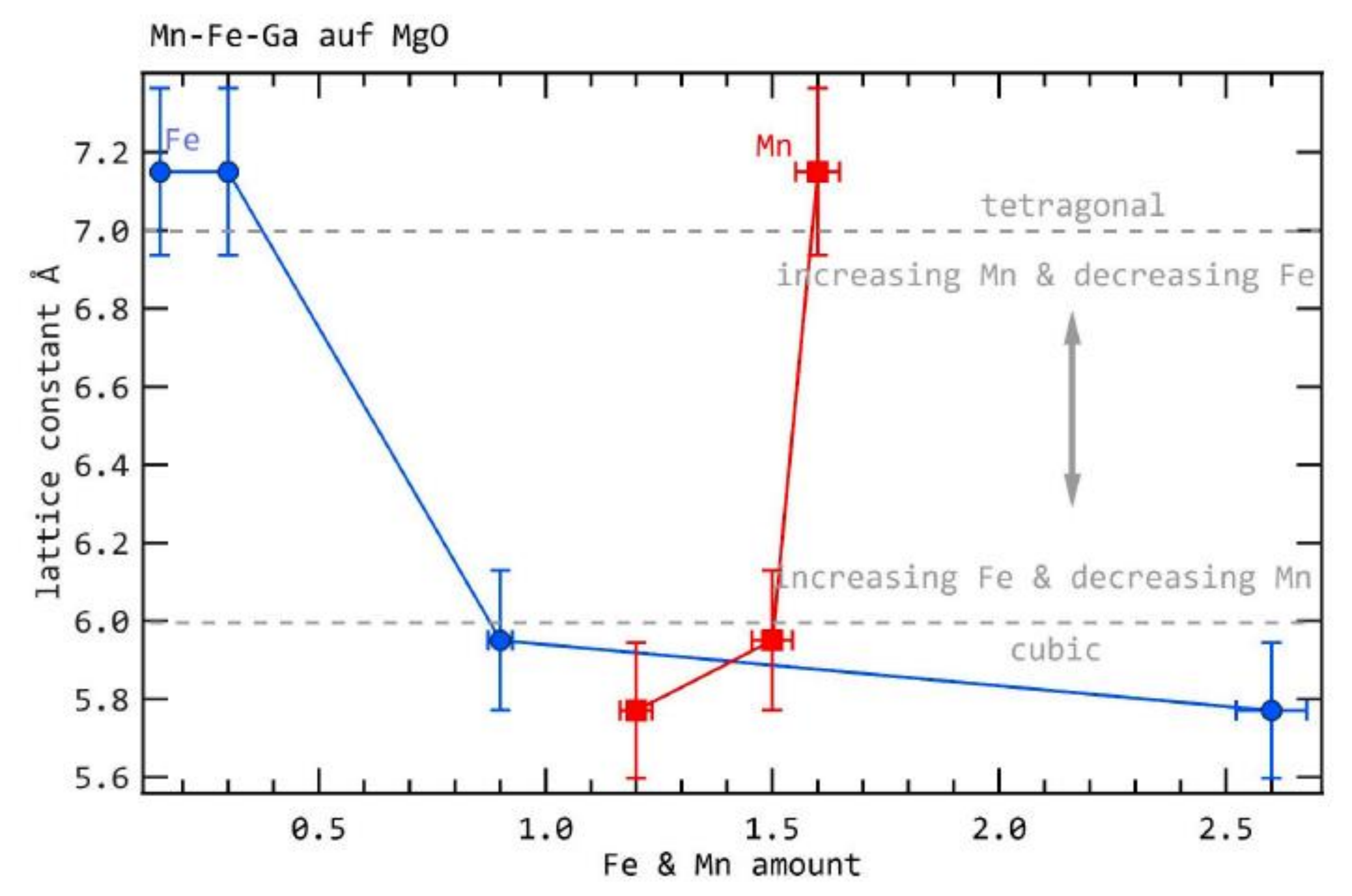
high Fe amount



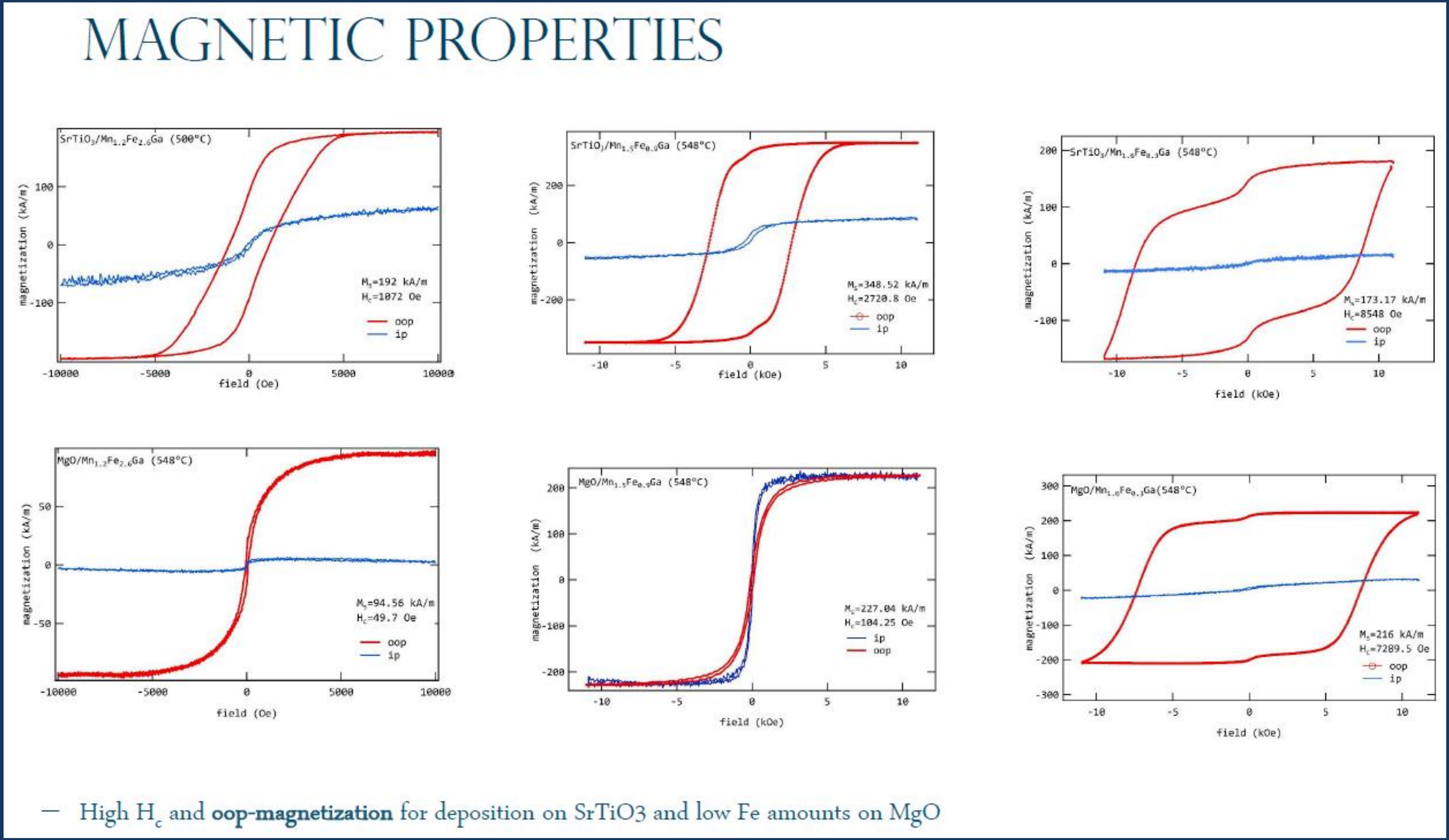
low Fe amount



New Materials: MnFeGa



New Materials: MnFeGa



MAGNETIC PROPERTIES

- High H_c and **oop-magnetization** for deposition on SrTiO3 and low Fe amounts on MgO

New Materials: MnN

Markus Meinert group:

MnN

of the „Mn_xY - family“ is anti-ferromagnetic and shows excellent exchange bias to CoFe:

A new exchange bias system !

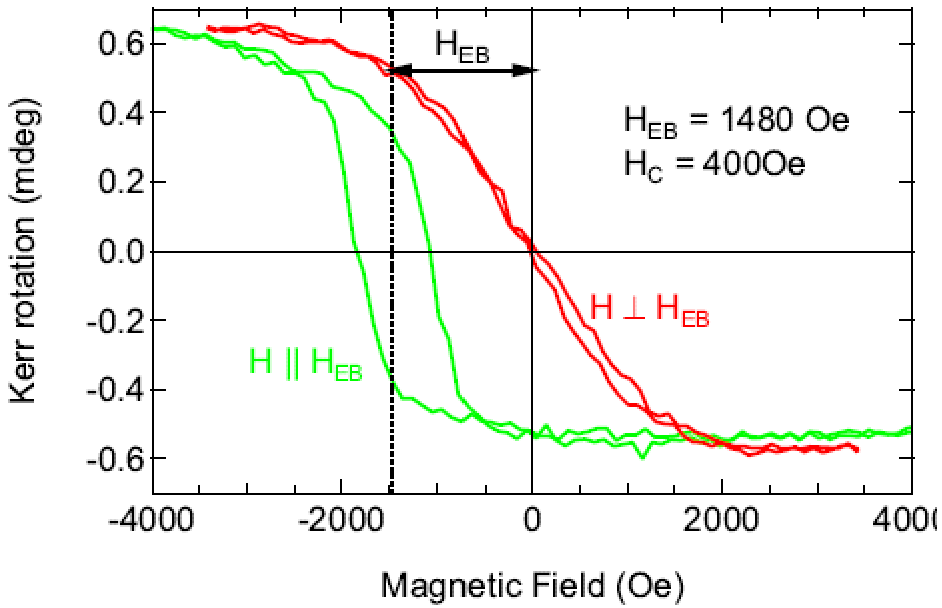


FIG. 1. **Hysteresis loops of a MnN/CoFe stack at RT.** The film thicknesses were $t_{\mathrm{MnN}} = 30\,\mathrm{nm}$ and $t_{\mathrm{CoFe}} = 1.5\,\mathrm{nm}$. The sample was annealed at 325° for 15 min and field cooled. The definition of the exchange bias field H_{EB} is shown. With the external magnetic field perpendicular to the exchange biasing field no hysteresis shift is observed.

MnN grown by reactive sputtering with Ar/N_2 mixture

Can be grown on single crystalline substrates and on Si/SiO_2

See M. Meinert, M. Dunz, arXiv:1501.05162v1 [cond-mat.mtrl-sci]

New Materials: MnN

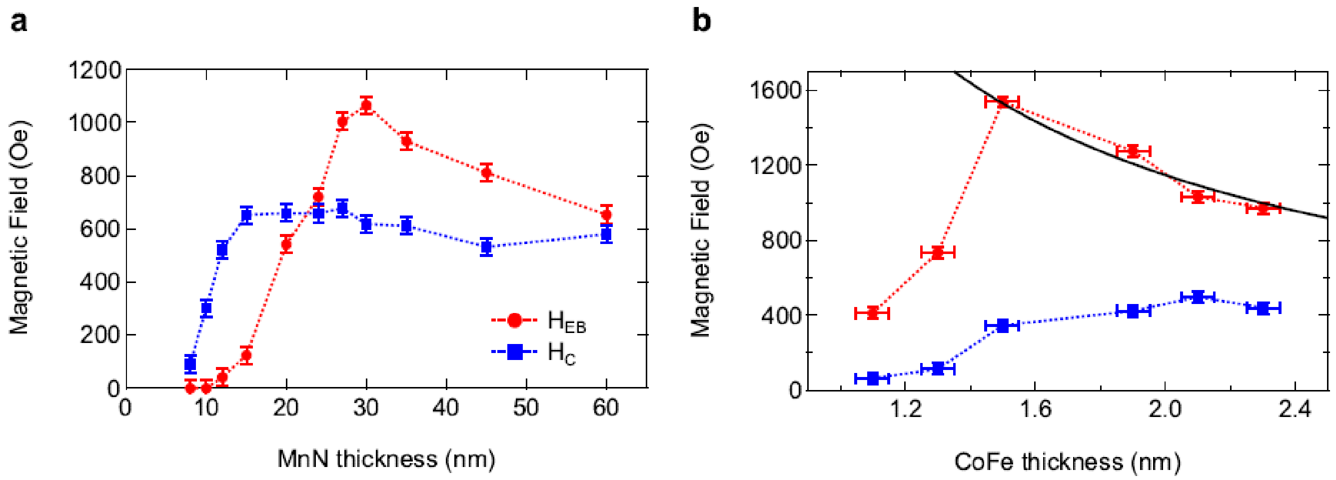
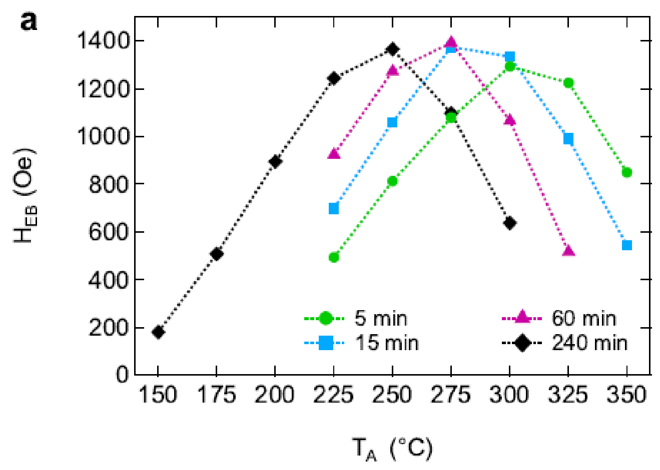


FIG. 2. **Dependence of exchange bias field and coercive field on the film thicknesses.** **a**, The dependence of H_{EB} and H_C on the MnN thickness. The CoFe thickness was 2.0 nm. **b**, The dependence of H_{EB} and H_C on the CoFe thickness. The MnN thickness was 30 nm. The samples were annealed at 325° for 15 min. Dotted lines are guides to the eye throughout this letter.



Dependence of the exchange bias field on annealing temperature and duration. The samples with MnN 30nm / CoFe 1.9nm were annealed and field cooled at temperature T_A for different times t_A.

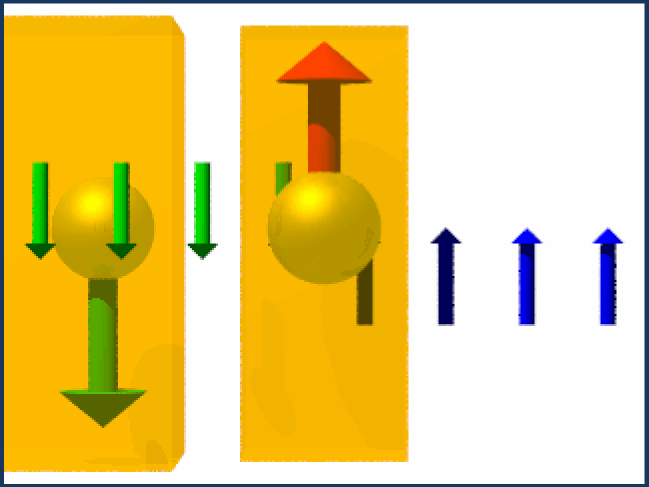
a, Samples successively heated for t_A with increasing temperature T_A.

b, Same data, parametrized with annealing temperature T_A.

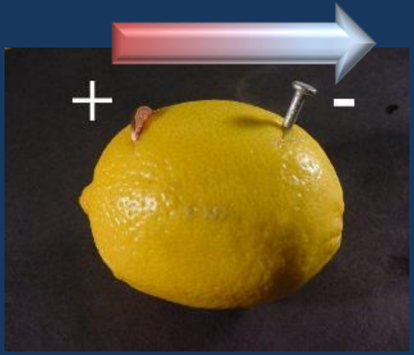
See M. Meinert, M. Dunz, arXiv:1501.05162v1 [cond-mat.mtrl-sci]

Spincaloritronics

Remember: Spin Transfer Torque:



Spin Current is carried by electrons
Electrons are driven by voltage

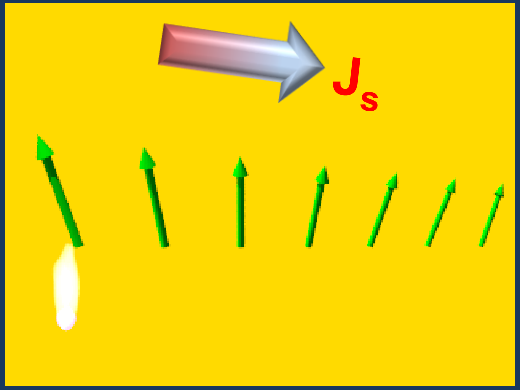


$$\vec{E} = -\nabla\Phi(\vec{r})$$

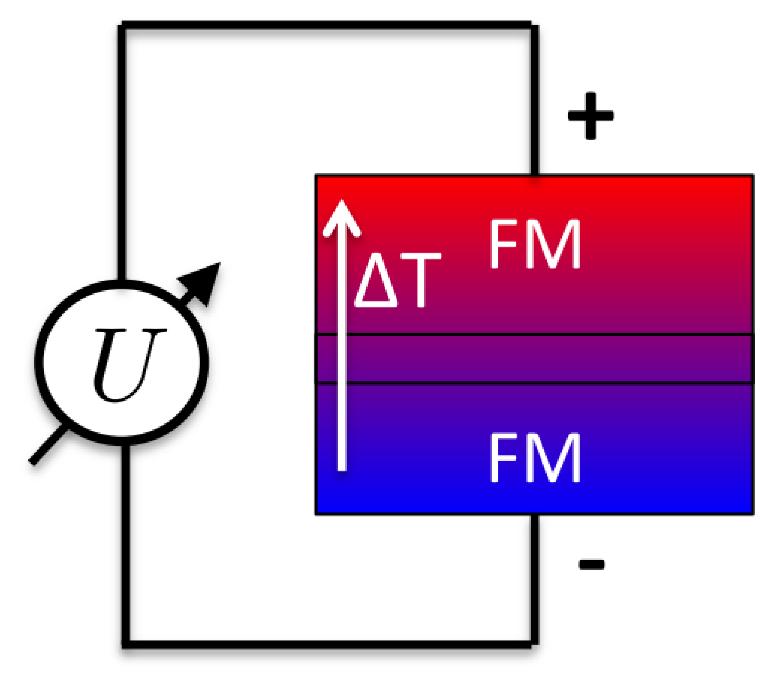
Can we drive Spin Currents also by other external forces?

YES: Temperature differences

$$\Delta T = \nabla T(\vec{r})$$



Spincaloritronics: MTJs



Seebeck coefficient for such tunnel devices:

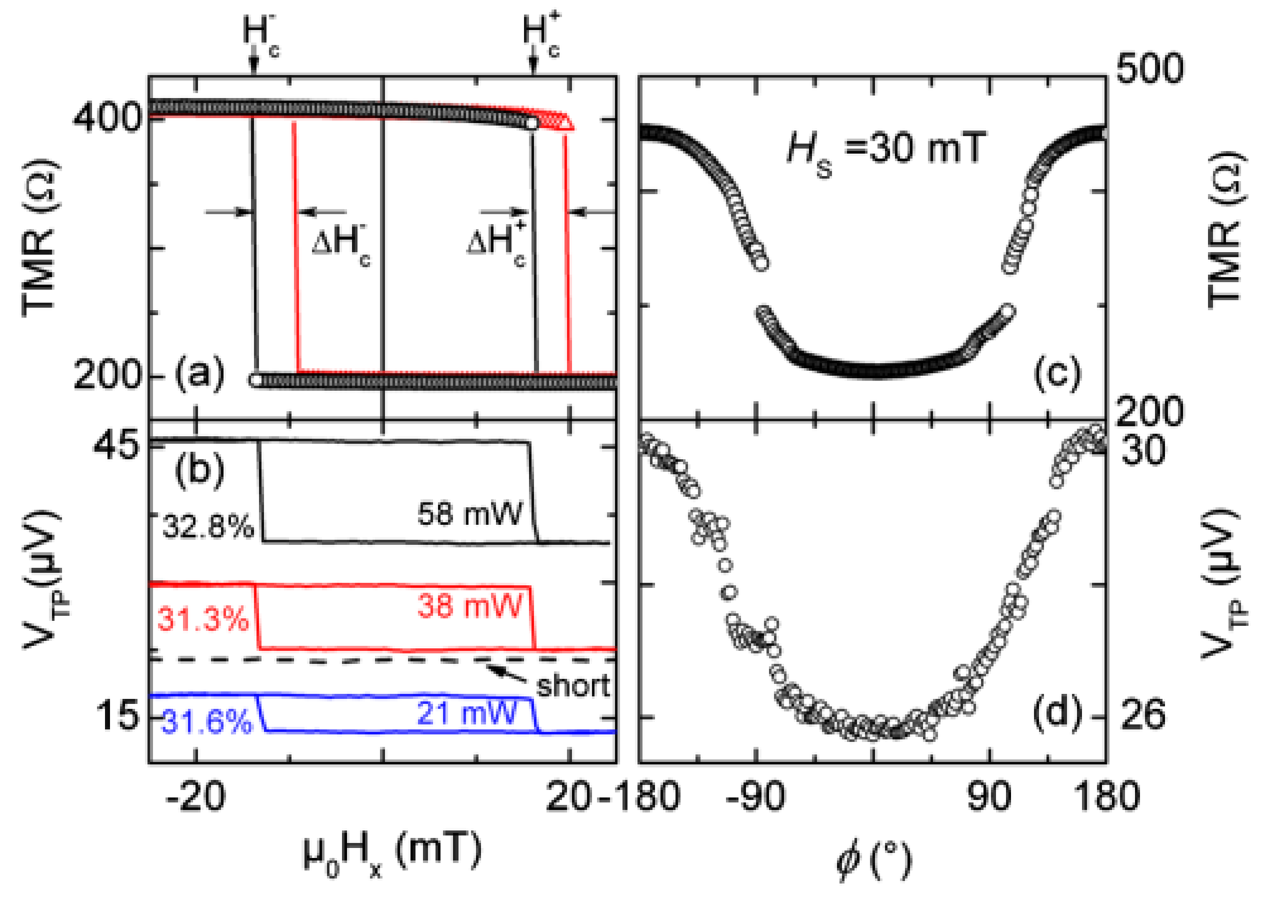
$$S = \frac{\int T(E)(E-\mu)\left(-\partial_E f(E,\mu,T)\right)dE}{e\,T\int T(E)\left(-\partial_E f(E,\mu,T)\right)dE}$$

$\partial_E f(E,\mu,T)$: Derivative of occupation function

Thermovoltage should depend on magnetization directions

Important: S unequal to conductivity $g = \frac{e^2}{h}\int T(E)\left(-\partial_E f(E,\mu,T)\right)dE$

Spincaloritronics : MTJs



Tunneling Magnetoresistance

and

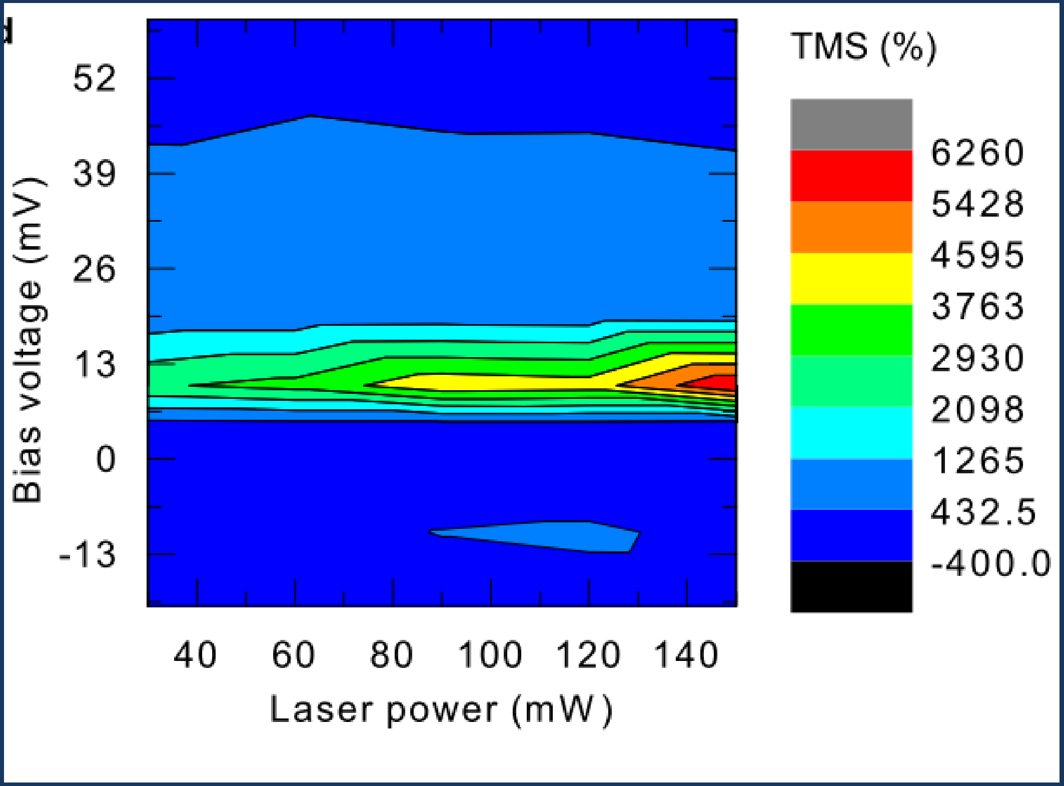
Thermovoltage

of Magnetic Tunnel Junctions

M. Walter et.al., Nature Materials, 10 (2011) 742
(Münzenberg group Göttingen)

N. Liebing et.al., Phys. Rev. Lett. 107 (2011) 177201
(Schumacher group Braunschweig)

Spincaloritronics : MTJs



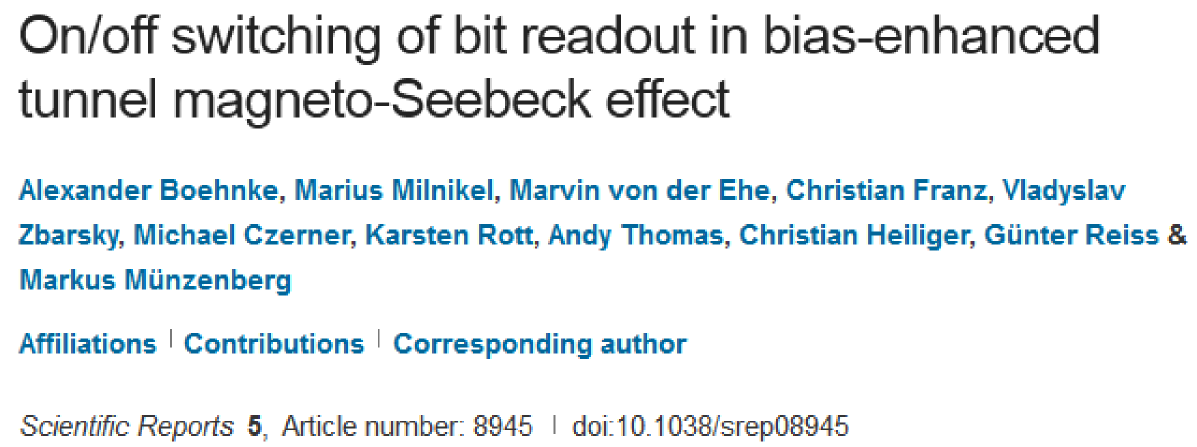
Additional bias voltage at

CoFeB/MgO/CoFeB

tunnel-junctions:

Tunnel Magneto Seebeck effects > 6000%

.. ongoing work



On/off switching of bit readout in bias-enhanced tunnel magneto-Seebeck effect

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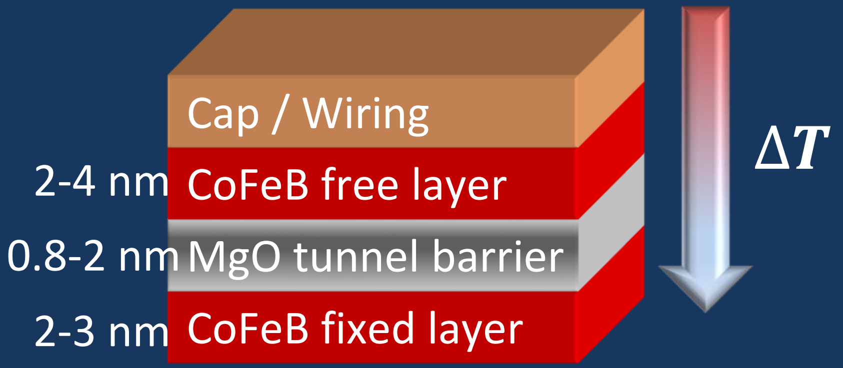
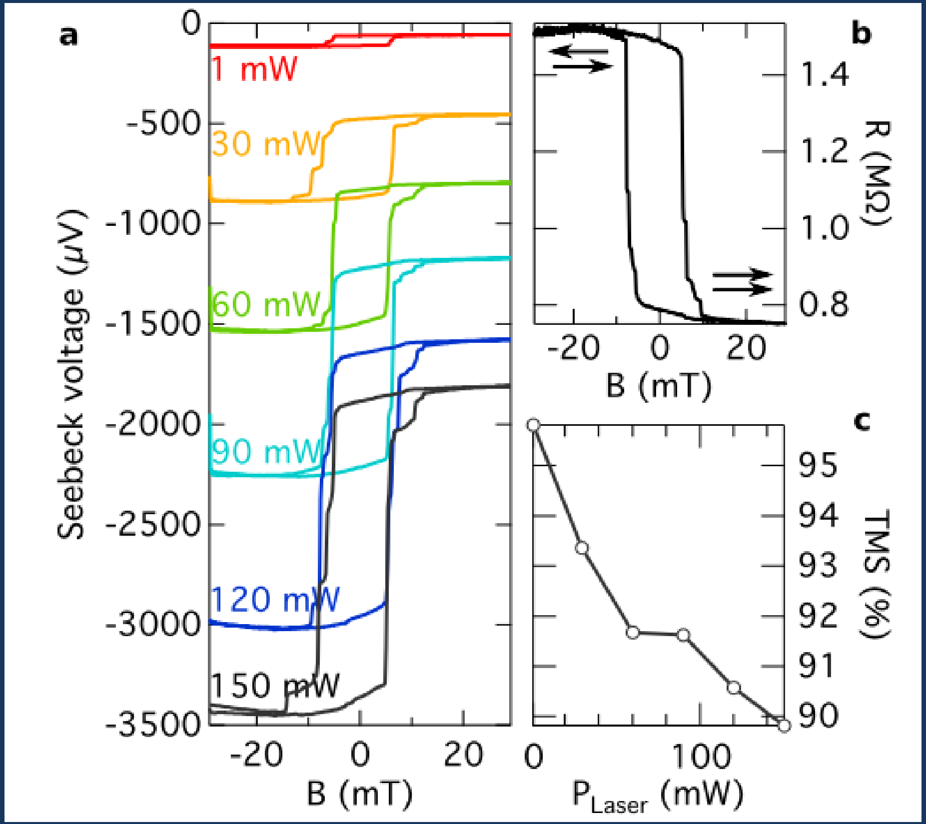
Scientific Reports **5**, Article number: 8945 | doi:10.1038/srep08945

Spincalorics : MTJs + Heuslers



replace by highly spin polarized Heusler alloy

→ Co_2FeSi



Gap in one spin direction should increase not only TMR but also

$$S = \frac{\int T(E)(E-\mu)\left(-\partial_E f(E,\mu,T)\right)dE}{e\,T\int T(E)\left(-\partial_E f(E,\mu,T)\right)dE}$$

(large asymmetry of DOS at E_F)

a) TMS reaches 90 … 96 % comparable to TMR (b)
c) Dependence of TMS ratio on applied laser power.

… ongoing experimental and theoretical work

Thanks

All coworkers in Bielefeld

Siemens AG, Sensitec, Prema,
Qiagen, Singulus, …

M. Münzenberg, Göttingen
H.-W. Schumacher, PTB Braunschweig
S. Demokritov, Münster
H. Ebert, München
C. Felser, G. Jacob, G. Fecher, Mainz
B. Hillebrands, Kaiserslautern
J. Moodera, Cambridge
C. Chappert, Paris
R. Cowburn, London, …

.. and you for listening !

Funding:

BMBF (German Ministry of Education and Research)
DFG (German Research Society)
NRW (Northrhine Westfalia)
European Commission
Thyssen Krupp Foundation
Humboldt Foundation