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Recent progress in large tunnel magnetoresistance junctions

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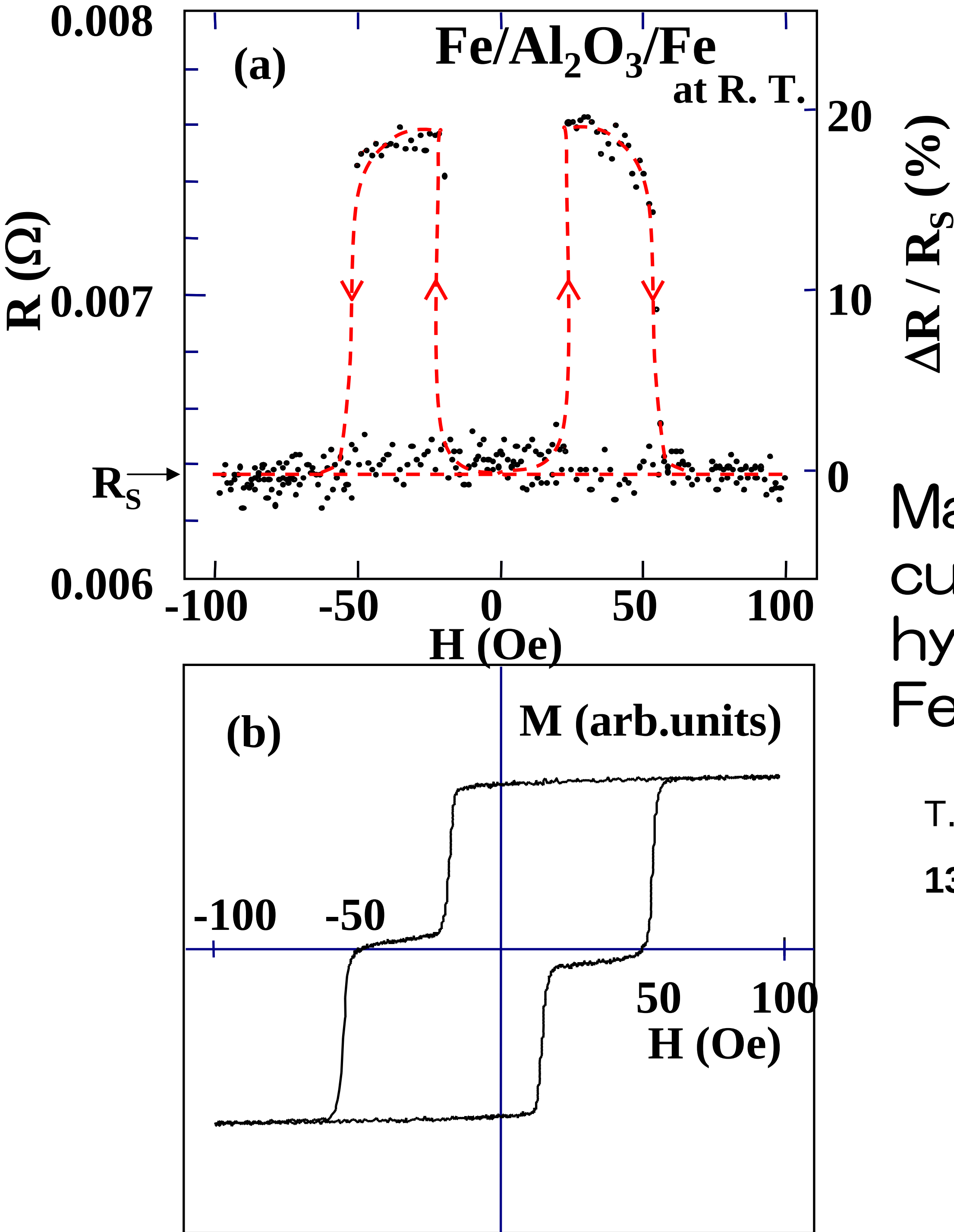
1. **A little review of MTJs**
2. **Heusler electrode TMR junctions**
   - **Relationship between degree of order and TMR ratio**
   - **Temperature and bias dependences of TMR ratio**
3. **Applications of TMR junctions (Gilbert damping constant α)**

1. A little review of MTJs

History of TMR research

(Typical reports)

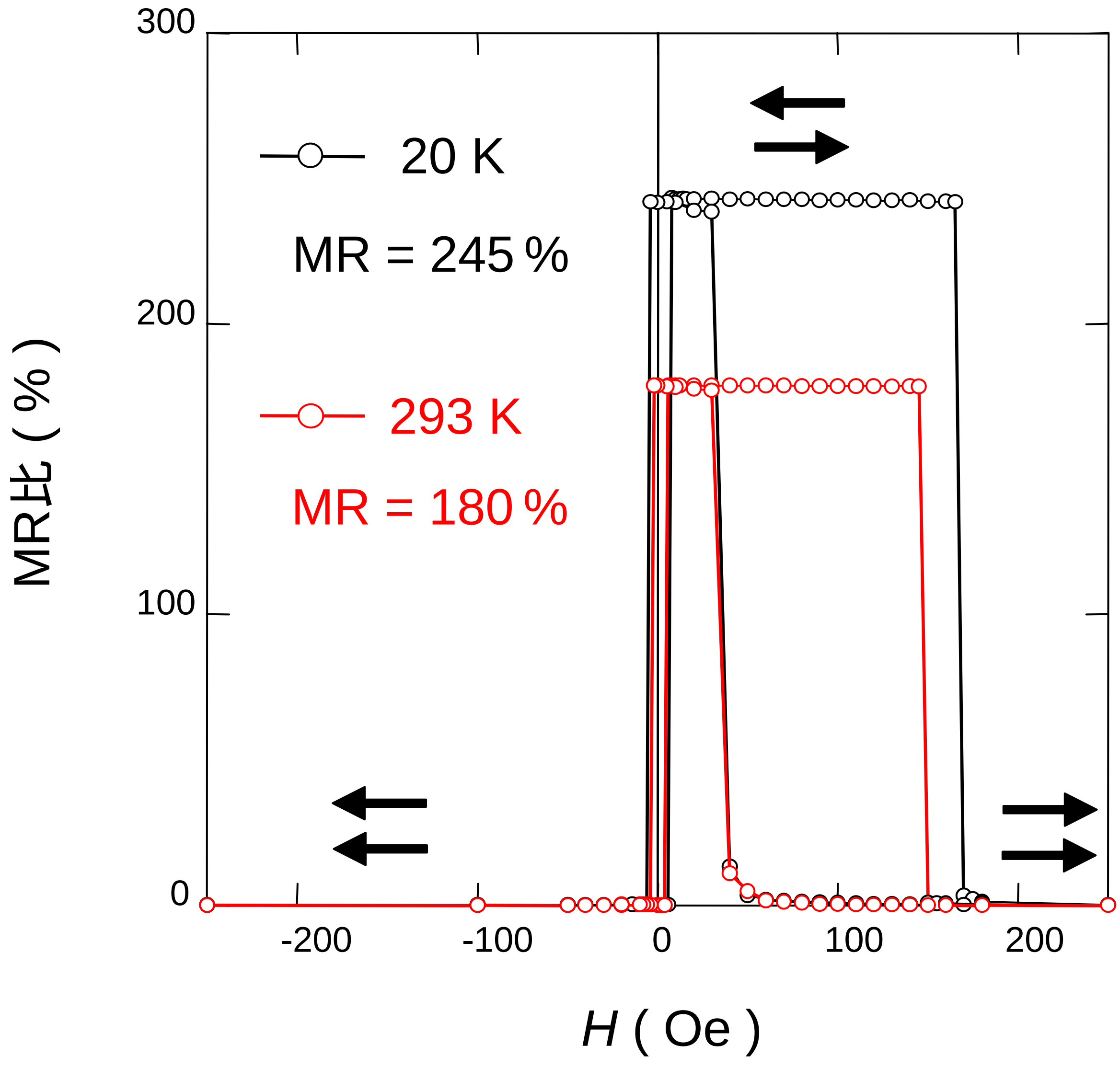
- Julliere (Phys. Lett., **54A**,225(1975)) Gd-O
- T.Miyazaki et al. (J.Magn.Magn.Mater.139,**L231**(1995)) Al-O
- J.S.Moodera et al. (Phys.Rev.Lett., **74**, 3273(1995)) Al-O
-
- S.Yuasa et al. (Jpn.J.Appl.Phys., **43**, L588(2004)) MgO
- S.S.P.Parkin et al. (Nature Materials, **3**, 863(2004)) MgO
-
- Y.Sakuraba et al. (Appl.Phys.Lett., **88**, 192508(2006)) Heusler
- N.Tezuka et al. (Appl.Phys. Lett.,**89**, 252508-1(2006)) Heusler
- T.Ishikawa et al. (Appl.Phys.Lett.,**89**,192505-1(2006)) Heusler



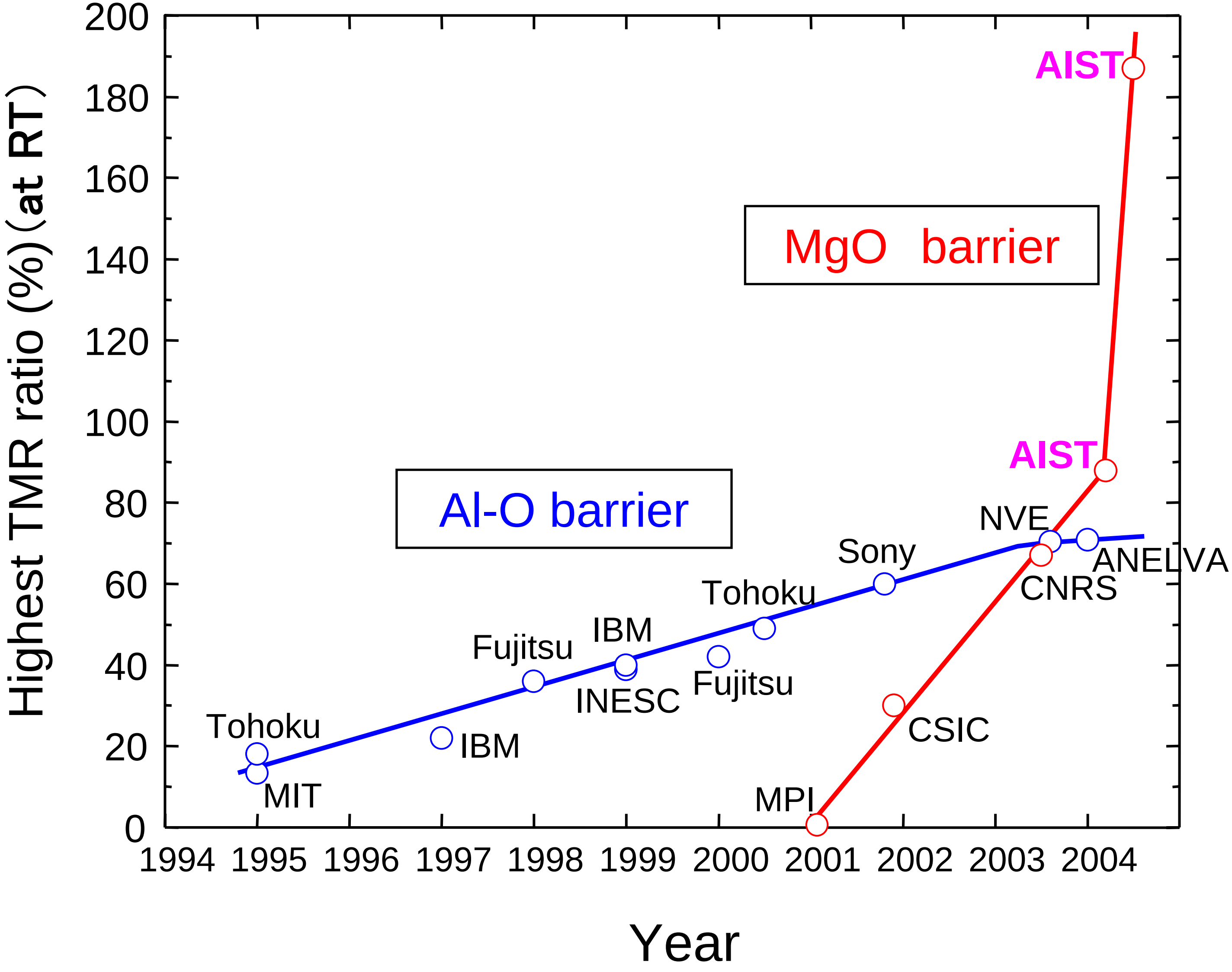
Magnetoresistance curve (a) and magnetic hysteresis loop (b) for $Fe/Al_2O_3/Fe$ junction

T.Miyazaki and N.Tezuka, J.3M, **139**,L2341(1995).

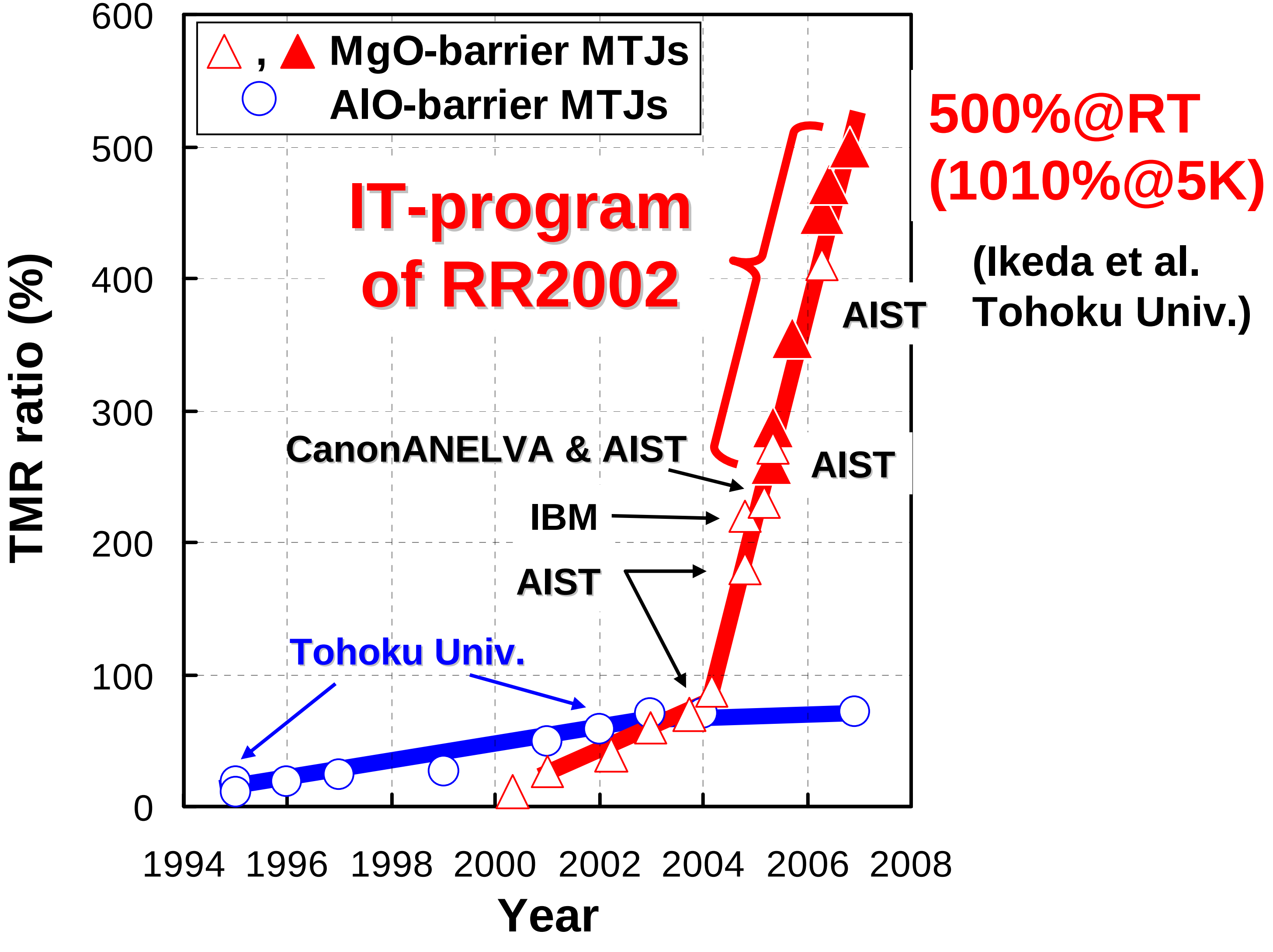
Fe(001)/MgO(001)/Fe(001)



Yuasa et al. Nature Materials 3 (2004).



Highest TMR ratio (%) (at RT)
Year
200
180
160
140
120
100
80
60
40
20
0
1994
1995
1996
1997
1998
1999
2000
2001
2002
2003
2004
MgO barrier
Al-O barrier
AIST
AIST
NVE
ANELVA
CNRS
Sony
Tohoku
IBM
Fujitsu
INESC
Fujitsu
CSIC
Tohoku
IBM
MIT
MPI



MgO-barrier MTJs
AlO-barrier MTJs
500%@RT
(1010%@5K)
IT-program
of RR2002
(Ikeda et al.
Tohoku Univ.)
AIST
CanonANELVA & AIST
AIST
IBM
AIST
Tohoku Univ.
TMR ratio (%)
Year
0
100
200
300
400
500
600
1994
1996
1998
2000
2002
2004
2006
2008

2. TMR in Heusler alloy electrode junction

Kristallstruktur und Ferromagnetismus der Mangan–Aluminium–Kupferlegierungen[1])

Von Otto Heusler

(Mit 20 Figuren)

Inhalt: A. Problemstellung. — B. Experimentelles. — C. Röntgenographische Strukturbestimmung. — D. Messungen des elektrischen Widerstandes. — E. Magnetische Messungen. — F. Versuch zu einer Deutung des Ergebnisses. — G. Zusammenfassung.

A. Problemstellung

Seit es Heisenberg[2]) gelungen ist, eine quantenmechanische Deutung des Ferromagnetismus zu geben, ist der Zusammenhang zwischen Kristallstruktur und Ferromagnetismus zu einem entscheidenden Problem für den weiteren Ausbau der Theorie geworden. Denn Heisenberg zeigte, daß neben der Elektronenkonfiguration der beteiligten Atome in erster Linie die Kristallstruktur maßgebend für das Zustandekommen dieser Erscheinung ist. Welcher Art diese strukturellen Vorbedingungen im einzelnen sind, ist bislang noch nicht hinreichend erfaßt. Es ist daher eine wichtige Aufgabe, diese Frage auf experimentellem Wege anzugreifen. In diesem Zusammenhang dürfen die an der Grenze des ferromagnetischen Erscheinungsgebiets stehenden Heuslerschen Legierungen, die aus unmagnetischen Komponenten aufgebaut sind, einiges Interesse beanspruchen.

Besonders wichtig sind hier die sogenannten Alterungserscheinungen, die schon von Anfang an die Aufmerksamkeit der Forscher auf sich lenkten: Offenbar ganz geringfügige Änderungen der Kristallstruktur, die sich durch thermische

1) Der erste Teil der vorliegenden Arbeit (röntgenographische Strukturbestimmung) wurde am 28. Januar 1933 in der Sitzung des Gauvereins Hessen der D. Phys. Ges. in Darmstadt vorgetragen (Referat Verh. D. Phys. Ges. [3] **14**. S. 7. 1933), der zweite Teil (elektrische und magnetische Messungen) am 27. Mai 1933 auf der Tagung der D. Bunsengesellschaft in Karlsruhe (Referat Ztschr. Elektrochem. **39**. S. 645. 1933).

2) W. Heisenberg, Ztschr. f. Phys. **49**. S. 619. 1928.

Annalen der Physik Band 19 (1934) 155-201.

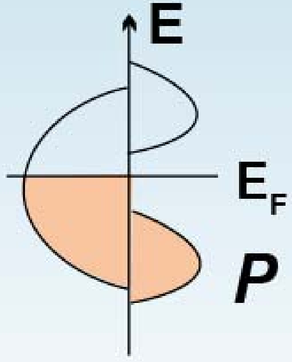
Functions of Heusler Compounds

1905	• Magnetic material:	Cu_2MnAl
1983	• Halfmetallic ferromagnet:	NiMnSb
	• Magneto-optical:	PtMnSb
	• Magneto-mechanic:	Ni_2MnGa
	• Superconductor:	Pd_2YSn
	Semiconductor:	CoTiSb
	• Heavy fermion:	Fe_2VAl
	• Li-conductor:	LiMnSb
2001	• Magneto-electronic:	Co_2FeSi
	• Thermo-electric:	TiNiSn
	• Magneto-caloric:	CoMnSb:Nb
	▪ **Spintronics Material :**	**Co_2MnSi**

From
Prof. Felser

MTJs with Half-metallic electrode

- **Half-metals (Heusler alloys, $La_{2/3}Sr_{1/3}MnO_3$, Fe_3O_4 *etc.*)**



LSMO/STO/LSMO MR ~ 0 % (RT), 1800% (4 K)

LSMO T_C = 370 K **P = 0.95**

M. Bowen et al. Appl. Phys. Lett. ***82*** *(2003) 233*

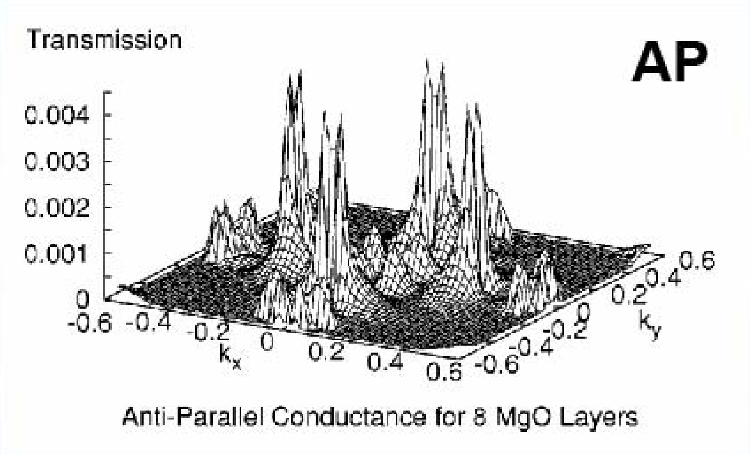
Fe/MgO/Fe,CoFeB/MgO/CoFeB-MTJ

Finite tunnel conductance in AP

⇒ P < 1,Maximum TMR ~ 1200%

Butler et al. PRB ***63*** *056614 (2001)*

Mathon & Umerski PRB ***63*** *220403 (2001)*



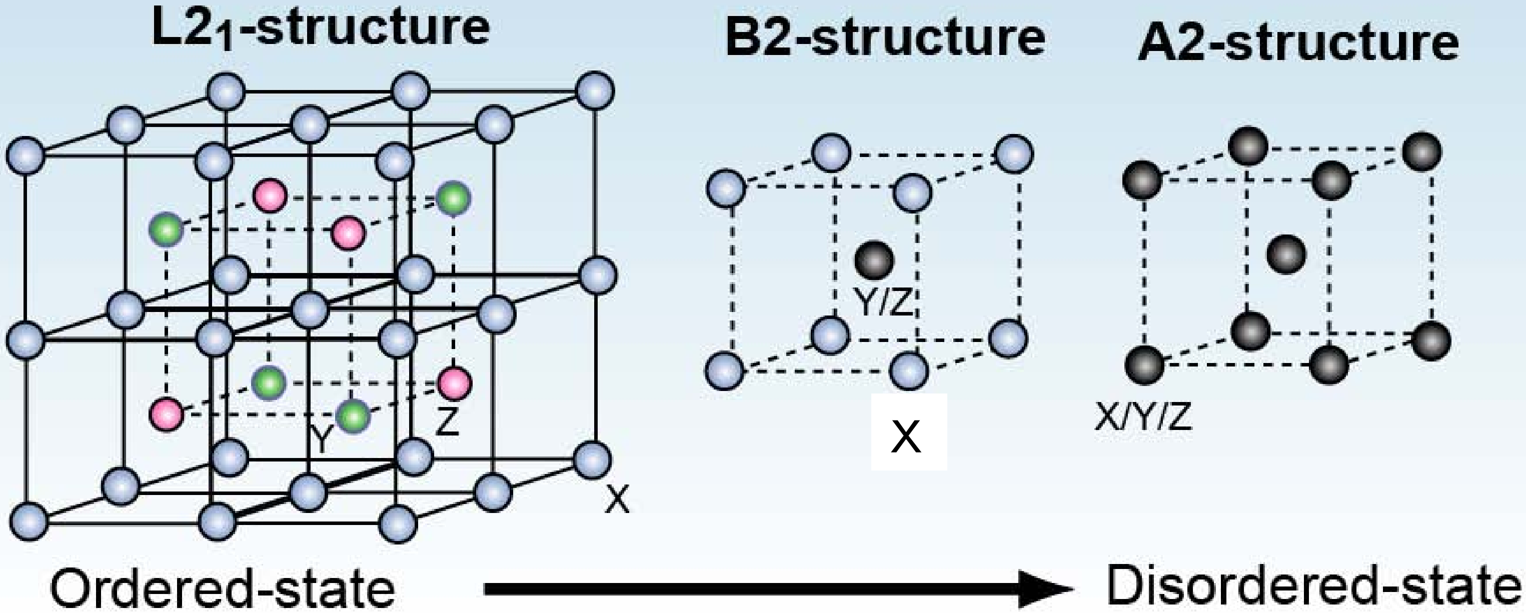
Half-metallic full-Heusler alloys : Co_2MnSi,Co_2MnGe, etc.

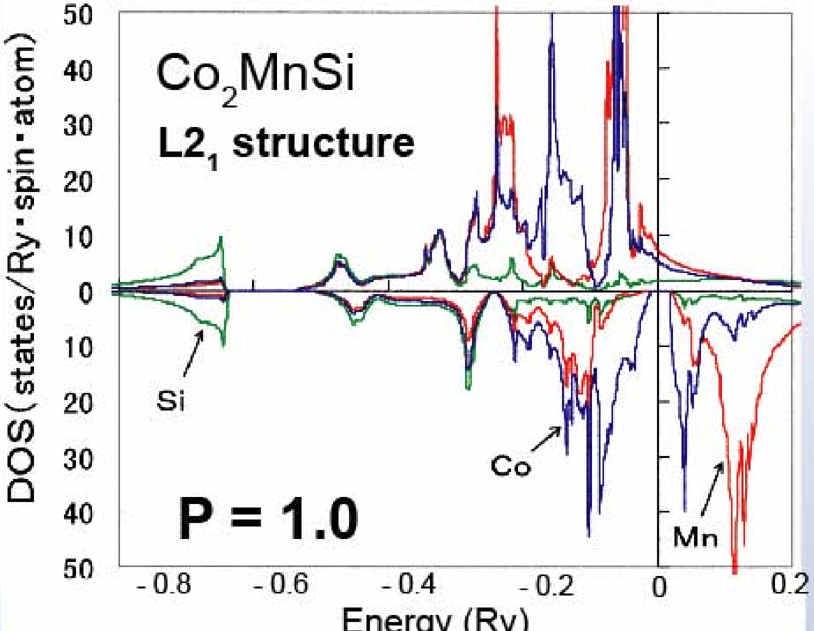
★High T_C well above RT ⇒ large spin-polarization even at RT

★ High spin injection efficiency *g*, Small damping constant α

⇒ reduce J_c in spin-transfer switching

Half-metallic full-Heusler alloy : X_2YZ





Co_2MnSi (CMS)

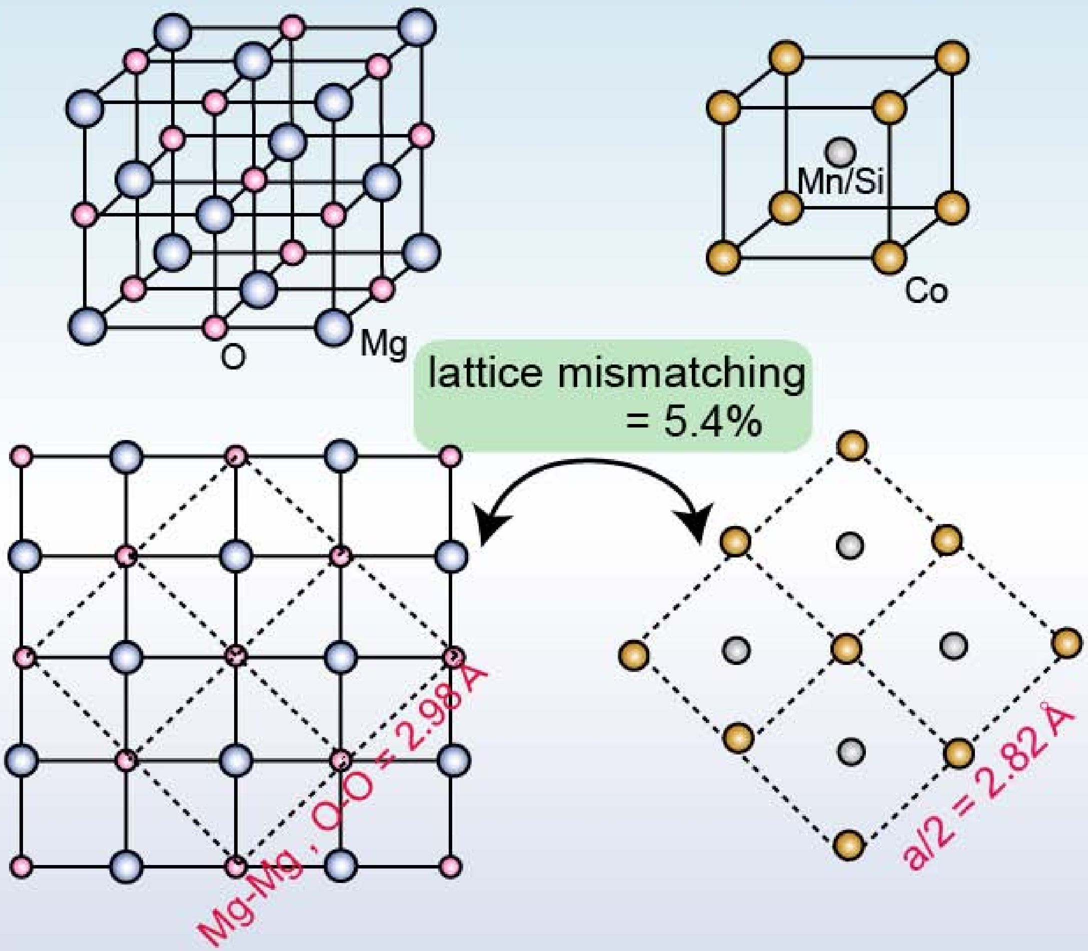
- Half-metallic band gap : 400 - 600 meV
- *L*2_1-structure is easily obtained by annealing process
- High T_C (~ 985 K)

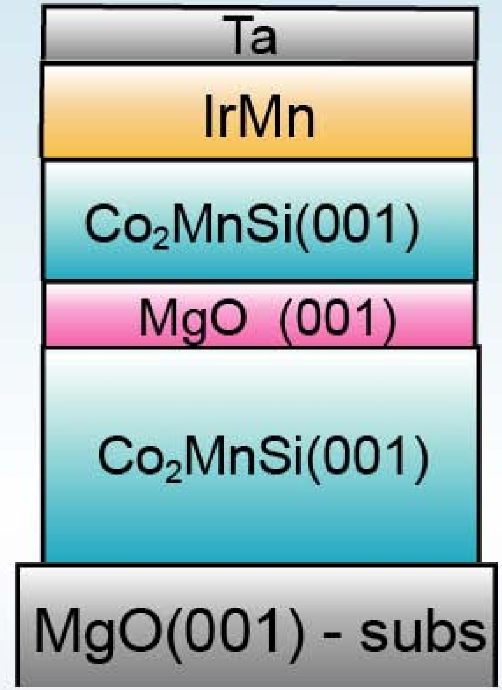
⇒ ***Co_2MnSi is expected as the most promising HMF***

Introduction - *Epitaxial relation between MgO and Co_2MnSi* -

MgO (NaCl-type) a = 4.22 Å

Co_2MnSi a = 5.65 Å



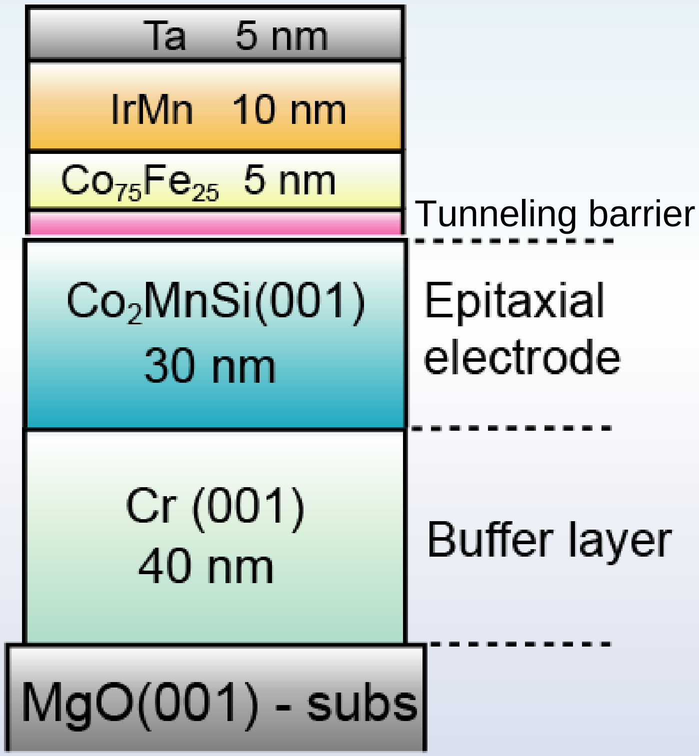


Full-epitaxial MTJ can be fabricated on MgO (001) single crystalline substrate.

Epitaxial relation:
$Co_2MnSi(001)[100]$ //$MgO(001)[110]$

Experimental method

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



Stacking structure

MTJ

Al-O barrier : O_2 plasma oxidation of Al layer

MgO barrier : rf-sputtering from MgO target

micro-fabrication : photolithography & Ar ion milling

(element size : 5 x 5 ~ 100 x 100 μm^2)

Annealing : 200 – 300℃ at 300 Oe

MR, I-V curve : DC 4-probe method

G-V curve : AC lock-in amplifer technique

Co_2MnSi bottom epitaxial electrode

Target : $Co_{2.00}Mn_{1.28}Si_{1.30}$ ⇒ Film : $Co_{2.00}Mn_{1.00}Si_{1.08}$ (ICP)

Deposited @ RT ⇒ Post-annealed : T_a = 250 - 650℃

Crystal structure, $L2_1$ ・ B2 ordered level : XRD

Magnetization : SQUID

Surface image : AFM

Cr buffer layer

Substrate temp : T_S, Post-annealing temp : T_{ann}

Result 1 - *Epitaxial bottom Co_2MnSi electrode* -

XRD (θ-2θ scan)

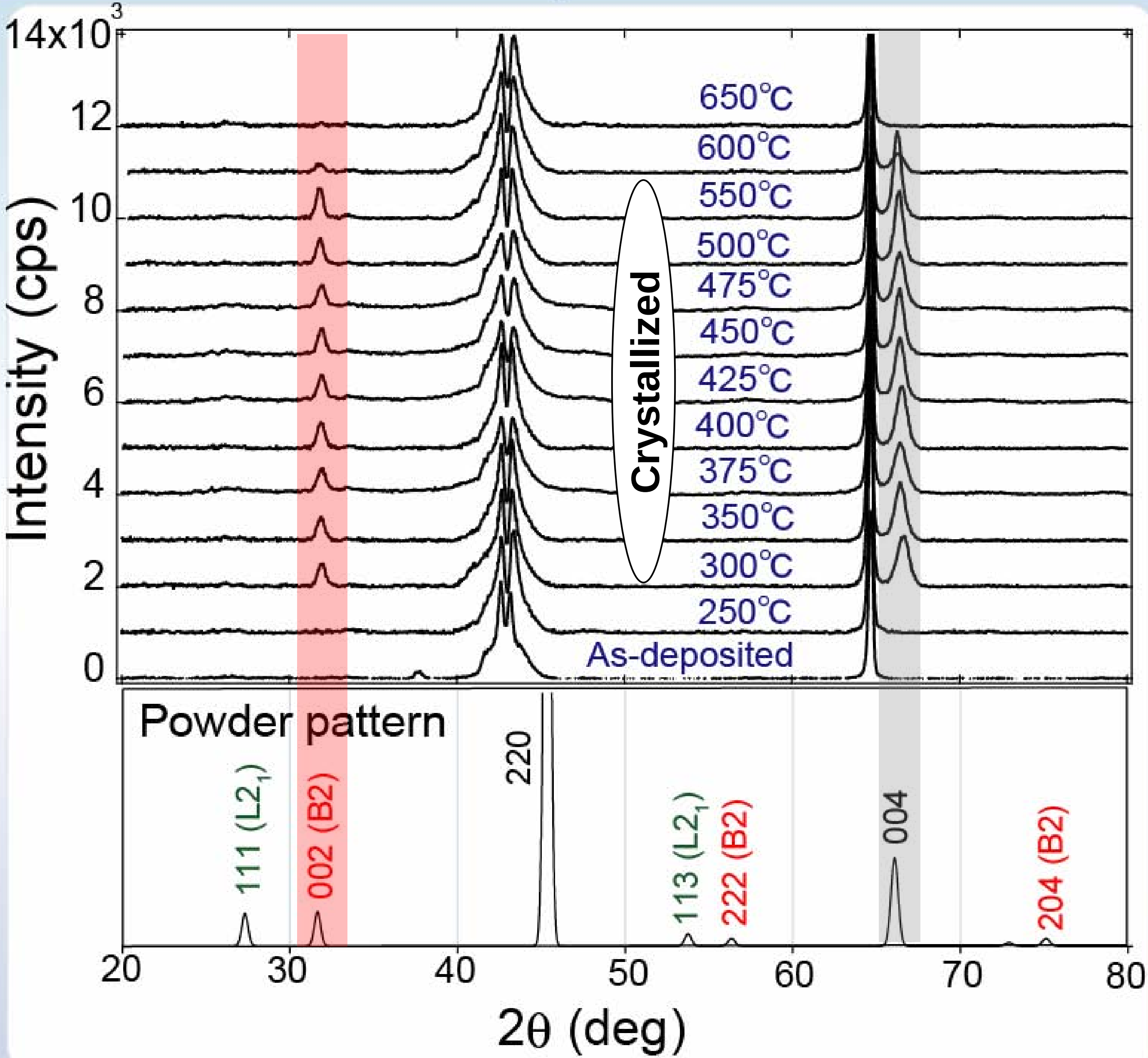
- **Superlattice peaks**

  $L2_1$-structure
  h,k,l : all odds

  B2-structure
  h+k+l = 4n+2

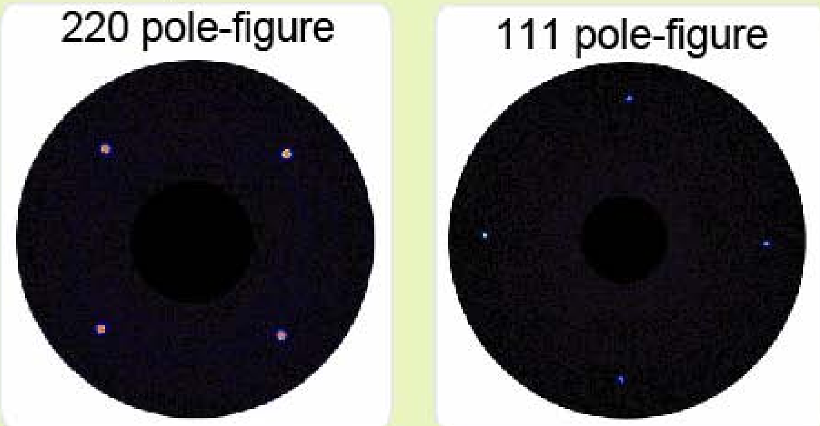
- **Fundamental peaks**

  A2-structure
  h+k+l = 4n

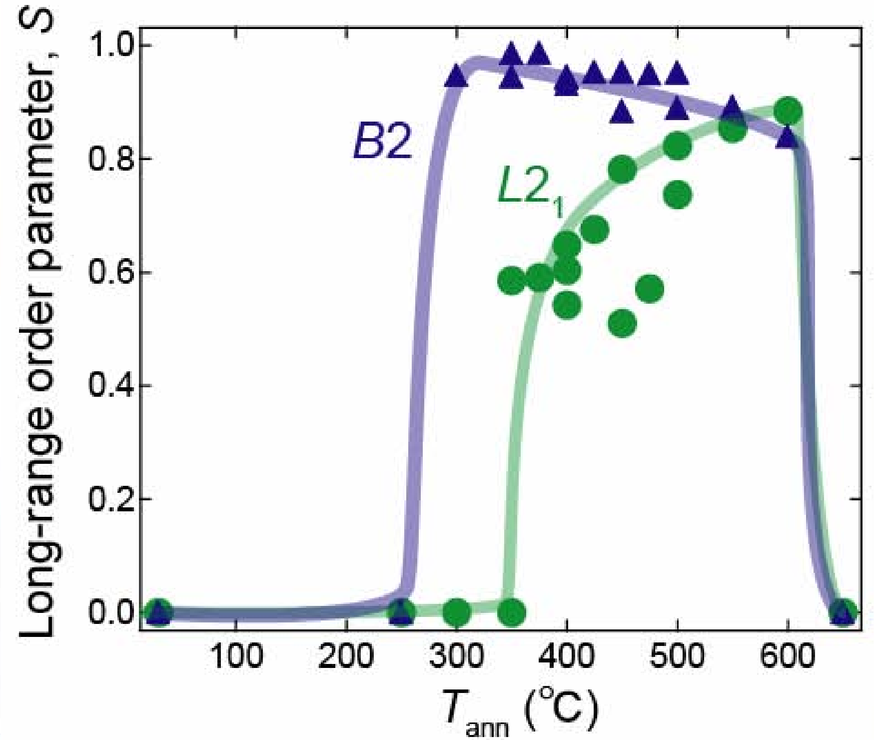


Result 1 - Epitaxial bottom Co_2MnSi electrode -

$L2_1$-order: I(111)/I(220)



BRUKER DISCOVER D8 / 2 dimentional detector



Long-range order parameter, S

$$S_{B2}^2 = \frac{I_{obs}(200)/I_{obs}(400)}{I_{cal}(200)/I_{cal}(400)}$$

$$S_{L2_1}^2 = \frac{I_{obs}(111)/I_{obs}(220)}{I_{cal}(111)/I_{cal}(220)}$$

$$I_{cal} = I_{sim} \cdot m^{-1} \cdot G_t$$

m^{-1}: **multiple factor**; G_t: **Film thickness factor**

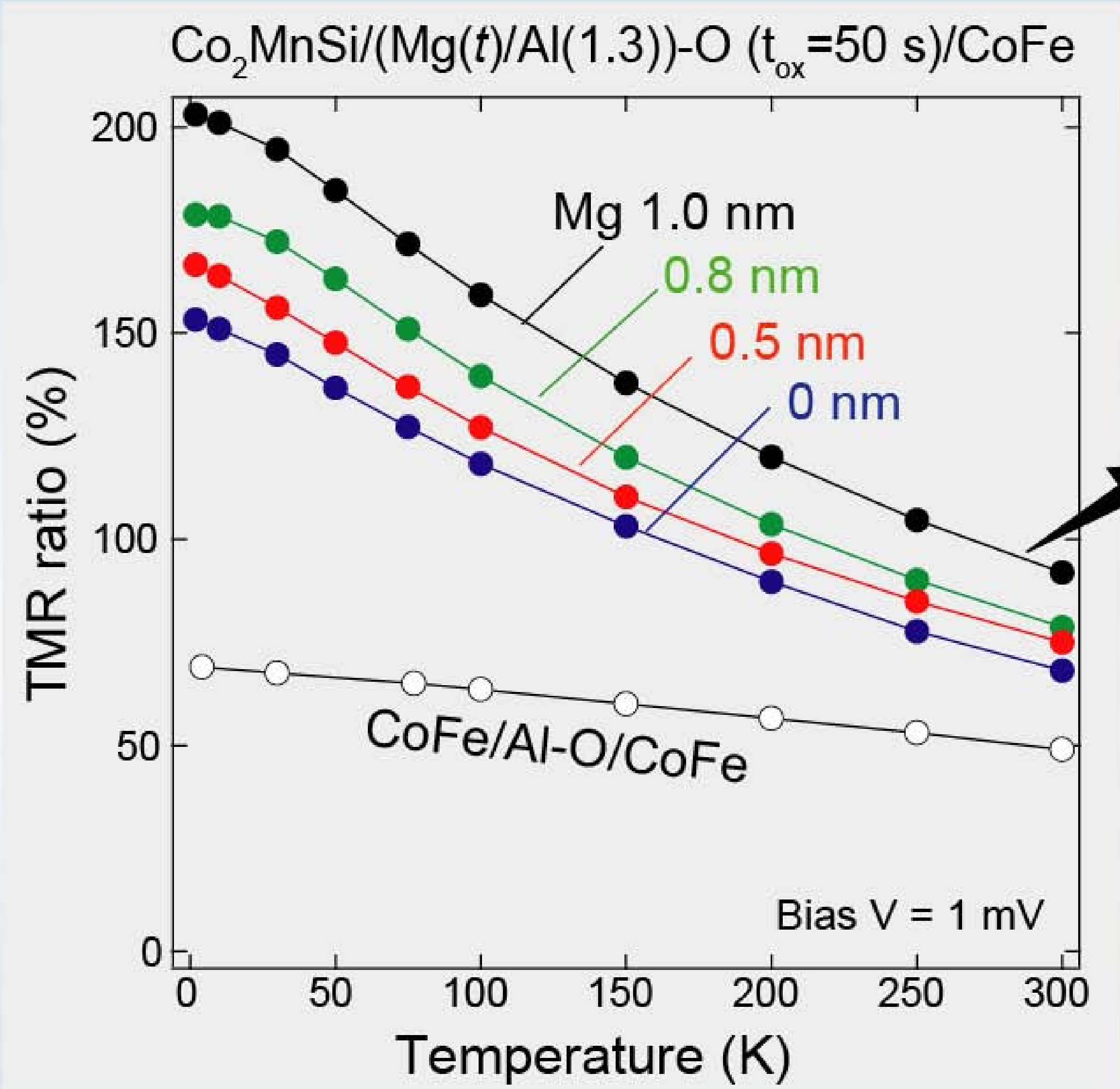
$$G_t = 1 - \exp\left(\frac{-2\mu t}{\sin\theta}\right)$$

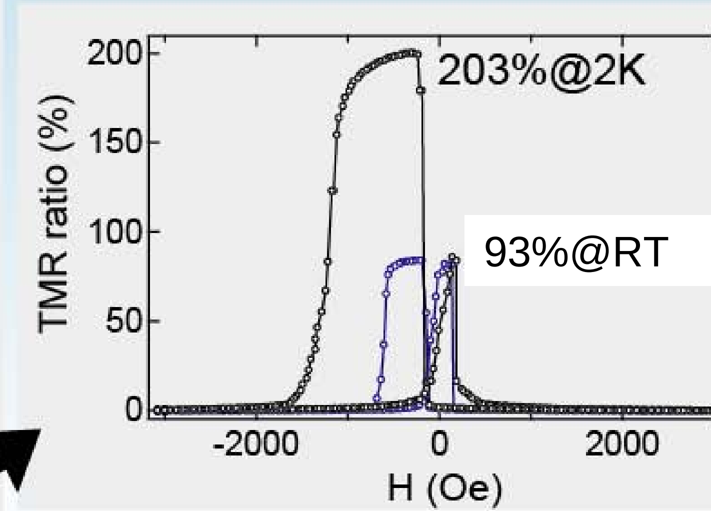
B2-order : T_{ann} > 300°C, S ~ 1
$L2_1$-order : T_{ann} = 600°C, S ~ 0.9

We successfully fabricated highly ordered Co_2MnSi

Sakuraba et al. Jpn. J. Appl. Phys. **44** 6536 (2005)

Result 2: Co_2MnSi/(Mg/Al)-O/CoFe - MTJs





CMS/(Mg/Al)-O/CoFe

TMR ~ 93% @RT

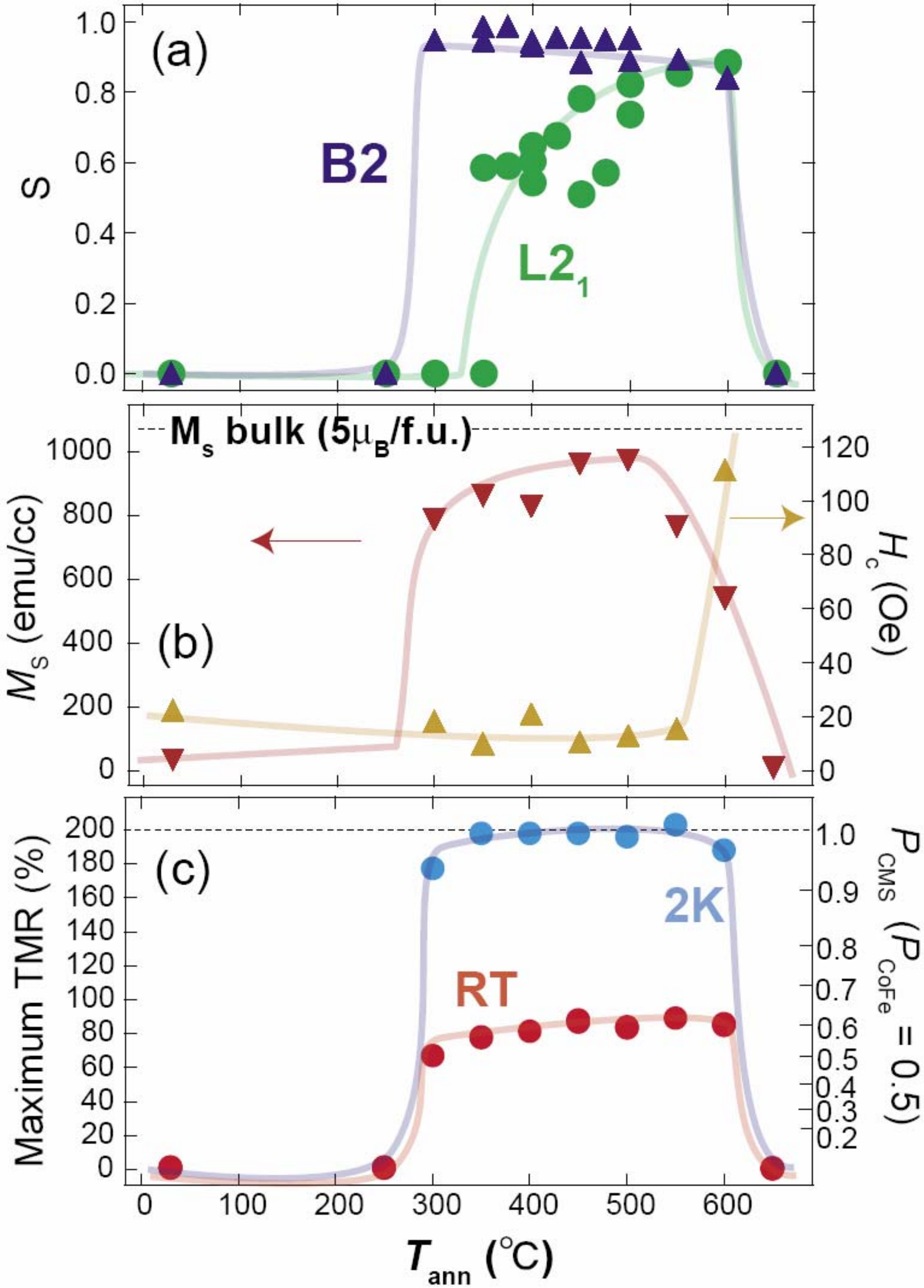
~ 203% @2K

⇒ P_{CMS} **~ 1.0** (P_{CoFe} ~ 0.50)

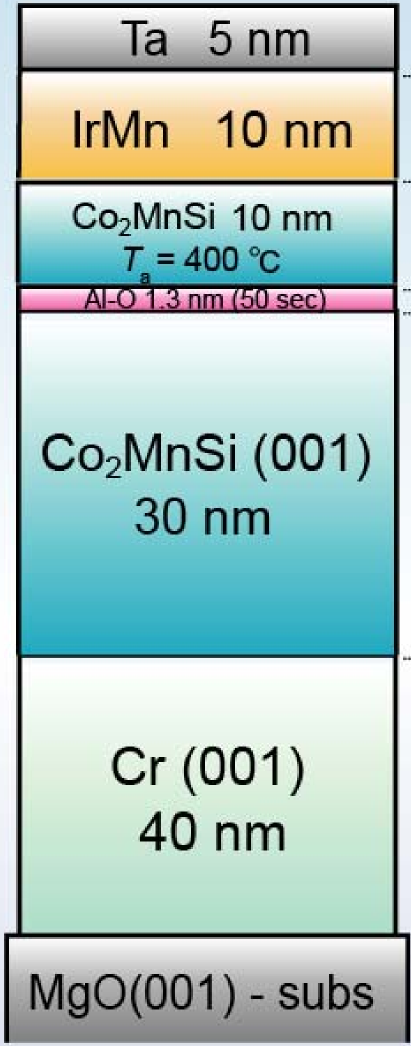
↓

Ideal half-metallic spin-polarization

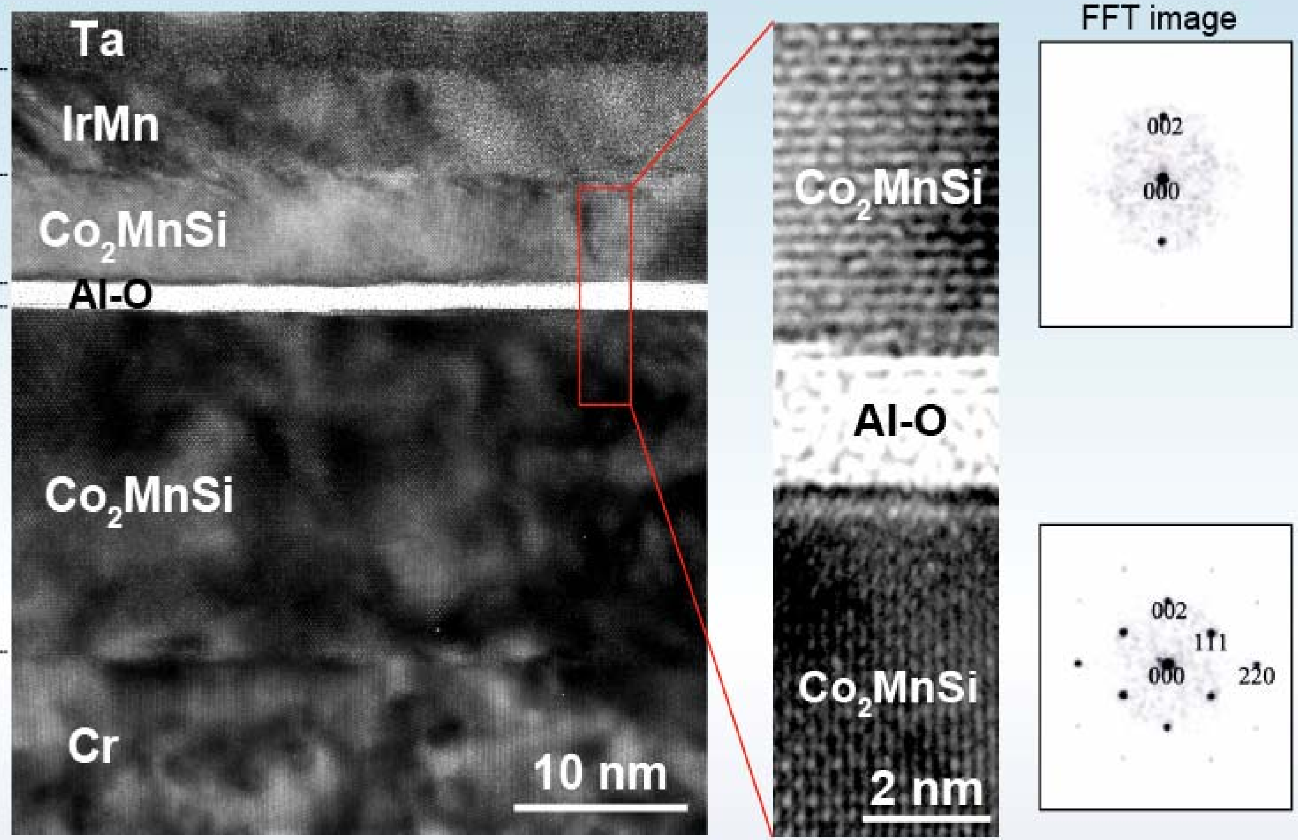
Giant TMR ratio over 1000% at LT is possible in CMS/(Mg/Al/Mg)-O/CMS structure


(a)
B2
$L2_1$
S
M_s bulk ($5\mu_B$/f.u.)
(b)
M_S (emu/cc)
H_c (Oe)
(c)
Maximum TMR (%)
2K
RT
P_{CMS} (P_{CoFe} = 0.5)
T_{ann} (°C)

MTJ - Co_2MnSi/Al-O/Co_2MnSi

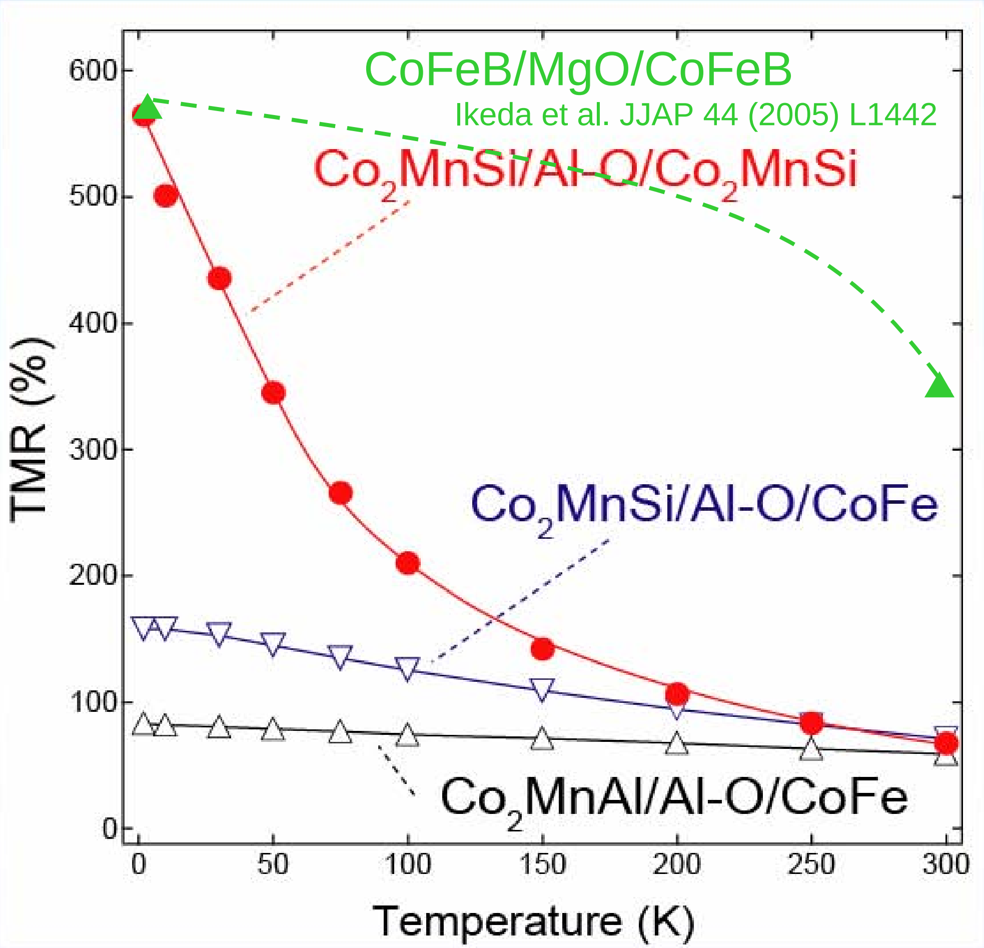


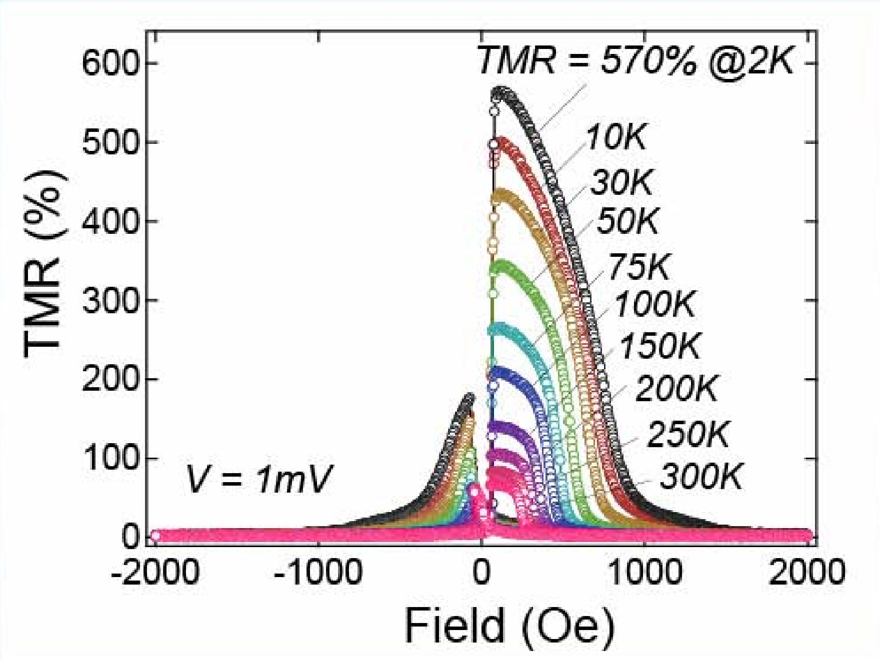
Junction structure



- Upper Co_2MnSi was annealed at 400°C before the deposition of IrMn AF layer.
- Upper Co_2MnSi electrode shows (001)-oriented growth
- Flat and sharp interface around Al-O interface

MTJ - $Co_2MnSi/Al\text{-}O/Co_2MnSi$





TMR ratio = 67%@RT, 570%@2K

⇒The largest TMR ratio in MTJ using Al-O barrier

Y. Sakuraba et al. Appl. Phys. Lett. vol.88 192508 (2006)

Large temperature dependence of TMR ratio should be solved.

Non-quasiparticle states in Co_2MnSi evidenced through magnetic tunnel junction spectroscopy measurements

L. Chioncel,[1,2] Y. Sakuraba,[3] E. Arrigoni,[1] M.I. Katsnelson,[4]
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[3] *Department of Applied Physics, Graduate School of Engineering, Tohoku University, Sendai 980-8579, Japan*
[4] *Institute for Molecules and Materials, Radboud University of Nijmegen, NL-6525 ED Nijmegen, The Netherlands*
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[6] *Institute of Theoretical Physics, University of Hamburg, DE-20355 Hamburg, Germany*

We investigate the effects of electronic correlations in the full-Heusler Co_2MnSi, by combining a theoretical analysis of the spin-resolved density of states with tunneling-conductance spectroscopy measurements using Co_2MnSi as electrode. Both experimental and theoretical results confirm the existence of so-called non-quasiparticle states and their crucial contribution to the finite-temperature spin polarisation in this material.

To be published in Phys. Rev. Lett.

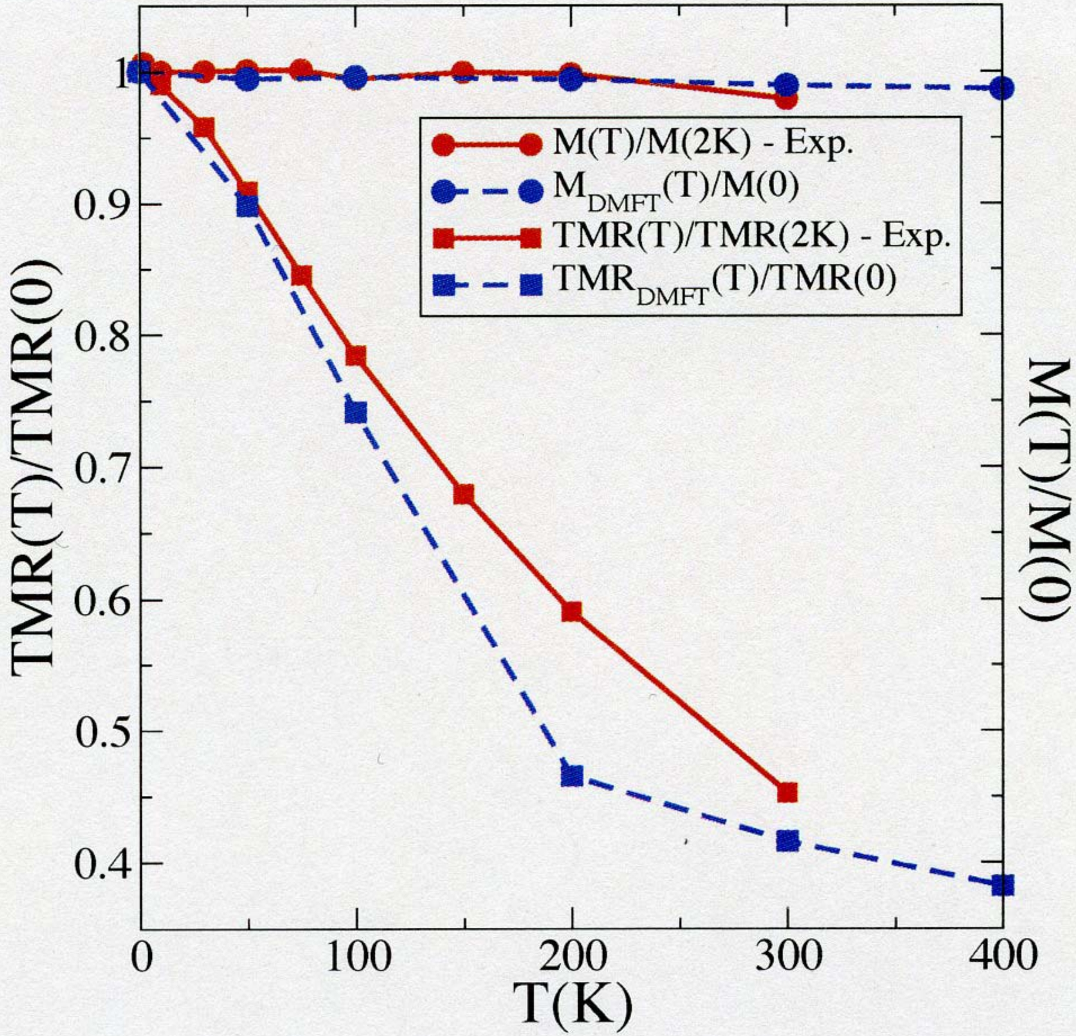


FIG. 3: (Color online) Comparison of the TMR ratio and magnetisation (M) as a function of temperature for a FeCo/Al-O/Co_2MnSi tunnel juction as obtained from experimental data and from LSDA+DMFT.

LSDA : Local Spin Density Appro.

DMFT : Dynamical Mean Field Theory

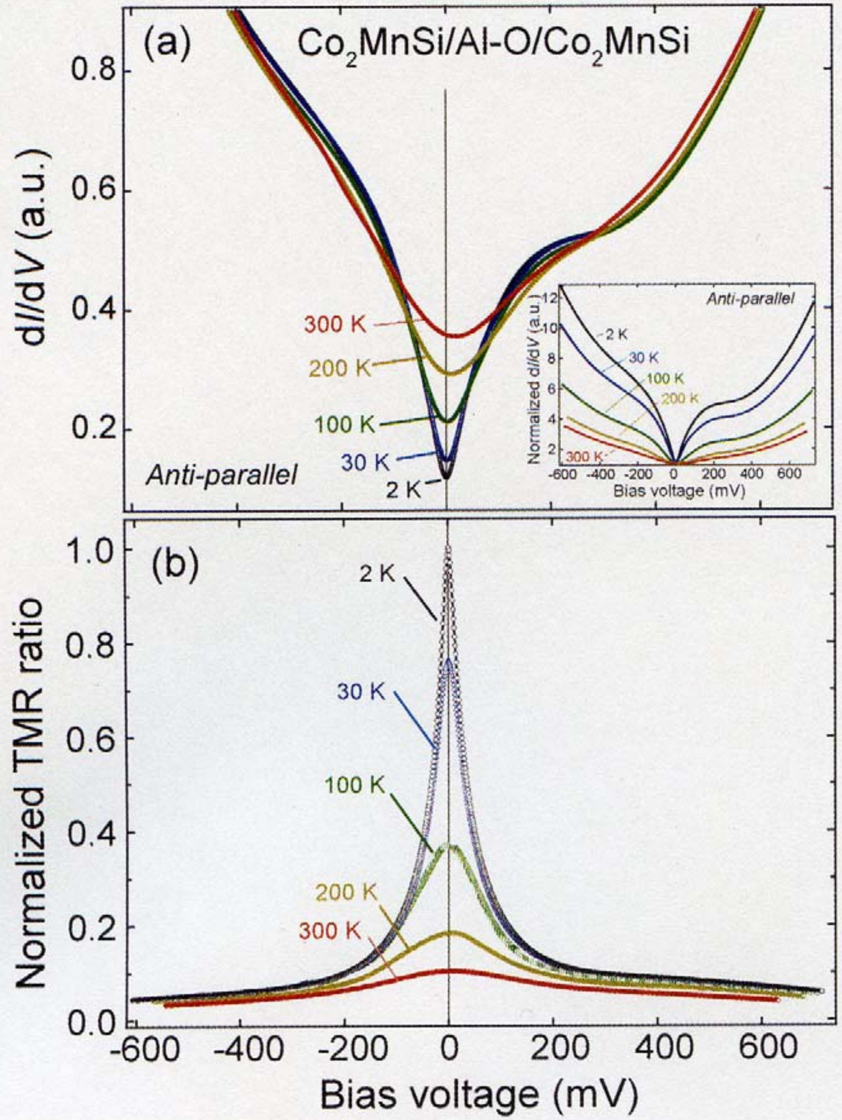


FIG. 2: (Color online) (a) Tunnel conductance for the Co_2MnSi/Al-O/Co_2MnSi anti-parallel magnetic tunnel junction and (b) TMR ratio (normalized to $T = 2K, V = 0$), as a function of voltage for different temperatures. The inset of panel (a) shows the conductance normalized to zero bias voltage.

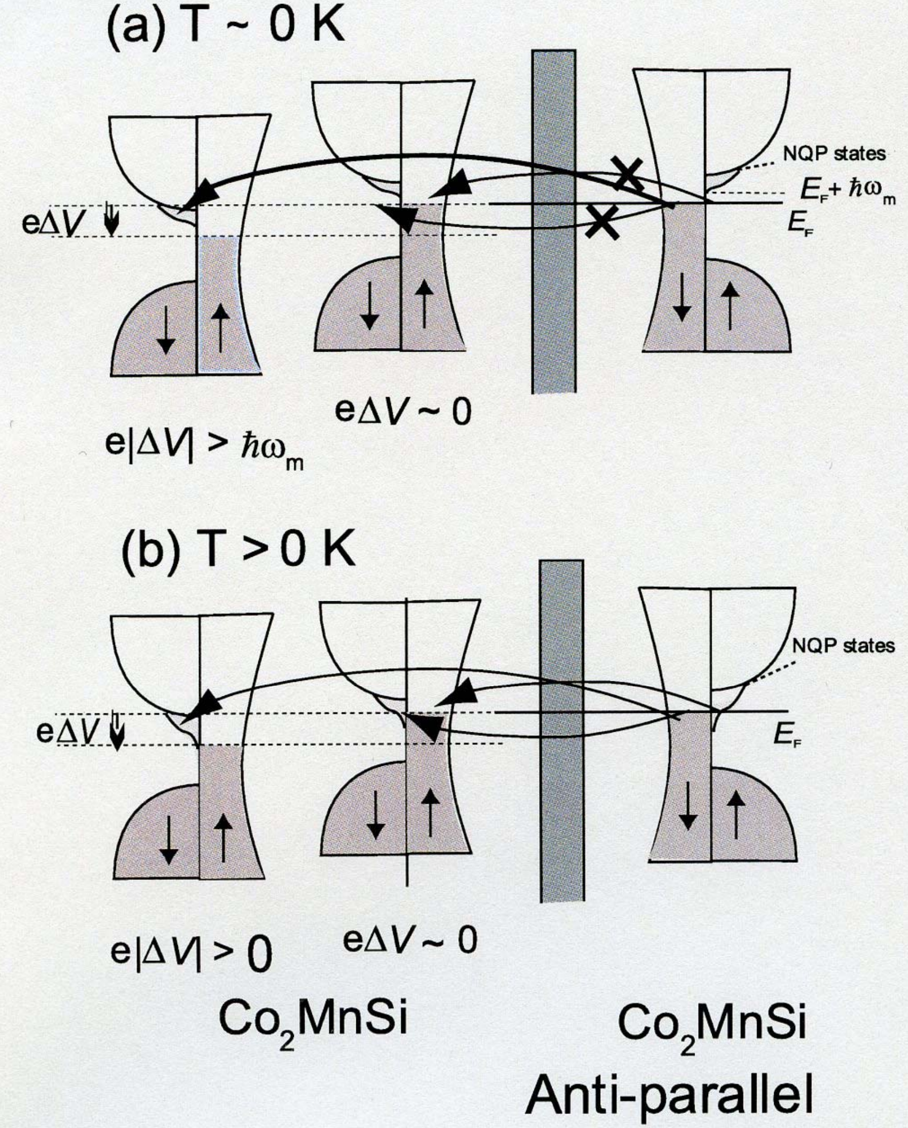


FIG. 4: Schematic representation of an electron tunneling process at $\Delta V \approx 0$ and at finite bias voltage in a Co_2MnSi/Al-O/Co_2MnSi magnetic tunnel junction. At $T \approx 0K$ (a), NQP states vanish at E_F, while for $T > 0$ (b) (see text) NQP states extends across E_F and an additional tunneling channel opens.

3. Applications of TMR junction

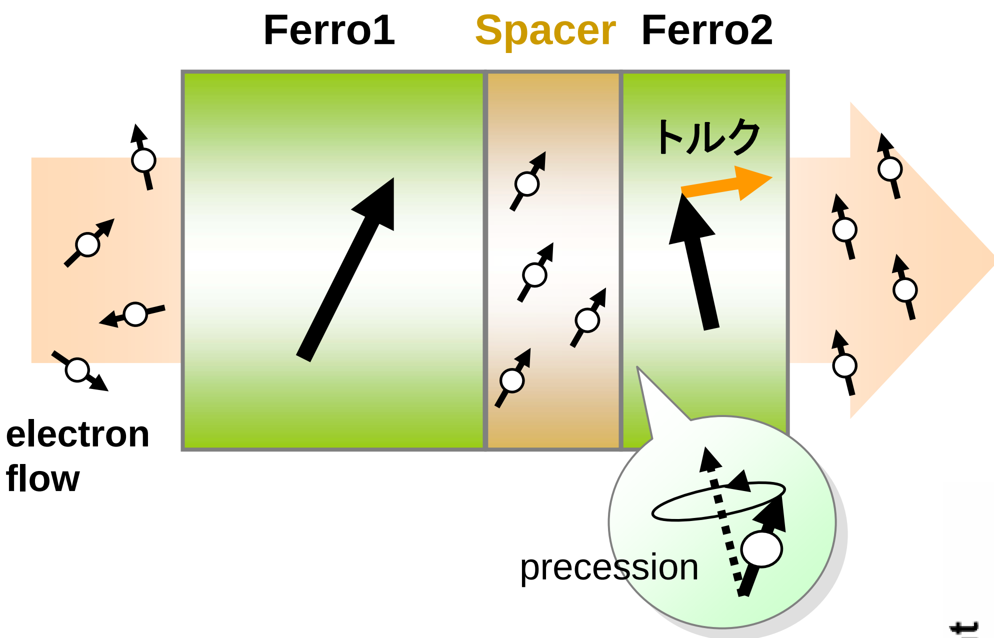
Application of large TMR junctions

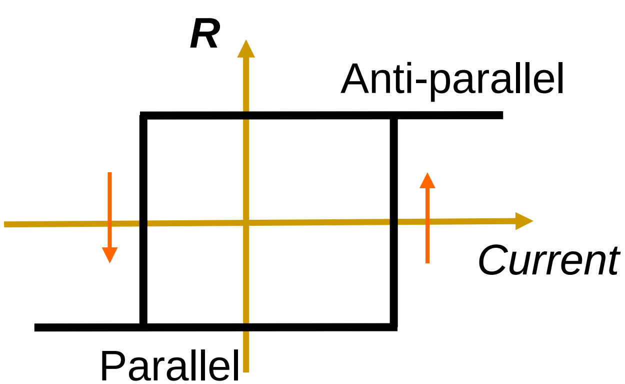
- MRA (Magnetization switching : bit and word lines)
- Spin-RAM (Switching by spin transfer torque)
- Magnetic reading head of HDD
- GPS sensor devices
- High frequency oscillator devices
-
-
-

Magnetization switching by direct current

Spin-transfer switching (STS)

Slonzcewski, JMMM**159** (1996)L1

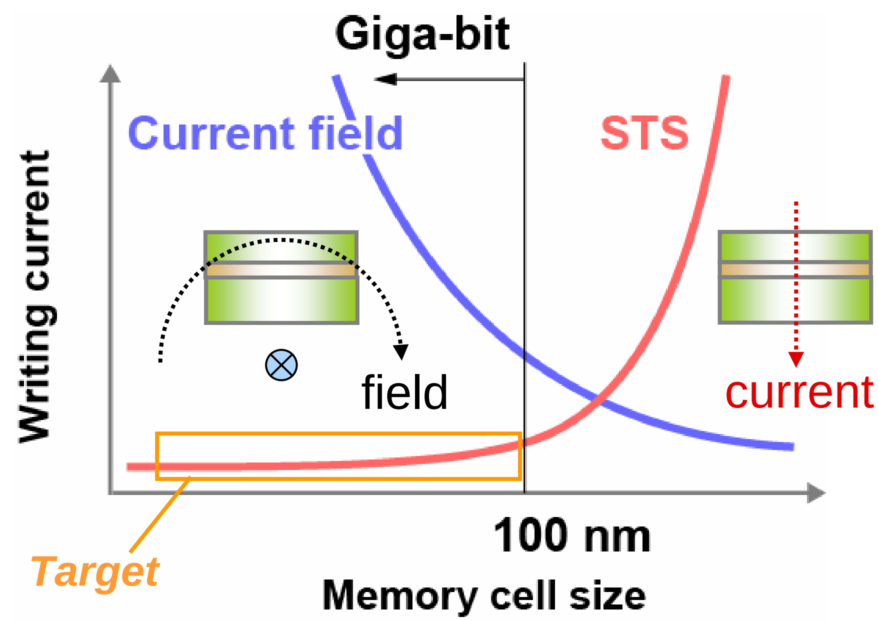




STS + TMR

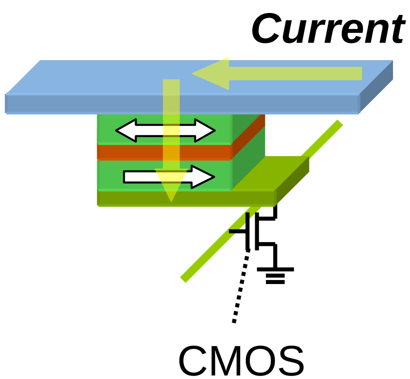
G bit + low power MRAM

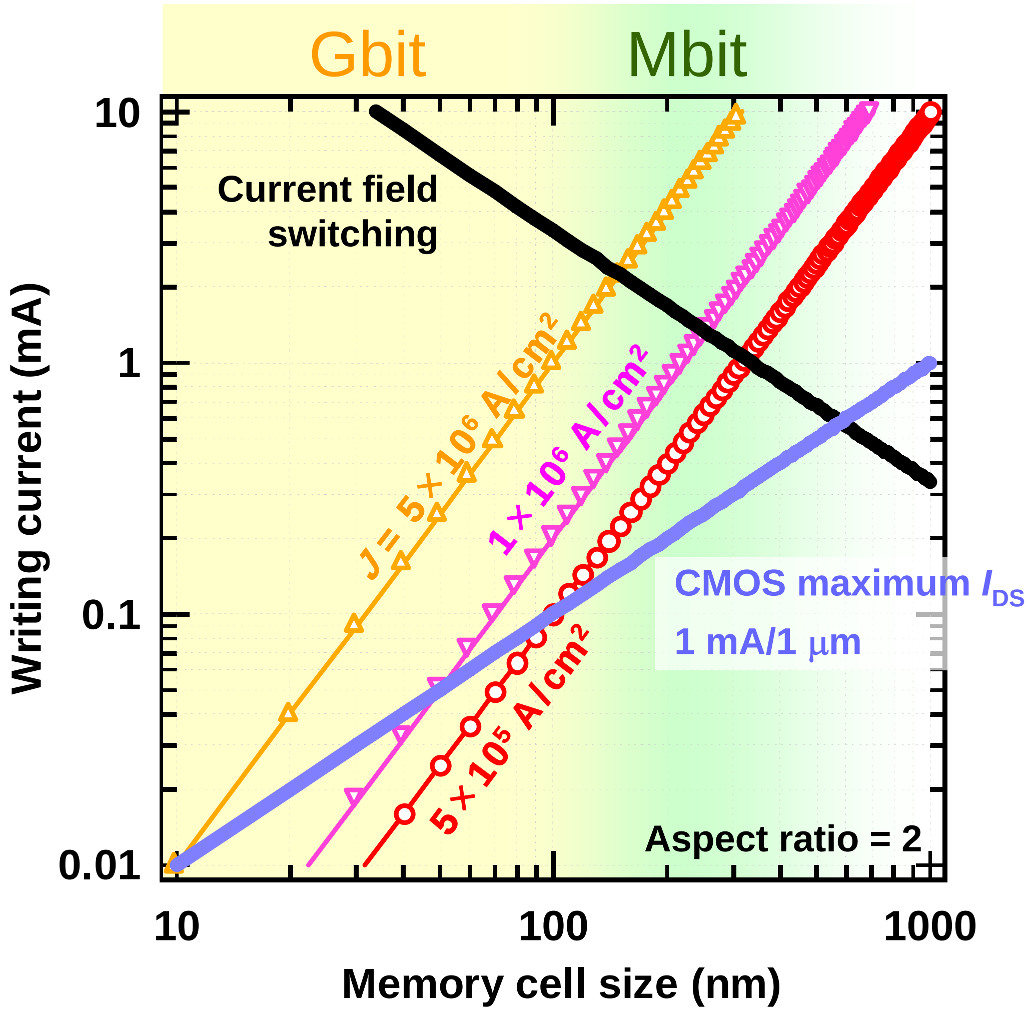
New devices



Issues for Spin-RAM

Spin transfer switching





Switching current : J_{C0} ~ 5×10^{5} A/cm^{2}

Issues for device application

Requirement for devices

Junction Resis. 20 kΩ

Area 80 × 160 nm^2

R・A ~ 200 Ωμm^2

J_{C0} ~ 5 × 10^5 A/cm^2

Switching current density

J. C. Slonzcewski, JMMM159 (1996)L1，PRB71(2005)024411

$$J_{c0}^{\pm} = \frac{\alpha \gamma e M_S d}{\mu_B g(P,\theta)} \left[H_{ext} \pm (H_{eff} + 2\pi M_S)/2 \right]$$

$$g_{tunnel}(P,\theta) = \frac{1}{2} \frac{P}{(1+P^2 \cos\theta)}$$

Thermal stability

R. H. Koch, PRL92(2004)088302-1

$$J_c = J_{c0} \left\{ 1 - \frac{k_B T}{K_u V} \ln \frac{\tau_p}{\tau_0} \right\}$$

Pulse width

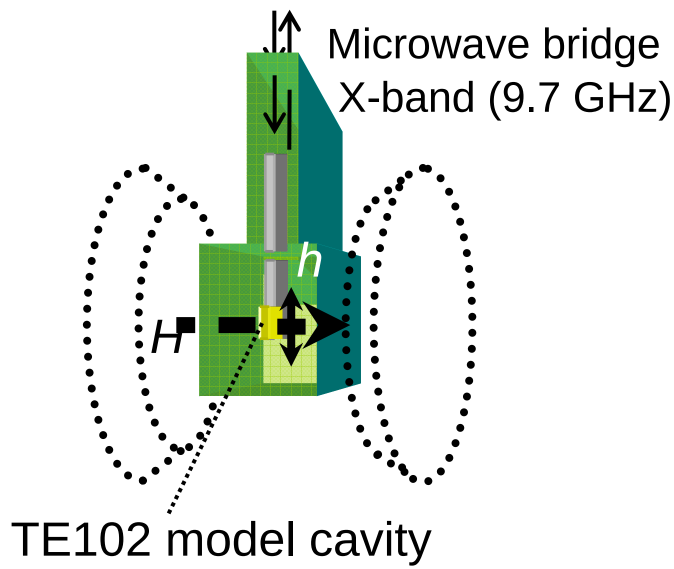
τ_0 ~ 1 ns

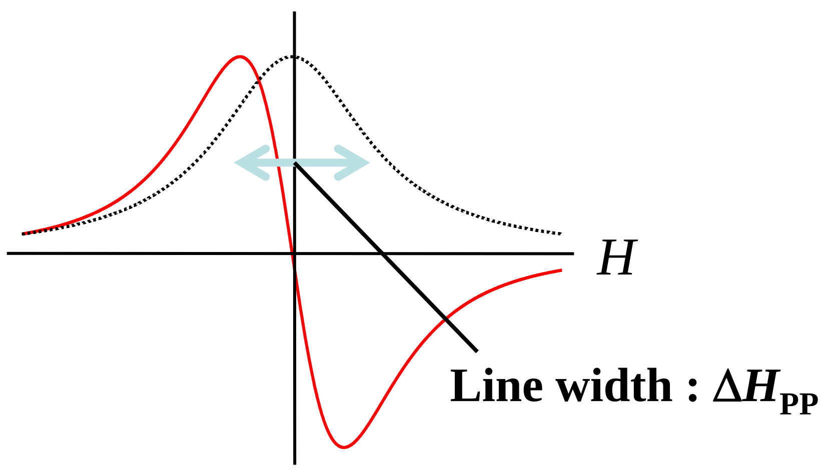
α : damping constant, M_s : saturation magnetization

d : free layer thickness, P : spin polarization of pinned layer

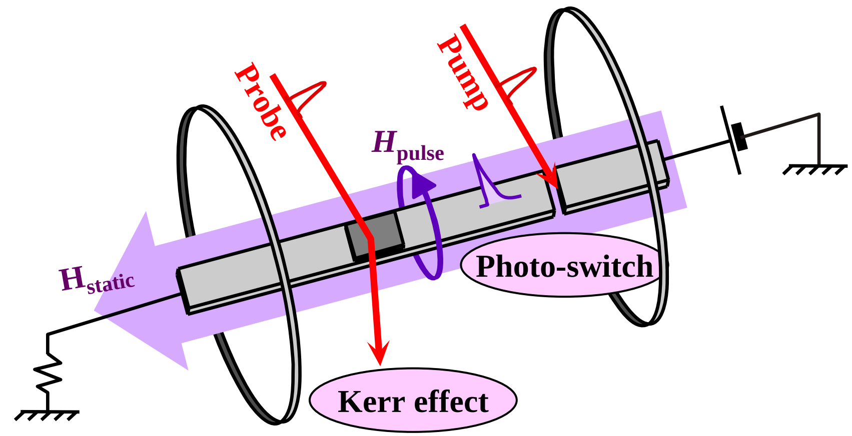
Measurement of damping constant α

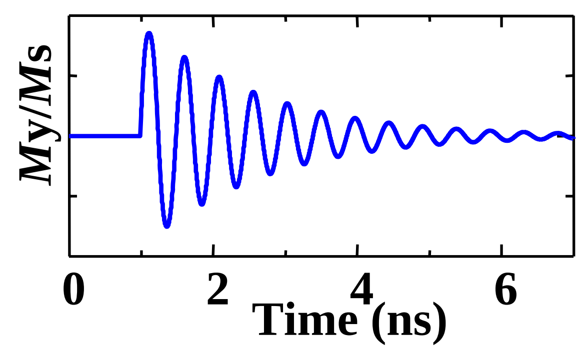
FMR



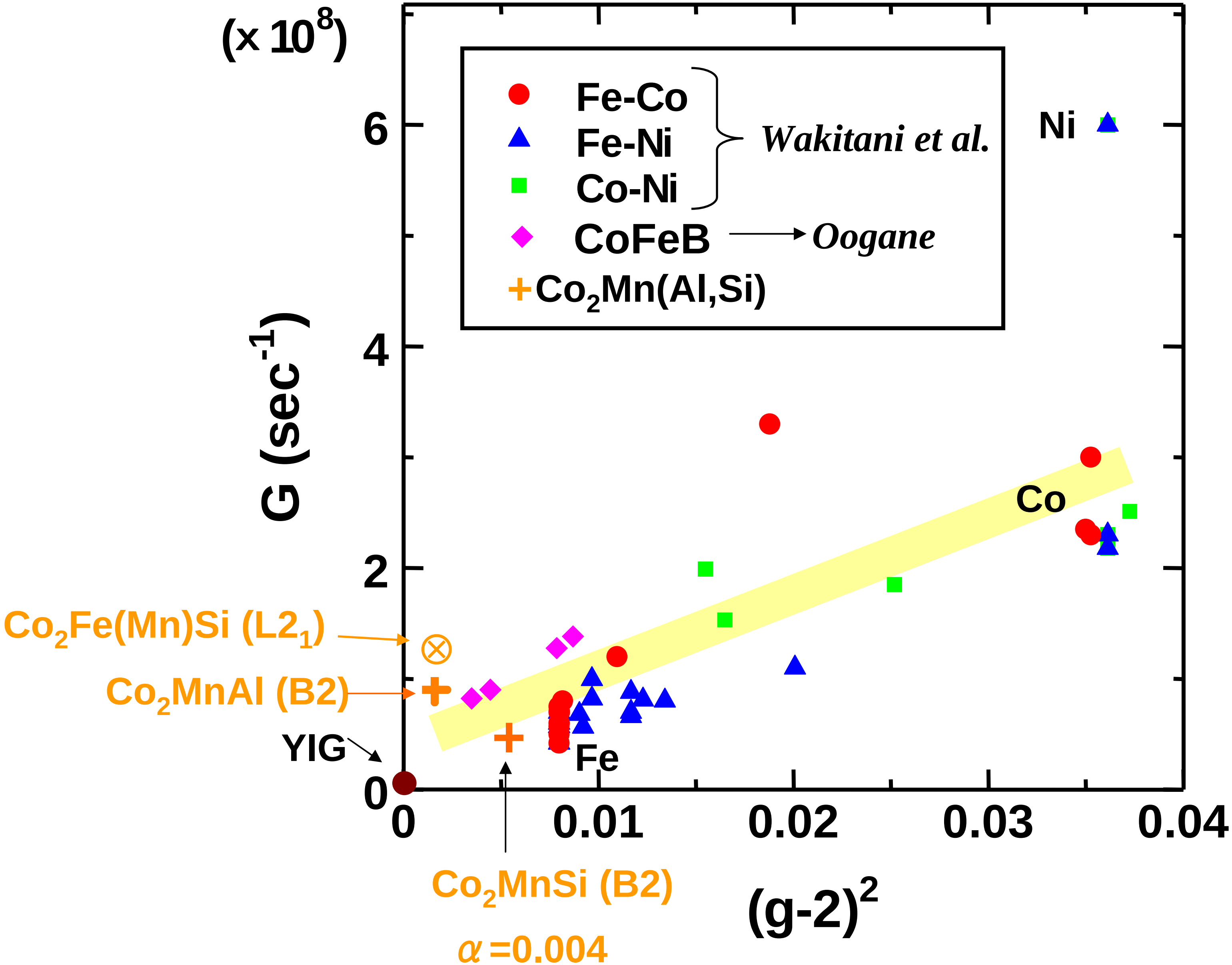


Pump-probe method





$G \equiv \alpha \gamma M_S$; $G \propto (g-2)^2$; g-2 $\approx$ L/S



Summary

- TMR ratio more than 100 % (200 %) at room temperature has been obtained for Heusler electrode and Al-O (MgO) barrier tunnel junctions.
- Damping constant α of Heusler alloys (Co_2MnSi) is 0.004.
- Rapid decrease of TMR ratio with raising temperature and increasing voltage can be explained by Non-quasiparticle states.
- Heusler alloys are excellent candidate for spintronics materials.

ACKNOWLEDGMENTS

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