Advanced Spintronic Materials: for Generation and Control of Spin Current

Koki Takanashi
Magnetic Materials Laboratory
Institute for Materials Research (IMR)
Tohoku University
Sendai, Japan
• IEEE Magnetics Society Home Page: www.ieemagnetics.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
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• 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/Education Staffs
16,000 Students
10 Faculties
18 Graduate Schools
6 Research Institutes
Institute for Materials Research (IMR)
Tohoku University

Founded in 1907

Honda Memorial Hall

Kotaro Honda
1st Director
KS magnet (1917)

KIN KEN

Founded in 1916

We will have a centennial anniversary in 2016!
First experimental evidence for magnetocrystalline anisotropy

**Figure 30** Magnetization curves for single crystals of iron, nickel, and cobalt. From the curves for iron we see that the [100] directions are easy directions of magnetization and the [111] directions are hard directions. The applied field is $B_a$. (After Honda and Kaya.)

*C. Kittel, “Introduction to Solid State Physics”*
Institute for Materials Research (IMR)
Tohoku University

Founded in 1907

Kotaro Honda
1st Director
KS magnet (1917)
President of Tohoku Univ. (1931 – 1940)

Honda Memorial Hall

Founded in 1916

Presently,
120 Research Staffs
200 Students
27 Laboratories
9 Research Centers

We will have a centennial anniversary in 2016!
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)
    Jinhyeok Kim (Korea)
+ 8 MC students

Collaborators
Seiji Mitani (NIMS, Tsukuba)
Toshiyuki Shima (Tohoku-gakuin Univ.)
Masato Kotsugi (Spring-8)
Sadamichi Maekawa, Eiji Saitoh, Gerrit Bauer (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?

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

A concept that has attracted much attention in recent years

\[ J_s = J_\uparrow - J_\downarrow \]

Origin of magnetism
Angular momentum

\( S \)

\( e^- \)

Origin of electricity

\[ J_e = J_\uparrow + J_\downarrow \]

Conserved

Charge

\( \rightarrow \) Electric current

Not conserved

Spin

\( \rightarrow \) Spin current

Electron
What is spin current?

Generation

Spin current

Spin relaxation
Spin diffusion

Transformation

Annihilation

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)

\[
J = J_\uparrow - J_\downarrow
\]

Spin current

Spin diffusion length

10 nm ~ 100 nm ~ 1 µm
Example of spin current -2

- Without electric current (pure spin current)

With electron motion

\[ J_e = J^\uparrow + J^\downarrow = 0 \]

Without electron motion

**Non-local spin injection**

**Spin Hall effect**

**Spin pumping**

**Spin Seebeck effect, etc.**

Spin waves

(magnon spin current)

Magnetic insulator
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

**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

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

At the award ceremony (Dec. 10, 2007)

On the Noble lecture (Dec. 8, 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**

   - Upper electrode
   - Lower electrode
   - Interlayer or Insulator: Tunnel magnetoresistance (TMR)
   - Metal: Giant magnetoresistance (CPP-GMR)

   \[ \text{Magnitude of MR: } \frac{\Delta R}{R} \propto P_A \cdot P_B \]

   - \( P_{A(B)} \): spin polarization of conduction electrons in A (B)
Typical device structures in spintronics

2. Lateral structure type

Applications: spin transistor, etc.

Non-local geometry

Key: efficiency of Spin injection from ferromagnetic metal (FM) to nonmagnetic metal (NM)

control of Spin relaxation

\[ J_e = J_{\uparrow} + J_{\downarrow} = 0 \]
\[ J_s = J_{\uparrow} - J_{\downarrow} \neq 0 \]
Research on *pure* spin current

**Generation of pure spin current**
- Non-local spin injection (electric current $\rightarrow$ spin current)
- Spin Hall effect (electric current $\rightarrow$ spin current)

![Diagram of Direct spin Hall effect and Inverse spin Hall effect](image)

$J_q$: Charge current (positive charge)

$J_s$: Spin current (Flow of moment)

$\alpha_H$: Spin Hall angle ($=\sigma_{SH}/\sigma$)
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 $\rightarrow$ spin current)
- Spin Hall effect (electric current $\rightarrow$ spin current)
- Spin pumping (electromagnetic wave $\rightarrow$ spin current)
- Spin Seebeck effect (heat current $\rightarrow$ spin current)

![Diagram showing Ni$_3$Fe and Pt with spin Hall effect and temperature gradient](image)

**Inverse spin Hall effect**

**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


Spin Seebeck effect appears even in a magnetic insulator

Magnon spin current

Temperature difference dependence of spin voltage

LaY$_2$Fe$_5$O$_{12}$

Similar behavior to Ni$_3$Fe

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

Spatial distribution

Development of Spin Caloritronics Application to Energy Harvesting
Transmission of pure spin current

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


Transmission of pure spin current; metal $\rightarrow$ insulator $\rightarrow$ metal

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$_2$MnSi, Co$_2$Fe(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$_3$Fe$_5$O$_{12}$, etc.

- **Spin relaxation**
  
  Nanoparticles \( \rightarrow \) *size effect*

  Molecular / carbon-based materials \( \rightarrow \) *weak LS coupling*

  Magnetic insulator \( \rightarrow \) *low magnetization damping*
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- **Spin relaxation**

  Nanoparticles $\rightarrow$ *size effect*

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  Magnetic insulator $\rightarrow$ *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, \( \cdots \)
  \[ P = 0.4\sim0.5 \text{ typically} \]

- Half metals
  \[ P = 1 \]

Heusler alloys:
  NiMnSb, Co\(_2\)MnSi, Co\(_2\)MnAl, etc.

Transition metal oxides:
  CrO\(_2\), Fe\(_3\)O\(_4\), LSMO, etc.

→ Efficient spin injection
  High performance of spintronics devices
**Co$_2$MnSi (CMS)**

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

Heusler-MTJ

- Red circles: RT
- Blue triangles: LT

**Co$_2$MnSi/MgO/Co$_2$MnSi**
TMR ratio = 1995% @ 10K
= 330% @ RT
H-x Liu *et al.*, APL (2012)

**Co$_2$FeAl$_{0.5}$Si$_{0.5}$/MgO/Co$_2$FeAl$_{0.5}$Si$_{0.5}$**
Tezuka *et al.* (Tohoku Univ.)

**Co$_2$MnSi/Al-O/Co$_2$MnSi**
Sakuraba *et al.* (Tohoku Univ.)

**Co$_2$Cr$_{0.6}$Fe$_{0.4}$/Al-Al-O/CoFe**
Inomata *et al.* (Tohoku Univ.)

Heusler-based MTJ: Large temperature dependence of MR ratio is still a serious problem especially in CMS.
Giant TMR in MgO-MTJ

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

Band structure of Fe
Δ₁ band: half metallic nature

MR = 180% (RT)
247% (4.2K)

S. Yuasa et al.,
MTJs with half-metallic Heusler alloys

Heusler-based MTJ: Large temperature dependence of MR ratio is still a serious problem especially in CMS.
High MR and low resistance

Reported MR ratio in small RA region

Essential decrease in TMR with reducing resistance

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

CPP-GMR

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

Development of CPP-GMR for Heusler alloys
CoFeₓMn₁₋ₓSi(20)/Ag(5)/CoFeₓMn₁₋ₓSi(7)

CoFe₀.₄Mn₀.₆Si(4)/Ag(3)/CoFe₀.₄Mn₀.₆Si(2)

Best composition ratio: CoFe₀.₄Mn₀.₆Si

<table>
<thead>
<tr>
<th>Average MR ratio</th>
<th>RA</th>
<th>ΔRA</th>
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<tbody>
<tr>
<td>48 %</td>
<td>24.3 mΩ·µm²</td>
<td>11.8 mΩ·µm²</td>
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Large MR ratio even in very thin trilayer structure!
High MR and low resistance

Reported MR ratio in small RA region

Essential decrease in TMR with reducing resistance

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

MR ratio (%)

R×A (Ω µm²): resistance area product

TMR

CPP-GMR
Rf oscillation in Heusler alloys by spin transfer torque

Non-local spin injection in lateral spin valves

\[ \text{Co}_{2}\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si(CFMS)/Cu} \]

Y. Sakuraba et al., unpublished.

NLSV signal@RT, gap = 350 nm

\[ \Delta R_S \sim 4.8 \text{ m\ensuremath{\Omega}} \]

\[ \Delta R_S \sim 0.1 \text{ m\ensuremath{\Omega}} \]

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 $\rightarrow$ Thermal stability of magnetization

Negative shape anisotropy $\rightarrow$ Easy magnetization switching

No restriction on aspect ratio

Examples of perpendicularly magnetized films

Co-based granular films such as CoCrPt-SiO$_2$
RE-TM amorphous alloy films such as TbFeCo
Metallic multilayers or ultrathin films such as Ni/Co, Co/Pd, CoFeB/MgO, etc.
$L1_0$ ordered alloy films such as FePt, FePd, CoPt, etc.
Phase diagram of Fe-Pt system

**L1\textsubscript{0} ordered FePt alloy**

- **L1\textsubscript{0} ordered phase**

![Phase diagram of Fe-Pt system](image)

- Large uniaxial magnetic anisotropy
  \[ K_u = 7 \times 10^7 \text{ erg/cm}^3 \]
  - Perpendicular magnetic recording media
  - Patterned media
  - Spintronics

- **Fe\textsubscript{3}Pt**
- **FePt**
- **FePt\textsubscript{3}**

- Pt concentration, \( c_{\text{Pt}} \) (mol\%)
- Temperature, \( T \) / °C

- 3.836 Å
- 3.714 Å
- c-axis

- Intersection points:
  - 1519°C (δFe)
  - 700°C (γFe, Pt)
  - 550°C (αFe)
  - 1300°C
  - 1350°C

- Phase diagram of Fe-Pt system
Spin-torque switching of magnetization for $L1_0$-FePt

Fully epitaxial

FePt / Au / FePt CPP-GMR pillars
Spin-torque switching of magnetization


$[\text{Co/Pt}]_4 / [\text{Co/Ni}]_2 / \text{Cu} / [\text{Co/Ni}]_4$

$[\text{CoFe/Pt}]_5 / \text{Cu} / [\text{CoFe/Pt}]_7$
Coercivity control by electric field for $L1_0$-FePt

Perpendicularly magnetized $L1_0$-FePt

FePt / MgO / Al-O Hall device

Anomalous Hall resistance curve

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

Spin wave-assisted magnetization switching


**FePt / Permalloy (Py) Exchange-Coupled Bilayer**

Utilization of Perpendicular Standing Spin Wave Mode in the Bilayer
Spin wave-assisted magnetization switching

Without spin wave excitation, \( H_{sw} \sim 1900 \text{ Oe} \).
Spin wave-assisted magnetization switching

$t_{Py} = 100 \text{ nm} \ (H_{rf} = 145 \text{ Oe})$
Spin wave assisted magnetization switching

Time evolution of magnetic structure by micromagnetics simulation

Time Evolution of Magnetic Structure

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


Perpendicularly magnetized FePt injector

Au Hall cross

Spin Hall angle $\alpha_H \sim 0.1$

Electrical detection of giant spin Hall effect at room temperature
Theoretical discussion

G.Y. Guo, S. Maekawa, and N. Nagaosa

Spin Hall Effect by Kondo singlet state

**Orbital selective Kondo**

\[ e_g \text{ Kondo limit} \rightarrow T_K \approx 0.4K \]
\[ t_{2g} \text{ Mixed valence } d^6 \text{ and } d^7 \]

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: $\alpha_H = 0.05$


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


Otani’s group (Univ. Tokyo)

Ir-doped Cu: $\alpha_H = 0.02$


Bi-doped Cu: $\alpha_H = 0.24$


Ralph’s group (Cornell Univ.) Mechanism?

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

**L1₀ ordered alloy and element strategy**

**FePt, FePd, CoPt, etc.**

High uniaxial magnetic anisotropy

\[ K_u = 10^7 \sim 10^8 \text{ erg/cm}^3 \]

Spintronics

Magnetic storages

Permanent magnets

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

Expensive

High damping constant

**Requirement for a noble-metal-free L1₀ ordered alloy**
$L1_0$ ordered FeNi alloy

Order-disorder transformation temperature $\sim 320^\circ C$

Requires annealing for an astronomically long time
Naturally found only in meteorites

Neutron irradiation: $K_u = 1.3\times10^7$ erg/cm$^3$

$L1_0$-FeNi fabricated by alternate monatomic layer deposition

- Optimization of growth temperature
- Optimization of buffer
  - Lattice matching
  - Surface flatness
  - Nonmagnetic

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

**GI-XRD** (SPring-8 BL46XU)

**Equation**:

$$2\pi M_s^2$$

**Results**:

- $S = 0.5$
  - $K_u = 7 \times 10^6$ erg/cm$^3$

**References**:
- $\alpha = 0.013$ ($L1_0$)
- $0.009$ (disordered)
  (in collaboration with Prof. Mizukami’s group.
  unpublished)
Summary

*Spin current and spintronics*

- Recent progress of research on pure spin current
  - Spin Hall effect
  - Spin pumping
  - Spin Seebeck effect, etc.

- Materials for spintronics
  - Half-metallic Heusler alloys (Co$_2$MnSi)
    - *Enhanced CPP-GMR*
  - High magnetic anisotropy $L1_0$-ordered alloys (FePt)
    - *Perpendicular spin polarizer*
    - *Magnetization switching*
    - *Noble metal free → FeNi*