ハーフメタルのスピントロニクスデバイス応用と軟X線MCD解析

- Applications of half-metals to spintronics devices and utilizations of soft X-ray magnetic circular dichroism -

桜庭裕弥, 岩瀬拓, 高梨弘毅　東北大学金属材料研究所
Y. Sakuraba, T. Iwase, K. Takanashi　*Institute for Materials Research (IMR), Tohoku university*

平塚喬士, 窪田崇秀, 大兼幹彦, 安藤康夫　東北大学工学研究科
T. Hiratsuka, T. Kubota, M. Oogane, Y. Ando　*Graduate School of Engineering,Tohoku university*

児玉謙司, 中村哲也,　高輝度光科学研究センター/SPring-8
K. Kodama, T. Nakamura　*JASRI/Spring-8*

IMR TOHOKU UNIVERSITY

Outline

- ***Introduction*** *: Half-metallic materials*
  Half-metallic Heusler alloys

- ***Experiments***

  Experiment 1 *Co_2MnSi-based magnetic tunnel junctions*

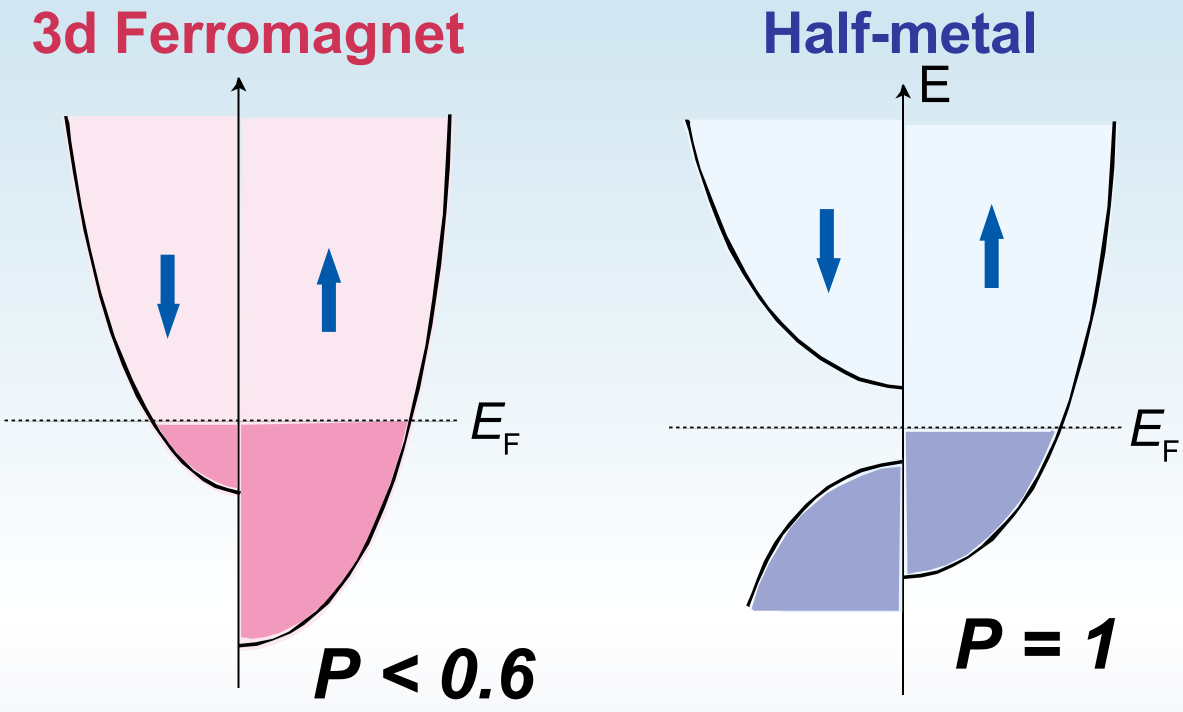
  Experiment 2 *MTJs with $L1_0/L2_1$-ordered alloy hybrid electrodes*

  Experiment 3 *Half-metallic Ferri-magnets : Mn_2VAl*

  Experiment 4 *Co_2MnSi-based CPP-GMR devices*

- ***Summary***

Half-metallic materials



Spin-polarization

$$P = \frac{D_\uparrow(E_F) - D_\downarrow(E_F)}{D_\uparrow(E_F) + D_\downarrow(E_F)}$$

Candidates of half-metallic materials

Oxides : CrO_2, Fe_3O_4, $La_{2/3}Sr_{1/3}MnO_3$ *etc*.

Zinc-blend type compounds: CrAs, MnAs *etc*.

Half-Heusler alloys: NiMnSn, PtMnSb *etc*.

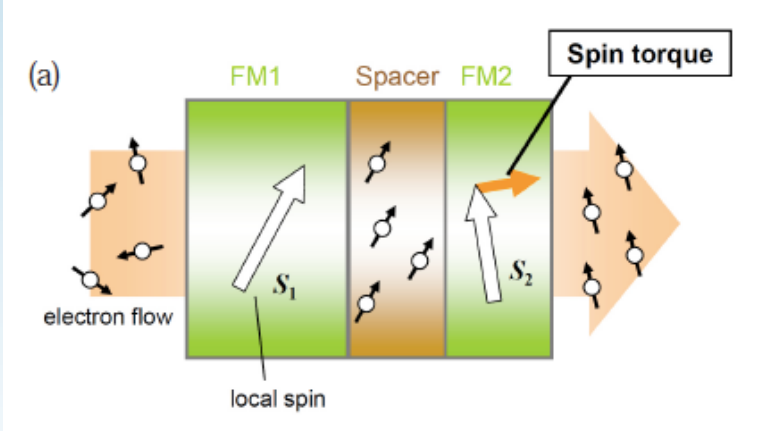
Full-Heulser alloys : Co_2MnSi, Co_2MnGe, etc.

Applications of half-metallic materials

TOHOKU

- **MTJs & CPP-GMR devices**

$$TMR\ \ ratio = \frac{2P_1P_2}{1-P_1P_2} \times 100\ (\%)$$



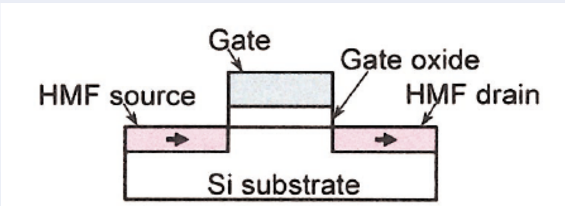
Spin-torque induced magnetization switching

$$J_C \propto \frac{\alpha}{g(\theta)} M_S^2 t$$

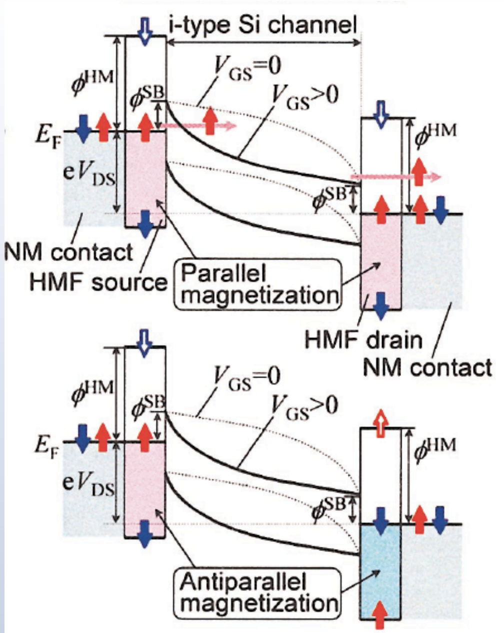
Spin-injection efficiency : $g(\theta) = P_1/(1+P_1P_2\cos\theta)$

- **Spin-Transistor**

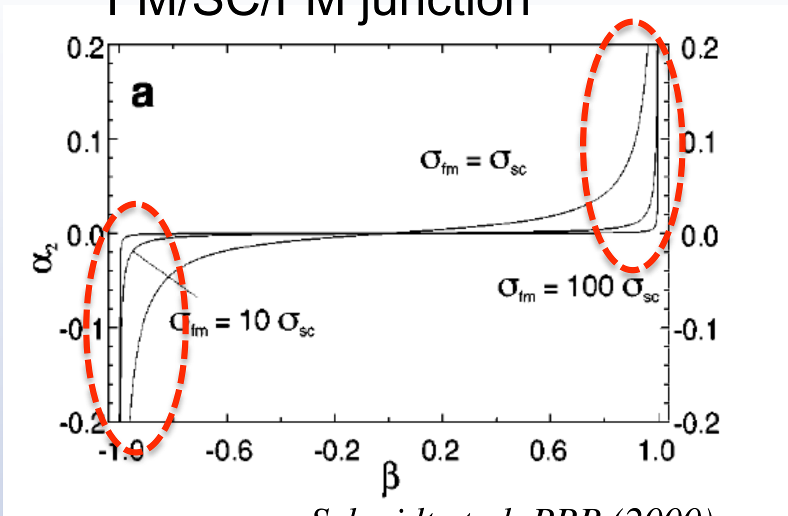
Spin-MOSFET based on
Half-metallic source and drain



Sugahara et al. APL (2004)



FM/SC/FM junction



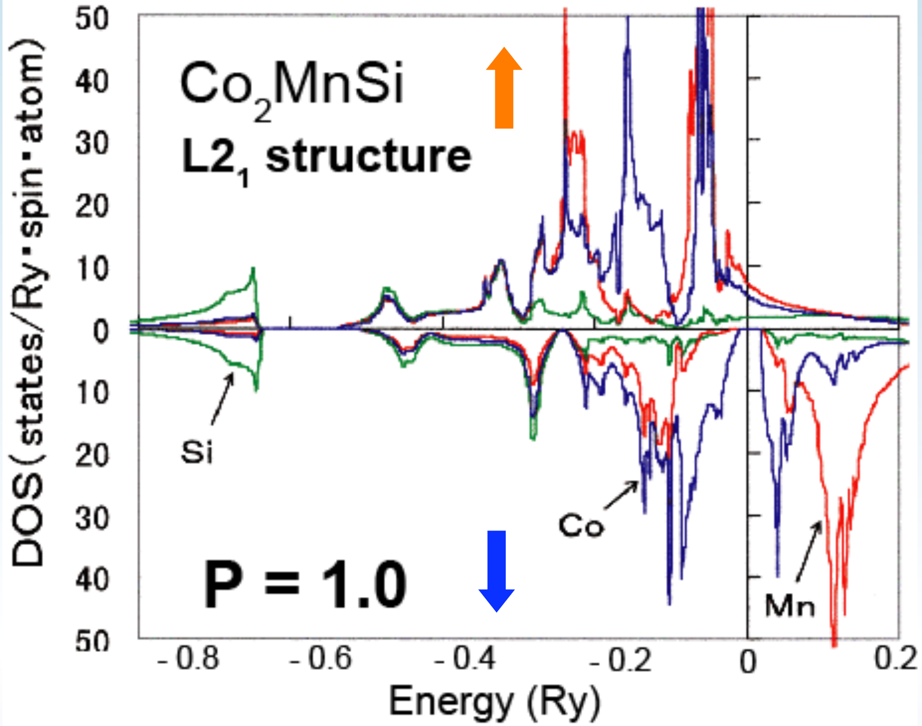
Schmidt et al. PRB (2000)

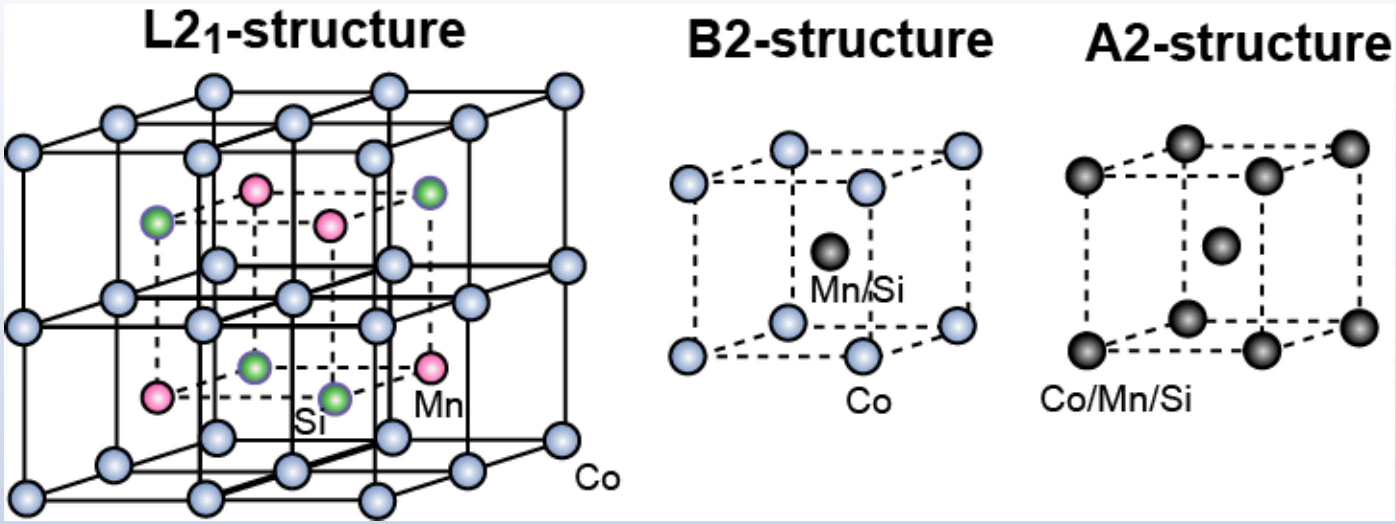
Half-metallic full-Heusler alloy Co_2MnSi

Co_2MnSi (CMS)

- High Curie temp.: T_C ~ 985 K
- Half-metallic energy gap E_G : 0.4 ~ 0.6 eV
- High degree of $L2_1$ chemical ordering is easily obtained

CMS is a promising candidate in the half-metal candidates.

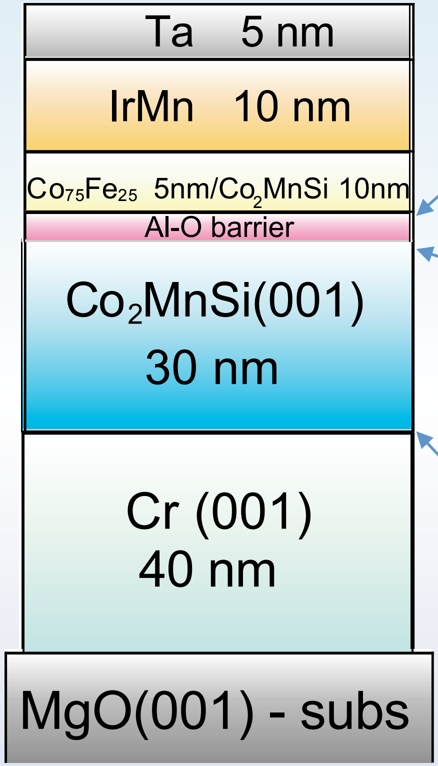




Experiment 1

Fabrication of CMS-based MTJs

UHV magnetron sputtering system (P < 1 x 10^{-7}Pa)



MTJ

Al-O barrier : Plasma oxidation of Al layer
Micro-fabrication : PL & Ar ion milling
(element size : 5 x 5 - 100 x 100μm^2)
Annealing : 200 -500°C with applying *H*
MR, I-V curve : DC 4-probe method

Co_2MnSi bottom epitaxial electrode

Composition :Target $Co_{2.00}Mn_{1.28}Si_{1.30}$ → Film : $Co_{2.00}Mn_{1.00}Si_{1.08}$
Deposited@RT → Post-annealed@T_a = 250 - 650°C
Crystal structure, the degree of ordering : XRD
Magnetization : SQUID,VSM
Surface image : AFM

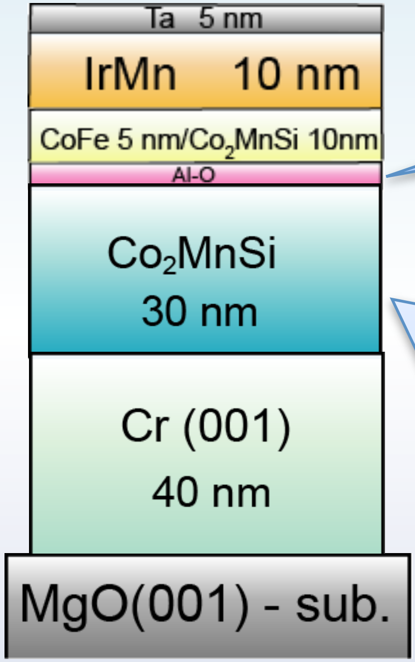
Cr buffer layer

Deposited@RT→Post-annealed@700°C

Epitaxial relation :
MgO(001)<100>/Cr(001)<110>/CMS(001)<110>
Lattice misfit < 5%

CMS-based MTJ

Magnetic tunnel junction (MTJs) with epitaxial CMS bottom electrode



Conventional Al-O amorphous barrier

Flat surface

Ra < 0.2 nm

Stoichiometry

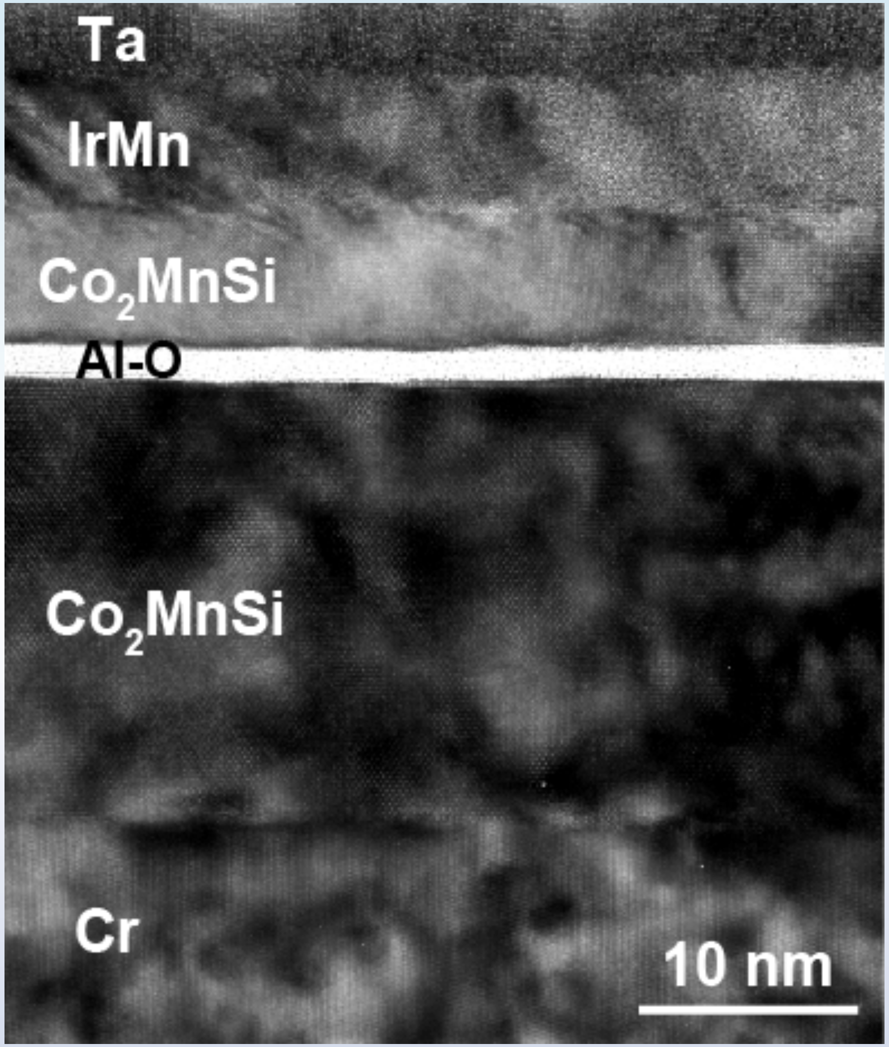
$Co_{2.00}Mn_{1.00}Si_{1.08}$ (ICP)

Highly $L2_1$-ordering

$S_{L21} > 0.8$ (XRD)

High-quality epitaxial CMS electrode

HR-TEM image

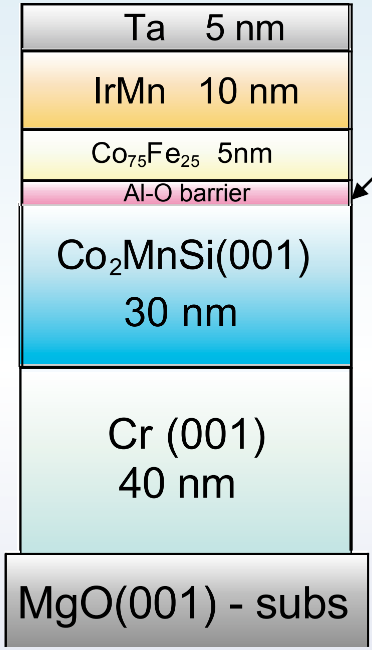


Extremely flat and sharp interfacial morphology

Experiment 1

CMS-based MTJs

Co_2MnSi : Mn has large affinity to oxygen compared to conventional 3d FM.



Magnetic impurities easily create between CMS/Al-O interface.

Optimization of interfacial chemical conditions is necessary.

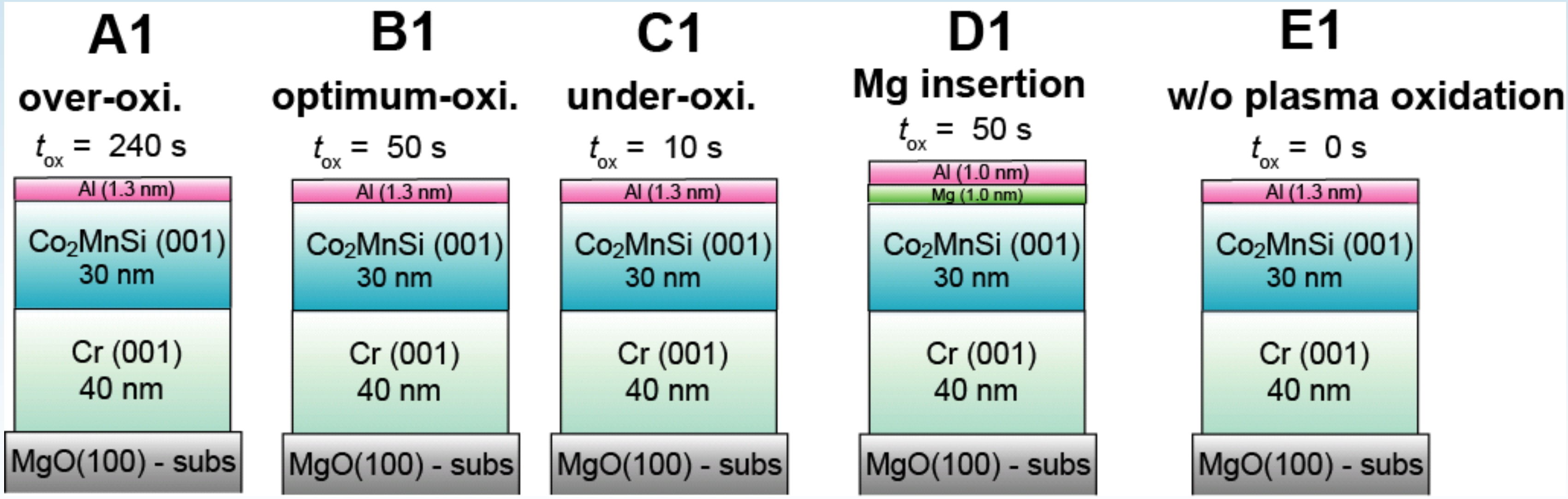
1. Plasma oxidation time for Al-O : t_{ox} = 10 - 240 s
2. Insert thin Mg layer between CMS and Al-O.
   (Formation energy of oxide : Mg < Al, Co, Mn, Si)

   Thickenss of Mg layer : t_{Mg} = 0 -1.3 nm

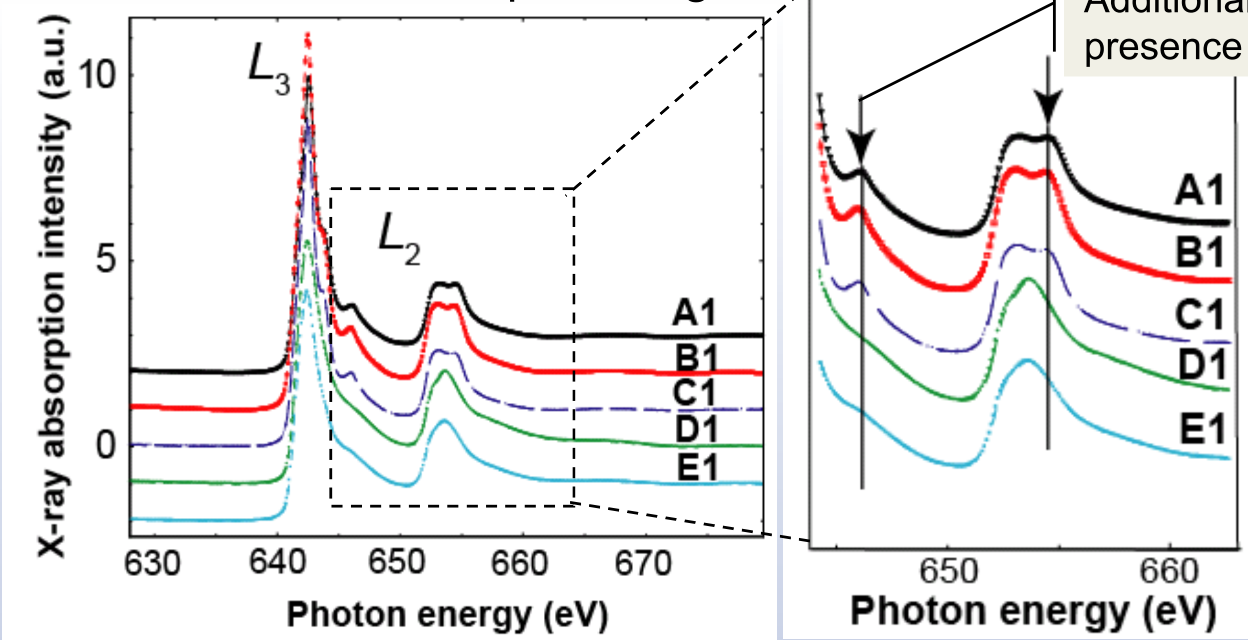
Experiment 1

CMS-based MTJs

Investigation of interfacial Mn impurities by XAS using TEY (ALS beamline 6.3.1)



XAS :Mn L-absorption edges



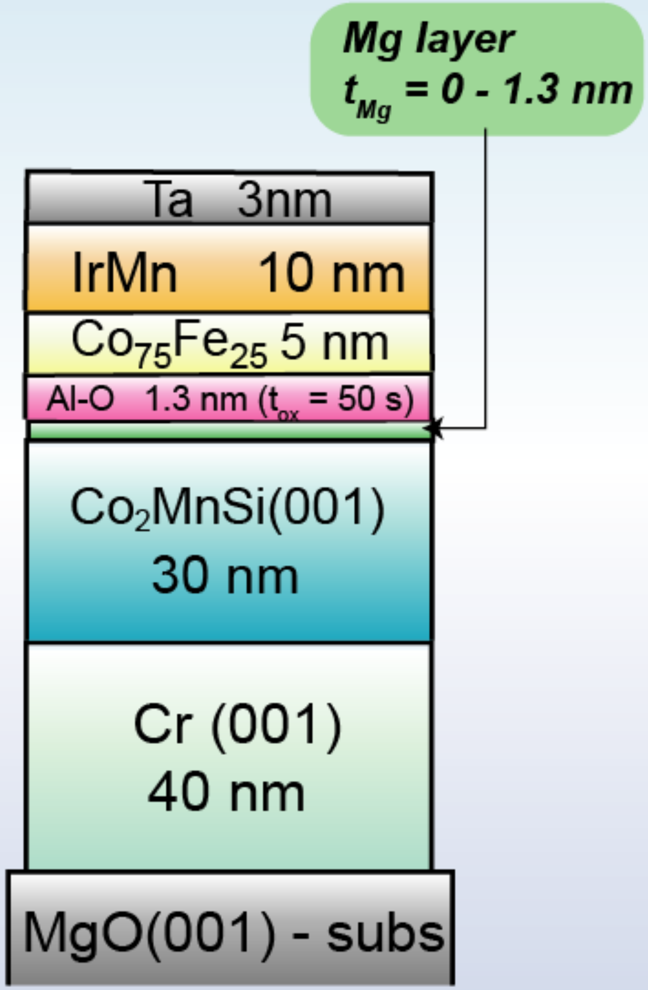
Additional multiplet structure indicates the presence of Mn-O impurities at interface.

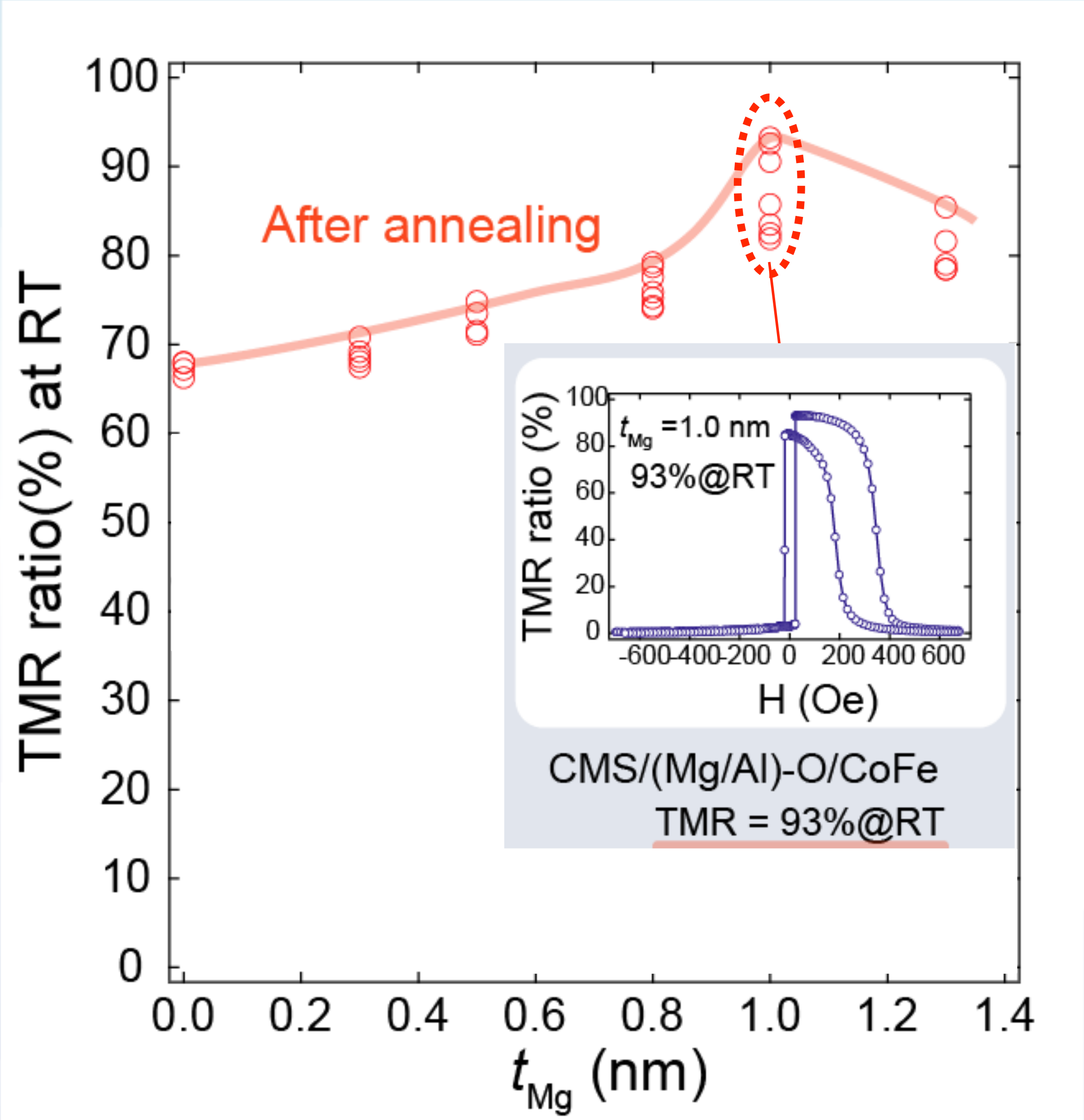
A1, B1, C1:
large amound of Mn-O at interface

D1, E1:
few Mn-O at interface

CMS-based MTJs

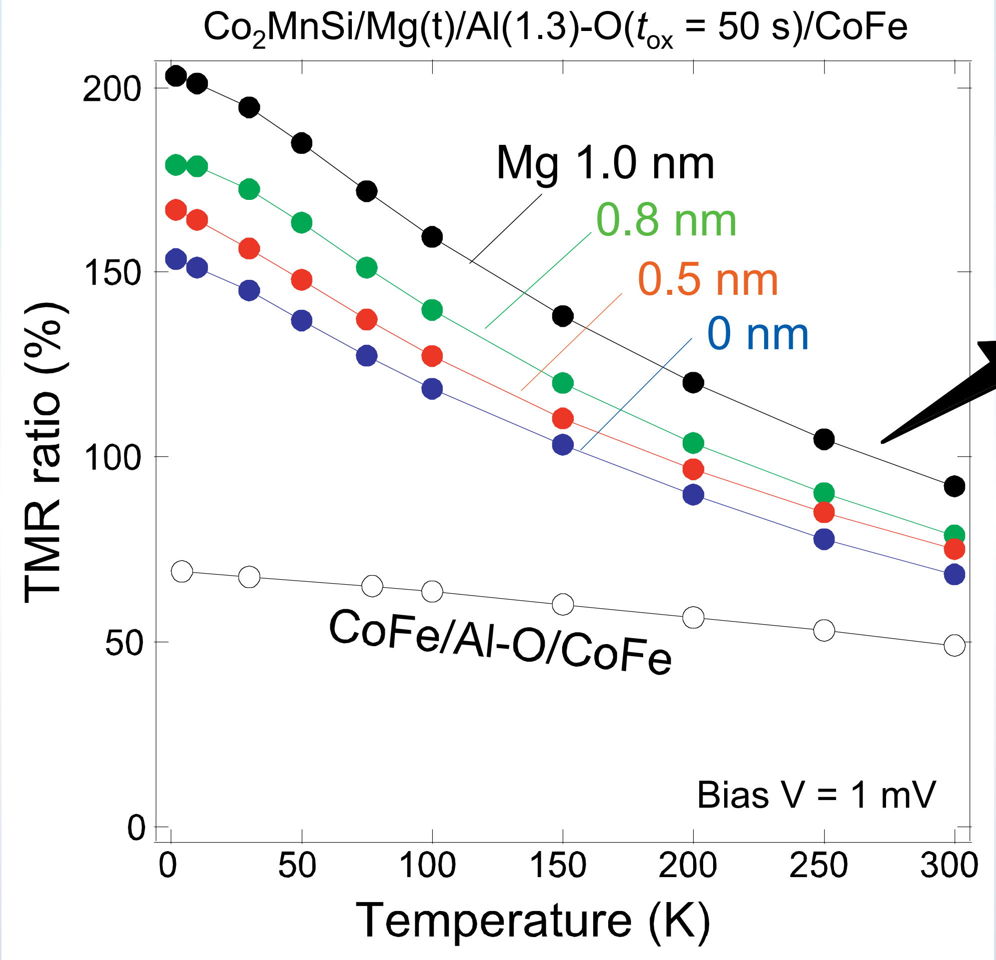
Mg thickness dependence of TMR ratio

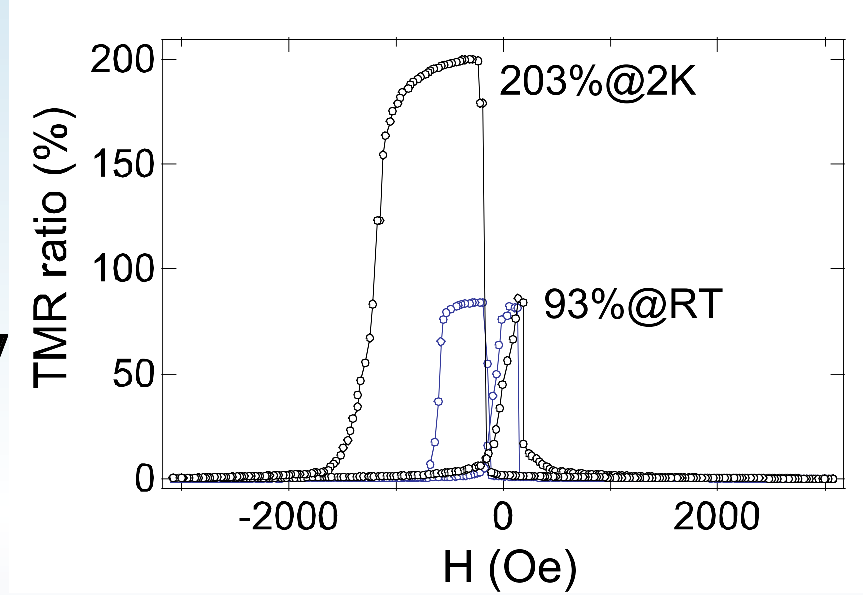




CMS-based MTJs

Temperature dependence of TMR ratio





CMS/Mg/Al-O/CoFe
TMR = 203% (T = 2K)

$0.50 < P_{CoFe} < 0.52$

$0.97 < P_{Co2MnSi} < 1.00$

Nearly ideal half-metallic spin-polarization

Sakuraba et al. J. Mag. Soc. Jpn. (2007)

XAS using TEY is an useful technique to investigate barrier interface quality in MTJs

CMS-based MTJs

TOHOKU

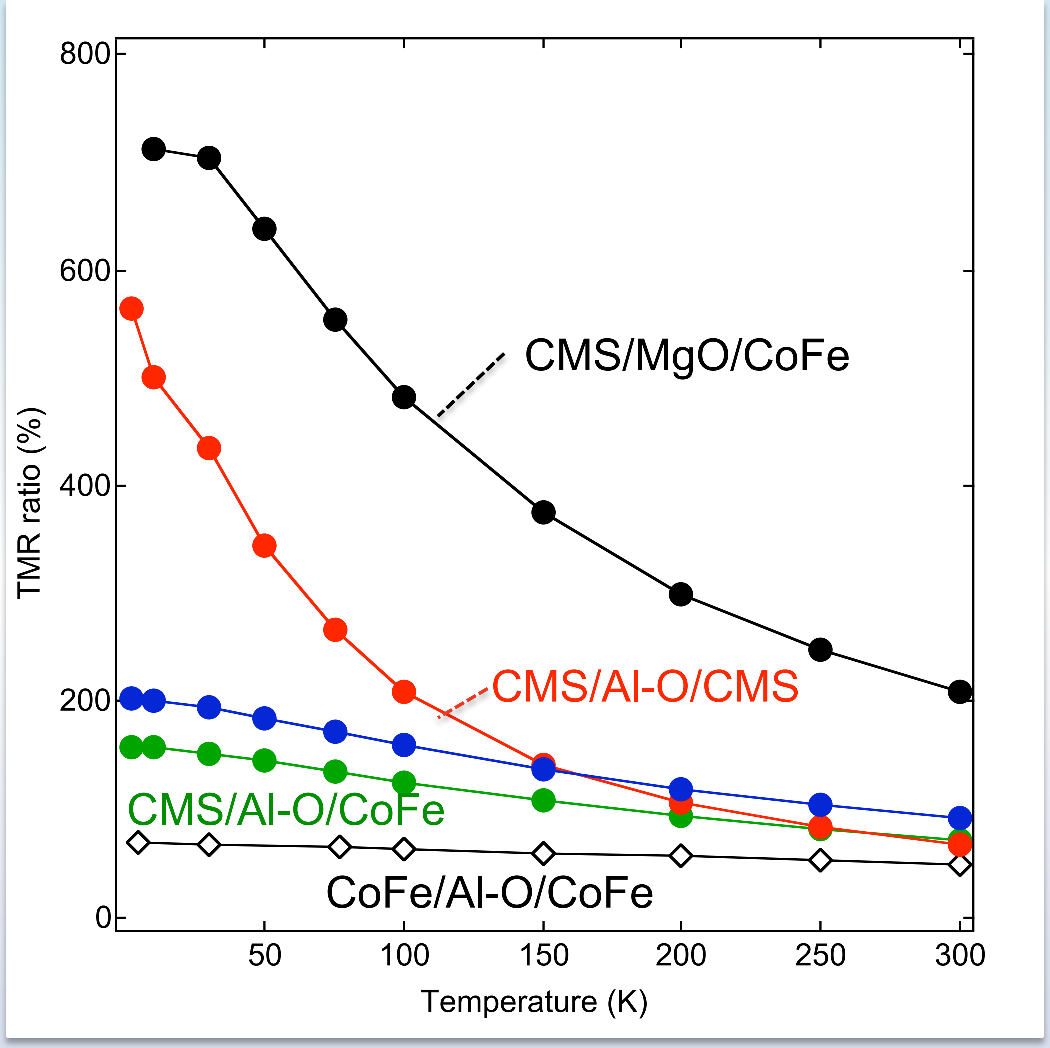
Co_2MnSi based-MTJs

$Co_2MnSi/Al\text{-}O/Co_2MnSi$-MTJ
TMR = 67%(RT), 570%(2K)
Sakuraba *et al. APL 88, 192508 (2006)*

$Co_2MnSi/MgO/CoFe$-MTJ
TMR = 217%(RT), 753%(10K)
Tsunegi *et al. APL 93, 112506 (2008)*

$Co_2MnSi/MgO/Co_2MnSi$-MTJ
TMR = 182%(RT), 705%(4.2K)
Ishikawai *et al. JAP (2009)*

cf. CoFeB/MgO/CoFeB-MTJ
TMR = 604%(RT), 1144%(4.2K)



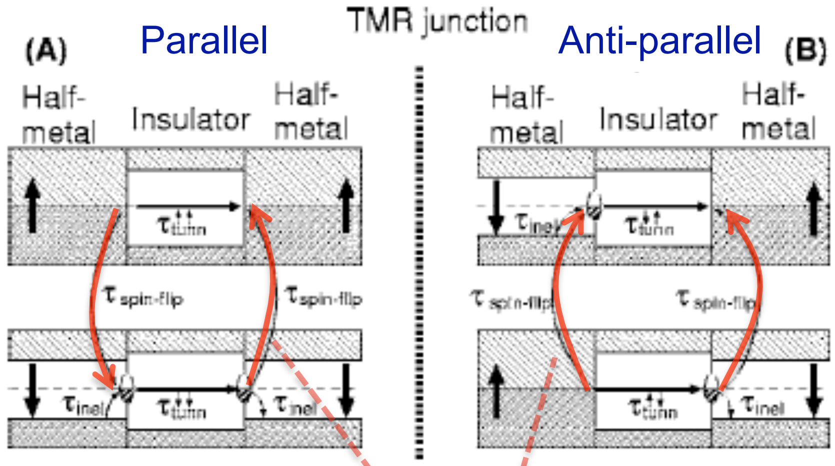
Disappearance of half-metallicity at RT is the largest problem in CMS-based MTJs.

Origin for disappearance of half-metallcity at RT

PHYSICAL REVIEW B 72, 174428 (2005)

Half-metallic ferromagnets for magnetic tunnel junctions by *ab initio* calculations

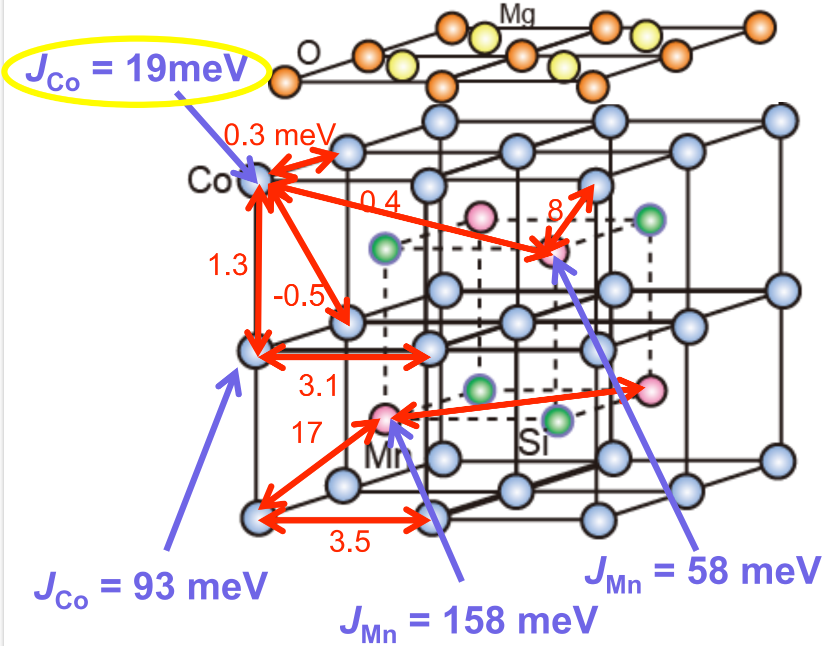
Phivos Mavropoulos,* Marjana Ležaić, and Stefan Blügel
Institut für Festkörperforschung, Forschungszentrum Jülich, D-52425 Jülich, Germany
(Received 23 August 2005; published 29 November 2005)



Spin-flip at interface due to magnon excitation.

Sakuma et al. J. Appl. Phys. 105, 07C910 (2009)

Co_2MnSi/MgO



Weak exchange energy of Co at the interface.

Large magnon excitation by thermal energy is the most possible origin for disappearance of half-metallicity at RT in CMS-based MTJs.

Experiment 2 MTJs using $L1_0/L2_1$-ordered alloy hybrid electrodes

For developing Spin-RAM

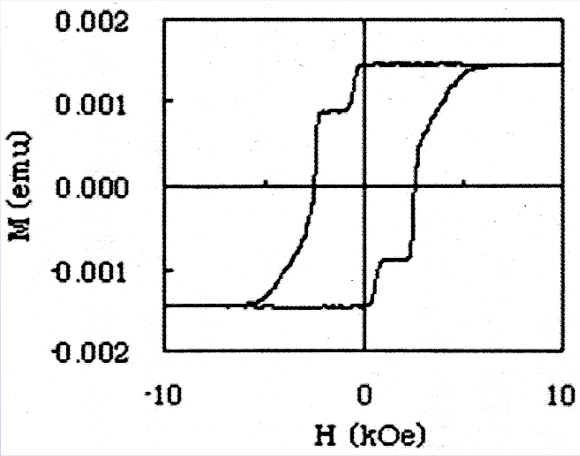
MTJs with perpendicular magnetized electrodes are recent trend.

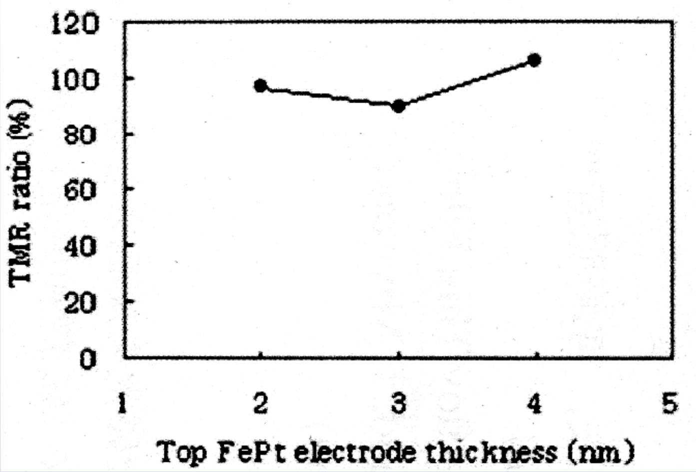
- High thermal stability : $K_u V/k_B T > 60$
- Small critical current density of spin-transfer torque switching : $J_{C0} < 10^6$ A/cm^2

FePt/Fe/MgO/FePt – MTJ

(MMM 2008, TOSHIBA)

→TMR ratio > 100% at R. T.

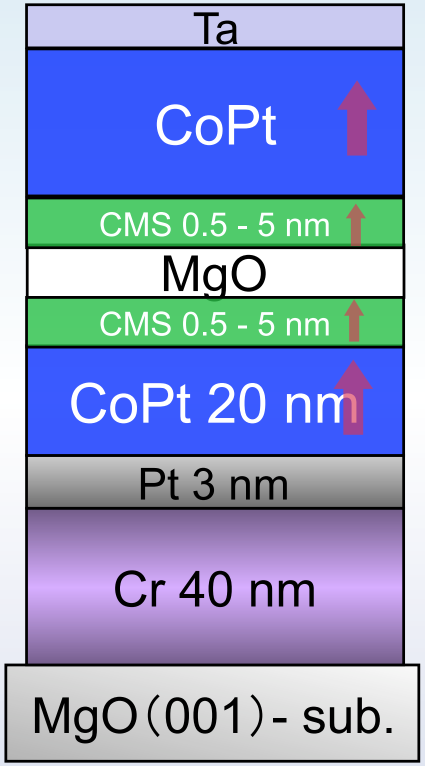




Experiment 2 MTJs using $L1_0$/$L2_1$-ordered alloy hybrid electrodes

Lattice misfit between CMS(001) and CoPt(001) ~ 4%

(001)-CoPt/CMS/MgO/CMS/CoPt fully-epitaxial MTJ can be fabricated.



★ High thermal stablity due to large magnetic anisotropy

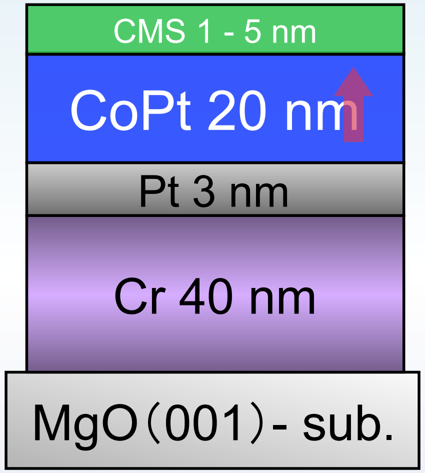
Suppression of magnon excitation at CMS/barrier interface.

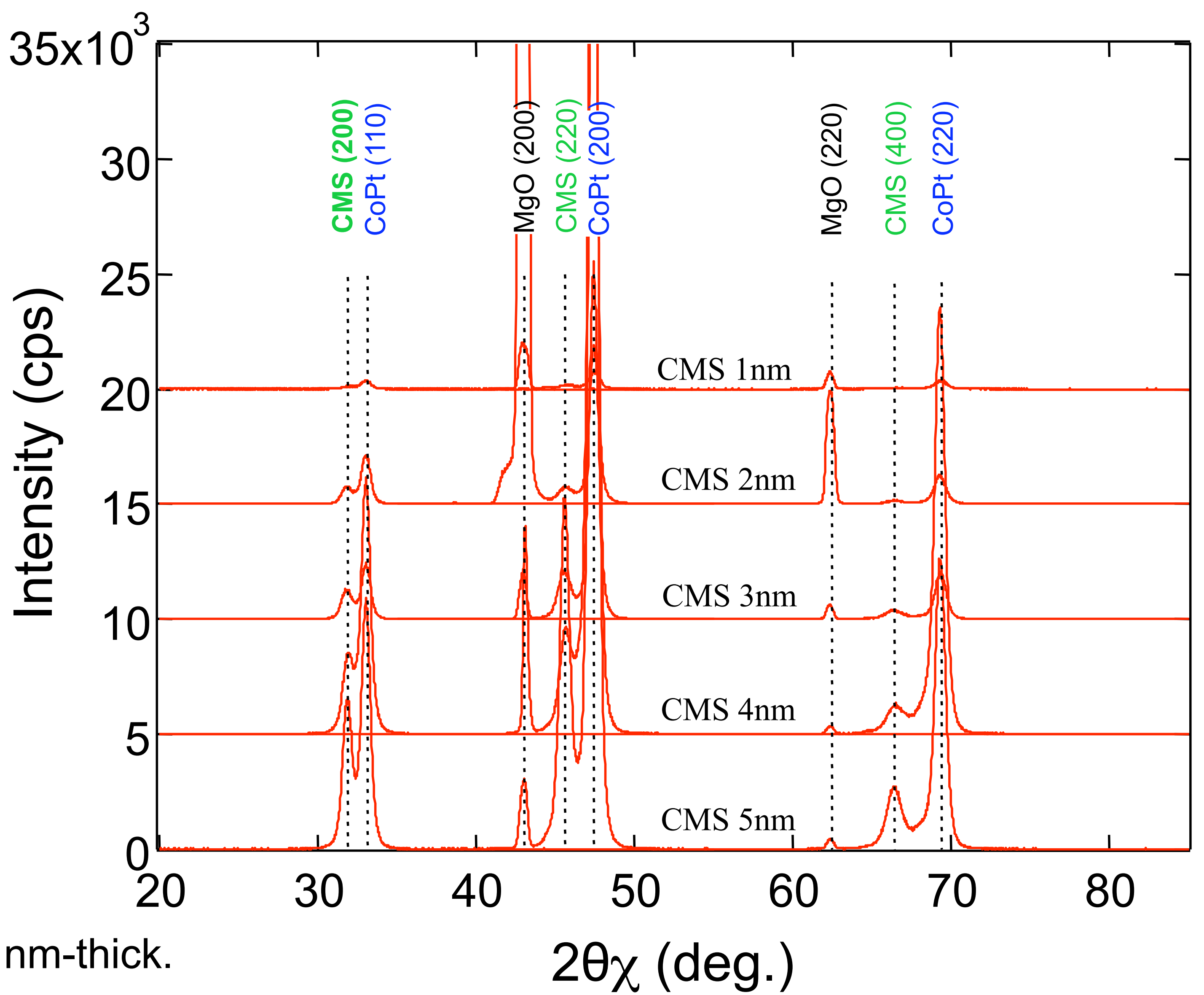
★ Large spin-polarization (TMR ratio) at RT

Experiment 2 MTJs using $L1_0$/$L2_1$-ordered alloy hybrid electrodes

Structural analysis : XRD in-plane geometry (X-ray incident angle 2θ < 0.4deg.)

CMS: Deposited@RT
→annealed@450℃

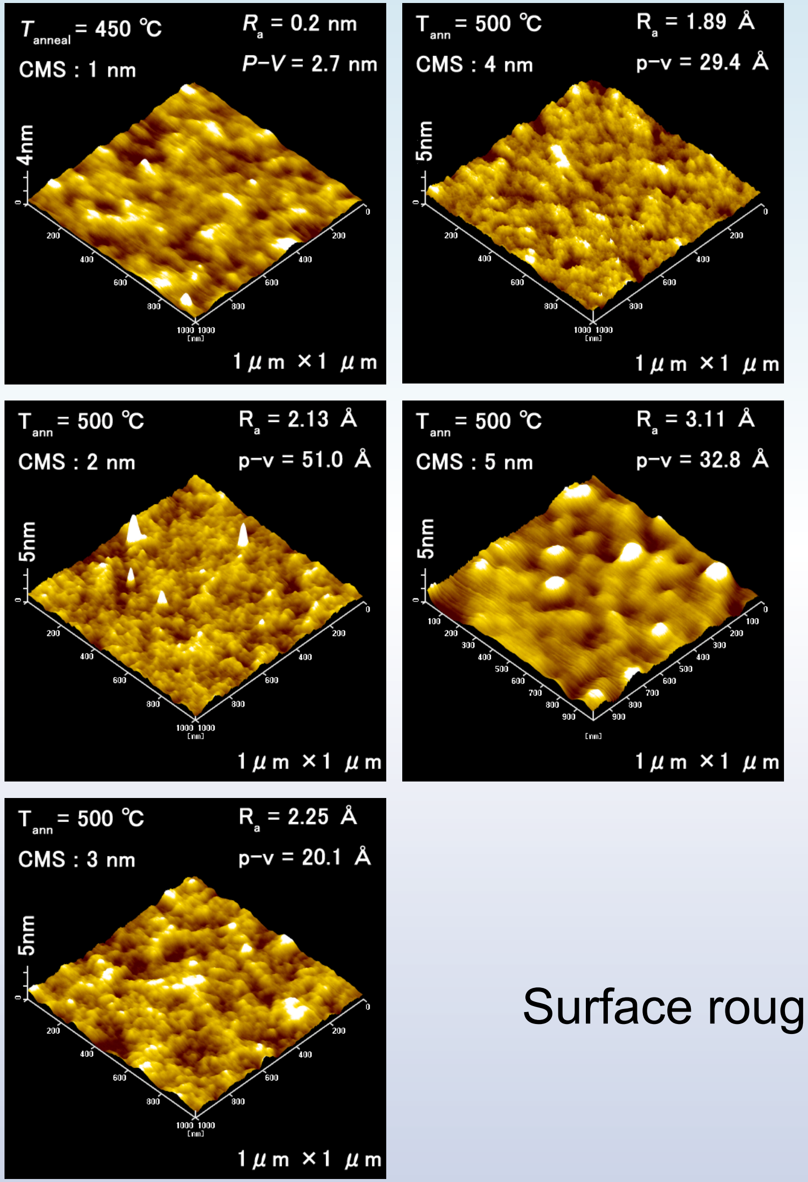


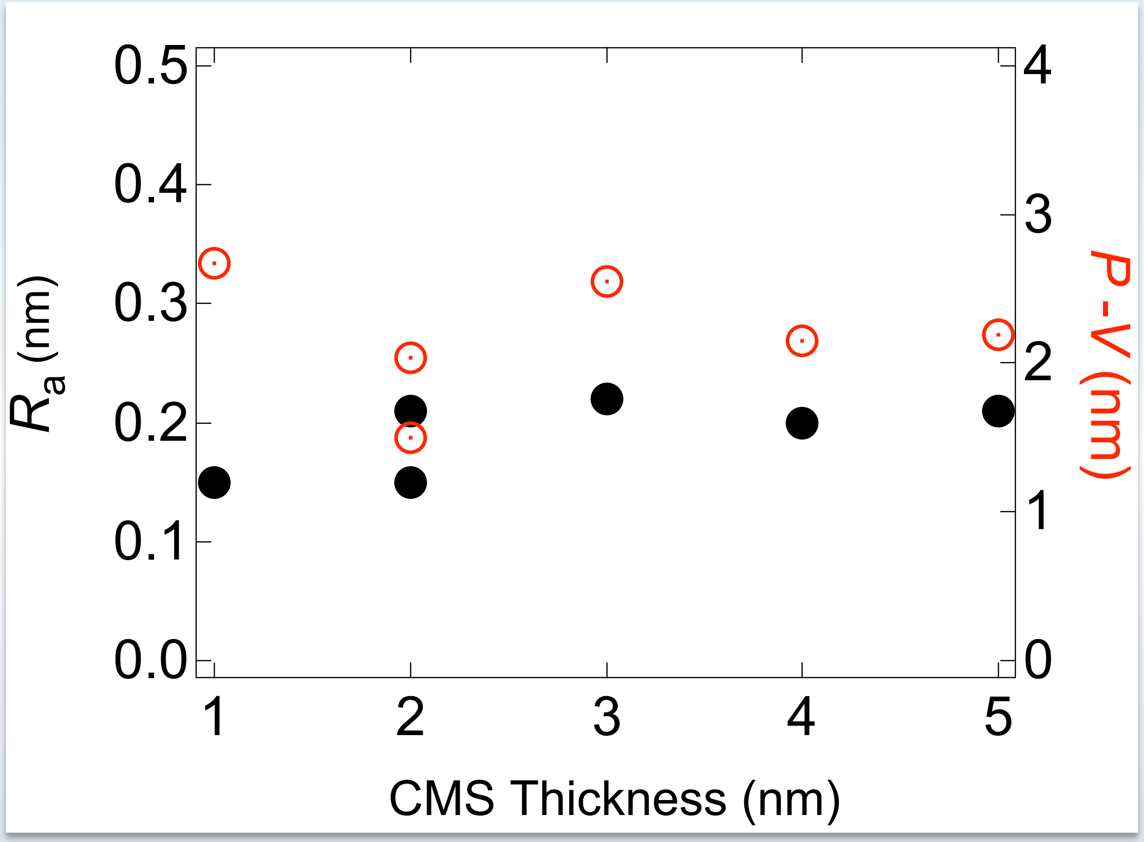


- fully-epitaxial growth
- B2-ordering even in 1nm-thick.

Experiment 2 MTJs using $L1_0/L2_1$-ordered alloy hybrid electrodes

Surface flatness : AFM

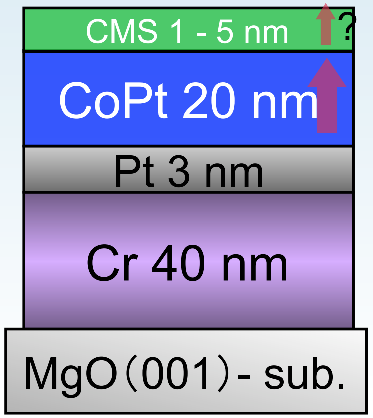


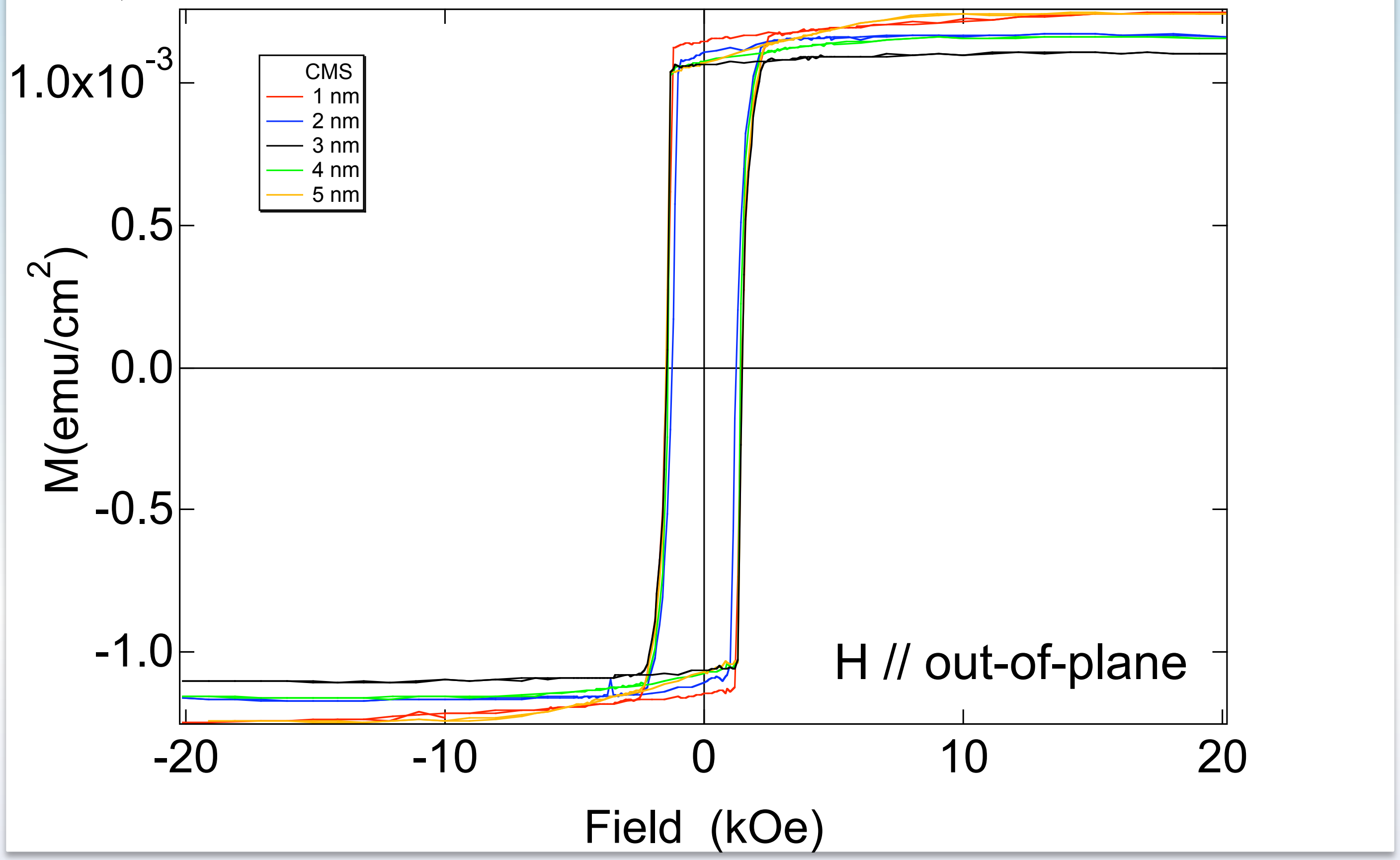


Surface roughness is enough small to deposit MgO barrier.

Experiment 2 MTJs using $L1_0/L2_1$-ordered alloy hybrid electrodes

M-H curves : SQUID, VSM





Magnetic behavior of CMS layer can not be identified because of…

(1) too small M_{CMS} compared with M_{CoPt}

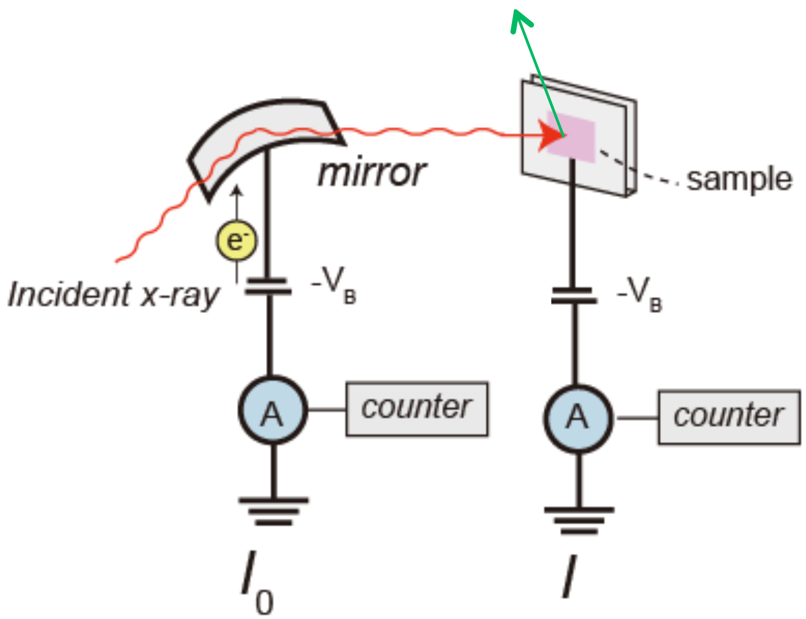
(2) the error in subtraction procedure of MgO-subs contribution

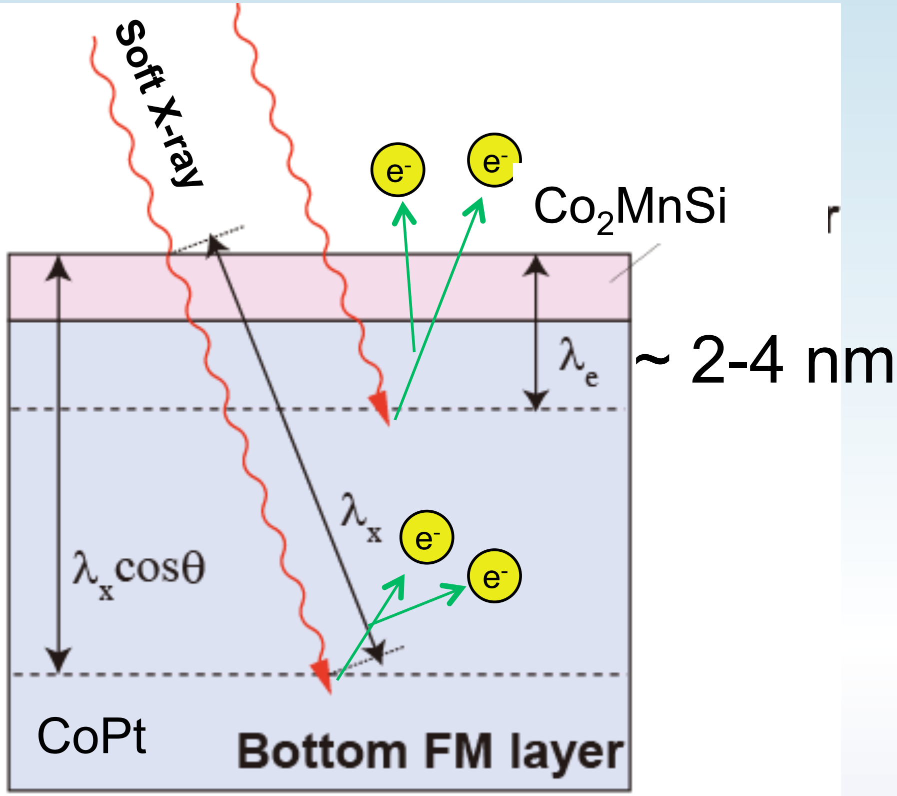
Other experimental techniques to observe surface moment is needed.

XMCD

Experiment 2 MTJs using $L1_0/L2_1$-ordered alloy hybrid electrodes

Total electron yield (TEY) method





XMCD : SPring-8 BL25SU

λ_x : penetration length of X-ray

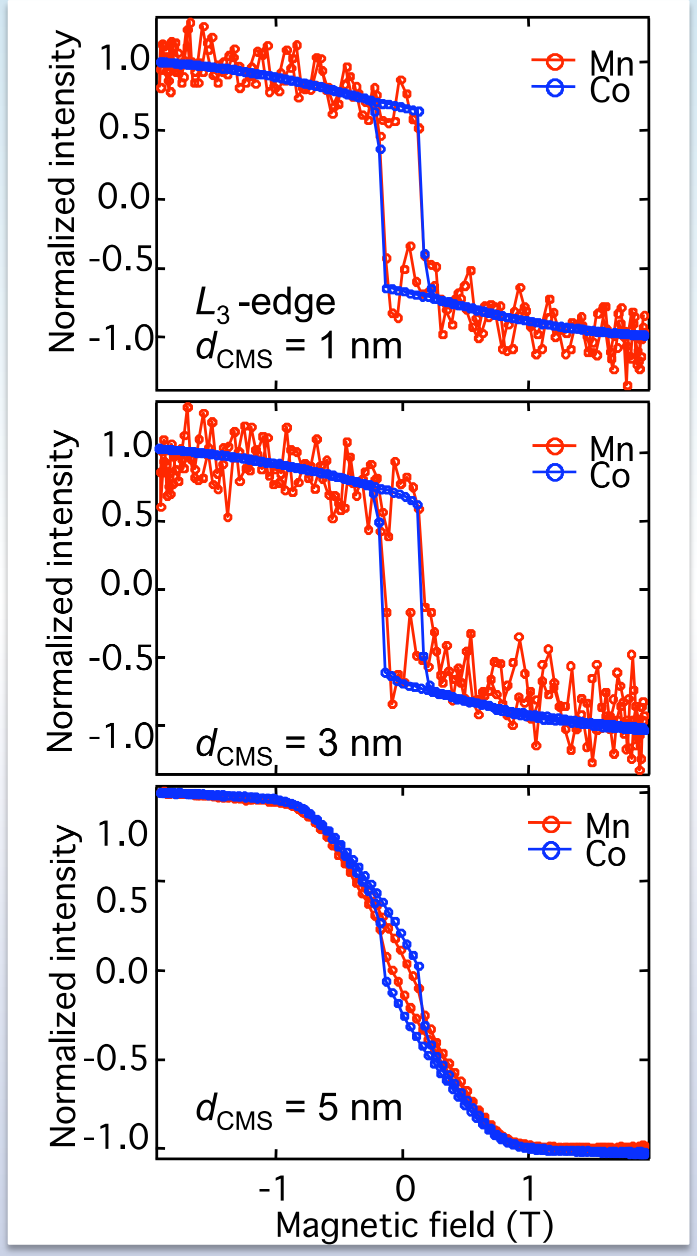
$\lambda_x \cos\theta$: penetration depth of X-ray

λ_e : escape depth of photo electron

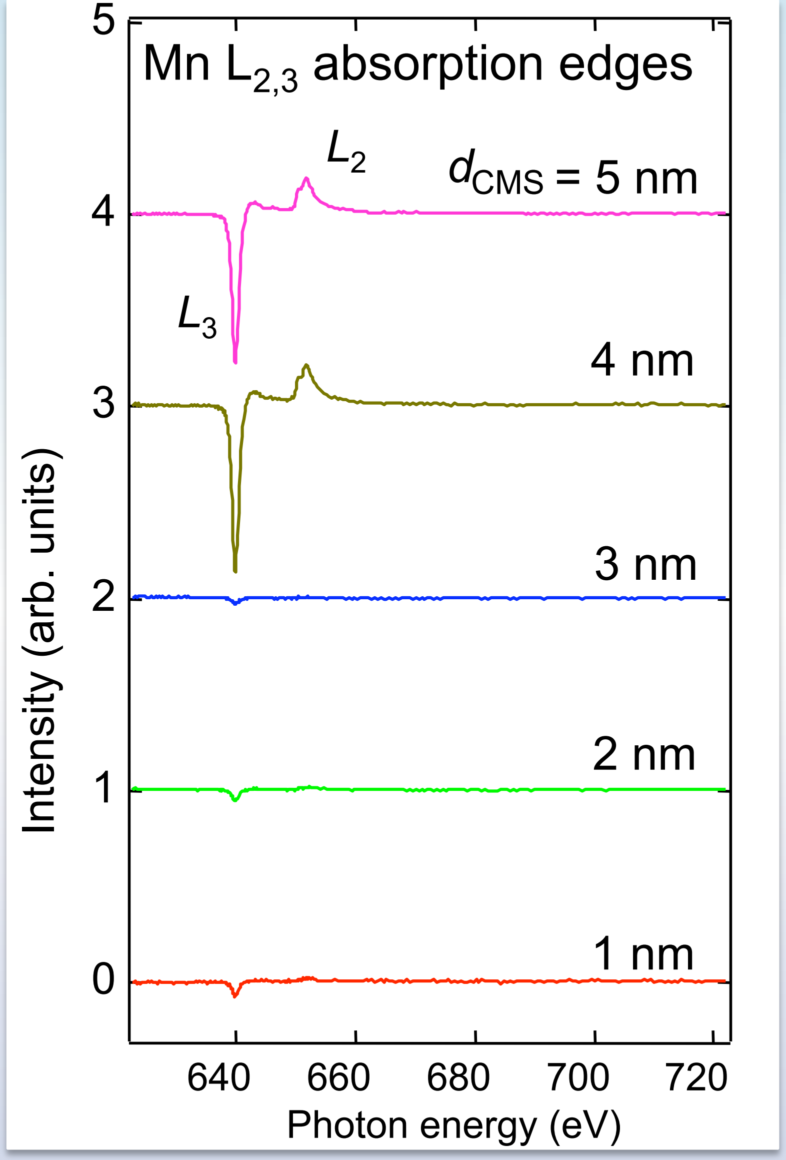
Elements specific (Co and Mn) M-H curves can be measured.

Experiment 2 MTJs using $L1_0/L2_1$-ordered alloy hybrid electrodes

Element specific M-H (H//out-of-plane)



XMCD spectra (H//out-of-plane)



- d_{CMS} < 3nm : CMS has perpendicular magnetic anisotropy, but magnetic moment is very small.

CMS-based CPP-GMR devices

TOHOKU

What is the next generation reading head?

Requirements : **Large MR ratio** for high S/N ratio, **Small RA** for high speed reading

- ***MgO-MTJs***

Large MR ratio, but difficult to achieve small RA.

CoFeB/MgO/CoFeB : RA = 0.4 Ω•μm², MR = 50%
Nagamine et al. APL(2006)

RA = 2.1 Ω•μm², MR = 206%
Isogami et al. MSJ conf.(2008)

- ***CPP-GMR***

Small RA, but difficult to achieve large MR ratio.

CoFe/Cu/CoFe : RA = 0.12 Ω•μm², MR = 1.3%
Yuasa et al. JAP(2002)

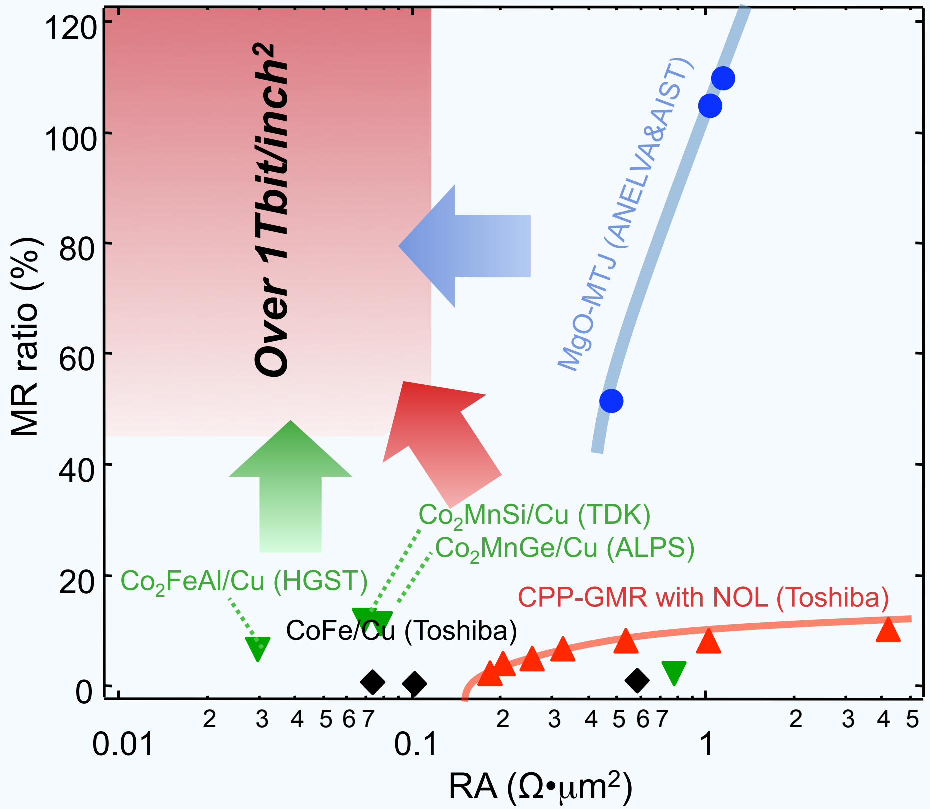
(i)Using Nano-oxide layer(NOL)

CoFe/NOL/CoFe : RA = 0.58 Ω•μm², MR = 8.2%
Fukuzawa et al. JPD(2007)

(ii)Using half-metallic full-Heusler alloy

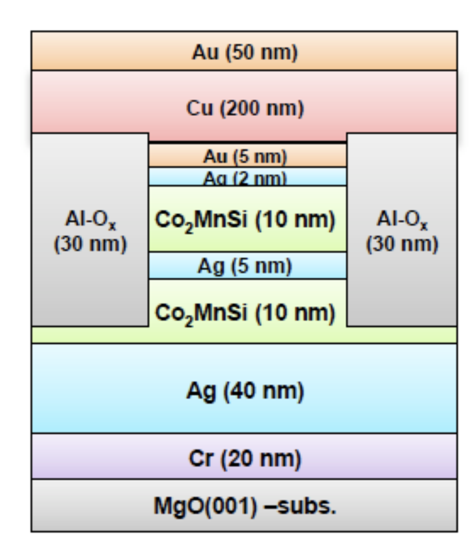
$Co_2MnGe/Cu/Co_2MnGe$: RA = 0.079 Ω•μm², MR = 11.5%
Saito et al. Intermag(2004)

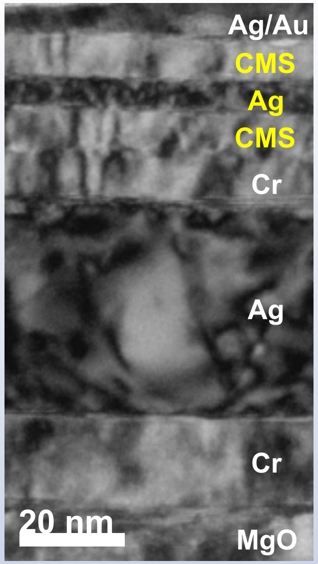
Reported MR ratio in small RA region

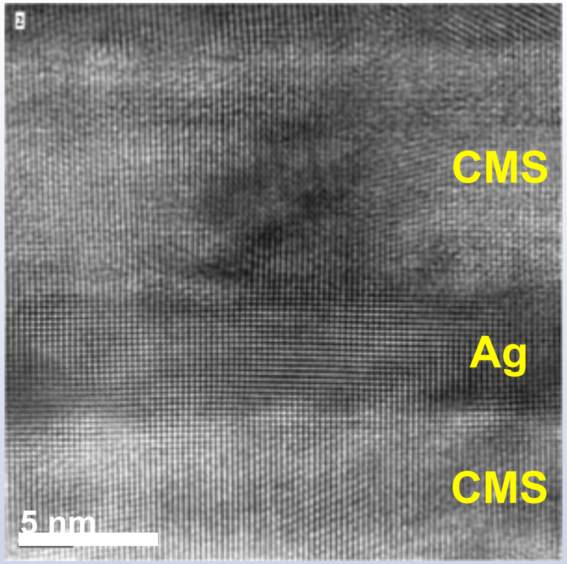


Experiment 4 CMS-based fully-epitaxial CPP-GMR devices

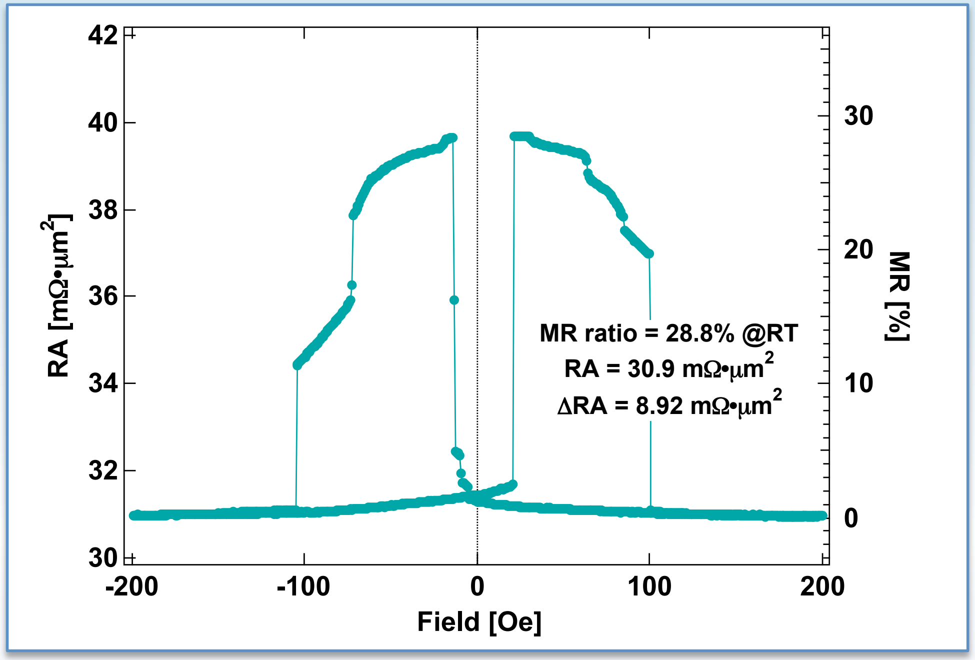
CMS/Ag/CMS fully-epitaxial CPP-GMR device







HR-TEM image



◆ The highest MR ratio (28.8%@RT) is the best record in all of the reports to date.

cf. CoFe/Cu/CoFe : MR ~ 2.3%@RT

Summary

● XAS based on TEY is an useful technique to observe interfacial chemical state between barrier and FM electrodes in MTJs.

● Element specific magnetization measurement based on XMCD is an helpful technique to fabricate new types of half-metals such as, perpendicular magnetized half-metals and ferri-magnetic half-metals.