at Japan-EU Workshop (November 21-22, 2011)

Materials Design for Magnets -Focused on Magnetic Semiconductors-

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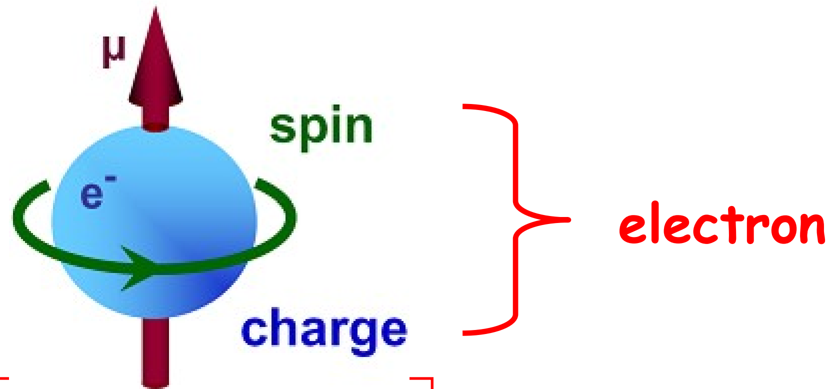
Spintronics: spin-based electronics

Beyond conventional charge-based electronics !



Advantages:

Fast data processing speed

Low electric power consumption

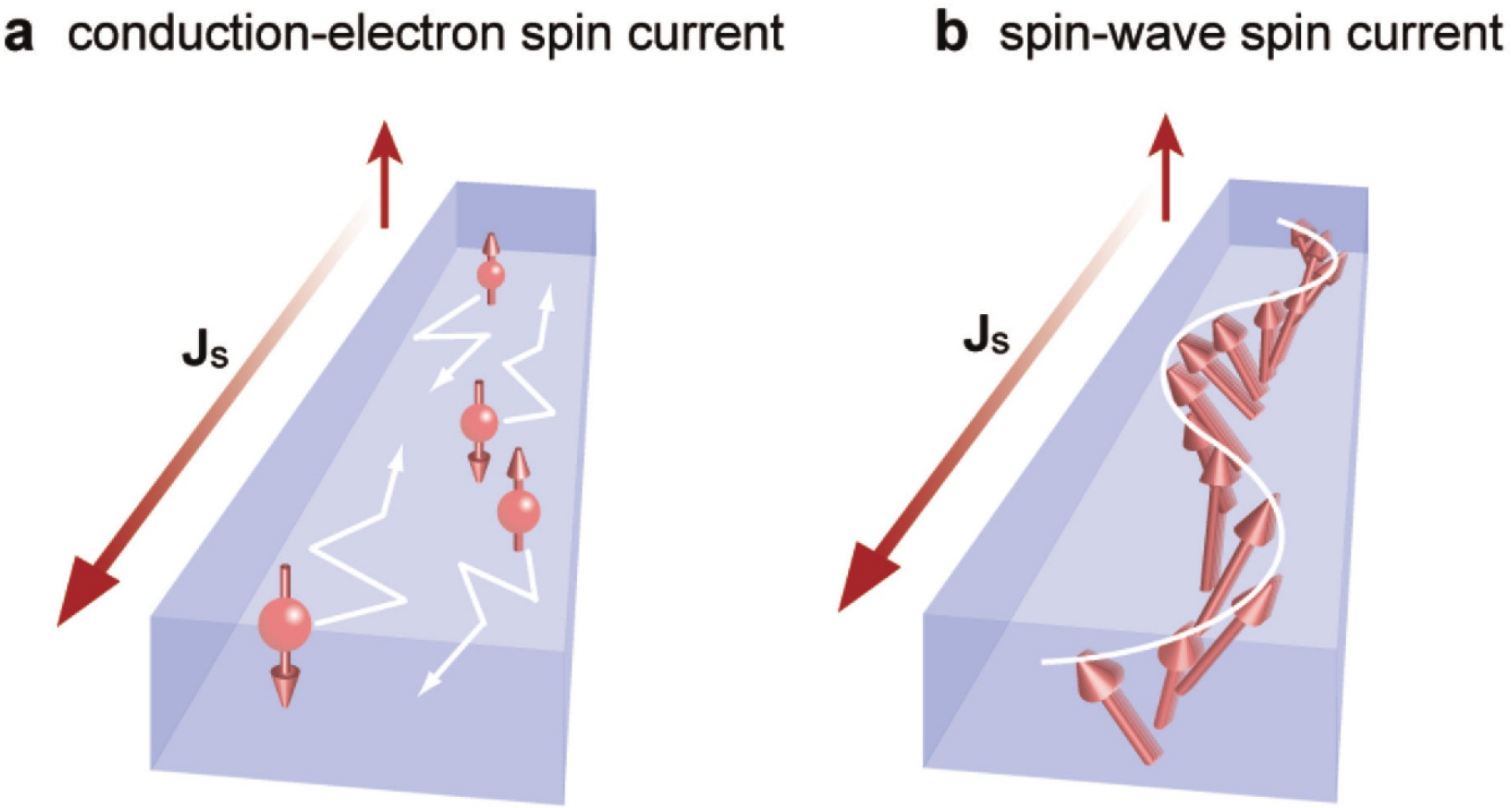
Increased integration density

Our aim is to use spin current and charge current on an equal footing!

Challenge:

* room temperature magnetic semiconductors!!*
* p-type and n-type magnetic memiconductors!!*
* enhanced spin-orbit interaction !!*

spin currents :



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

$$\begin{cases} \boldsymbol{j} = \boldsymbol{j}_\uparrow + \boldsymbol{j}_\downarrow \\ \boldsymbol{j}_{\text{spin}} = \boldsymbol{j}_\uparrow - \boldsymbol{j}_\downarrow \end{cases}$$

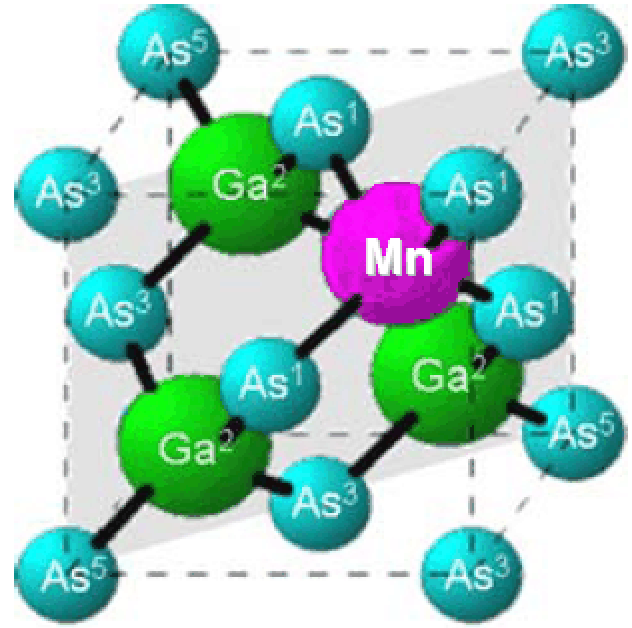
Insulating magnets are good spin current conductors!!

Research Field: Magnetic Semiconductors

Charge + Spin = Charge & Spin

Semiconductor   *Magnet*   *Magnetic Semiconductor*

GaAs + Mn = (Ga,Mn)As



Control magnetism (spin) by electric way (charge) !

***Challenge*:**

For present magnetic semiconductors, Curie temperature (~ 120K) << room temperature

Seek magnetic semiconductors with Curie temperature > room temperature !

Room temperature magnetic semiconductors !!

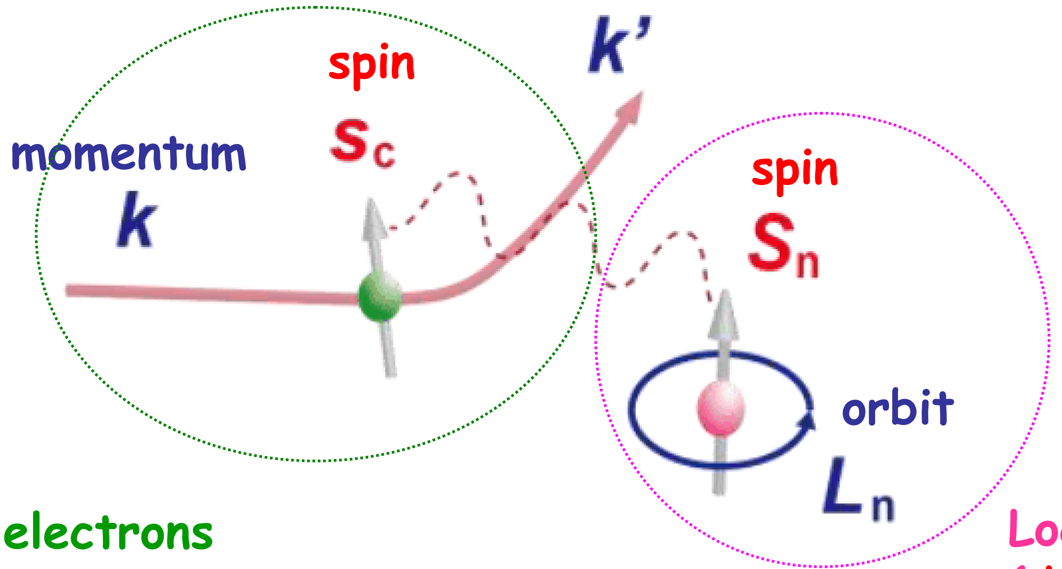
Magnetic Semiconductors:

	host	impurity	structure	T_c[K](concentration)	E_g[eV](〜300K)	m_e*[m_e]	m_h*[m_e]
II-VI	ZnTe	Mn		10(0.019)	2.391	0.122	0.42,0.17,0.72,0.14,0.89,0.14
		Cr		300(0.2)			
	BeTe	Mn		2.4(0.1)			
	ZnO	V	Wurtzite	>350(0.15,0.25)	3.2	0.24	1.8
		Mn		>300(0.002+N)			
		CoFe		>300(0.15)			
		Fe,Cu		550(0.05Fe+0.01Cu)			
III-V	GaAs	Mn	Zinc-blende	140(0.06)	1.429	0.0667	0.71,0.12
	InAs	Mn		35(0.14)	0.359	0.024	0.41,0.026
	InSb	Mn		85(0.028)	0.18	0.0139	0.32,0.016
	GaP	Mn		250(0.094)	2.261	1.7,0.254	0.55,0.13
	GaN	Mn	Wurtzite	300(0.03)	3.39	0.236	>0.6
		Cr		280(0.03)			
	AlN	Cr		>350			
IV-VI	TiO_2	Co	Rutile	>400	3.03-3.54		
			Anatase	>400	3.1-3.46		
d^0	MgO	N	Rocksalt	?	7.7	?	?
	SrO	N	Rocksalt	RT(Sawatzky, 2007)	5.3	?	?

UFO (Unidentified Ferromagnetic Objects)

(c.f., USO for High Tc Superconductors)

Our theoretical approach :



Conduction electrons
(s orbital)

Localized electrons
(d orbital)

Periodic crystal potentials

Coulomb interaction
(Ferrmomagnetism)

band theory
(density functional theory)

+

Non-perturbative theory
(quantum Monte Carlo)

A general & powerful combined method !
First established

The Method:

Host semiconductor

→ ***detailed band structure (LDA band calculation)***

Magnetism

→ ***strong electron correlation (QMC)***

(Coulomb interaction)

⬇

QMC method with Hirsch – Fye algorism

$$H = \sum_{\alpha \mathbf{k} \sigma} \left(\varepsilon_{\mathbf{k}}^{\alpha} - \mu \right) c_{\mathbf{k}\sigma}^{\alpha}{}^{\dagger} c_{\mathbf{k}\sigma}^{\alpha} + \sum_{i\sigma} \left(\varepsilon_{d} - \mu \right) d_{i\sigma}{}^{\dagger} d_{i\sigma} + \sum_{\alpha \mathbf{k} i \sigma} \left(V_{\mathbf{k}i}^{\alpha} c_{\mathbf{k}\sigma}^{\alpha}{}^{\dagger} d_{i\sigma} + h.c. \right) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

Material dependence
(LDA or tight binding band calculation)

Materials design of magnetic semiconductors

PHYSICAL REVIEW B 79, 024407 (2009)

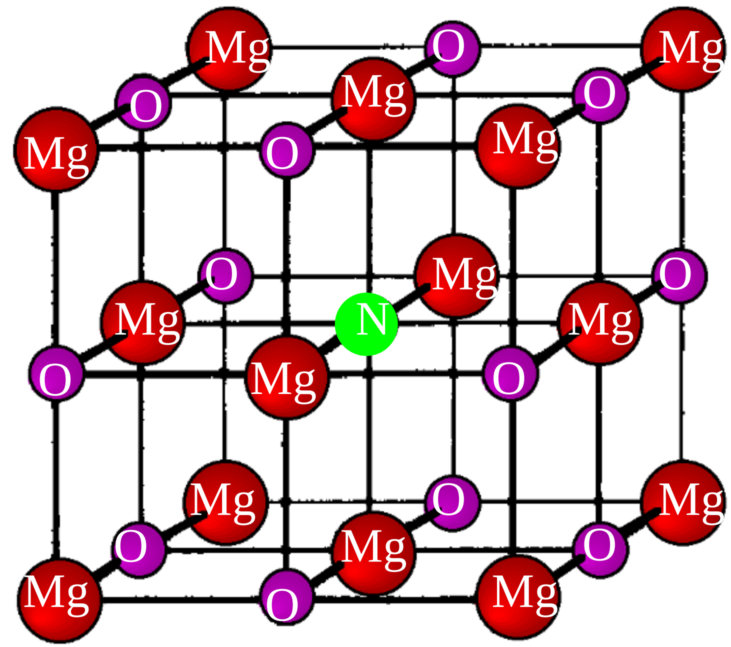
Possible d^0 ferromagnetism in MgO doped with nitrogen

Bo Gu,[1] Nejat Bulut,[1,2] Timothy Ziman,[3] and Sadamichi Maekawa[1,2]
[1]*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

Wide-gap Semiconductor (MgO): Charge

Non-Magnet (N): No active spin

⇨ *Magnetic semiconductor Mg(O,N): Charge & Spin*



O : $2s^2\ 2p^4$
N: $2s^2\ 2p^3$

⇨ *A unpaired p-orbital hole at N site*

Predict a new magnetic semiconductor without magnetic ion !

c.f., Experiment of Mg(O,N): S. Parkin at IBM Almaden (unpublised).

Possible d_0 ferromagnetism:

Target materials:

Sr(O,N), Mg(O,N), Ca(O,v), Hf(O,v),…..

(v: vacancy)

N-doped diamond :

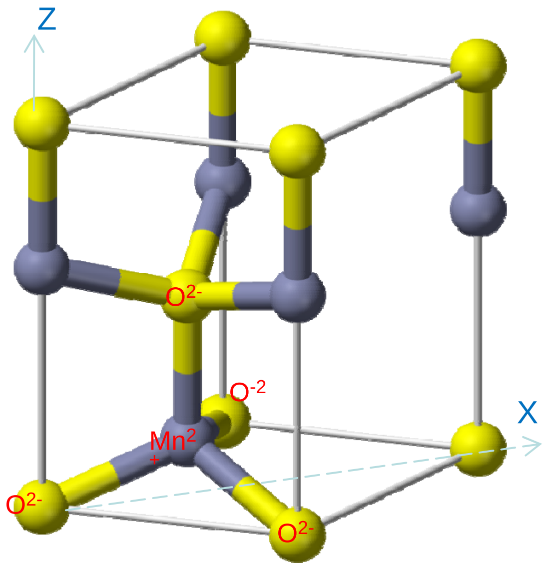
- *n-type semiconductor*
- *N-impurity has spin ½,*
- *deep impurity level,*
- *N-concentration.*

(B-doped diamond: p-type $\longrightarrow$ superconductor)

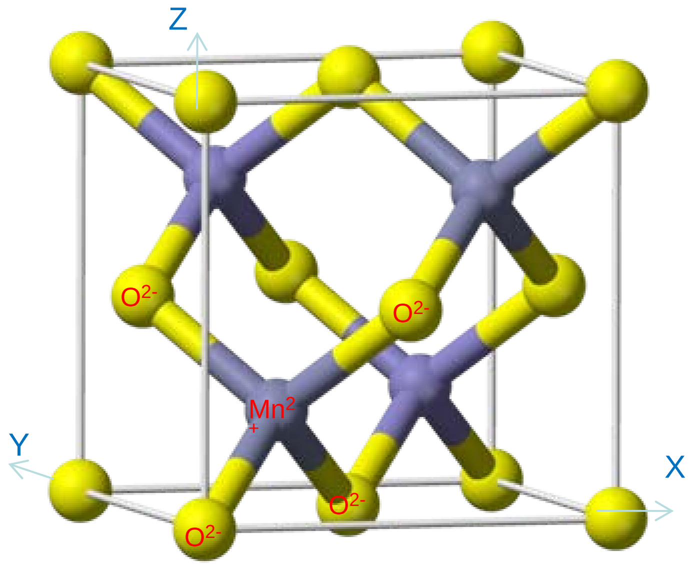
(Zn,Mn)O

Crystal structures

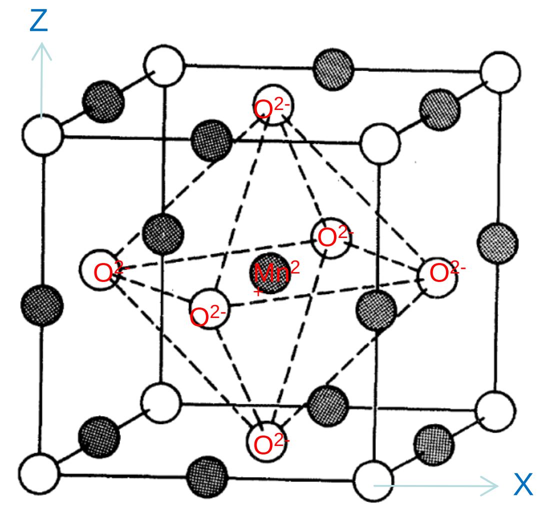
Wurtzite(hexagonal)



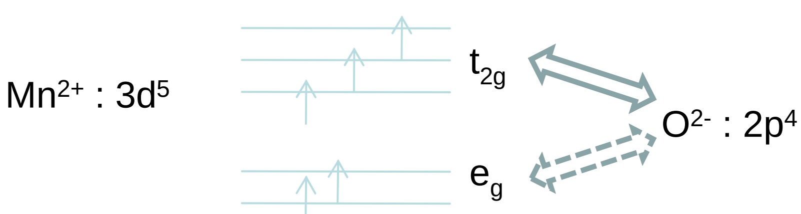
Zincblende (fcc)



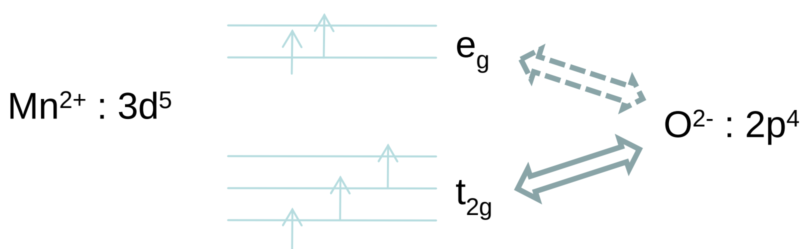
Rocksalt(fcc)



(Zn,Mn)O Zn^{2+} : $4s^2$ conduction band Mn^{2+} : $3d^5$ O^{2-} : $2p^4$ valence band



wurtzite and zincblende
(tetrahedral crystal field)



rocksalt
(octahedral crystal field)

Zincblende sturucture is better for ferromagnetism!!

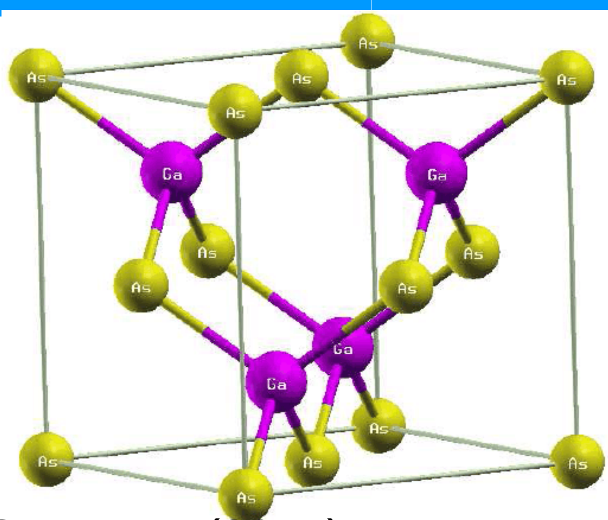
Li(Zn,Mn)As (bulk)

I-II-V DMS

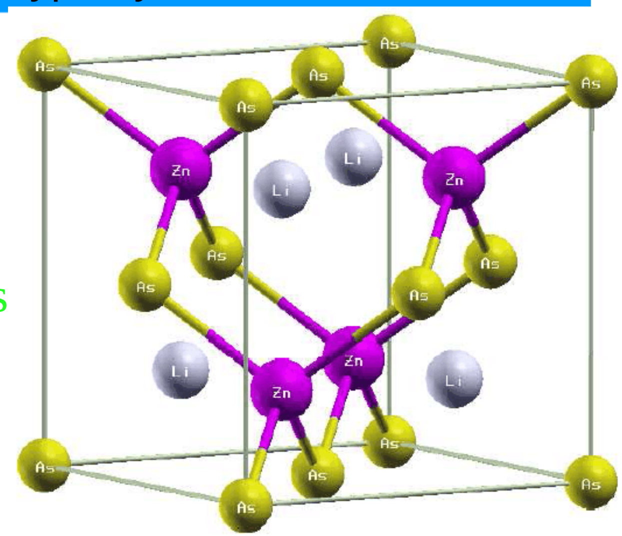
p-type and n-type (?!)

Experiments	(Ga,Mn)As	Li(Zn,Mn)As
Crystal structure	Zinc blende (ZB)	ZB + filled tetrahedral
Lattice constant	5.65 Å	5.94 Å
Energy gap	1.52 eV (direct)	1.61 eV (direct)
Substitutional Mn	Mn2+ / Ga3+	Mn2+ / Zn2+
Chemical solubility limit	< 1%	NO
Concentration of Mn	~ 5% in very thin film	~ 15 % in bulk poly crystal
Curie Temperature	~120 K	~ 40 K
Moment	4 ~ 5 μ B / Mn	~ 5 μ B / Mn
Carriers type	p type (hole)	n type by excess Li+ (?) p type by less Li+

GaAs



LiZnAs

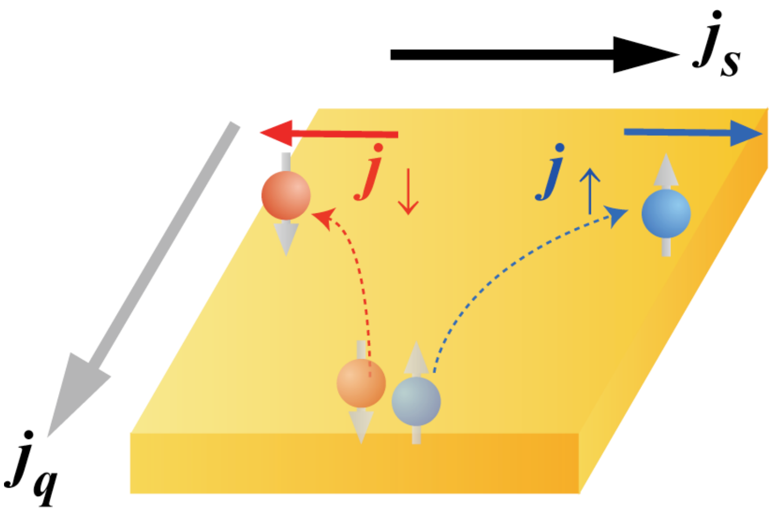


Nature Commun. (2011)

Spin Hall effect due to spin-orbit interaction:
(Conversion between spin current and charge current)

spin-Hall effect

$$\hat{j}_s \propto \hat{z} \times \hat{j}_q$$



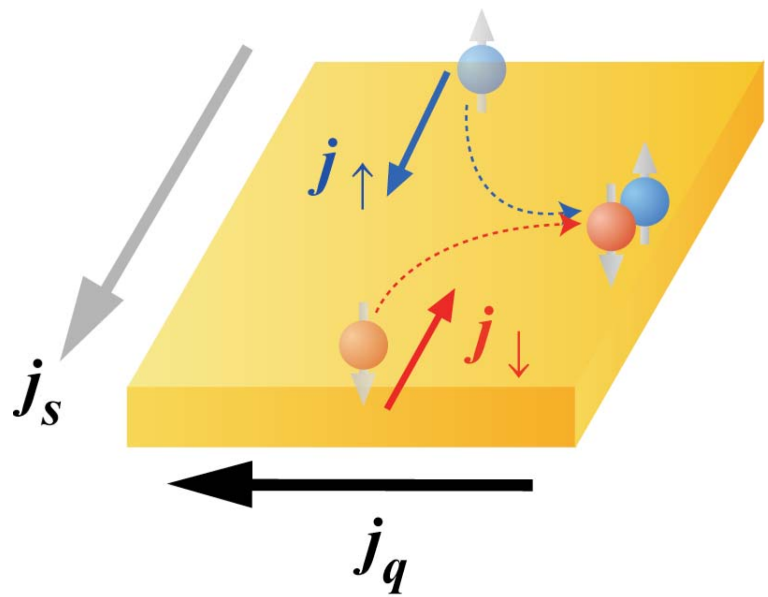
Charge current

↓

Spin current

Inverse spin-Hall effect

$$\hat{j}_q \propto \hat{z} \times \hat{j}_s$$

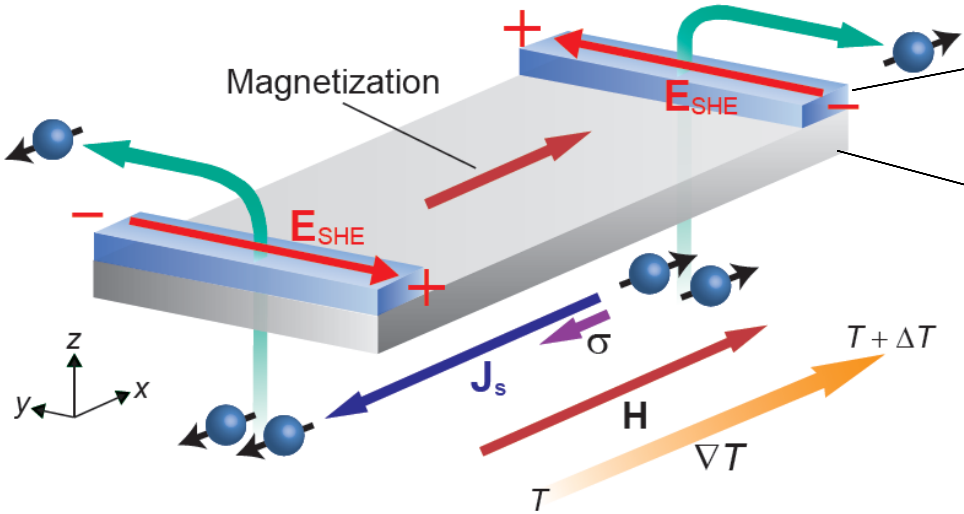


Spin current

↓

Charge current

Spin Seebeck Effect:



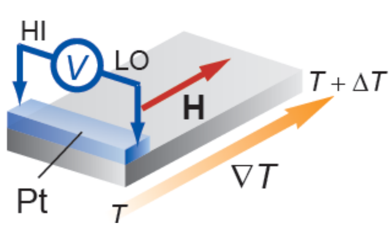
Pt: spin detector
(4 mm x100 μm x10 nm)

NiFe: thermo-spin generator
(4 mm x6 mm x20 nm)

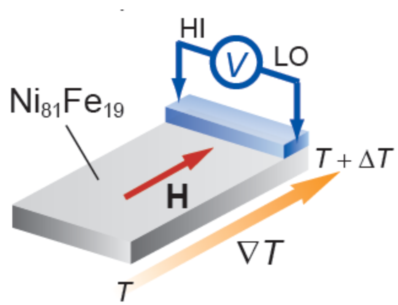
spin Hall effect:

$$\mathbf{E}_{SHE} = D_{ISHE}\, \mathbf{J}_s \times \boldsymbol{\sigma}$$

magnitude & polarization of $\mathbf{J}_s$



Lower *T* end

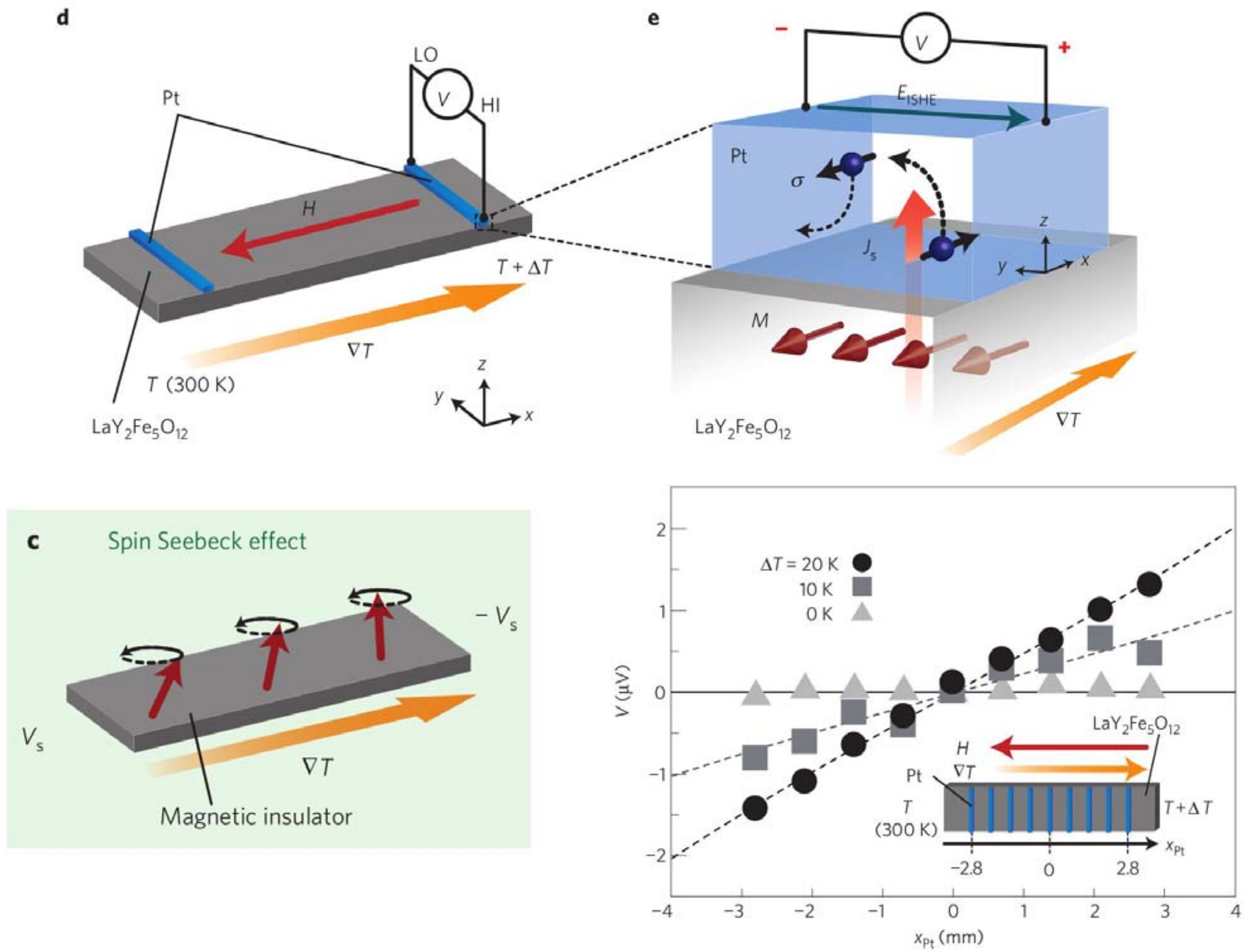


Higher *T* end

Experiment (Spin Seebeck effect: SSE)

Spin Seebeck insulator (Nature Materials in press)

K. Uchida[1], J. Xiao[2,3], H. Adachi[4,5], J. Ohe[4,5], S. Takahashi[1,5], J. Ieda[4,5], T. Ota[1], Y. Kajiwara[1], H. Umezawa[6], H. Kawai[6], G. E. W. Bauer[3], S. Maekawa[4,5] and E. Saitoh[1,4,7]★



Research Field: Spin Hall Effect

Charge current ⟷ Spin current

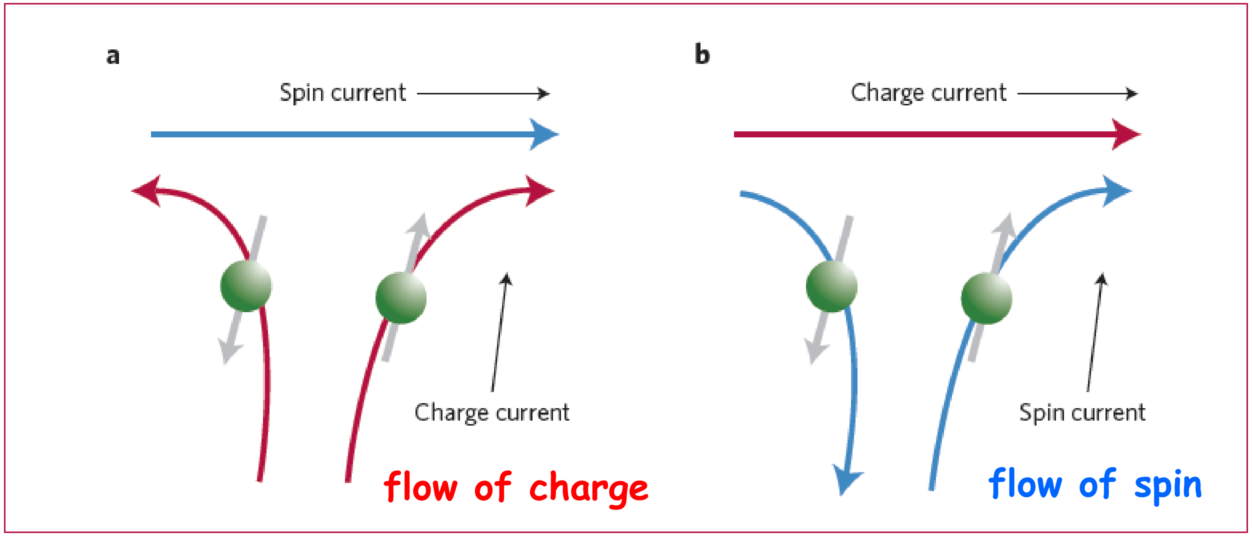


Figure 1 | Interacting charge and spin currents. **a**,**b**, Spin Hall effect (**a**) and the inverse spin Hall effect (**b**), where the longitudinal charge current is converted into the transverse spin current and vice versa.

Conversion efficiency is determined by spin-orbit interaction !

***Challenge*:**

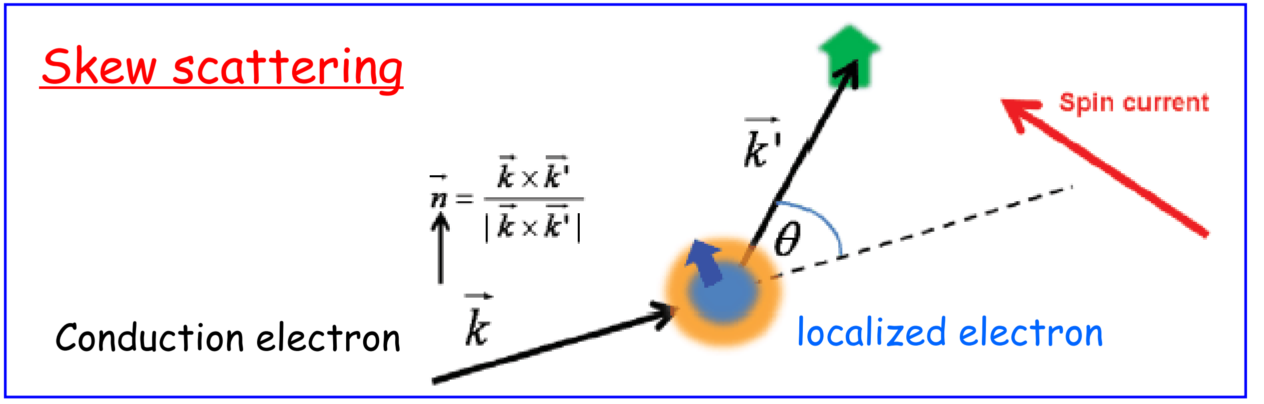
In many materials, spin-orbit interaction is very small.

Seek materials with large spin Hall effect at room temperature !

PRL 105, 216401 (2010) PHYSICAL REVIEW LETTERS week ending 19 NOVEMBER 2010

Surface-Assisted Spin Hall Effect in Au Films with Pt Impurities

B. Gu,[1,2] I. Sugai,[3] T. Ziman,[4] G. Y. Guo,[5,6] N. Nagaosa,[7,8] T. Seki,[3] K. Takanashi,[3] and S. Maekawa[1,2]
[1]Advanced Science Research Center, Japan Atomic Energy Agency, Tokai 319-1195, Japan



Impurities on surface → **Strong valence fluctuation** → **Impurity levels shifted to Fermi level** → **Enhanced skew scattering**

Find a new way to enhance skew scattering (spin Hall effect) !

Spintronics: New treasure in a gold mine

NPG Asia Materials research highlight | doi:10.1038/asiamat.2011.24
Published online 14 February 2011

Nature Publishing Group (NPG) Asia Materials research highlight !

Enhanced spin-orbit coupling:

Au with Fe impurities, Cu with Ir impurities,….

Summary:

Targets:

Higher Tc in magnetic semiconductors

d0 magnets such as Mg (O,N)

Enhanced spin Hall effect

Strategy:

Band structure of the host materials (band (LDA) thoery),

Strong electron correlation (fcoulomb interaction) for magnetism (QMC).

> LDA → Anderson impurity Model → QMC → Materials design

→ *This simulation program may be applied to a variety of materials design.*

→ *Good supercomputer facilities.*