

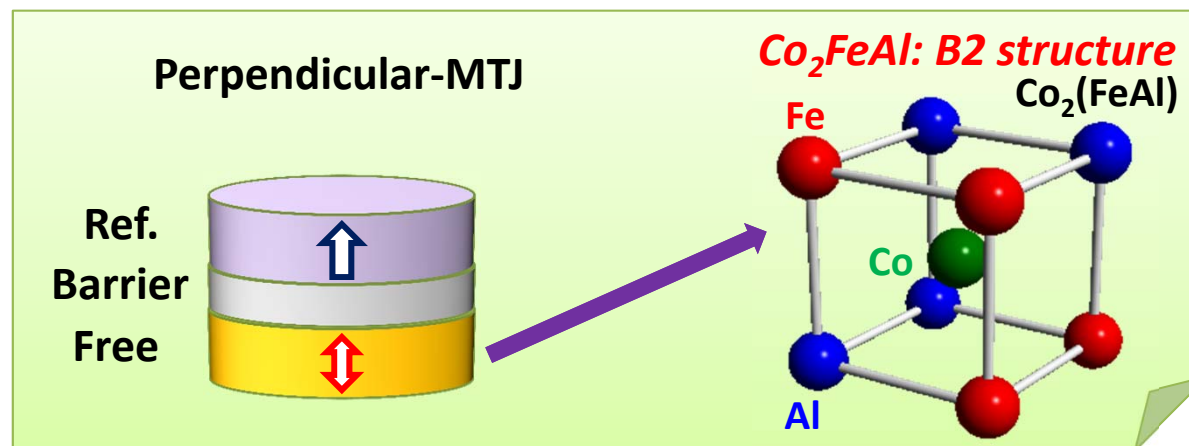
Friday, July 31, 2015

## “Heusler Alloys for Spintronic Devices”

organized by The Center for Spintronic Materials, Interfaces,  
and Novel Architectures (C-SPIN)



# Interface-induced perpendicular magnetic anisotropy and giant tunnel magnetoresistance in $\text{Co}_2\text{FeAl}$ Heusler alloy based heterostructures



Hiroaki Sukegawa

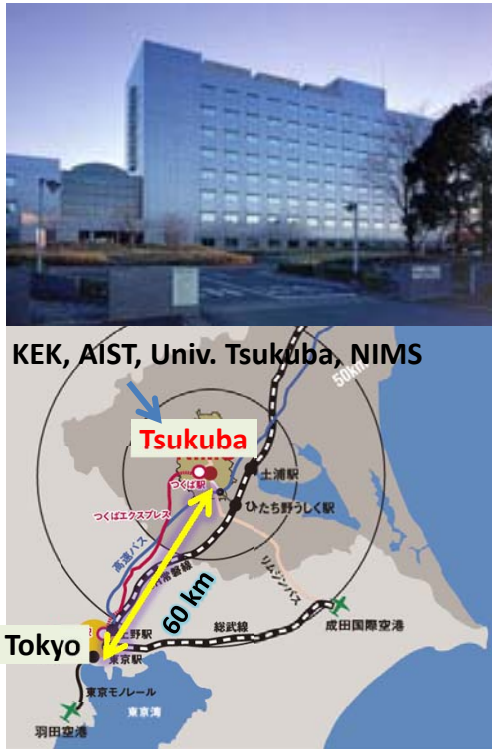
*National Institute for Materials Science (NIMS), Tsukuba, Japan*

In collaboration with Magnetic Materials Unit members in NIMS:

S. Mitani, K. Inomata, Z. C. Wen (~2015.4 now Tohoku Univ. Japan), T. Scheike, T. Furubayashi, J. P. Hadorn, T. Ohkubo and K. Hono

University of Minnesota, Minneapolis, Minnesota, USA, July 30-31, 2015

NIMS: National Institute for Materials Science, Tsukuba, Japan (permanent staffs: 410, total ~1,500)



**Magnetic Materials Unit**

















Director



Kazuhiro Hono

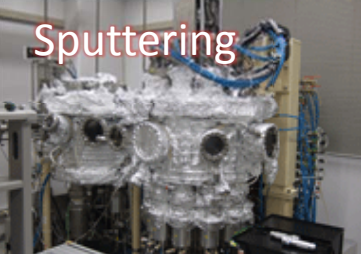
Magnetic Materials Group  
Spintronics Group  
Nanostructure Analysis Group

**Spintronics group members**

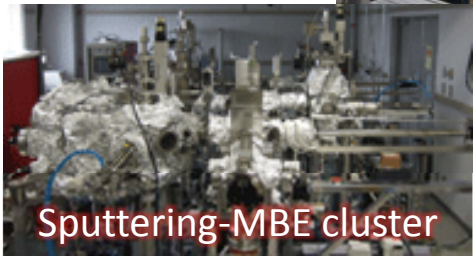
GL Leader	NIMS Staff				Emeritus Fellow
 <a href="#">S. Mitani</a>	 <a href="#">S. Kasai</a>	 <a href="#">M. Hayashi</a>	 <a href="#">H. Sukegawa</a>	 <a href="#">K. Inomata</a>	
Post-Doc		Graduate Student (Univ. of Tsukuba)			Visiting Researcher
 <a href="#">P. Sheng</a>	 <a href="#">M. Belmoubarik</a>	 <a href="#">S. Hirayama</a>	 <a href="#">T. Scheike</a>	 <a href="#">Q. Xiang</a>	 <a href="#">S. Ichikawa</a>
		<b>Admin.</b>			
Visiting Researcher	Internship	Technical Assist.			
 <a href="#">R. Pérez</a>	 <a href="#">Y. Kato</a>	 C. Choi	 <a href="#">H. Shimadu</a>	 <a href="#">A. Tomaru</a>	

**Equipment**

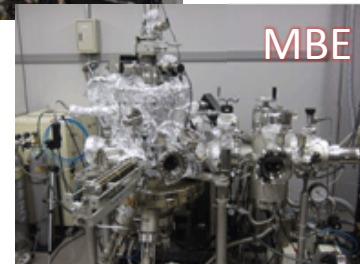
Thin-film deposition systems



Sputtering



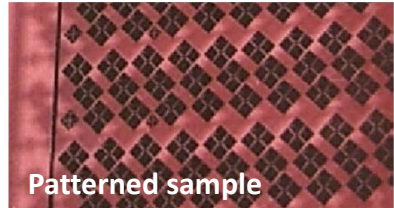
Sputtering-MBE cluster



MBE

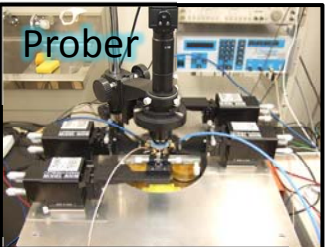
**Microfab. facility**

E-beam lithography

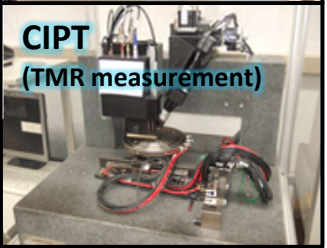


Patterned sample

**Electric measurement**



Prober



CIPT (TMR measurement)

# Outline

## 1. Background

Co<sub>2</sub>FeAl Heusler alloy based magnetic tunnel junctions (MTJs) for MRAM applications

## 2. Perpendicular anisotropy in Co<sub>2</sub>FeAl ultra-thin films

2.1 Perpendicular magnetic anisotropy (PMA) and TMR in ultrathin Co<sub>2</sub>FeAl/MgO structures

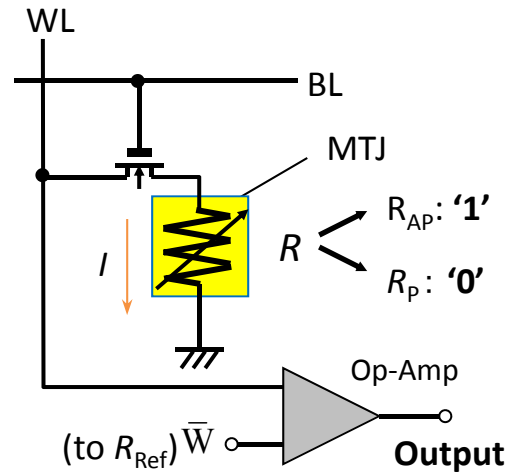
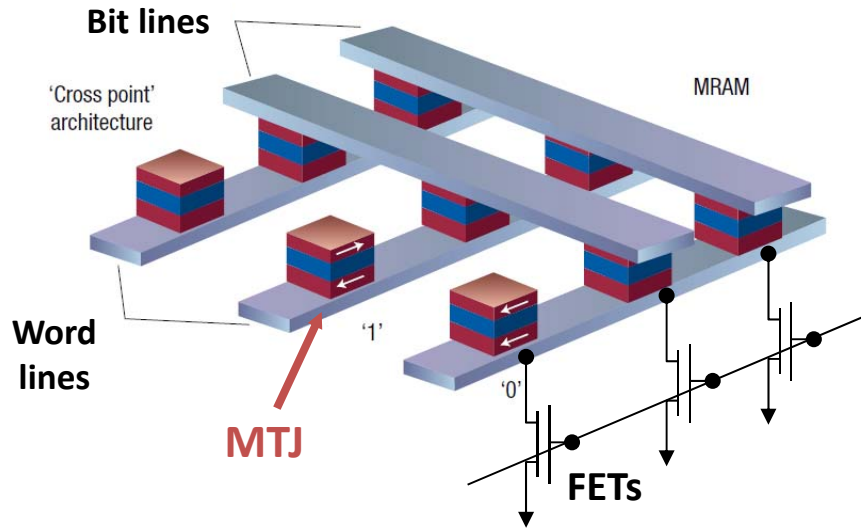
2.2 Enhanced PMA by Ru(0001) underlayer

2.3 Possible mechanism of PMA

## 3. Co<sub>2</sub>FeAl MTJs with a lattice-matched MgAl<sub>2</sub>O<sub>4</sub> coherent barrier

# MRAM (magnetoresistive random access memory)

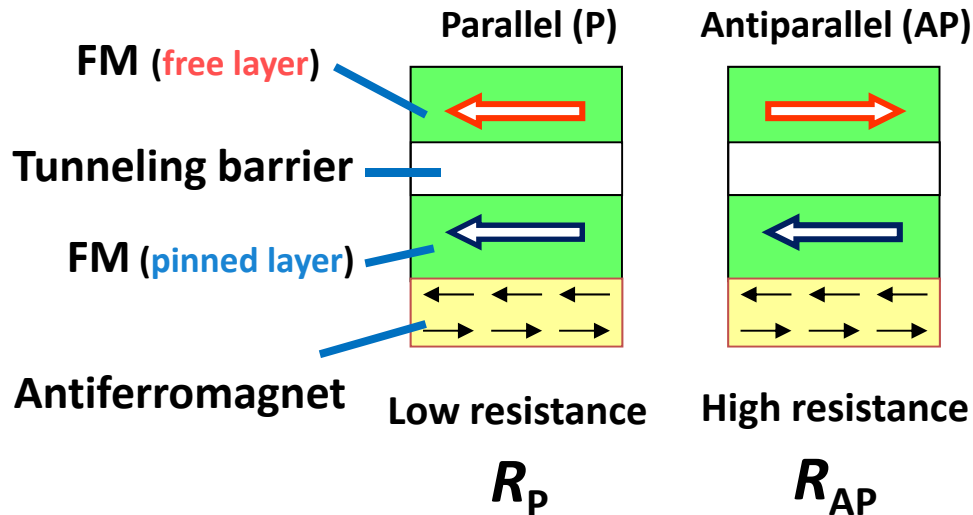
C. Chappert, A. Fert and F. N. V. Dau, Nat. Mater. 6, 813 (2007)



- Nonvolatile
- Infinite endurance
- High speed
- High density

## MTJ (magnetic tunnel junction)

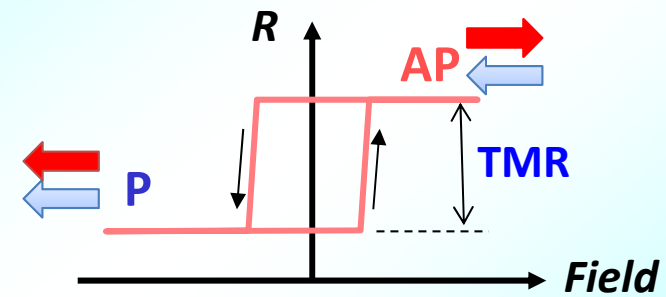
(in-plane mag. type)



FM: Ferromagnet

## TMR (tunnel magnetoresistance)

$$\text{TMR ratio} = \frac{R_{AP} - R_P}{R_P}$$



High TMR  $\Rightarrow$  High output (high speed reading)

# Properties needed for MRAM application

## (1) High signal output (TMR) with tunable device resistance

- ✓ High spin polarization materials
  - Co-based Heusler alloys
- ✓ New (coherent) tunneling barrier materials
  - $\text{MgAl}_2\text{O}_4$  based barrier

## (2) High thermal stability of stored information

- ✓ High perpendicular magnetic anisotropy (PMA)
  - Bulk-induced anisotropy
    - Multilayers,  $L1_0/\text{D0}_{22}$  based alloys...
  - Interface-induced anisotropy
    - ultrathin FM/insulator interface

## (3) Low writing energy (low current density for STT writing)

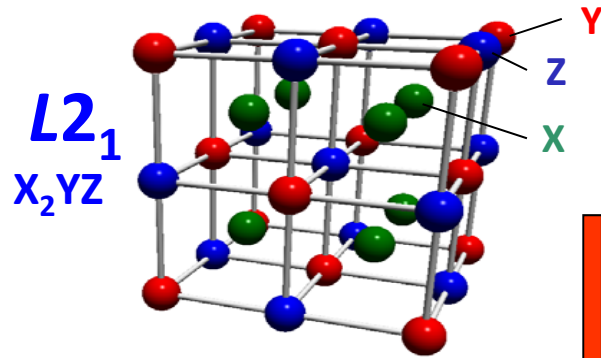
- ✓ Low magnetic damping materials
  - Co-based Heusler alloys
  - Mn-based PMA alloys

## (4) New writing technology

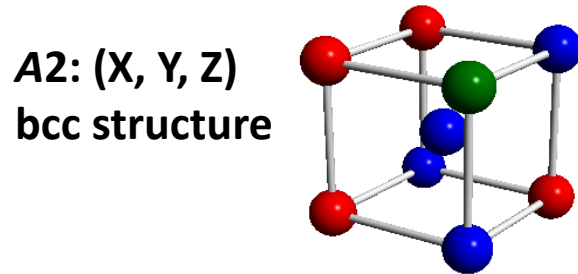
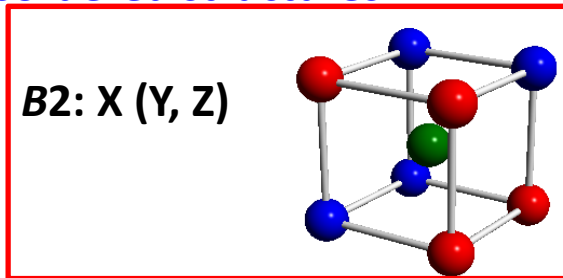
- ✓ Magnetic anisotropy control by electric field
- ✓ Spin-orbit torque
  - Interface PMA system



# Half-metallic full Heusler alloys



Disordered structures



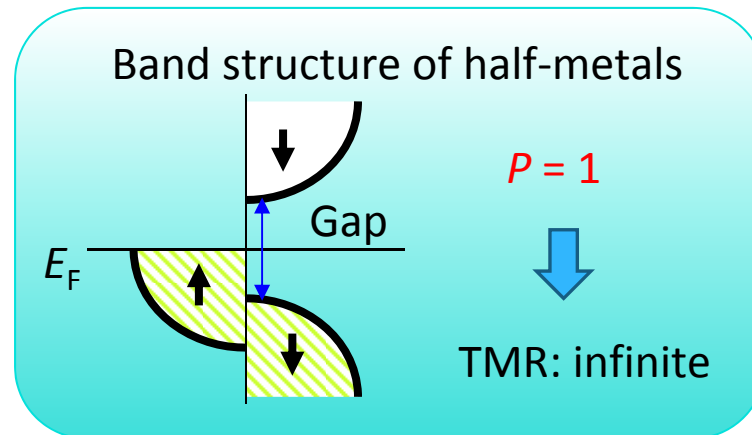
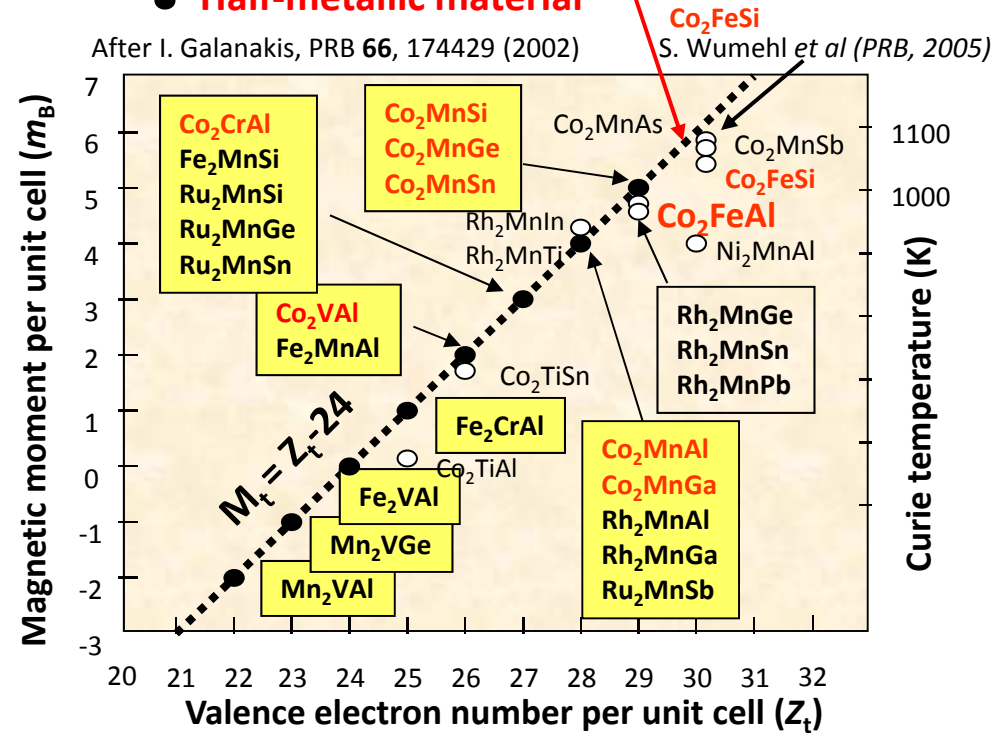
Lower  $P$

## Co-based Heusler alloy: $Co_2YZ$

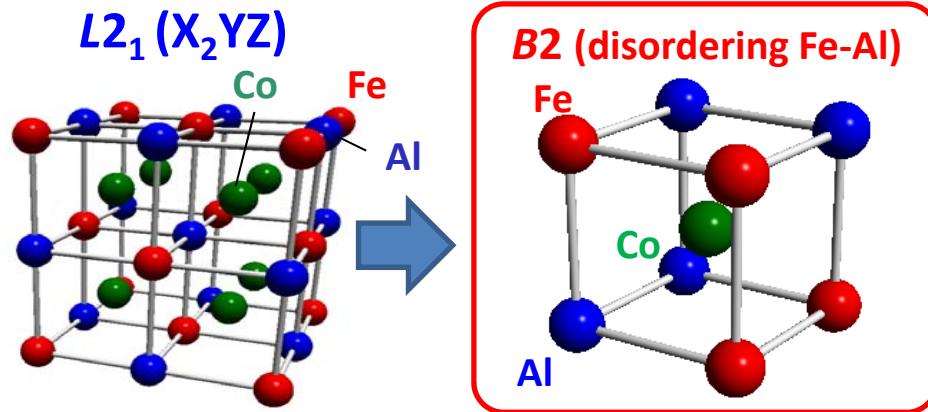
- High Curie temperatures
- High spin polarization
- Low magnetic damping  
even in  $B2$  structure

Slater-Pauling rule  $M_t = Z_v - 24$

• Half-metallic material

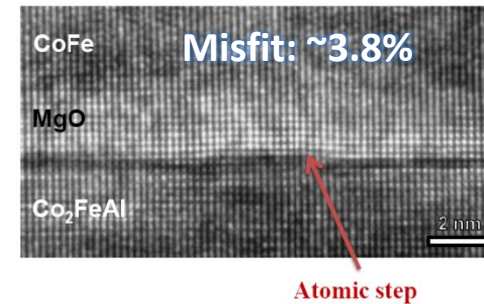


# Co<sub>2</sub>FeAl (CFA) Heusler alloy for spintronics



B2-ordered CFA is generally obtained by sputtering deposition.

## 1. Small lattice misfit of CFA/MgO(001)



High-quality CFA/MgO-MTJs can be fabricated using a **sputtering method**.

## 2. Large TMR in CFA/MgO MTJs

Epitaxial **CFA/MgO/CoFe(0.5 nm)/CFA** (in-plane mag.)

**TMR: 785% (10 K), 360% (RT)**

W. H. Wang, HS *et al.*, PRB82, 092402 (2010).

- High spin polarization and coherent tunneling
- Less temperature dependence of TMR ratio

## 3. Possible low damping constant $\alpha$

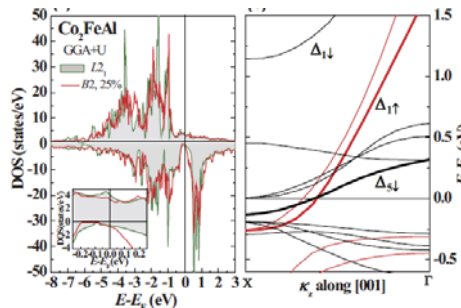
$\alpha \sim 0.001$  for 50-nm-thick CFA(001)

S. Mizukami *et al.*, JAP105, 07D306 (2009).  
(Tohoku group)

Effective for spin-transfer torque (STT) switching

### Band calculation

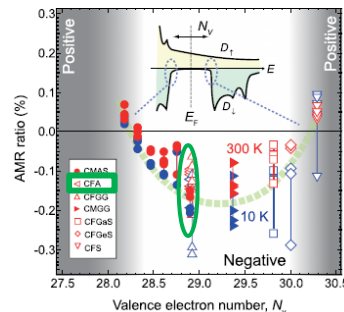
W. H. Wang *et al.*, PRB81, 140402(R) (2010).



Not a perfect half-metal, but fully spin-polarized  $\Delta_1$  band

### AMR

Y. Sakuraba *et al.*, APL104, 172407 (2014).



Negative AMR in B2-CFA film: a nearly half-metallic band structure

# Outline

1. Background  
Co<sub>2</sub>FeAl Heusler alloy based magnetic tunnel junctions (MTJs) for MRAM applications
2. Perpendicular anisotropy in Co<sub>2</sub>FeAl ultra-thin films
  - 2.1 Perpendicular magnetic anisotropy (PMA) and TMR in ultrathin Co<sub>2</sub>FeAl/MgO structures
  - 2.2 Enhanced PMA by Ru(0001) underlayer
  - 2.3 Possible PMA mechanism
3. Co<sub>2</sub>FeAl MTJs with a lattice-matched MgAl<sub>2</sub>O<sub>4</sub> coherent barrier





# Perpendicular MTJs

## Perpendicular magnetic anisotropy (PMA)

PMA: higher thermal stability, lower STT switching current density

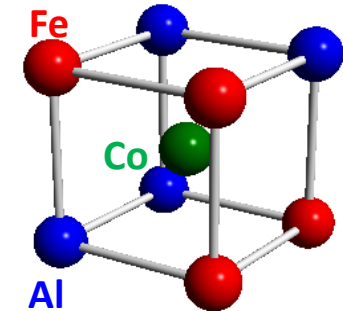
### Bulk-induced PMA: b-PMA

Bulk PMA materials, Multilayers

$L1_0$ -FePt,  $D0_{22}$ -MnGa, amorphous-TbFeCo,  $(Co/Pt)_n$ ,  $(Co/Pd)_n$ ...

Small uniaxial magnetocrystalline anisotropy can be expected in bulk of **cubic** Heusler alloys.

→ Use of Interface effect

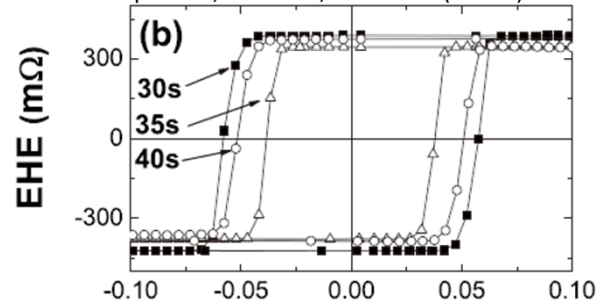


### Interface-induced PMA: i-PMA

Interface at ultrathin layer and oxide

Pt/Co/AlOx trilayers

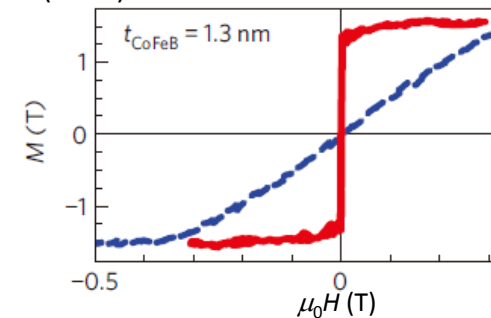
B. Rodmacq *et al.*, PRB **79**, 024423 (2009).



CoFeB(1)/MgO(0.9)/CoFeB(1.3 nm) p-MTJ

S. Ikeda *et al.*, Nat. Mater. **9**, 721 (2010).

TMR ratio = 120%  
 $K_u = 2.1 \times 10^6 \text{ erg/cm}^3$   
 $J_{co} = 3.6 \text{ MA/cm}^2$



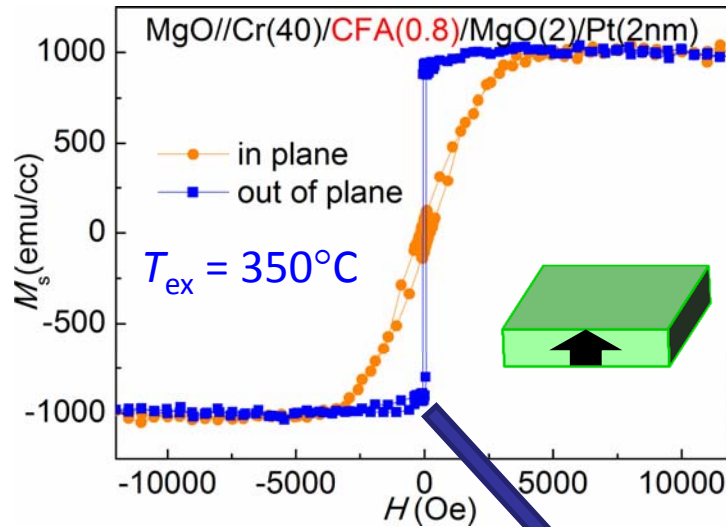
Ultra-thin Co<sub>2</sub>FeAl (CFA)/MgO structure

# Perpendicular magnetization of Co<sub>2</sub>FeAl (CFA) ultrathin films

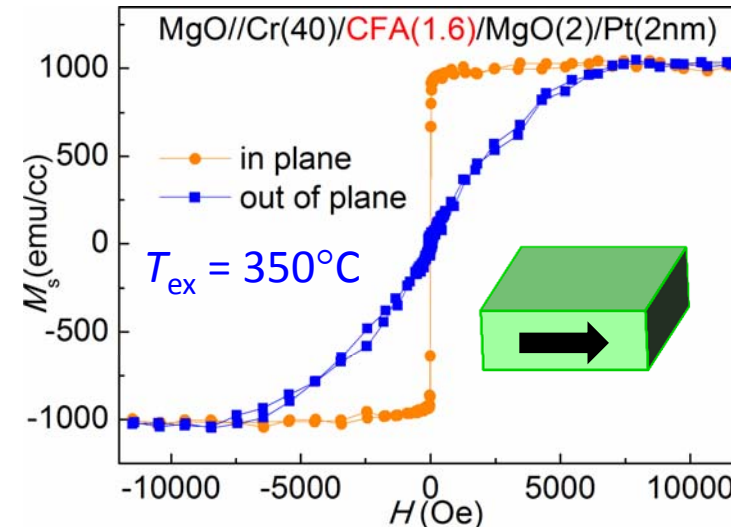
MgO(001) sub./Cr/CFA(*t* nm)/MgO/Pt

**M-H curves**

***t* = 0.8 nm: perpendicular**

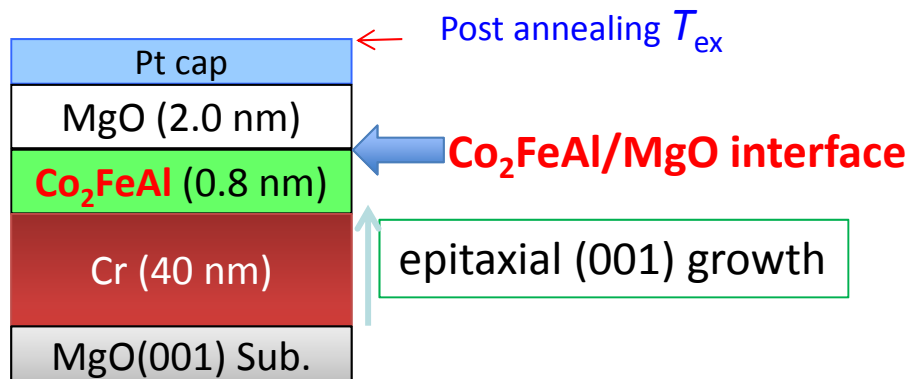


***t* = 1.6 nm: in-plane**



**Sputter deposition**

**Area enclosed by the curves: PMA energy density ( $K_u$ )**



$$K_u = 2 \times 10^6 \text{ erg/cm}^3 (\times 10^5 \text{ J/m}^3)$$

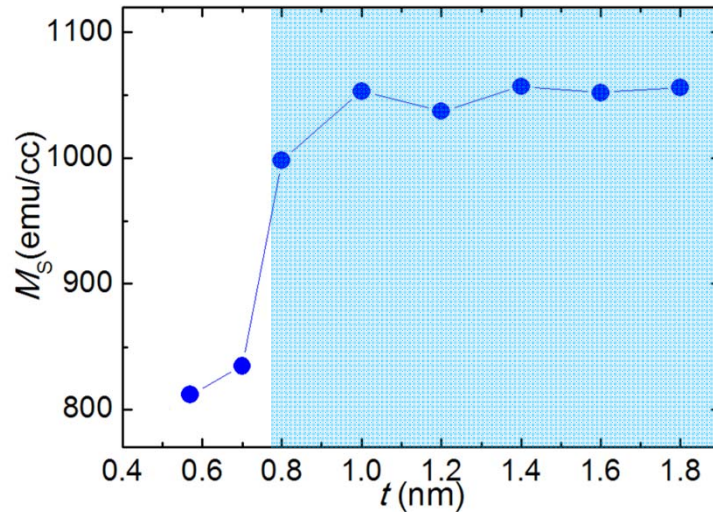
Cf. CoFeB/MgO:  $2 \sim 5 \times 10^6 \text{ erg/cm}^3 (\times 10^5 \text{ J/m}^3)$   
 \*M. Yamanouchi *et al.*, J. Appl. Phys. **109**, 07C712 (2011).

# i-PMA at an ultrathin Co<sub>2</sub>FeAl (CFA)/MgO interface

$T_{ex} = 300^\circ\text{C}$

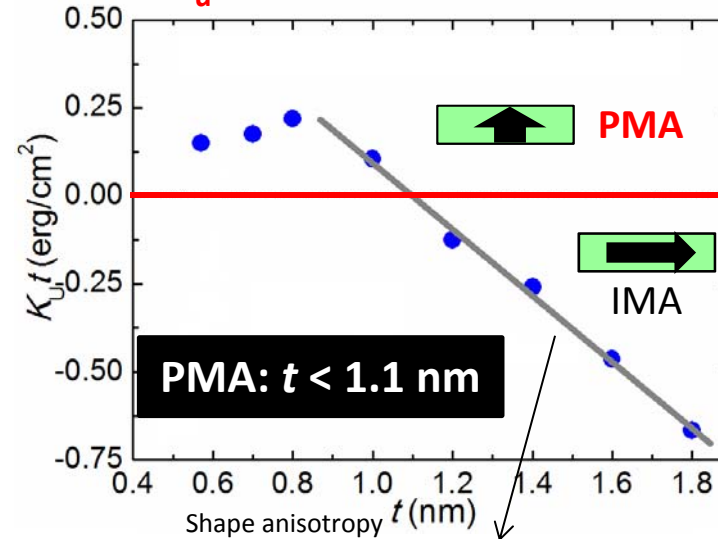
MgO(001) sub./Cr/CFA( $t$  nm)/MgO/Pt

Saturation magnetization  $M_s$

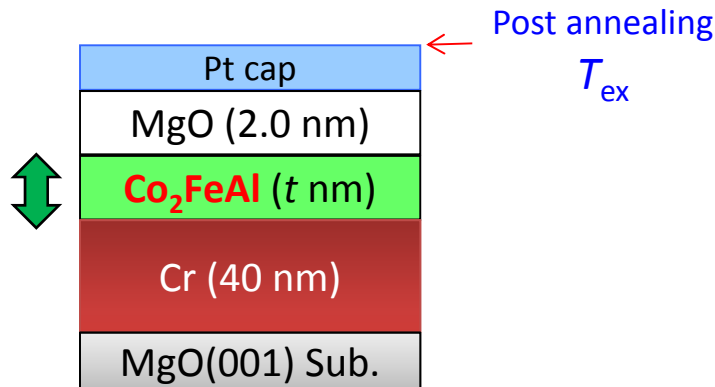


$t > 0.8$  nm:  $M_s > 1000$  emu/cm<sup>3</sup>

$K_u t$  vs.  $t$



$$K_u t = \underbrace{(K_V - 2\pi M_s^2)}_{\text{slope}} t + \underbrace{K_s}_{\text{intercept}}$$



Bulk PMA energy density  $K_V$

$K_V = -2.6 \times 10^6$  erg/cm<sup>3</sup> ( $\times 10^5$  J/m<sup>3</sup>)

In-plane  
(mainly lattice distortion)

Interfacial PMA energy density  $K_s$

$K_s = +1.04$  erg/cm<sup>2</sup> (mJ/m<sup>2</sup>)

Perpendicular

CFA/MgO interface induces PMA

# CFA/MgO/CoFeB p-MTJs on MgO(001)

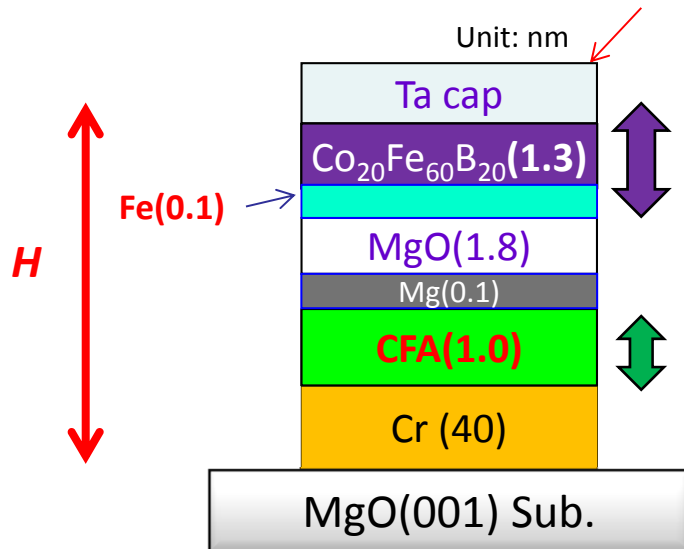


Wen et al., APEX5, 063003 (2012).

MTJs: Cr/CFA(1.0 nm)/MgO/Fe/Co<sub>20</sub>Fe<sub>60</sub>B<sub>20</sub>

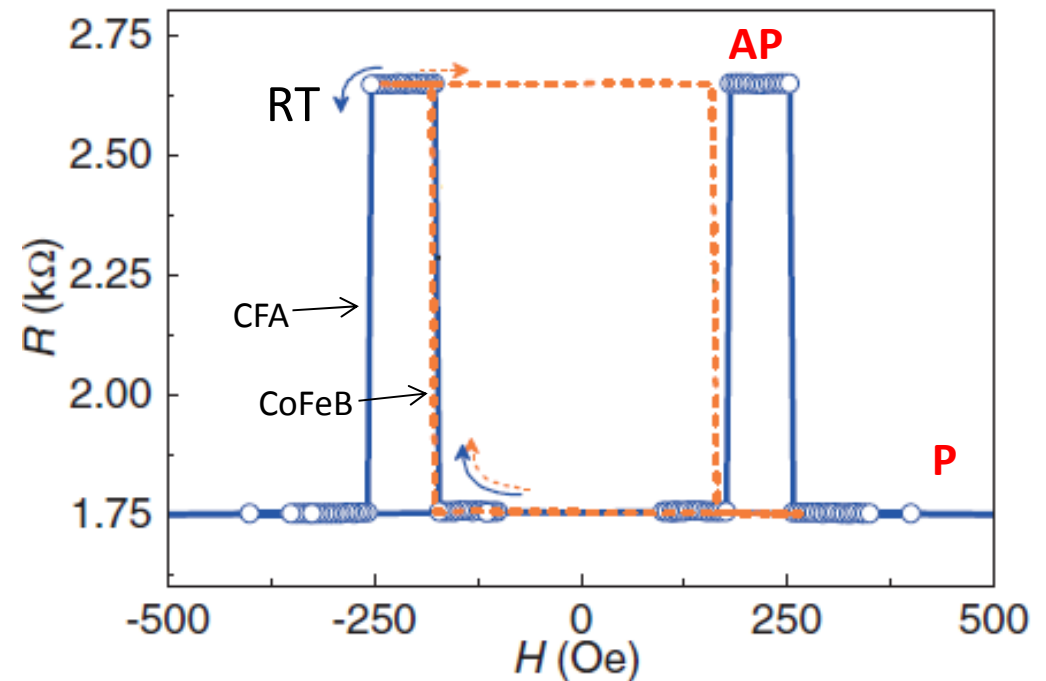
improvement of the interface structure

$T_{ex} = 325^{\circ}C$



TMR curve

Area: 10x5μm<sup>2</sup>



perp-TMR at RT = 53% ~ 91%

Ultra-thin CFA (~1 nm) still has a high effective spin polarization.

# Outline

1. Background  
Co<sub>2</sub>FeAl Heusler alloy based magnetic tunnel junctions (MTJs) for MRAM applications
2. **Perpendicular anisotropy in Co<sub>2</sub>FeAl ultra-thin films**
  - 2.1 Perpendicular magnetic anisotropy (PMA) and TMR in ultrathin Co<sub>2</sub>FeAl/MgO structures
  - 2.2 Enhanced PMA by Ru(02 $\bar{2}$ 3) underlayer**
  - 2.3 Possible PMA mechanism
3. Co<sub>2</sub>FeAl MTJs with a lattice-matched MgAl<sub>2</sub>O<sub>4</sub> coherent barrier

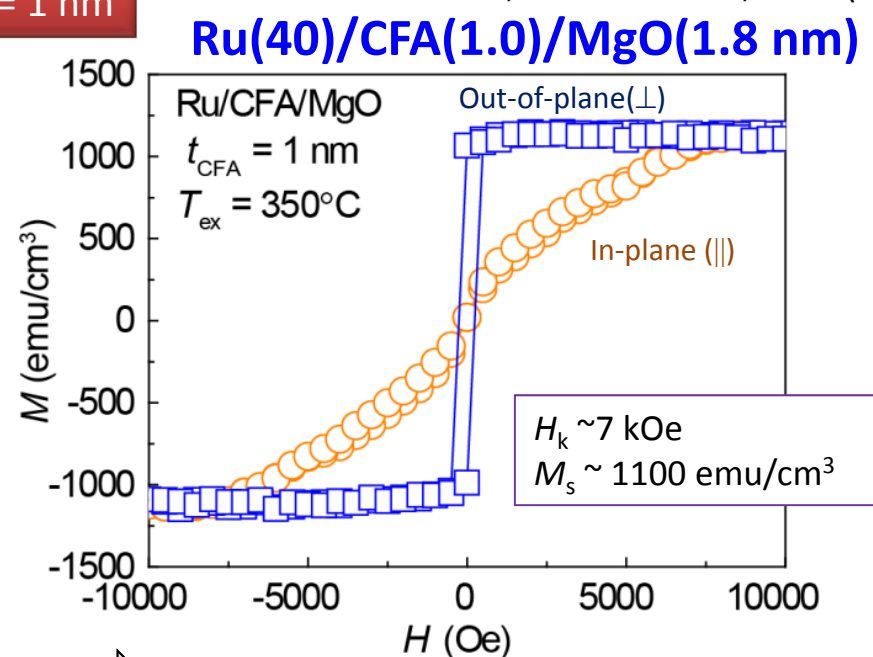
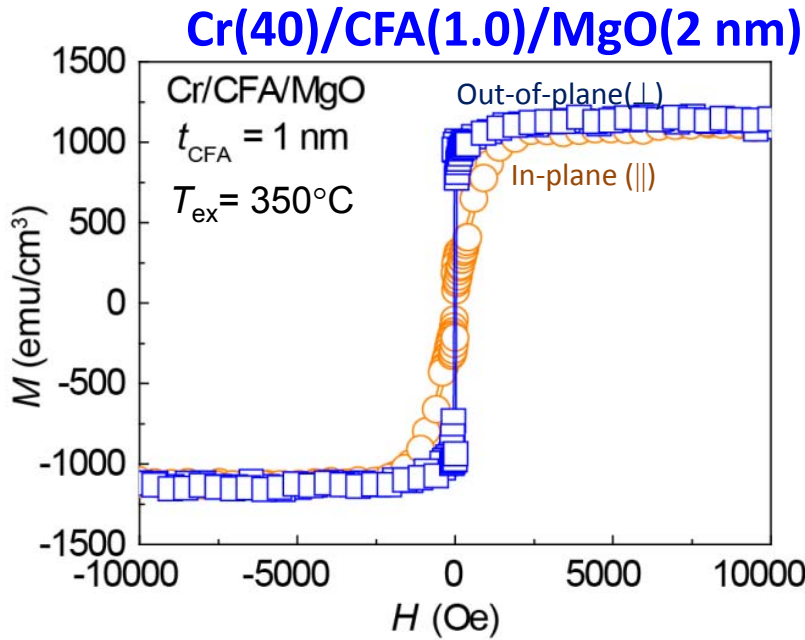




# Enhanced PMA in a Ru-buffered CFA/MgO

Z. C. Wen *et al.*, Adv. Mater. **26**, 6483 (2014).

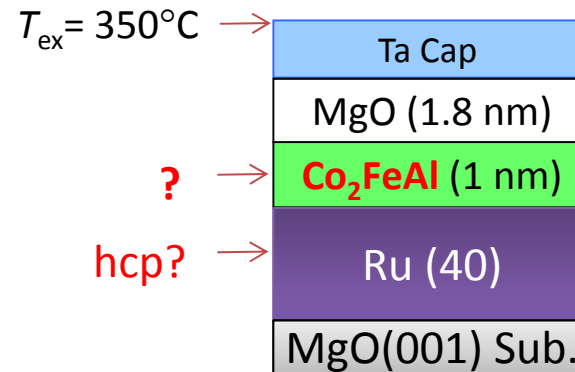
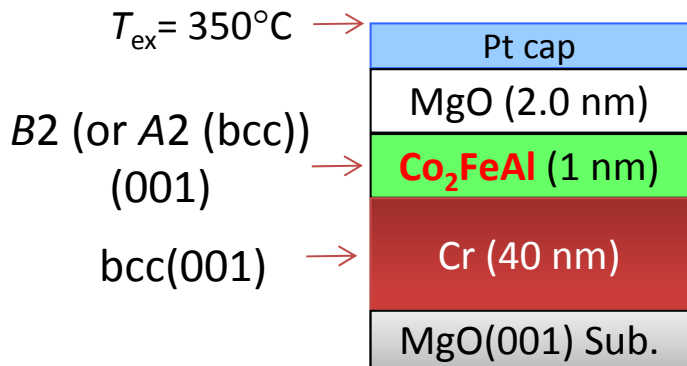
$t_{\text{CFA}} = 1 \text{ nm}$



$K_u$  Cr-buffer:  $0.8 \times 10^6 \text{ erg/cm}^3$  ( $\times 10^5 \text{ J/m}^3$ )

4 times

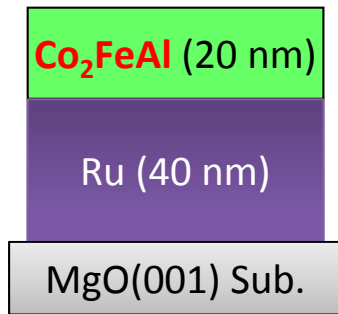
$K_u$  Ru-buffer:  $3.1 \times 10^6 \text{ erg/cm}^3$  ( $\times 10^5 \text{ J/m}^3$ )



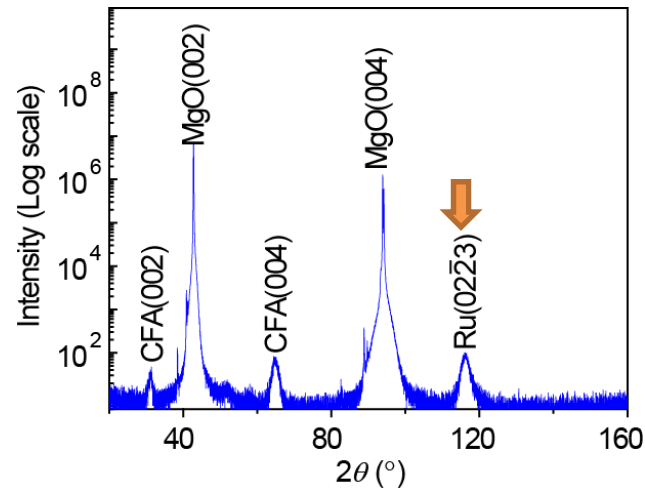
Enhancement of PMA using the Ru buffer

$K_s(\text{Cr-buffered}) = 1.0 \text{ erg/cm}^2$   
 $K_s(\text{Ru-buffered}) = 2.2 \text{ erg/cm}^2$

# XRD profiles of Ru-buffered CFA on MgO

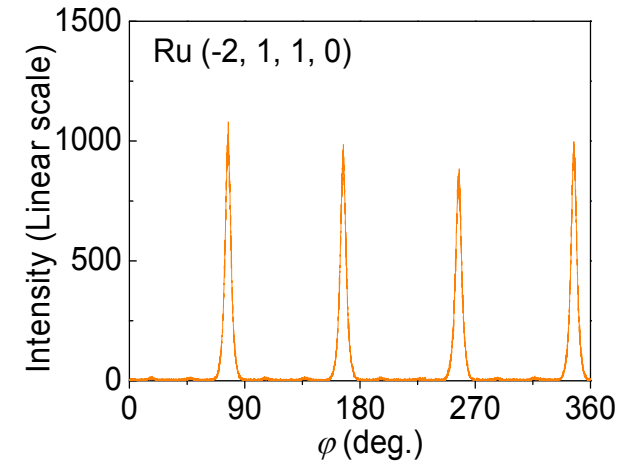


Out-of-plane ( $2\theta$ - $\omega$  scan)



Ru: Only  $(02\bar{2}3)$ , CFA: only  $(00L)$

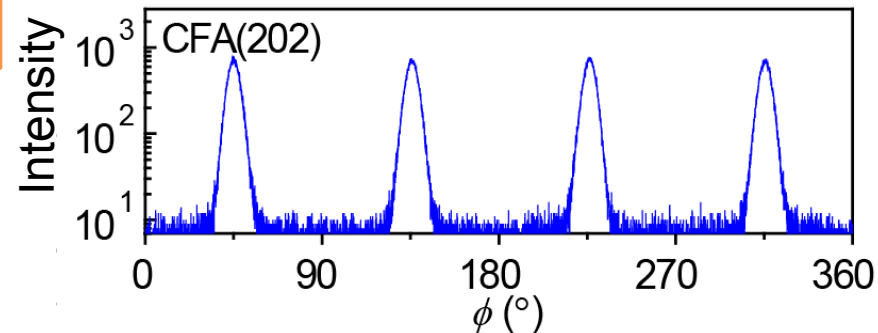
Pole scan for Ru



4-fold peaks for Ru: mixture of variants

**Epitaxial growth of 4-fold symmetry Ru $(02\bar{2}3)$  on MgO(001)**

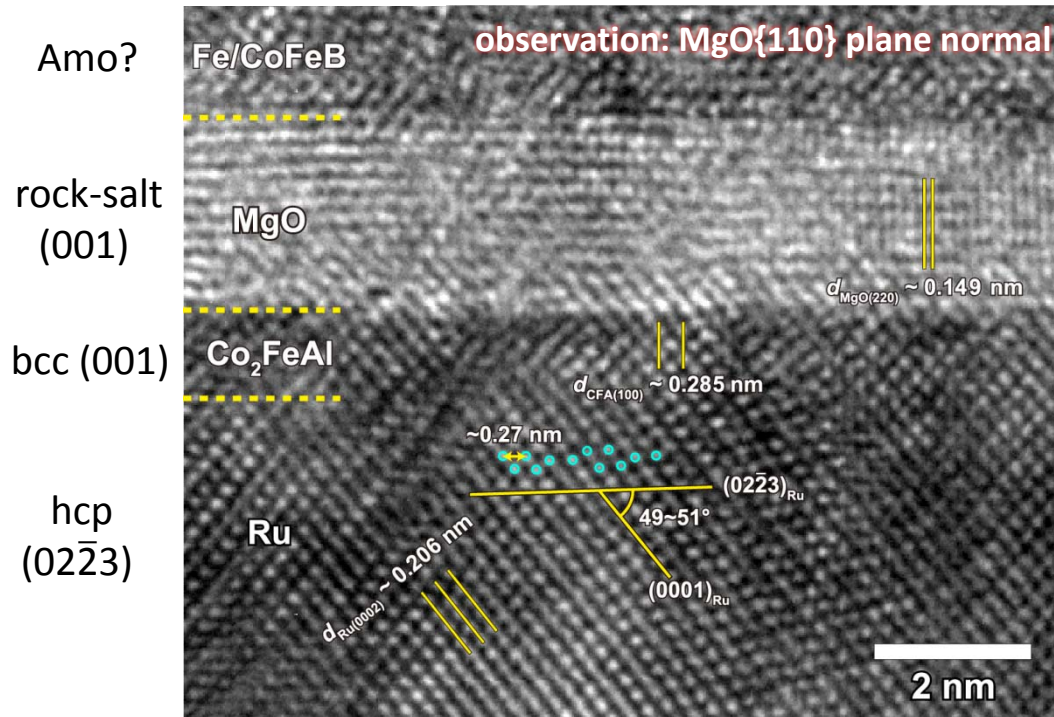
Pole scan for CFA



**Epitaxial growth of B2-ordered CFA(001) on Ru $(02\bar{2}3)$**

# HRTEM image of Ru-buffered CFA on MgO

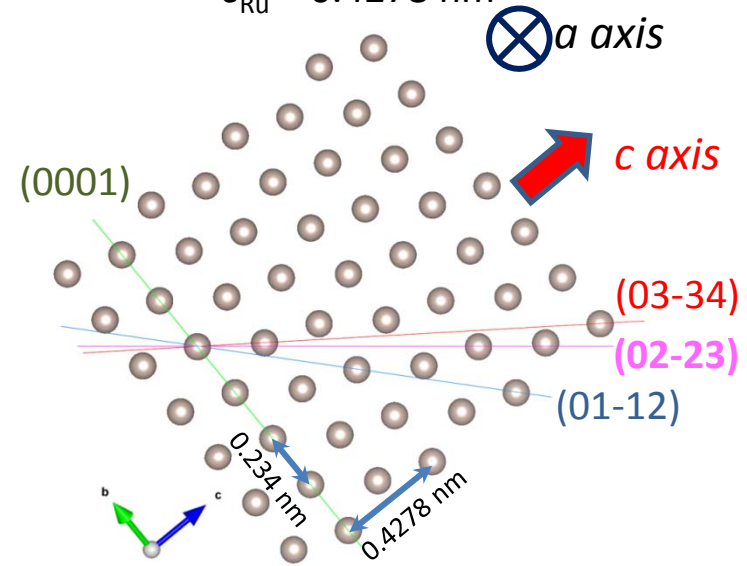
Ru (40)/CFA (1.2)/MgO (1.8)/[Fe (0.1)/CoFeB (1.3 nm)]



## Model (Bulk hcp-Ru)

$$a_{\text{Ru}} = 0.2704 \text{ nm}$$

$$c_{\text{Ru}} = 0.4278 \text{ nm}$$

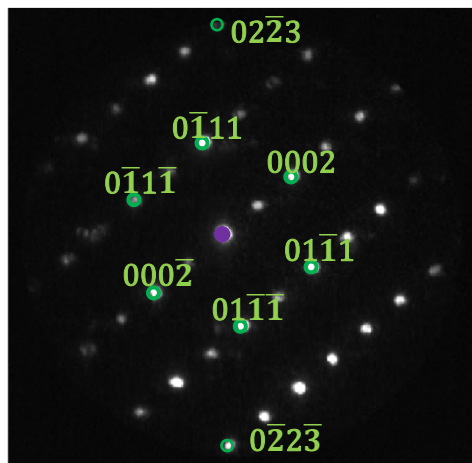


$$\angle(0001)-(02-23): 50.61^\circ$$

$$\angle(02-23)-(03-34): 3.26^\circ$$

$$\angle(02-23)-(01-12): 8.20^\circ$$

Nano electron-beam diffraction



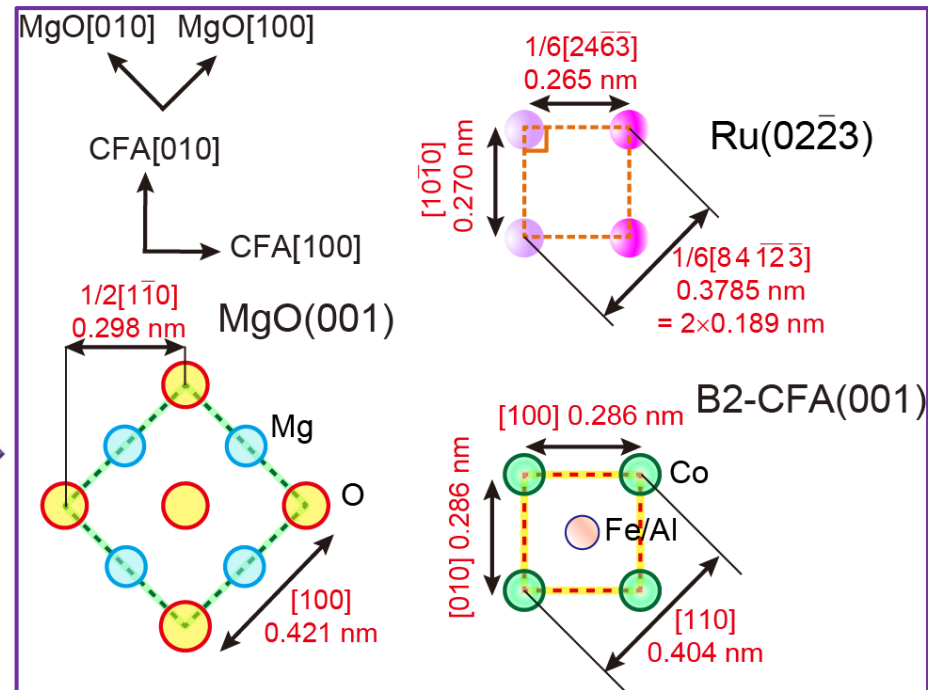
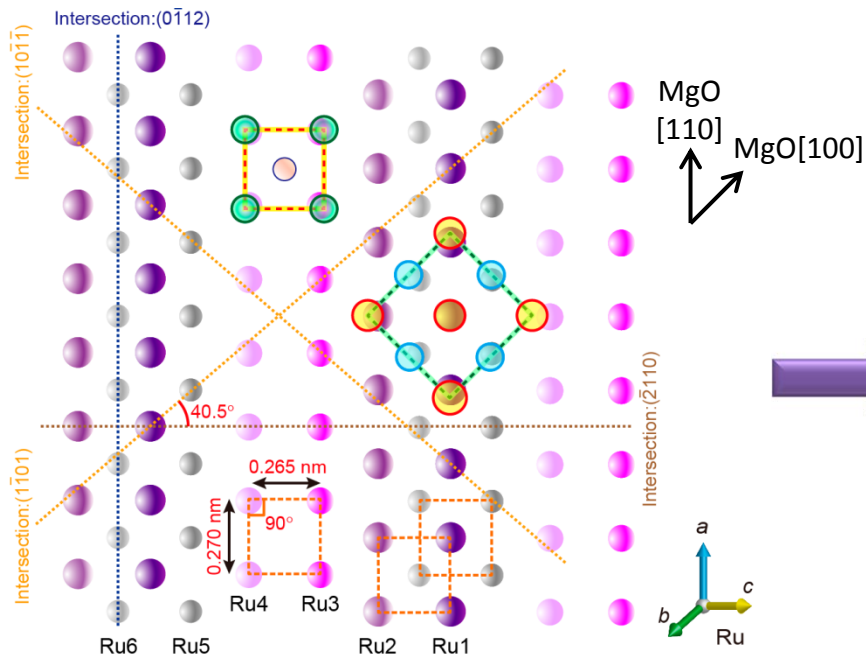
**Nearly Ru(02-23) growth**

(To be more accurate, (03-35) growth)

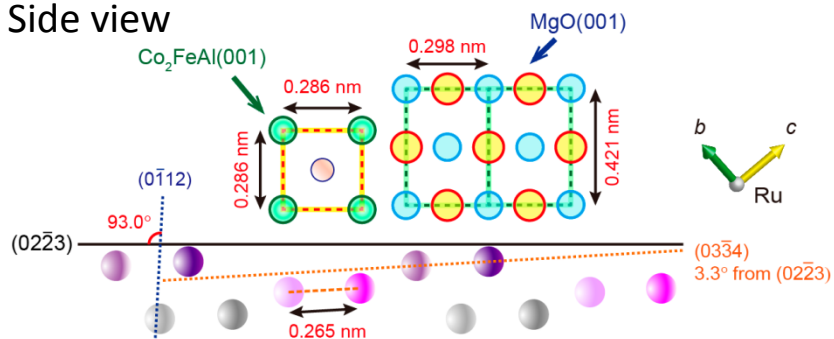
Why do CFA and MgO grow epitaxially on it?

# Epitaxial relationship of MgO/Ru/CFA heterostructure

Plan view for Ru(02 $\bar{2}$ 3)



Side view

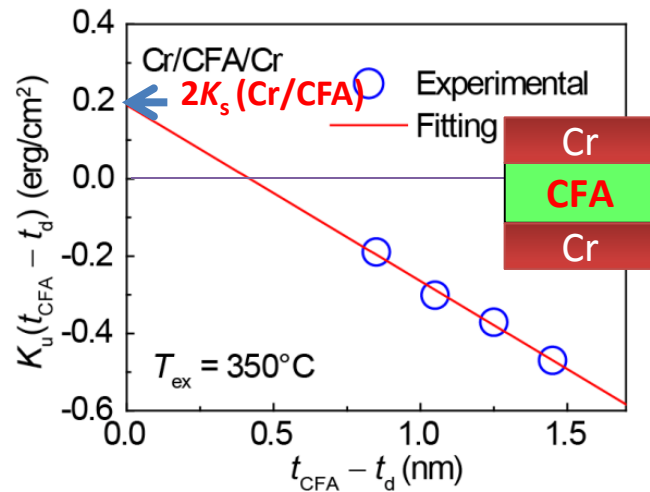


- Epitaxial relationship: MgO(001)/Ru(02 $\bar{2}$ 3)/CFA(001)
- Lattice mismatch  $\begin{cases} \sim 11\% \text{ (MgO/Ru)} \\ \sim 8\% \text{ (Ru/CFA)} \end{cases}$   
cf.  $\sim 5\%$  (MgO/Cr),  $\sim 1\%$  (Cr/CFA)

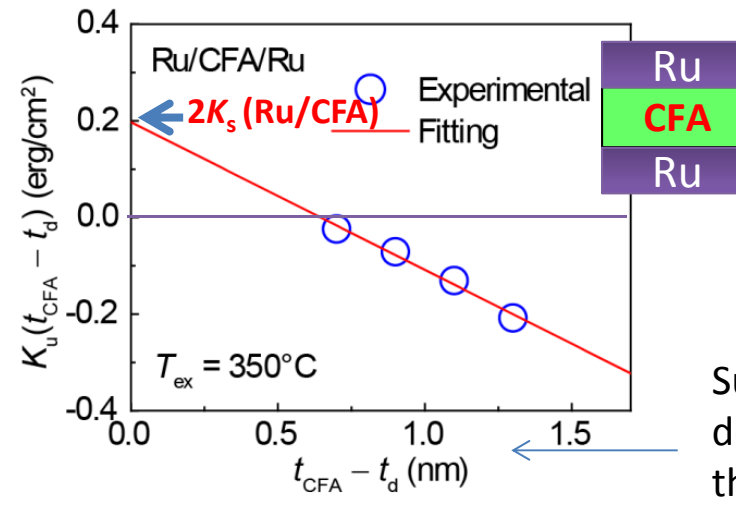
**Nearly square lattice structure of the Ru plane can act as a template of the CFA(001) growth**

# Negligible effect of buffer/CFA interface to i-PMA

## Cr/CFA/Cr



## Ru/CFA/Ru



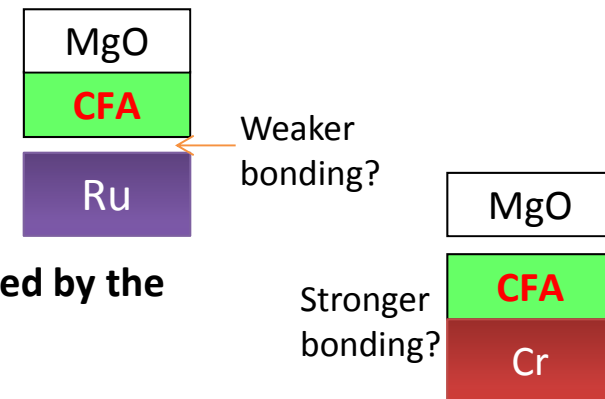
Subtraction of dead layer thickness:  $t_d$

Both Cr/CFA and Ru/CFA interfaces show negligible contribution to interface-PMA

$$K_s (\text{Cr or Ru/CFA}) \sim 0.1 \text{ erg/cm}^2$$

cf. CFA/MgO interface:  $K_s = 1.0 \text{ erg/cm}^2$  (Cr-buffer),  $2.2 \text{ erg/cm}^2$  (Ru buffer)

Possible origin of the enhanced PMA:  
**Improvement of CFA/MgO lattice combination due to a weaker effect from the Ru lattice**



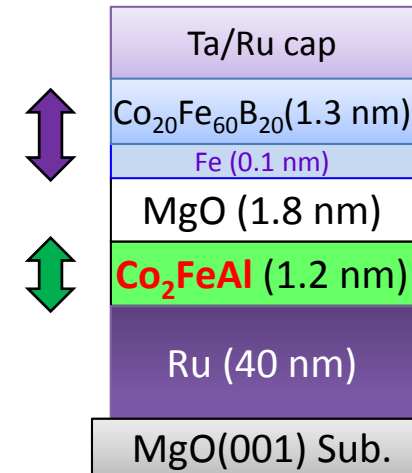
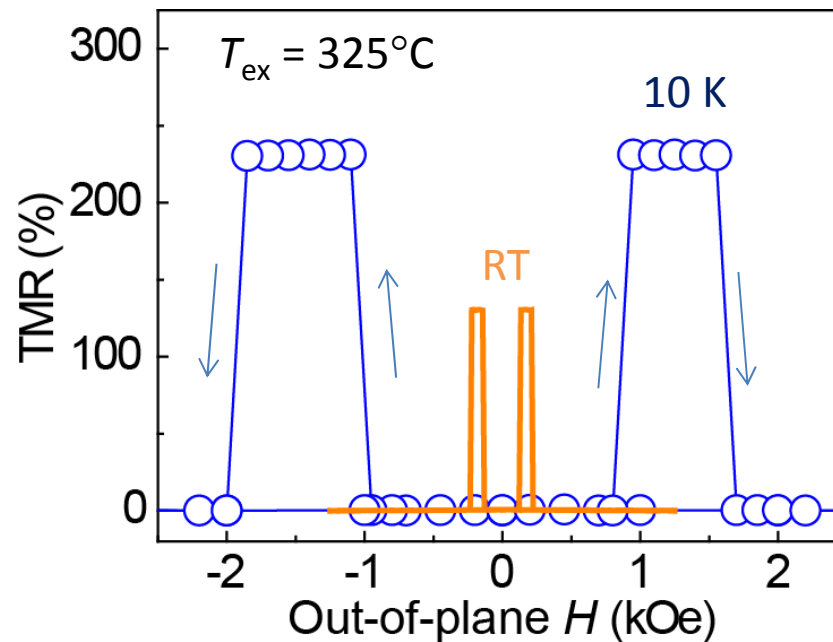
cf. Cr buffer: CFA lattice with a structure of Cr/CFA/MgO is dominated by the **Cr(001) lattice** owing to the very small lattice mismatch  
 = not a perfect CFA/MgO interface (related to lattice distortion)



# Large TMR ratio in a $\text{Co}_2\text{FeAl}/\text{MgO}/\text{CoFeB}$ p-MTJ

Z. C. Wen *et al.*, Adv. Mater. **26**, 6483 (2014).

Ru/CFA (1.2)/MgO (1.8)/Fe (0.1)/ $\text{Co}_{20}\text{Fe}_{60}\text{B}_{20}$ (1.3 nm) p-MTJ



TMR: 132% at RT (237% at 10 K)  
(Cr-buffer: 91% at RT)

Over 130% TMR at RT  
in p-MTJ using the interface PMA of  $\text{Co}_2\text{FeAl}/\text{MgO}$

The epitaxial Ru with a high crystal index opens up a new path in the development of engineered heterostructures combining **hcp and cubic or tetragonal materials**.

# Outline

1. Background  
Co<sub>2</sub>FeAl Heusler alloy based magnetic tunnel junctions (MTJs) for MRAM applications
2. **Perpendicular anisotropy in Co<sub>2</sub>FeAl ultra-thin films**
  - 2.1 Perpendicular magnetic anisotropy (PMA) and TMR in ultrathin Co<sub>2</sub>FeAl/MgO structures
  - 2.2 Enhanced PMA by Ru(02 $\bar{2}$ 3) underlayer
  - 2.3 Possible PMA mechanism**
3. Co<sub>2</sub>FeAl MTJs with a lattice-matched MgAl<sub>2</sub>O<sub>4</sub> coherent barrier

# Possible origin of i-PMA in bcc-Fe/MgO system

K. Nakamura *et al.*, PRB **81**, 220409(R) (2010).

H. X. Yang *et al.*, PRB **84**, 054401 (2011).

**Hybridization between oxygen and Fe atoms shifts up  $m = 0$  ( $3d_{z^2}$ ) above  $E_F$**

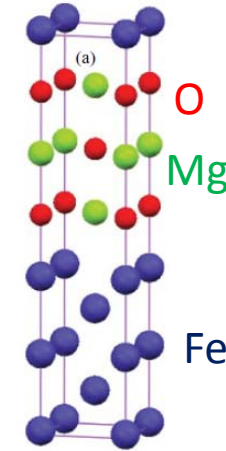
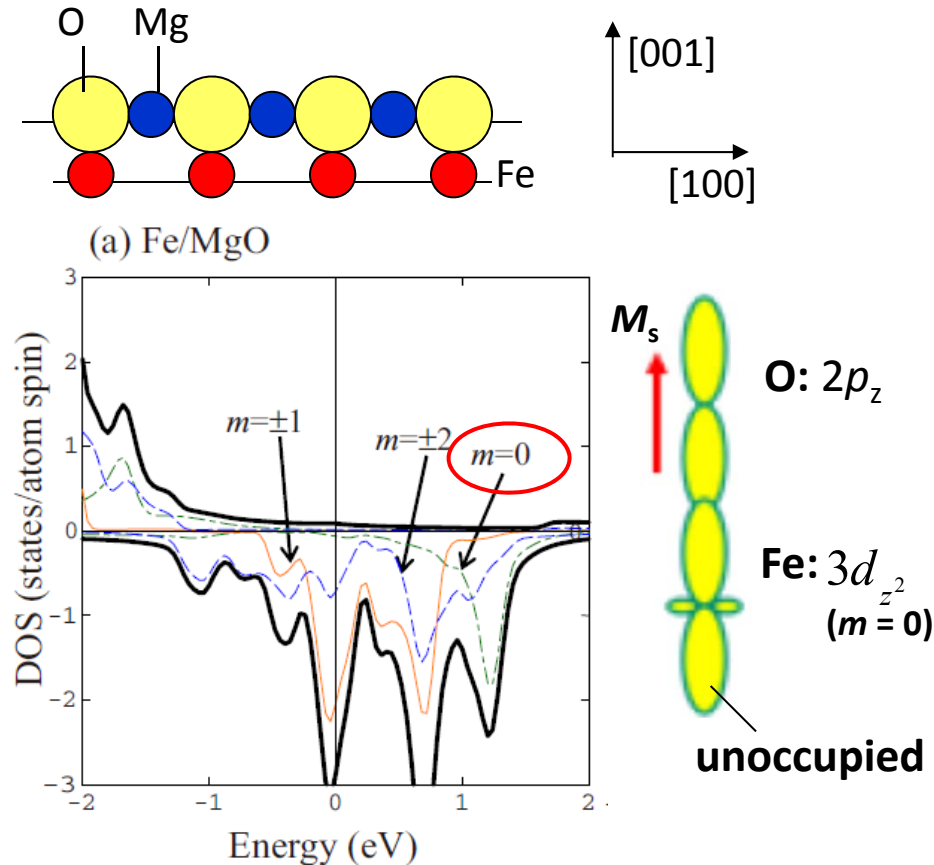


TABLE I. PMA value ( $\text{erg}/\text{cm}^2$ ) and magnetic moment  $m$  [ $\mu_B$  per Fe(Co) atom] for different layers of Fe(Co) in Fe(Co)|MgO magnetic tunnel junctions under different oxidation conditions.

		Fe MgO			Co MgO:
		Pure	Underoxidized	Overoxidized	pure
PMA	<b>Fe</b>	2.93	2.27	0.98	0.38
$m$	( $\mu_B$ )				
	Interfacial	2.73	2.14	3.33	1.67
	Sublayer	2.54	2.41	2.70	1.84
	Bulk	2.56	2.55	2.61	1.60

Hybridization between **oxygen  $2p_z$**  and **Fe  $3d_{z^2}$**  orbitals plays an important role for PMA in the Fe/MgO system

The calculated PMA energy density of **Fe/MgO** is much larger than that of **Co/MgO**

# Co<sub>2</sub>FeAl = Co rich (50 at.%) alloy

However, the band structures of Co-based Heusler alloys are considerably different from those of Fe or Co owing to their half-metallic properties.



## Bruno model:

spin-orbit interaction is treated as a perturbation for the tight binding approximation

Anisotropy of orbital magnetic moments between perpendicular ( $m_{\text{orb}}^{\perp}$ ) and in-plane directions ( $m_{\text{orb}}^{\parallel}$ ) is proportional to PMA (for 3d transition metals).

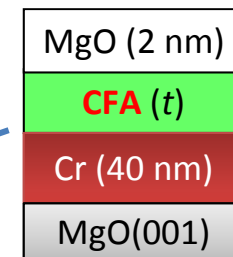
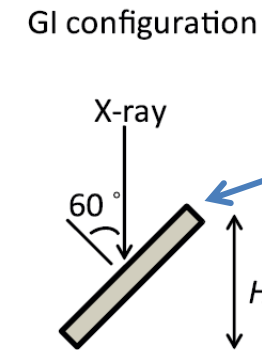
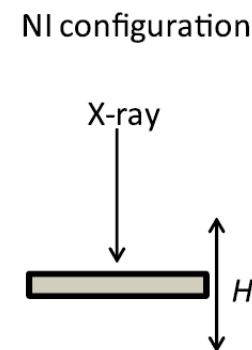
Angular-dependent x-ray magnetic circular dichroism (XMCD) study

To deduce the **orbital magnetic moments** along perpendicular and in-plane directions

$$K = \alpha \xi (m_{\text{orb}}^{\perp} - m_{\text{orb}}^{\parallel})$$

↑  
Element specific  
PMA energy

↑  
Spin-orbit splitting  
(33 meV for Fe)

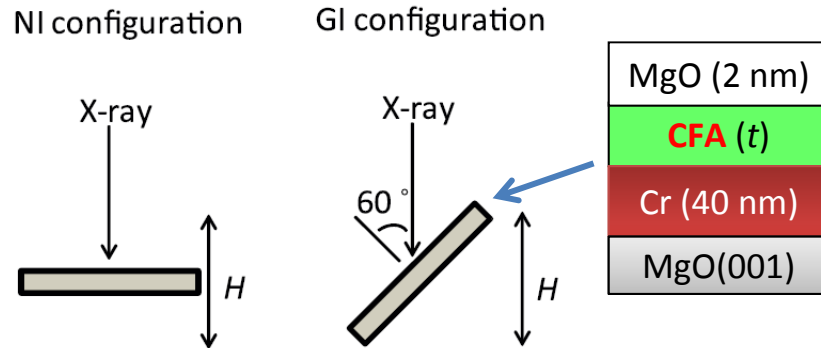


$$m_{\text{orb}}(\theta) = m_{\text{orb}}^{\parallel} \sin^2 \theta + m_{\text{orb}}^{\perp} \cos^2 \theta$$

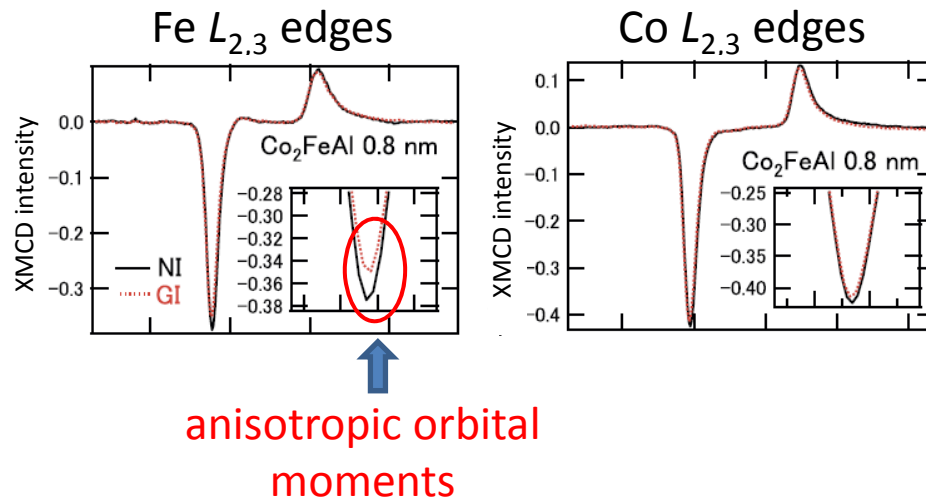
# Angular-dependent XMCD study: hybridization Fe-O

In collaboration with Prof. Okabayashi (Univ. Tokyo)  
BL-7A in Photon Factory, KEK

J. Okabayashi, HS *et al.*, APL103, 102402 (2013).



Spectra taken in normal incidence (NI) and grazing incidence (GI) revealed the anisotropic orbital moments.



$$m_{\text{orb}}(\theta) = m_{\text{orb}}^{\parallel} \sin^2 \theta + m_{\text{orb}}^{\perp} \cos^2 \theta$$

TABLE I. Spin magnetic moments ( $m_{\text{spin}}$ ) and orbital magnetic moments of parallel and perpendicular components  $m_{\text{orb}}^{\perp}$ ,  $m_{\text{orb}}^{\parallel}$ , respectively, of Fe and Co in  $\text{Co}_2\text{FeAl}$  layers of different thickness. The unit is  $\mu_B$ .

	0.8 nm PMA		2 nm ← in-plane anisotropy → 10 nm			
	Fe	Co	Fe	Co	Fe	Co
$m_{\text{spin}}$	1.83	1.21	2.08	1.22	1.91	1.21
$m_{\text{orb}}^{\perp}$	0.32	0.16	0.22	0.14	0.21	0.13
$m_{\text{orb}}^{\parallel}$	0.24	0.16	0.21	0.15	0.22	0.13

Large **anisotropic Fe orbital moments** at the CFA/MgO interface ( $m_{\text{orb}}^{\perp}(\text{Fe}) > m_{\text{orb}}^{\parallel}(\text{Fe})$ )

Bruno model: 0.37 erg/cm<sup>2</sup> (mJ/m<sup>2</sup>)

XMCD result suggests **contribution of Fe atoms** (negligible small for Co) in CFA



# Theoretical predictions of PMA at CFA/MgO interface

*Ab initio* calculation by Prof. Shirai Group (Tohoku U)

D. Mori *et al.*, EB-07, Intermag 2012

PMA arises from **Co-terminated interface** for CFA/MgO(001) system (MgO/L<sub>21</sub>-CFA/MgO)

$K_s$ : Interfacial PMA energy density

**Co-terminated case**

$$K_s^{\text{Co}} = 0.99 \text{ erg/cm}^2 \text{ (mJ/m}^2\text{)} \text{ --- PMA}$$

**FeAl-terminated case**

$$K_s^{\text{FeAl}} = -0.49 \text{ erg/cm}^2 \text{ --- In-plane MA}$$

$$K_s(\text{exp. Cr-buffered}) = 1.0 \text{ erg/cm}^2$$

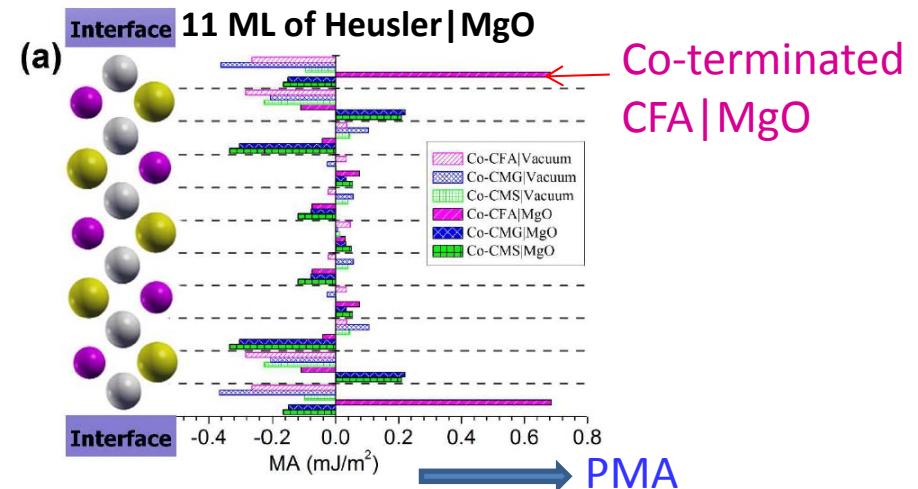
$$K_s(\text{exp. Ru-buffered}) = 2.2 \text{ erg/cm}^2$$

**Relevant report**

R. Vadapoo, A. Hallal, M. Chshiev (SPINTEC group)  
arXiv:1404.5646v1

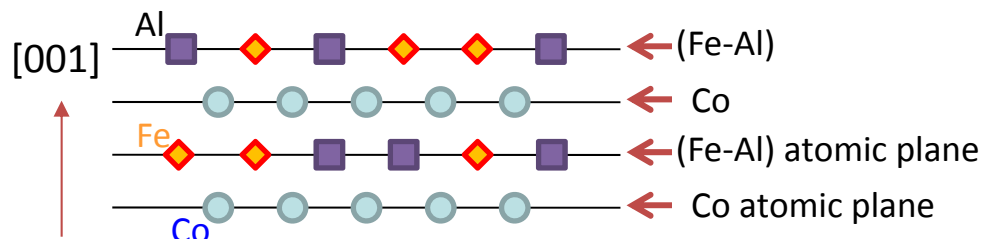
**Co-terminated case**

$$K_s^{\text{Co}} \sim 1.3 \text{ erg/cm}^2 \text{ (mJ/m}^2\text{)} \text{ --- PMA}$$



PMA is theoretically expected only in **Co-terminated CFA**, however **FeAl-terminated CFA** is more thermodynamically stable

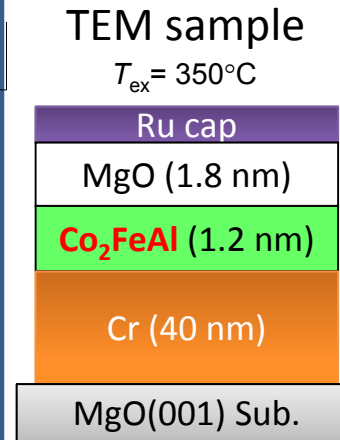
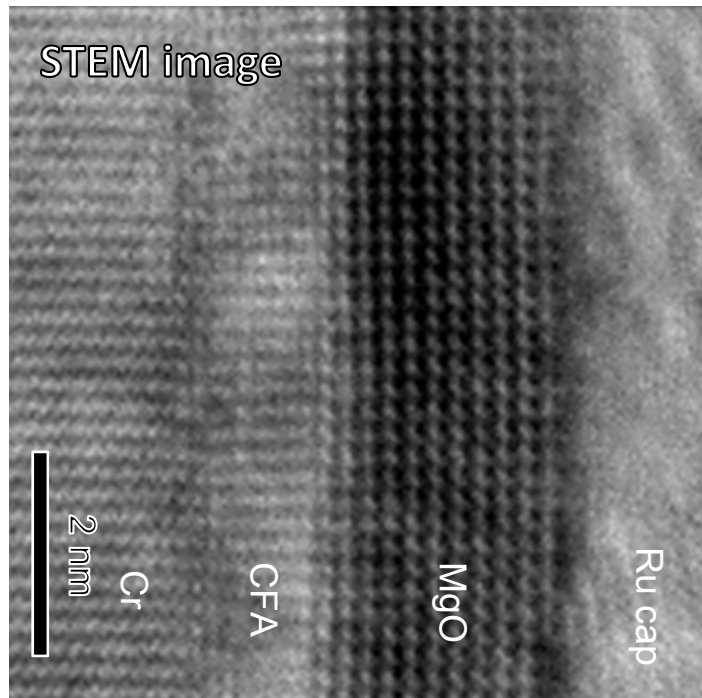
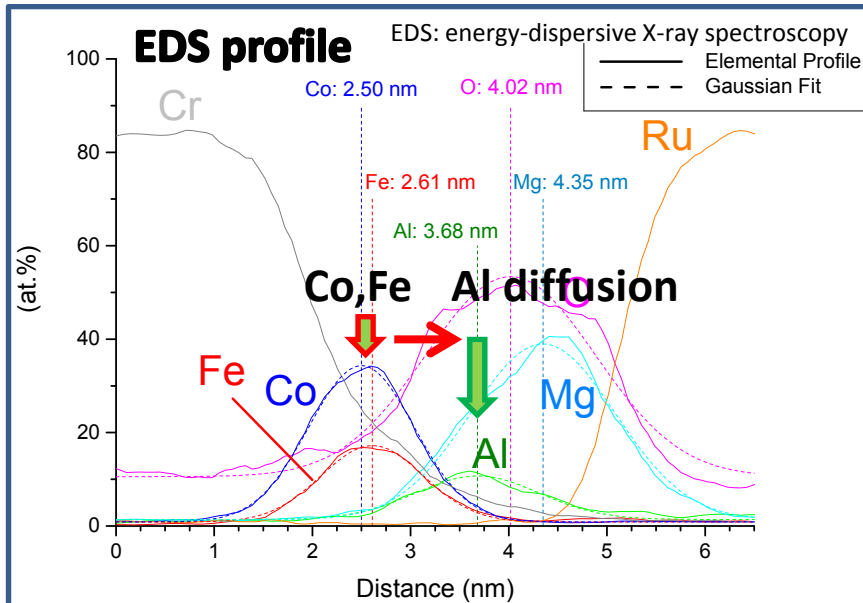
**Atomic model of B2-Co<sub>2</sub>FeAl(001)**



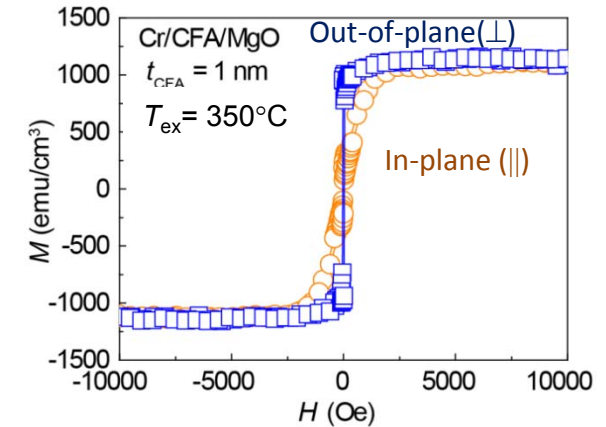
**Termination layer of CFA?**

**Determination of the interfacial structure is needed**

# Al interdiffusion from CFA layer to MgO barrier

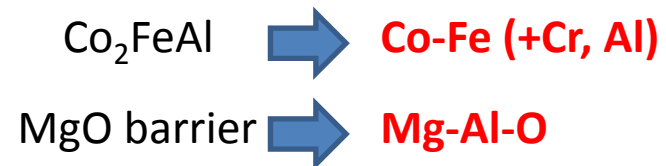


Wen *et al.*, unpublished.



- Significant Al diffusion from CFA into MgO
- The CFA layer is no longer a “Heusler alloy”

## Solid-phase reaction at the CFA/MgO interface



Enhancement of hybridization between Fe and O orbitals? (under investigation)

Importance of sub-nm scale structural analyses

# Outline

1. Background  
Co<sub>2</sub>FeAl Heusler alloy based magnetic tunnel junctions (MTJs) for MRAM applications
2. Perpendicular anisotropy in Co<sub>2</sub>FeAl ultra-thin films
  - 2.1 Perpendicular magnetic anisotropy (PMA) and TMR in ultrathin Co<sub>2</sub>FeAl/MgO structures
  - 2.2 Enhanced PMA by Ru(002̄3) underlayer
3. Co<sub>2</sub>FeAl MTJs with a lattice-matched MgAl<sub>2</sub>O<sub>4</sub> coherent barrier (In-plane magnetized MTJ)



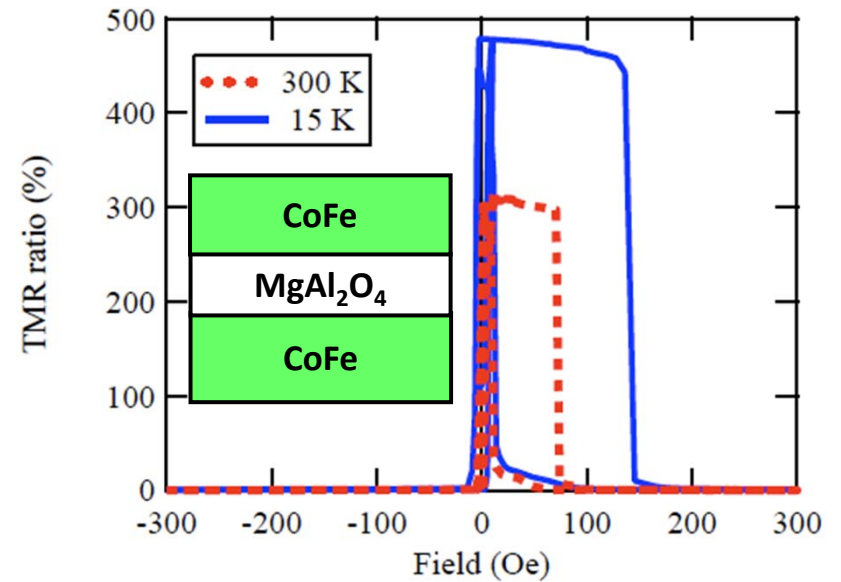
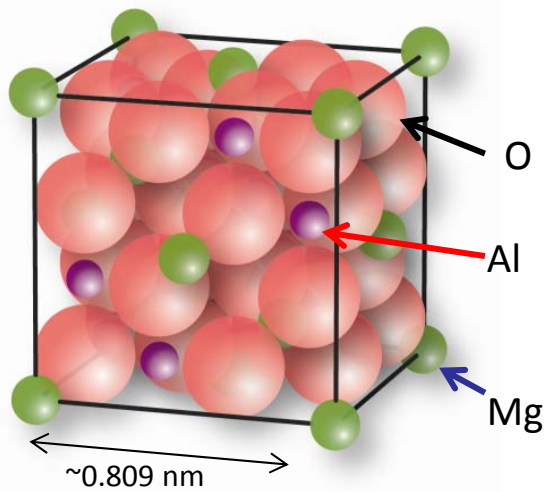
[T. Scheike](#)

# New coherent barrier material: Spinel $MgAl_2O_4$

- Giant RT tunnel magnetoresistance (TMR)
- Non-deliquescence
- Tunable lattice parameter: Good lattice matching with 3d ferromagnetic electrodes

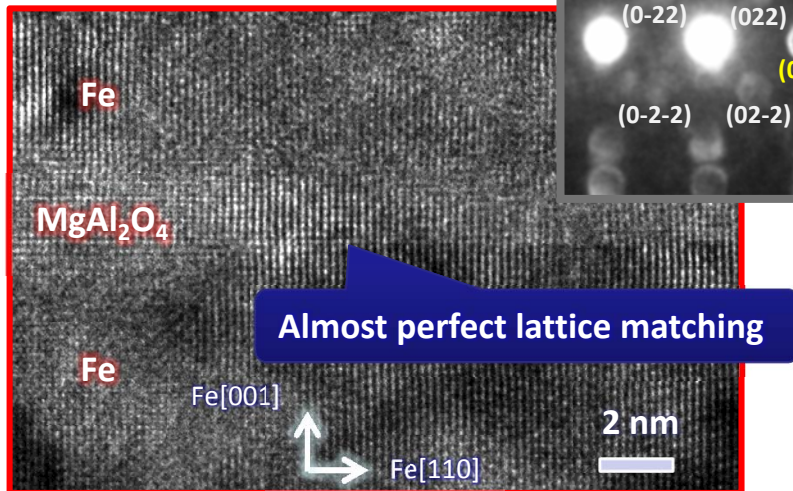
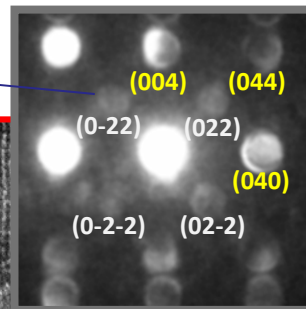
R. Shan, HS *et al.*, Phys. Rev. Lett. **102**, 246601 (2009)

H. Sukegawa *et al.* Appl. Phys. Lett **96**, 212505 (2010), Phys. Rev. B **86**, 184401 (2012)



**Max TMR ratio : 328 % (RT)**  
**: 494 % (4 K)**

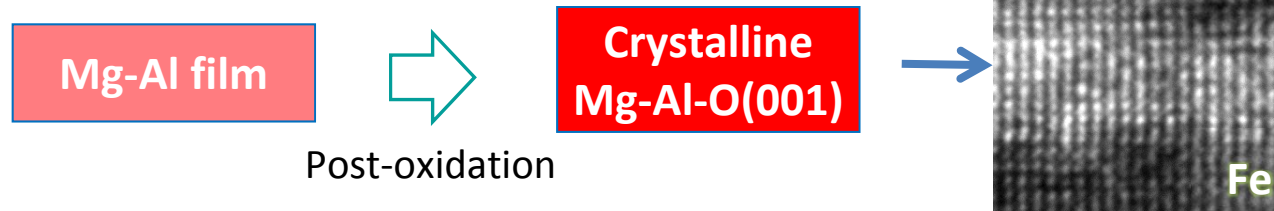
{220} spots: Spinel structure



**Giant TMR owing to coherent tunneling effect in non-MgO barrier**

$\Delta_1$  transport mechanism is still valid!

# Current status of spinel-based MTJs



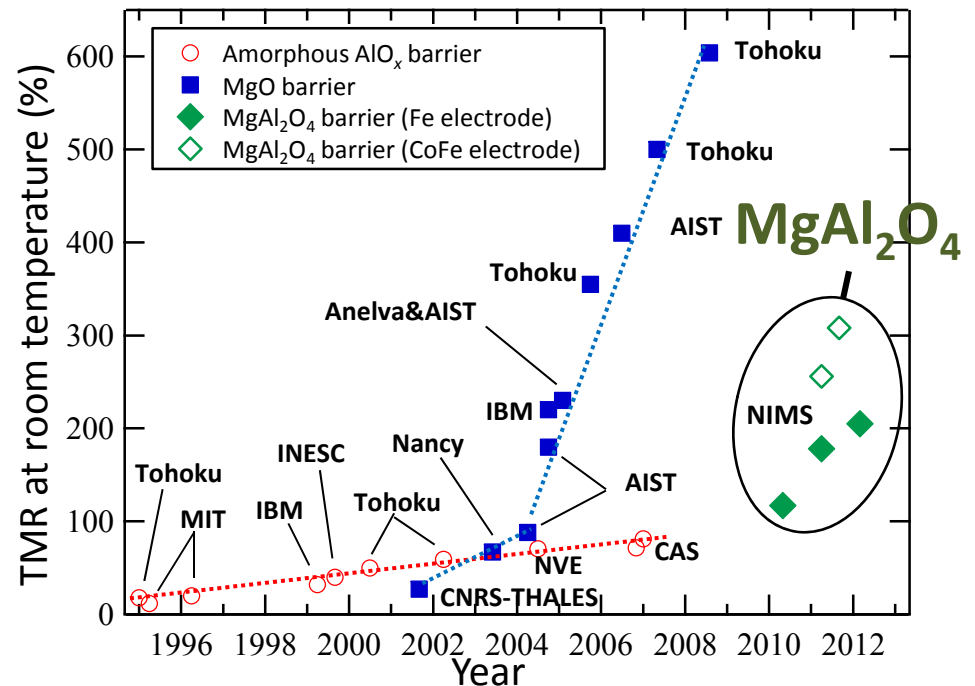
- Perfect lattice matching with 3d FM/Co-based Heuser alloys Phys. Rev. Lett. **102**, 246601 (2009).
- Large TMR: >300% Phys. Rev. B **86**, 184401 (2012).
- Wide RA range:  $10^0$ - $10^6 \Omega \cdot \mu\text{m}^2$  (STT switching) Appl. Phys. Lett. **103**, 142409 (2013); ibid.**105**, 092403 (2014).
- Interface-induced PMA Status Solidi RRL **8**, 841 (2014).

## Lattice mismatch

Electrode materials	Mismatch (%) for (001) growth	
	vs. MgO (0.421 nm)	vs. <b>MgAl<sub>2</sub>O<sub>4</sub>-based</b> (0.396 ~ 0.404 nm)
bcc-Fe ( $a = 0.2866$ nm)	-3.8	<b>+0.3 ~ +2.5</b>
<b>L2<sub>1</sub>-Co<sub>2</sub>FeSi</b> ( $a = 0.564$ nm)	-5.3	<b>-1.4 ~ +0.8</b>
L1 <sub>0</sub> -FePt ( $a = 0.385$ nm)	-8.6	<b>-4.8 ~ -2.7</b>
DO <sub>22</sub> -MnGa ( $a = 0.390$ nm)	-7.4	<b>-3.4 ~ -1.4</b>

Next generation spintronics materials

## TMR at RT (CoFe(B) electrodes)

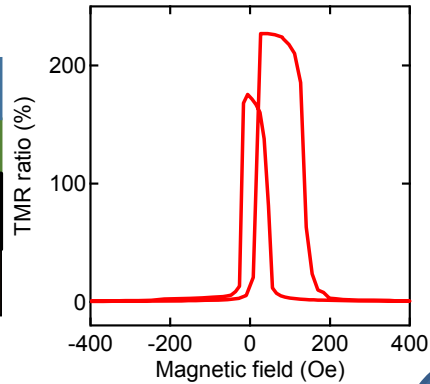
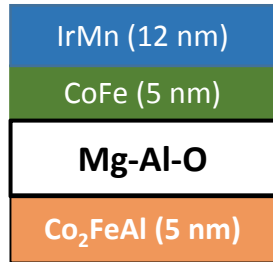




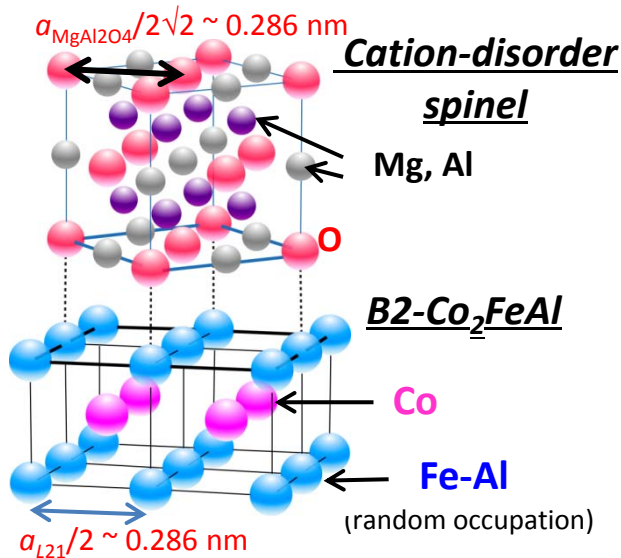
# Lattice-matched Heusler $\text{Co}_2\text{FeAl}/\text{MgAl}_2\text{O}_4$ MTJs

T. Scheike *et al.*, APL105, 242407 (2014).

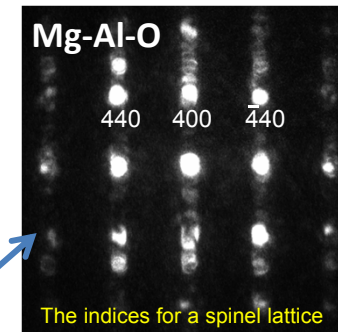
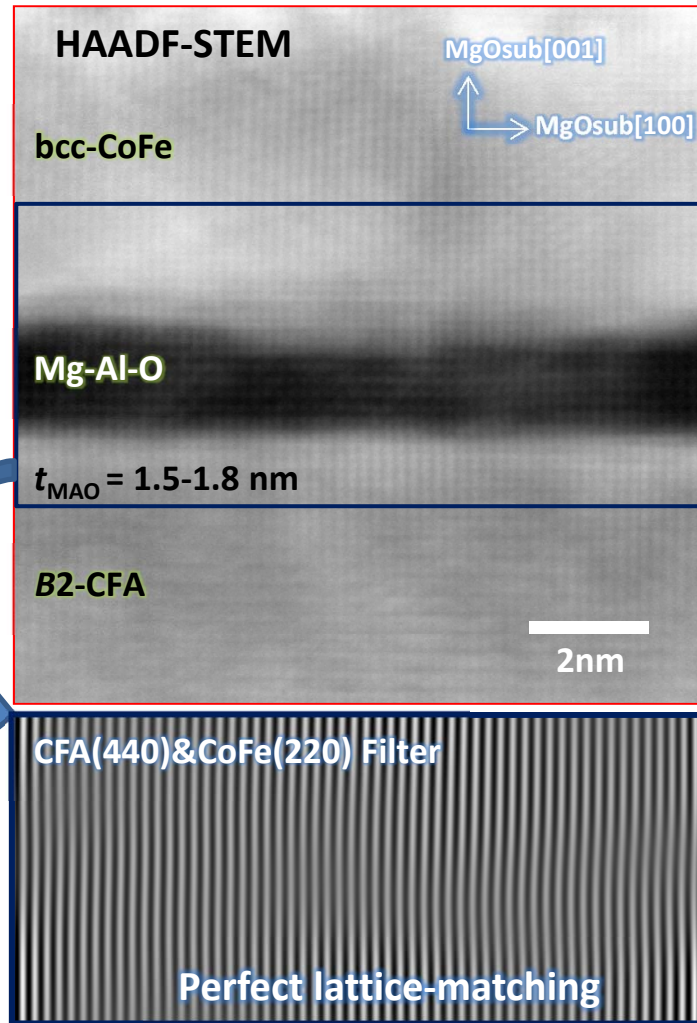
In-plane magnetized MTJ



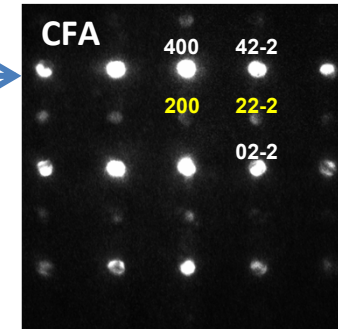
TMR: 236%  
RA: 1.8  $\text{k}\Omega \cdot \mu\text{m}^2$



Misfit: ~0% (cf. MgO/CFA ~3.8%)



Cation-disorder spinel structure



B2 CFA

**Giant TMR effect in CFA/Mg-Al-O/CoFe MTJs**

236-280% TMR at room temperature

Latest data > 340%



# Conclusion

## Co<sub>2</sub>FeAl-based heterostructures

### Interface-induced PMA

- (1) Perpendicular magnetic anisotropy in a Cr/Co<sub>2</sub>FeAl/MgO ( $K_s \sim 1 \text{ erg/cm}^2$ ) and p-TMR of **91%** at RT.
- (2) Ru buffer layer as a new ferromagnetic layer growth: Unusual crystallographic orientation Ru(02-23) buffer enhanced PMA ( $K_s \sim 2.2 \text{ erg/cm}^2$ ) and p-TMR (**132%** at RT)
- (3) **Al interdiffusion** from CFA layer to barrier was confirmed; the Fe-O hybridization owing to the interfacial reaction could be responsible for the strong PMA.

	Cr buffer	Ru buffer
Buffer orientation	bcc (001)	hcp ~(02 $\bar{2}$ 3)
$K_u$ (Merg/cm <sup>3</sup> ) for 1-nm	0.8	3.1
$K_s$ (erg/cm <sup>2</sup> )	1.0	2.2
TMR (%)	91 (RT)	132 (RT)

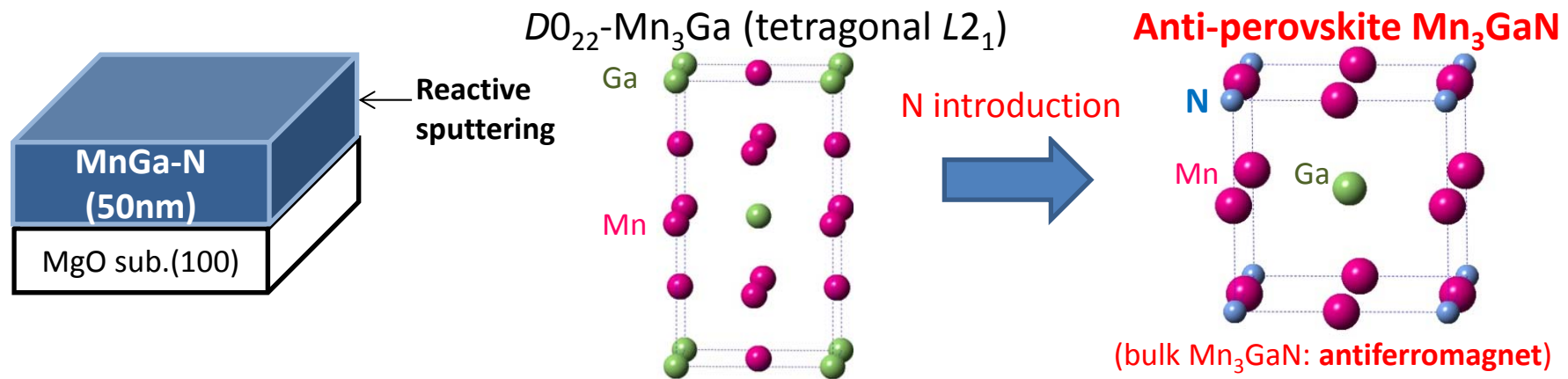
Review of PMA in CFA-based heterostructures → Sukegawa *et al.*, SPIN **4**, 1440023 (2014).

### Lattice-matched MTJ using Spinel barrier

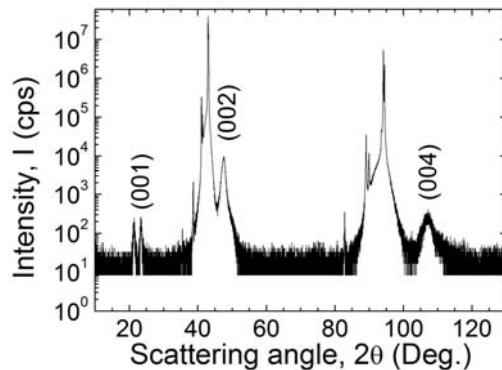
Lattice-matched CFA/MgAl<sub>2</sub>O<sub>4</sub>/CoFe MTJs were successfully developed and giant TMR more than 280% at RT was demonstrated.

**CFA-based (or CFA-derived) heterostructures will be a promising candidate for future spintronics applications**

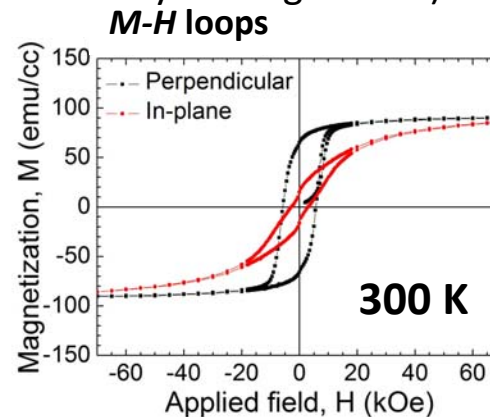
# New perpendicular magnetization material: *N-deficient MnGaN*



Mn<sub>57</sub>Ga<sub>32</sub>N<sub>11</sub> (**N-deficient**, but structurally homogeneous)

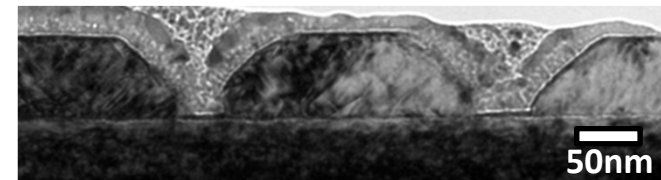


**Antiperovskite ( $E2_1$ ) single phase**

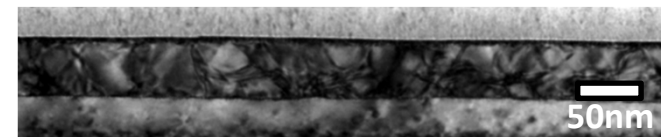


**PMA film**

**Cross-sectional TEM**



$DO_{22}$ -MnGa: very rough



**$E2_1$ -MnGaN: atomically flat**

- Ferromagnetic MnGaN with PMA ( $K_u \sim 0.2$  MJ/m<sup>3</sup>)
- Flat film structure
- Low saturation magnetization ( $\sim 100$  emu/cc)
- Curie temperature  $\gg$  room temperature
- Relatively high spin-polarization  $\sim 57\%$  (PCAR)

**New PMA material for spintronics**

Lee *et al.*, APL**107**, 032403 (2015).