

# Spintronics Design Course (in CMD-WS37) 5<sup>th</sup> lecture

5th Lecture:

Spintronics, Design, Magnetization Control I

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Spintronics Design Course in CMD-WS37

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(90 minutes)

第5講義:スピントロニクス・デザイン・磁化制御 I :90分 小田竜樹(金沢大理工)

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# Contents in 5th lecture

- (5-1) Electronics structure: Ferromagnetism, Antiferromagnetism**
- (5-2) Magnetic moment: spin, orbital, localized and itinerant**
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- (5-5) Spin-orbit interaction, spin-texture in the reciprocal space**
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- (5-7) Magnetic anisotropy energy: electron orbital, magnet shape**
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- (5-10) Landau–Lifshitz–Gilbert equation**
- (5-11) Design on magnetic anisotropy in magnetic materials**

- (5-1) 電子構造(強磁性、反強磁性)、
- (5-2) 磁気モーメント(スピン、軌道、局在・遍歴)
- (5-3) ゼーマンエネルギー
- (5-4) 磁性担体距離と磁気相互作用、磁気双極子相互作用
- (5-5) スピン軌道相互作用、逆格子空間スピントクスチャ
- (5-6) スピントランスファートルク
- (5-7) 磁気異方性エネルギー(電子軌道、磁性体形状)
- (5-8) 磁気異方性(面内、面直)
- (5-9) 電圧スピントルク
- (5-10) ランダウ＝リフシッツ＝ギルバート方程式
- (5-11) 磁性薄膜材料の磁気異方性デザイン

# (5-1) Electronics structure: Ferromagnetism,

# Anti-ferromagnetism

One electron approx.

$$\left\{ -\frac{\hbar^2}{2m} \nabla^2 + V(\mathbf{r}) \right\} \Psi_{n\mathbf{k}\sigma}(\mathbf{r}) = \varepsilon_{n\mathbf{k}\sigma} \Psi_{n\mathbf{k}\sigma}(\mathbf{r})$$

$(n\mathbf{k}\sigma)$  : quantum number

$\Psi_{n\mathbf{k}\sigma}(\mathbf{r})$  : wave function

$\varepsilon_{n\mathbf{k}\sigma}$  : eigenvalue

$n$  : band index

$\mathbf{k}$  : wave number

$\sigma$  : spin index ( $\sigma = \uparrow, \downarrow$ )

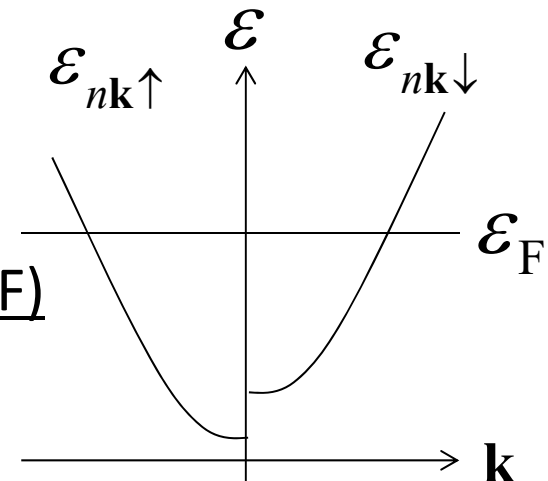
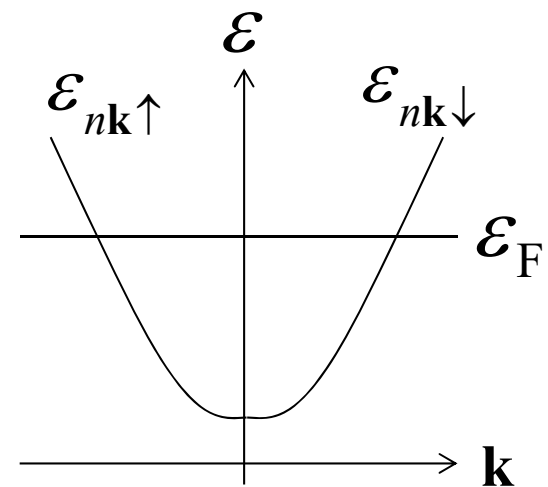
Ferromagnet(F)

$$\varepsilon_{n\mathbf{k}\uparrow} \neq \varepsilon_{n\mathbf{k}\downarrow}$$

Anti-Ferromagnet(AF)

$$\varepsilon_{n\mathbf{k}\uparrow} = \varepsilon_{n\mathbf{k}\downarrow}$$

Band dispersion



## (5-1-2) Electronics structure: Density of states

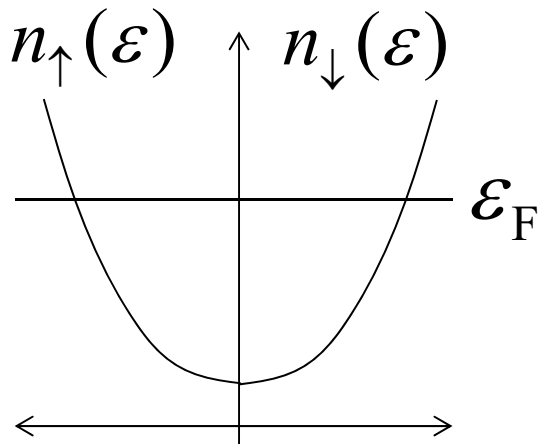
Density of states(DOS)

$$n(\mathbf{r}) = \sum_{\sigma} \sum_{nk} \delta(\varepsilon - \varepsilon_{nk\sigma})$$

$$n_{\sigma}(\mathbf{r}) = \sum_{nk} \delta(\varepsilon - \varepsilon_{nk\sigma})$$

$\sigma$  : spin index ( $\sigma = \uparrow, \downarrow$ )

Spin-up state      Spin-down state



DOS

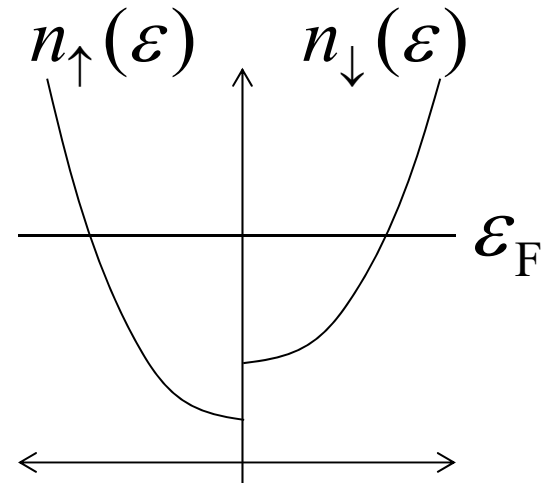
DOS

[states/(energy unit)/cell]

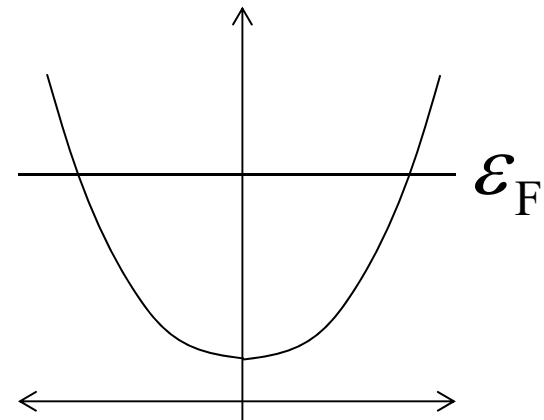
Ferromagnet(F)

Majority spin state

Minority spin state



Anti-Ferromagnet(AF)



**Effective  
potential  
 $V(\mathbf{r})$**

## (5-2) Magnetic moment: spin, orbital, localized and itinerant

**Origin of magnetism** Most of magnetism in materials comes from the contained electrons.

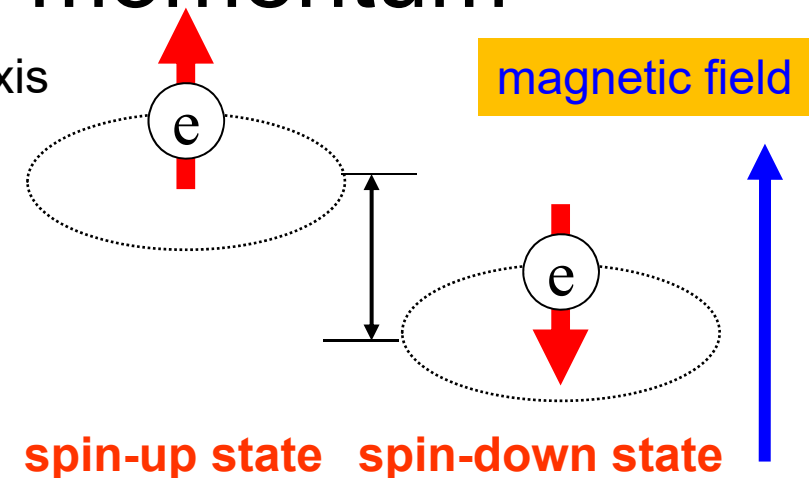
**main origin of magnetism**

### electron : spin angular momentum

like being supposed to rotate on its own axis

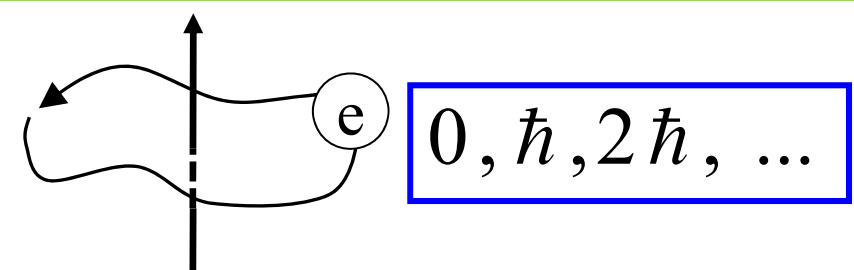
$\frac{1}{2} \hbar$

two different spin states in an electron, different energy levels for external magnetic field



**spin-up state    spin-down state**

note) The orbital angular momentum appears from the orbit motion of electrons



$0, \hbar, 2\hbar, \dots$

## (5-2-2) Orbital magnetic moment

Coulomb gauge  
 $\vec{\nabla} \cdot \vec{A} = 0$

An electron in a magnetic field along z-axis

$$\mathcal{H} = \frac{1}{2m} \left( \vec{p} + \frac{e}{c} \vec{A} \right)^2 + V(\vec{r}) \quad \vec{p} = -i\hbar \vec{\nabla} \quad \vec{A} = \frac{1}{2} H (-y \vec{e}_x + x \vec{e}_y)$$

$$\mathcal{H} = -\frac{\hbar^2}{2m} \nabla^2 + \frac{e\hbar H}{2imc} \left( x \frac{\partial}{\partial y} - y \frac{\partial}{\partial x} \right) + \frac{e^2 H^2}{8mc^2} (x^2 + y^2) + V(\vec{r}) \quad (e > 0)$$

$$-\frac{\partial \mathcal{H}}{\partial H} = \frac{e}{2mc} \left( x \frac{\hbar}{i} \frac{\partial}{\partial y} - y \frac{\hbar}{i} \frac{\partial}{\partial x} \right) - \frac{e^2 H}{4mc^2} (x^2 + y^2) \quad \xrightarrow{\text{blue arrow}} \quad -\mu_B \vec{\ell} \cdot \vec{H} = -\vec{\mu}_\ell \cdot \vec{H}$$

$$\vec{\mu}_\ell = -\frac{e\hbar}{2mc} \vec{\ell} = -\mu_B \vec{\ell}$$

orbital magnetic  
moment

$$\frac{\ell}{\hbar} \rightarrow \ell$$

$$\mu_d = -\frac{e^2 H}{4mc^2} (x^2 + y^2)$$

$$\mu_d = -\frac{e^2 H}{4mc^2} \langle x^2 + y^2 \rangle = -\frac{e^2 H}{6mc^2} \langle r^2 \rangle$$

diamagnetic moment

## (5-2-3) Spin magnetic moment

Spin angular momentum

Stern-Gerlach, anomalous Zeeman's effect,

Doublet of the D-line in sodium

Zeeman's energy term in Dirac equation

$$\frac{\hbar e}{2mc} \vec{\sigma} \cdot \vec{H} = \mu_B 2\vec{s} \cdot \vec{H} = g\mu_B \vec{s} \cdot \vec{H}$$

spin angular momentum  $\vec{\mu}_s = -g\mu_B \vec{s} \quad \frac{s}{\hbar} \rightarrow s$

$$s_z |m_s\rangle = \begin{cases} \frac{\hbar}{2} |m_s\rangle & m_s = \frac{\hbar}{2} \\ -\frac{\hbar}{2} |m_s\rangle & m_s = -\frac{\hbar}{2} \end{cases} \quad (\vec{s})^2 = s(s+1)$$

---

Contribution from the  $i$ 'th electron  $\vec{\mu}_i = -(2\vec{s}_i + \vec{l}_i)\mu_B$

## Dirac equation (as a reference, see Appendix 4)

$$\left\{ p_0 + \frac{e}{c} A_0 - \rho_1 \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right) - \rho_3 mc \right\} \Psi = 0$$

$$p_0 = i\hbar \frac{\partial}{\partial(ct)}$$

$$\vec{p} = -i\hbar \frac{\partial}{\partial \vec{r}}$$

$$- eA_0 (\equiv V(\vec{r}))$$

$A_0$  **scalar potential**

$\vec{A}$  **vector potential**

$\rho_1, \rho_3, \sigma$  **4 × 4 matrixes**

$$\sigma_1 = \begin{pmatrix} \sigma_x & 0 \\ 0 & \sigma_x \end{pmatrix} \quad \sigma_2 = \begin{pmatrix} \sigma_y & 0 \\ 0 & \sigma_y \end{pmatrix} \quad \sigma_3 = \begin{pmatrix} \sigma_z & 0 \\ 0 & \sigma_z \end{pmatrix}$$

$$\rho_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \rho_2 = \begin{pmatrix} 0 & i \\ i & 0 \end{pmatrix} \quad \rho_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

$$\Psi = \begin{pmatrix} \varphi_1 \\ \varphi_2 \\ \varphi_3 \\ \varphi_4 \end{pmatrix} = \begin{pmatrix} \varphi_L \\ \varphi_S \end{pmatrix} \quad \varphi_L = \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} \quad \varphi_S = \begin{pmatrix} \varphi_3 \\ \varphi_4 \end{pmatrix}$$



### (5-3) Zeeman energy

$$\left[ \frac{1}{2m} \left( \vec{p} + \frac{e}{c} \vec{A} \right)^2 + V + \frac{\hbar e}{2mc} \vec{\sigma} \cdot \vec{H} \quad \vec{H} = \vec{\nabla} \times \vec{A} \right.$$

**Zeeman energy**

$$+ \frac{\hbar}{4m^2 c^2} \vec{\sigma} \cdot \left\{ (\text{grad } V) \times \left( \vec{p} + \frac{e}{c} \vec{A} \right) \right\}$$

**Spin-orbit interaction**

$$+ \frac{\hbar^2}{8m^2 c^2} \text{div} (\text{grad } V) \Big] \varphi_L = (\varepsilon - mc^2) \varphi_L$$

**Darwin term**

$$\varphi_L = \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix}$$

Wave function of  
2-component spinor

## (5-4) Distance and interaction between magnetic carriers, magnetic and crystal structures

Magnetic dipole term in Breit's interaction

$$E_{\text{Breit}}^{\text{magnetic dipole}} = \frac{e^2}{4m^2c^2} \frac{\vec{\sigma}_1 \cdot \vec{\sigma}_2 - 3(\vec{\sigma}_1 \cdot \hat{r}_{12})(\vec{\sigma}_2 \cdot \hat{r}_{12})}{r_{12}^3}$$

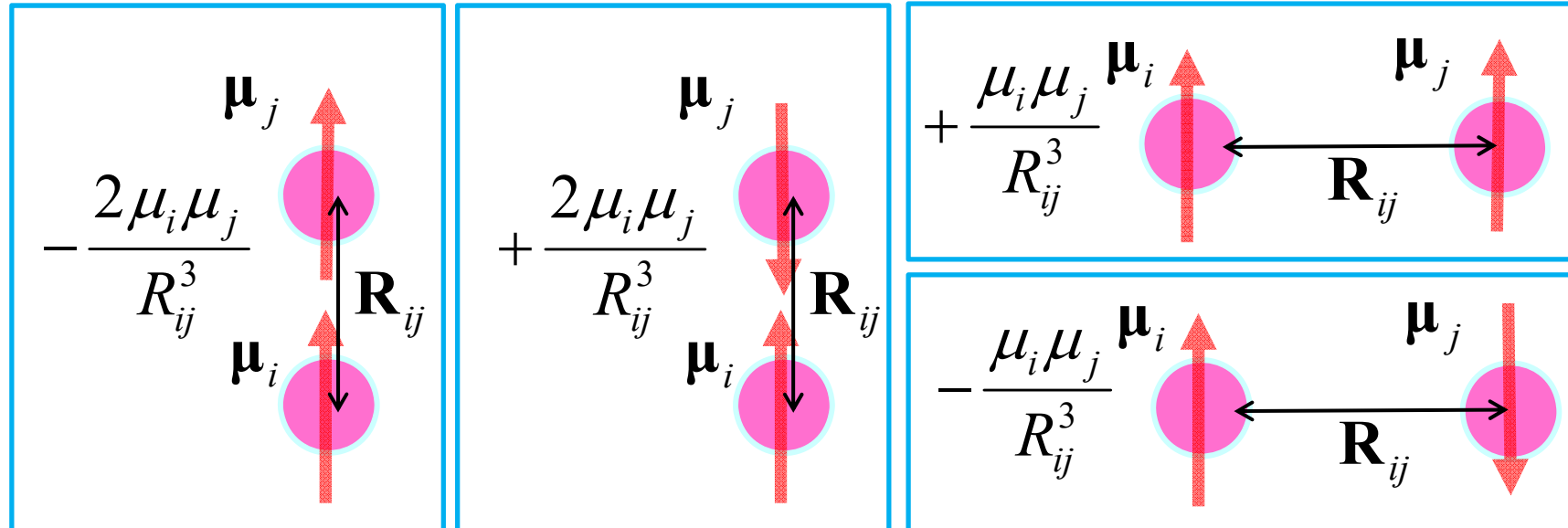
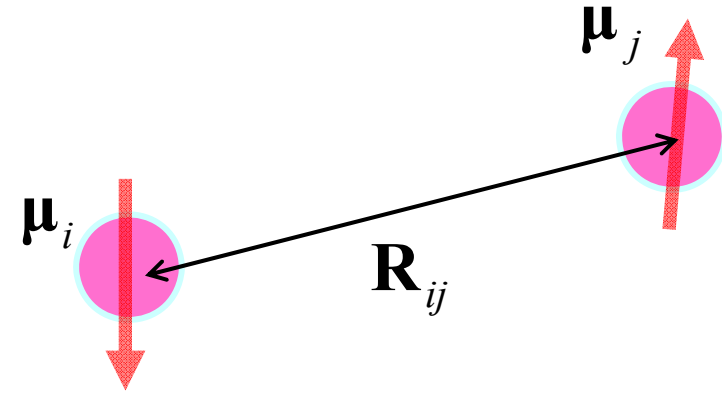
Interaction between atomic magnetic moments

$$E_{ij}^{\text{dipole}} = \frac{e^2}{4m^2c^2} \frac{\vec{\mu}_i \cdot \vec{\mu}_j - 3(\vec{\mu}_i \cdot \hat{R}_{ij})(\vec{\mu}_j \cdot \hat{R}_{ij})}{R_{ij}^3}$$

## (5-4-2) Magnetic dipole-dipole interaction (MDDI)

Classical magnetic interaction

$$E_{ij}^{\text{dipole}} = \frac{\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j - 3(\boldsymbol{\mu}_i \cdot \hat{\mathbf{R}}_{ij})(\boldsymbol{\mu}_j \cdot \hat{\mathbf{R}}_{ij})}{R_{ij}^3}$$



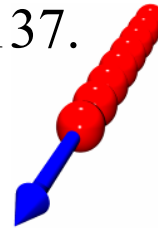
## (5-4-3) MDDI: Ferromagnetic 1D-chain and 2D-square lattice

### ➤ 1D-chain

$$E_{\text{dd}}(\theta) = \frac{e^2}{4m^2c^2} \frac{C_M \mu^2}{a^3} \left( \frac{3}{2} \cos^2 \theta - \frac{1}{2} \right) \quad C_M = 4.804$$

$\mu$  : Magnetic moment in  $\mu_B$

$$E_{\text{dd}}(\theta) [\text{Ryd}] \quad m = e = 1 \quad c = 137.$$



$$E_{\text{MAE}}^{\text{dd}} = 0.08 \text{ meV/Fe}$$

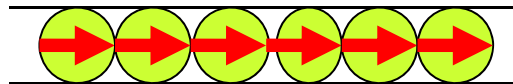
$$\mu = 3 \mu_B \quad a = 2.77 \text{ \AA}$$

### ➤ 2D-dimensional case

2D Madelung constant approach

Ref.) L. Szunyogh et al., Phys. Rev. B, **51**, 9552, (1995)

2D square lattice



$$E_{\text{MAE}}^{\text{dd}} = 0.15 \text{ meV/Fe}$$

$$\mu = 3.1 \mu_B$$

$$a = 2.87 \text{ \AA}$$

$$E_{\text{dd}}(\theta) = \frac{e^2}{4m^2c^2} \frac{C_M \mu^2}{a^3} \left( \frac{3}{2} \cos^2 \theta - \frac{1}{2} \right)$$

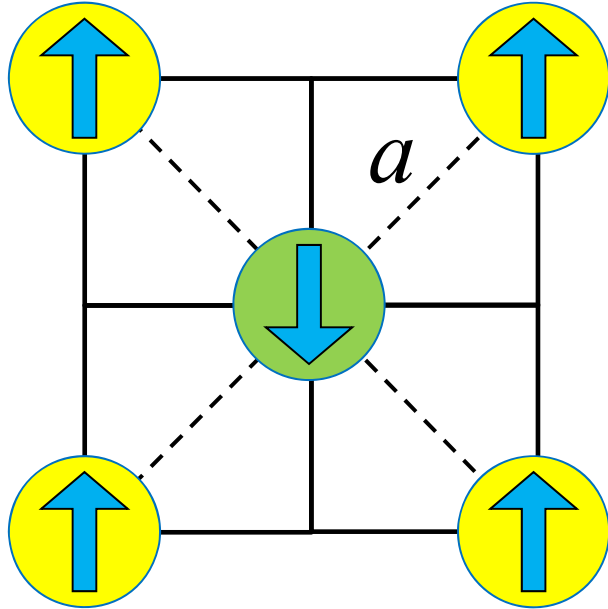
$$C_M = 9.03362$$

$\mu$  : Magnetic moment in  $\mu_B$

$a$  : Lattice constant in  $a_B$

$$E_{\text{dd}}(\theta) [\text{Ryd}] \quad m = e = 1 \quad c = 137.$$

## (5-4-4) MDDI: Antiferromagnetic 2D-square lattice



Perpendicular anisotropy

$$E_{\text{MAE}}^{\text{dd}} = -0.082 \text{ meV/Mn}$$

$$\mu = 4.2 \mu_{\text{B}}$$

$$a = 2.83 \text{ \AA}$$

Note)

$$E_{\text{MAE}}^{\text{dd}} = -0.175 \text{ meV/Mn (SDA)}$$

SDA: Spin density approach (see Appendix 7).

TO, I. Pardede et al, IEEE Trans. Magn., 55(2), Art. Num. 1300104, (2019).

## (5-5) Spin-orbit interaction

$$H_{\text{SOI}} = \frac{\hbar}{4m^2c^2} \vec{\sigma} \cdot \left( \text{grad } V(\vec{r}) \times \vec{p} \right) \quad \vec{\sigma} \text{ Pauli's matrix}$$

$$V(\mathbf{r}) \approx -\frac{Ze^2}{r} \quad \text{grad } V(r) \approx \frac{dV}{dr} \frac{\mathbf{r}}{r}$$

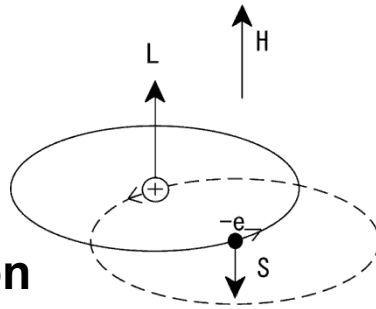
The effect is emphasized at surface/interface: becoming not spherical.

$$H_{\text{SOI}} = \xi \vec{l} \cdot \vec{\sigma} = \xi (l_x \sigma_x + l_y \sigma_y + l_z \sigma_z)$$

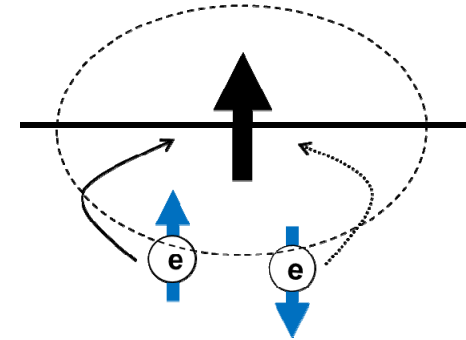
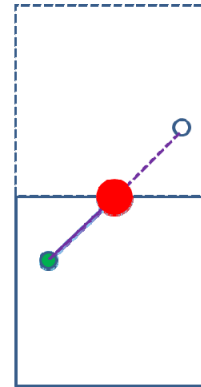
$$\xi(r) = \frac{\hbar^2}{4m^2c^2r} \frac{dV}{dr} \quad \text{connects orbital and spin spaces}$$

2

**Biot-Savart law  
in the classical  
electromagnetics  
spin-orbit interaction**



$$E_{\text{SO}}^j = \lambda \vec{l} \cdot \vec{s} = \frac{\lambda}{2} \{j(j+1) - s(s+1) - l(l+1)\}$$



Due to the surface/interface the inversion symmetry breaks.

➔ Rashba effect, etc.

## (5-5-2) Spin-texture in the reciprocal space: Rashba

effect 1

2 dimensional free electron with spin-orbit interaction

$$H = -\frac{\hbar^2}{2m} \nabla^2 + \vec{\sigma} \cdot (\vec{\alpha} \times \vec{p}) \quad \vec{\alpha} \propto \left\langle \text{grad } V(r) = \frac{1}{r} \frac{dV(r)}{dr} \mathbf{r} \right\rangle$$

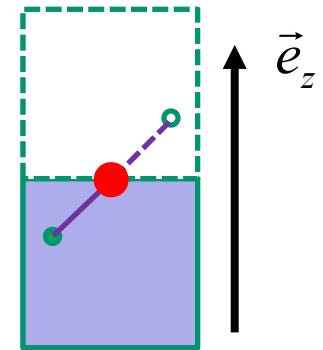
Plane wave  $\varphi_{\vec{k}} = \frac{1}{\sqrt{\Omega}} e^{i\vec{k} \cdot \vec{r}} \begin{pmatrix} u_1 \\ u_2 \end{pmatrix} \propto \vec{e}_z$

$$H = \frac{\hbar^2 k^2}{2m} + \vec{\sigma} \cdot (\vec{\alpha} \times \vec{k}) = \frac{\hbar^2 k^2}{2m} + \alpha_z (\vec{k} \times \vec{\sigma})_z$$

$$= \frac{\hbar^2 k^2}{2m} + \alpha_z (k_x \sigma_y - k_y \sigma_x) \quad \vec{k} = (k_x, k_y) \quad k = \sqrt{k_x^2 + k_y^2}$$

$$E_+ = \frac{\hbar^2 k^2}{2m} + \alpha_z k \quad \varphi_{\vec{k}}^+ = \frac{1}{\sqrt{\Omega}} e^{i\vec{k} \cdot \vec{r}} \frac{1}{\sqrt{2}} \begin{pmatrix} -1 \\ -ie^{i\theta_k} \end{pmatrix} \propto \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i(\theta_k + \pi/2)/2} \\ e^{i(\theta_k + \pi/2)/2} \end{pmatrix}$$

$$E_- = \frac{\hbar^2 k^2}{2m} - \alpha_z k \quad \varphi_{\vec{k}}^- = \frac{1}{\sqrt{\Omega}} e^{i\vec{k} \cdot \vec{r}} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -ie^{i\theta_k} \end{pmatrix} \propto \frac{1}{\sqrt{2}} \begin{pmatrix} -e^{-i(\theta_k + \pi/2)/2} \\ e^{i(\theta_k + \pi/2)/2} \end{pmatrix}$$

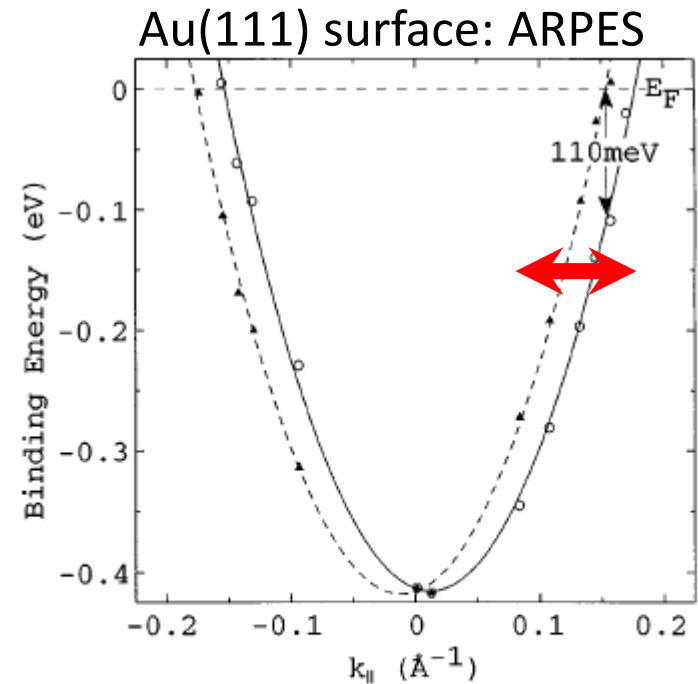
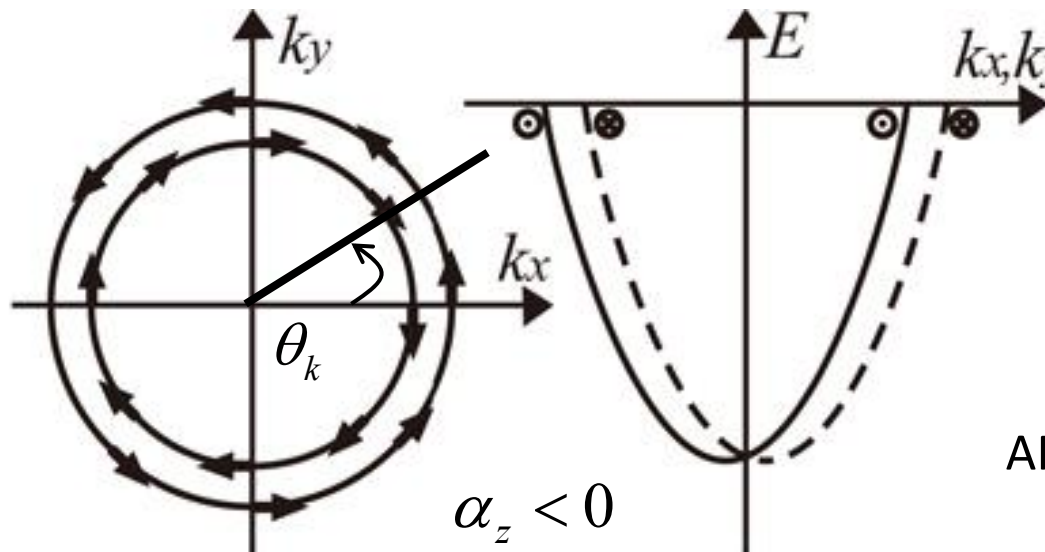


## (5-5-3) Spin-texture in the reciprocal space: Rashba effect 2

$$\langle \varphi_{\vec{k}}^+ | \vec{\sigma} | \varphi_{\vec{k}}^+ \rangle = (-\sin \theta_k, \cos \theta_k, 0)$$

$$\langle \varphi_{\vec{k}}^- | \vec{\sigma} | \varphi_{\vec{k}}^- \rangle = (\sin \theta_k, -\cos \theta_k, 0)$$

$$E_{\pm} = \frac{\hbar^2}{2m} \left( k \pm \frac{m\alpha_z}{\hbar^2} \right)^2 - \frac{\hbar^2}{2m} \left( \frac{m\alpha_z}{\hbar^2} \right)^2$$



➤ High resolution

Lashell et al., Phys. Rev. Lett.  
77, 3419 (1996)

ARPES: Angle Resolved  
Photoelectron Spectroscopy



## (5-5-4) Spin-orbit interaction 2

$$H_{\text{SOI}} = \frac{\hbar}{4m^2c^2} \vec{\sigma} \cdot \left\{ \left( \vec{\nabla} V(\vec{r}) \right) \times \vec{p} \right\}$$

$$= \frac{\hbar}{4m^2c^2} \left\{ \left( -\frac{\partial V}{\partial z} \right) (\sigma_x p_y - \sigma_y p_x) + \sigma_z \left( \frac{\partial V}{\partial x} p_y - \frac{\partial V}{\partial y} p_x \right) + \left( \sigma_x \frac{\partial V}{\partial y} - \sigma_y \frac{\partial V}{\partial x} \right) p_z \right\}$$

**Normal Rashba term**

**General Rashba term**

**Effective in-plane potential gradient**

**Spin-orbit interaction (Rashba term + damping rate term)**

## (5-5-5) Interpretation of Spin-orbit interaction

$$H_{\text{SOI}} = \frac{\hbar}{4m^2c^2} \vec{\sigma} \cdot \left\{ \left( \vec{\nabla} V(\vec{r}) \right) \times \vec{p} \right\}$$

$$H_{\text{SOI}} = 2 \frac{\hbar e}{2mc} \frac{\vec{\sigma}}{2} \cdot \left\{ \frac{1}{2mce} \left( \vec{\nabla} V(\vec{r}) \right) \times \vec{p} \right\} = 2\mu_{\text{B}} \frac{\vec{\sigma}}{2} \cdot \left\{ \frac{1}{2mce} \left( \vec{\nabla} V(\vec{r}) \right) \times \vec{p} \right\}$$

**Bohr magneton**

**Effective magnetic field**

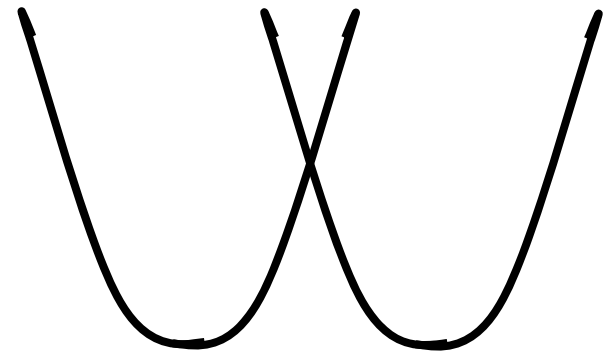
$$H_{\text{SOI}} = \frac{\hbar}{4m^2c^2} \left\{ \vec{\sigma} \times \vec{\nabla} V(\vec{r}) \right\} \cdot \vec{p} = \frac{\hbar}{m} \frac{1}{4mc^2} \left\{ \vec{\sigma} \times \vec{\nabla} V(\vec{r}) \right\} \cdot \vec{p}$$

**Effective wave number**

$$H_{\text{SOI}} = -e \frac{\hbar}{4m^2c^2} (\vec{\sigma} \times \vec{p}) \cdot \frac{1}{e} \vec{\nabla} V(\vec{r})$$

**Effective electric dipole**

**Electric field**



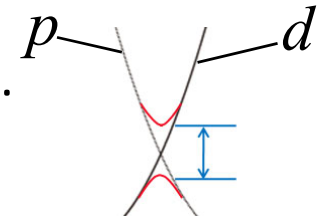
## (5-5-6) Electric field effects in electronic structures of the surface/interface

### ➤ Stark effect

$$H_{\text{ED}} = -e\vec{r} \cdot \vec{E}_{\text{ext}}$$

Electric dipole

Orbital coupling between the states of different angular quantum number  $\Delta\ell = \pm 1$ .



### ➤ Rashba (SOI) effect

$$H_{\text{SOI}} = -e \frac{\hbar}{4m^2 c^2} (\vec{\sigma} \times \vec{p}) \cdot \vec{E}_{\text{ext}}$$

Modification of the Rashba effect.

### ➤ Electron depletion (accumulation)

When imposing the EF,

for PDOS  $\sim 1$  states/eV

Induced change in the number of electrons  $\sim 0.01$   $\sim 0.01$  eV

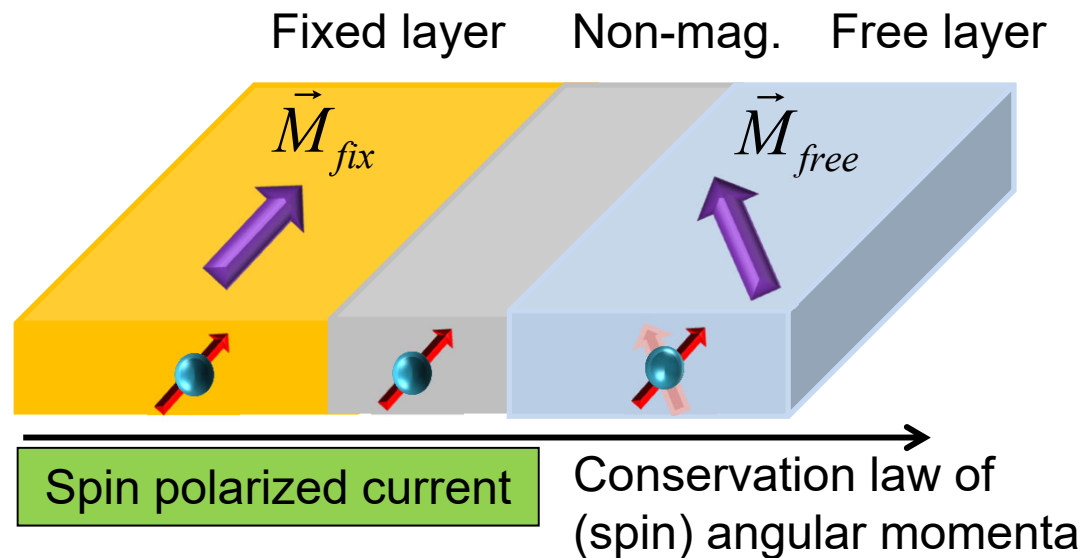
Induced change in the number of electrons  $\sim 0.1$   $\sim 0.1$  eV

by lattice constant, or modification surface/interface, etc.

## (5-6) Spin transfer torque

**Spin current** produced in fixed layer  $\longrightarrow$  **Spin torque** provided to free layer

$$N_{\text{stt}} = \frac{d\vec{M}_{\text{free}}}{dt} \propto -I_s \vec{M}_{\text{free}} \times (\vec{M}_{\text{free}} \times \vec{M}_{\text{fix}})$$



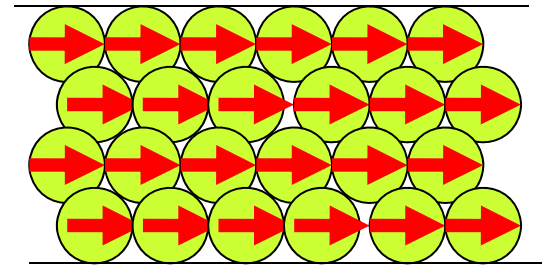
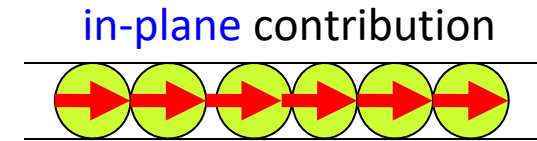
(5-7) Magnetic anisotropy energy:  
electron orbital, magnet shape

**Magnetostatic contribution**

$$E_{d-d} = \frac{1}{c^2} \sum_{\vec{R}_i, \vec{R}_j}^{i \neq j} \left\{ \frac{\vec{m}(\vec{R}_i) \cdot \vec{m}(\vec{R}_j)}{R_{ij}^3} - 3 \frac{[\vec{m}(\vec{R}_i) \cdot (\vec{R}_i - \vec{R}_j)][\vec{m}(\vec{R}_j) \cdot (\vec{R}_i - \vec{R}_j)]}{R_{ij}^5} \right\}$$

2D square lat.  
Shape aniso.

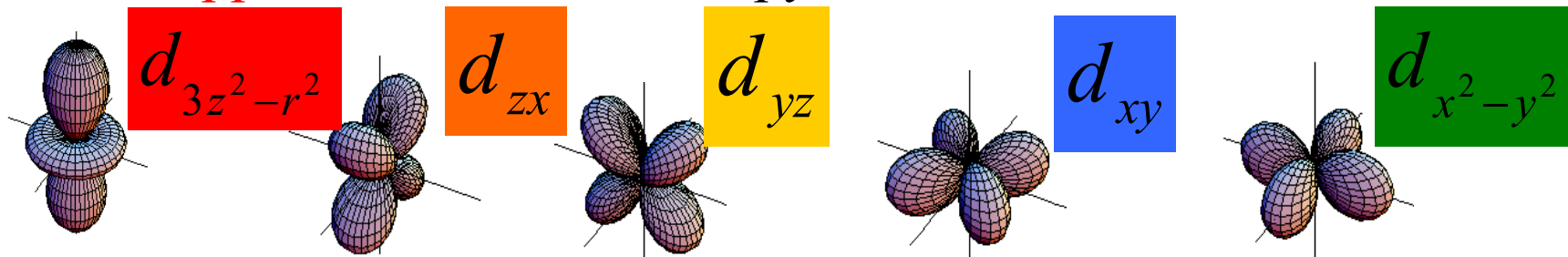
This depends on the arrangement of magnetic atoms, not so depend on electric field.



**Electronic structure contribution**

perturbation of spin-orbit interaction,  
MA **appears** from an anisotropy of orbitals

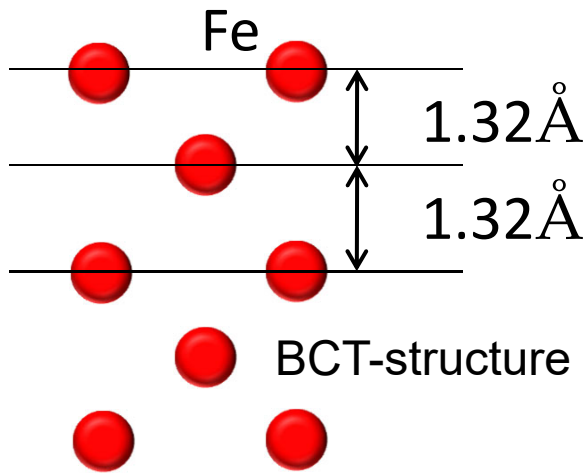
$$H_{\text{SOI}} = \xi \vec{\ell} \cdot \vec{\sigma}$$



It is important to see the behavior of each angular orbitals.  
Anisotropic occupation of electrons leads to MA.

# MAE from d-d interaction for Fe-multilayers

In-plane lattice constant  
MgO 4.21Å



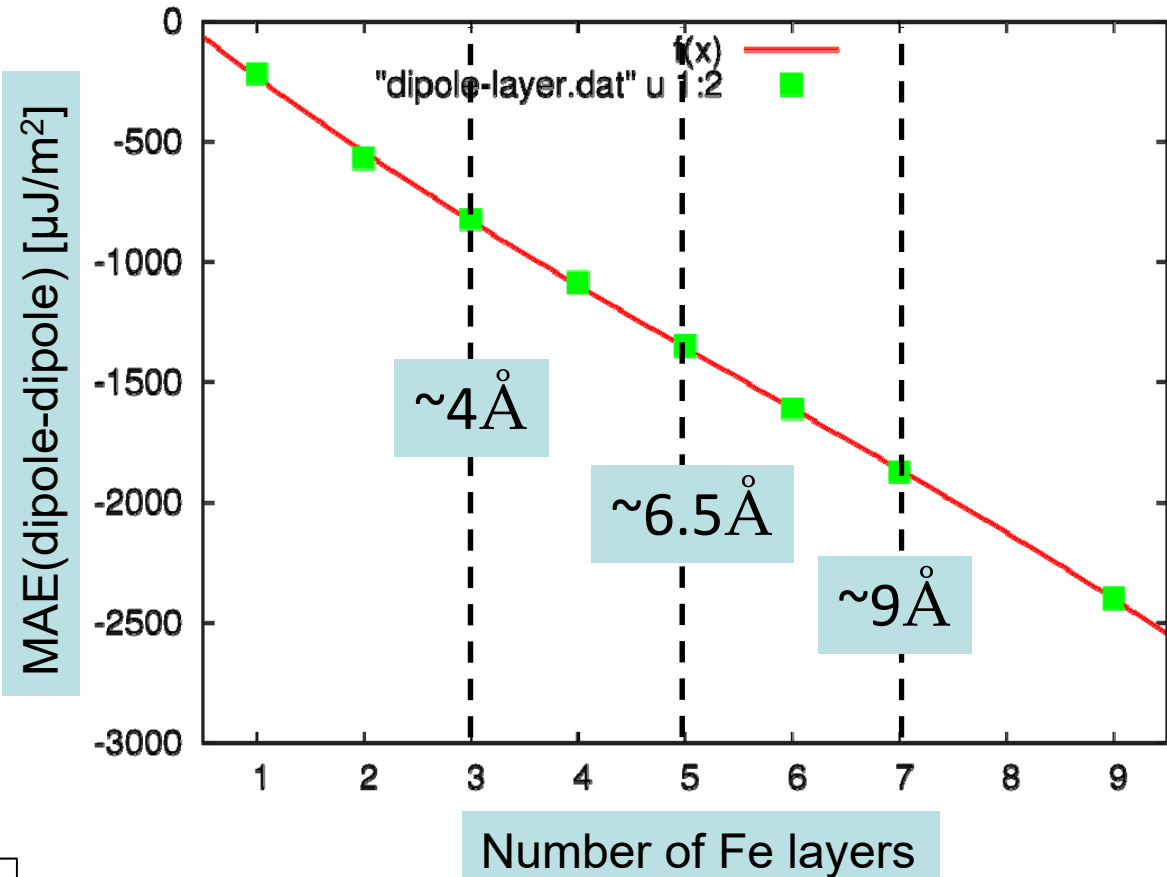
Fe: 2.96  $\mu_B$  (interface)  
2.63  $\mu_B$  (inside)

FeCo layer

> 6.3 Å in-plane

< 6.3 Å out-of-plane

Y. Shiota et. al., Appl. Phys.  
Exp., 4, 043305(2011).



In-plane MAE increases  
by 250-300 $\mu\text{J}/\text{m}^2$  per Fe-layer

Ref.)L. Szunyogh et al., Phys. Rev. B, **51**, 9552, (1995)

# Origin of MAE in electronic structure

$$H_{\text{SOI}} = \xi \vec{l} \cdot \vec{\sigma}$$

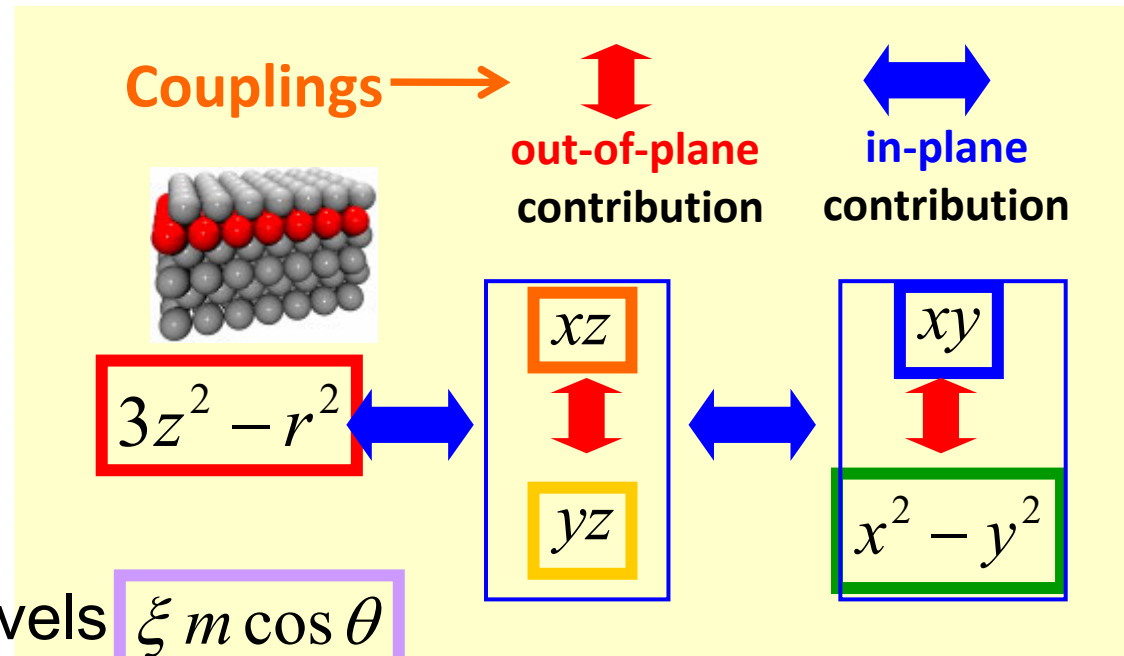
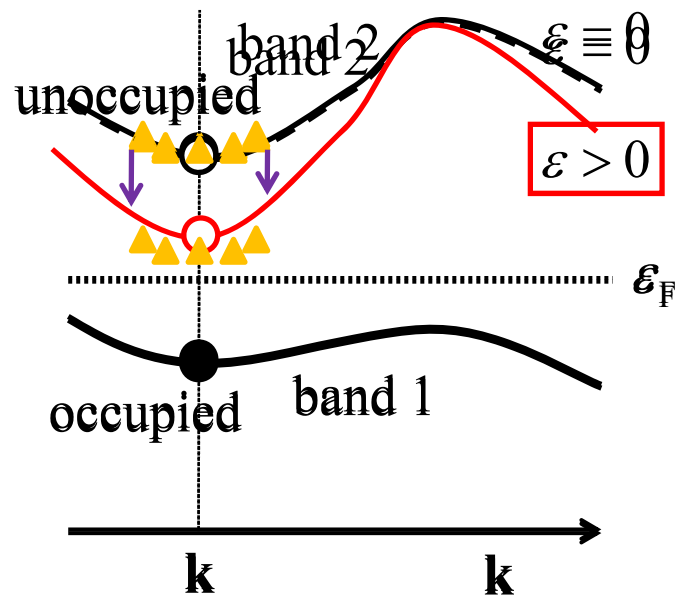
• spin-orbit coupling contribution from band electrons

matrix element

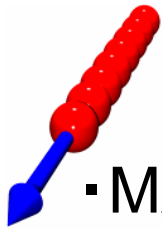
(A) 2nd perturbative contributions

$$\text{MAE} = E_x - E_z \approx (\xi)^2 \sum_{o,u} \frac{|\langle o_{\downarrow} | l_z | u_{\downarrow} \rangle|^2 - |\langle o_{\downarrow} | l_x | u_{\downarrow} \rangle|^2}{\epsilon_u^{\downarrow} - \epsilon_o^{\downarrow}}$$

z	$\langle xz   l_z   yz \rangle = 1$ $\langle x^2 - y^2   l_z   xy \rangle = 2$
x	$\langle 3z^2 - r^2   l_x   yz \rangle = \sqrt{3}$ $\langle xy   l_x   xz \rangle = 1$ $\langle x^2 - y^2   l_x   yz \rangle = 1$
y	$\langle 3z^2 - r^2   l_y   xz \rangle = \sqrt{3}$ $\langle xy   l_y   yz \rangle = 1$ $\langle x^2 - y^2   l_y   xz \rangle = 1$



(B) Existence of partly-occupied degenerate levels  $\xi m \cos \theta$

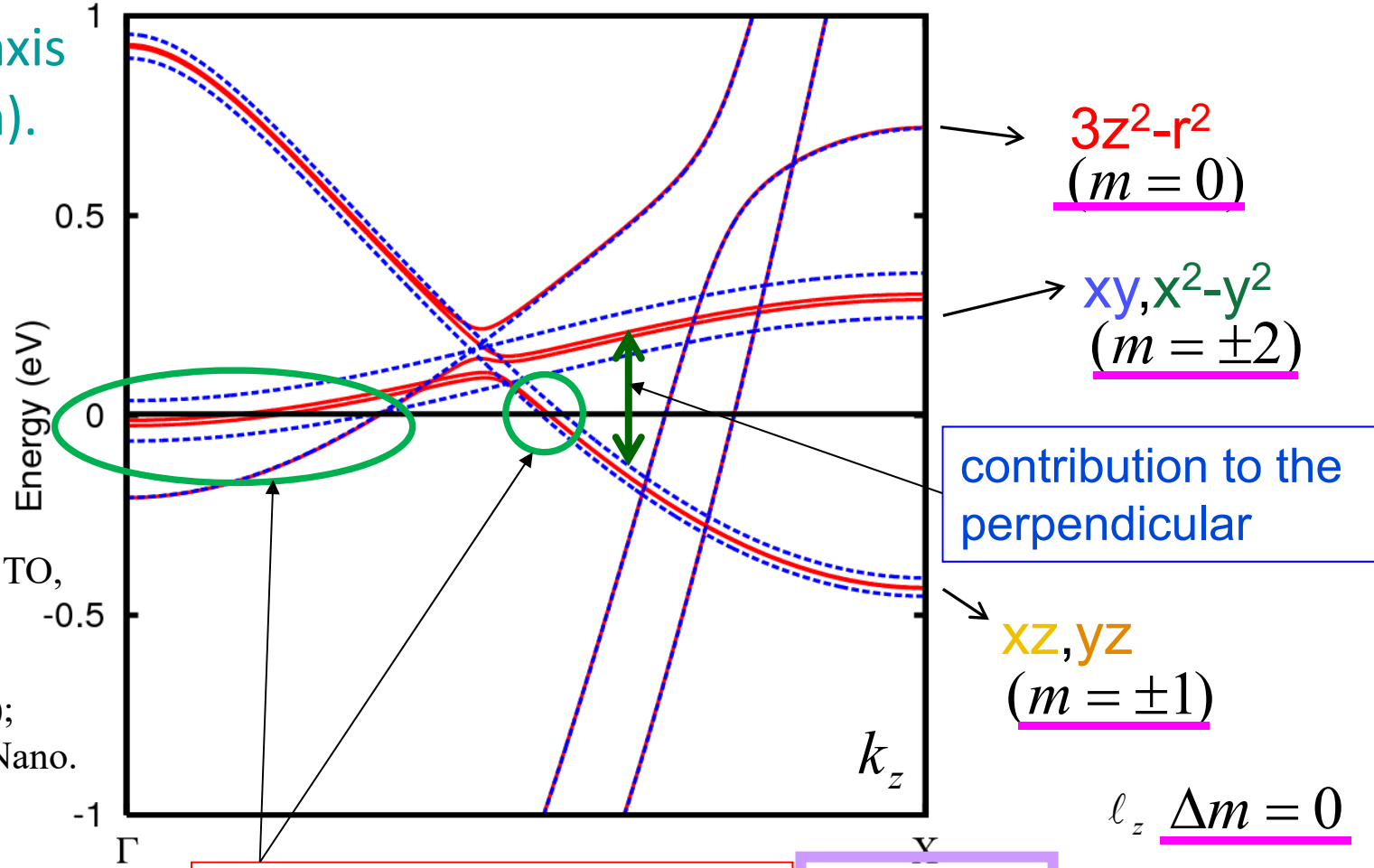


# example: Isolated iron chain

• MAE=3.23 meV/Fe

**red: mag. perpendicular to the axis**  
**blue: mag. parallel to the axis**

• Easy axis: parallel to the chain-axis (z direction).



M. Tsujikawa and TO,  
 J. Phys.: Condens. Matter,  
 21, 064213 (2009);  
 J. Compt. Theor. Nano.  
 6, 2597 (2009).

dipole: 0.05meV/Fe

contribution to the parallel

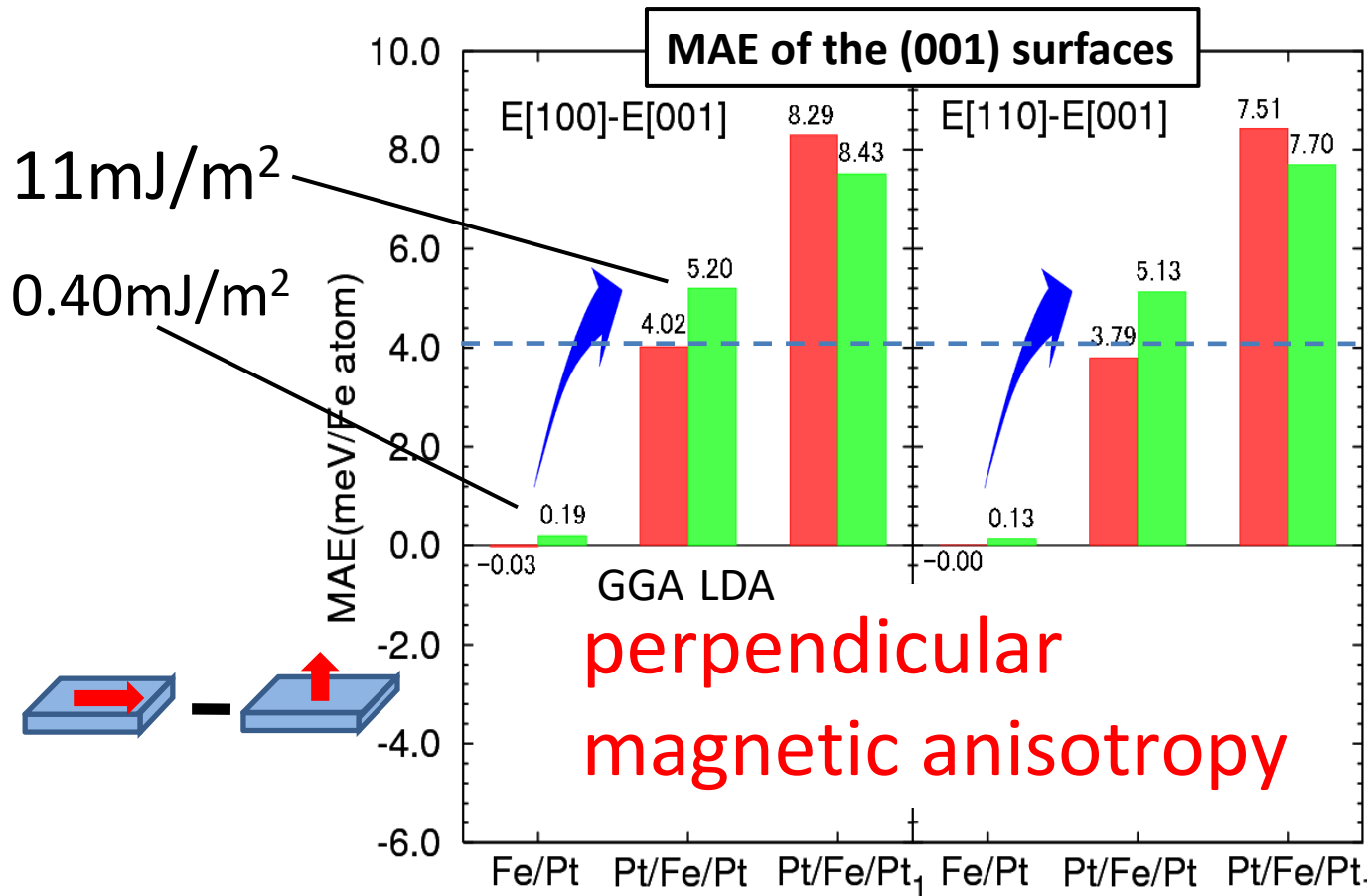
$$\xi m \cos \theta$$

$$l_z \Delta m = 0$$

$$l_x \Delta m = \pm 1$$



# (5-8) Magnetic anisotropy: in-plane, perpendicular



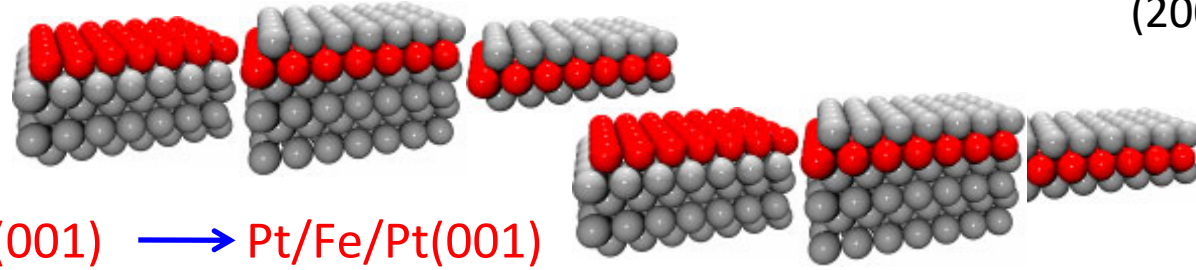
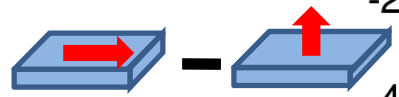
L1<sub>0</sub>-FePt regular alloy(LDA)  
MAE

2.61meV<sup>a</sup>、  
2.724meV<sup>b</sup>

<sup>a</sup>P. Ravindran *et.al*, (2001).

<sup>b</sup>TO *et.al*, (2005).

Tsujikawa *et al.* (2008, 2009)

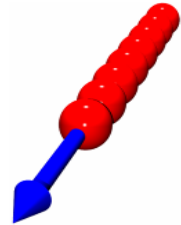


Fe/Pt(001) → Pt/Fe/Pt(001)

dipole: -0.32mJ/m<sup>2</sup>

# MAE of Fe-chain/Pt(664) surface

-3.23 meV/Fe

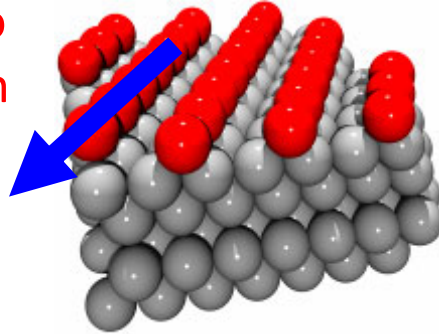


bare  
Fe-chain

parallel to  
the chain



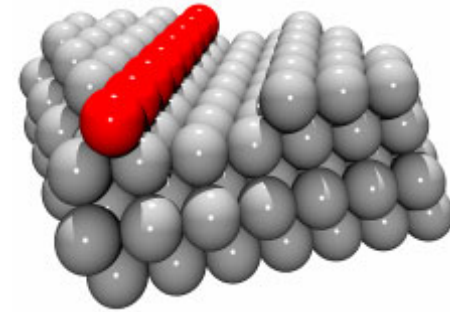
-2.25 meV/Fe



Fe-chain/Pt(111)



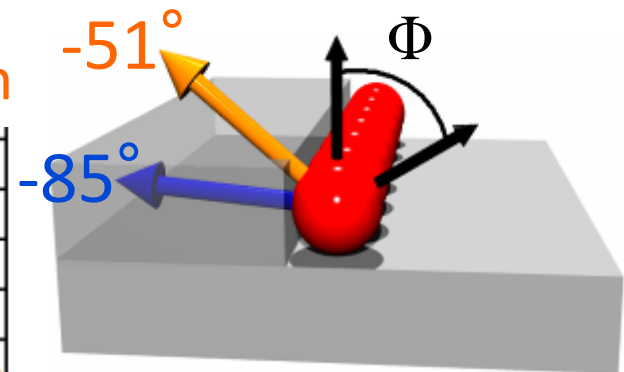
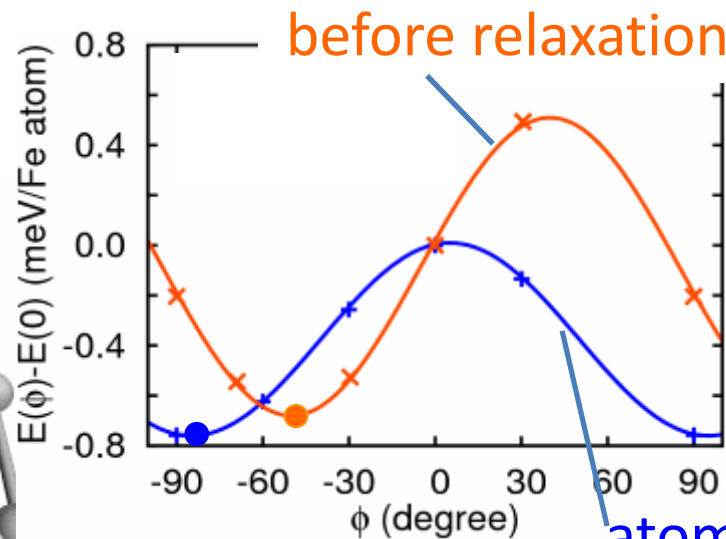
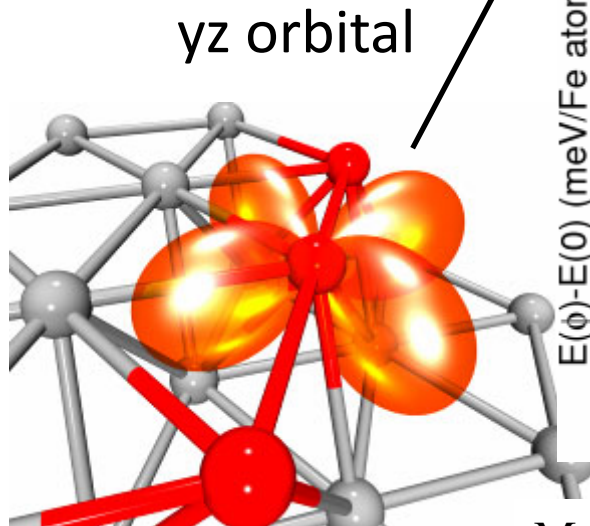
1.19 meV/Fe



Fe-chain/Pt(664)

perpendicular to the chain

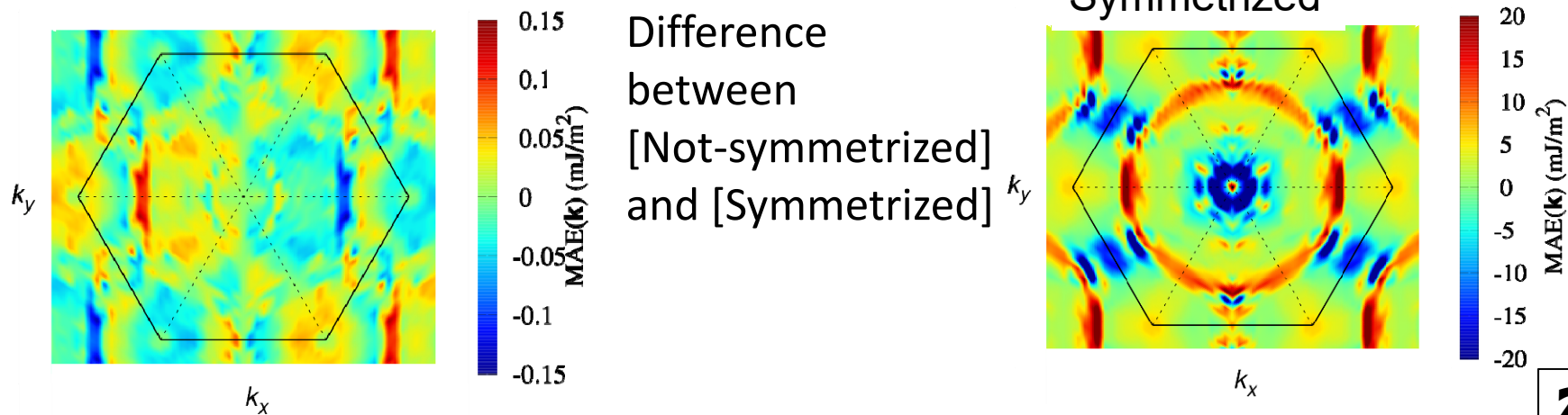
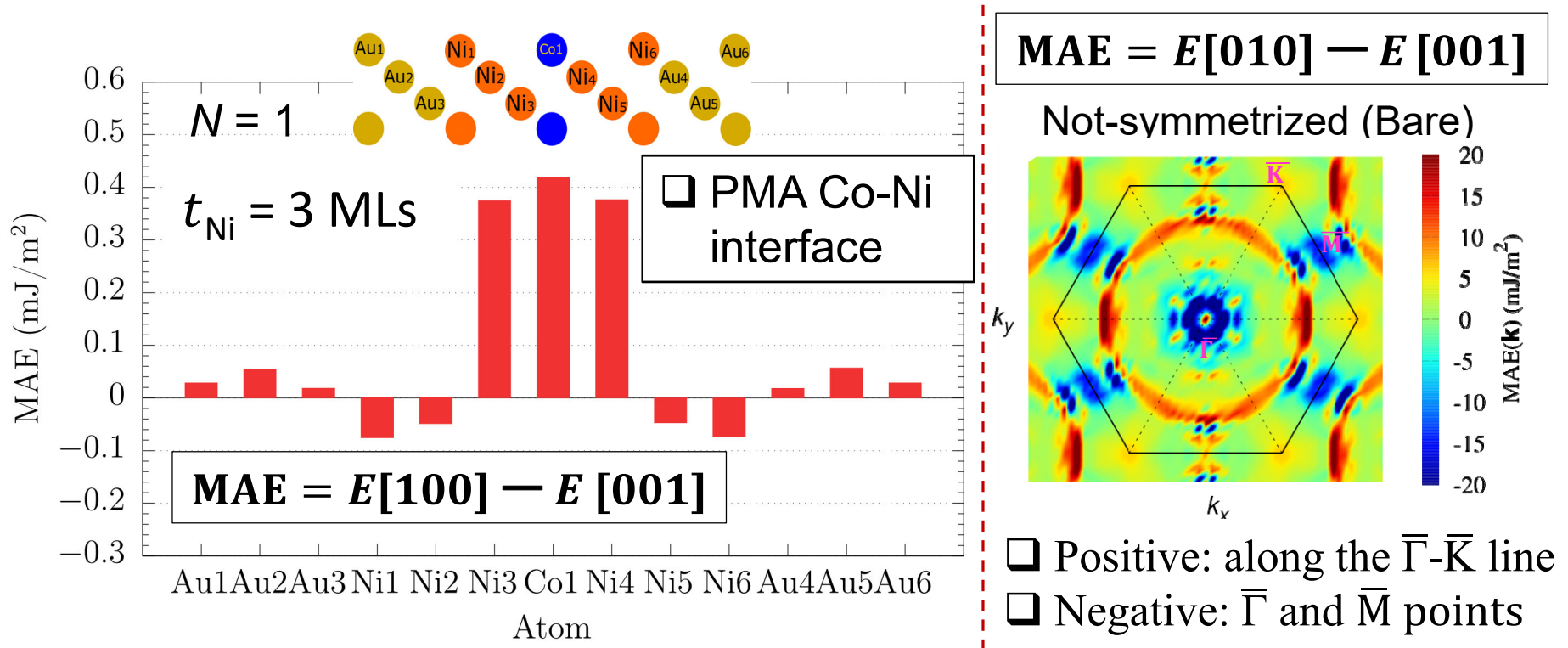
hybridization of 3d orbitals between Fe and Pt atoms



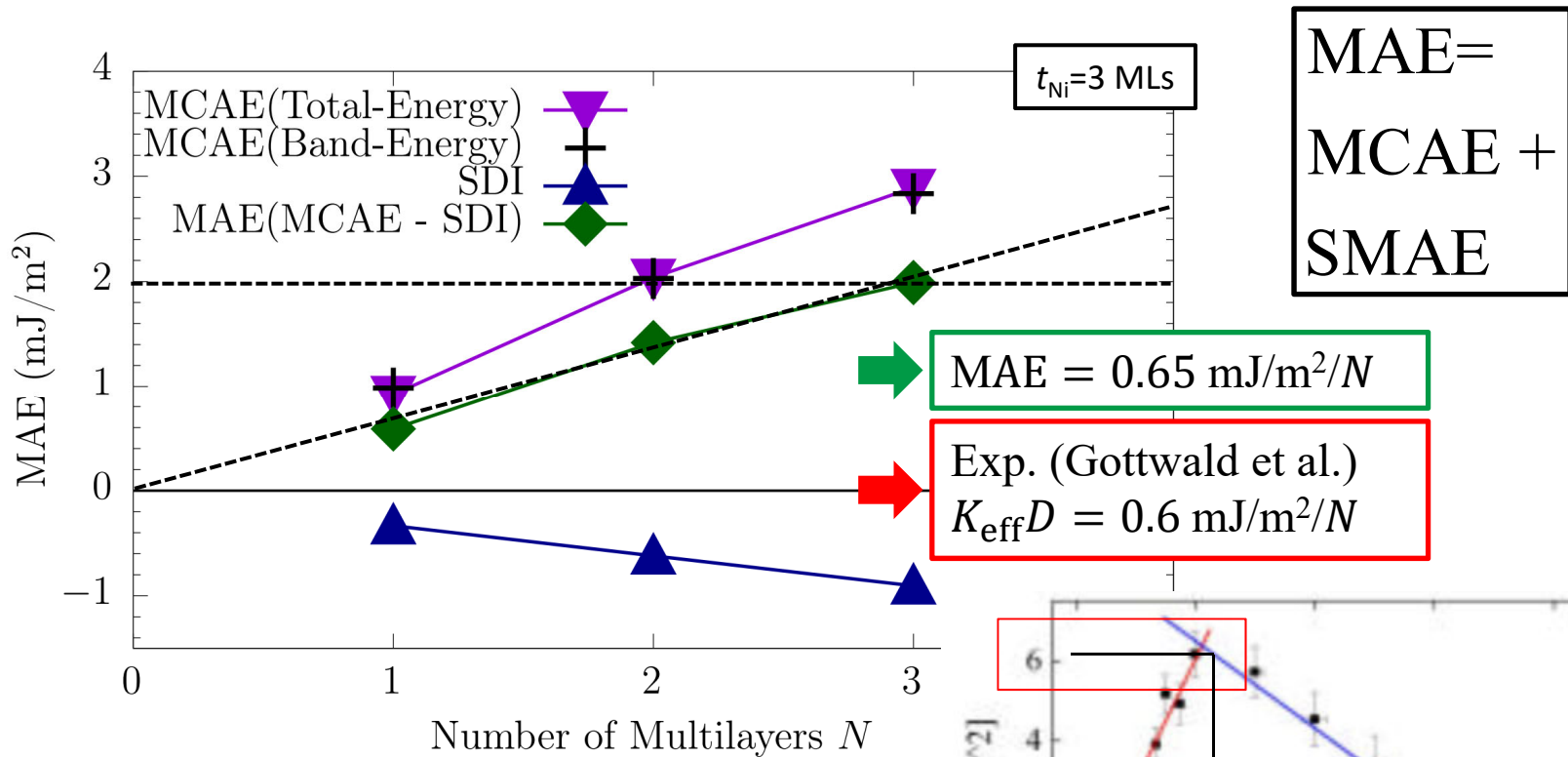
exp.  $-80^\circ$   
(Repetto et al., 2006)

atomic relaxation

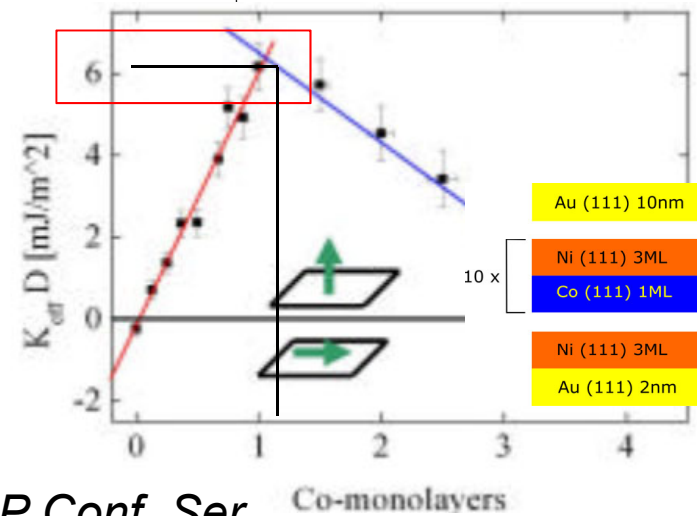
# Atom- and $\mathbf{k}$ -resolved MCAE: Au/[Ni( $t_{\text{Ni}}$ )/Co(1ML)]<sub>N</sub>/Ni( $t_{\text{Ni}}$ )/Au



# Total magnetic anisotropy (MAE): Au/Ni/Co/Ni/Au

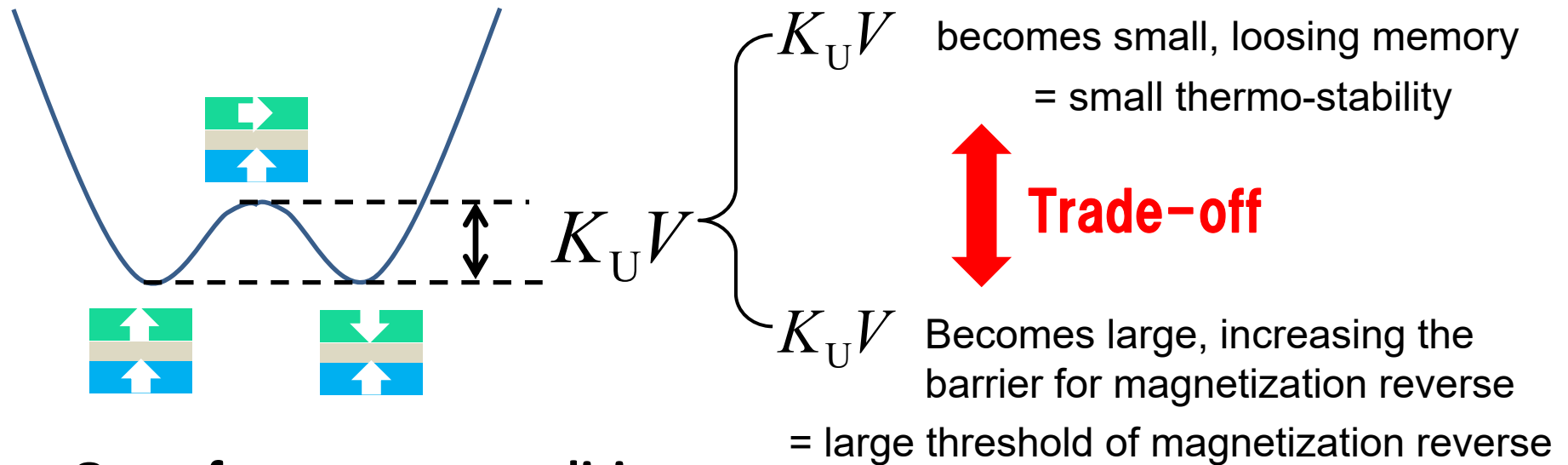


- ☐ Positive MAE indicates a PMA.
- ☐ For  $N = 3$ , the MAE reaches to the value of 2 mJ/m<sup>2</sup>.



M. Gottwald *et al*, *IOP Conf. Ser. Mater. Sci. Eng* **12**, 012018, (2010).

# Trade-off property in magnetic memory



**One of necessary condition**

**Non-volatile**

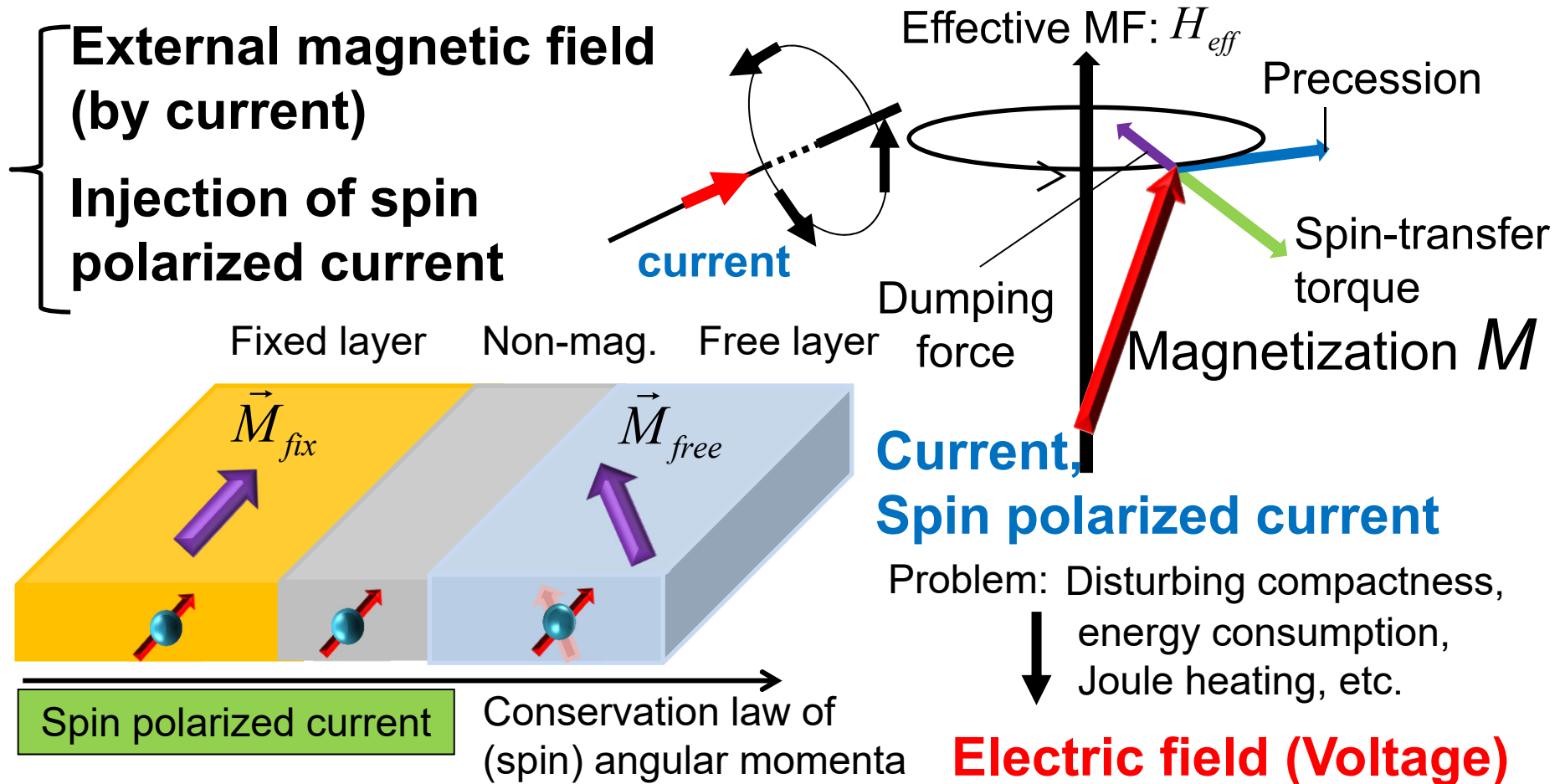
$$\Delta \geq 60 \quad (\text{ten years}) \quad \Delta = \frac{K_U V}{k_B T}$$

Heat energy

Magnetic anisotropy energy of magnetic memory

$$\left\{ \begin{array}{l} K_U \\ V \end{array} \right. \begin{array}{l} \text{Magnetic anisotropy energy per volume} \\ \text{Volume of magnet} \end{array}$$

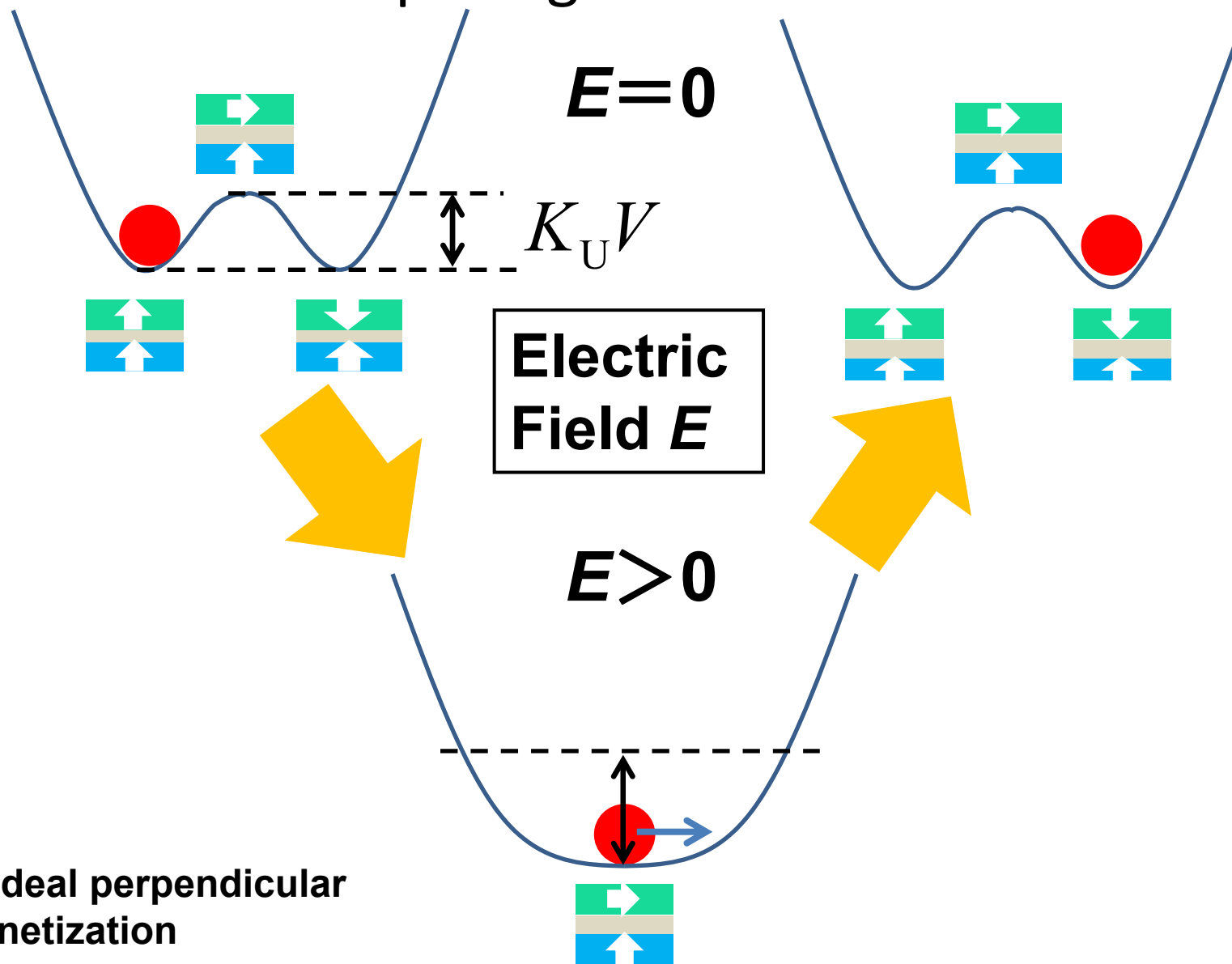
# Driving forces in dynamical control of magnetization



$$\vec{H}_{eff} = \vec{H}_{ext} + \vec{H}_{stt} + \vec{H}_{shape} + \vec{H}_{aniso}$$

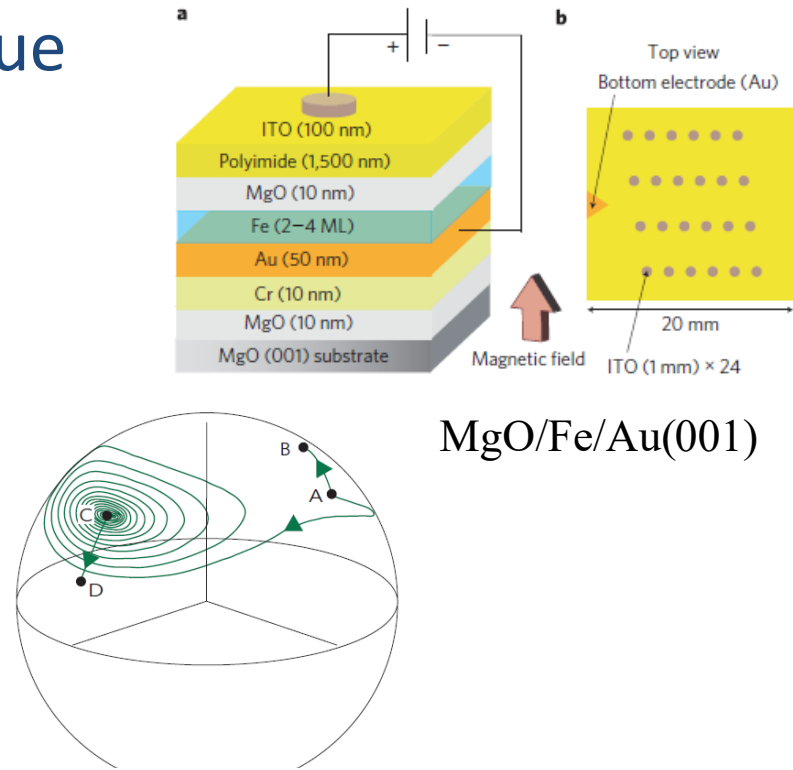
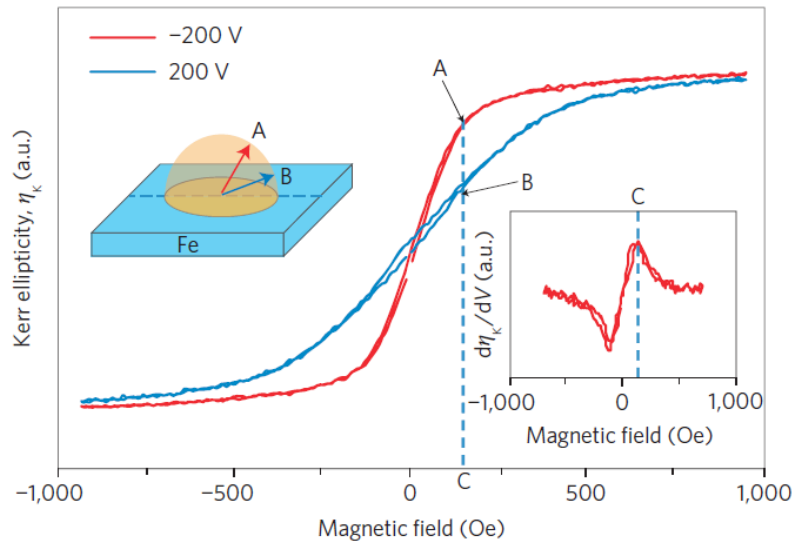
Expectation: ultra-low energy consumption, non-volatile property, compactness (high density memory), enough high speed in reading & writing

# Schematic diagram of magnetization reversal in imposing electric field



For ideal perpendicular magnetization

# (5-9) Voltage-induced spin torque



$$\frac{d\vec{M}}{dt} = \gamma\vec{M} \times \vec{H}_{eff} + \frac{\alpha\vec{M}}{M_s} \times \frac{d\vec{M}}{dt}$$

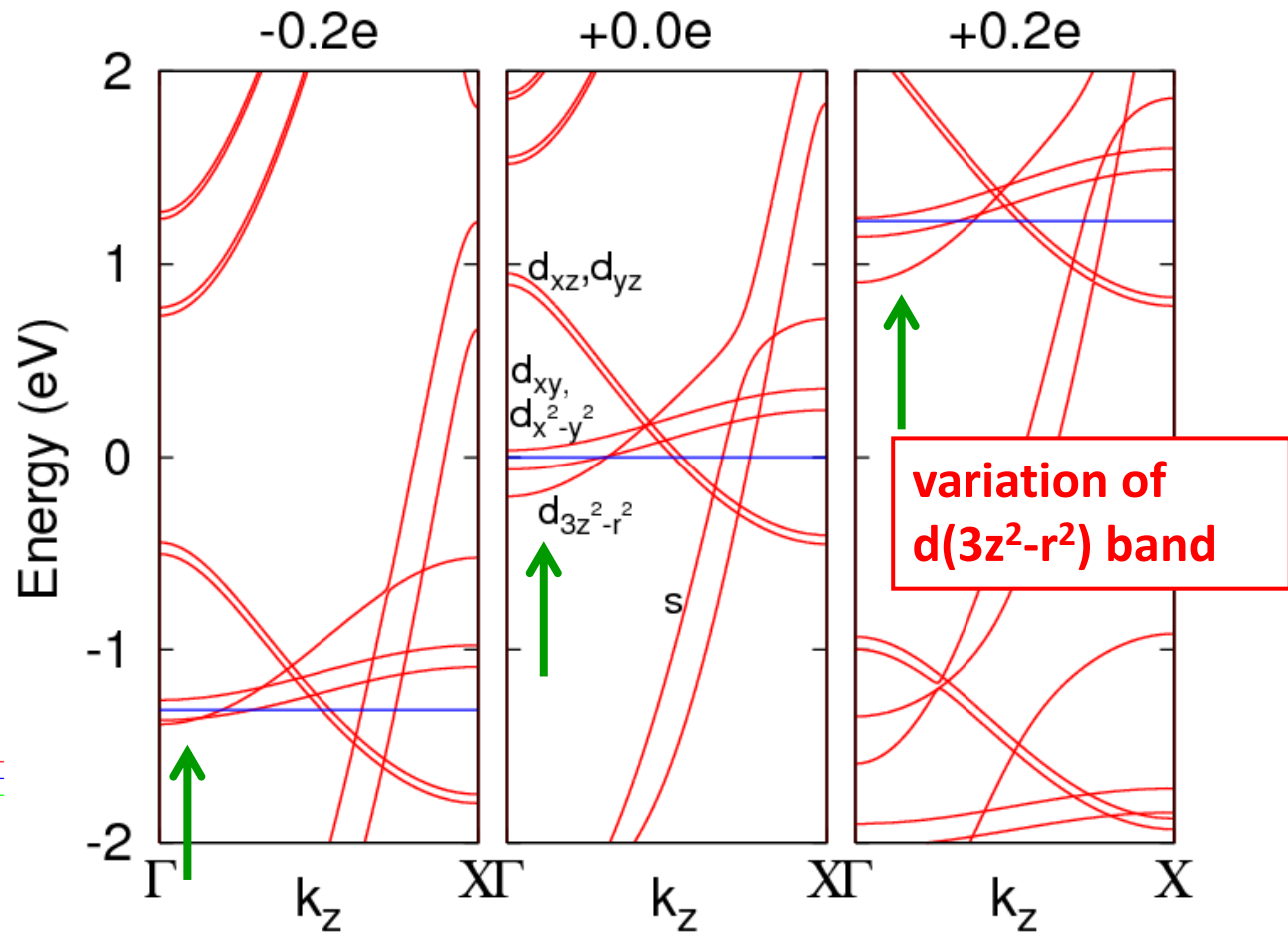
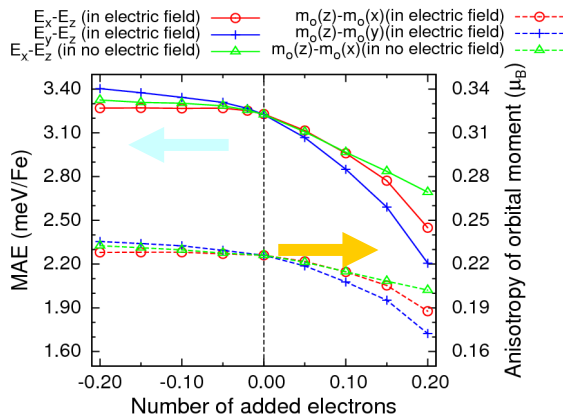
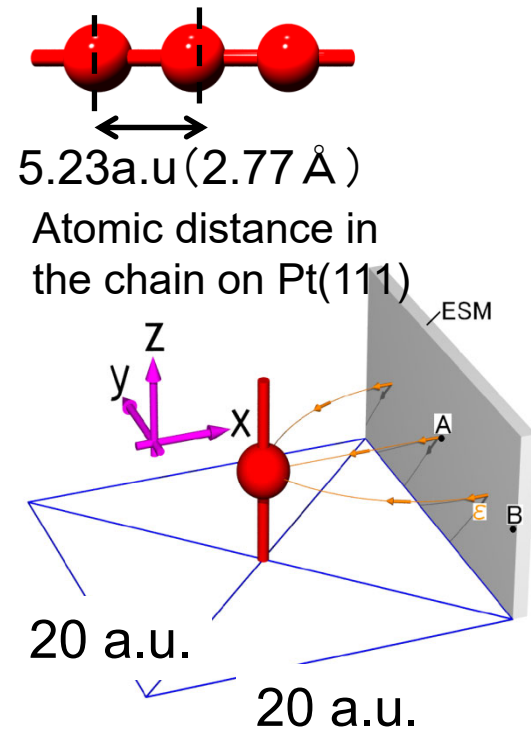
$$\vec{H}_{eff} = -\frac{1}{\mu_0 M_s} \vec{\nabla} E_{mag}$$

$$E_{mag} = -\mu_0 M_z H_{ext} + \frac{1}{2} K_u (V) \left( \frac{M_z}{M_s} \right)^2 + \frac{1}{2} K_{//} \left( \frac{M_x}{M_s} \right)^2$$

T. Maruyama et. al., Nature Nanotech. 4, 158 (2009)



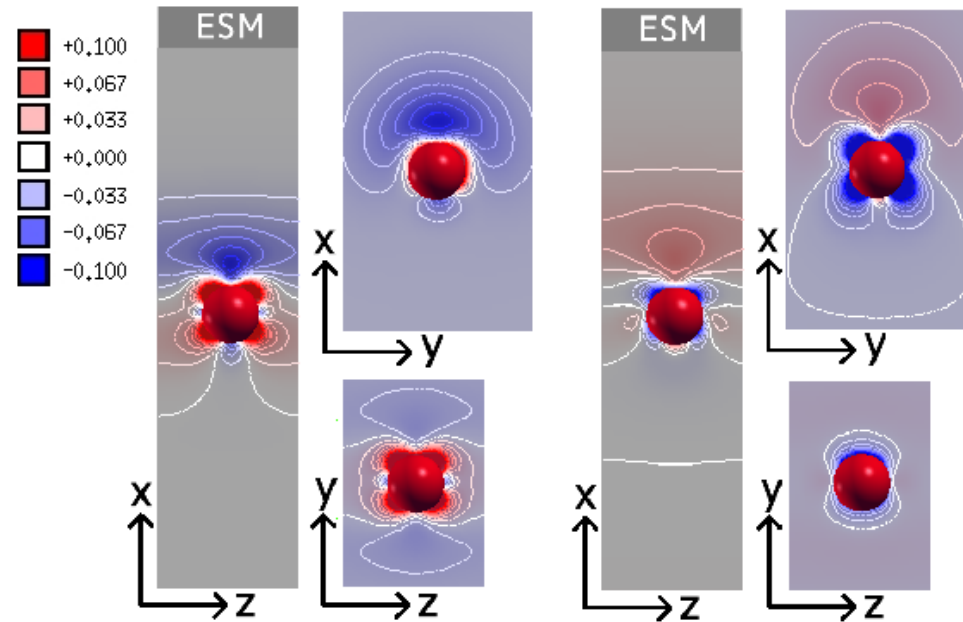
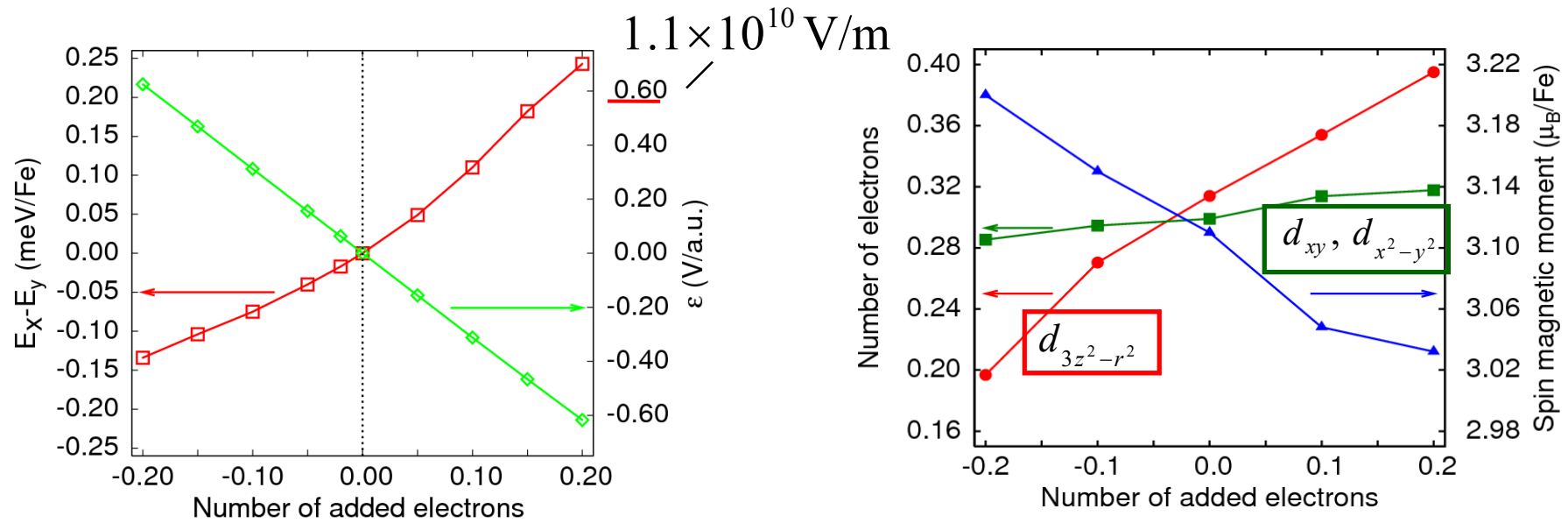
# A1-5. Applying the electric field (EF) on iron chain



EF dependence depends on the orbital direction

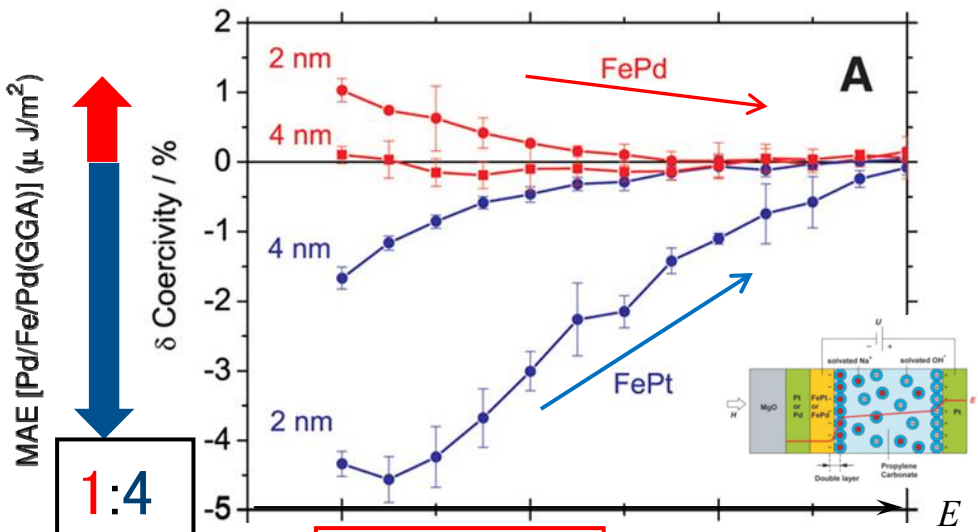
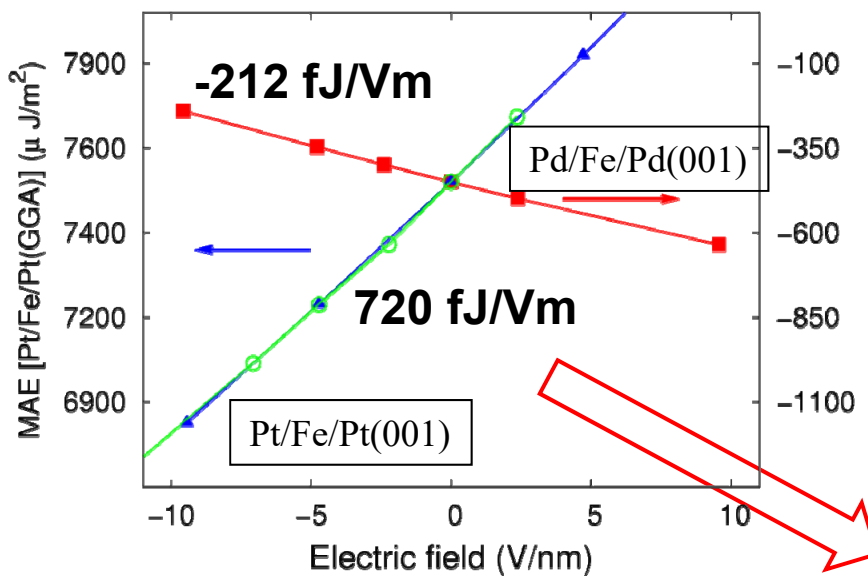
causes variations of MAE

# A1-6. Imposing the electric field (EF) on iron chain (part 2)



# MAE and EF effects (comparison with exp.)

S. Haraguchi et. al., J. Phys. D: Appl. Phys. **44**, 064005 (2011). M. Weisheit et. al., Science **315**, 349 (2007).



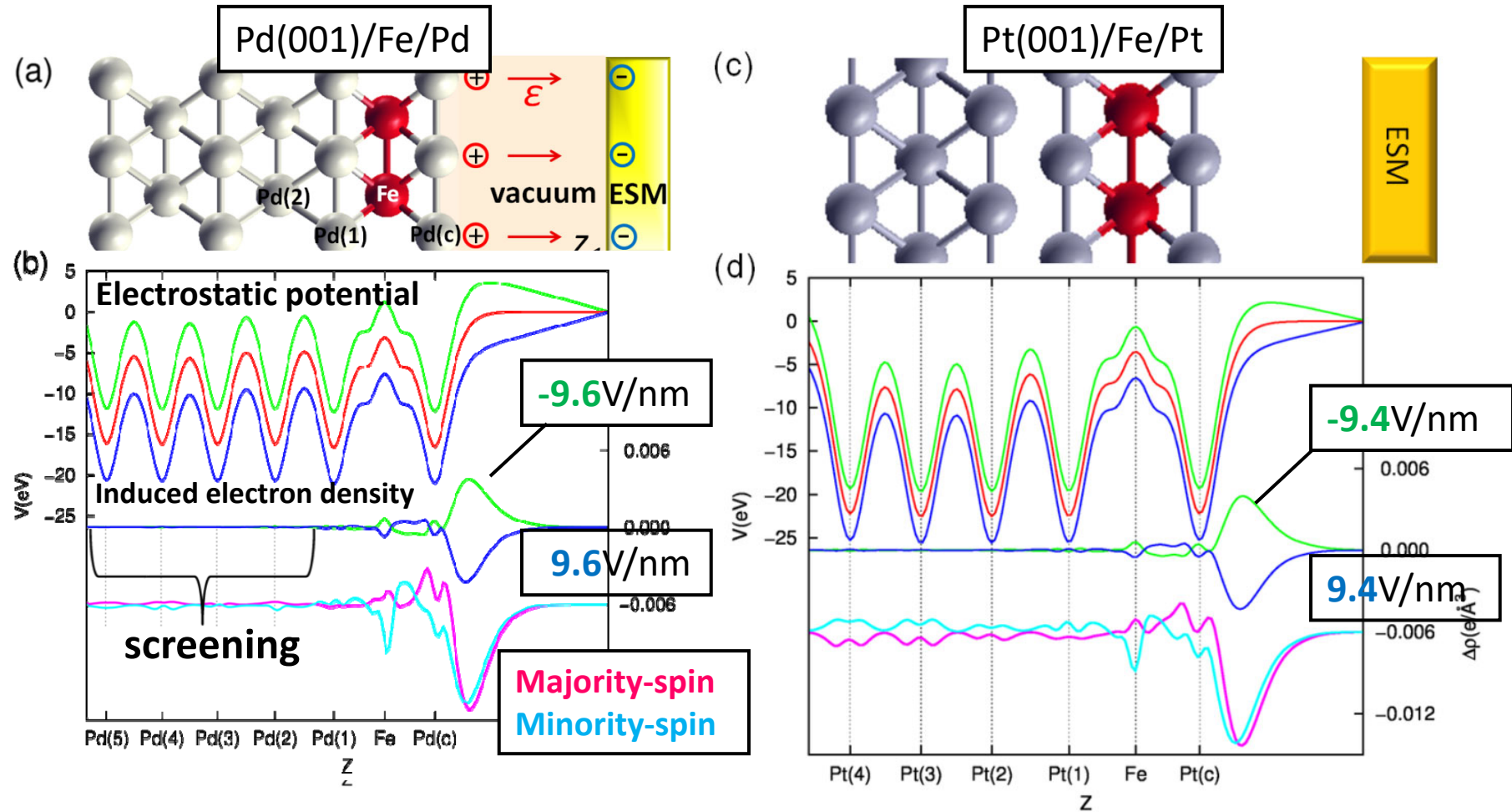
Ratio of slopes  $\frac{\gamma_{\text{Pd}}}{\gamma_{\text{Pt}}} \approx -0.27$

- Pd/Fe/Pd(001)  
MAE:  $-425 \mu\text{J}/\text{m}^2$  ( $E=0.0$  V/nm)  
EF-effect:  $-21.2$  fJ/Vm
- experiment  
EF-effect:  $-602$  fJ/Vm  
F. Bonell et al., APL, **98**, 232510(2011).

MgO  $\epsilon_r \approx 10$  (low frequency limit)  
EF-effect =  $-21.2 \times 10$  fJ/Vm =  $-212$  fJ/Vm  
↕ comparable  
 $-602$  fJ/Vm (exp.)

These signs of slopes and the ratio are in good qualitative agreement with the available experimental data.

# EF-Induced modulation on density

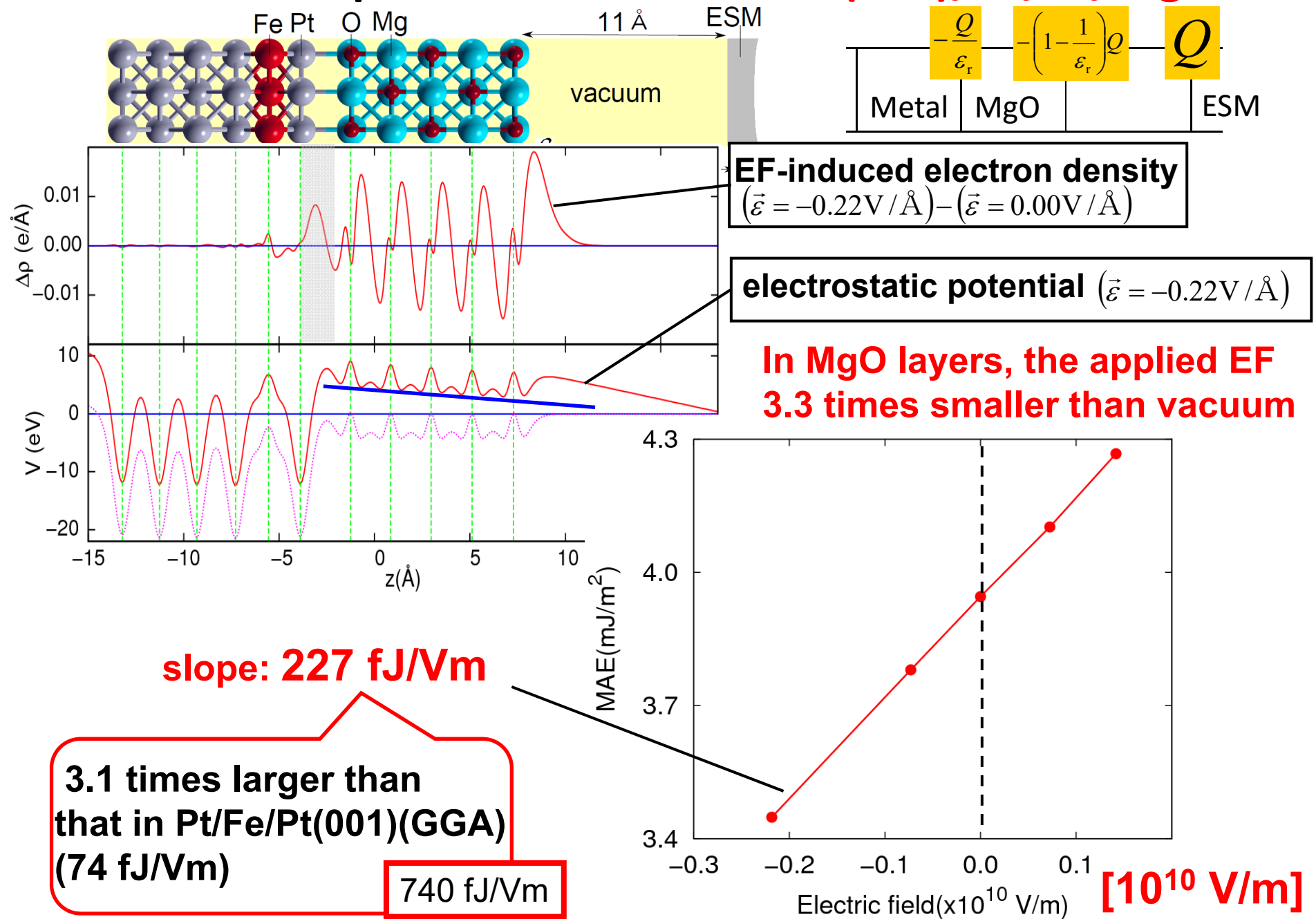


- The electric field is screened in a few number of surface layers.
- In these layers, EF effects are induced.

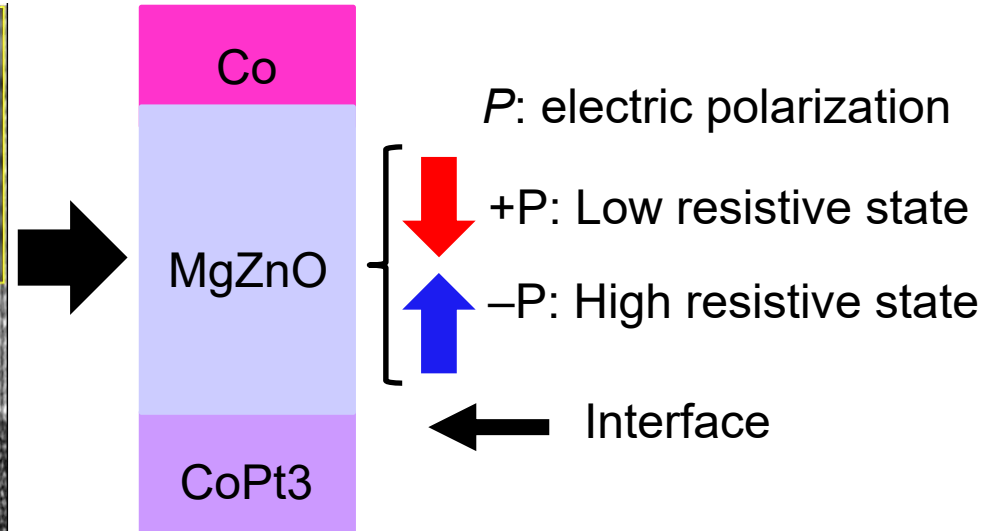
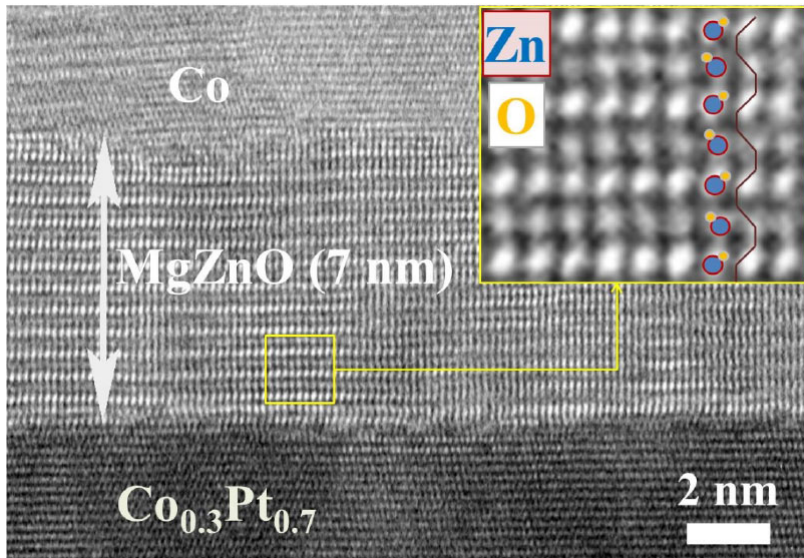
S. Haraguchi et. al., J. Phys. D: Appl. Phys. **44** (2011) 064005.

Effective Screening Medium(ESM method): Otani et al., PRB **73**,115407 (2006).

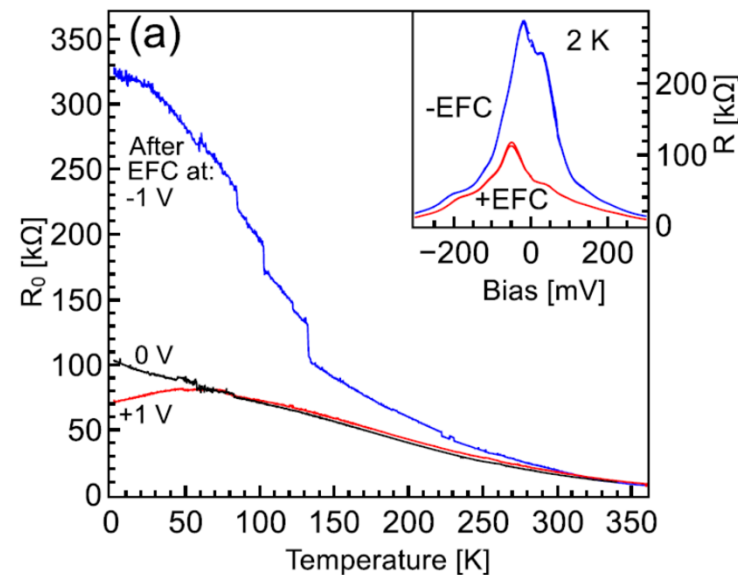
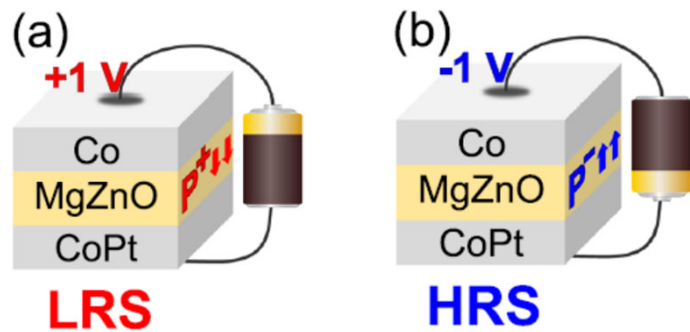
# Electric field dependence of MAE in Pt(001)/Fe/Pt/MgO film



# Interface: magnetic metal and dielectric insulator (Exp.) magnetic anisotropy change by electric polarization switching



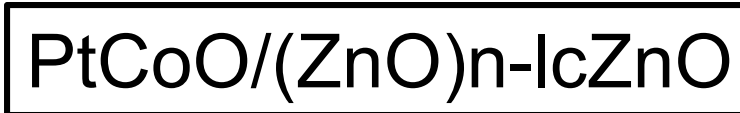
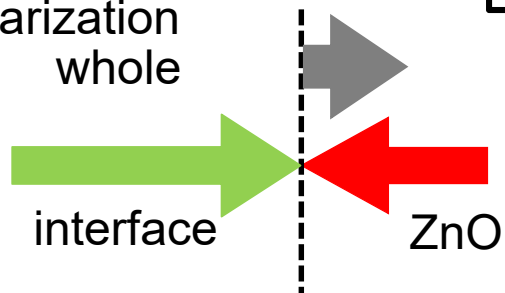
- Theory: “Multi-ferroic-like”
- Experiment: “New type MTJ”



# Interface: magnetic metal and dielectric insulator (DFT)

Low resistive state(LRS)

Polarization whole

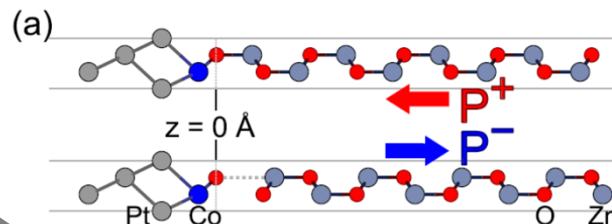


MAE(SOI, int)  
0.25 meV/cell

**0.44 mJ/m<sup>2</sup>**

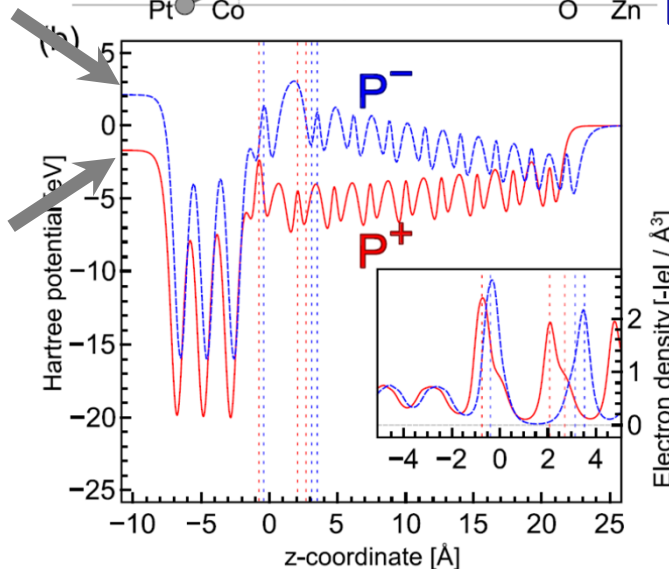
**-2.16 mJ/m<sup>2</sup>**

-1.23 meV/cell

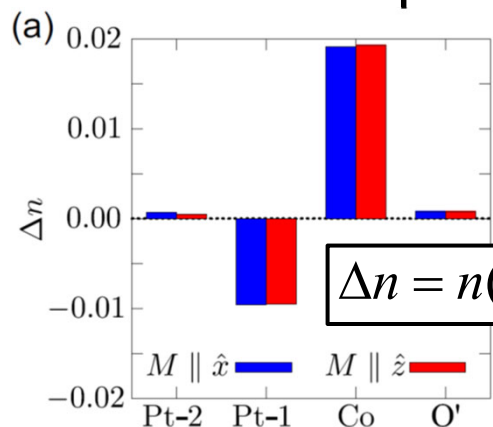


High resistive state(HRS)

Polarization whole



$$\mathbf{D}(z) = \epsilon_0 \mathbf{E}(z) + \mathbf{P}(z)$$



$$\Delta n = n(P-) - n(P+)$$

M. Al-Mahdawi et al., Phys. Rev. B **100**, 054423 (2019).

## (5-10) Landau–Lifshitz–Gilbert equation (LLG-equation)

$$\frac{d\vec{M}}{dt} = \underbrace{-\gamma(\vec{M} \times \vec{H}_{\text{eff}})}_{\text{precessional term}} + \underbrace{\frac{\alpha}{M_s} \vec{M} \times \frac{d\vec{M}}{dt}}_{\text{dumping term}} - \underbrace{\eta(\theta) \frac{\mu_B I}{eV} \frac{\vec{M}}{M_s} \times \left( \frac{\vec{M}}{M_s} \times \frac{\vec{M}_{\text{fix}}}{M_{\text{fix}}} \right)}_{\text{spin transfer torque}}$$

$\vec{M}$  :Magnetization vector at free layer       $\alpha$       :Gilbert magnetic damping factor

$\gamma$  :gyromagnetic factor       $\vec{M}_{\text{fix}}$  :Magnetization vector at fixed layer

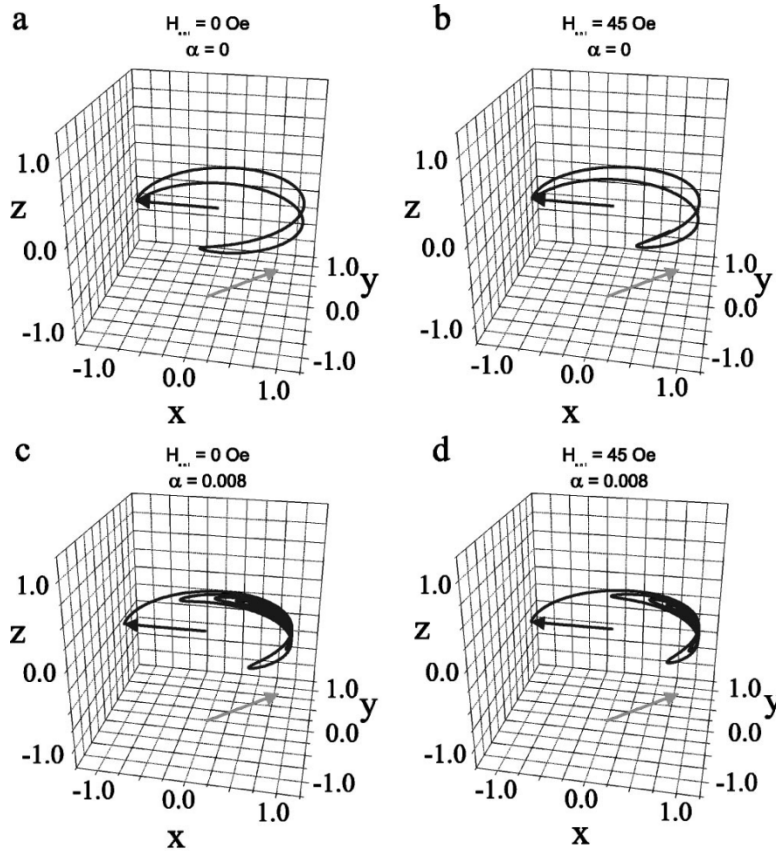
$\eta(\theta)$  :spin transfer efficiency

$$\vec{H}_{\text{eff}} = \vec{H} + \vec{H}_{\text{shape}} + \vec{H}_{\text{cry-aniso}}$$

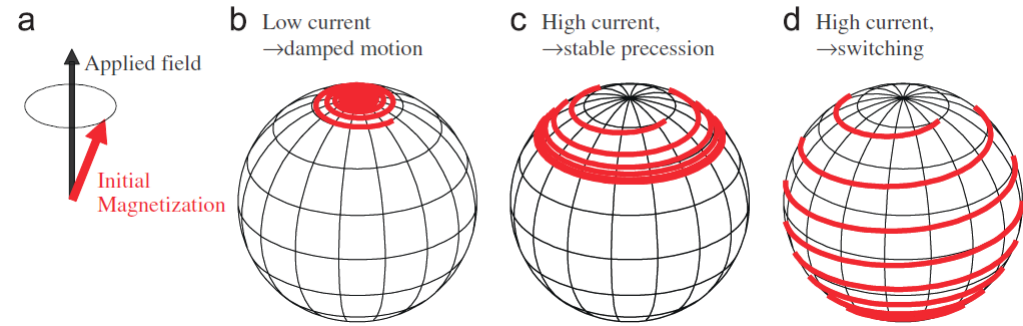
J. C. Slonczewski. Journal of Magnetism and Magnetic Materials 159(1996)L1-L7



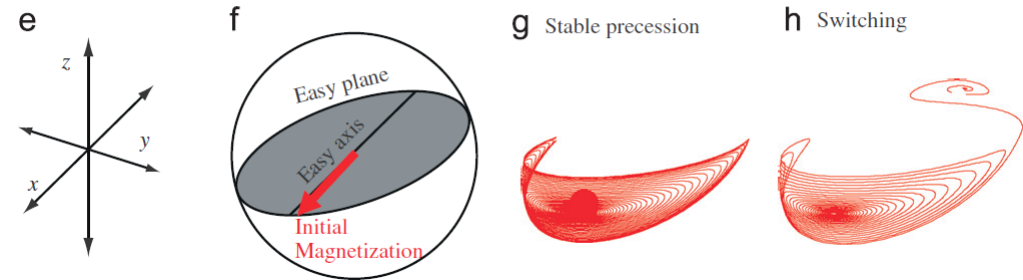
# Dynamics of the magnetization on the free layer



Moment in an applied field along z with no anisotropy



Thin-film sample with biaxial anisotropy, easy axis in-plane along x, hard direction along z



M. Bauer et al., *Phy. Rev.* B**61**, 3410-3416(2000)

D.C. Ralph, M.D. Stiles, *J. Magn. Magn. Materials* 320, 1190 (2008)

# Shreshold current (derived from LLG equation)

$$I_c = \frac{2e}{\hbar} \frac{\alpha}{\eta(0)} V \mu_0 M_s \left( \underset{\text{external}}{H} + \underset{\text{anisotropy}}{H_k} + \underset{\text{diamagnetism}}{\frac{M_s}{2}} \right) \propto \frac{\alpha}{\eta(0)} M_s$$

$V$  Free layer volume

$\alpha$  about 0.01

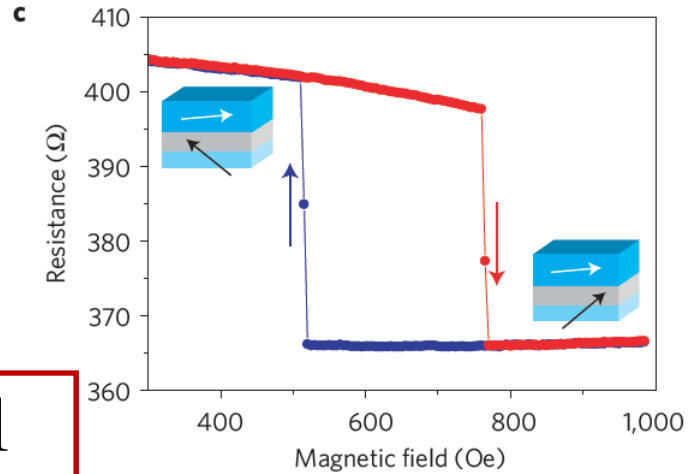
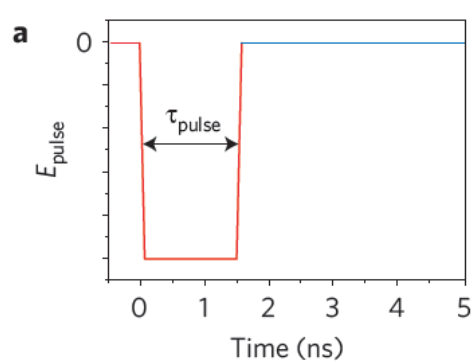
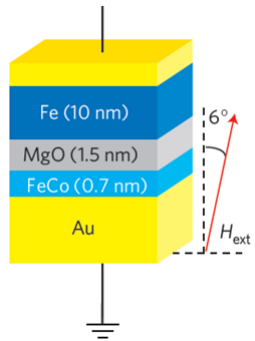
$$\eta(\theta) = \frac{q}{A + B \cos \theta} \quad \text{Anisotropy function}$$

$\theta$  : the angle between the directions of spin current  
and fixed magnetization  $M_s$

# Proto-type of the magnetic device for a **voltage driven MRAM**

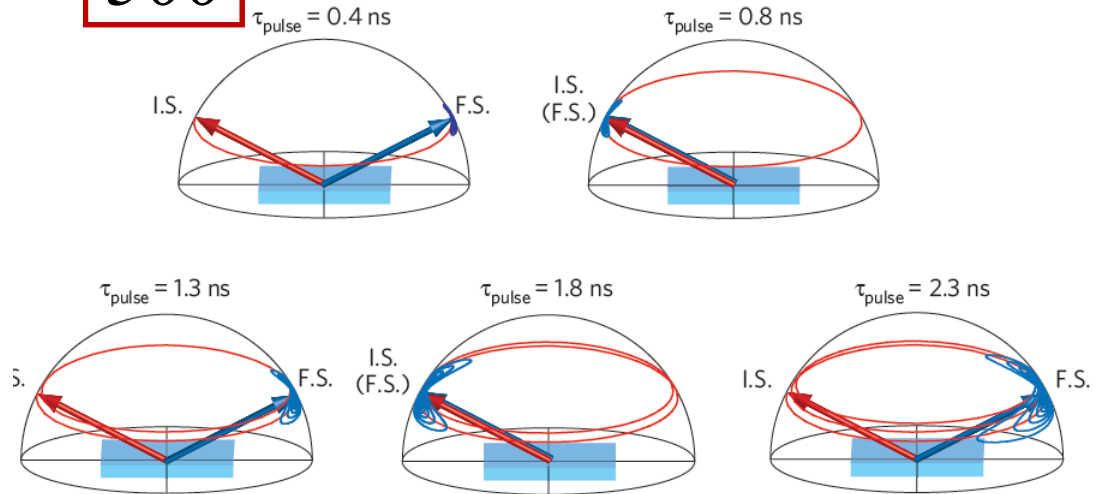
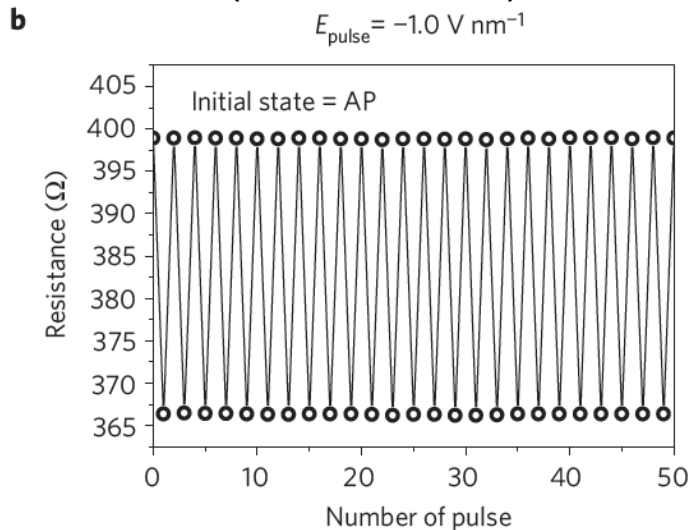
## Magnetization switching using voltage pulse

Y. Shiota, T. Nozaki, F. Bonell, S. Murakami, T. Shinio and Y. Suzuki. Nat. Mat.. **11**. 39(2012)



Consumption energy ratio  
with respect to a typical device  
of spin-transfer-torque driven  
MRAM(similar scale)

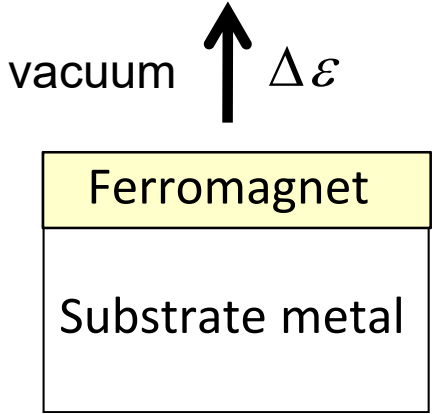
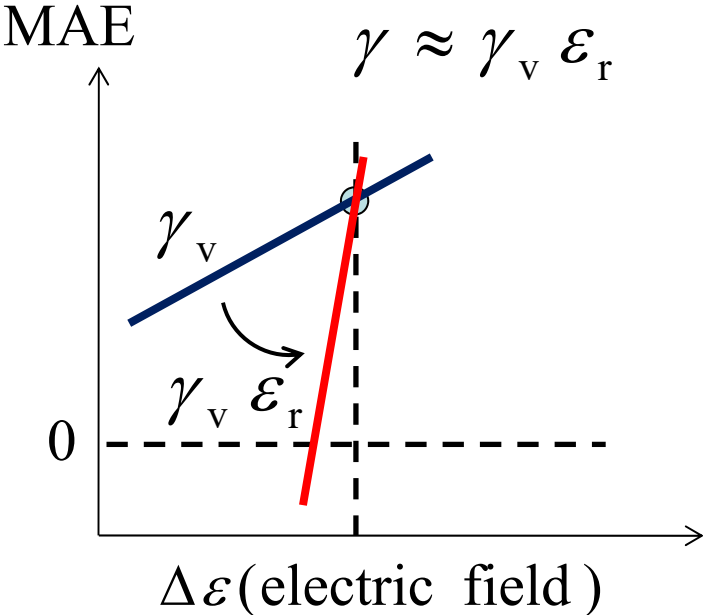
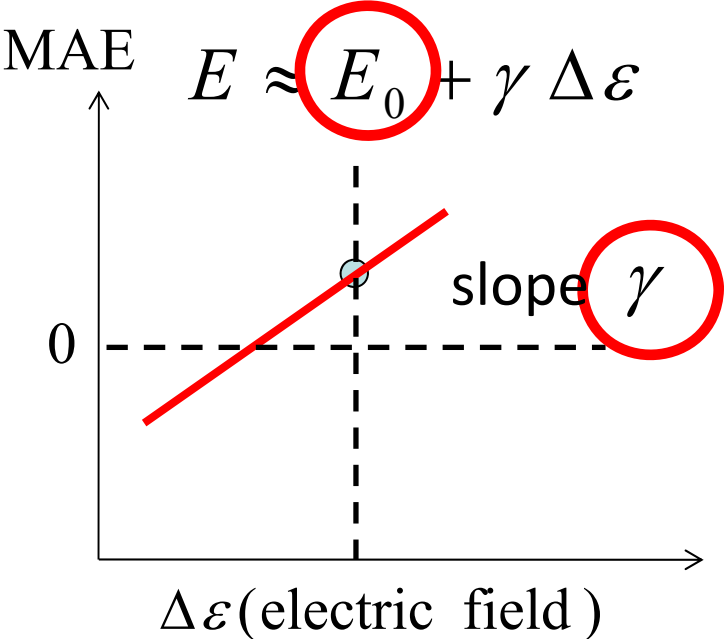
$$\frac{1}{500}$$



Macro-spin model simulation

# (5-11) Design on magnetic anisotropy in magnetic materials

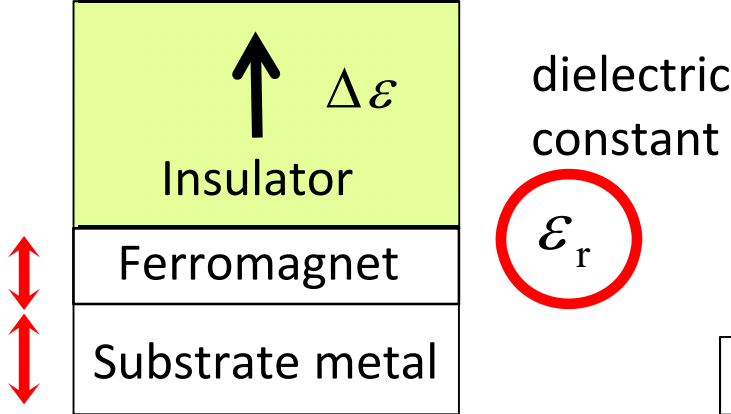
➤ Parameters for controlling MAE and its EF effect



thickness

vicinity effect

Detailed description: Two vertical red double-headed arrows are shown. The top arrow is labeled 'thickness' and the bottom arrow is labeled 'vicinity effect'.



# Elements for controlling the MAE and EF variation in thin magnetic layers

## Magnetic materials and interfaces.

Fe Fe/Pt Fe/Pd Fe/Au MgO/Fe Fe/Ni

MgO/FeCo MgO/Co<sub>2</sub>FeAl (Huesler alloy)

Mn<sub>3</sub>Ga Mn<sub>3</sub>Ge

L1-0 MnGa: K. Z. Suzuki et al.,  
Scientific Reports, 6, 30249 (2016).

## Number of magnetic layers, layer-stacking alignment, etc.

$(\text{Fe}_n/\text{Ni}_m)_\ell$

Ref.) K. Hotta et al., Phys. Rev. Lett., **110**, 267206 (2013).

## Insulating materials: larger dielectric constant : $\epsilon_r$

insulator/Fe

$\epsilon_r(\text{MgO}) = 9.8 \approx 10$

## Magnetic interaction with neighboring layers.

Exchange bias between ferro- and antiferr-magnets.

# Threshold to magnetic anisotropy transition

Films	$E_b$ (mJ/m <sup>2</sup> )	$E_s$ (mJ/m <sup>2</sup> )	$E_b + E_s$ (mJ/m <sup>2</sup> )	MAE slope $\gamma$ (fJ/Vm)	spin rotation EF (V/nm)
MgO/Fe/Pd(001)	0.36	-0.34	0.02	130	-0.2
MgO/Fe/Pt(001)	-1.18	-0.32	-1.50	615	2.4
MgO/Fe/Au(001)	0.96	-0.25	0.71	18	-40.2
MgO/Fe(2ML)/Au(001)	2.11	-0.57	1.54	-114	13.5
MgO/Pd/Fe/Au(001)	-0.68	-0.28	-0.96	-388	-2.5
MgO/Pt/Fe/Au(001)	1.52	-0.28	1.24	846	-1.5
MgO/Au/Fe/Au(001)	2.06	-0.26	1.80	-196	9.2
MgO/Pt/Fe(3ML)/Au(001)	0.69	-1.17	-0.48	633	0.8

$$\text{MAE} (\varepsilon) \approx E_b + E_s + \gamma \Delta \varepsilon$$

$$\varepsilon_c = -(E_b + E_s) / \gamma$$

# Summary

- (5-1) Electronics structure: DOS**
- (5-2) Magnetic moment: spin, orbital, localized and itinerant, spin-texture**
- (5-3) Zeeman energy**
- (5-4) Distance and interaction between magnetic carriers, magnetic and crystal structures**
- (5-5) Spin-orbit interaction, electric field effects**
- (5-6) Spin transfer Torque**
- (5-7) Magnetic anisotropy energy: electron orbital, magnet shape**
- (5-8) Magnetic anisotropy: in-plane, perpendicular**
- (5-9) Voltage-induced spin torque**
- (5-10) Landau–Lifshitz–Gilbert equation**
- (5-11) Design on magnetic anisotropy in magnetic materials**

(Appendix 1)  
Imposing the electric field  
perpendicular to  
surface/interface



A1-1. Imposing electric field

ESM (effective screening medium) method  
(M.Otani and O.Sugino, Phys.Rev.B73,115407,2006)

**Energy functional**

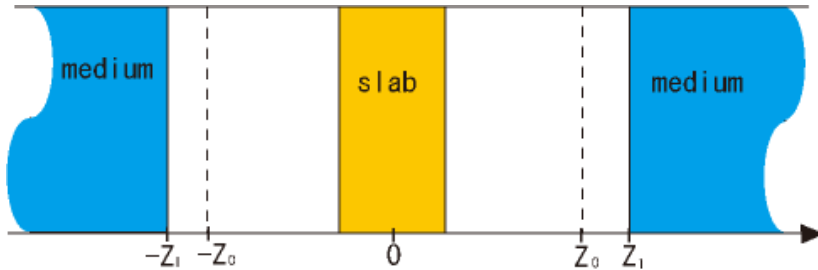
$$E[n_e, V] = K[n_e] + E_{xc}[n_e] - \int d\vec{r} \frac{\epsilon(\vec{r})}{8\pi} |\nabla V(\vec{r})|^2 + \int d\vec{r} [n_e(\vec{r}) + n_I(\vec{r})] V(\vec{r})$$

variation with the to static potential

variation with the orbital

**Poisson equation**

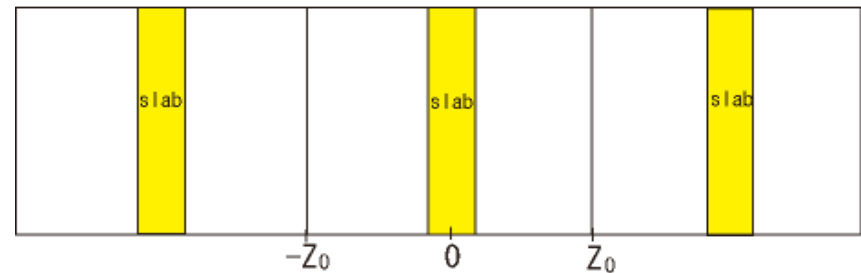
$$\begin{aligned} \nabla \cdot [\epsilon(\vec{r}) \nabla] V(\vec{r}) &= -4\pi n_{tot}(\vec{r}) \\ \nabla \cdot [\epsilon(\vec{r}) \nabla] G(\vec{r}) &= -4\pi \delta(\vec{r} - \vec{r}') \end{aligned}$$



$$V(r) = \int d\vec{r}' G(\vec{r}, \vec{r}') n_{tot}(\vec{r}')$$

**Kohn-Sham equation**

$$\left[ -\frac{1}{2} \nabla^2 + V(\vec{r}) + \hat{V}_{NL} + V_{xc}(\vec{r}) \right] \phi_\alpha(\vec{r}) = \epsilon_\alpha \phi_\alpha(\vec{r})$$



periodic boundary condition

Total energy representation by the Green's function

$$\begin{aligned} E[n_e] &= K[n_e] + E_{xc}[n_e] + \frac{1}{2} \iint d\vec{r} d\vec{r}' n_e(\vec{r}) G(\vec{r}, \vec{r}') n_e(\vec{r}') \\ &+ \iint d\vec{r} d\vec{r}' n_e(\vec{r}) G(\vec{r}, \vec{r}') n_I(\vec{r}') + \frac{1}{2} \iint n_I(\vec{r}) G(\vec{r}, \vec{r}') n_I(\vec{r}') \end{aligned}$$

## A1-2.

Usual first-principles approach  
Electrostatic potential (solution of  
Poisson's equation) ;

$$V_H(\vec{r}) = \int G(\vec{r}, \vec{r}') n(\vec{r}') d\vec{r}'$$

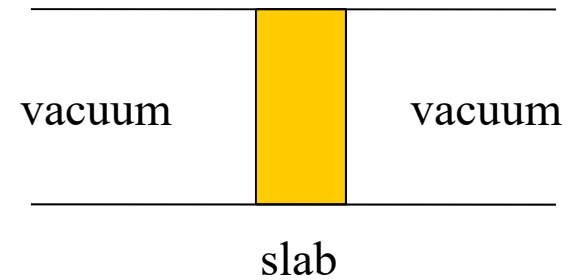
$$G(\vec{r}, \vec{r}') = \frac{1}{|\vec{r} - \vec{r}'|}$$

In practical, the above is calculated with  
the Fourier transformation

$$V_H(\vec{r}) = \sum_{\vec{g}(\neq 0)} \frac{4\pi}{\vec{g}^2} n(\vec{G}) e^{i\vec{g}\cdot\vec{r}}$$

**This procedure can not be applied for  
the system on which the electric field is  
imposed because of the breaking for the  
periodic boundary condition.**

Imposing the electric field  
on a slab

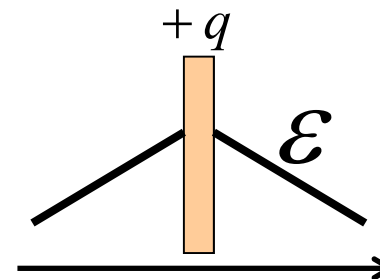


$$G(\vec{g}_{\parallel}, z, z') = \frac{4\pi}{2g_{\parallel}} e^{-g_{\parallel}|z-z'|}$$

$$V_H(\vec{g}_{\parallel}, z) = \int_{z_1}^{z_2} dz' G(\vec{g}_{\parallel}, z, z') \tilde{n}(\vec{g}_{\parallel}, z')$$

For small  $g_{\parallel}$

$$G(\vec{g}_{\parallel}, z, z') = \frac{4\pi}{2g_{\parallel}} - 2\pi |z - z'|$$



Green's function  
for a charged sheet

# A1-3. ESM method (when the electrode is placed at one side of the cell.)

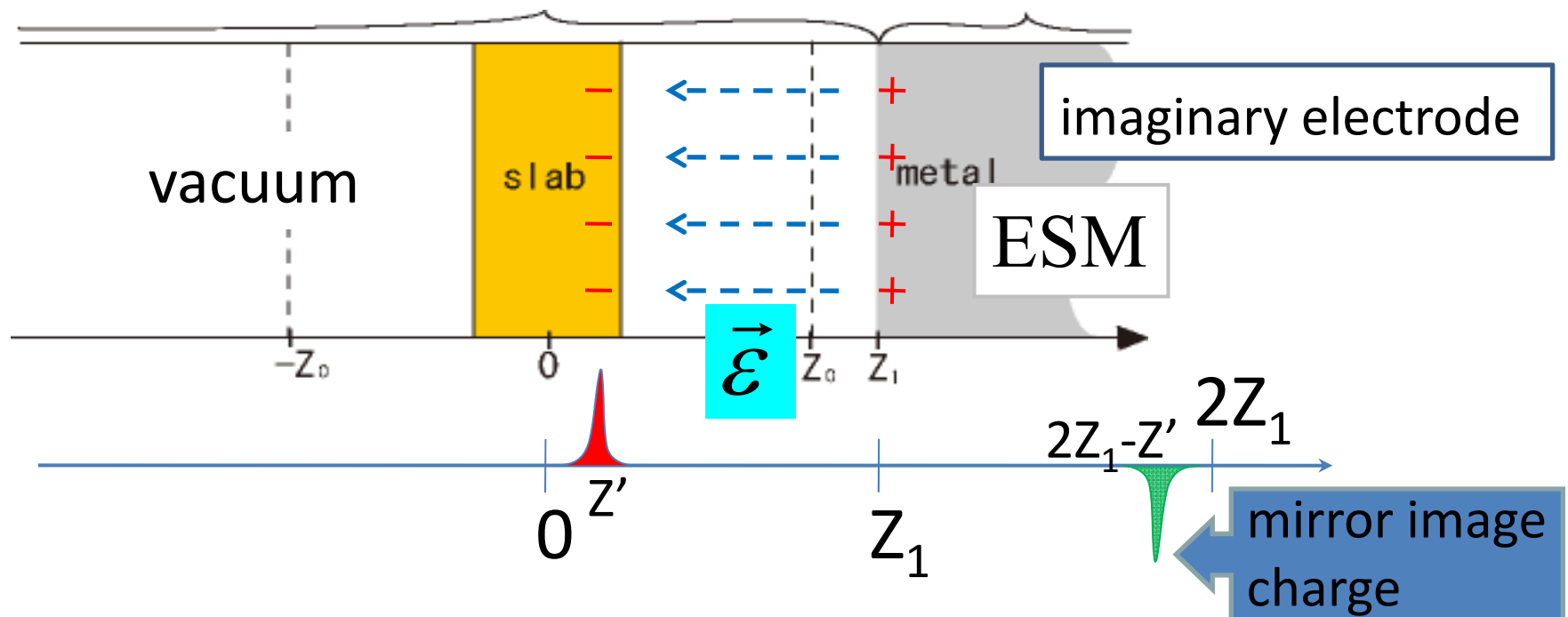
(M.Otani and O.Sugino, Phys.Rev.B73,115407,2006)

electrostatic potential  $V(r) = \int d\vec{r}' G(\vec{r}, \vec{r}') n_{tot}(\vec{r}')$

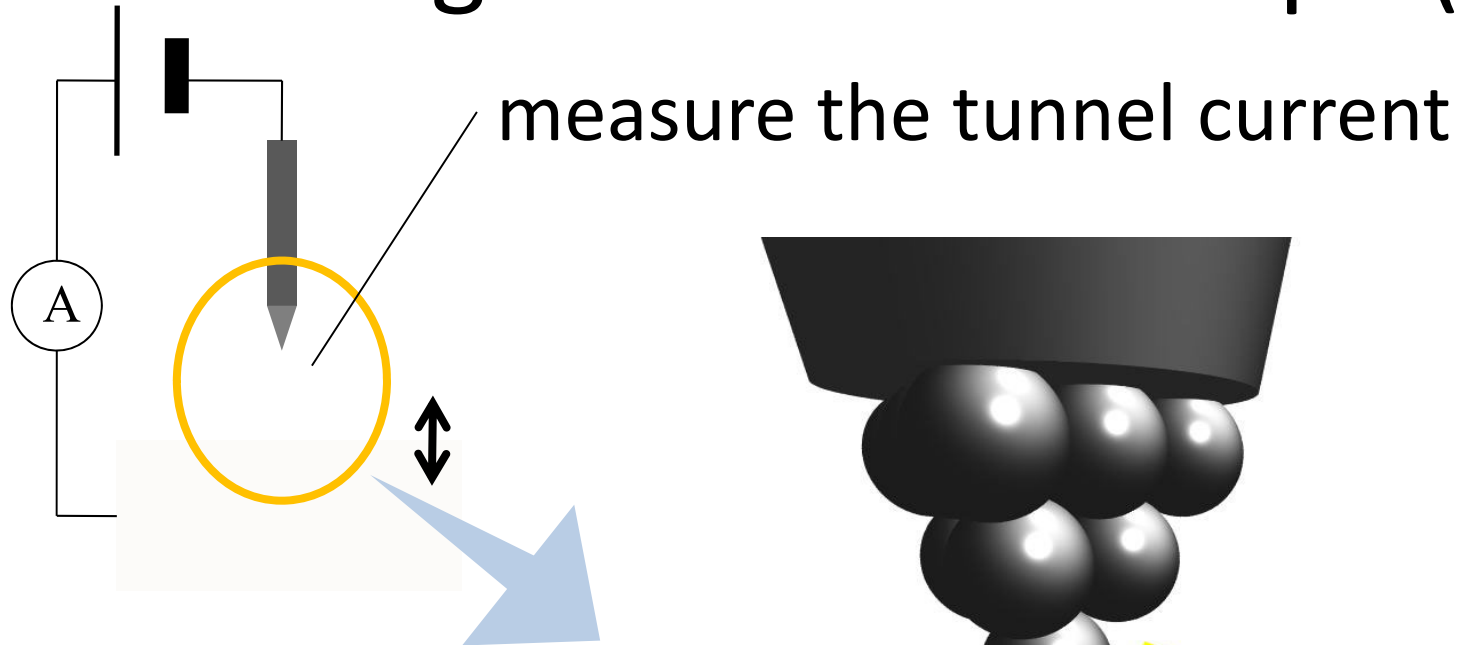
boundary condition  $\begin{cases} V_H(\mathbf{g}_{\parallel}, z)|_{z=z_1} = 0 \\ \frac{\partial}{\partial z} V_H(\mathbf{g}_{\parallel}, z)|_{z=-\infty} = 0 \end{cases} \quad \epsilon(z) = \begin{cases} 1 & \text{if } z \leq z_1 \\ \infty & \text{if } z \geq z_1 \end{cases}$

$$G(\mathbf{g}_{\parallel}, z, z') = \frac{4\pi}{2g_{\parallel}} e^{-g_{\parallel}|z-z'|} - \frac{4\pi}{2g_{\parallel}} e^{-g_{\parallel}(2z_1-z-z')}$$

the contribution from mirror image charge

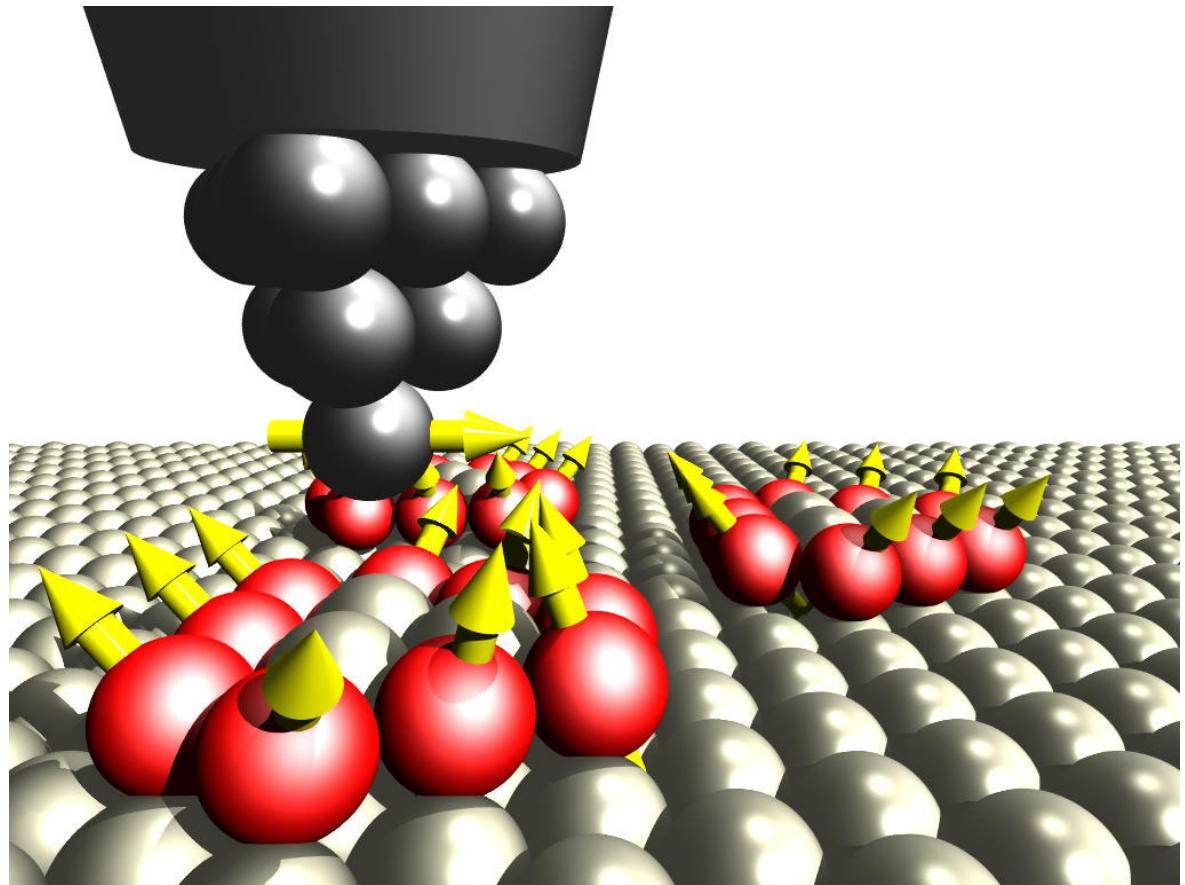


# A1-4. Electric field induced in scanning tunneling microscope (STM)



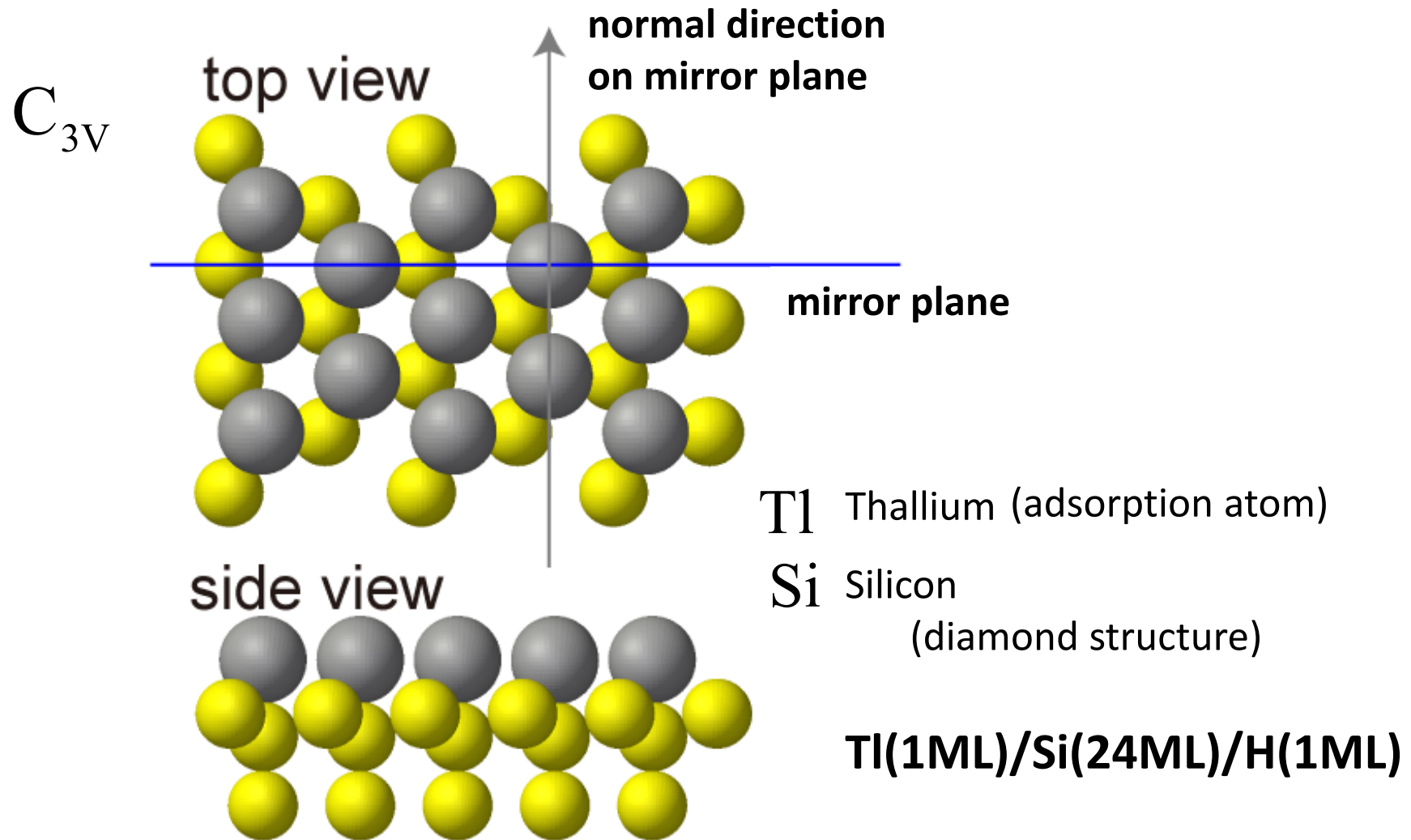
Scale of electric field

$$\approx 10^8 - 10^{10} \text{ V/m}$$



(Appendix 2)  
Single spin-state valley  
in the surface states of  
Tl/Si(111) and Tl/Si(110)

# Tl/Si(111)-1 × 1 surface



Full spin-orbit interaction other than the Rashba term

$$H_{SOI} = \frac{\hbar}{4m^2c^2} \vec{\sigma} \cdot (\text{grad } V(\vec{r}) \times \vec{p}) \quad \left( \varphi_{\vec{k}} \left| H_{SOI} \right| \varphi_{\vec{k}} \right)$$

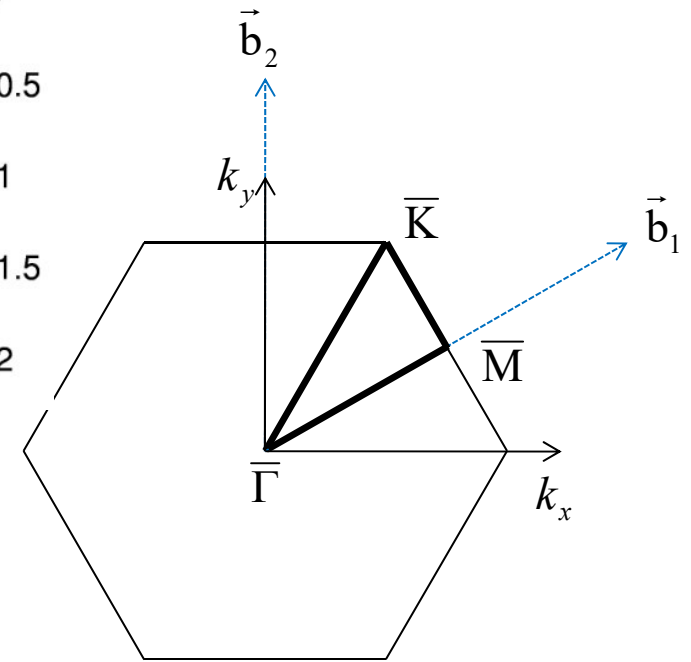
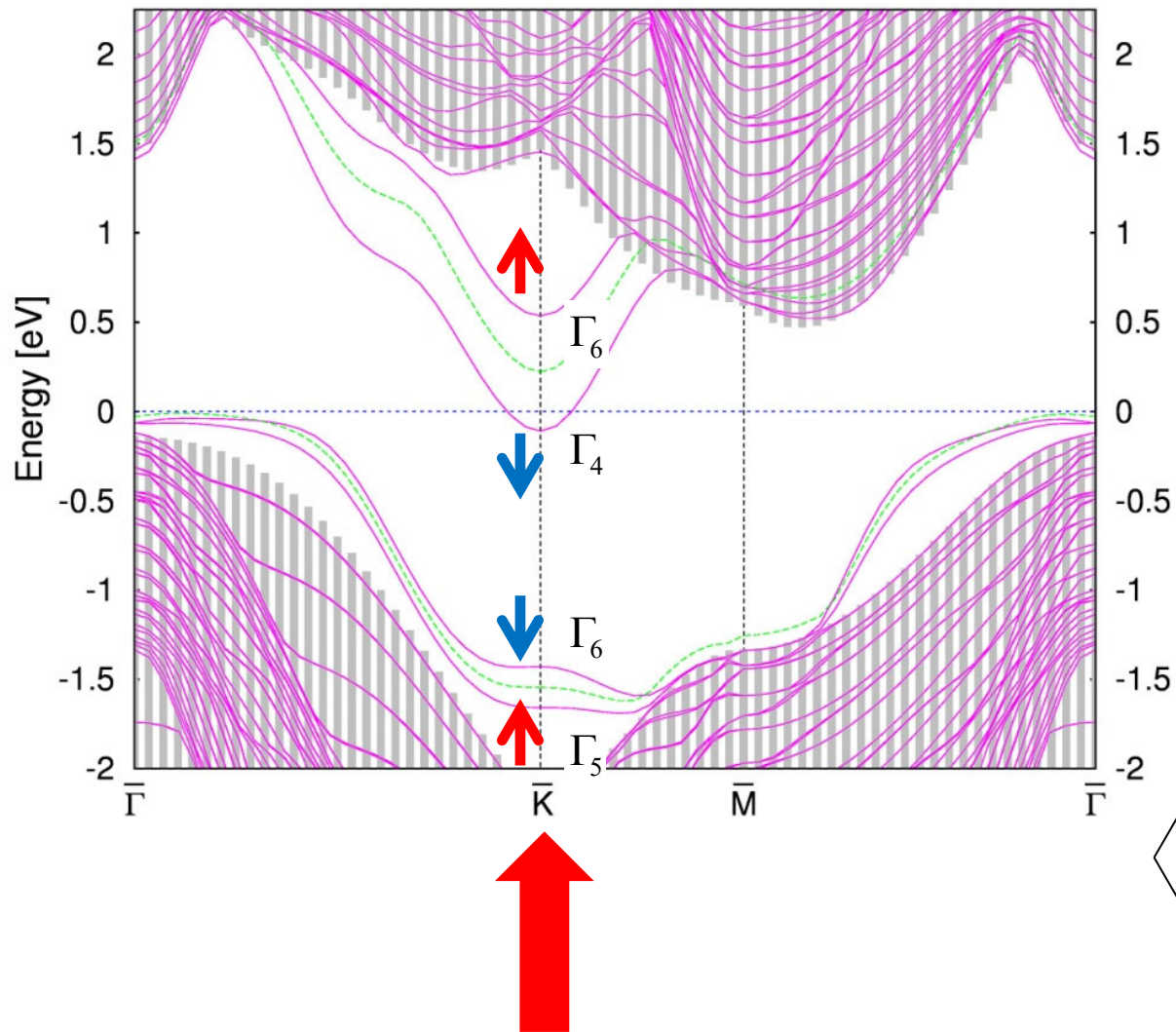
$$\left( H_{SOI} \right) = \underbrace{\vec{\sigma} \cdot \left\{ \vec{\alpha}_n(\vec{k}) \times \vec{k} \right\}}_{\text{Rashba term}} + \underbrace{\vec{\sigma} \cdot \vec{B}_n(\vec{k})}_{\text{(Zeeman term)}}$$

$$\varphi_{n\vec{k}}(\vec{r}) = \frac{1}{\sqrt{\Omega}} \exp(i\vec{k} \cdot \vec{r}) \underline{u_{n\vec{k}}(\vec{r})}$$

$$\vec{\alpha}_n = \frac{\hbar^2 N}{4m_e^2 c^2 \Omega} \int_{\text{cell}} d\vec{r} \left| u_{n\vec{k}}(\vec{r}) \right|^2 \vec{\nabla} V(\vec{r}) \quad \longrightarrow \quad \vec{\alpha}_n = \alpha \vec{e}_z$$

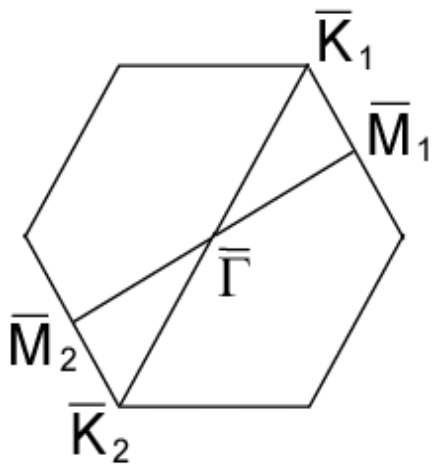
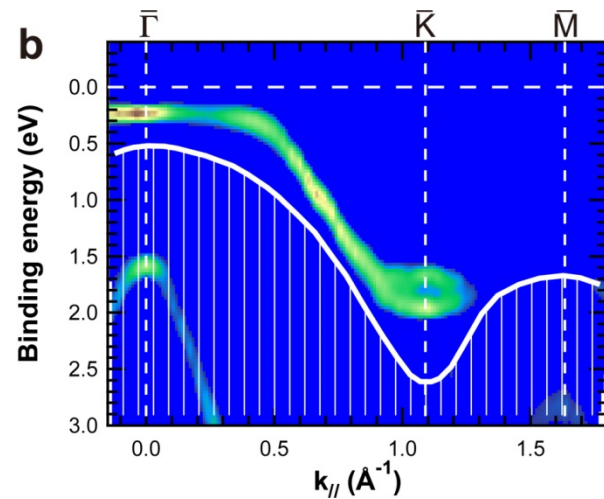
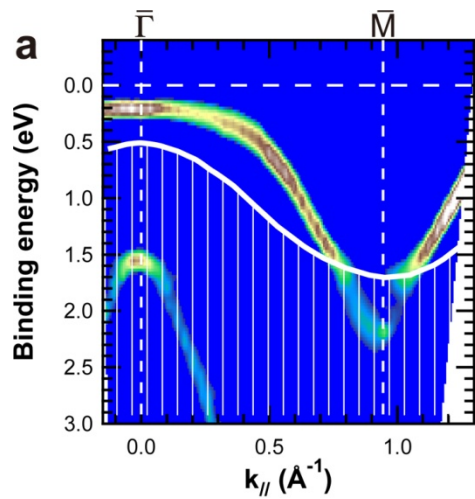
$$\vec{B}_n(\vec{k}) \approx \frac{\hbar^2 N}{4m_e^2 c^2 \Omega} \int_{\text{cell}} d\vec{r} \frac{1}{r} \frac{dV}{dr} \left\{ u_{n\vec{k}}^*(\vec{r})(\vec{\ell}) u_{n\vec{k}}(\vec{r}) \right\}$$

# Surface band structure in Tl/Si(111)

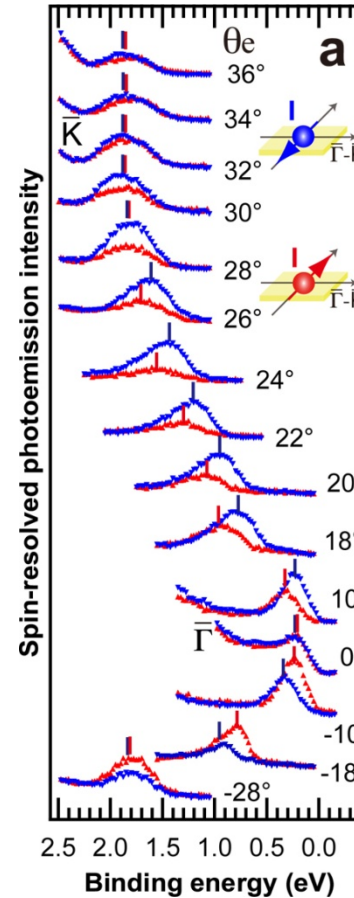




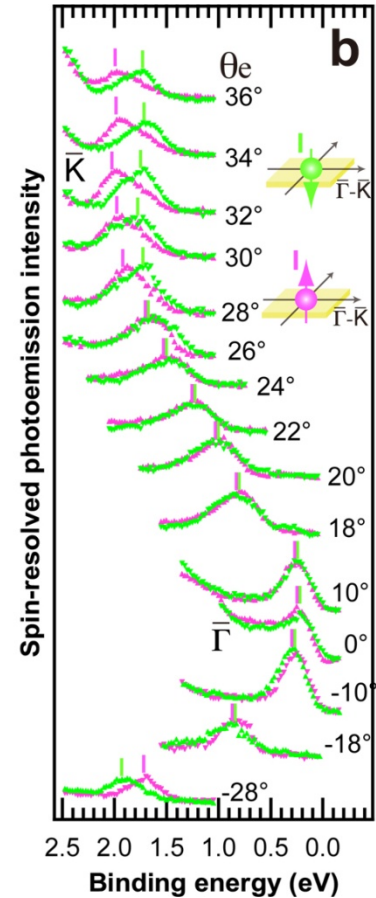
# Spin splitting of Tl/Si(111) 1 × 1 surface band



Clear splitting of electron levels in spin states at  $\bar{K}$

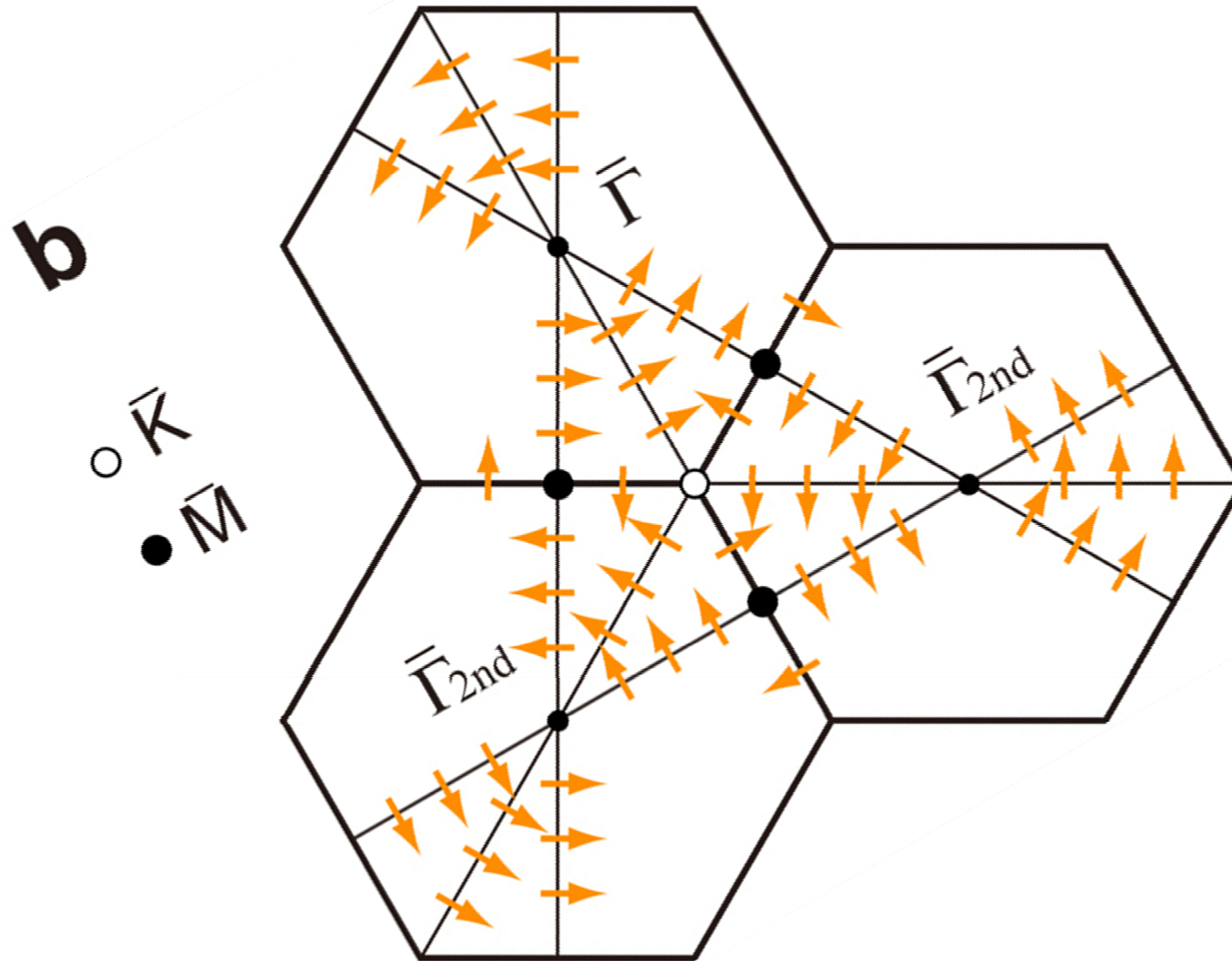


Spin splitting along Rashba direction

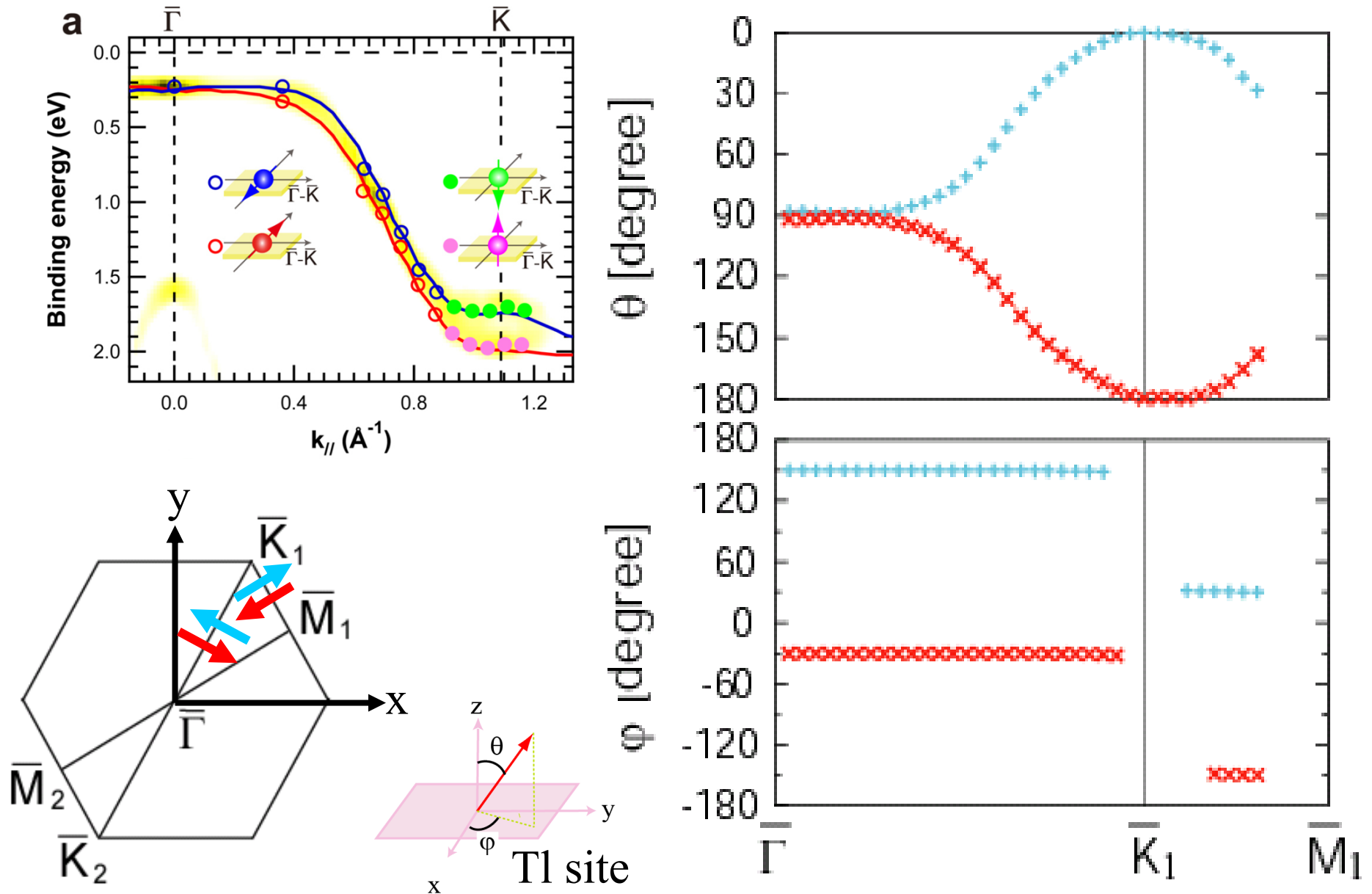


Normal to Rashba direction

Wave vector dependence of spin direction:  
Vortical spin polarization



# Wave vector dependence of electron spin direction : Tl site



# Atomic orbital components in the eigenstates at $\bar{K}$

$\bar{K}$ point	$\langle \ell_z \rangle_{\text{Tl}}$	Tl( $p_x, p_y$ ) ( $p_z$ )	Si(1)( $p_x, p_y$ ) ( $p_z$ )	Si(2)( $p_x, p_y$ ) ( $p_z$ )
$\Gamma_6$ -1.53eV	-0.047	↓ 0.060	---	↓ 0.105
$\Gamma_5$ -1.73eV	-0.068	↑ 0.090	↓ 0.002	↑ 0.024

$T_4$  site

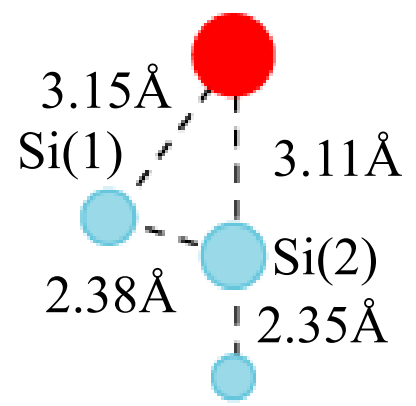
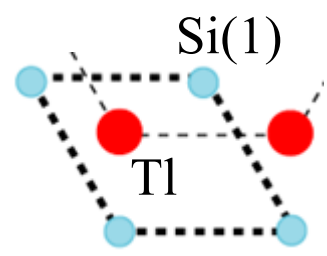
$C_3$  double group  $\langle \ell_z \rangle_{\text{Si(2)}} = -0.023$

Good quantum state of  $\ell_z$

**Tl :  $p_x - i p_y$**

basis function of atomic orbital

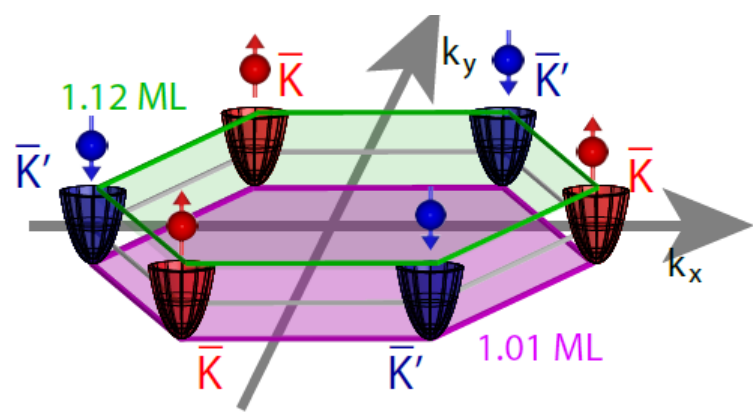
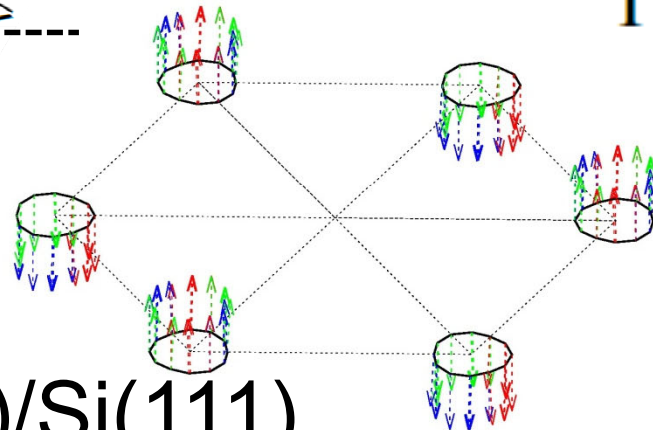
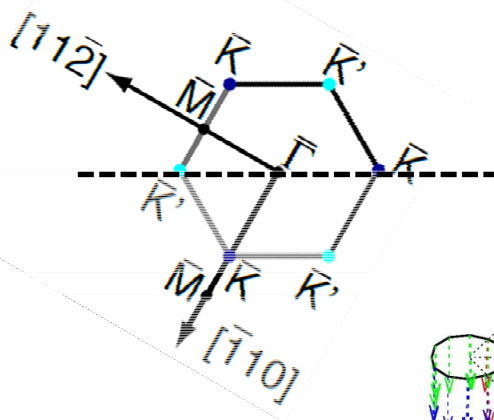
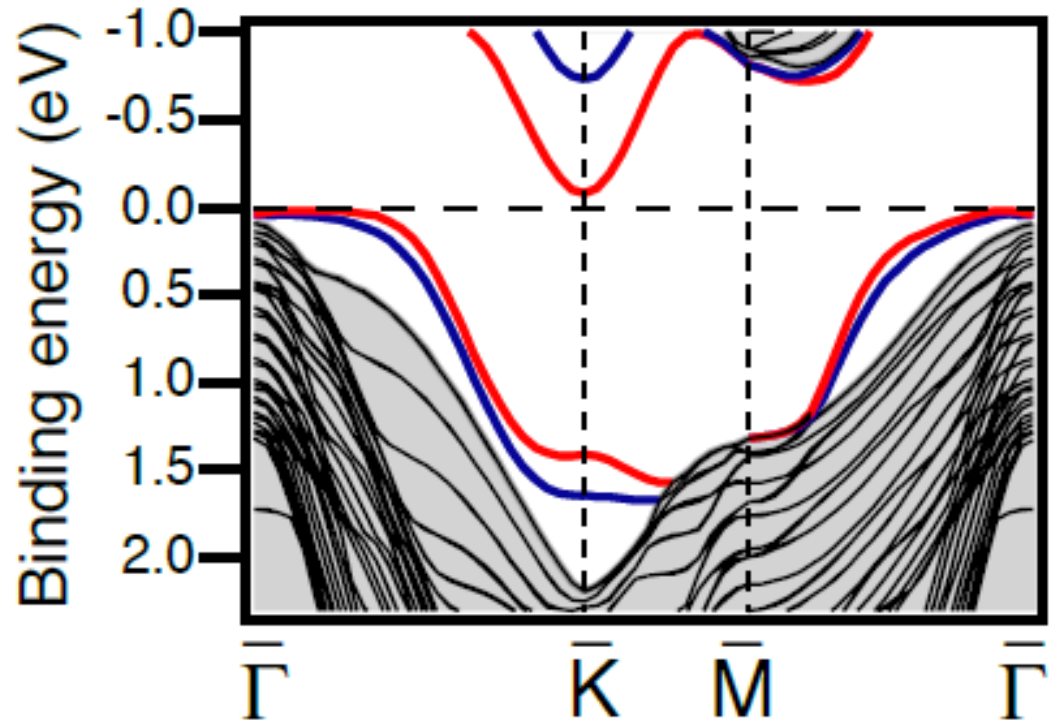
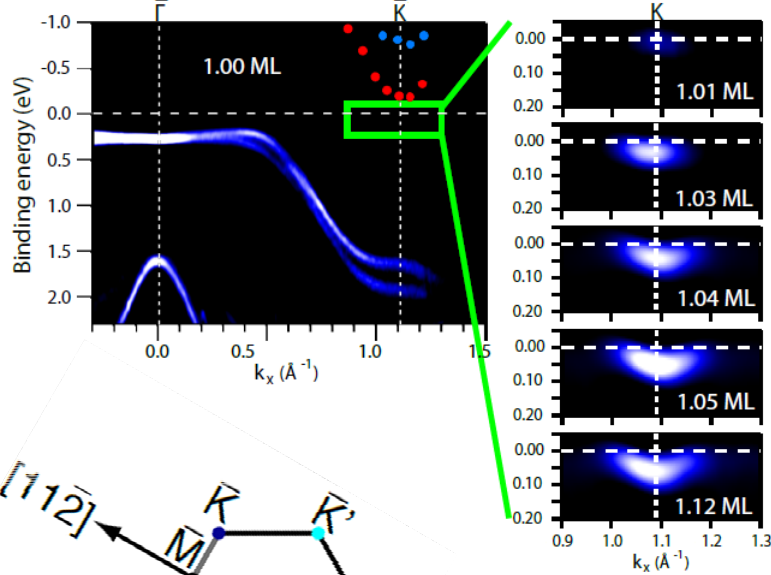
Tl site, Si(2)	Si(1) site
$\Gamma_6 : \{(x + iy)\alpha, (x - iy)\beta\}$	$\{(x - iy)\alpha, z\beta\}$
$\Gamma_5 : \{(x - iy)\alpha, z\beta\}$	$\{z\alpha, (x + iy)\beta\}$
$\Gamma_4 : \{z\alpha, (x + iy)\beta\}$	$\{(x + iy)\alpha, (x - iy)\beta\}$



**A. Araki, T. Nishijima, M. Tsujikawa and TO, J. Phys.: Conf. Ser., 200 (2010) 062001.**

# Single spin-state valley with vertical spin direction

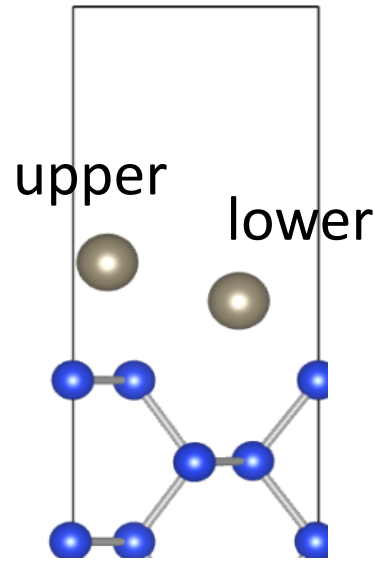
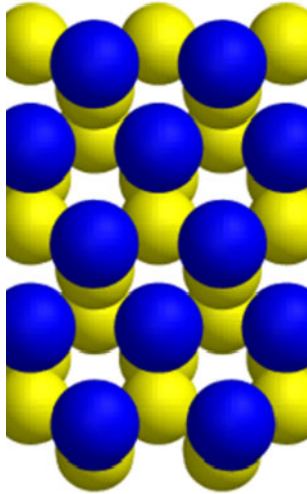
Angle resolved photoemission spectroscopy (ARPES)



Tl( $x$  ML)/Si(111)

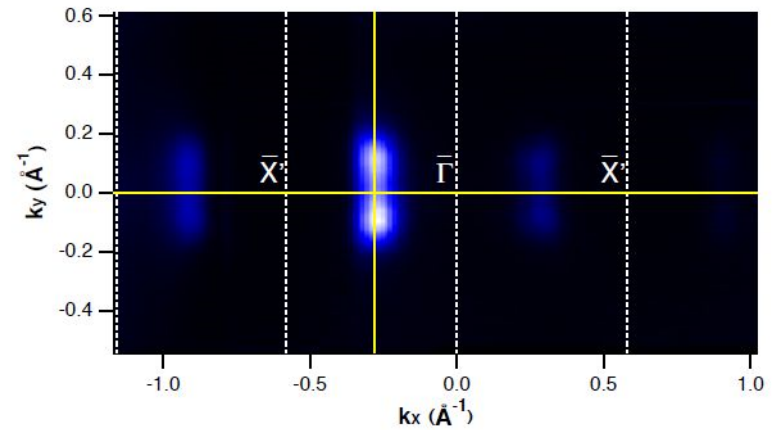
# Spin-splitting of the surface Tl/Si(110)

Structure

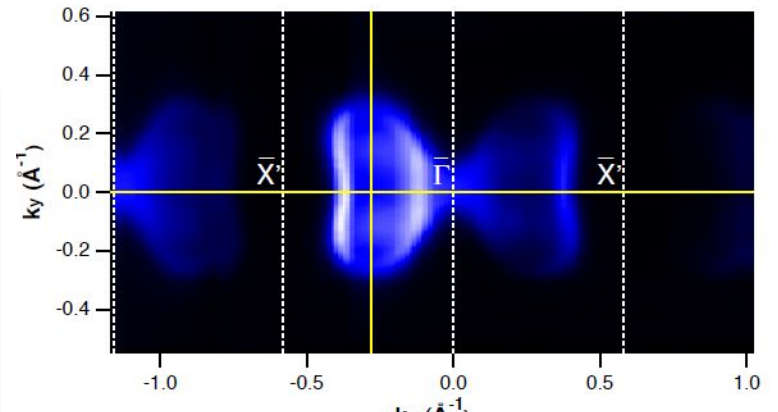


ARPES

$E_{\kappa}=35.96$  eV

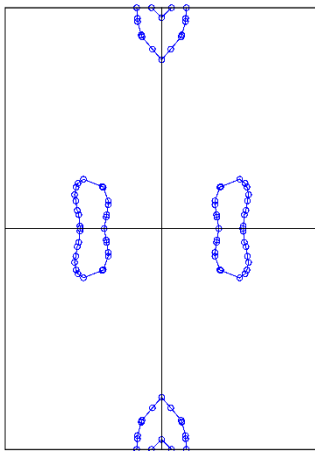


$E_{\kappa}=35.78$  eV

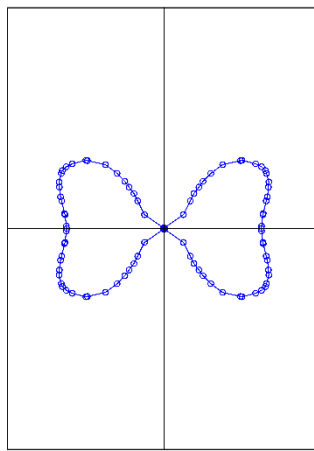


$E-E_F$

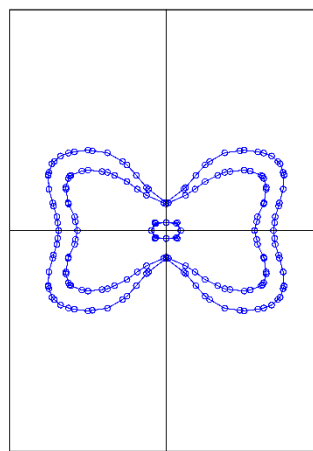
-0.04 eV



-0.13 eV



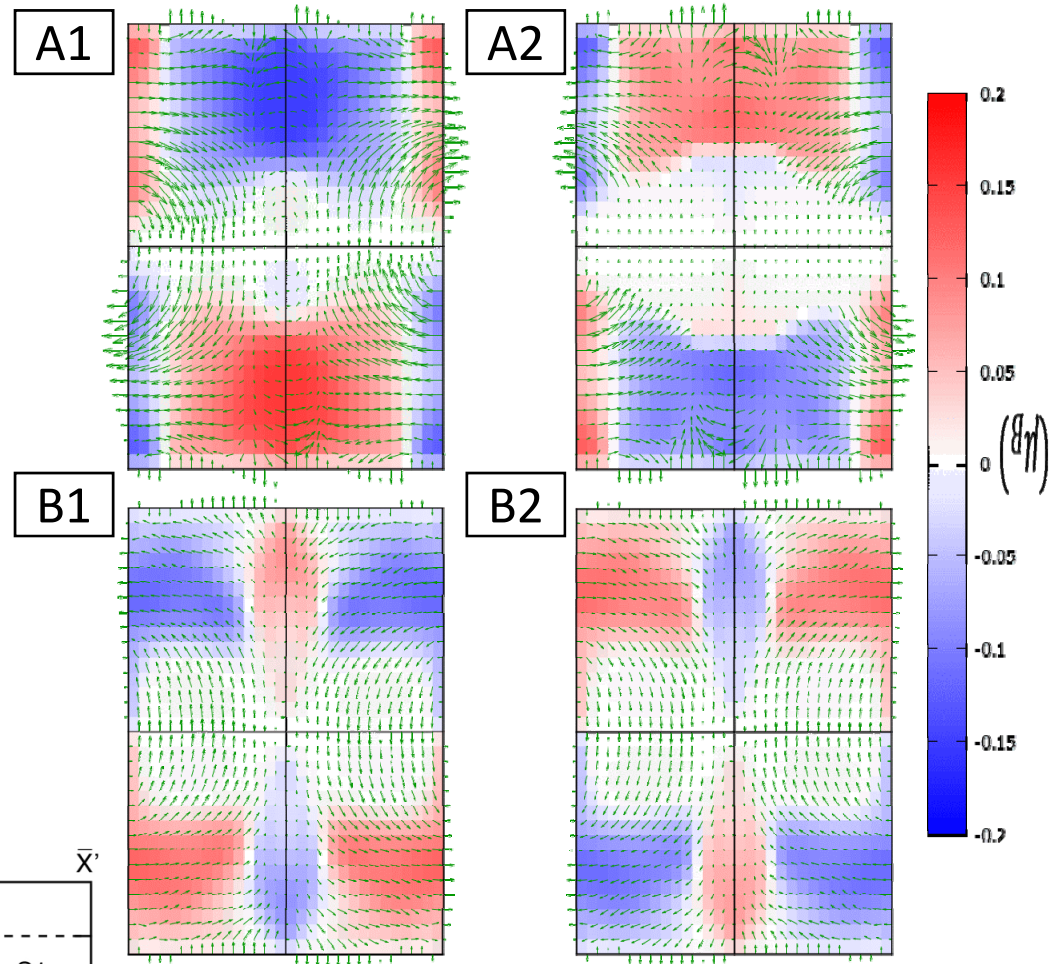
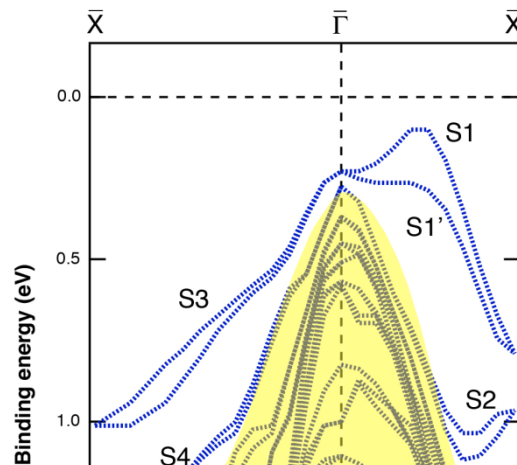
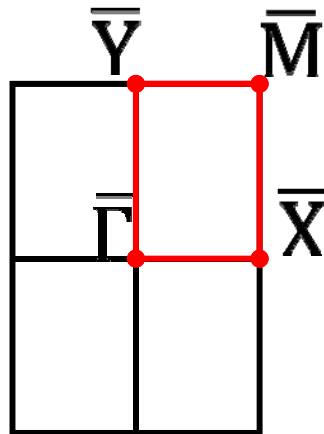
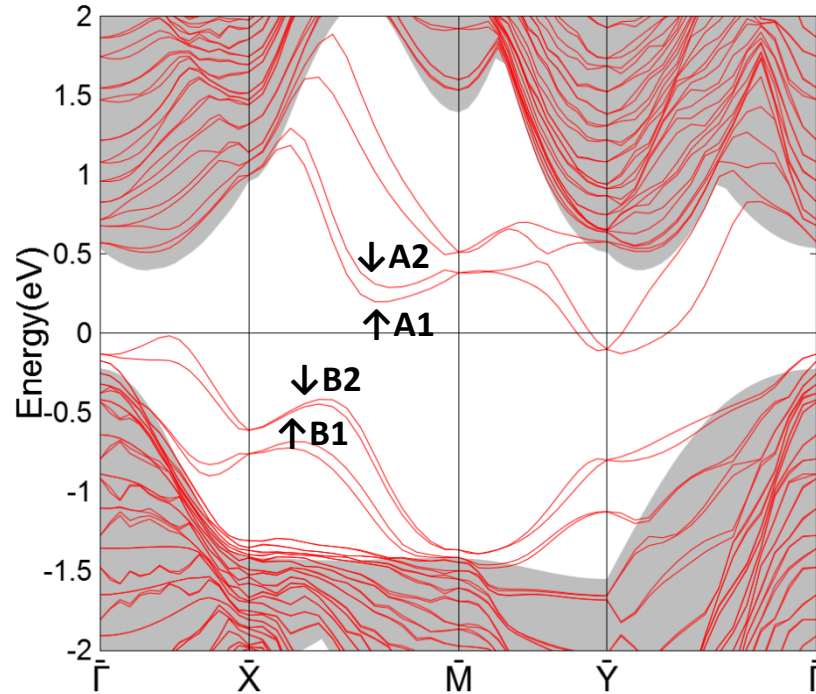
-0.22 eV



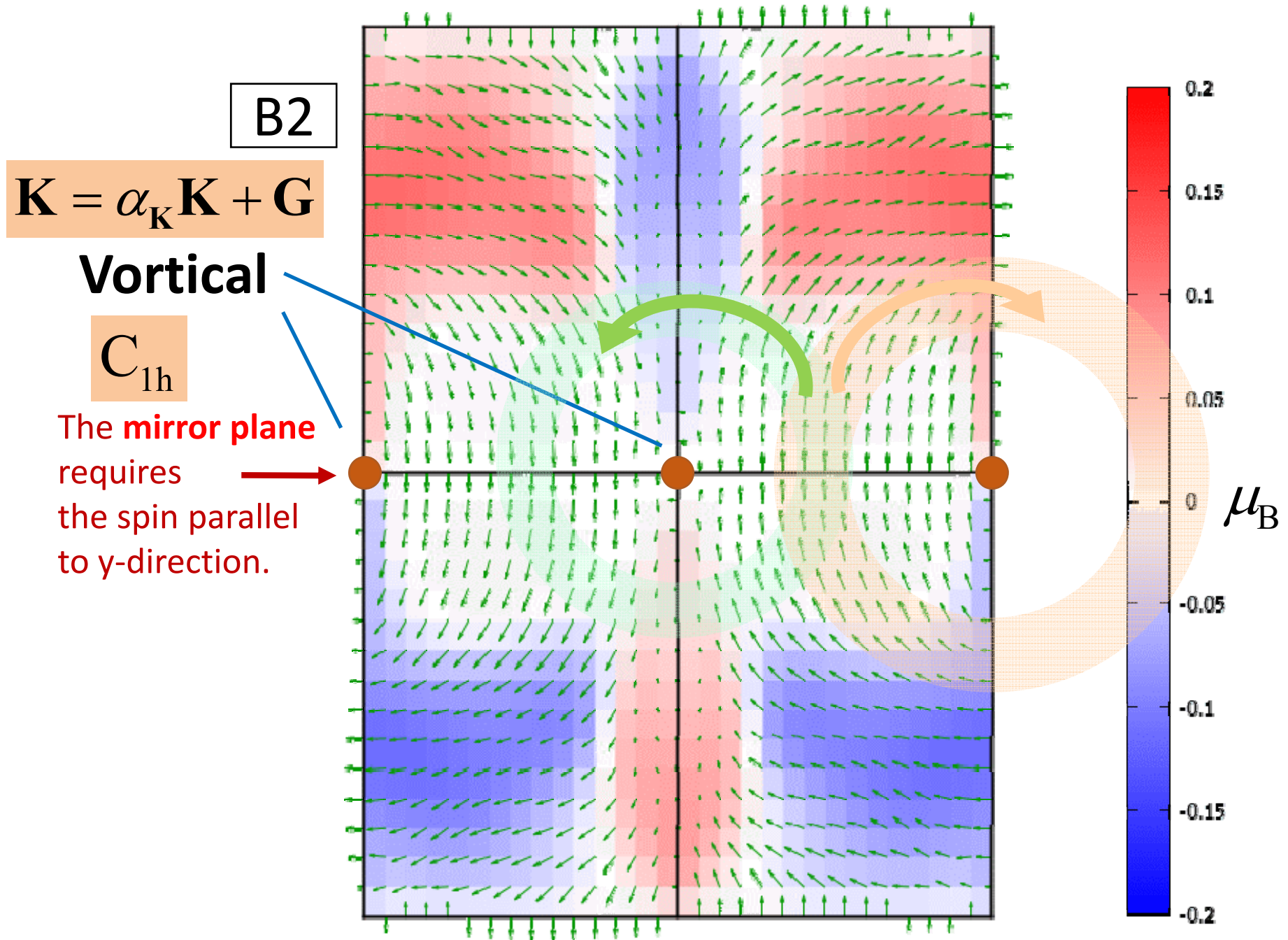
E. Annese, et al., Phys. Rev. Lett.,  
117, 16803(2016)

# Band dispersion and Spin texture

Tl/Si(110)



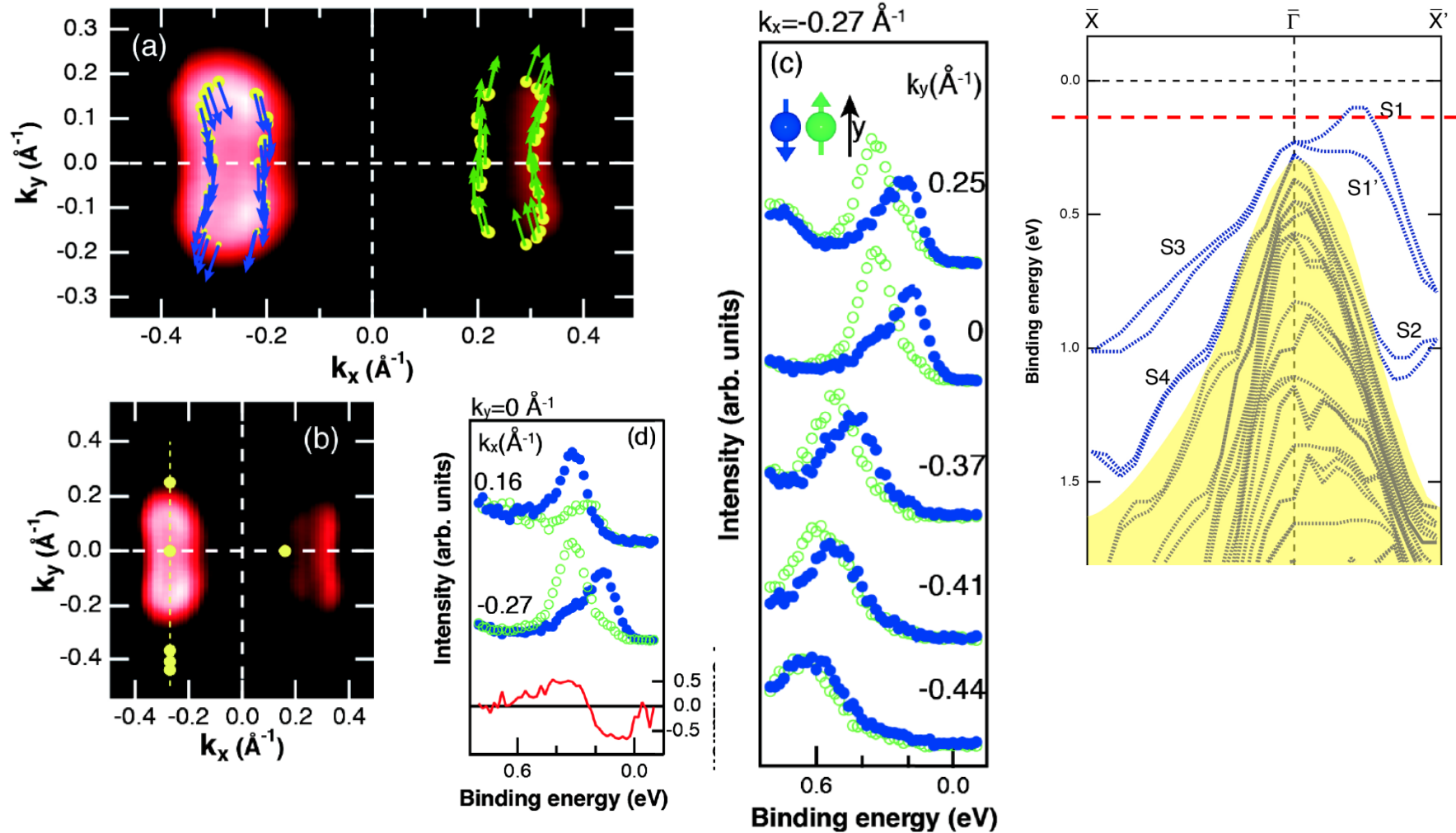
- The electron spins at



Discussion on group theory, see the paper, Nagano, Kodama, Shishido, Oguchi, JPCM,2009.



# Single spin-state valley with in-pane spin direction in Tl/Si(110)



(Appendix 3)  
Spin and orbital magnetic  
moments  
in an electronic structure  
calculation

# Spin and orbital magnetic moments

spin magnetic moment

$$m_{spin,k}^I = -\mu_B \langle m_k(\vec{r}) \rangle_I$$

orbital magnetic moment

$$m_{orb,k}^I = -\mu_B \langle \ell_k \rangle_I$$

expansion with the local basis

$$\Psi_{i\alpha}(\vec{r}) = \sum_{\ell m} Y_{\ell m}(\hat{r}_I) R_{\ell m, i\alpha}(r)$$

$$\langle \ell_k \rangle_I = \langle \ell_k \rangle_{I, PW} + \langle \ell_k \rangle_{I, VB}$$

$$\langle \ell_k \rangle_{I, PW} = \sum_i f_i \langle \Psi_i | \ell_k | \Psi_i \rangle_I$$

$$\langle \ell_k \rangle_{I, VB} = \sum_{ipq} f_i \langle \Psi_i | \beta_q^I \rangle \ell_{k,pq}^I \langle \beta_p^I | \Psi_i \rangle$$

$$\ell_{k,pq}^I = \int_0^{r_c} \phi_p(r_I) \phi_q(r_I) r_I^2 dr_I \left\langle Y_{j_q \mu_q}^{\text{sgn}(\kappa_q)} \middle| \ell_k \middle| Y_{j_p \mu_p}^{\text{sgn}(\kappa_p)} \right\rangle$$

# Atomic magnetic moments in CoPt and FePt

$r_c=2.5$ a.u.		spin ( $\mu_B$ )			orbital( $\mu_B$ )			
		USPP	AE		USPP	AE		
CoPt	[001]	Co	1.926	1.91	1.803	0.102	0.11	0.089
		Pt	0.377	0.38	0.394	0.061	0.07	0.056
CoPt	[110]	Co	1.929	1.809		0.069	0.057	
		Pt	0.377	0.398		0.078	0.073	
FePt	[001]	Fe	3.016	2.93	2.891	0.067	0.08	0.067
		Pt	0.338	0.33	0.353	0.046	0.05	0.042
FePt	[110]	Fe	3.020	2.893		0.062	0.061	
		Pt	0.340	0.355		0.059	0.055	

AE: Sakuma,JPSJ(1994), Ravindran *et al.*,PRB(2001)

(Appendix 4)  
Approximation in Dirac equation

## Dirac equation (part 2)

(eq. in stationary-state)

$$p_0 = i\hbar \frac{\partial}{\partial(ct)} \longrightarrow p_0 = \frac{\varepsilon}{c}$$

Coupled equations for the large and small components;

$$\left(\varepsilon - mc^2 + eA_0\right)\varphi_L = c \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right) \varphi_S$$

$$\left(\varepsilon + mc^2 + eA_0\right)\varphi_S = c \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right) \varphi_L$$

Eliminate the small component;

$$\left(\varepsilon - mc^2 + eA_0\right)\varphi_L = c^2 \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right)$$

$$\left(\varepsilon + mc^2 + eA_0\right)^{-1} \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right) \varphi_L$$

Commute the parts with underline:

## Dirac equation (part 3)

$$\begin{aligned} (\varepsilon - mc^2 + eA_0)\varphi_L = c^2 \underbrace{(\varepsilon + mc^2 + eA_0)^{-1}} \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right)^2 \\ + c^2 \underbrace{(\varepsilon + mc^2 + eA_0)^{-2}} (\vec{\sigma}, (-e\vec{p}A_0)) \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right) \varphi_L \end{aligned}$$

For not heavy elements, the eigenvalue at valence electrons is larger than the rest energy by a small value. Therefore, we cant take a following approximation;

$$\varepsilon' = \varepsilon - mc^2, \quad \frac{\varepsilon' + eV_0}{mc^2} \lll 1 \quad \begin{aligned} \varepsilon + mc^2 + eV_0 \\ \approx 2mc^2 \end{aligned}$$

Using this approximation, the equation for the large component;

## Formula 1

$(\vec{a}, \vec{b})$  : Scalar product

Using following general properties;

$$(\vec{\sigma}, \vec{B})(\vec{\sigma}, \vec{C}) = (\vec{B}, \vec{C}) + i(\vec{\sigma}, \vec{B} \times \vec{C})$$

$$\vec{H} = \vec{\nabla} \times \vec{A}$$

We obtain the following formula related with Zeeman term;

$$\left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right)^2 = \left( \vec{p} + \frac{e}{c} \vec{A} \right)^2 + \frac{\hbar e}{c} (\vec{\sigma}, \vec{H})$$

$$\begin{aligned} (\vec{\sigma}, (-e\vec{p}A_0)) \left( \vec{\sigma}, \vec{p} + \frac{e}{c} \vec{A} \right) &= \left( (-e\vec{p}A_0), \vec{p} + \frac{e}{c} \vec{A} \right) + i \left( \vec{\sigma}, (-e\vec{p}A_0) \times \left( \vec{p} + \frac{e}{c} \vec{A} \right) \right) \\ &= \left( (-e\vec{p}A_0), \vec{p} + \frac{e}{c} \vec{A} \right) + \left( i\vec{\sigma} \times (-e\vec{p}A_0), \vec{p} + \frac{e}{c} \vec{A} \right) \end{aligned}$$

$$c^2 (\varepsilon + mc^2 + eA_0)^{-1} = \frac{1}{2m} \left( 1 + \frac{\varepsilon' + eA_0}{2mc^2} \right)^{-1} = \frac{1}{2m} \left( 1 - \frac{\varepsilon' + eA_0}{2mc^2} + \dots \right)$$

$$c^2 (\varepsilon + mc^2 + eA_0)^{-2} = \frac{1}{4m^2 c^2} \left( 1 + \frac{\varepsilon' + eA_0}{2mc^2} \right)^{-2} = \frac{1}{4m^2 c^2} \left( 1 - \frac{\varepsilon' + eA_0}{mc^2} + \dots \right)$$



## Formula 2

$$(\varepsilon' + eA_0)\varphi_L =$$

$$\left(1 - \frac{\varepsilon' + eA_0}{2mc^2} + \dots\right) \left\{ \frac{1}{2m} \left( \vec{p} + \frac{e}{c} \vec{A} \right)^2 + \frac{\hbar e}{2mc} (\vec{\sigma}, \vec{H}) \right\} \varphi_L +$$

$$\left(1 - \frac{\varepsilon' + eA_0}{mc^2} + \dots\right) \times$$

$$\left\{ \frac{1}{4m^2c^2} \left( (-e\vec{p}A_0), \vec{p} + \frac{e}{c} \vec{A} \right) + \frac{1}{4m^2c^2} \left( i\vec{\sigma} \times (-e\vec{p}A_0), \vec{p} + \frac{e}{c} \vec{A} \right) \right\} \varphi_L$$

Taking the leading terms in the approximation;  
the approximation formula in (5-3).

(Appendix 5)  
Spin-orbit splitting in the heavy  
element

$$E_{\text{SO}}^j = \lambda \vec{\ell} \cdot \vec{s} = \frac{\lambda}{2} \{j(j+1) - s(s+1) - \ell(\ell+1)\}$$

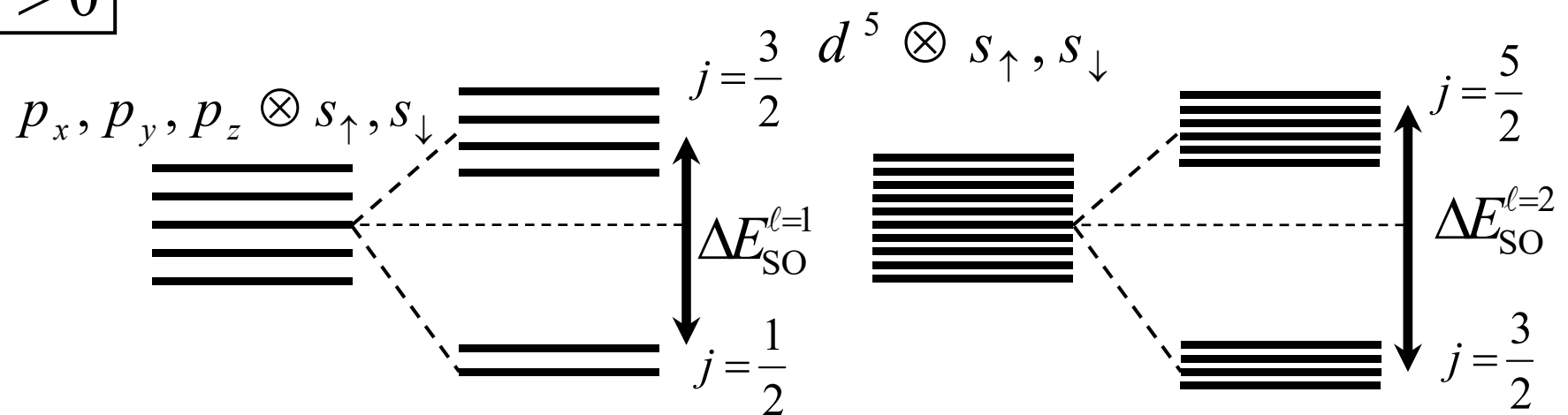
$$\left. \begin{array}{l} s, s_z = s, s-1, \dots, -s \\ \ell, m = \ell, \ell-1, \dots, -\ell \end{array} \right\} j = \ell + s, \dots, |\ell - s|$$

$$s = \frac{1}{2}, \quad s_z = \frac{1}{2}, -\frac{1}{2}$$

$$\ell = 1 \quad \longrightarrow \quad j = \frac{3}{2}, \frac{1}{2}$$

$$\ell = 2 \quad \longrightarrow \quad j = \frac{5}{2}, \frac{3}{2}$$

$$\boxed{\lambda > 0}$$



# Eigenvalues of Pb atom

(in Ry energy unit)

Ref. State:

(Kr Core)(5d)<sup>10</sup>(6s)<sup>2</sup>(6p)<sup>2</sup>

$n\ell$	$j$	all electron	pseudo ( $\Delta E$ )	
$5d$	$3/2$	-1.6734	-1.6733 (+0.0001)	$\Delta E_{so}(d)$ 0.1912 2.601 eV
$5d$	$5/2$	-1.4823	-1.4821 (+0.0002)	
$6s$	$1/2$	-0.9016	-0.9014 (+0.0002)	
$6p$	$1/2$	-0.3547	-0.3545 (+0.0002)	$\Delta E_{so}(p)$ 0.1106 1.505 eV
$6p$	$3/2$	-0.2440	-0.2439 (+0.0001)	

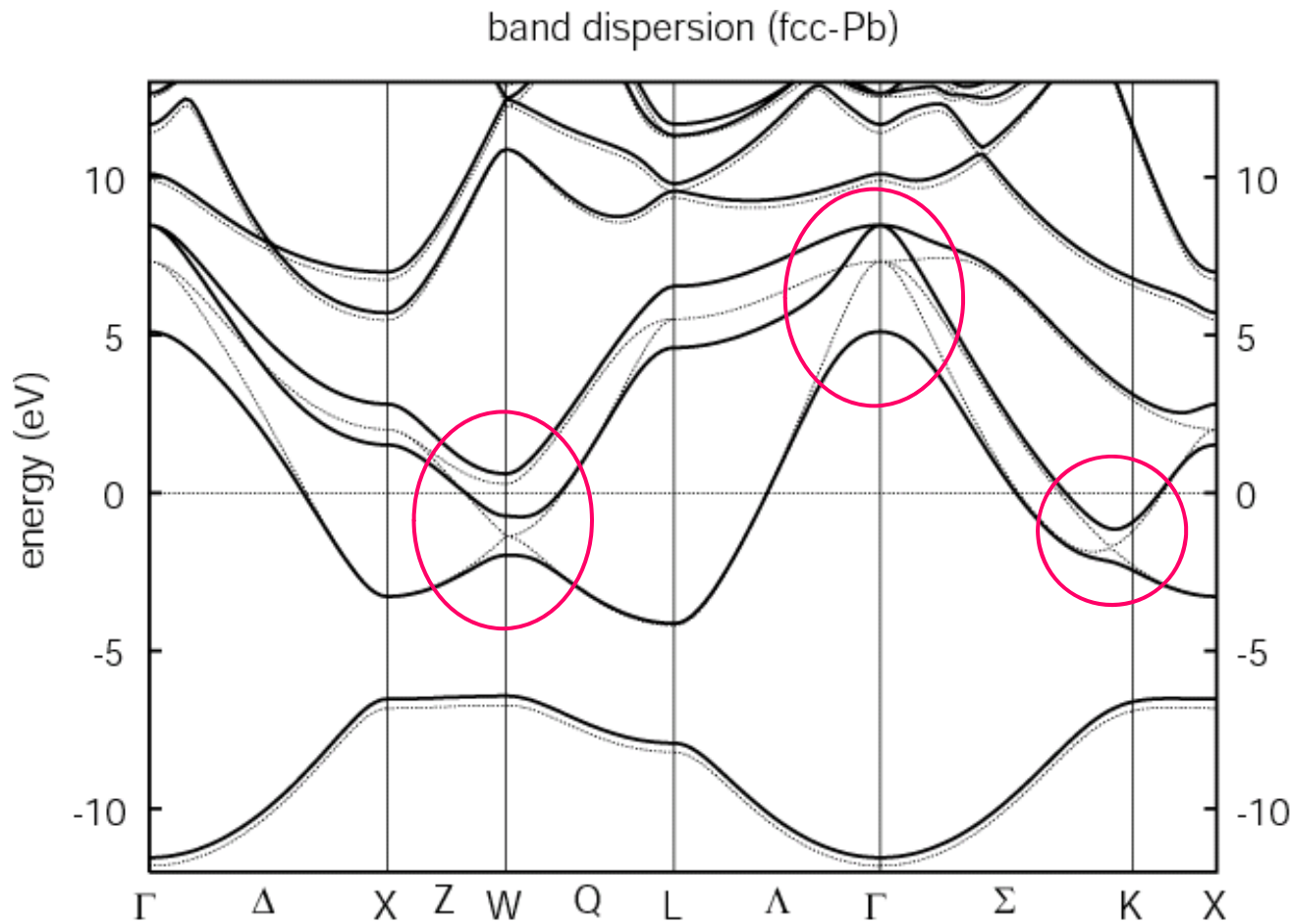
These values are in good agreement with the previous data.

$$\Delta E_{SO}^{\ell} = E_{SO}^{\ell+1/2} - E_{SO}^{\ell-1/2} = \frac{\lambda}{2}(2\ell+1) \quad \lambda = \frac{2}{2\ell+1} \Delta E_{SO}^{\ell} \quad \lambda_{\ell=2}^{Pb} = 1.04 \text{ eV}$$

$$\lambda_{\ell=1}^{Pb} = 1.00 \text{ eV}$$

# Band dispersion of fcc Pb

$a=9.35$  a.u.(exp.)



USPP (RAPW)

$\Gamma$ :	-11.55	(-11.4)
$L$ :	-7.92	(-8.2)
	-4.13	(-4.5)
$K$ :	-6.61	(-6.7)
	-2.44	(-2.8)
	-0.93	(-1.2)
$W$ :	-6.43	(-7.2)
$X$ :	-6.52	(-6.7)
	-3.28	(-3.6)

Thick lines : Fully-Relativistic  
Thin lines : Scalar-Relativistic

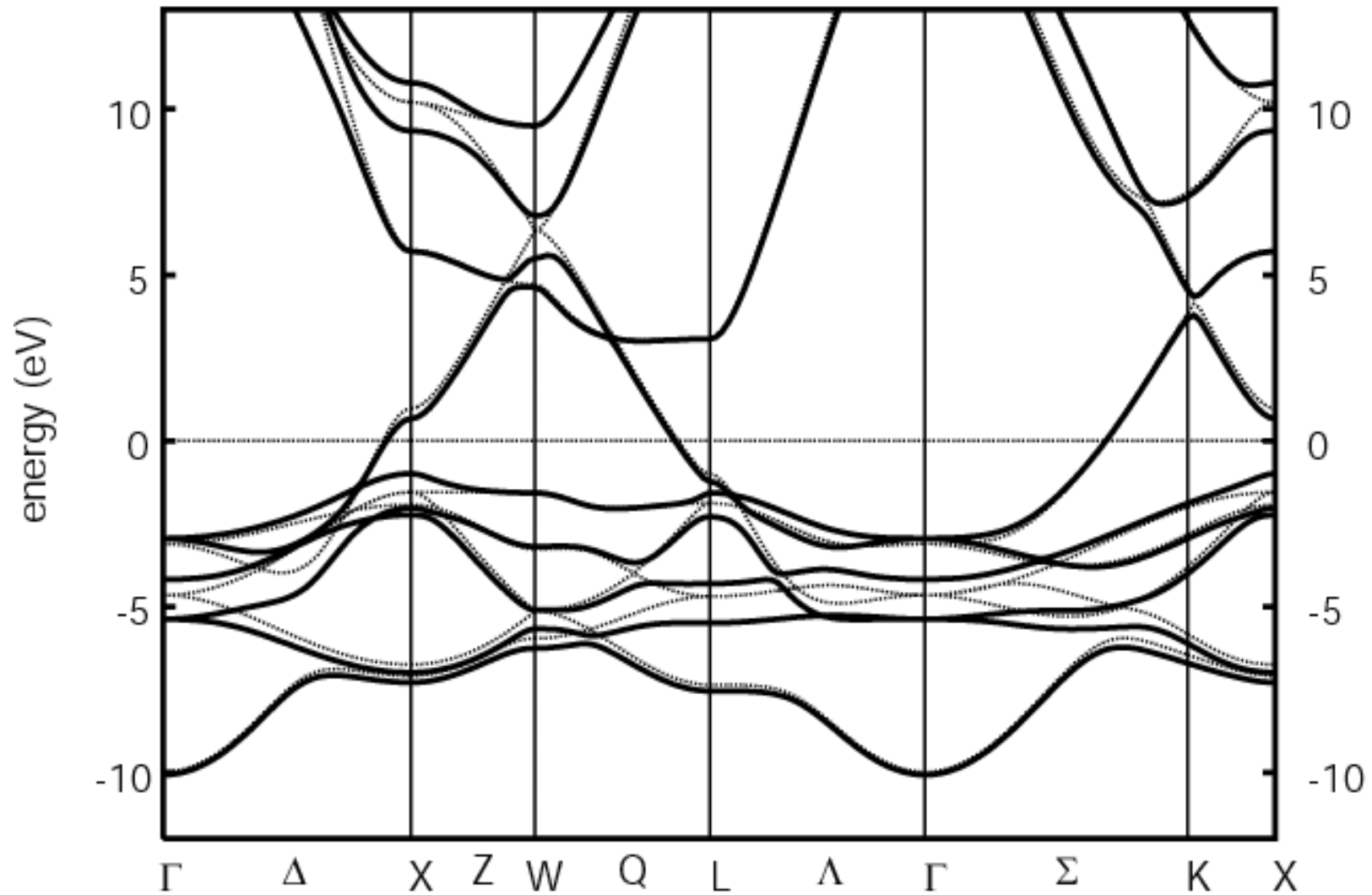
At  $\Gamma$  point,  $\Delta E_{so}$

$5d$ : 2.50 eV

$6p$ : 3.38 eV

# Band dispersion of fcc noble metals, Au

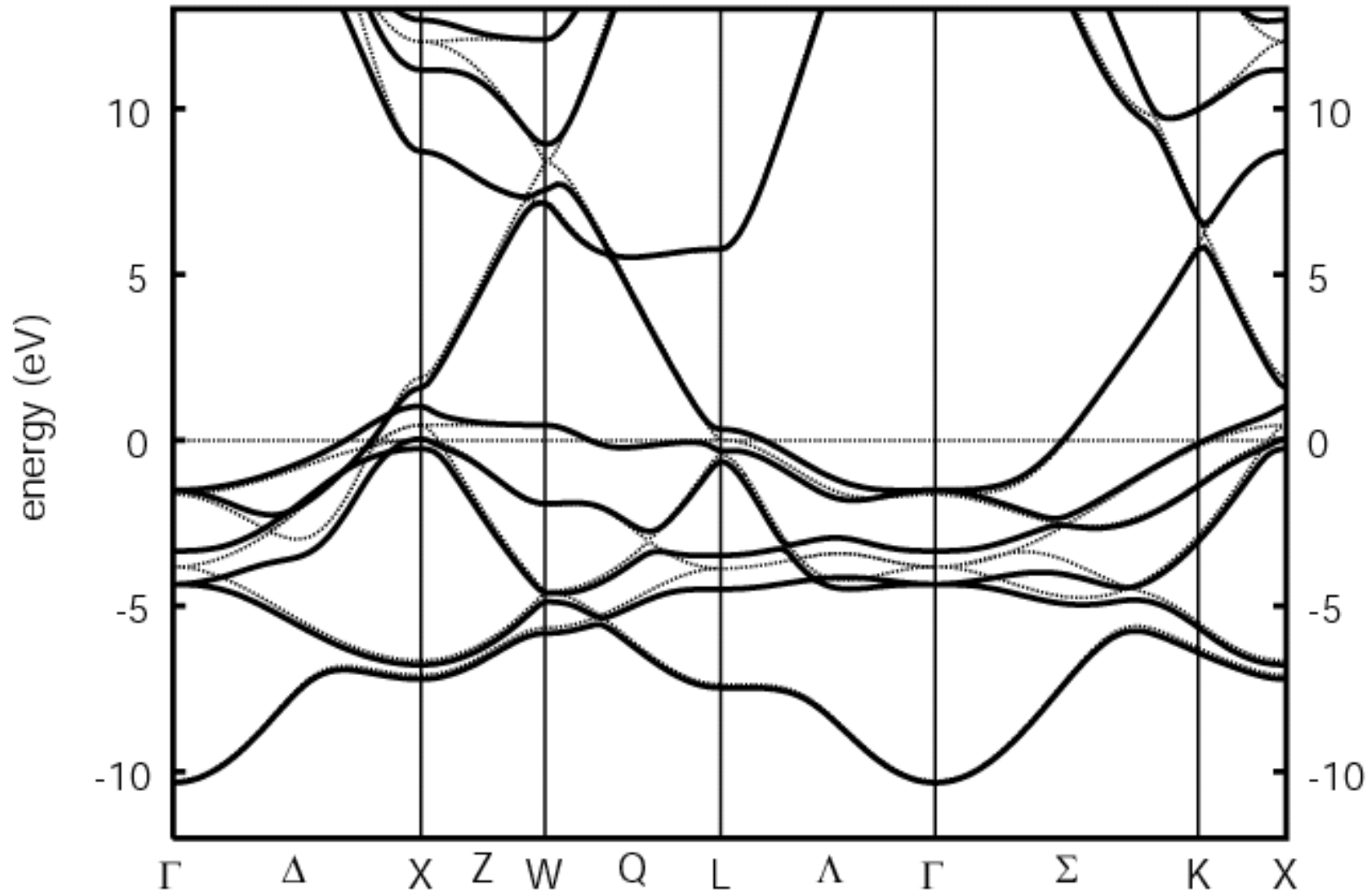
band dispersion (fcc-Au)



Au:  $a=7.71$  a.u.(exp.)

# Band dispersion of fcc noble metals, Pt

band dispersion (fcc-Pt)



Pt:  $a=7.41$  a.u.(exp.)

(Appendix 6)  
Kohn-Sham equation,  
variational principles,  
Car-Parrinello molecular dynamics,  
fully relativistic pseudo potential



# Density functional theory: Kohn&Sham equation

## Variational principles

$$\tilde{E}[n(\mathbf{r})] = E[n(\mathbf{r})] - \mu \left( \int n(\mathbf{r}) d\mathbf{r} - N_e \right)$$

$$\frac{\delta \tilde{E}}{\delta n(\mathbf{r})} = 0$$

Kohn&Sham equation  $\longrightarrow$  Electron density

$$\left\{ -\frac{1}{2} \nabla^2 + V_{\text{eff}}(\mathbf{r}) \right\} \Psi_i(\mathbf{r}) = \varepsilon_i \Psi_i(\mathbf{r}) \quad n(\mathbf{r}) = \sum_i^{\text{occ.}} |\Psi_i(\mathbf{r})|^2$$

Electron potential  $\longleftarrow$  Poisson equation

$$V_{\text{eff}}(\mathbf{r}) = V_{\text{ext}}(\mathbf{r}) + \frac{1}{2} \int \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' + \frac{\delta E_{\text{xc}}}{\delta n(\mathbf{r})}$$

# Car-Parrinello Molecular Dynamics for Noncollinear Magnetism

- Bispinor Wave Functions for Single Electron States

$$\Phi_k(r) = \begin{pmatrix} \phi_{k1}(r) \\ \phi_{k2}(r) \end{pmatrix}$$

Soft part

Augmented part  
(hard part)

- Density Matrix

$$\begin{aligned} \rho_{\alpha\beta}(r) &= \sum_k f_k \{ \phi_{k\alpha}(r) \phi_{k\beta}^*(r) + \sum_{Inm} Q_{nm}^I(r) \langle \beta_n^I | \phi_{k\alpha} \rangle \langle \phi_{k\beta} | \beta_m^I \rangle \} \\ &= \frac{1}{2} (n(r) \sigma_0 + m_x(r) \sigma_x + m_y(r) \sigma_y + m_z(r) \sigma_z)_{\alpha\beta} \end{aligned}$$

unit matrix  $\sigma_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$

Charge density  $n(r)$   
Spin density vector  $m_\alpha(r)$

Pauli matrix  $\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$   $\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$   $\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$

$$n(r) = \rho_{11} + \rho_{22}$$

$$m_x(r) = 2 \operatorname{Re} \rho_{12} \quad m_y(r) = -2 \operatorname{Im} \rho_{12} \quad m_z(r) = \rho_{11} - \rho_{22}$$

- Total Energy(Energy Functional)

$$E_{tot}[\{\Phi_k\}, \{R_I\}] = \sum_k f_k \langle \Phi_k | (-\frac{1}{2} \nabla^2 \sigma_0 + V_{NL}) | \Phi_k \rangle + \frac{1}{2} \iint \frac{n(r)n(r')}{|r-r'|} dr dr' \\ + \int V_{loc}^{ion}(r) n(r) dr + \underline{E_{XC}[n(r), m(r)]} + U_{ion}[\{R_I\}]$$

$$V_{loc}^{ion}(r) = \sum_I V_{loc}^I(r - R_I) \quad V_{NL} = \left( \sum_{nm} |\beta_m^I\rangle D_{nm}^{(0)I} \langle \beta_n^I| \right) \sigma_0$$

$$U_{ion}[\{R_I\}] = \frac{1}{2} \sum_{IJ} \frac{Z_I Z_J}{|R_I - R_J|} \quad \underline{m(r) = |\vec{m}(r)|}$$

$$\beta_n^I(r) \quad D_{nm}^{(0)I} \quad Q_{nm}^I(r) \quad V_{loc}^I(r)$$

These quantities are transferred from an atomic reference configuration. The pseudo potential is the ultra-soft type.

- Density Functional for the Exchange-Correlation Term.  
Local spin density approximation(LDA),  
Generalized gradient approximation(GGA,PW91)  
Van der Waals density functional (vdW-DF)

- Lagrangian (Car-Parrinello Molecular Dynamics)

$$L = m_{\Phi} \sum_k f_k \langle \dot{\Phi}_k | \dot{\Phi}_k \rangle + \frac{1}{2} \sum_I M_I \dot{R}_I^2 - E_{tot} [\{\Phi_k\}, \{R_I\}] + \sum_{kl} \Lambda_{kl} (\langle \Phi_k | S | \Phi_l \rangle - \delta_{kl})$$

- Molecular Dynamics (Euler-Lagrange equation)

$$m_{\Phi} \ddot{\Phi}_k(r) = -H \Phi_k(r) + \sum_{\ell} \frac{1}{f_k} \Lambda_{kl} S \Phi_{\ell}(r)$$

$$M_I \ddot{R}_I = F_I + \sum_{kl} \Lambda_{kl} \langle \Phi_k | \frac{\partial S}{\partial R_I} | \Phi_{\ell} \rangle$$

$$H = \frac{1}{f_k} \frac{\delta E_{tot}}{\delta \Phi_k} \quad \begin{pmatrix} \bar{\phi}_{k1}(r) \\ \bar{\phi}_{k2}(r) \end{pmatrix} = - \begin{pmatrix} H11 & H12 \\ H21 & H22 \end{pmatrix} \begin{pmatrix} \phi_{k1}(r) \\ \phi_{k2}(r) \end{pmatrix}$$

$$F_I = - \frac{\partial E_{tot}}{\partial R_I}$$

R. Car and M. Parrinello, Phys. Rev. Lett. 55, 2471 (1985).

# Install to the plane wave method (I)

spinor wave function

$$\Phi_k(r) = \begin{pmatrix} \phi_{k1}(r) \\ \phi_{k2}(r) \end{pmatrix}$$

density matrix

$$\rho_{\alpha\beta}(r) = \sum_k f_k \{ \phi_{k\alpha}(r) \phi_{k\beta}^*(r) + \sum_{I pq} \underline{Q}_{pq,\alpha\beta}^I(r) \langle \beta_p^I | \Phi_k \rangle \langle \Phi_k | \beta_q^I \rangle \}$$

spinor type projector function

$$\beta_p^I(r) = b_{j\kappa\tau}^I(r) Y_{j\mu}^{\text{sgn}(\kappa)}(r_I) \quad p = \{j\mu\kappa\tau\}$$

$$\begin{array}{l} j = \ell + \frac{1}{2}, \mu = m + \frac{1}{2} \\ \kappa = -\ell - 1 < 0 \end{array} \quad Y_{j,\mu}^{(-)} = \left( \frac{l+m+1}{2l+1} \right)^{1/2} Y_{l,m} \begin{pmatrix} 1 \\ 0 \end{pmatrix} + \left( \frac{l-m}{2l+1} \right)^{1/2} Y_{l,m+1} \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

$$\begin{array}{l} j = \ell - \frac{1}{2}, \mu = m - \frac{1}{2} \\ \kappa = \ell > 0 \end{array} \quad Y_{j,\mu}^{(+)} = \left( \frac{l-m+1}{2l+1} \right)^{1/2} Y_{l,m-1} \begin{pmatrix} 1 \\ 0 \end{pmatrix} - \left( \frac{l+m}{2l+1} \right)^{1/2} Y_{l,m} \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

# Install to the plane wave method (II)

nonlocal potential

$$V_{NL} = \sum_{I pq} |\beta_q^I\rangle D_{pq}^{(0)I} \langle \beta_p^I |$$

transfer from the atomic generation code

$$\beta_p^I(r) \quad D_{pq}^{(0)I} \quad Q_{pq,\alpha\beta}^I(r) \quad V_{loc}^I(r)$$

Kohn-Sham equation

$$H \Phi_k(r) = \varepsilon_k S \Phi_k(r)$$

$$H = \left( -\frac{1}{2} \nabla^2 \right) \sigma_0 + \bar{V}_{eff} + \sum_{I pq} |\beta_p^I\rangle D_{pq}^I \langle \beta_q^I | \quad S = 1 + \sum_{I pq} |\beta_q^I\rangle q_{pq}^I \langle \beta_p^I |$$

$$\bar{V}_{eff}(r) = \left( V_{loc}^{ion}(r) + \int \frac{n(r')}{|r-r'|} dr' + V_{xc}^N(r) \right) \sigma_0 + V_{xc}^M(r) \frac{1}{m(r)} \vec{m}(r) \cdot \vec{\sigma}$$

$$D_{pq}^I = D_{pq}^{(0)I} + \sum_{\alpha\beta} \int Q_{pq,\alpha\beta}^I(r) (V_{eff}(r))_{\alpha\beta} dr \quad V_{xc}^N(r) = \frac{\delta E_{XC}}{\delta n(r)} \quad V_{xc}^M(r) = \frac{\delta E_{XC}}{\delta m(r)}$$

TO and A. Hosokawa, PRB, **72**, 224428 (2005)

(Appendix 7)  
Magnetic anisotropy:  
shape magnetic anisotropy  
and  
magnetocrystalline anisotropy

# Shape Magnetic Anisotropy Energy (SMAE)

From magnetic dipole interaction(MDI)

$$\text{SMAE} = E_{\text{MDI}}^{[001]} - E_{\text{MDI}}^{[100]}$$

Continuum approach (CA)

$$\text{MAE} = E_{\text{MDI}}^{[001]} - E_{\text{MDI}}^{[100]} = \frac{1}{2} \mu_0 \frac{M^2}{\Omega}$$

$M$  : Total magnetization

$\Omega$  : Volume of magnetic material

$\mu_0$  : Permeability of vacuum

Discrete approach (DA) [1,2]

$\mathbf{m}(\mathbf{R}_i)$  : Atomic magnetic moment

$$E_{\text{MDI}}^{\mathbf{m}} = \frac{e^2}{4m^2c^2} \sum_{\mathbf{R}_i, \mathbf{R}_j}^{i \neq j} \left[ \frac{\mathbf{m}(\mathbf{R}_i) \cdot \mathbf{m}(\mathbf{R}_j)}{R_{ij}^3} - 3 \frac{\mathbf{m}(\mathbf{R}_i) \cdot (\mathbf{R}_i - \mathbf{R}_j) \mathbf{m}(\mathbf{R}_j) \cdot (\mathbf{R}_i - \mathbf{R}_j)}{R_{ij}^5} \right]$$

Spin density approach (SDA) [3]

$$E_{\text{MDI}}^{\mathbf{m}} = \frac{e^2}{4m^2c^2} \iint d\mathbf{r}_1 d\mathbf{r}_2 \left[ \frac{\mathbf{m}(\mathbf{r}_1) \cdot \mathbf{m}(\mathbf{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|^3} - 3 \frac{\mathbf{m}(\mathbf{r}_1) \cdot (\mathbf{r}_1 - \mathbf{r}_2) \mathbf{m}(\mathbf{r}_2) \cdot (\mathbf{r}_1 - \mathbf{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|^5} \right]$$

➤ High precision shape magnetic anisotropy from spin density distribution: magnetic interface/surface

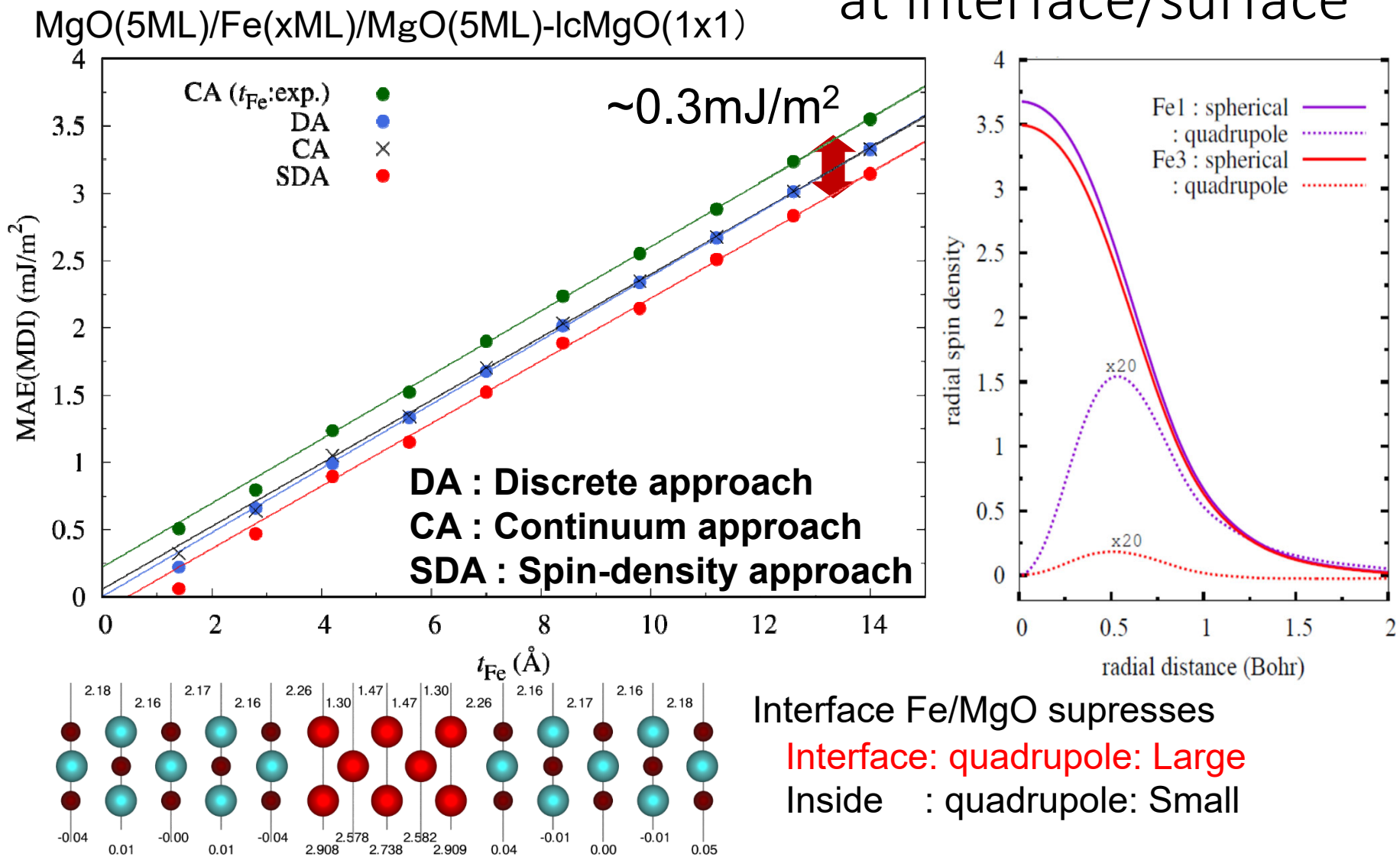
[1] H. J. G. Draaisma and W. J. M. de Jonge, J. Appl. Phys. 64, 1988.

[2] L. Szunyogh et al, Phys. Rev. B 51, 9552, 1995.

[3] T. Oda and M. Obata, J. Phys. Soc. Jpn. 87, 064803, 2018.



# Quadrupole atomic spin density distribution at interface/surface



TO, I. Pardede, et al., IEEE Trans. Magn. **55**, 1300104 (2018).

# Magnetic anisotropy energy (MAE)

$$\boxed{\text{MAE} = \text{SMAE} + \text{MCAE}}$$

$$\text{SMAE} = E_{\text{MDI}}[100] - E_{\text{MDI}}[001] \quad \text{MCAE} = E_{\text{SDFT}}[100] - E_{\text{SDFT}}[001]$$

Total Energy (Energy Functional: SDFT)

$$E_{\text{tot}}[n(\mathbf{r}), \mathbf{m}(\mathbf{r})] = E_{\text{MDI}}[\mathbf{m}(\mathbf{r})] + E_{\text{SDFT}}[n(\mathbf{r}), \mathbf{m}(\mathbf{r})]$$

Kohn-Sham equation

$$\left[ -\frac{1}{2} \nabla^2 + V_{\text{eff}}^{\text{SDFT}}(\mathbf{r}) + V_{\text{MDI}}^{\text{SDI}}(\mathbf{r}) \right] \Phi_i(\mathbf{r}) = \varepsilon_i \Phi_i(\mathbf{r})$$

Grand-canonical force theorem (GCFT) for MCAE

$$\delta E_{\text{SDFT}}^{\mathbf{m}, \text{GCFT}} = \sum_{\mathbf{k}} \sum_{\ell} \delta \varepsilon_{\ell \mathbf{k}}^{\mathbf{m}, \text{GCFT}}$$

$$\delta \varepsilon_{\ell \mathbf{k}}^{\mathbf{m}, \text{GCFT}} = f_{\ell \mathbf{k}}^{\mathbf{m}} (\varepsilon_{\ell \mathbf{k}}^{\mathbf{m}, \text{GCFT}} - \mu_0) - f_{\ell \mathbf{k}}^0 (\varepsilon_{\ell \mathbf{k}}^0 - \mu_0)$$

**MCAE**

- Atom-resolved
- $\mathbf{k}$ -resolved

SAME: Shape magnetic anisotropy energy

MCAE: Magnetocrystalline anisotropy energy

GCFT: D. Li et al., Phys. Rev. B 88, 214413 (2013);

I. Pardede et al., J. Magn. Magn. Mater., 500, 166357 (2020).