

筑波大学  
University of Tsukuba

# Heusler alloy CPP spin-valves for ultrathin reader applications

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<http://www.nims.go.jp/apfim/>

## Acknowledgments



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S. Mitani, A. Rajanikanth, S. V. Karthik, A. Srinivasan



T. Taniguchi, H. Imamura



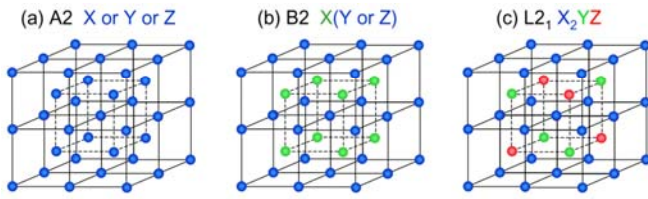
O. Mryasov



H. Iwasaki, M. Takagishi, K. Koi

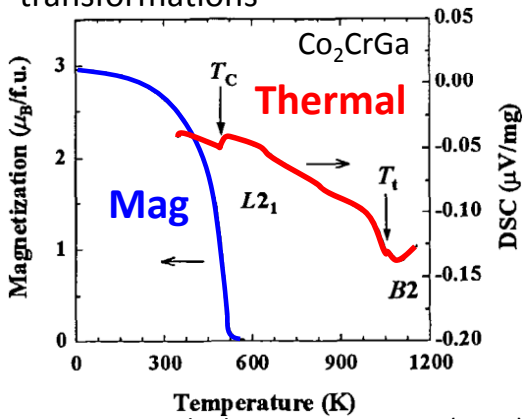
Hitachi Global Storage Technologies J. R. Childress

# Half-metallic Heusler alloy

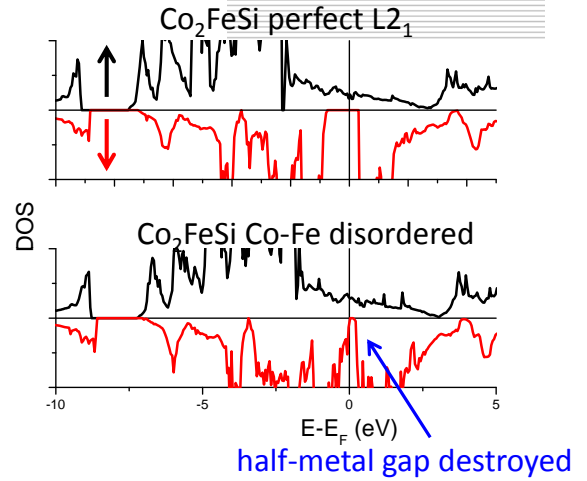


Co<sub>2</sub>MnGe, Co<sub>2</sub>FeSi etc.

Magnetic and order-disorder transformations



Umetsu Appl. Phys. Lett. 85, 2011 (2004)

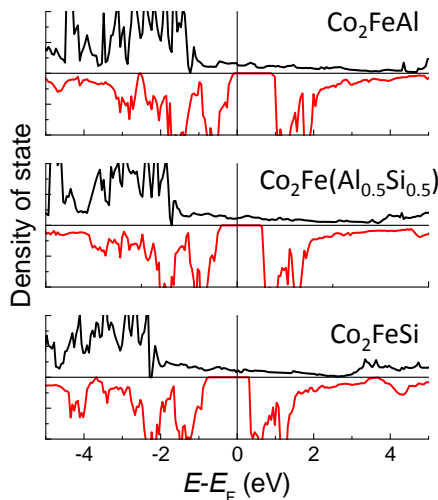


Criterion for alloy choice

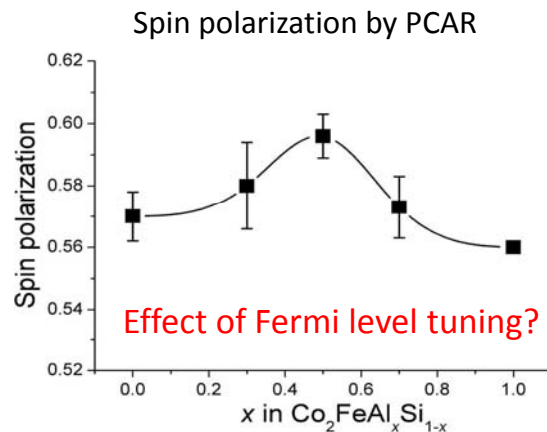
- Half-metallicity in calculation.
- High Curie temperature.
- High order-disorder temperature  
→ high driving force for ordering

# Quaternary Heusler alloy

- Balance different properties  
e.g. Co<sub>2</sub>(Cr,Fe)Al: Co<sub>2</sub>CrAl is HM but low T<sub>c</sub>,  
Co<sub>2</sub>FeAl has high T<sub>c</sub> but not HM
- Tune the Fermi level  
e.g. Co<sub>2</sub>Fe(Al,Si), Co<sub>2</sub>(Mn,Fe)Si



Systematic change of E<sub>F</sub>



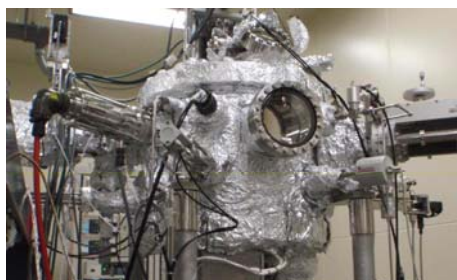
Fecher, J. Phys. D, 40, 1572 (2007); Tezuka, Appl. Phys. Lett. 89, 112514 (2006); Balke, Phys. Rev. B 89, 252508 (2006); Nakatani, J. Appl. Phys. 102, 033916 (2007); Gercsi, J. Phys. Cond. Mat. 19, 326216 (2007).

# Purpose & outline

1. Study physical aspects of Heusler based-CPP-GMR using a  $\text{Co}_2\text{Fe}(\text{Al}_{0.5}\text{Si}_{0.5})$  alloy.
  - Spin-dependent scattering
  - FM and spacer thickness for CPP-GMR
  - Annealing temperature for Heusler ordering
  - Spin torque noise
2. Search and develop new Heusler composition with large CPP-GMR output.

## Experimental methods

### UHV Magnetron sputtering



3 inch DC/RF cathode, 10 targets

- base  $1 \times 10^{-7}$  Pa
- RT deposition
- Deposited from alloy target
- Substrate heater

### Fully epitaxial on MgO(001)

- Single crystalline  $\rightarrow$  more intrinsic property
- Smooth interfaces

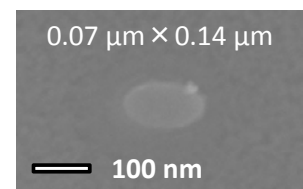
MgO//Cr(10 nm)/Ag(100 nm) underlayer

lead electrode

Roughness  $R_a = 1 \text{ \AA}$

### Device fabrication

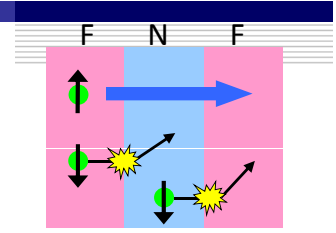
Photolithography + EB lithography



1 cm substrate

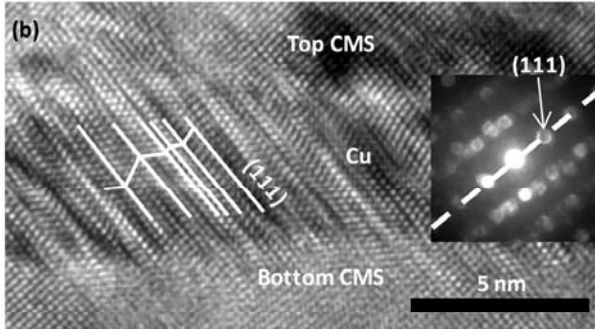
# Material for the spacer layer

- Good conductivity → spin diffusion length
- Small lattice mismatch
- Thermal stability for annealing
- Band matching



Large interfacial spin-dependent scattering

Twin in **Cu** spacer: Co<sub>2</sub>MnSi/Cu 001 epi



Large lattice mismatch with Heusler: 10%

Kodama, J. Appl. Phys. 105, 07E905 (2009)  
Furubayashi, JAP 107, 113917 (2010)

## Ag

- $\rho = 2 \mu\Omega\text{cm}$
- SDL > 100 nm
- Lattice mismatch < 2% with Heusler
- Immiscible with Co, Fe or Si either
- Band matching ??

Furubayashi et al. APL (2008)

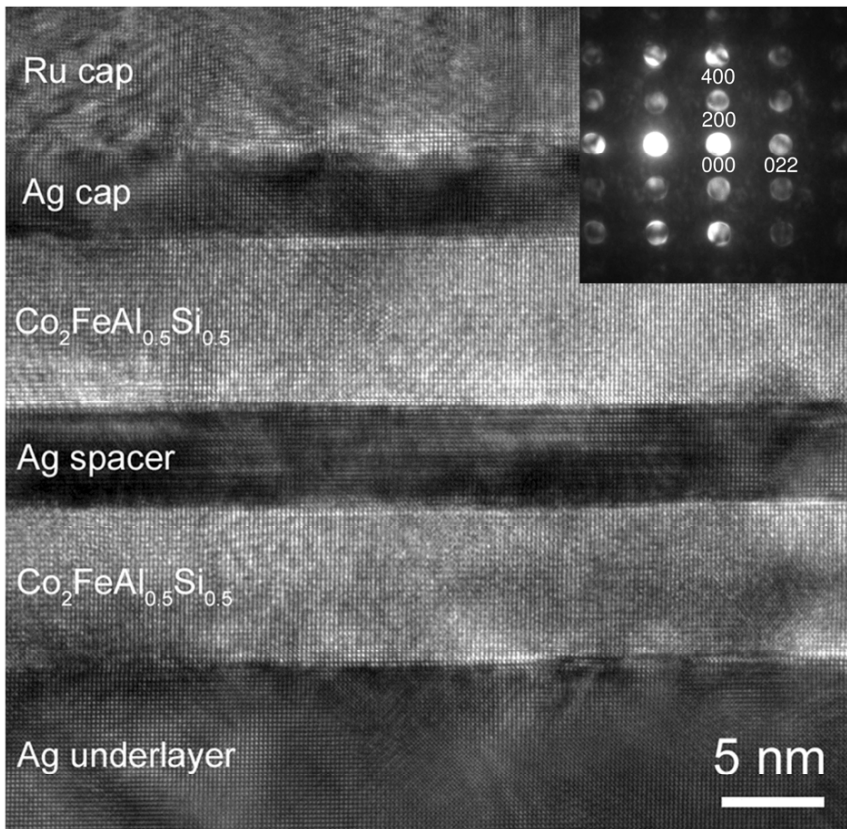
## NiAl B2 intermetallic compound

- $\Delta\text{RA}$  comparable to Ag
- Short spin diffusion length
- Strong ferromagnetic interlayer coupling

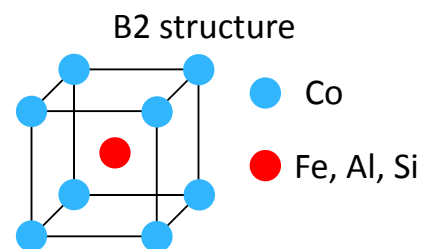
Nakatani, to be published

# Cross-sectional TEM

Annealed at 500 °C



- Coherent epitaxial at CAFS/Ag interface.
- Free from strain.
- Very flat and sharp interfaces.
- B2 ordered CFAS layers.

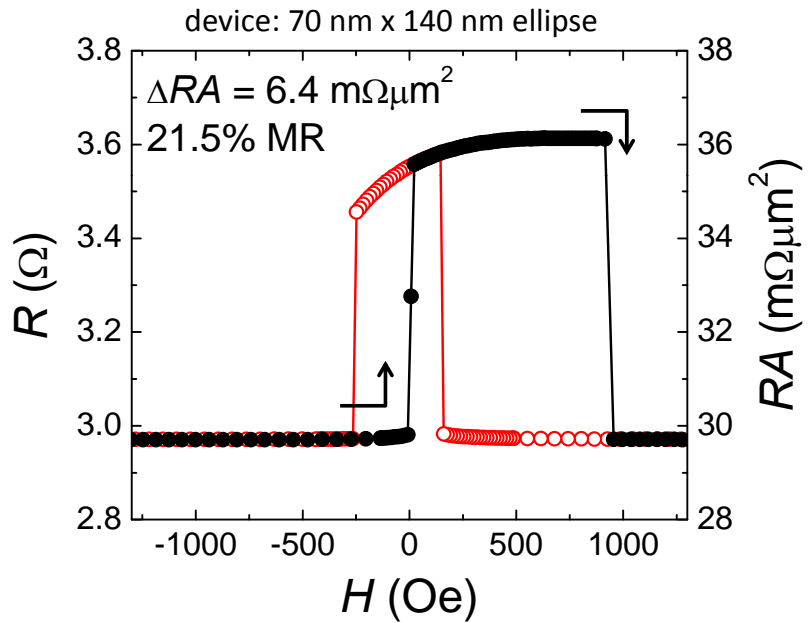


# Exchange biased spin-valve

Co<sub>2</sub>Fe(Al<sub>0.5</sub>Si<sub>0.5</sub>) Heusler (CFAS)



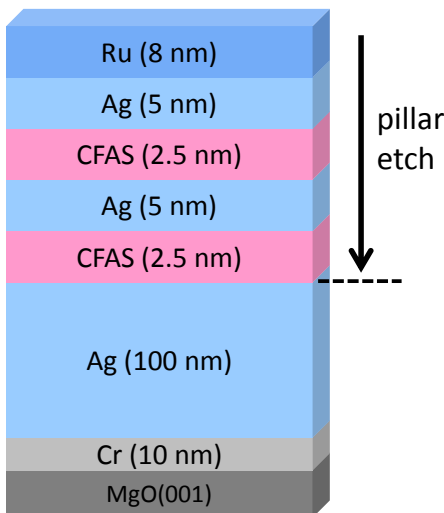
All deposition at RT  
 ↓  
 500 °C in 5 kOe



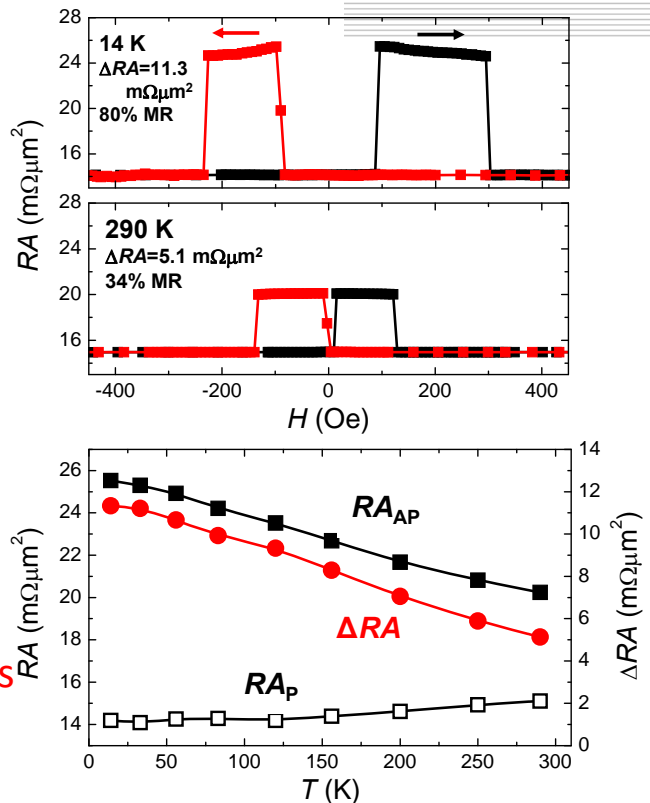
Large CPP-GMR outputs with CFAS/Ag  
 cf. poly-CoFe/Cu  $\Delta RA < 1 \text{ m}\Omega\mu\text{m}^2$   
 epi-CoFe/Ag  $\Delta RA = 2 \text{ m}\Omega\mu\text{m}^2$

# Pseudo spin-valve

annealed at 500 °C

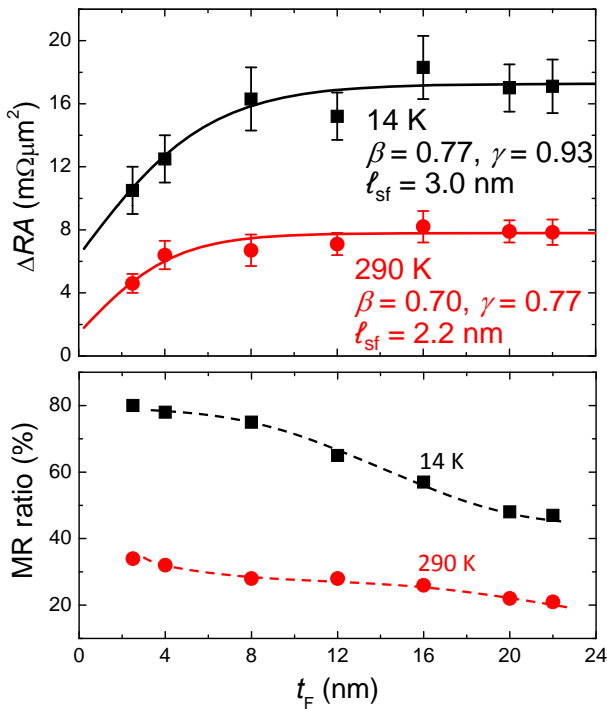


- AP state by dipole coupling
- Large  $\Delta RA$  with 2.5 nm FM layers
- Large temperature dependence of  $\Delta RA$ .



# Spin-dependent scattering

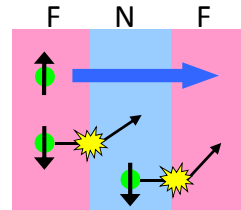
CFAS/Ag/CFAS PSV annealed at 500 C



- Large bulk scattering
- High resistivity  $70 \mu\Omega\text{cm}$
- Short spin diffusion length
- Large interfacial scattering



Large  $\Delta RA$  and  $\Delta R/R$  with thin FM layers.

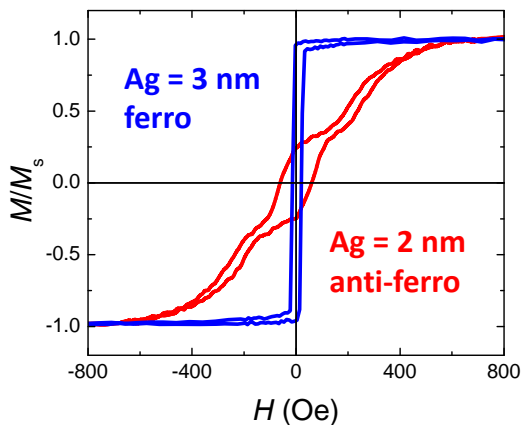
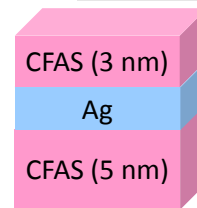
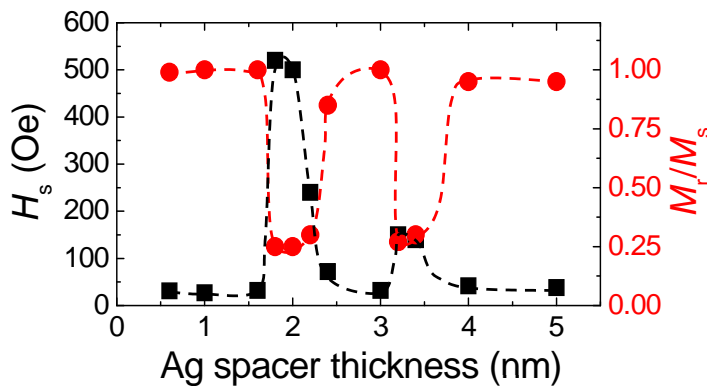


- Small interfacial R  
 $AR \sim 0.25 \text{ m}\Omega\mu\text{m}^2$
- Large  $\gamma$

→ Beneficial for ultrathin reader

Nakatani, Appl. Phys. Lett. 82, 094444 (2010), J. Appl. Phys. 109, 07B724 (2011)

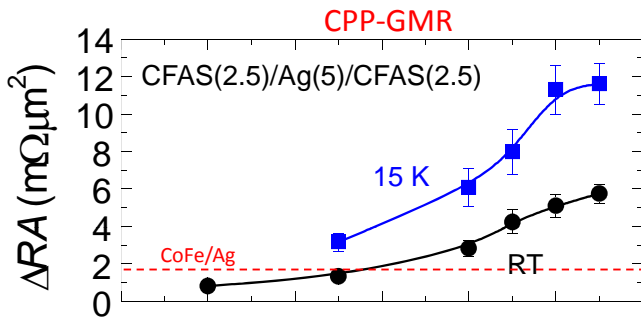
## Ag spacer thickness: How thin can we reduce?



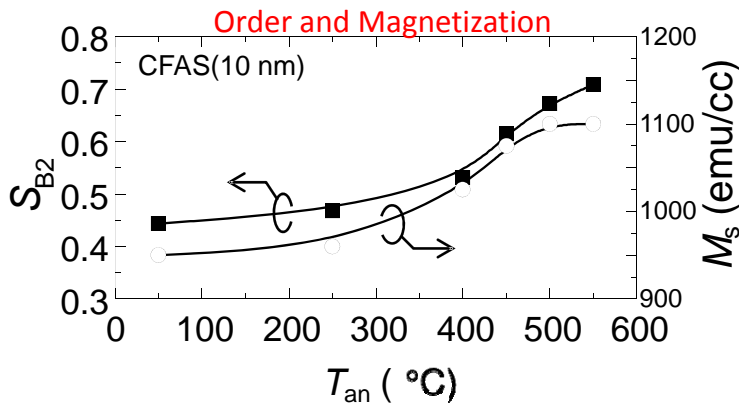
- Oscillatory AFM coupling
- Keep Ag > 4 nm for spin-valve.
- AFM coupled is usable for dual-free-layer sensor?

Presented at poster session of TMRC2011  
 To be presented at MMM by Nakatani (poster)

# Annealing temperature dependence



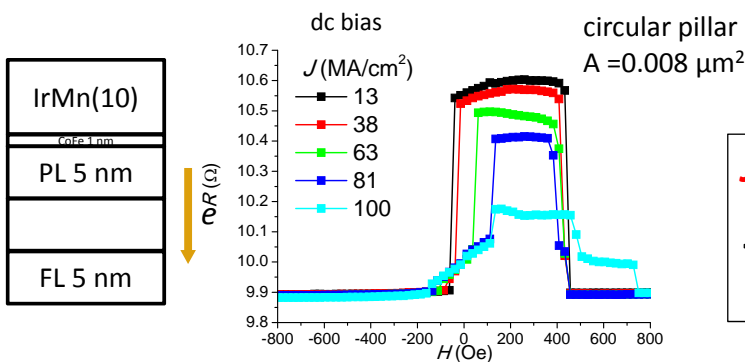
- $\Delta RA$  increases with  $T_{an}$ .
- Consistent with B2 order and  $M_s$ .
- $T_{an} < 300$  °C,  $\Delta RA$  lower than CoFe.



**Important to obtain highly ordered Heusler by low  $T_{an}$ .**

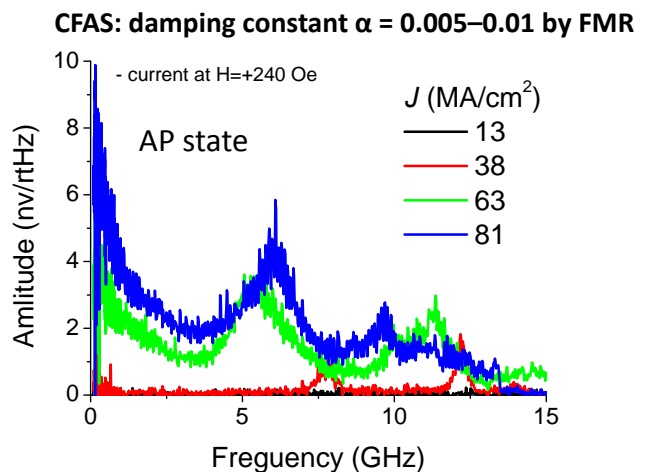
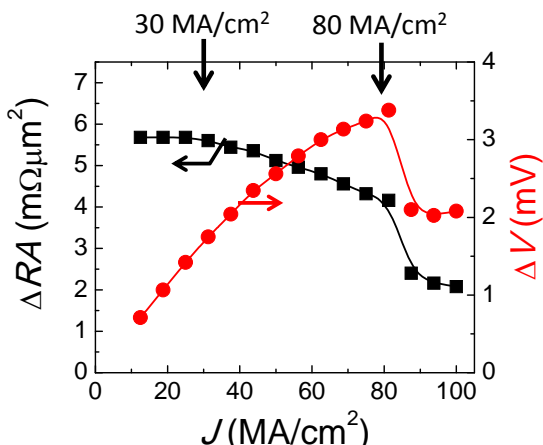
submitted to proceedings of TMRC2011

# Bias current density and spin torque noise



**$J_{critical} \sim 40 MA/cm^2$**

**→ Need to be improved.  
e.g. 100  $MA/cm^2$  for read head**



## Summary of $\text{Co}_2\text{Fe}(\text{Al}_{0.5}\text{Si}_{0.5})$ CPP-SV

### 001-epitaxial CFAS/Ag/CFAS SVs

- $\Delta RA = 6 \text{ m}\Omega\mu\text{m}^2$ ,  $\Delta R/R = 21\%$  in EBSV with 5 nm CFAS ( $T_{\text{an}} = 500 \text{ C}$ ).
- Large contribution of interfacial scattering.
  - Good band match at CFAS/Ag
  - Advantageous for ultrathin reader.
- Ag spacer for SV must  $> 4 \text{ nm}$  to decouple FL and PL.  
FM/NM/FM trilayer  $\sim 10 \text{ nm}$
- $\Delta RA$ : strong dependence on  $T_{\text{an}} \rightarrow T_{\text{an}} > 400 \text{ C}$  for B2 ordering.
- $J_{\text{critical}} = 40 \text{ MA/cm}^2$  for  $1/f$ -like spin-torque noise.

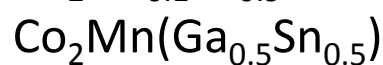
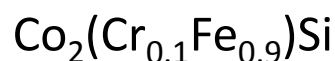
### Need Heusler materials with



- Larger  $\Delta RA$  and  $\Delta R/R$  with thin FM thickness.
- Lower annealing temperature for ordering.

## Purpose & outline

1. Study physical aspects of Heusler based-CPP-GMR using a  $\text{Co}_2\text{Fe}(\text{Al}_{0.5}\text{Si}_{0.5})$  alloy.
2. Search and development of new Heusler composition with large CPP-GMR output.



# Methods of alloy search

## Alloy synthesis

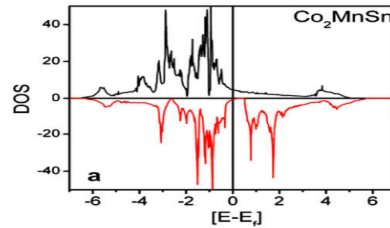
arc melting or induction melting



+ annealing

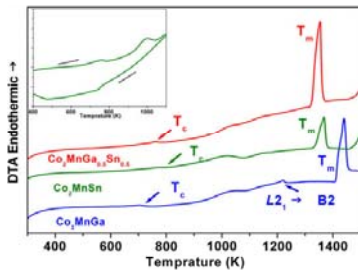
## DOS calculation

Commercial package: VASP



## Thermodynamically property

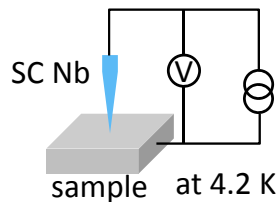
differential thermal analysis (DTA)



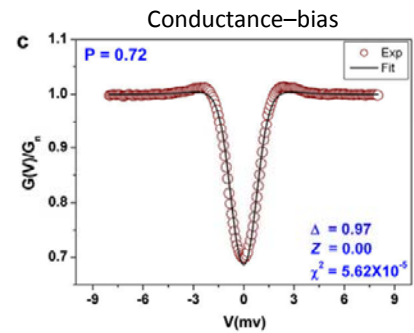
$T_c$ , order-disorder temperature, melting point

## Spin polarization

Point Contact Andreev Reflection (PCAR)



Lock-in amp  
Numerical  $dI/dV$

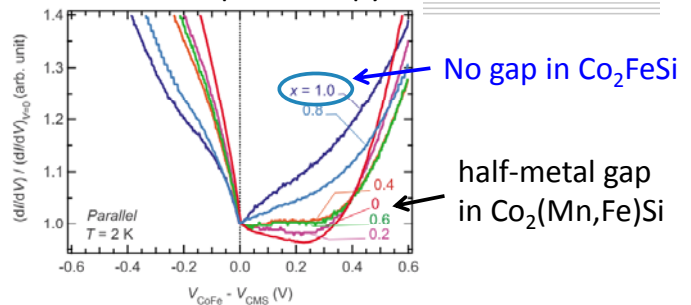


# Co<sub>2</sub>(Cr,Fe)Si alloy: effect of Cr doping

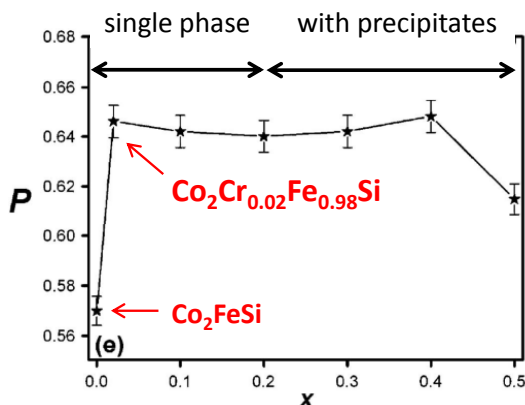
**Co<sub>2</sub>FeSi:**  $T_c = 827$  °C<sup>[1]</sup>  
 $T_{L2_1/B2} = 1145$  °C<sup>[2]</sup>  
 Predicted half-metal

[1] Wurmehl, Appl. Phys. Lett., 88, 032503 (2006).  
 [2] Umetsu, private communication.

## Tunnel spectroscopy



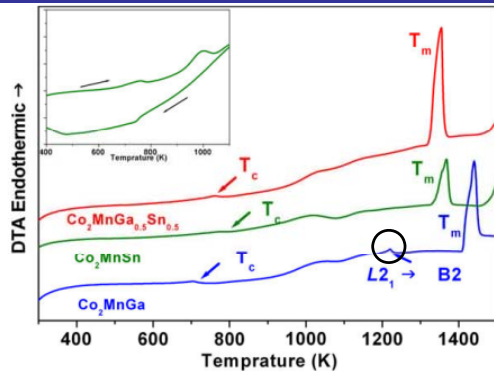
Kubota, Appl. Phys. Lett. 94, 122504 (2009).



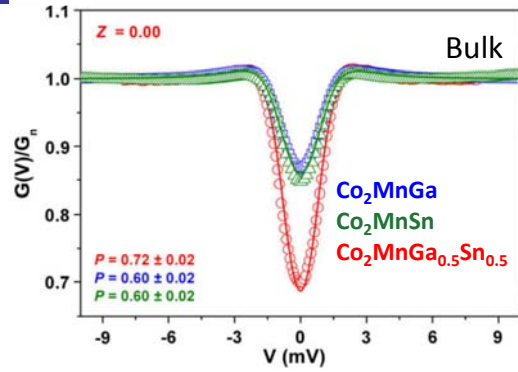
Karthik, J. Appl. Phys. 102, 043903 (2007).

- Enhancement of P by small Cr
- Improvement of ordering by Cr by XRD and Mössbauer

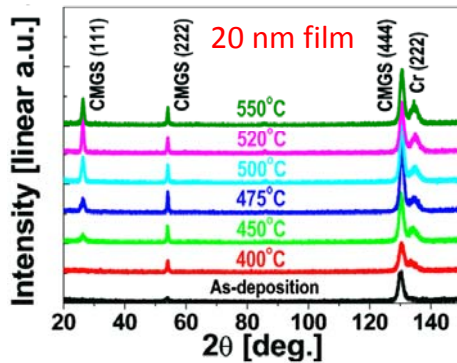
# Intermetallic $\text{Co}_2\text{MnGa}_{0.5}\text{Sn}_{0.5}$



$L_{21}$  is stable up to melting point  
 → high driving force of  $L_{21}$  order.



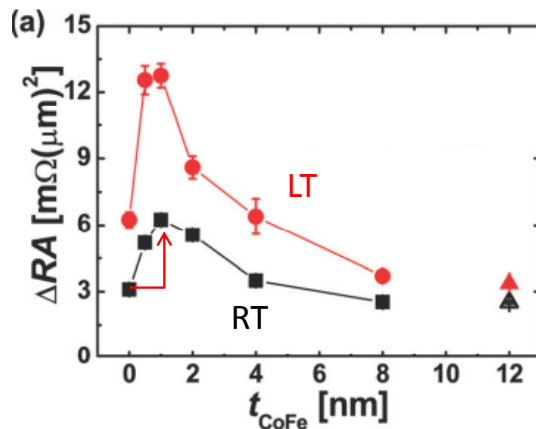
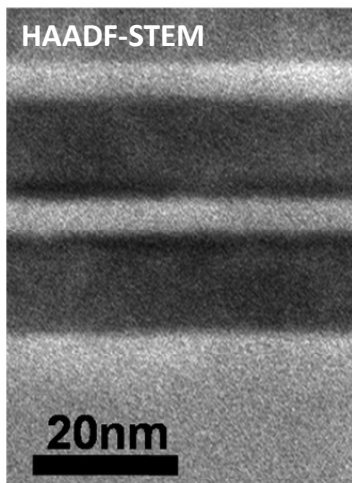
$P = 0.72$  for  $\text{Co}_2\text{MnGa}_{0.5}\text{Sn}_{0.5}$



$L_{21}$  order by  $>450^\circ\text{C}$

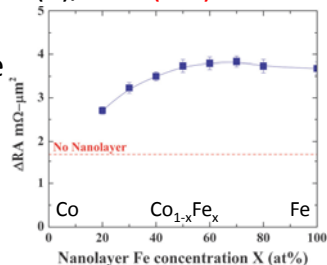
Varaprasad, Acta Mater. 57, 2702 (2009).  
 Hase, J. Appl. Phys. 108, 093916 (2010).

## Enhancement of CPP-GMR by inserting CoFe layers



Twofold enhancement of  $\Delta RA$  by 1 nm CoFe insertions.

Ref: RL(4)/CoFe(0.5)/Cu(4)/CoFe(0.5)  
 /FL(4)/CoFe(0.5)/cap  
 RL and FL =  $\text{Co}_2\text{MnGe}$



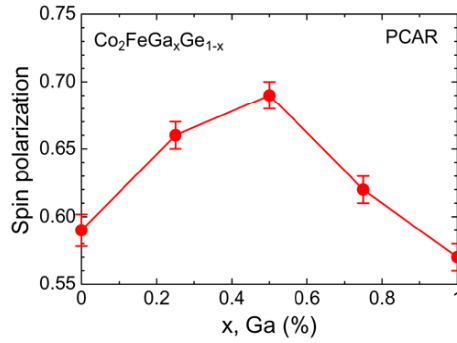
Carey, J. Appl. Phys.  
 109, 093912 (2011)

Practically useful to improve  $\Delta RA$   
 without increasing thickness much.

Hase, J. Appl. Phys. 109, 07E112 (2011)

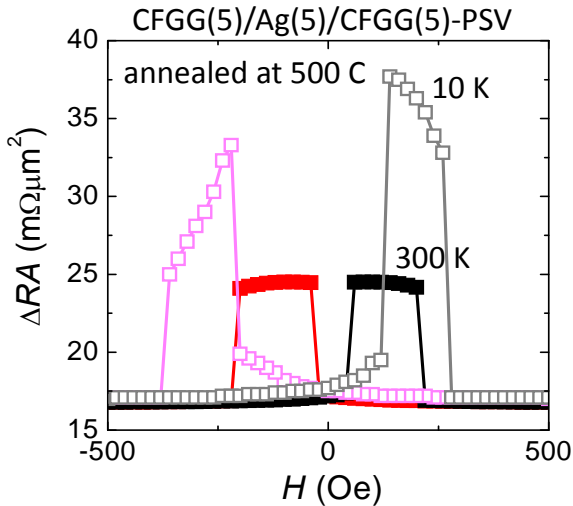
# Co<sub>2</sub>Fe(Ge<sub>0.5</sub>Ga<sub>0.5</sub>)

$T_c = 827 \text{ }^\circ\text{C}$   
 $T_{L2_1/B2} = 1027 \text{ }^\circ\text{C}$



$P = 0.69$

Co<sub>2</sub>Fe-based alloy  
 → high annealing possible  
 ~ 500 °C

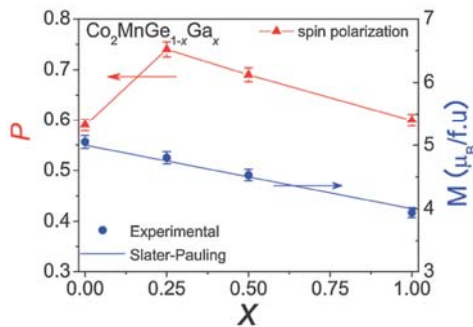


RT  $\Delta RA = 7.6 \text{ m}\Omega\mu\text{m}^2$   $\Delta R/R = 44\%$   
 10 K  $\Delta RA = 20 \text{ m}\Omega\mu\text{m}^2$   $\Delta R/R = 119\%$

Largest ever reported  
 due to large  $\beta = 0.77$  at RT.

Takahashi, Appl. Phys. Lett. 98, 072510 (2011).

# Co<sub>2</sub>Mn(Ge,Ga)

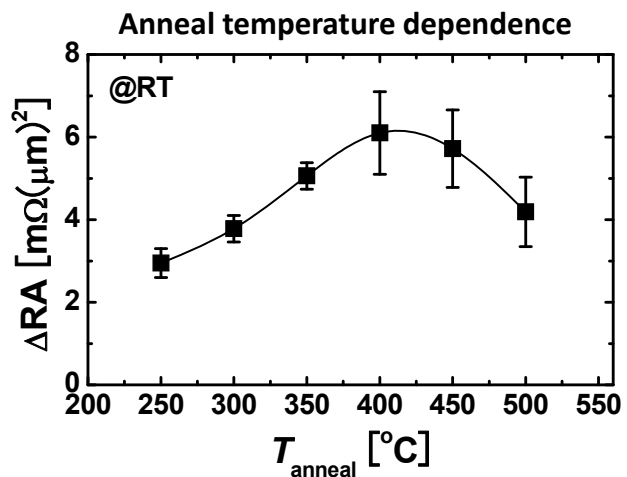
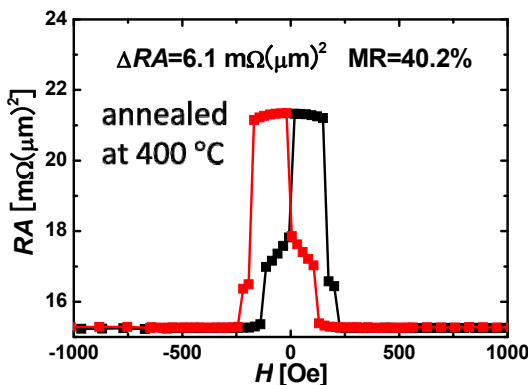


Bulk Co<sub>2</sub>Mn(Ge<sub>0.75</sub>Ga<sub>0.25</sub>)

$P = 74\%$  (highest ever!)

$T_c = 622 \text{ }^\circ\text{C}$

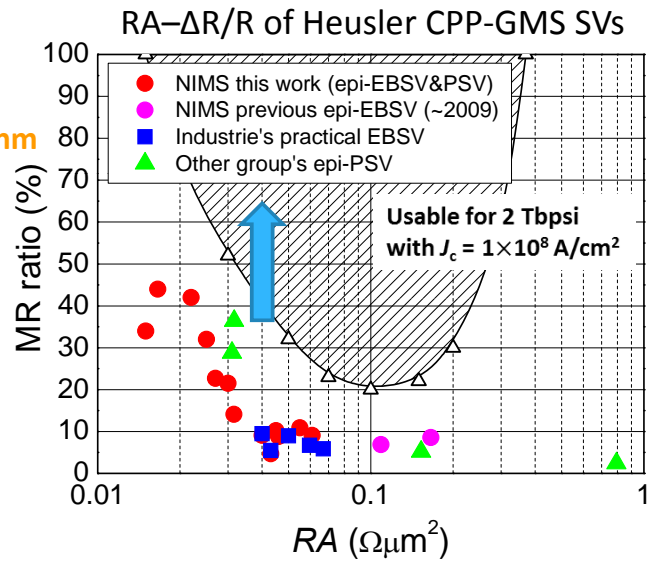
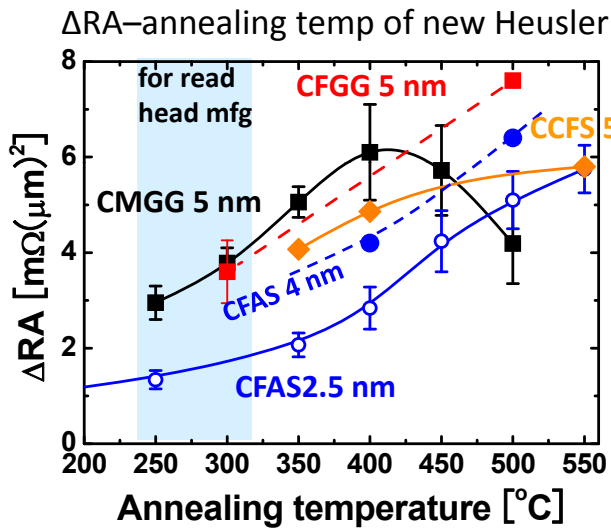
Intermetallic (no L<sub>2</sub><sub>1</sub>-B<sub>2</sub> transformation)



Large  $\Delta RA$  and  $\Delta R/R$  with low  $T_{\text{an}}$   
 → practically beneficial

To be presented at MMM by Naoki Hase (oral)

## Summary



CFGG =  $\text{Co}_2\text{Fe}(\text{Ge}_{0.5}\text{Ga}_{0.5})$   
 CMGG =  $\text{Co}_2\text{Mn}(\text{Ge}_{0.75}\text{Ga}_{0.25})$   
 CCFS =  $\text{Co}_2(\text{Cr}_{0.1}\text{Fe}_{0.9})\text{Si}$   
 CFAS =  $\text{Co}_2\text{Fe}(\text{Al}_{0.5}\text{Si}_{0.5})$

Further improvement of  $\beta$  and  $\gamma$  (also,  $\rho_F$  and  $AR_{\text{interface}}$ ) will enhance  $\Delta RA$  with low RA.

## Future direction

- Further search of new Heusler composition
  - High spin polarization
  - Ordering by low annealing temperature
- Spacer material with better band matching
- Fundamental questions:
  - Origin of temperature dependence
  - Mechanism of enhancement of  $\Delta RA$  by CoFe insertion
  - Role of interfacial structure at FM/NM
  - Effect of crystalline orientation
- Poly-crystalline CPP-GMR films on Si/SiO<sub>2</sub> substrate
- Alternative reader structure
  - Dual-free-layer (scissors-type trilayer)
  - Non-local spin-valve