The 6th IEEE Magnetics Society Summer School Program
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Advanced Spintronic Materials for Generation and Control of Spin Current

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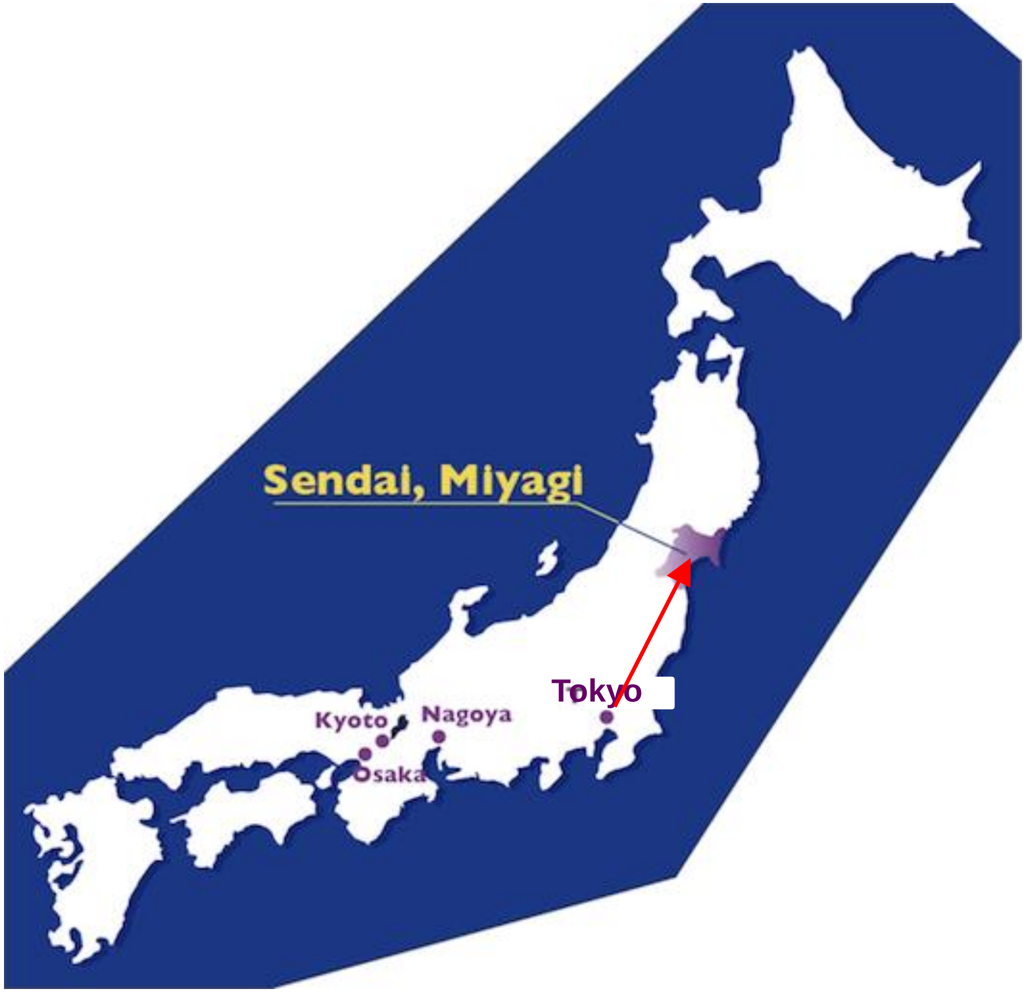
- **IEEE Magnetics Society Home Page:** ***www.ieeemagnetics.org***
  - 3000 full members
  - 300 student members

- **The Society**
  - Conference organization (INTERMAG, MMM, TMRC, etc.)
  - Student support for conferences
  - Large conference discounts for members
  - Graduate Student Summer Schools
  - Local chapter activities
  - Distinguished lectures

- ***IEEE Transactions on Magnetics***
  - ~2000 peer reviewed pages each year
  - Electronic access to all *IEEE Transactions on Magnetics* papers

- **Online applications for IEEE membership:** ***www.ieee.org/join***
  - 360,000 members
  - IEEE student membership IEEE full membership

Where is Tohoku University / Sendai?



Sendai

1 million population

350 km north from Tokyo

~2 h ride by “Shinkansen”

super-express

Tohoku University

Founded in 1907

3,000 Research Staffs

16,000 Students

10 Faculties

18 Graduate Schools

6 Research Institutes

KIN *KEN*

東北大学・金属材料研究所

Institute for Materials Research (IMR)
Tohoku University

Founded in 1907

Founded in 1916

We will have a centennial anniversary in 2016!

Honda Memorial Hall

Kotaro Honda
1st Director
KS magnet (1917)

120 Research Staffs
200 Students
27 Laboratories
9 Research Centers

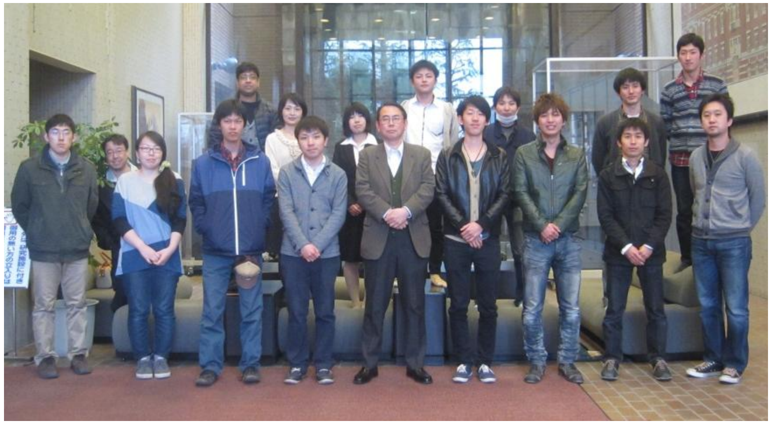
Magnetic Materials Laboratory (2012-2013)

Lab members

Professor	Koki Takanashi
Assoc. Prof.	Masaki Mizuguchi
Assist. Prof.	Yuya Sakuraba (~March 2013)
	Takeshi Seki
	Takahide Kubota (April 2013~)
Post-doc.	Bosu Subrojati (Bangladesh)
	Takayuki Kojima
	Hitomi Yako
DC student	Wei-Nan Zhou (China)

+ 8 MC students

FY 2012



FY 2013

Collaborators

Seiji Mitani (NIMS, Tsukuba)

Toshiyuki Shima (Tohoku-gakuin Univ.)

Sadamichi Maekawa, Eiji Saitoh, Saburo Takahashi (JAEA / IMR, Tohoku Univ.)

Masafumi Shirai (RIEC, Tohoku Univ.), Shigemi Mizukami (WPI, Tohoku Univ.)

Yasuo Ando, Junsaku Nitta (Faculty of Engng., Tohoku Univ.)

Outline

1. Introduction

   What is spin current?

   Relationship between spin current and spintronics

   Historical background: GMR

2. Recent progress in research on pure spin current

   Spin Hall effect / spin pumping / spin Seebeck effect

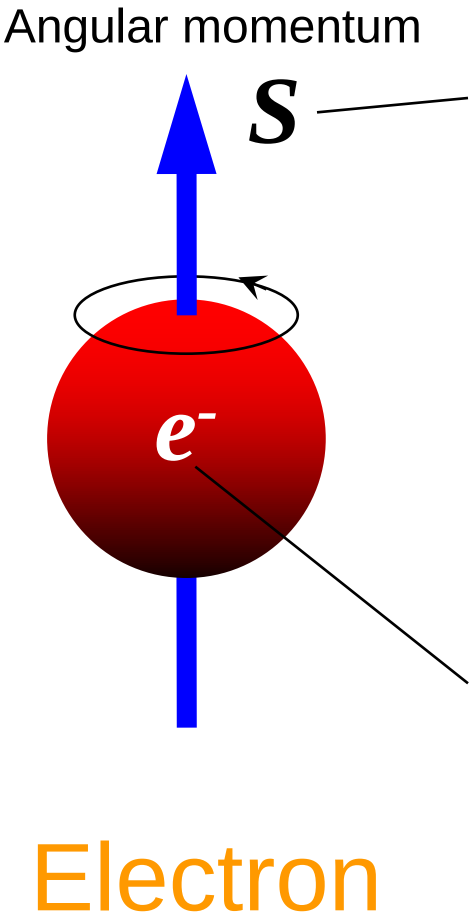
3. Topics of materials for spintronics

   Highly spin-polarized: half-metallic Heusler alloys

   Perpendicularly spin-polarized: high magnetic anisotropy $L1_0$-ordered alloys

4. Summary

What is spin current ?



not conserved

Spin → **Spin current**

Origin of magnetism

A concept that has attracted much attention in recent years

$$J_s = J_\uparrow - J_\downarrow$$

conserved

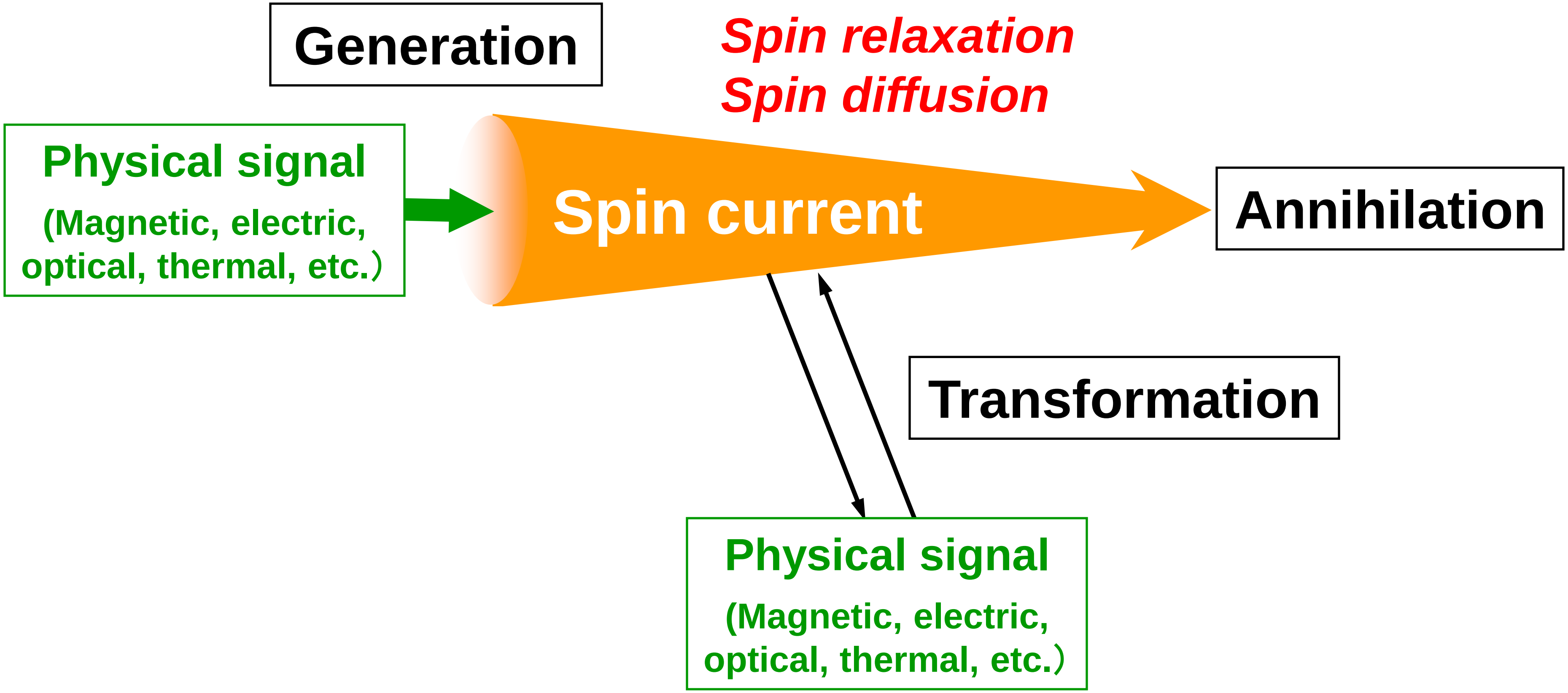
Charge → Electric current

Origin of electricity

A lot of studies since the 18th C.
Indispensable in daily life

$$J_e = J_\uparrow + J_\downarrow$$

What is spin current ?



Generation

Spin relaxation
Spin diffusion

Physical signal
(Magnetic, electric, optical, thermal, etc.）

Spin current

Annihilation

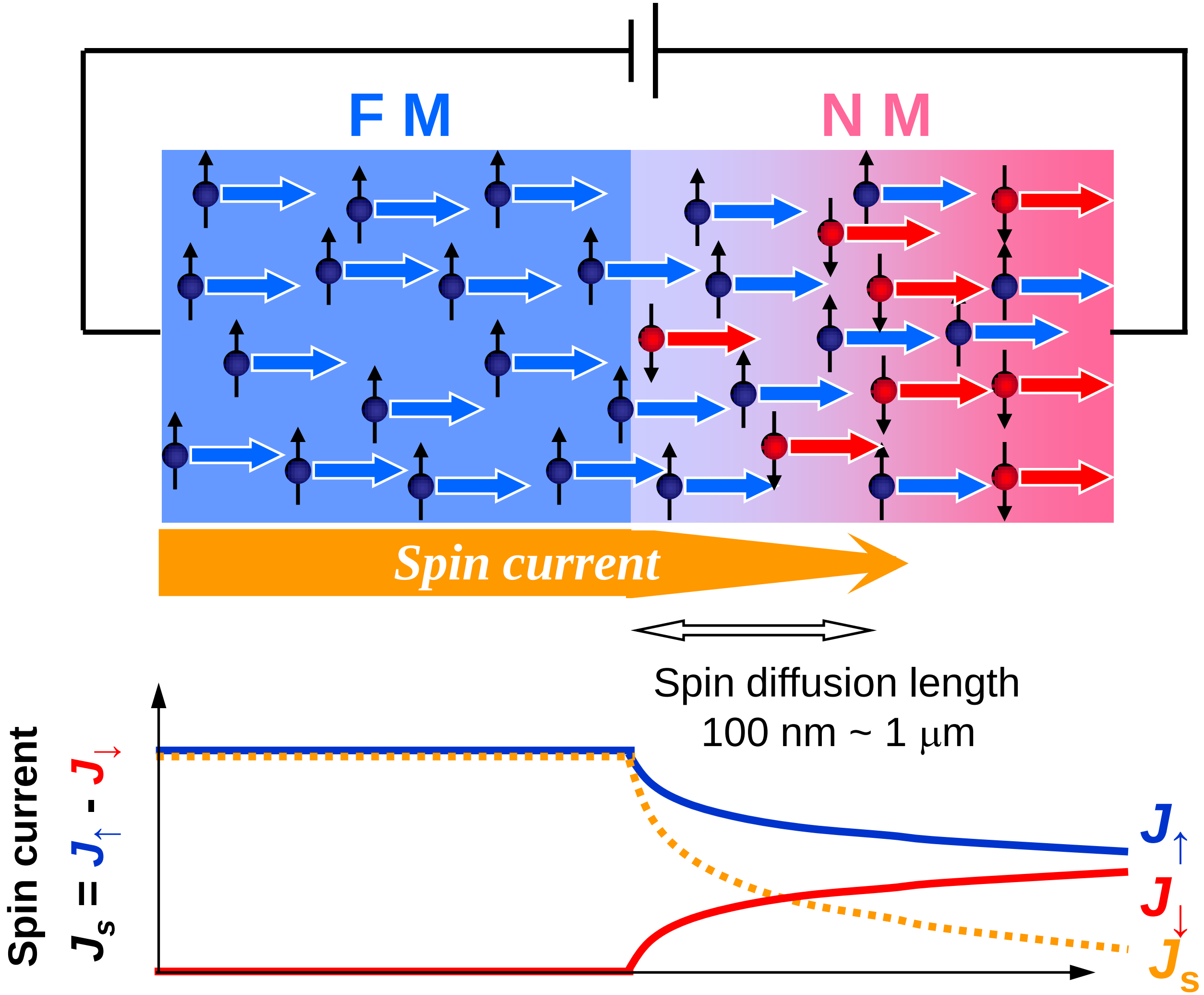
Transformation

Physical signal
(Magnetic, electric, optical, thermal, etc.）

Example of spin current -1

- With electric current

  Electrical *spin injection* from ferromagnetic material (FM) into nonmagnetic material (NM)



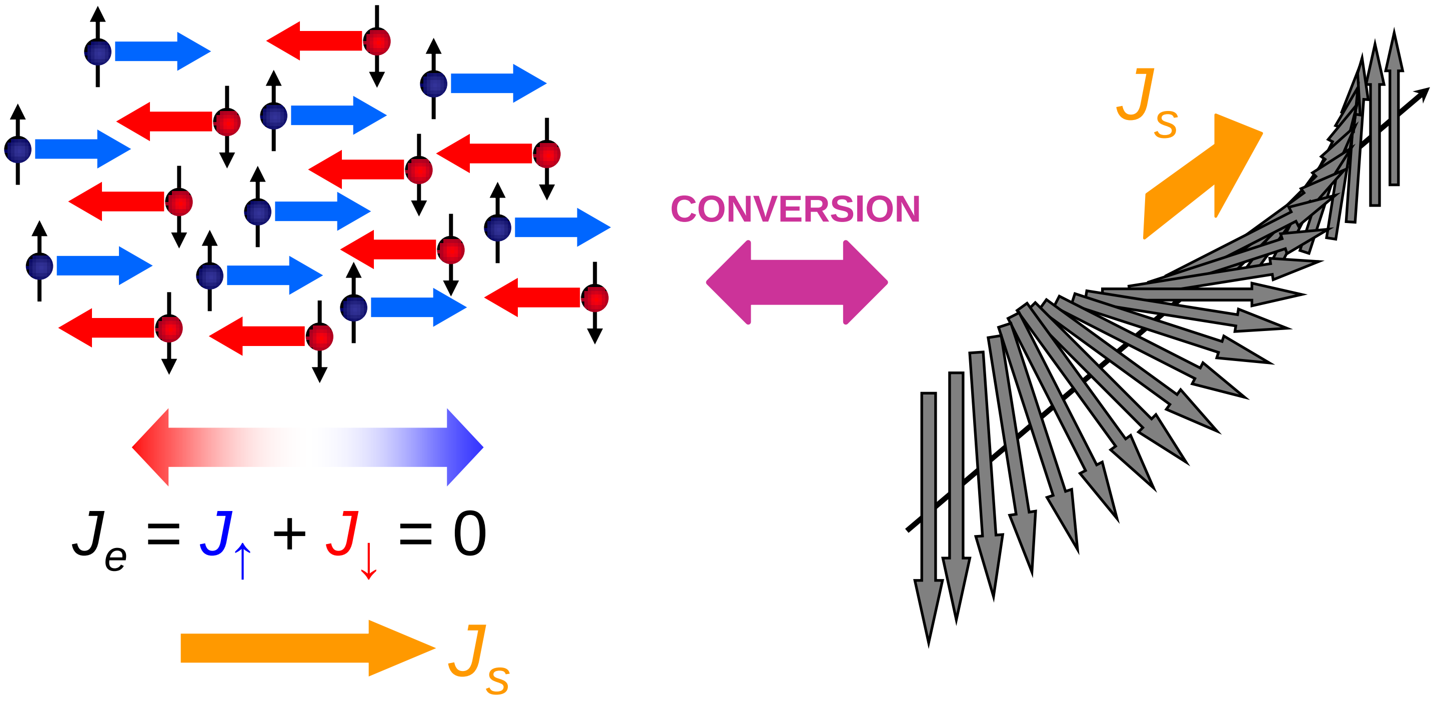
Spin diffusion length
100 nm ~ 1 μm

Example of spin current -2

- Without electric current (pure spin current)

With electron motion

Without electron motion



$$J_e = J_\uparrow + J_\downarrow = 0$$

Non-local spin injection
Spin Hall effect
Spin pumping
Spin Seebeck effect, etc.

Spin waves
(magnon spin current)

Feature of spin current

For electric current

Conductor
(metal/semiconductor)

Insulator

≠

For spin current

Conductor

Insulator

Spin current may flow in an electric insulator.

What is spintronics ?

Electron

Charge ＋ Spin

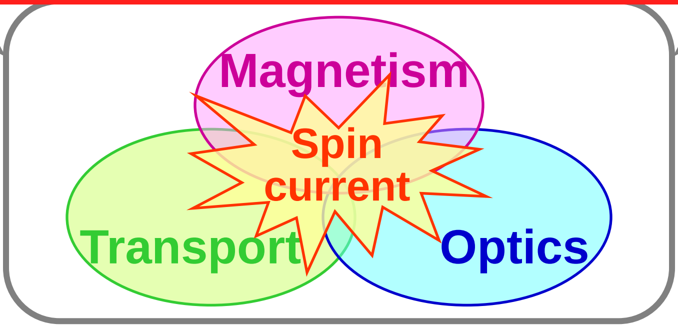
ELECTRONICS

Control of transport and optical properties (*s* & *p* electrons)

MAGNETICS

Control of magnetization (*d* & *f* electrons)

NANOTECHNOLOGY



Control of magnetization by electric and optical signals

Control of electric and optical signals by magnetization

Phenomena

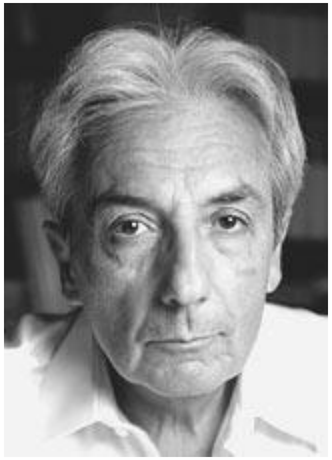
- Giant magnetoresistance (GMR)
- Tunnel magnetoresistance (TMR)
- Spin injection / accumulation
- Spin transfer phenomena
- Carrier or photo-induced magnetism

Devices

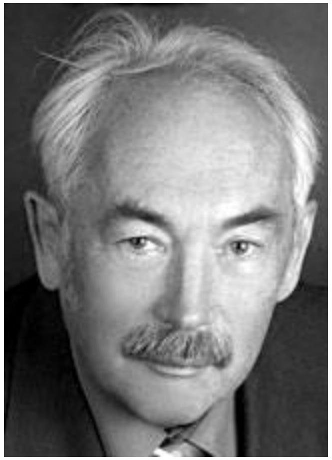
- GMR/TMR heads
- Magnetic sensors
- Magnetic random access memories (MRAM)
- Spin switches / transistors
- Spin logic circuits

Nobel prize in physics 2007

Albert Fert
(France)



Peter Grünberg
(Germany)



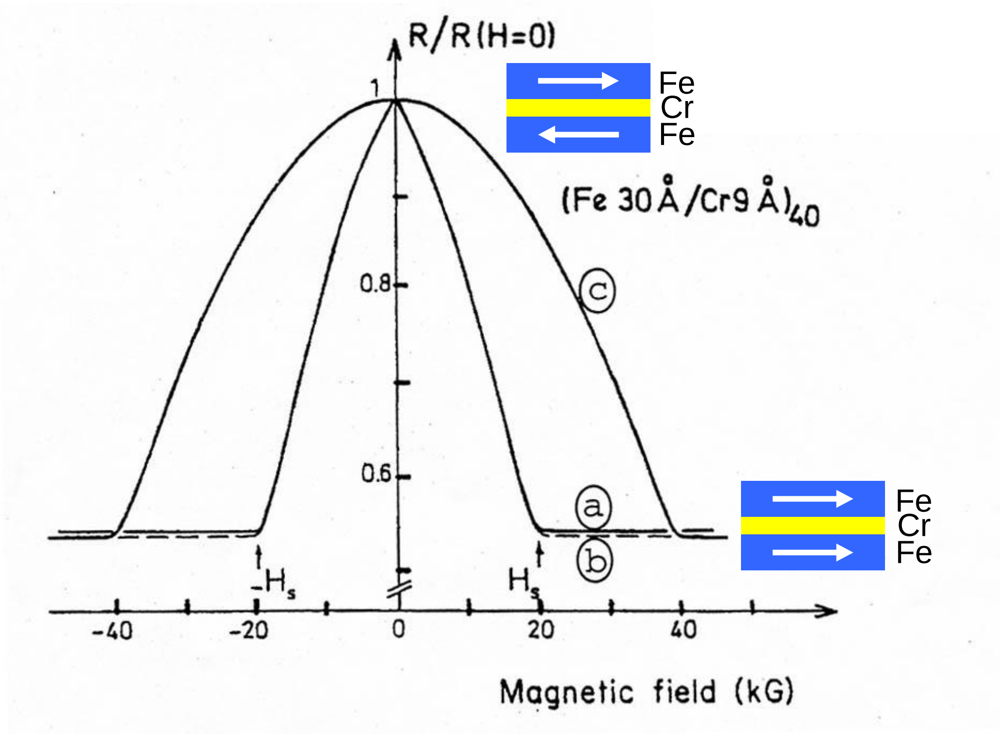
Discovery of GMR

↓

Remarkable enhancement of recording density of HDD

“The first major application of nanotechnology”

Development of spintronics



M. N. Baibich *et al*., Phys. Rev. Lett., **61** (1988) 2472.

Giant Magnetoresistance (GMR)
Large difference in electrical resistance between parallel and antiparallel alignments of magnetization.
（Spin-dependent transport）

Principle of spin-valve GMR head

***Nobel week** in Stockholm, December 2007*

On the Noble lecture (Dec. 8, 2007)

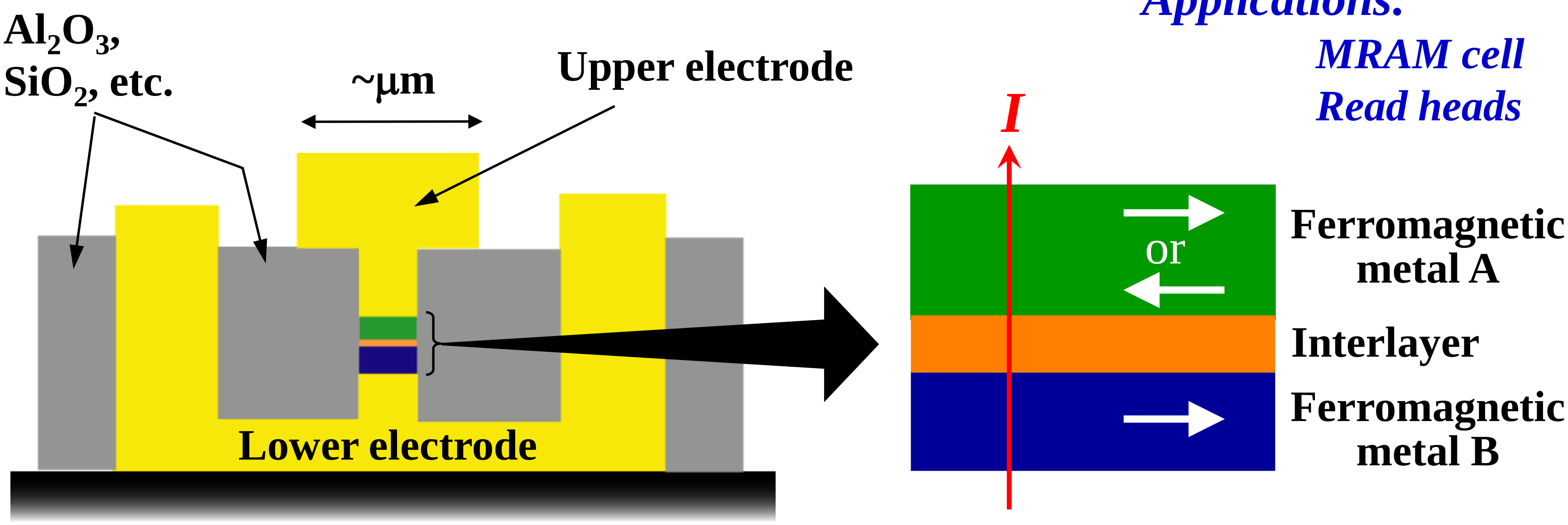
At the award ceremony
(Dec. 10, 2007)

At the Reception by the Royal Swedish Academy
(Dec. 7, 2007)

At the Nobel banquet
(Dec. 10, 2007)

Typical device structures in spintronics

1. CPP (Current-Perpendicular-to-Plane) type



Applications:
MRAM cell
Read heads

Interlayer = Insulator: Tunnel magnetoresistance（TMR）

Metal：Giant magnetoresistance（CPP-GMR）

Magnitude of MR : $\frac{\Delta R}{R} \propto P_A \cdot P_B$

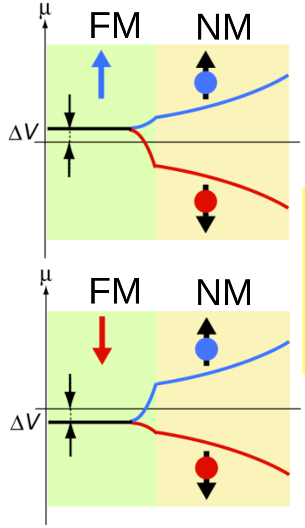
$P_{A(B)}$: spin polarization of conduction electrons in A (B)

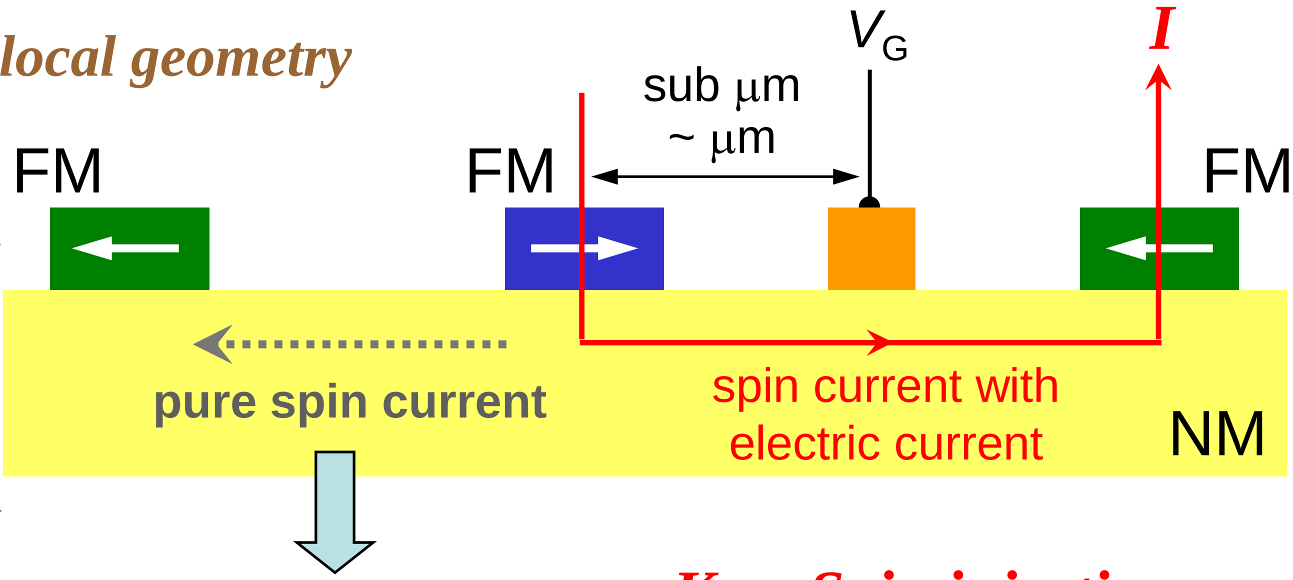
Typical device structures in spintronics

2. Lateral structure type

Applications: spin transistor, etc.

Non-local geometry





$$J_e = J_\uparrow + J_\downarrow = 0$$

$$J_s = J_\uparrow - J_\downarrow \neq 0$$

Key: Spin injection

from ferromagnetic metal (FM)
to nonmagnetic metal (NM)

Spin relaxation

Research on *pure* spin current

Generation of *pure* spin current

- Non-local spin injection (electric current → spin current)
- Spin Hall effect (electric current → spin current)

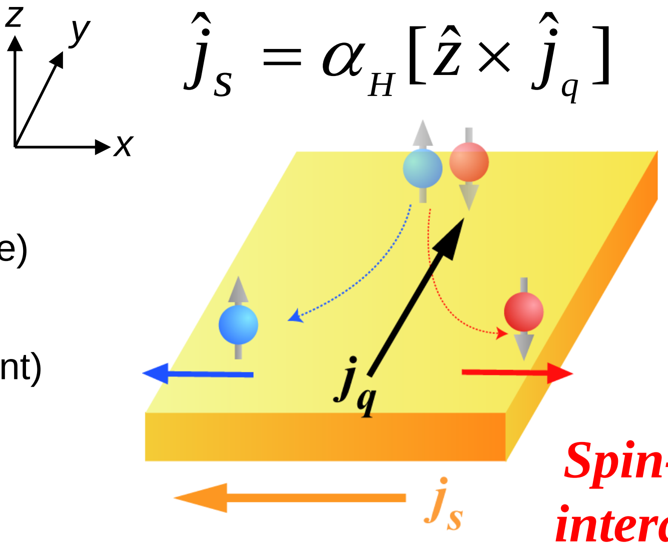
J_q: Charge current
(positive charge)

J_s: Spin current
(Flow of moment)

α_H: Spin Hall angle
($=\sigma_{SH}/\sigma$)

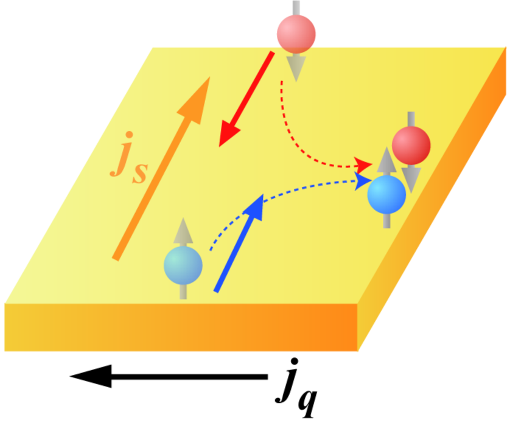
Direct spin Hall effect

$$\hat{j}_s = \alpha_H [\hat{z} \times \hat{j}_q]$$



Inverse spin Hall effect

$$\hat{j}_q = \alpha_H [\hat{z} \times \hat{j}_s]$$

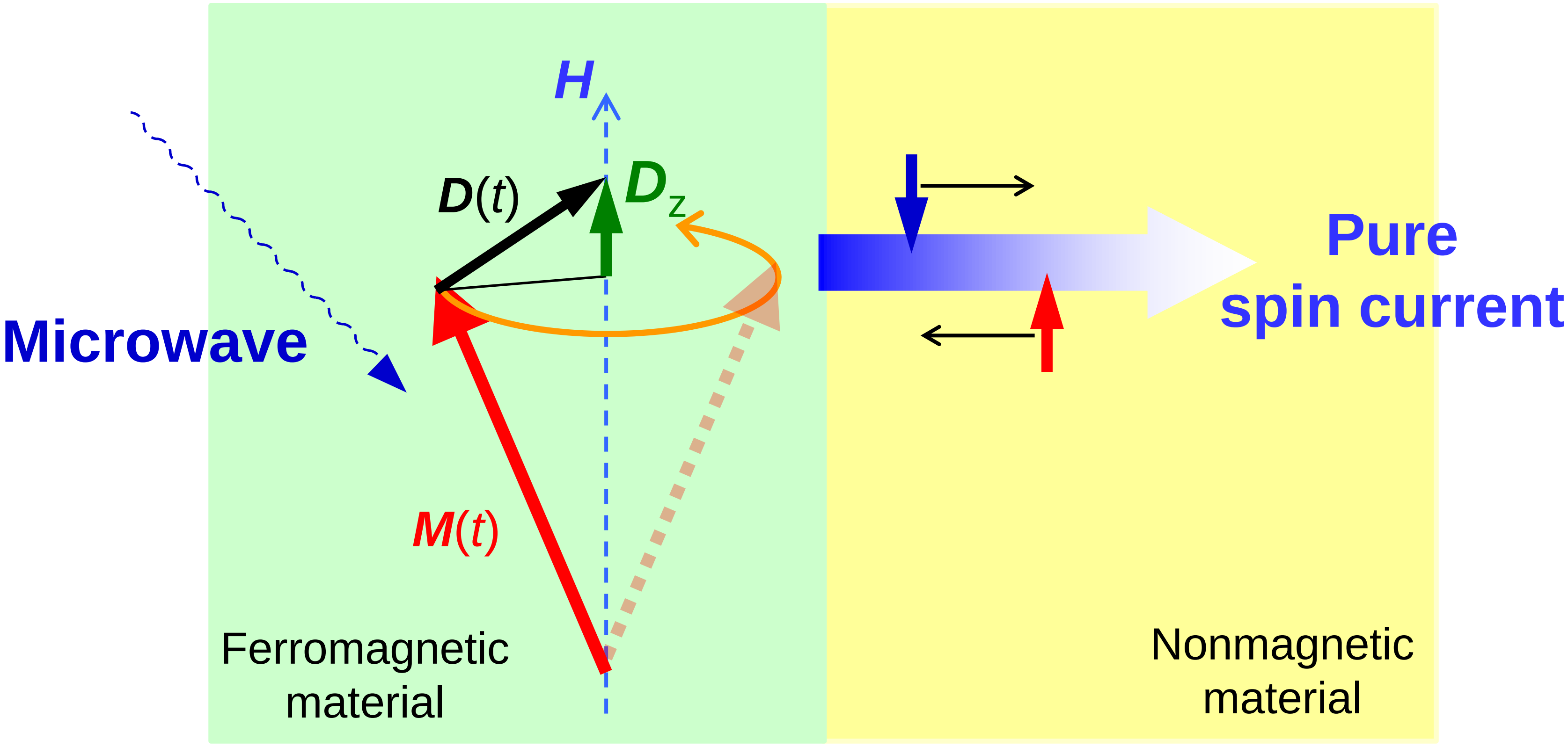


Spin-orbit interaction

Research on *pure* spin current

Generation of *pure* spin current

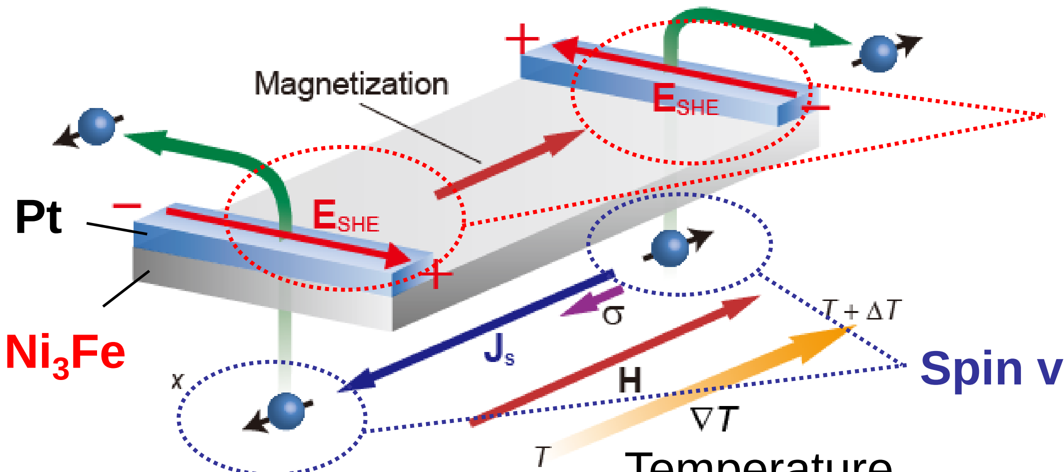
- Non-local spin injection (electric current → spin current)
- Spin Hall effect (electric current → spin current)
- Spin pumping (electromagnetic wave → spin current)



Research on *pure* spin current

Generation of *pure* spin current

- Non-local spin injection (electric current → spin current)
- Spin Hall effect (electric current → spin current)
- Spin pumping (electromagnetic wave → spin current)
- Spin Seebeck effect (heat current → spin current)



Temperature gradient

Electromotive force of opposite sign

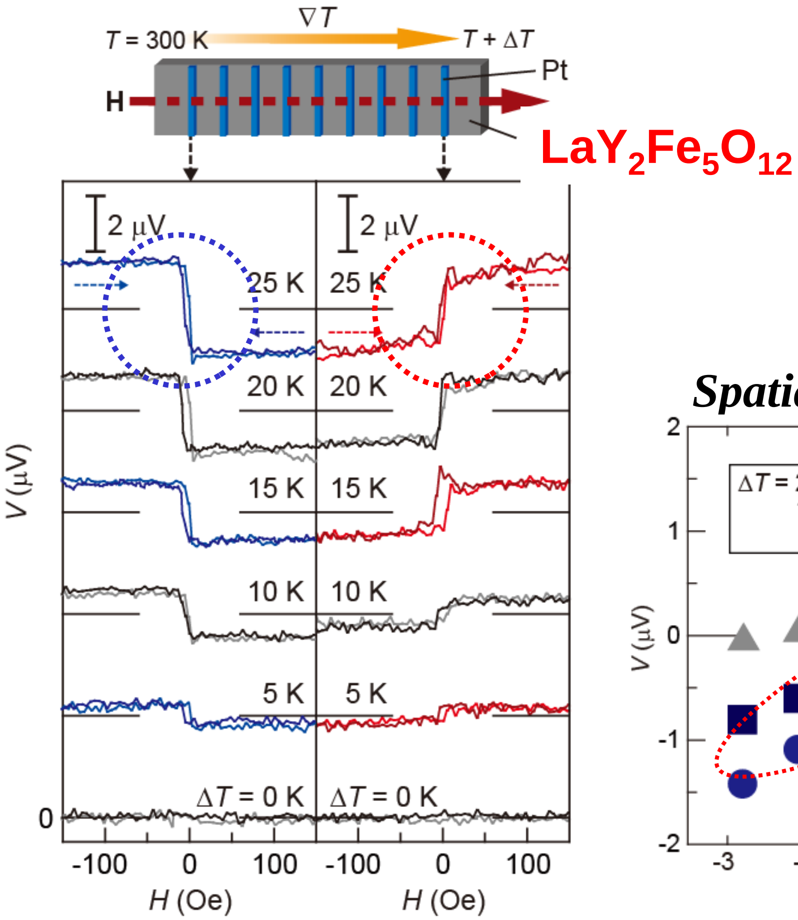
Spin voltage of opposite sign

K. Uchida et al., Nature, 455, 778 (2008).

(*Prof. E. Saito's group*)

Spin Seebeck insulator by E. Saitoh’s group

K. Uchida et al., Nature Mater., 9, 894 (2010).

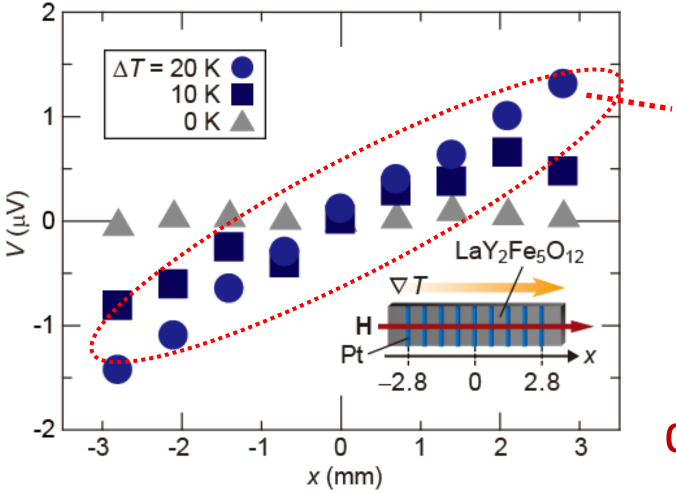


Temperature difference dependence
of spin voltage

Spin Seebeck effect appears even in a magnetic insulator

Magnon spin current

Spatial distribution



Similar behavior to Ni_3Fe

Opposite sign of spin voltage at the edges
+
distribution in mm scale

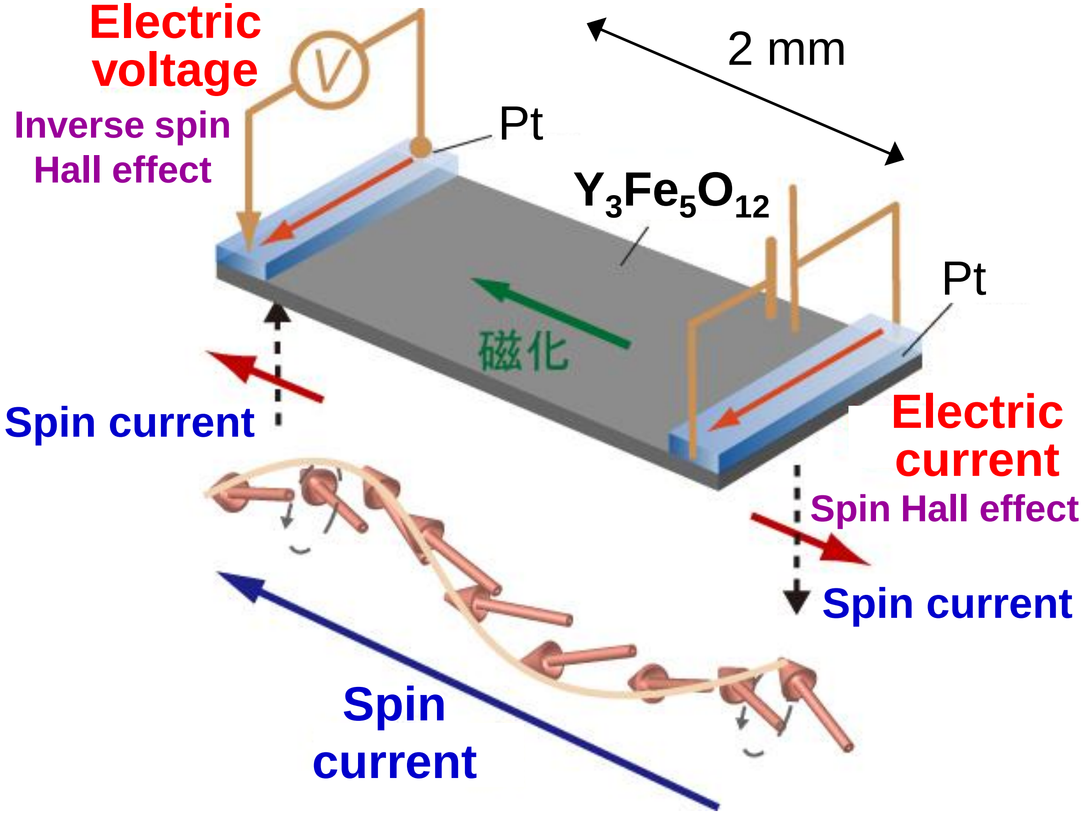
Development of Spin Caloritronics
Application to Energy Harvesting

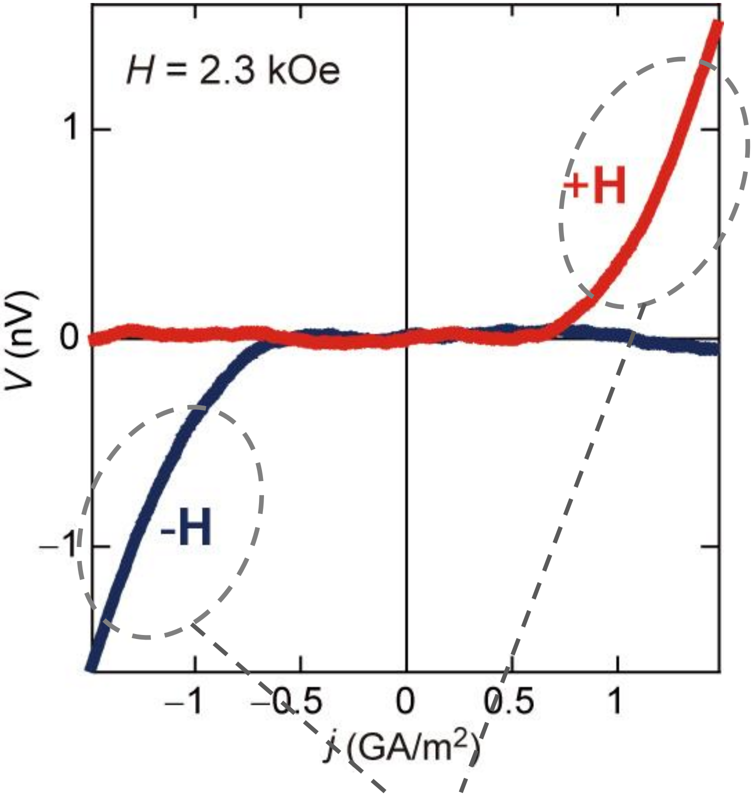
Transmission of pure spin current

by Saitoh's group in collaboration with Maekawa and Takanashi groups

Y. Kajiwara et al., Nature, 464 (2010) 262.

Transmission of pure spin current; metal→insulator→metal





Transmitted signal

Transmission of electric signal through spin current in a magnetic insulator

Keywords for spintronics

Spin polarization

Spin injection

Spin relaxation

⬇

Efficient generation and precise control of spin current

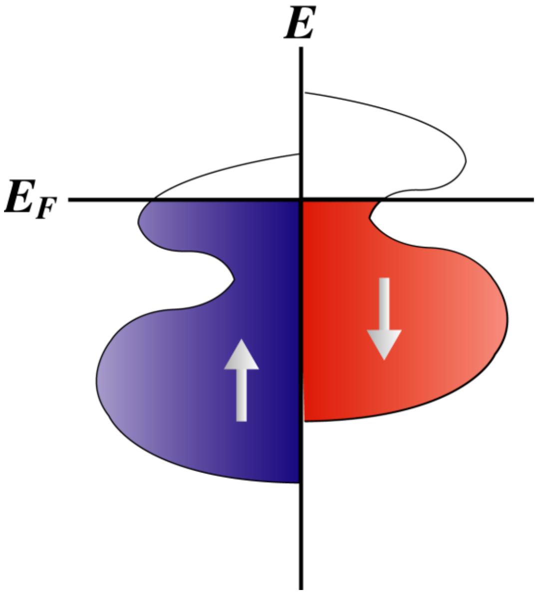
Topics of materials for spintronics

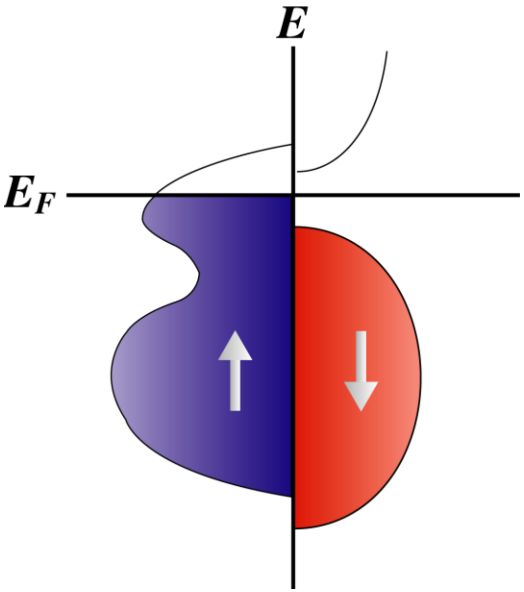
- **Spin polarization**
  - Highly spin polarized (half metallic)
    - Heusler alloys (Co_2MnSi, $Co_2Fe(Al,Si)$, etc.)
  - Perpendicularly spin polarized
    - High magnetic anisotropy: $L1_0$ ordered alloys (FePt, etc.)
- **Spin injection**
  - Magnetic metal / semiconductor junction
    - CoFe/Si, Fe/GaAs, etc.
  - Metal / magnetic insulator junction
    - Pt / $Y_3Fe_5O_{12}$, etc.
- **Spin relaxation**
  - Nanoparticles → ***size effect***
  - Molecular / carbon-based materials → ***weak LS coupling***
  - Magnetic insulator → ***low magnetization damping***

Highly spin-polarized materials:

half-metallic Heusler alloys

Half-metals





Spin polarization of conduction electrons

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

- Conventional 3d ferromagnetic metal and alloys: Fe, Co, Ni, NiFe, ･･･

  P = 0.4~0.5 typically

- **Half metals**

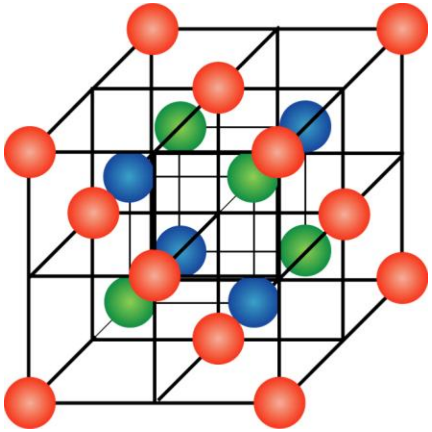
  ***P* = 1**

  Heusler alloys:
  NiMnSb, Co_2MnSi, Co_2MnAl, etc.

  Transition metal oxides:
  CrO_2, Fe_3O_4, LSMO, etc.

→ **Efficient spin injection**
High performance of spintronics devices

Half metallic Heusler alloys

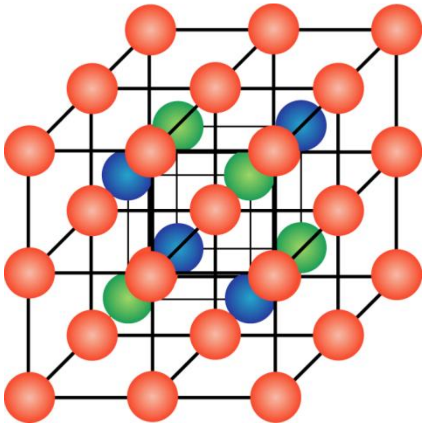


half-Heusler

XYZ

(NiMnSb etc.)

$C1_b$ structure



full-Heusler

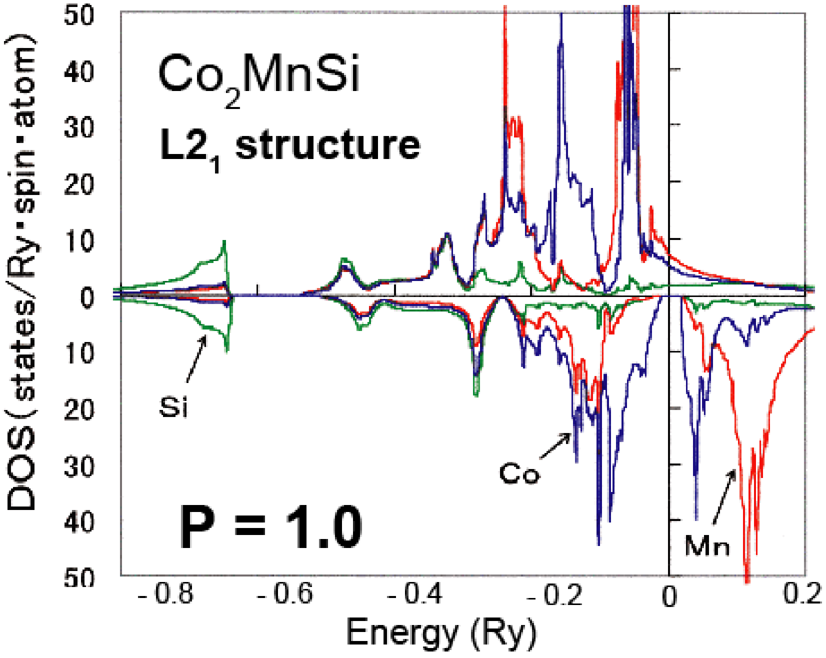
X_2YZ

(Co_2MnSi, Co_2MnGe etc.)

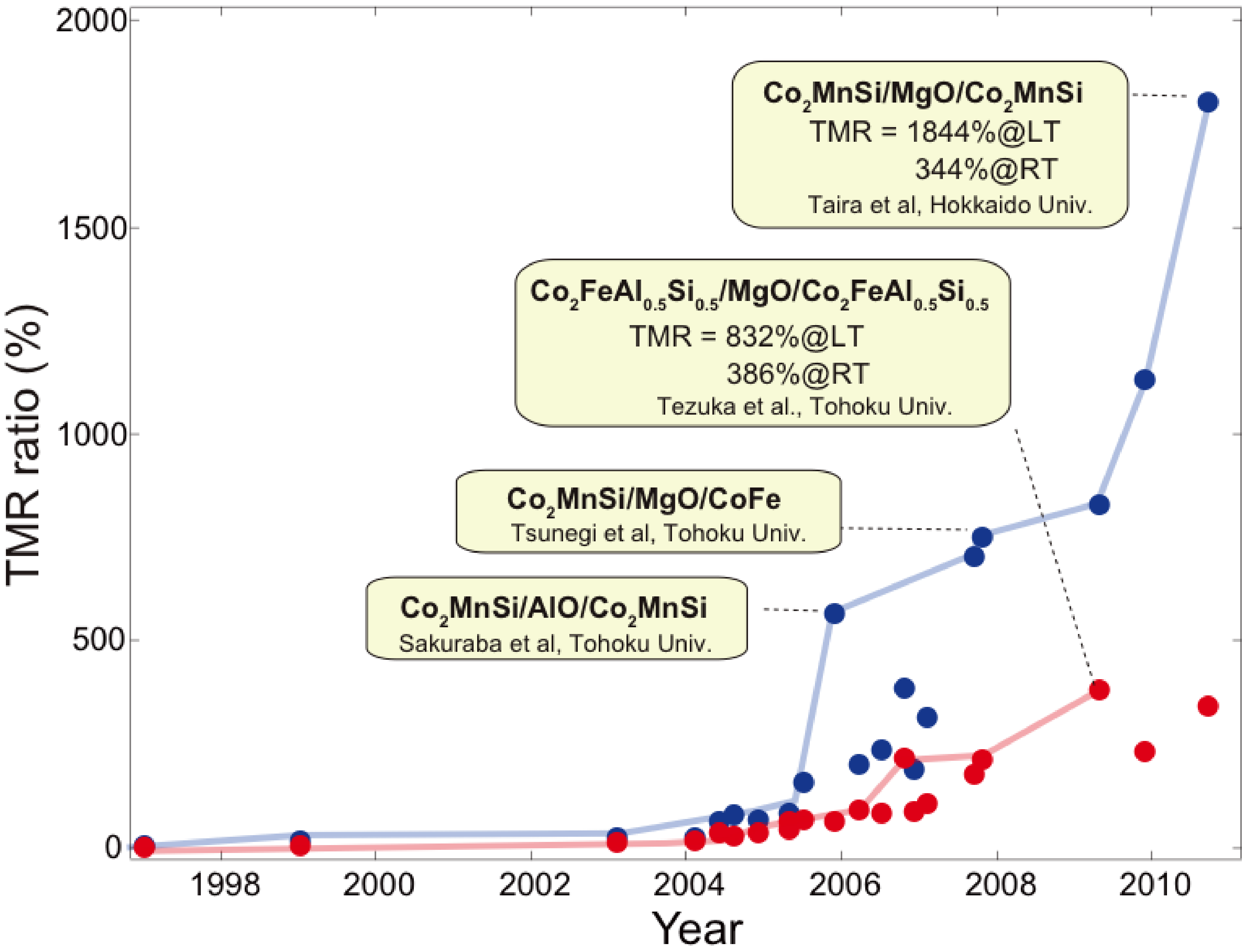
$L2_1$ structure

Co_2MnSi (CMS)

- **Half-metallic energy gap : 400 – 600 meV**
- **High T_c (~ 985K)**
- **Highly ordered $L2_1$-structure is easily obtained.**



Development of TMR for Heusler alloys



$Co_2MnSi/MgO/Co_2MnSi$
TMR = 1844%@LT
344%@RT
Taira et al, Hokkaido Univ.

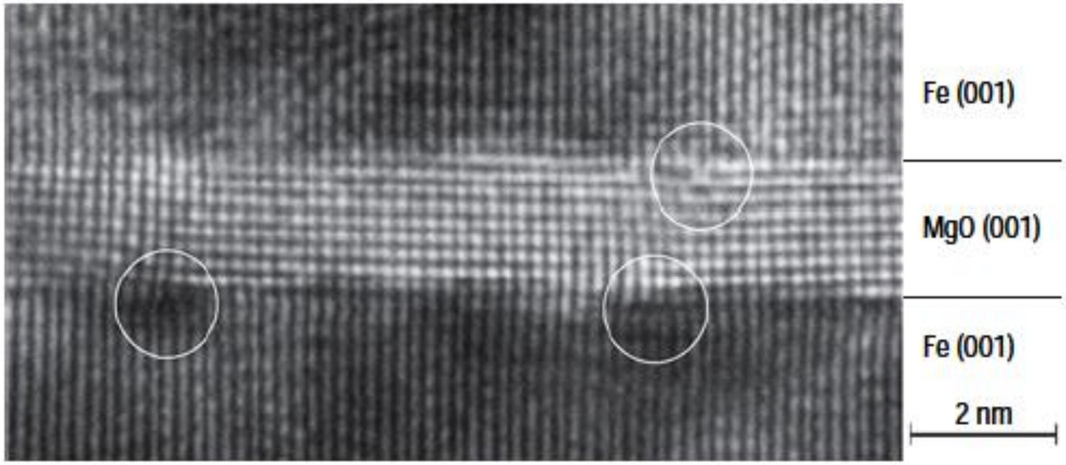
$Co_2FeAl_{0.5}Si_{0.5}/MgO/Co_2FeAl_{0.5}Si_{0.5}$
TMR = 832%@LT
386%@RT
Tezuka et al., Tohoku Univ.

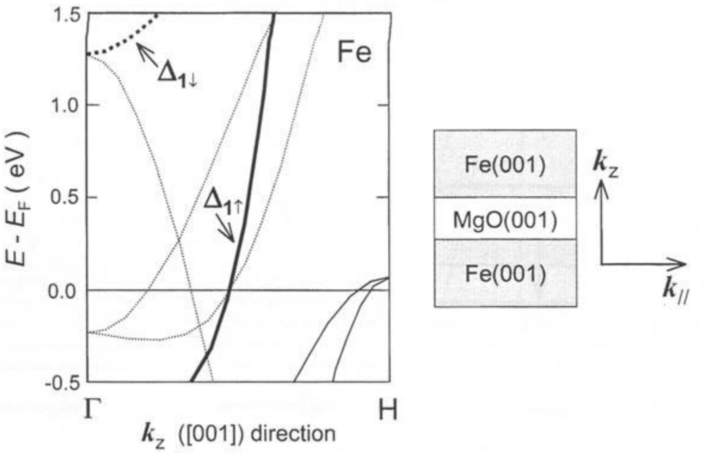
$Co_2MnSi/MgO/CoFe$
Tsunegi et al, Tohoku Univ.

$Co_2MnSi/AlO/Co_2MnSi$
Sakuraba et al, Tohoku Univ.

Giant TMR in MgO-MTJ

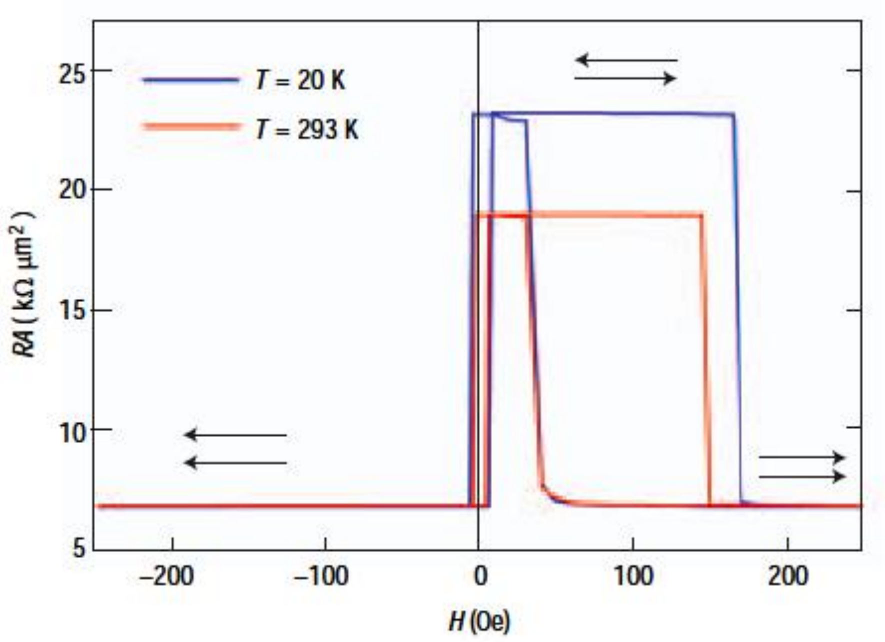
Fe (001) / MgO (001) / Fe (001) single crystal





Band structure of Fe

Δ_1 band: ***half metallic*** nature



MR = 180% (RT）

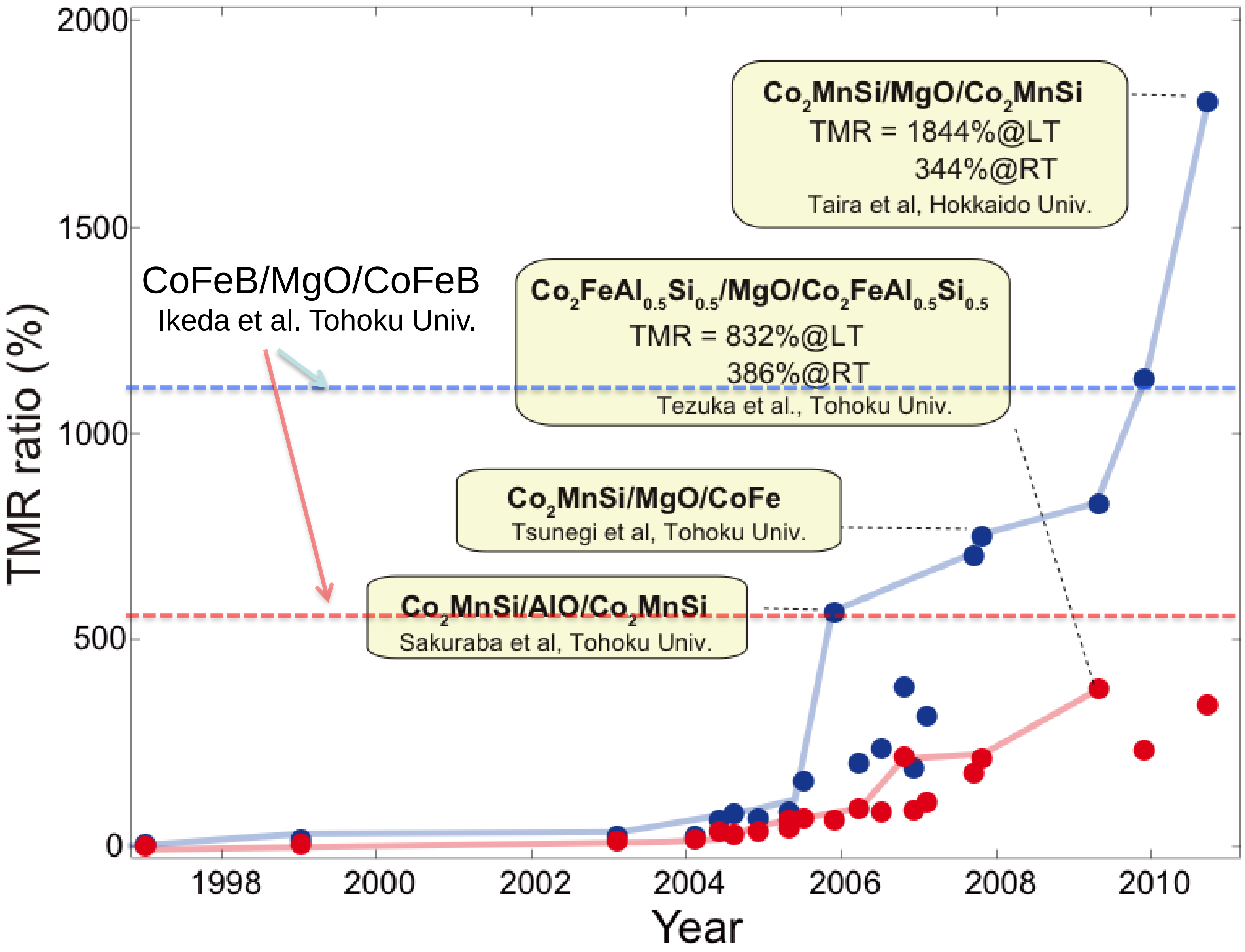
247% (4.2K)

S. Yuasa *et al.*,

Jpn. J. Appl. Phys., **43** (2004) L588.

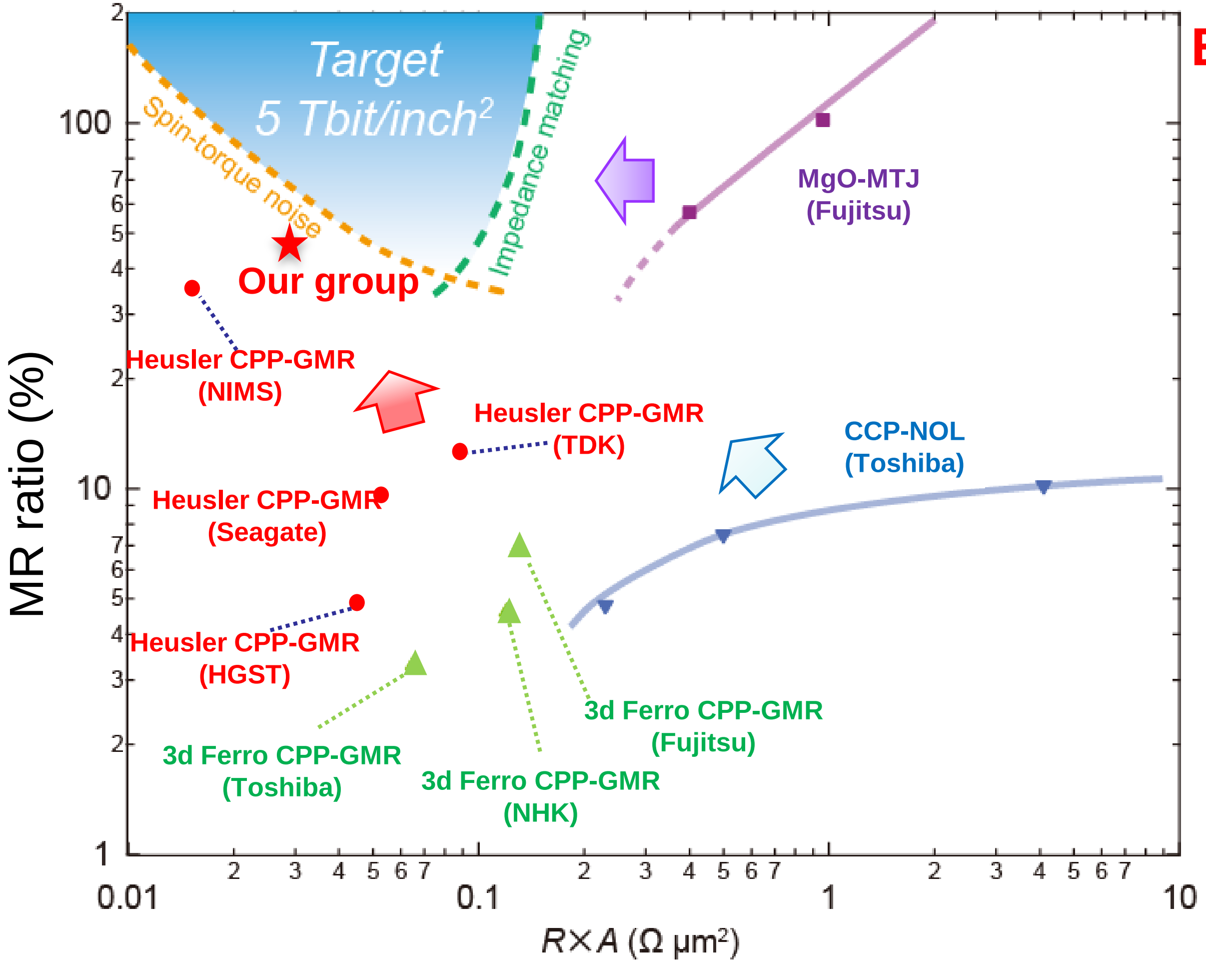
Nat. Mater., **3** (2004) 868.

Development of TMR for Heusler alloys



High MR and low resistance

Reported MR ratio in small *RA* region



R × *A*: resistance area product

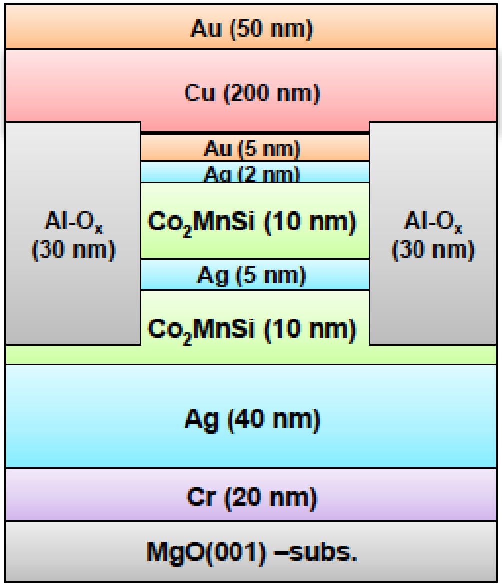
Essential decrease in TMR with reducing resistance

Half metal (P=100 %): Heusler alloys are still promising!

TMR

CPP-GMR

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



Au (50 nm)

Cu (200 nm)

Al-O_x (30 nm)

Au (5 nm)

Ag (2 nm)

Co_2MnSi (10 nm)

Ag (5 nm)

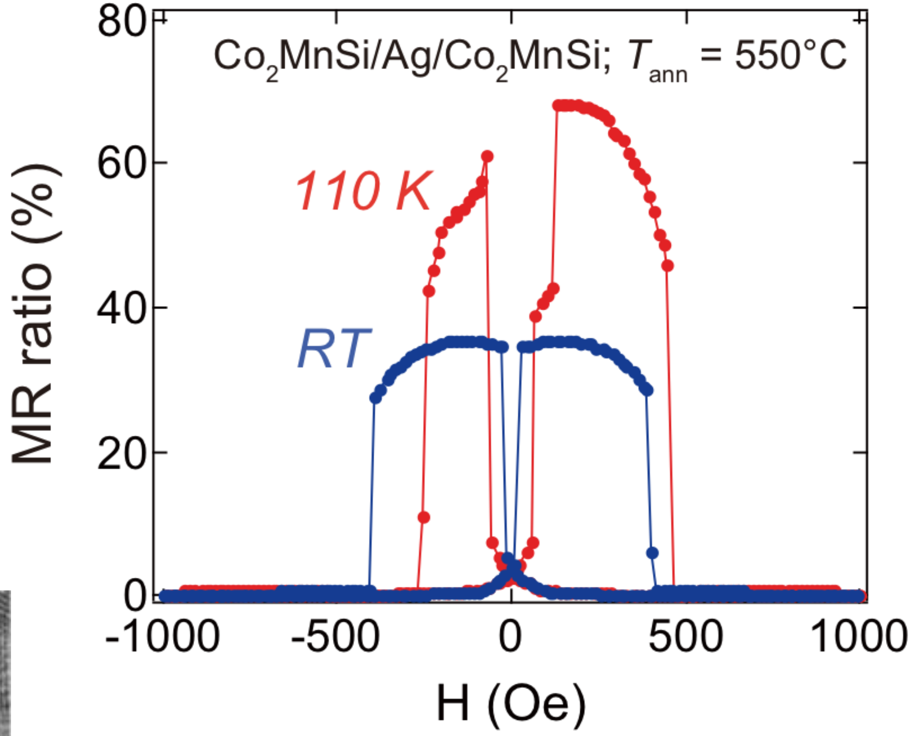
Co_2MnSi (10 nm)

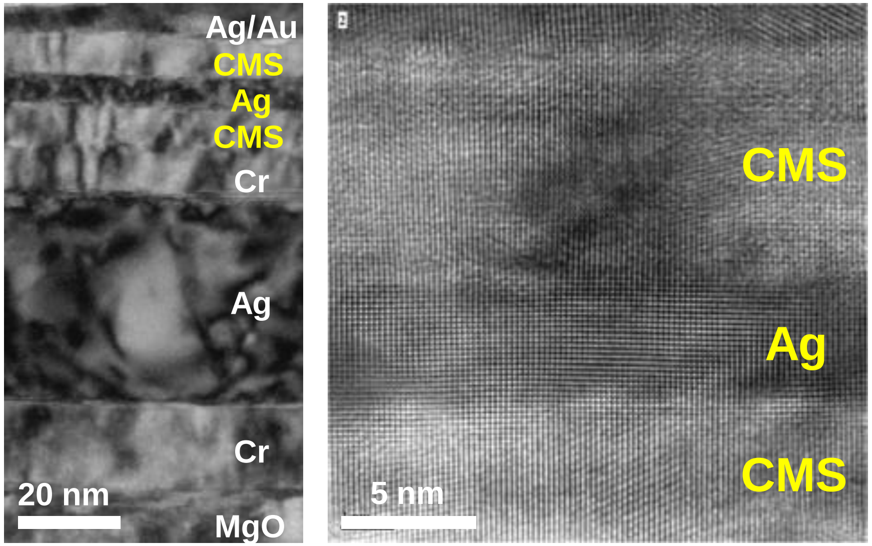
Al-O_x (30 nm)

Ag (40 nm)

Cr (20 nm)

MgO(001) –subs.





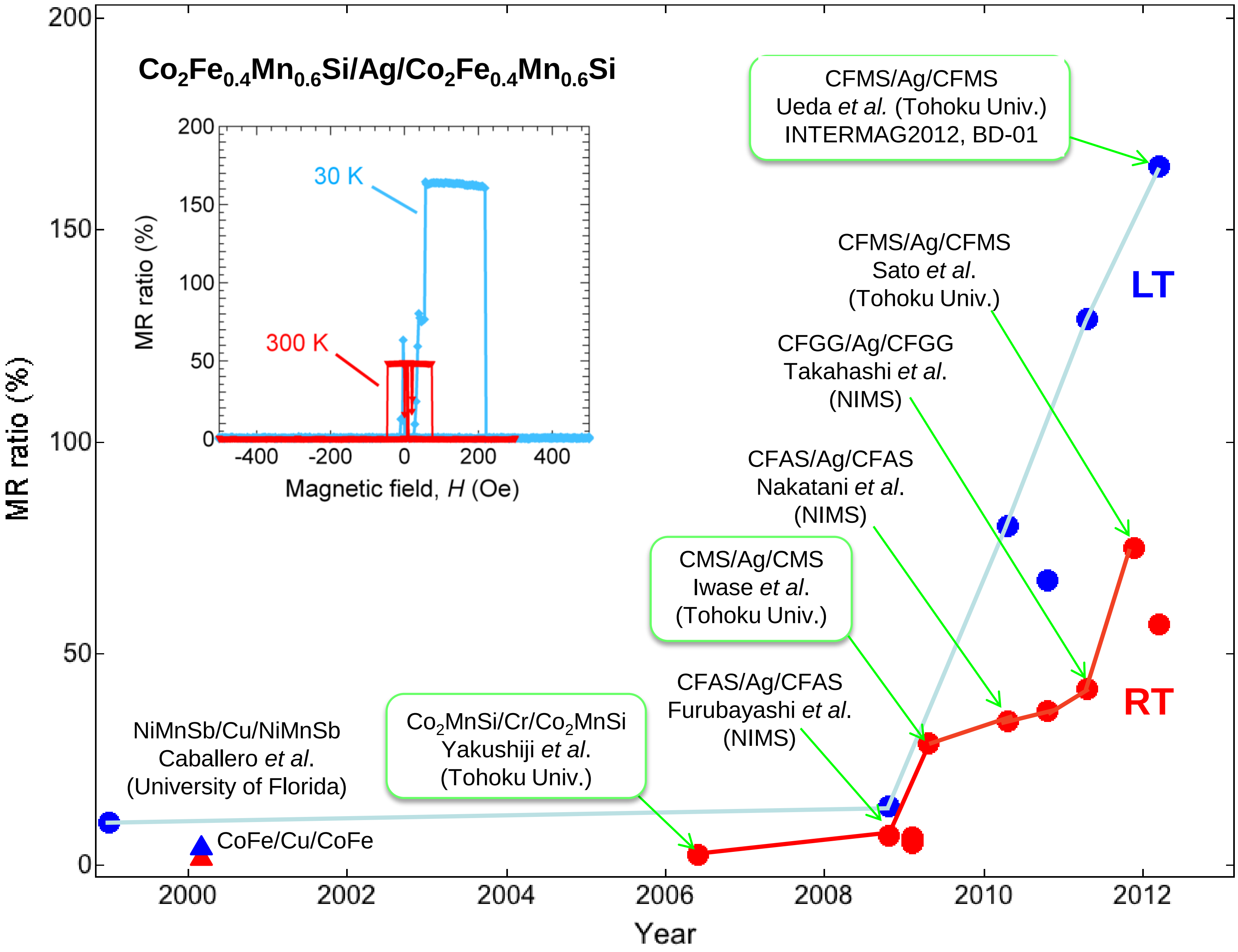
Fully-epitaxial growth in CMS/Ag/CMS

Breakthrough of CPP-GMR

A high MR ratio (36.4%@RT) was observed.

T. Iwase, K T *et al.*, Appl. Phys. Exp., 2 (2009) 063003.
Y. Sakuraba, KT *et al.*, Phys. Rev. B82 (2010) 094444.

Development of CPP-GMR for Heusler alloys



CFMS/Ag/CFMS
Ueda *et al.* (Tohoku Univ.)
INTERMAG2012, BD-01

CFMS/Ag/CFMS
Sato *et al*.
(Tohoku Univ.)

CFGG/Ag/CFGG
Takahashi *et al*.
(NIMS)

CFAS/Ag/CFAS
Nakatani *et al*.
(NIMS)

CMS/Ag/CMS
Iwase *et al*.
(Tohoku Univ.)

CFAS/Ag/CFAS
Furubayashi *et al*.
(NIMS)

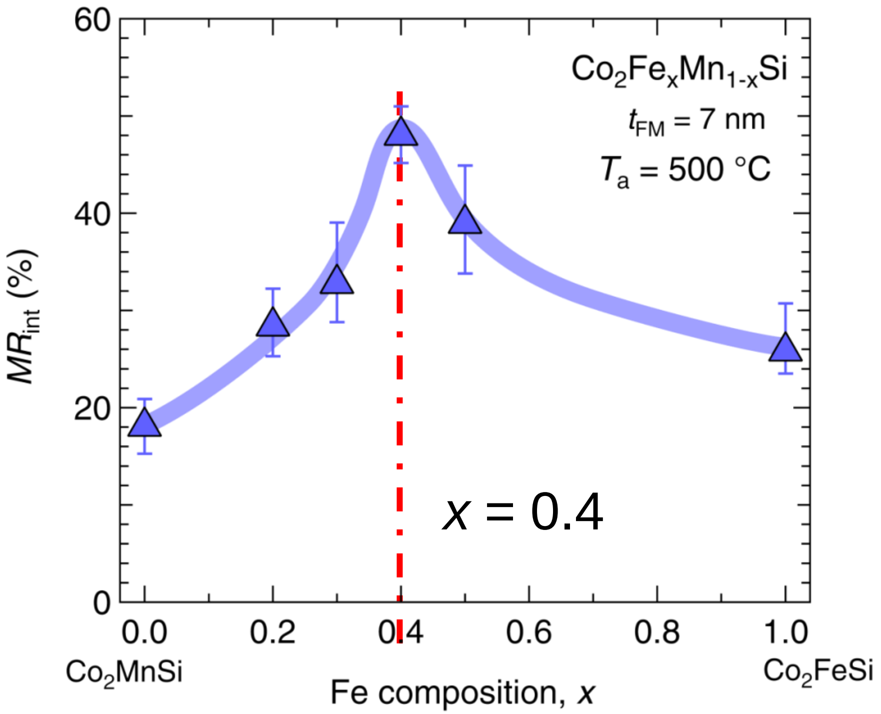
$Co_2MnSi/Cr/Co_2MnSi$
Yakushiji *et al*.
(Tohoku Univ.)

NiMnSb/Cu/NiMnSb
Caballero *et al*.
(University of Florida)

CoFe/Cu/CoFe

CPP-GMR in CFMS/Ag/CFMS

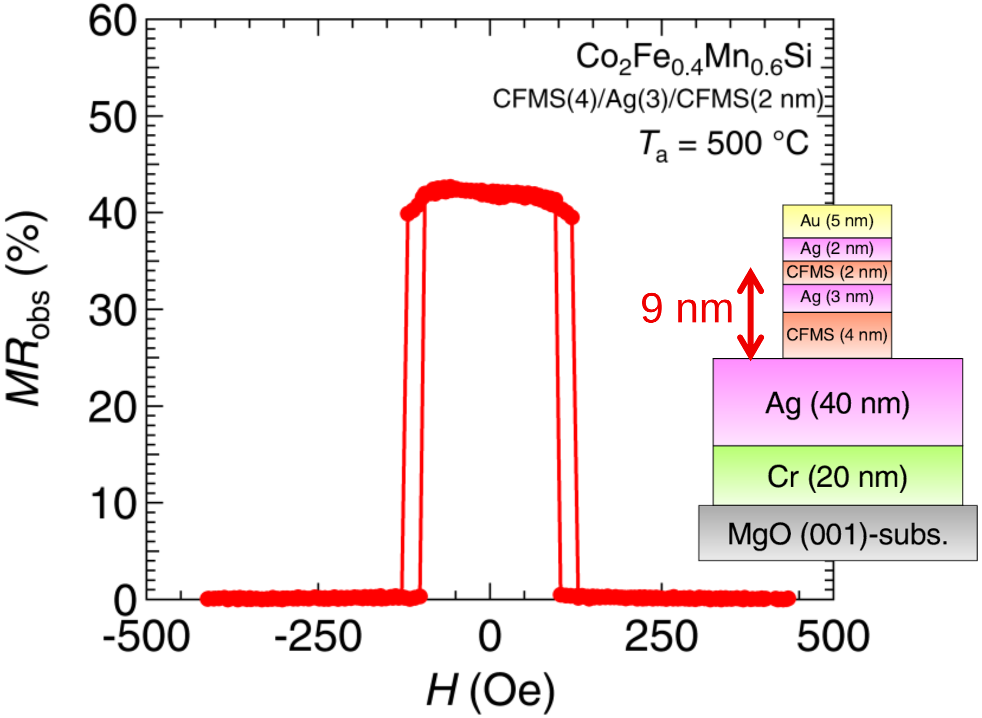
$Co_2Fe_xMn_{1-x}Si(20)/Ag(5)/Co_2Fe_xMn_{1-x}Si(7)$



Best composition ratio : $Co_2Fe_{0.4}Mn_{0.6}Si$

Average MR ratio	*RA*	*ΔRA*
48 %	24.3 mΩ·μm^2	11.8 mΩ·μm^2

$Co_2Fe_{0.4}Mn_{0.6}Si(4)/Ag(3)/Co_2Fe_{0.4}Mn_{0.6}Si(2)$



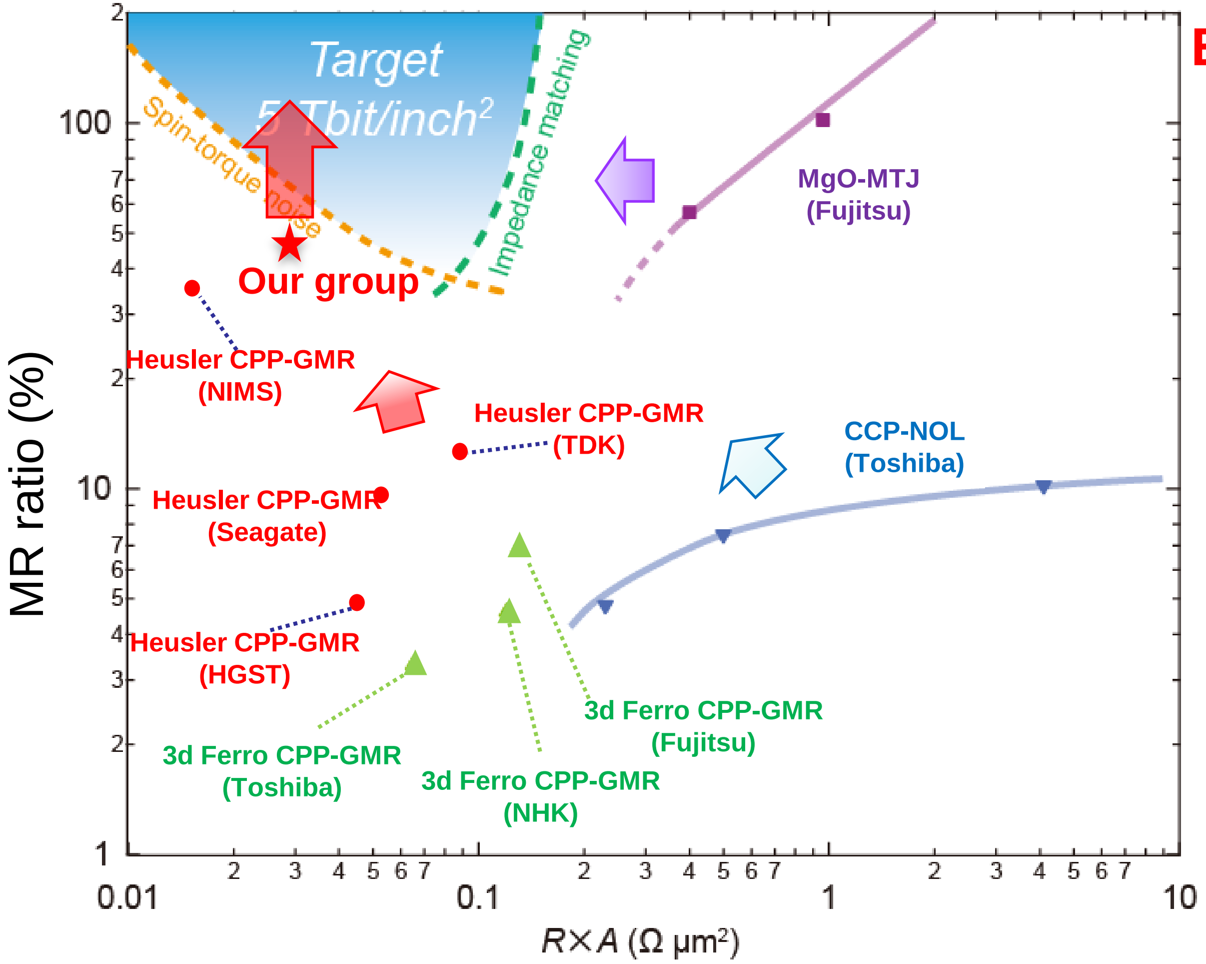
MR_{int}	*RA*	*ΔRA*
58%	21.7 mΩ·μm^2	12.5 mΩ·μm^2

Y.Sakuraba, KT, et al. Appl. Phys. Lett., 101 (2012) 252408.

Large MR ratio even in very thin trilayer structure !

High MR and low resistance

Reported MR ratio in small *RA* region



R × *A*: resistance area product

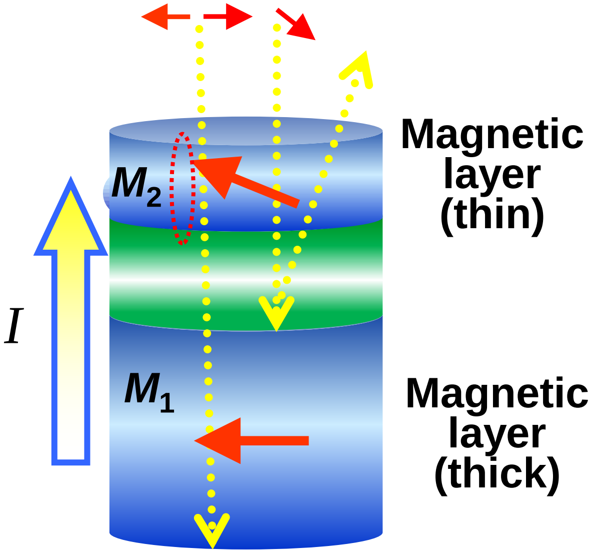
Essential decrease in TMR with reducing resistance

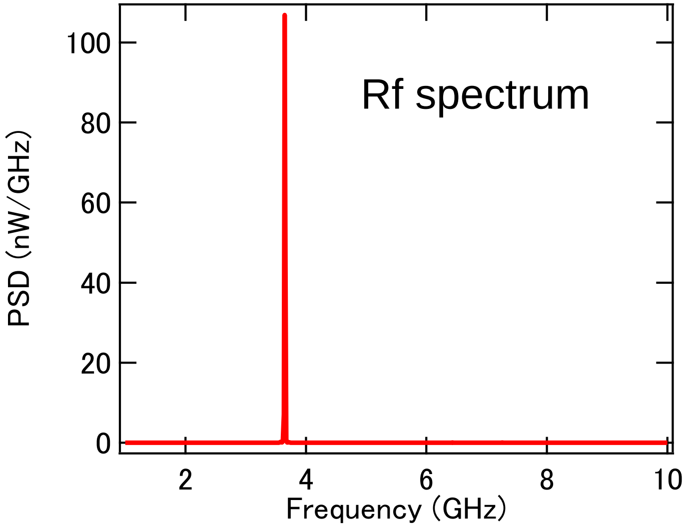
Half metal (P=100 %): Heusler alloys are still promising!

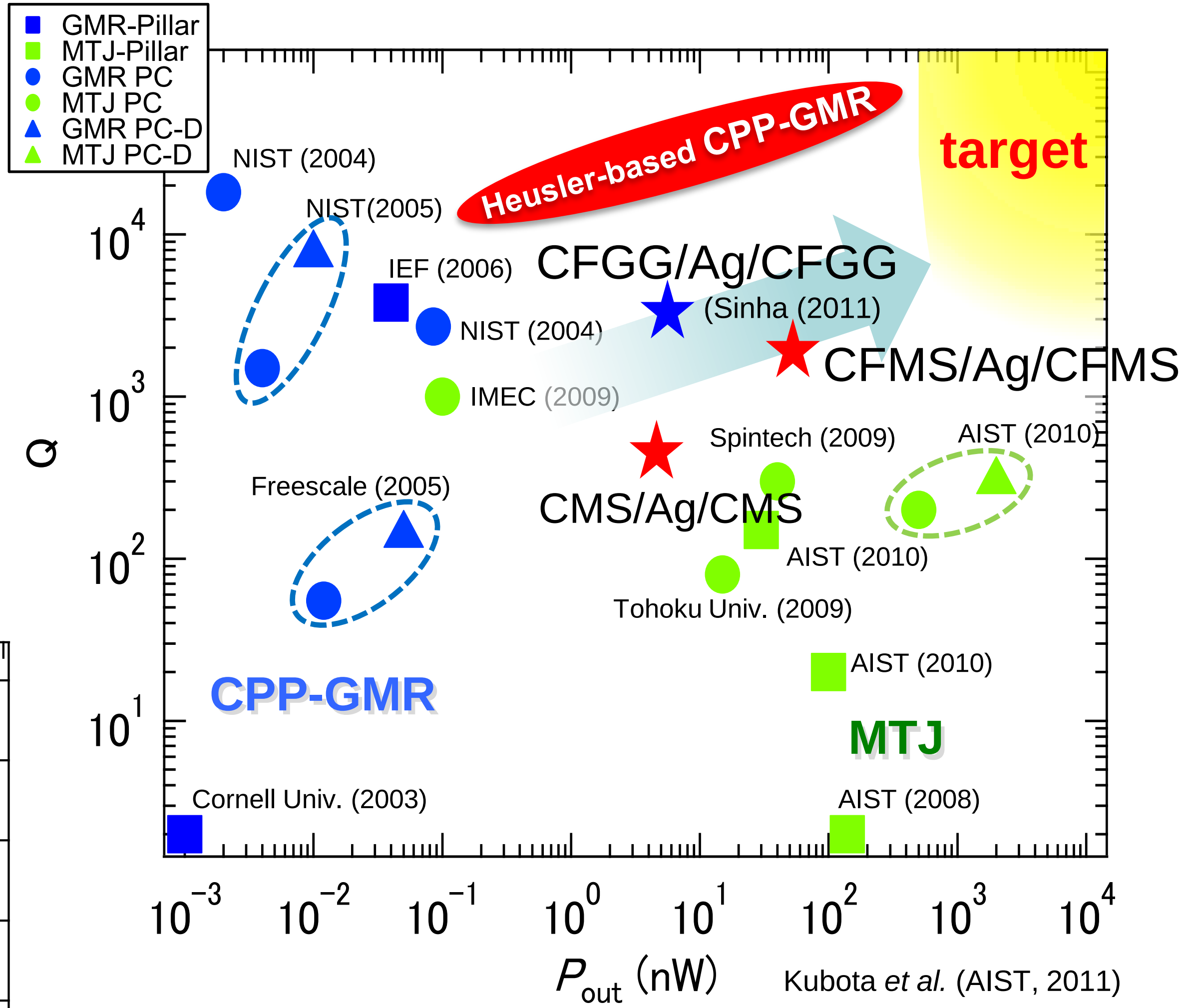
TMR

CPP-GMR

Rf oscillation in Heusler alloys by spin transfer torque





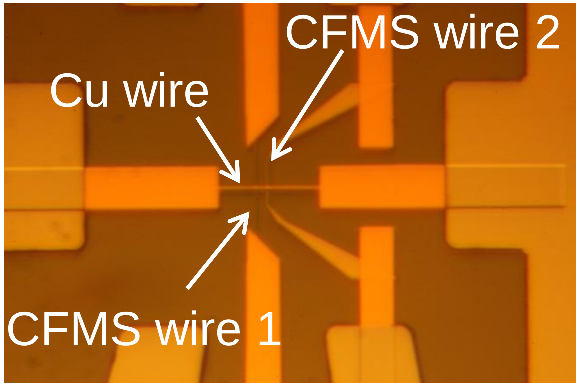


Kubota *et al.* (AIST, 2011)

R. Okura *et al*., Appl. Phys. Lett., 99 (2011) 052510.

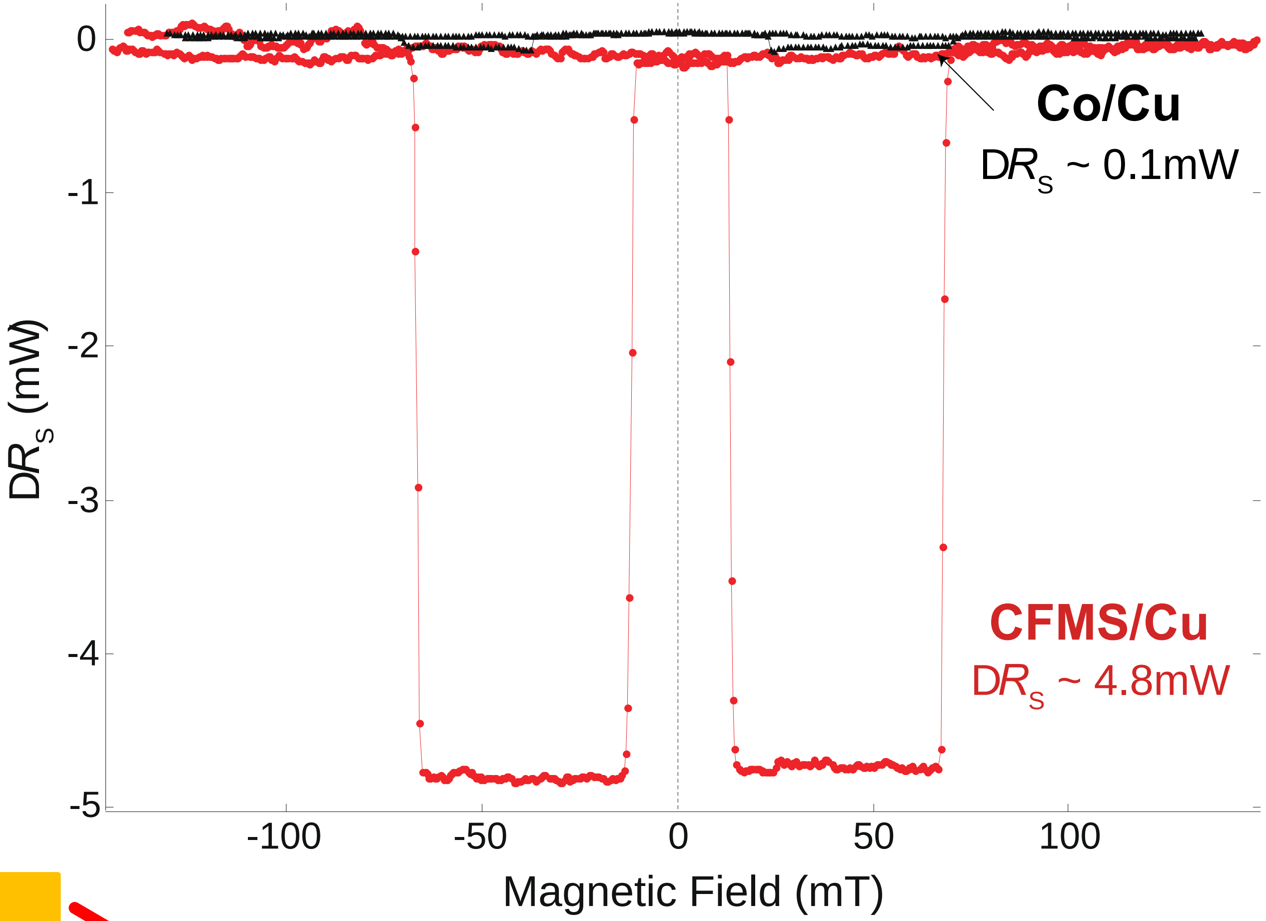
Non-local spin injection in lateral spin valves

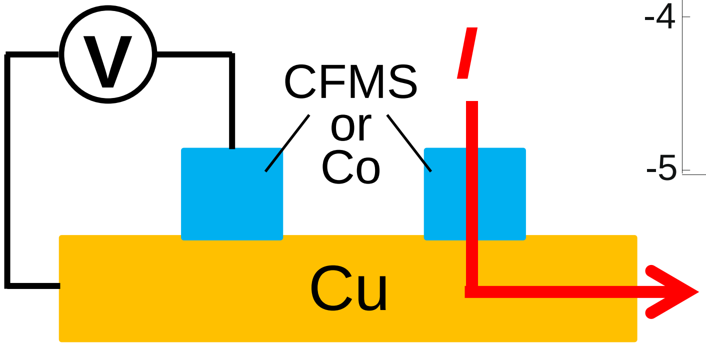
$Co_2Fe_{0.4}Mn_{0.6}Si$(CFMS)/Cu



Y. Sakuraba *et al.*,
unpublished.

NLSV signal@RT, gap = 350 nm





Non-local spin injection

Observation of large spin accumulation signal
Spin injection with high efficiency

Perpendicularly spin-polarized materials:
$L1_0$-ordered alloys
with high magnetic anisotropy

Perpendicular magnetization and spintronics

High magnetic anisotropy → Thermal stability of magnetization

Negative shape anisotropy → Easy magnetization switching

No restriction on aspect ratio

→ High Integration

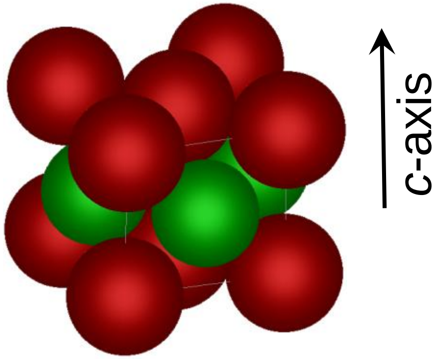
Examples of perpendicularly magnetized films

Co-based granular films such as CoCrPt-SiO_2

RE-TM amorphous alloy films such as TbFeCo

Metallic mutlilayers or ultrathin films such as Ni/Co, Co/Pd, CoFeB/MgO, etc.

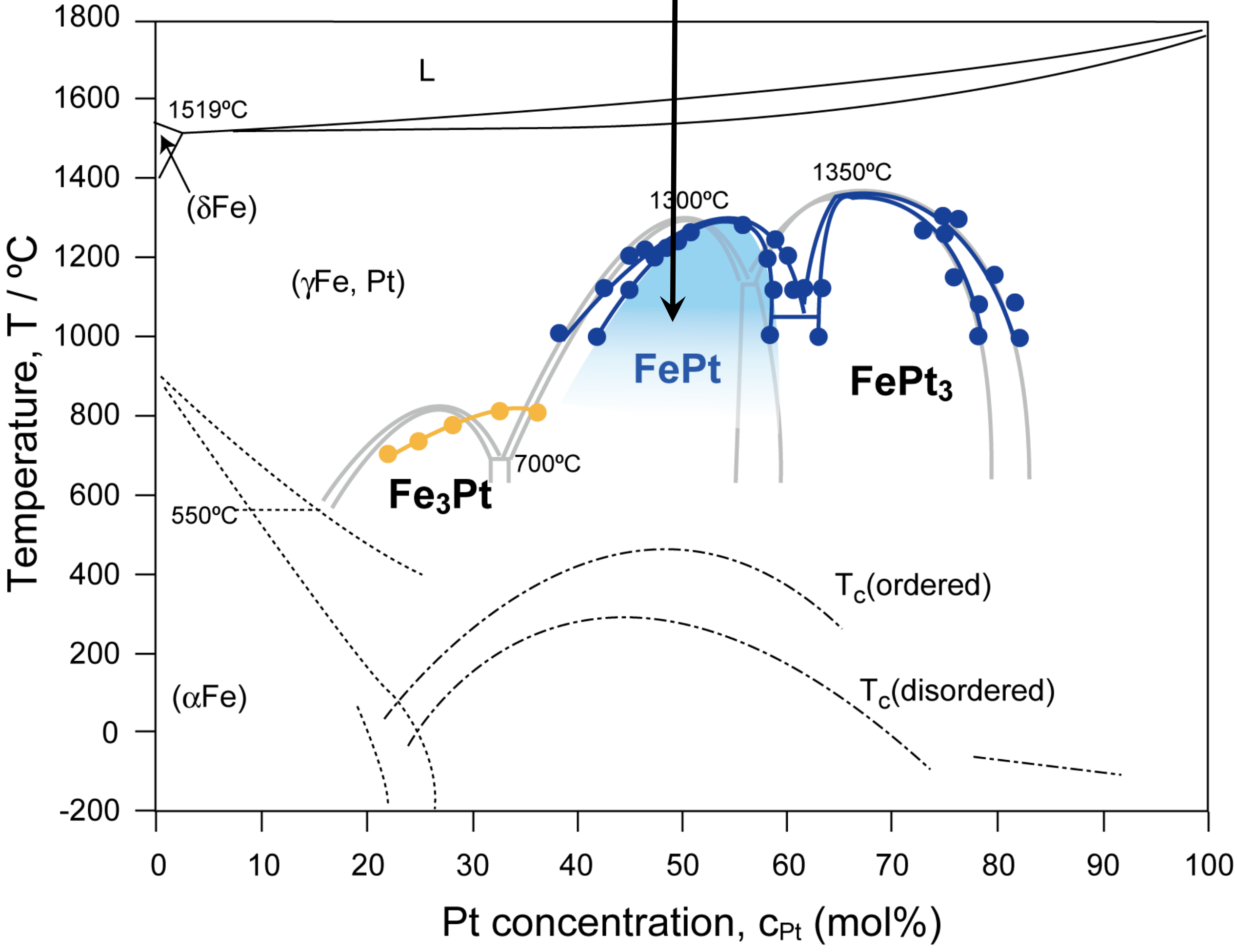
$L1_0$ ordered alloy films such as FePt, FePd, CoPt, etc.



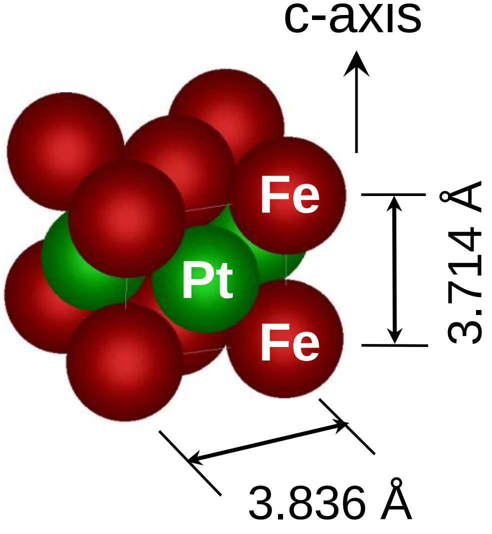
$L1_0$ structure

L1_0 ordered FePt alloy

L1_0 ordered phase



Phase diagram of Fe-Pt system

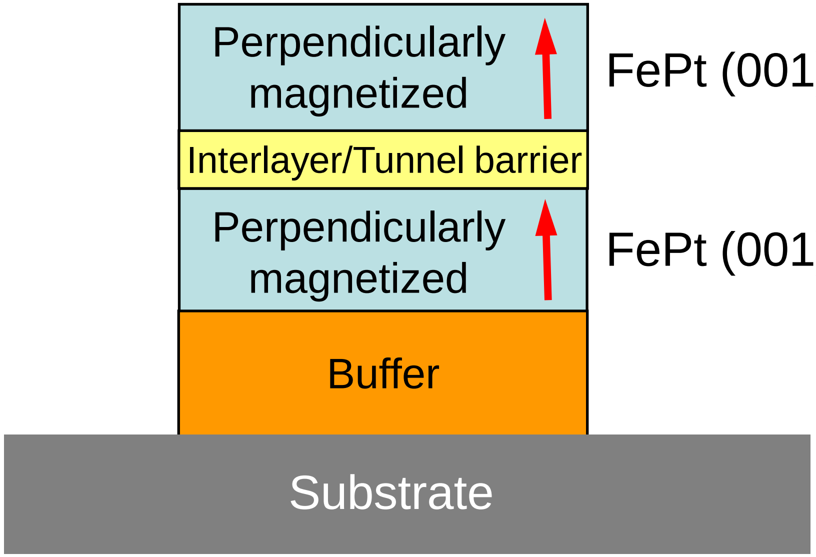


Large uniaxial magnetic anisotropy

$K_u = 7 \times 10^7$ erg/cm^3

- Perpendicular magnetic recording media
- Patterned media
- **Spintronics**

Spin-torque switching of magnetization for $L1_0$-FePt



Perpendicularly magnetized

FePt (001)

Interlayer/Tunnel barrier

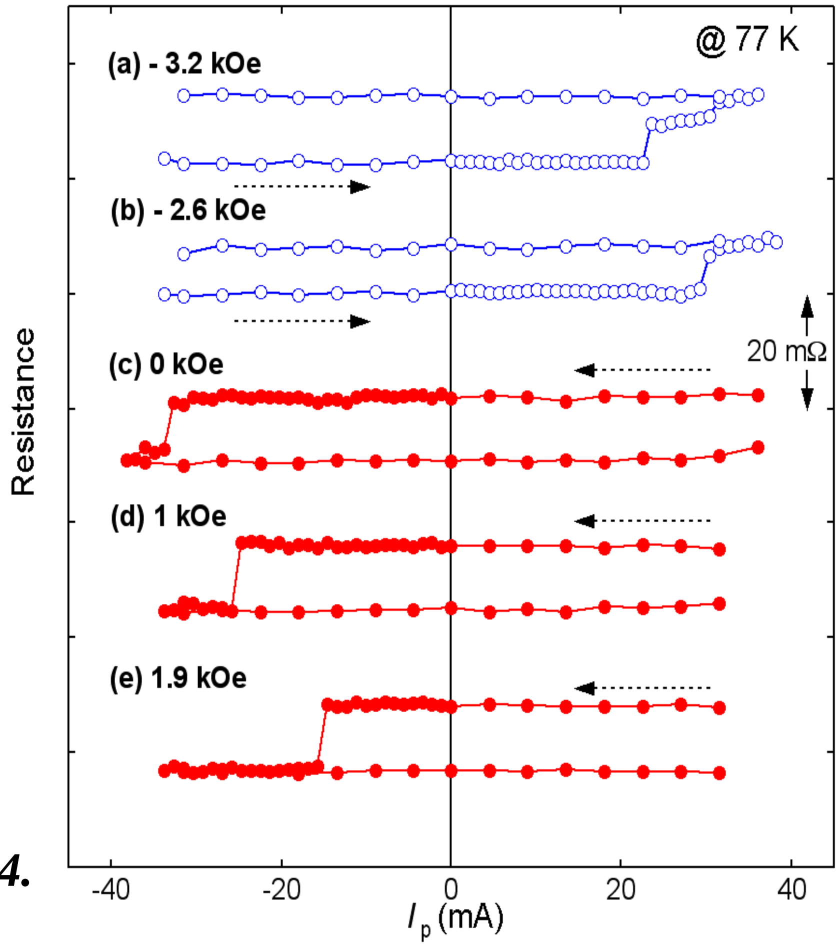
Perpendicularly magnetized

FePt (001)

Buffer

Substrate

Fully epitaxial



FePt / Au / FePt CPP-GMR pillars

Spin-torque switching of magnetization

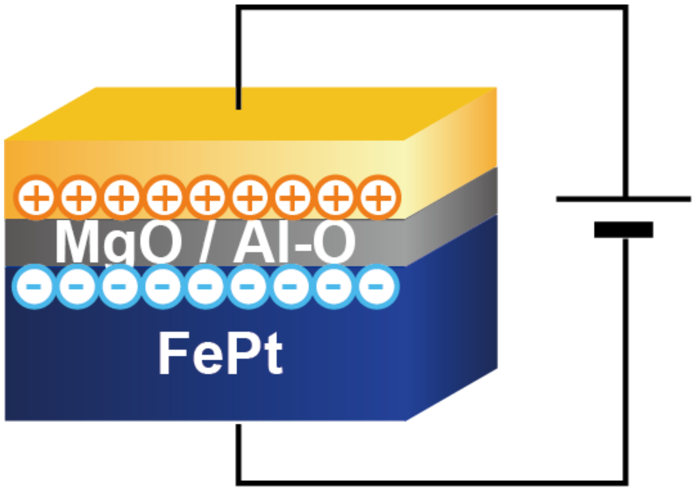
T. Seki, KT, et al., Appl. Phys. Lett. 88 (2006) 172504.

$[Co/Pt]_4$ / $[Co/Ni]_2$ / Cu / $[Co/Ni]_4$
S. Mangin et al., Nature Mater., 5 (2006) 210.

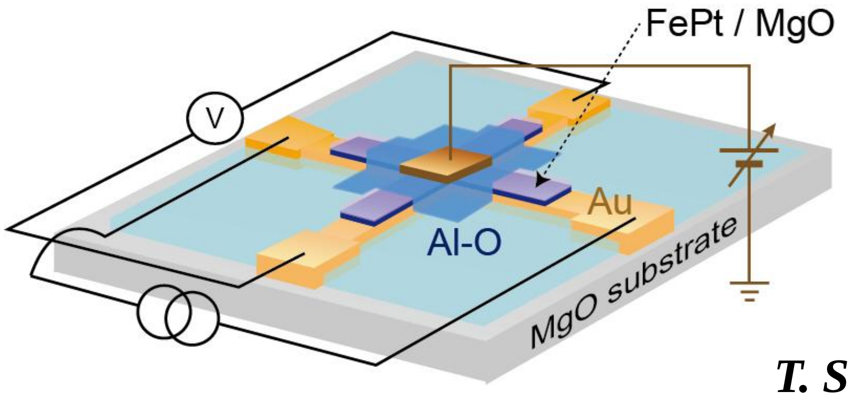
$[CoFe/Pt]_5$ / Cu / $[CoFe/Pt]_7$
H. Meng and J.-P. Wang, Appl. Phys. Lett., 88 (2006) 172506.

Coercivity control by electric field for $L1_0$-FePt

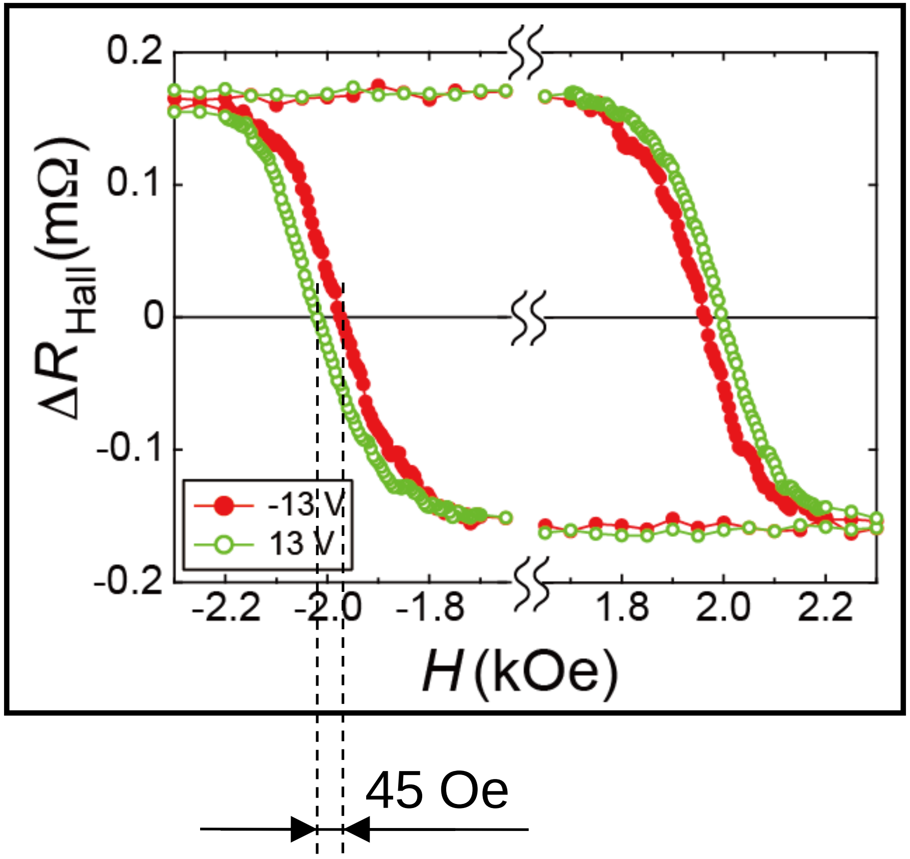
Perpencularly magnetized $L1_0$-FePt



FePt / MgO / Al-O Hall device



Anomalous Hall resistance curve



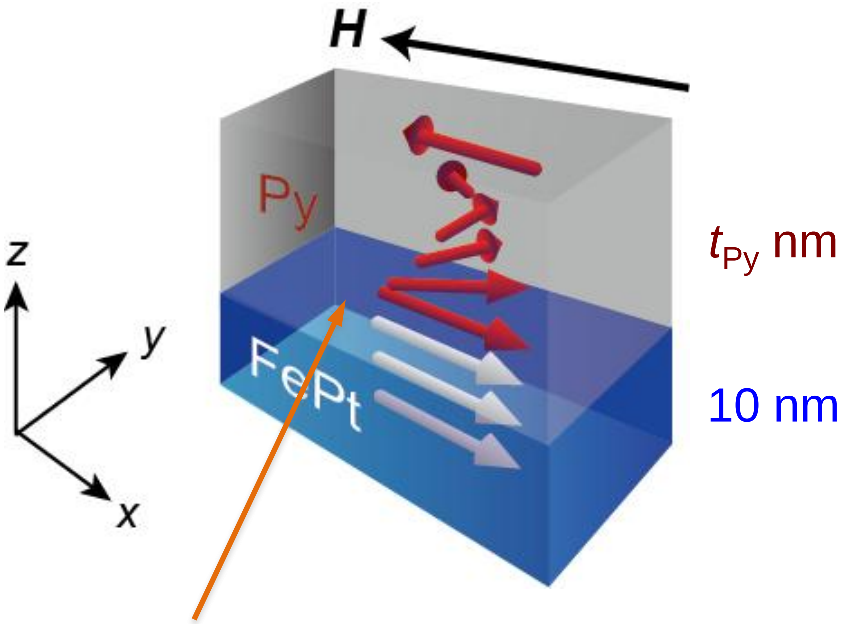
H_c modulation
by changing V_{app} (-13 ~ 13V)

T. Seki, KT, et al., Appl. Phys. Lett., 98 (2011) 212505.

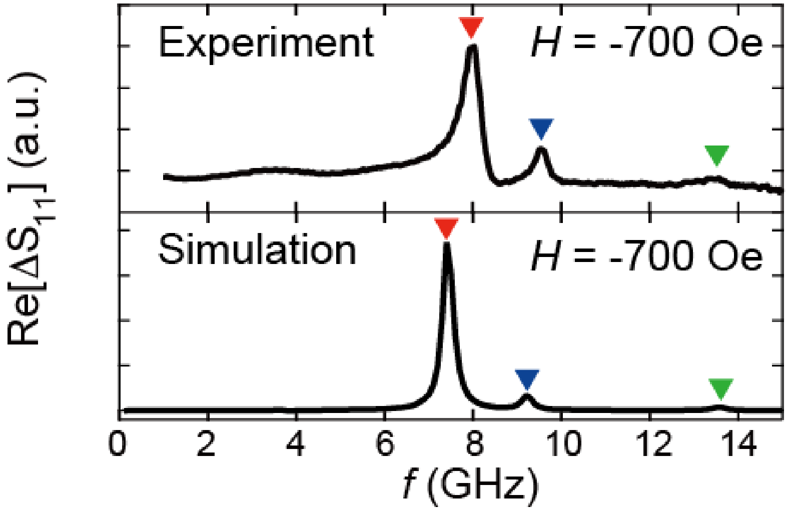
Spin wave-assisted magnetization switching

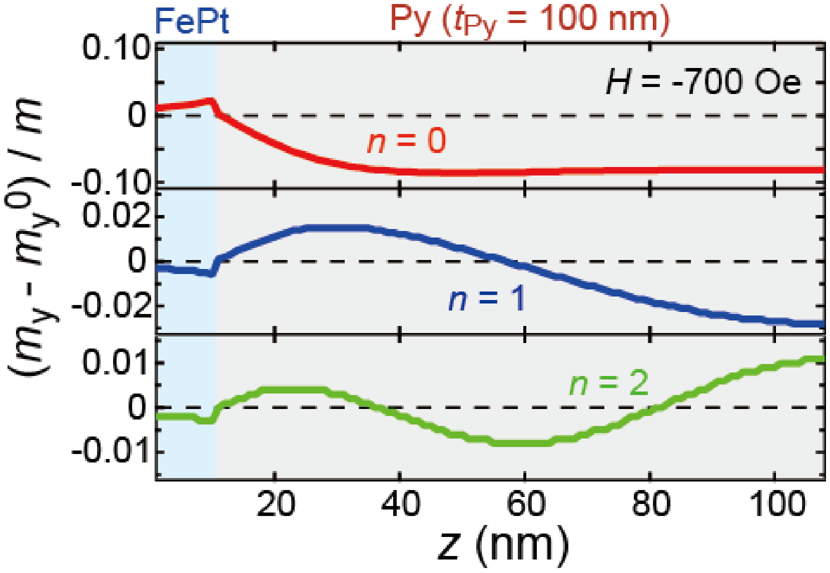
T. Seki, KT, et al., Nature Commun., **4:1726 doi: 10.1038/ ncomm2737 (2013).**

FePt / Permalloy (Py) Exchange-Coupled Bilayer

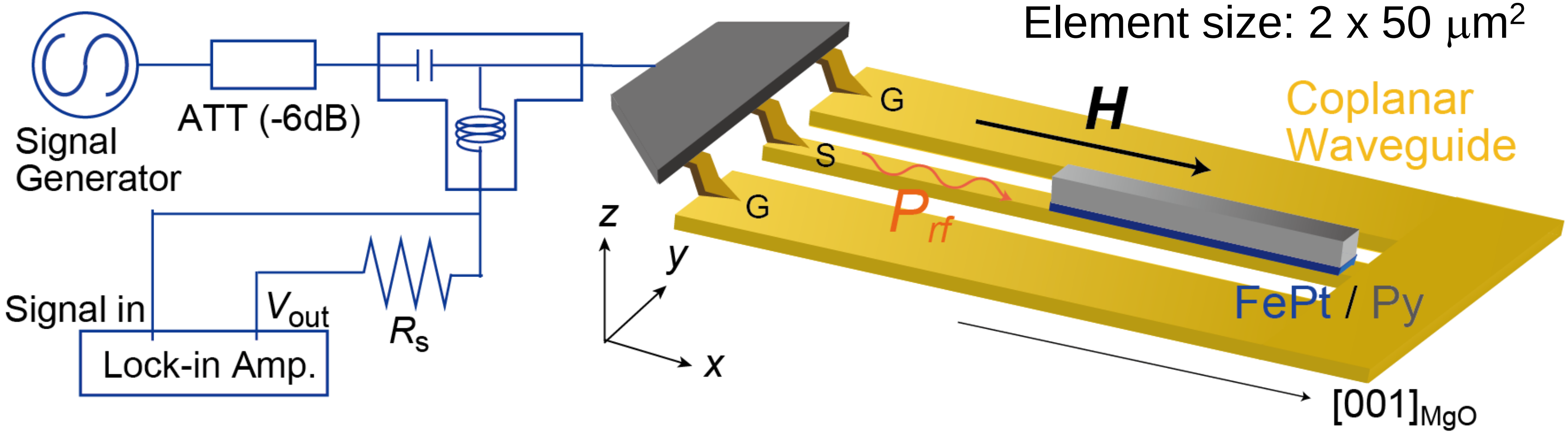


Utilization of Perpendicular Standing Spin Wave Mode in the Bilayer



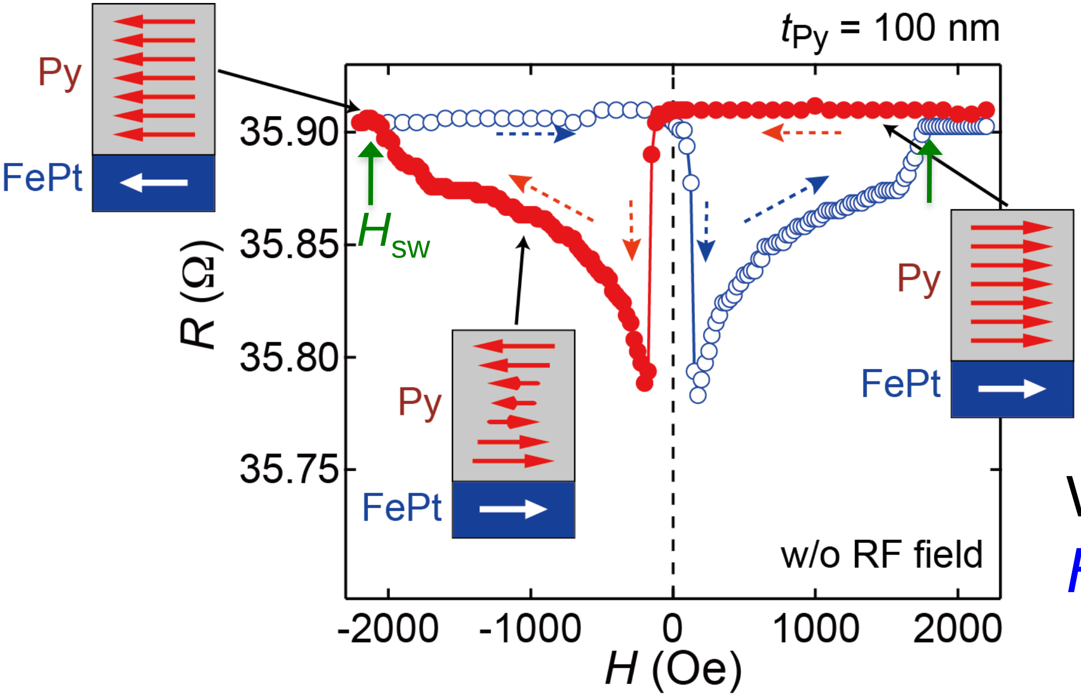


Spin wave-assisted magnetization switching



Element size: 2 x 50 μm^2

AMR curve

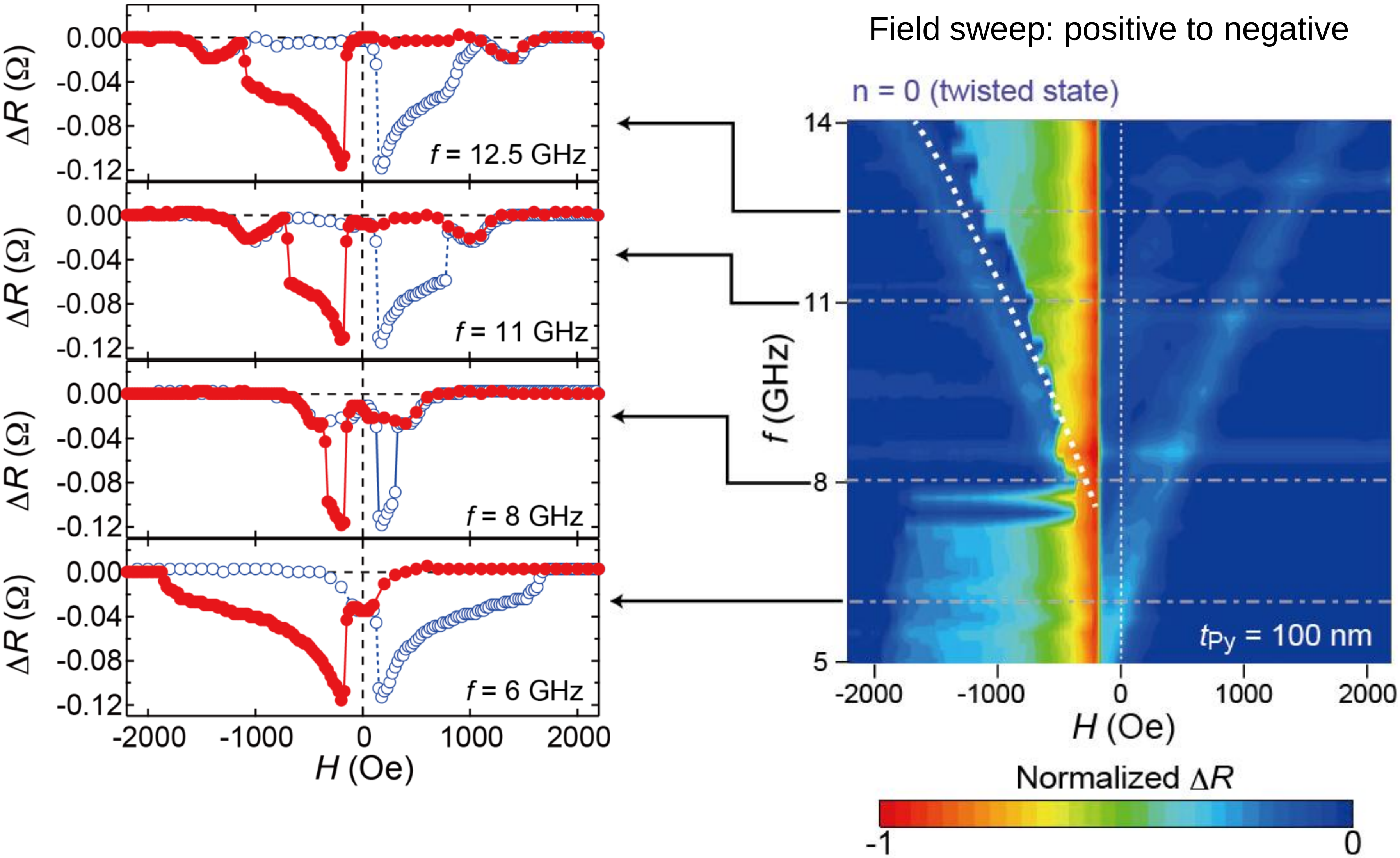


Without spin wave excitation,
H_{sw} ~ 1900 Oe.

Spin wave-assisted magnetization switching

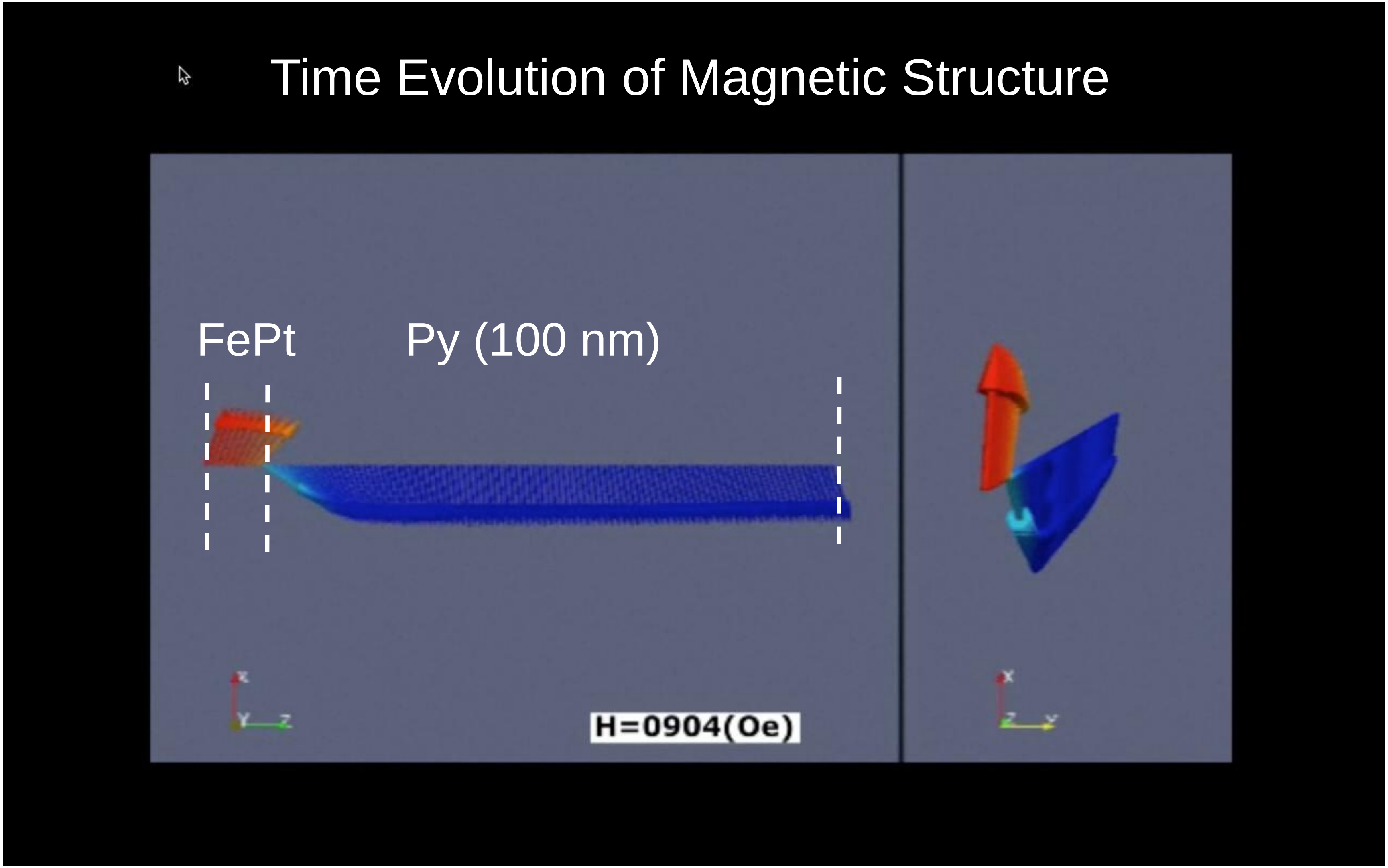
t_{Py} = 100 nm (H_{rf} = 145 Oe)

Field sweep: positive to negative



Spin wave assisted magnetization switching

Time evolution of magnetic structure by micromagnetics simulation



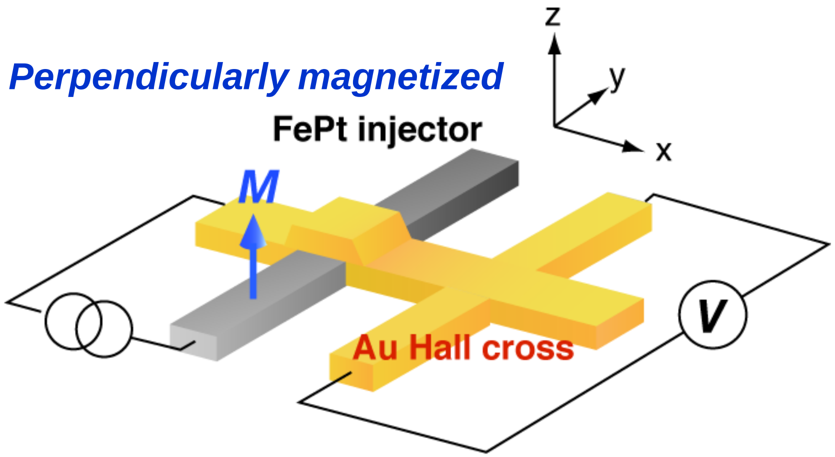
Magnetic Field Sweep: 50 Oe/nsec
H_{rf} = 90 Oe, f = 10 GHz

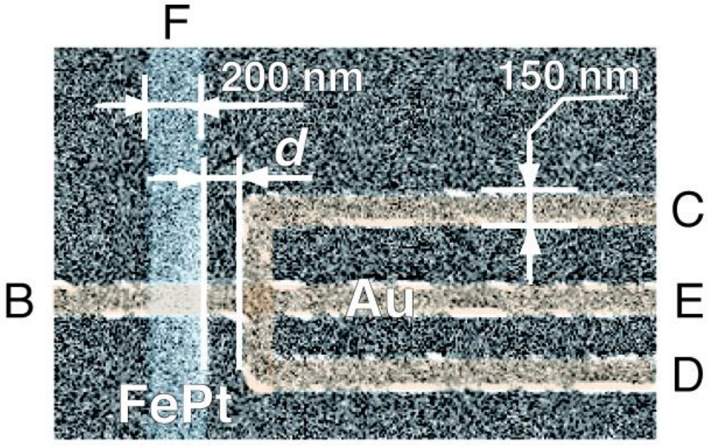
by Y. Nozaki, Keio Univ.

Observation of giant spin Hall effect in perpendicularly magnetized FePt/Au devices

T. Seki, K. T. et al., Nature Materials, 7 (2008) 125.

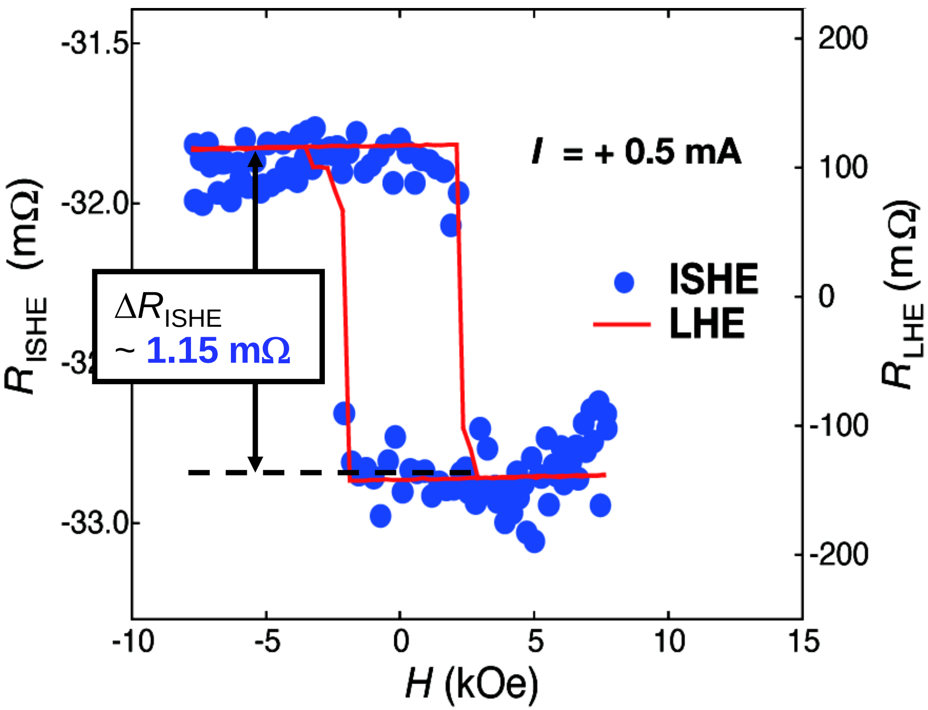
Perpendicularly magnetized





d = 70 nm ~ 400 nm

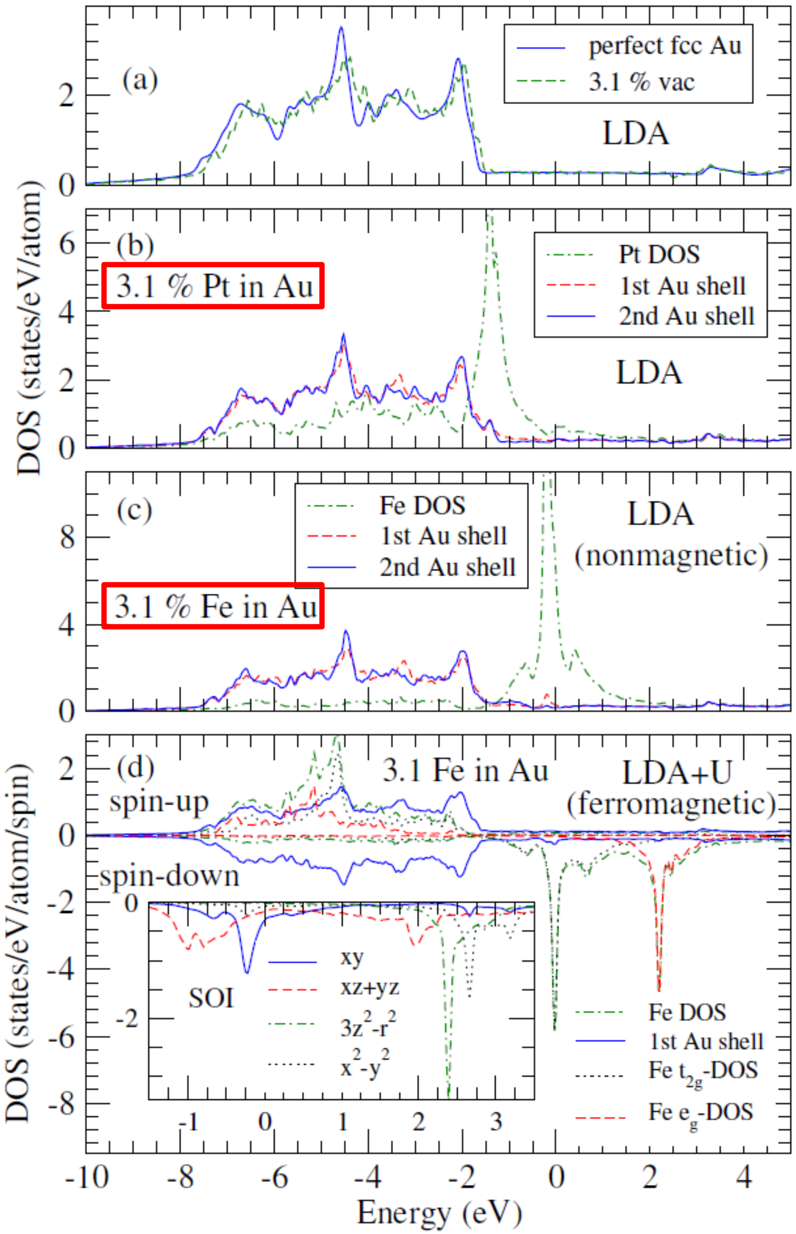
@ RT, d = 150 nm



Spin Hall angle α_H~0.1

Electrical detection of giant spin Hall effect at room temperature

Theoretical discussion



G.Y. Guo, S. Maekawa, and N. Nagaosa
Phys. Rev. Lett., 102 (2009) 036401.

Spin Hall Effect by Kondo singlet state

Orbital selective Kondo

e_g Kondo limit → $T_K \approx 0.4K$

t_{2g} Mixed valence d^6 and d^7

hybridization with Au
s- and d-orbitals

Renormalization effect due to electron correlation

$$\Delta = 1.4eV \Rightarrow \Delta^* = 0.3eV$$

$$10Dq = 0.1eV \Rightarrow 10Dq^* = 2.0eV$$

$$\lambda = 0.03eV \Rightarrow \lambda^* \approx 1eV$$

Resonant skew scattering

➡ Giant SHE

Recent development on giant SHE

- Enhancement due to skew scattering by impurities

Our study

Undoped Au : $\alpha_H = 0.05$ (corrected by geometrical effect)

Fe-doped Au : $\alpha_H = 0.05$

*I. Sugai, KT, et al., IEEE Trans Magn., **46** (2010) 2559.*

Pt-doped Au : $\alpha_H = 0.11$ ***Surface assisted skew scattering***

*B. Gu, KT, et al., Phys. Rev. Lett., **105** (2010) 216401.*

Otani's group (Univ. Tokyo)

Ir-doped Cu : $\alpha_H = 0.02$

*Y. Niimi et al., Phys. Rev. Lett., **106** (2011) 126601.*

Bi-doped Cu : $\alpha_H = 0.24$

*Y. Niimi et al., Phys. Rev. Lett., **109** (2012) 156602.*

Ralph's group (Cornell Univ.)

β-Ta : $\alpha_H = 0.15$ *L. Liu et al., Science, **336** (2012) 555.*

β-W: $\alpha_H = 0.33$ *C.-F. Pai et al., Appl. Phys. Lett., 101 (2012) 122404.*

$L1_0$ ordered alloy and element strategy

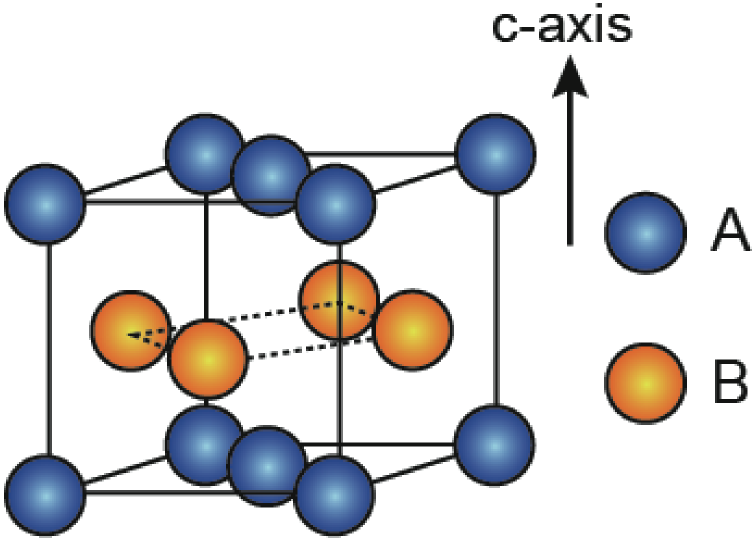
FePt, FePd, CoPt, etc.

High uniaxial magnetic anisotropy

$K_u = 10^7 \sim 10^8$ erg/cm^3

⬇

Spintronics
Magnetic storages
Permanent magnets



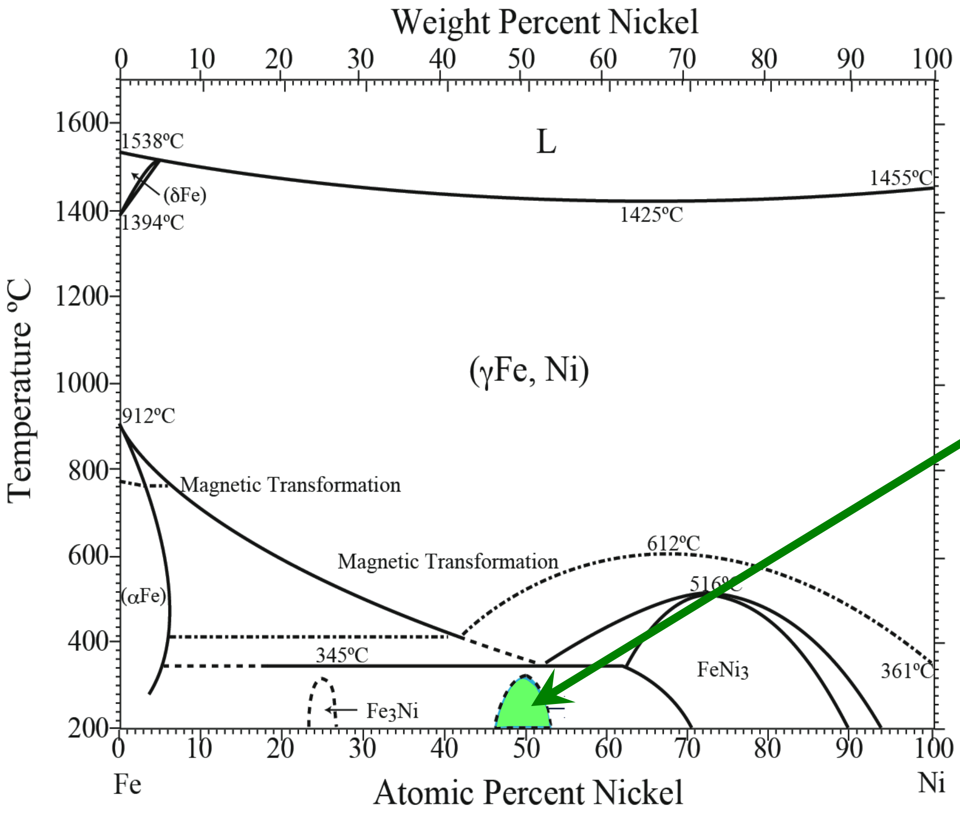
$L1_0$ ordered structure

However, a noble metal element is used in many cases!

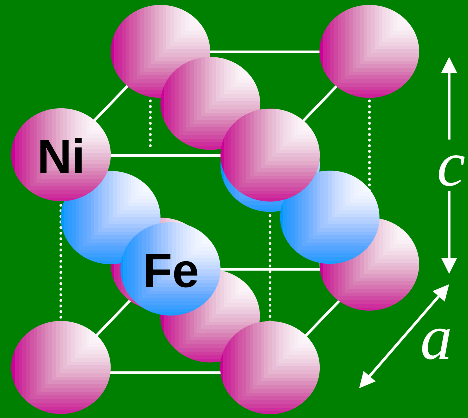
→ **Expensive**
High damping constant

→ **Requirement for a noble-metal-free $L1_0$ ordered alloy**

$L1_0$ ordered FeNi alloy



Order-disorder transformation temperature ~ 320ºC



$a \approx c = 3.582 \pm 0.002$ Å

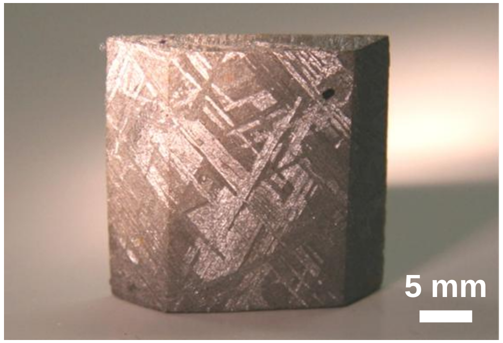
$L1_0$ ordered FeNi alloy

Requires annealing for an astronomically long time

Naturally found only in meteorites

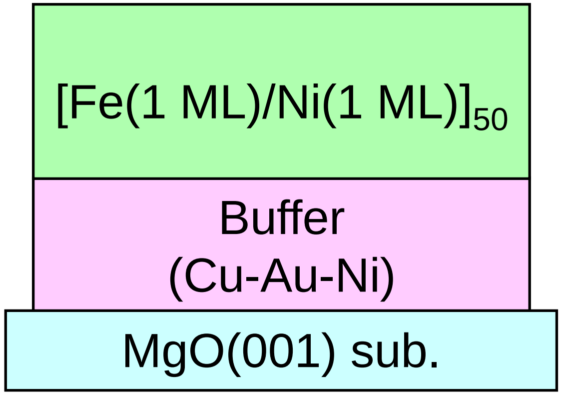
Neutron irradiation：$K_u = 1.3 \times 10^7$ erg/cm^3

J. Pauleve *et al*., J. Appl. Phys. **39**, 989 (1968).



meteorite

$L1_0$-FeNi fabricated by alternate monatomic layer deposition



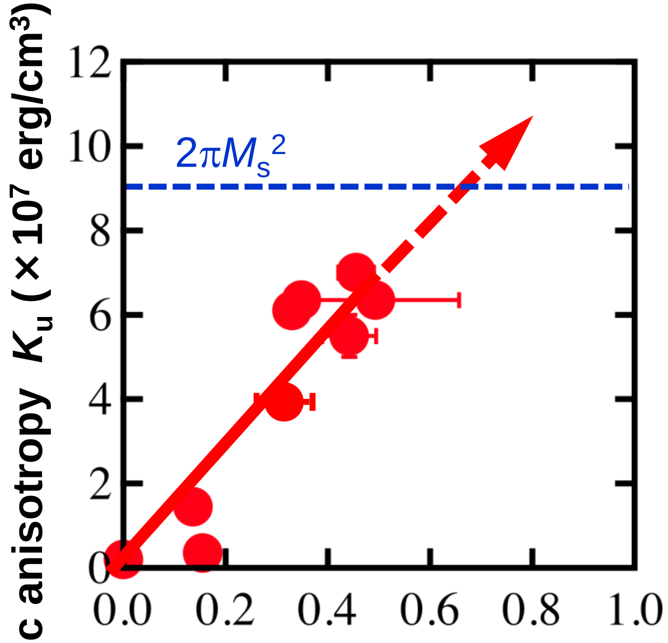
$[Fe(1\ ML)/Ni(1\ ML)]_{50}$

Buffer
(Cu-Au-Ni)

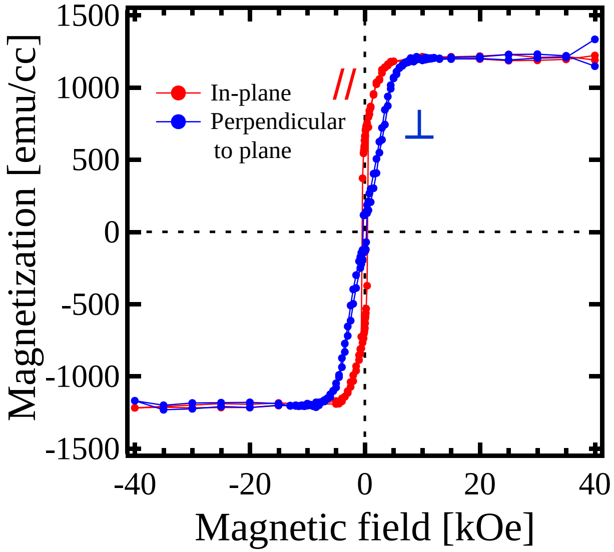
MgO(001) sub.

- Optimization of growth temperature
- Optimization of buffer

  Lattice matching
  Surface flatness
  Nonmagnetic



GI-XRD
(SPring-8 BL46XU)



S = 0.5
K_u= 7×10^6 erg/cm^3

T. Kojima, KT, et al., JJAP, 52 (2012) 010204.

Target : S > 0.9
K_u > 10^7 erg/cm^3
(perpendicularly magnetized)

Summary

Spin current and spintronics

- Recent progress of research on pure spin current

  Spin Hall effect → ***Enhancement by skew scattering***

  Spin pumping

  Spin Seebeck effect, etc.

- Materials for spintronics

  Half-metallic Heusler alloys (Co_2MnSi)

  → ***Enhanced CPP-GMR***

  High magnetic anisotropy $L1_0$-ordered alloys (FePt)

  → ***Perpendicular spin polarizer***

  Magnetization switching

  Noble metal free → FeNi