5th Lecture: Spintronics, Design, Magnetization Control I

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Spintronics Design Course in CMD-WS35

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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）

検索：

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(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
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(5-5) スピン軌道相互作用、逆格子空間スピンテクスチャ
(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

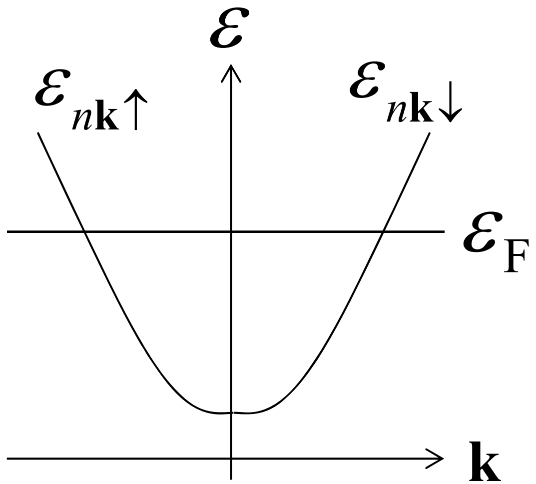
$\varepsilon_{n\mathbf{k}\sigma}$: eigenvalue

n : band index

$\mathbf{k}$: wave number

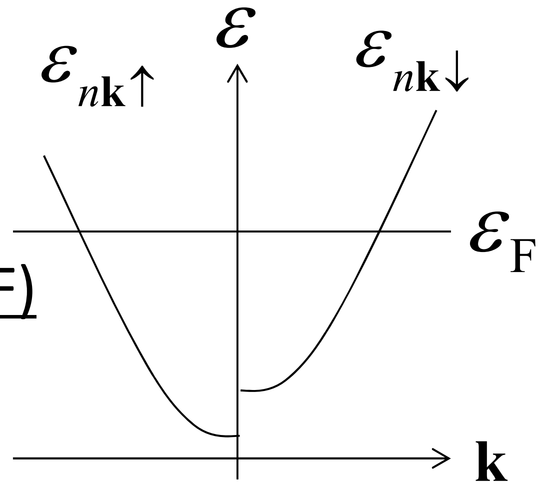
σ : spin index $(\sigma = \uparrow, \downarrow)$

Band dispersion



Ferromagnet(F)

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



Anti-Ferromagnet(AF)

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

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

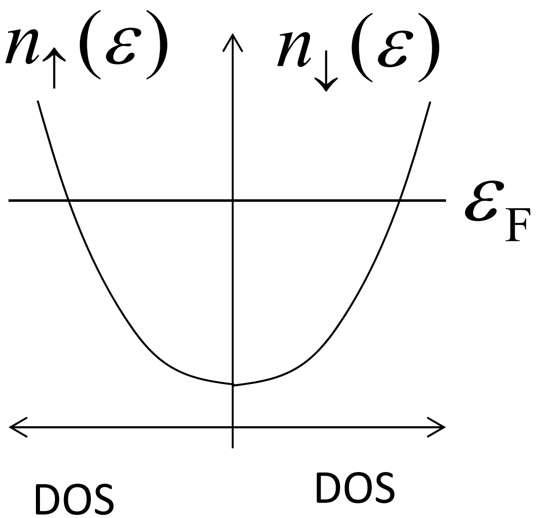
Density of states(DOS)

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

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

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

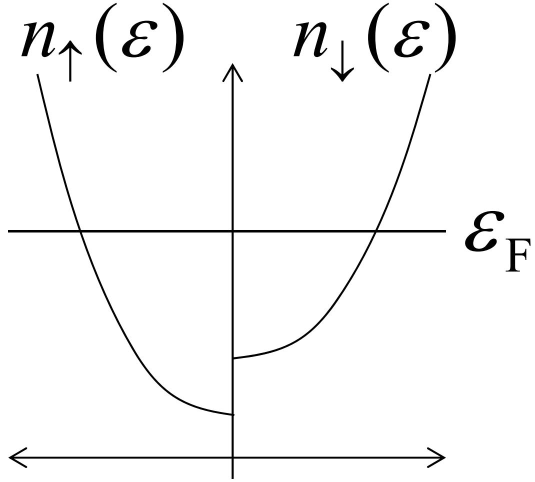
Spin-up state　　Spin-down state



[states/(energy unit)/cell]

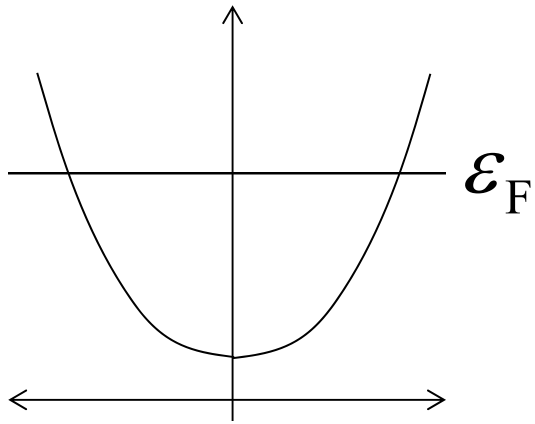
Ferromagnet(F)

Majority spin state　　Minority spin state



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

Anti-Ferromagnet(AF)



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

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

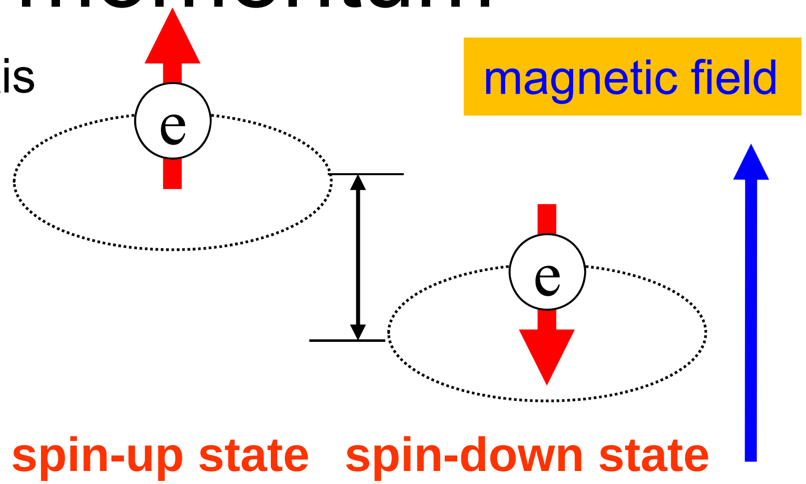
main orgin of magnetism

electron：spin angular momentum

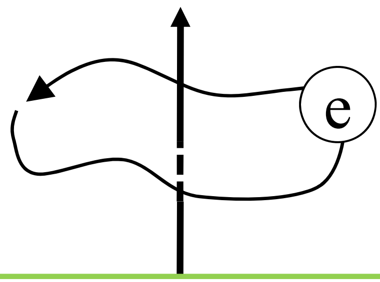
like being supposed to rotate on its own axis

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

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



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



$$\boxed{0\,,\hbar\,,2\hbar\,,\ \ldots}$$

(5-2-2) Orbital magnetic moment

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

An electron in a magnetic field along z-axis

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

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

$$-\mu_{\mathrm{B}}\vec{\ell}\cdot\vec{H}=-\overrightarrow{\mu_\ell}\cdot\vec{H}$$

$$-\frac{\partial\mathcal{H}}{\partial H}=\underline{-\frac{e}{2mc}\left(x\frac{\hbar}{i}\frac{\partial}{\partial y}-y\frac{\hbar}{i}\frac{\partial}{\partial x}\right)}\underline{-\frac{e^2H}{4mc^2}\left(x^2+y^2\right)}$$

$$\overrightarrow{\mu_\ell}=-\boxed{\frac{e\hbar}{2mc}}\vec{\ell}=-\underline{\mu_{\mathrm{B}}}\vec{\ell}$$

orbital magnetic moment

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

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

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

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

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

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

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

Contribution from the *i*'th electron $\vec{\mu}_i = -\left(2\vec{s}_i + \vec{\ell}_i\right)\mu_{\mathrm{B}}$

Dirac equation (as a reference, see Appendix 4)

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

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

ρ_1, ρ_3, σ **4 × 4 matrixes**

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

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

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

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

A_0 **scalar potential**

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

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

(5-3) Zeeman energy

$$\Bigg[\frac{1}{2m}\left(\vec{p}+\frac{e}{c}\vec{A}\right)^2+V+\underbrace{\frac{\hbar e}{2mc}\vec{\sigma}\cdot\vec{H}}_{\text{Zeeman energy}} \qquad \vec{H}=\vec{\nabla}\times\overline{A}$$

$$+\underbrace{\frac{\hbar}{4m^2c^2}\vec{\sigma}\cdot\left\{(\mathrm{grad}\ V)\times\left(\vec{p}+\frac{e}{c}\vec{A}\right)\right\}}_{\text{Spin-orbit interaction}}$$

$$+\underbrace{\frac{\hbar^2}{8m^2c^2}\mathrm{div}\,(\mathrm{grad}\ V)}_{\text{Darwin term}}\Bigg]\varphi_L=\left(\varepsilon-mc^2\right)\varphi_L$$

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

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

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

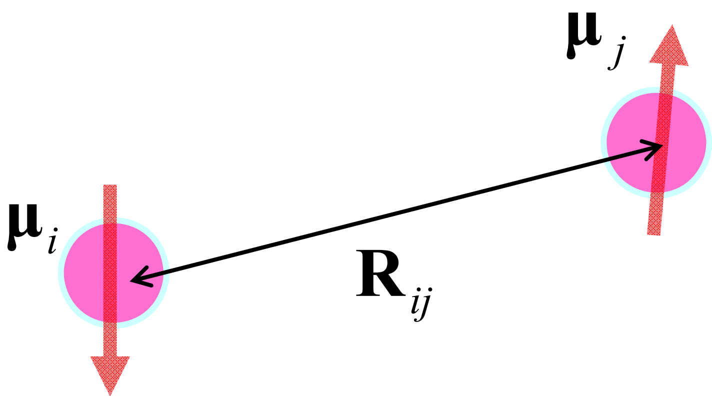
Interaction between atomic magnetic moments

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

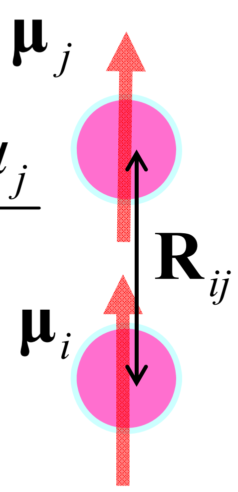
(5-4-2) Magnetic dipole-dipole interaction

Classical magnetic interaction

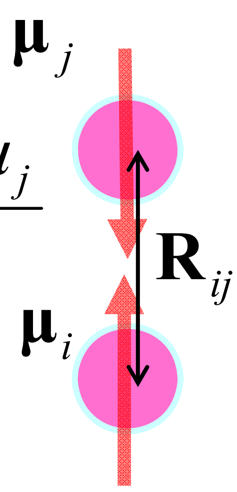
$$E_{ij}^{\text{dipole}} = \frac{\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j - 3(\boldsymbol{\mu}_i \cdot \hat{\mathbf{R}}_{ij})(\boldsymbol{\mu}_j \cdot \hat{\mathbf{R}}_{ij})}{R_{ij}^3}$$



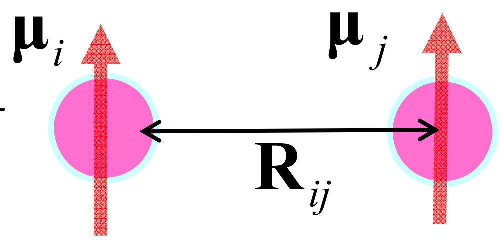
$$-\frac{2\mu_i\mu_j}{R_{ij}^3}$$



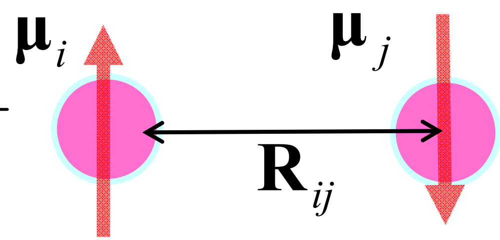
$$+\frac{2\mu_i\mu_j}{R_{ij}^3}$$



$$+\frac{\mu_i\mu_j}{R_{ij}^3}$$



$$-\frac{\mu_i\mu_j}{R_{ij}^3}$$



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

- 1D-chain

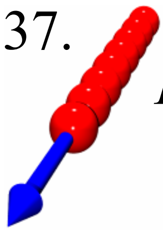
$$E_{\mathrm{dd}}(\theta) = \frac{e^2}{4m^2c^2}\frac{C_{\mathrm{M}}\mu^2}{a^3}\left(\frac{3}{2}\cos^2\theta - \frac{1}{2}\right)$$

$C_{\mathrm{M}} = 4.804$

μ : Magnetic moment in μ_{B}

a : Lattice constant in a_{B}

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



$E^{\mathrm{dd}}_{\mathrm{MAE}} = 0.08\ \mathrm{meV/Fe}$

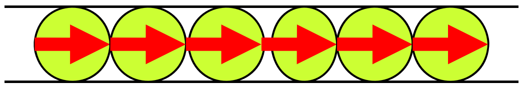
$\mu = 3\,\mu_{\mathrm{B}} \quad a = 2.77$ Å

- 2D-dimensional case

2D Madelung constant approach

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

2D square lattice



$E^{\mathrm{dd}}_{\mathrm{MAE}} = 0.15\ \mathrm{meV/Fe}$

$\mu = 3.1\mu_{\mathrm{B}}$

$a = 2.87$ Å

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

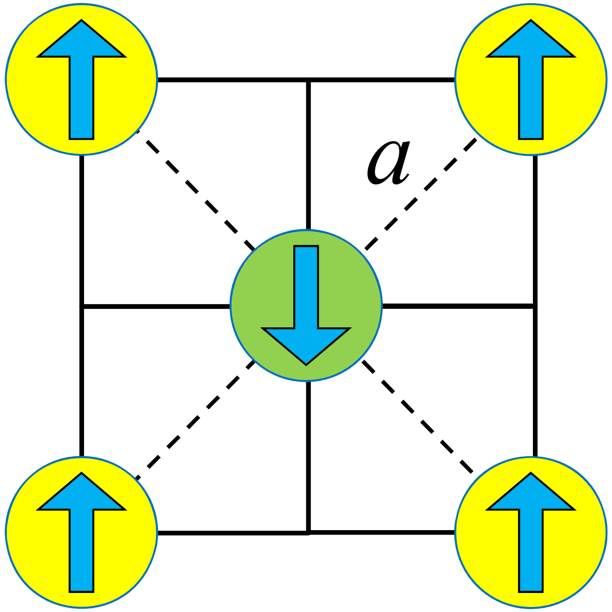
$C_{\mathrm{M}} = 9.03362$

μ : Magnetic moment in μ_{B}

a : Lattice constant in a_{B}

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

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



Perpendicular anisotropy

$$E^{\mathrm{dd}}_{\mathrm{MAE}} = -0.082 \text{ meV/Mn}$$

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

$$a = 2.83 \text{ Å}$$

Note)

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

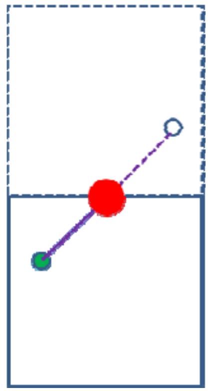
(5-5) Spin-orbit interaction

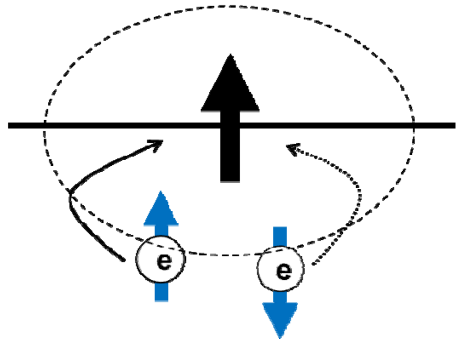
$$H_{\mathrm{SOI}} = \frac{\hbar}{4m^2c^2}\vec{\sigma}\cdot\left(\underline{\mathrm{grad}\ V(\vec{r})}\times\vec{p}\right)$$

$\vec{\sigma}$ Pauli's matrix

$$V(\mathbf{r}) \approx -\frac{Ze^2}{r} \qquad \mathrm{grad}\,V(r) \approx \frac{dV}{dr}\frac{\mathbf{r}}{r}$$

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





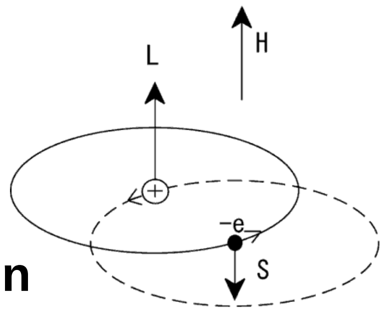
$$H_{SOI} = \xi\,\vec{\ell}\cdot\vec{\sigma} = \underline{\xi\,(\ell_x\sigma_x + \ell_y\sigma_y + \ell_z\sigma_z)}$$

connects orbital and spin spaces

$$\xi(r) = \frac{\hbar^2}{4m^2c^2r}\frac{dV}{dr}$$

2

Biot-Savart law in the classical electromagnetics



spin-orbit interaction

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

Due to the surface/interface the inversion symmetry breaks.

➡ Rashba effect, etc.

(5-5-2) Spin-texture in the reciprocal space: Rashba effect 1

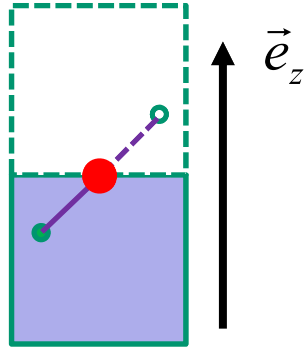
2 dimensional free electron with spin-orbit interaction

$$H = -\frac{\hbar^2}{2m}\nabla^2 + \vec{\sigma}\cdot(\vec{\alpha}\times\vec{p})$$

$$\vec{\alpha} \propto \left\langle \mathrm{grad}\, V(r) = \frac{1}{r}\frac{dV(r)}{dr}\mathbf{r} \right\rangle$$

$$\propto \vec{e}_z$$

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



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

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

$$\vec{k} = (k_x, k_y) \qquad k = \sqrt{k_x^2 + k_y^2}$$

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

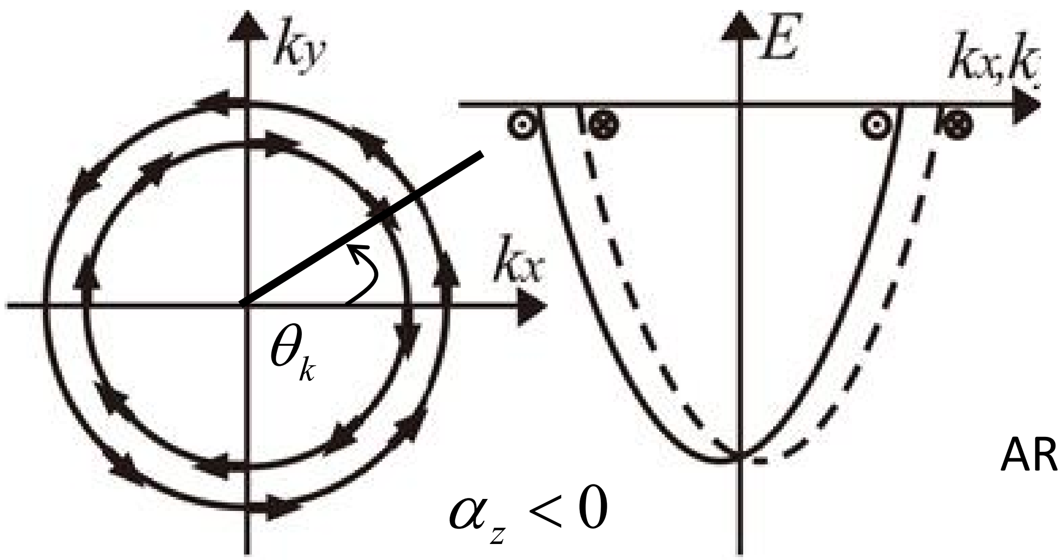
$$E_- = \frac{\hbar^2 k^2}{2m} - \alpha_z k \qquad \varphi_{\vec{k}}^- = \frac{1}{\sqrt{\Omega}} e^{i\vec{k}\cdot\vec{r}} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -ie^{i\theta_k} \end{pmatrix} \propto \frac{1}{\sqrt{2}} \begin{pmatrix} -e^{-i(\theta_k+\pi/2)/2} \\ e^{i(\theta_k+\pi/2)/2} \end{pmatrix}$$

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

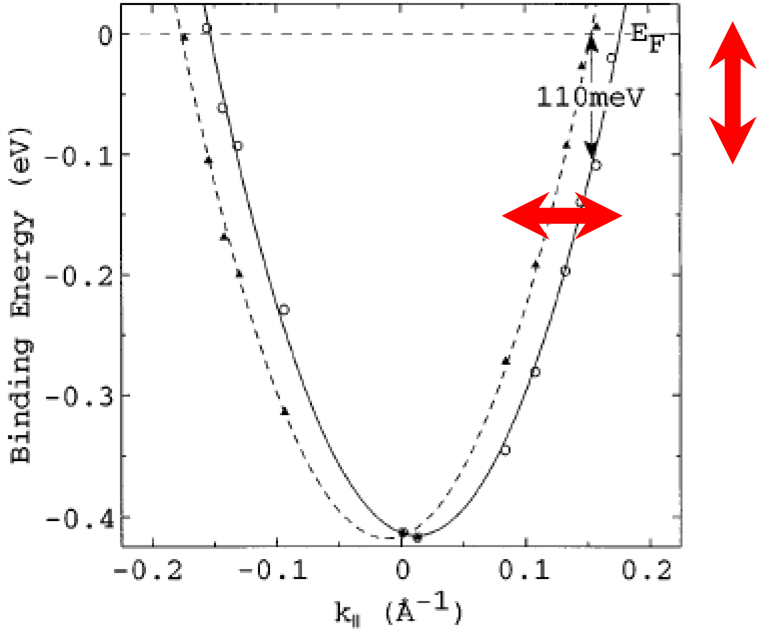
$$\left\langle \varphi_{\vec{k}}^{+} \middle| \vec{\sigma} \middle| \varphi_{\vec{k}}^{+} \right\rangle = (-\sin\theta_k, \cos\theta_k, 0)$$

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

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



Au(111) surface: ARPES



- High resolution

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

ARPES: Angle Resolved Photoelectron Spectroscopy

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

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

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

Normal Rashba term

General Rashba term **Effective in-plane potential gradient**

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

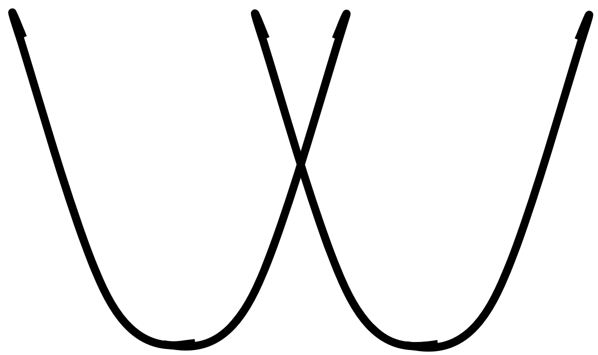
(5-5-5) Interpretation of Spin-orbit interaction

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

$$H_{\mathrm{SOI}} = 2\underbrace{\frac{\hbar e}{2mc}}_{\text{Bohr magneton}}\frac{\vec{\sigma}}{2}\cdot\left\{\frac{1}{2mce}\left(\vec{\nabla}V(\vec{r})\right)\times\vec{p}\right\} = 2\mu_{\mathrm{B}}\frac{\vec{\sigma}}{2}\cdot\underbrace{\left\{\frac{1}{2mce}\left(\vec{\nabla}V(\vec{r})\right)\times\vec{p}\right\}}_{\text{Effective magnetic field}}$$

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

$$H_{\mathrm{SOI}} = \underbrace{-e\frac{\hbar}{4m^2c^2}\left(\vec{\sigma}\times\vec{p}\right)}_{\text{Effective electric dipole}}\cdot\underbrace{\frac{1}{e}\vec{\nabla}V(\vec{r})}_{\text{Electric field}}$$



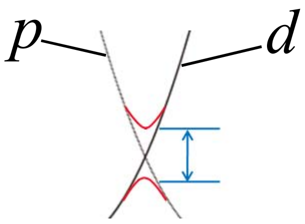
(5-5-6) Electric field effects in electronic structures of the surface/interface

- Stark effect

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

Electric dipole

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



- Rashba (SOI) effect

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

Modification of the Rashba effect.

- Electron depletion (accumulation)

When imposing the EF, for PDOS ~ 1 states/eV

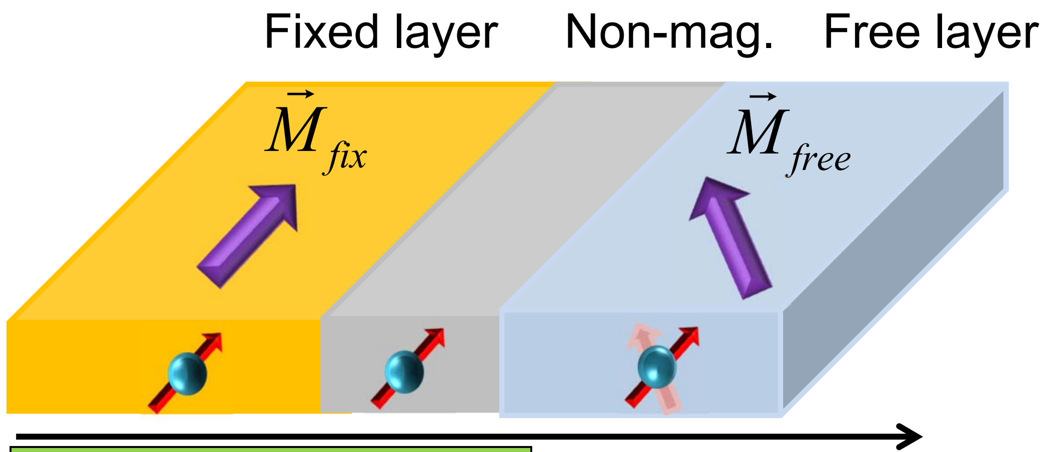
Induced change in the number of electrons	~0.01	~0.01 eV
Induced change in the number of electrons	~0.1	~0.1 eV

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

(5-6) Spin transfer torque

Spin current produced in fixed layer ⟶ **Spin torque** provided to free layer

$$N_{\mathrm{stt}} = \frac{d\vec{M}_{\mathrm{free}}}{dt} \propto -I_{\mathrm{s}}\vec{M}_{\mathrm{free}} \times \left(\vec{M}_{\mathrm{free}} \times \vec{M}_{\mathrm{fix}}\right)$$

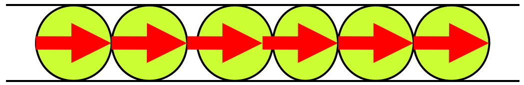


Spin polarized current

Conservation law of (spin) angular momenta

J.C. Slonczewski, J. Magn. Magn. Mater. 159, L1 (1996).

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

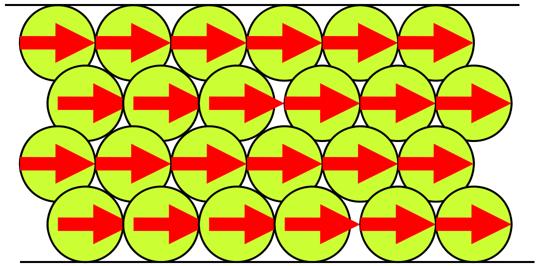


Magnetostatic contribution

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

2D square lat.

Shape aniso.

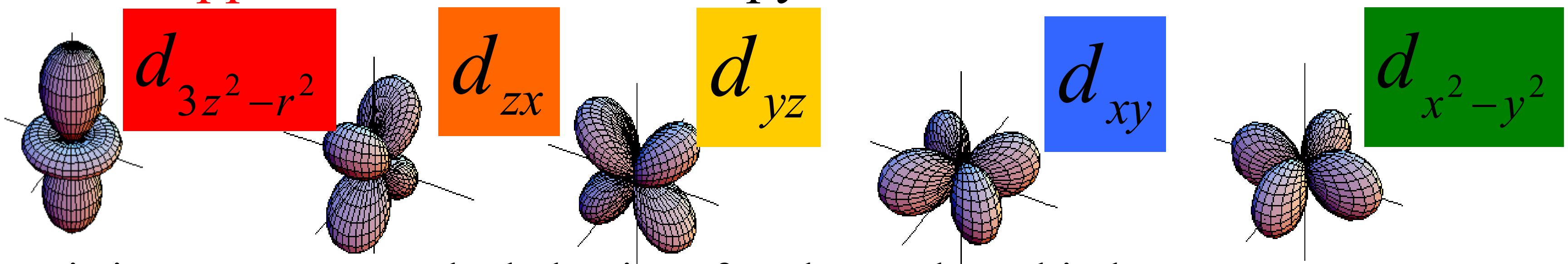


This depends on the arrangement of magnetic atoms, not so depend on electric field.

Electronic structure contribution

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

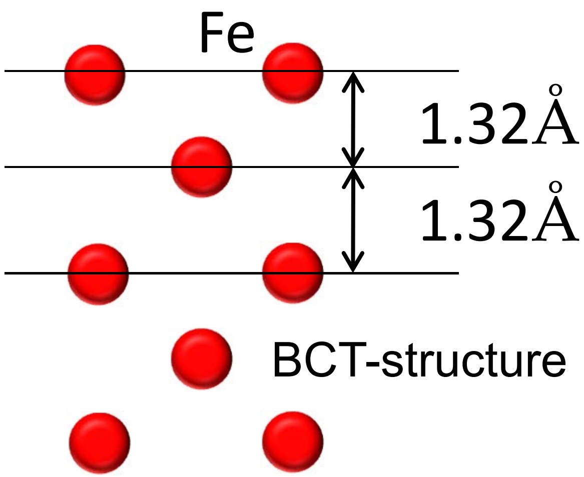
perturbation of spin-orbit interaction,
MA appears from an anisotropy of orbitals



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

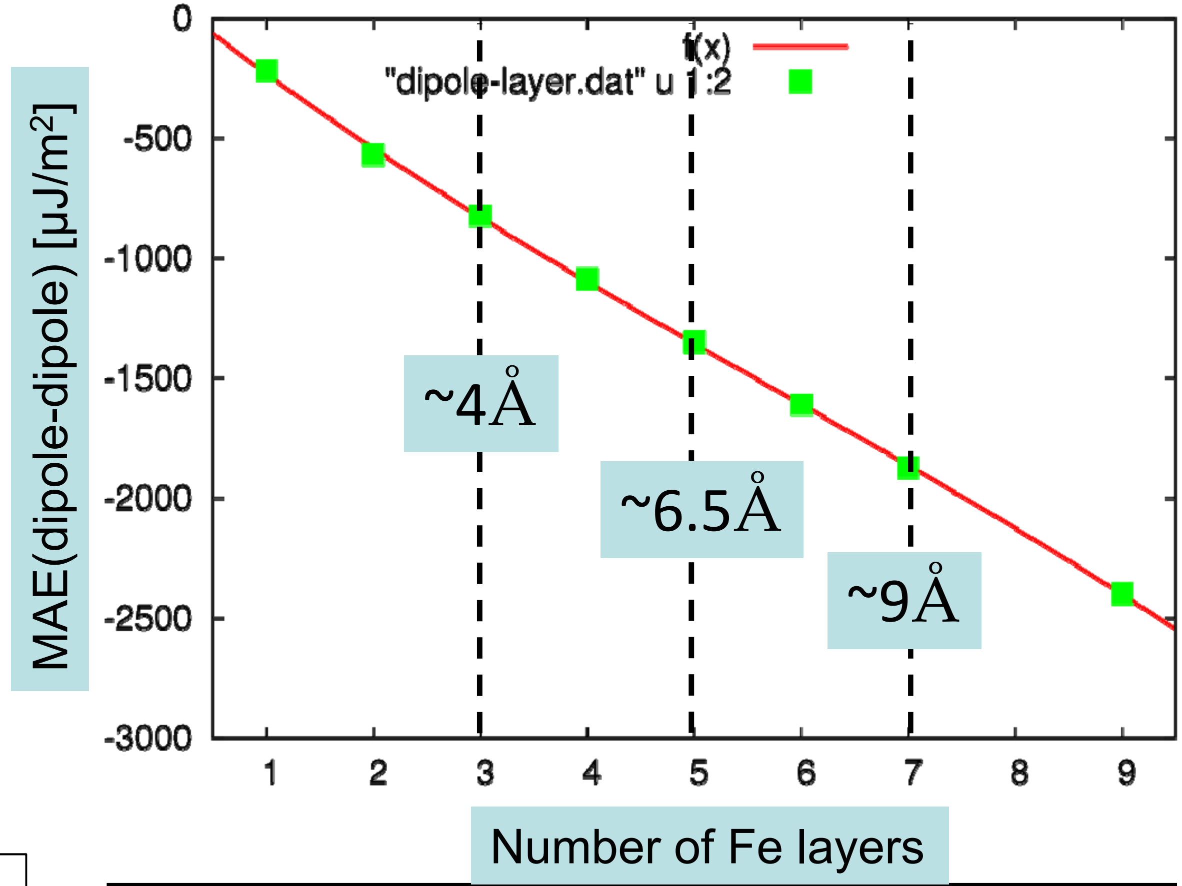
In-plane lattice constant
MgO 4.21Å



Fe: 2.96 μ_B(interface)
2.63 μ_B(inside)

FeCo layer
> 6.3Å in-plane
< 6.3Å out-of-plane

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



In-plane MAE increases by 250-300μJ/m² per Fe-layer

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

Origin of MAE in electronic structure

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

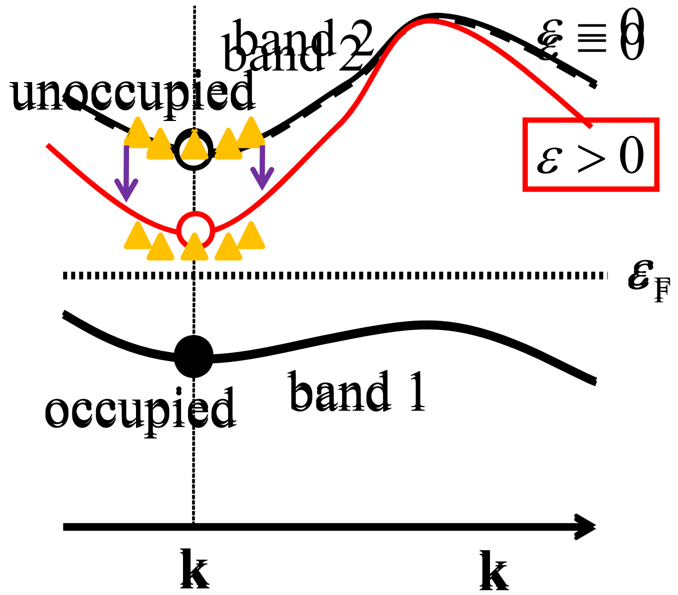
•**spin-orbit coupling contribution from band electrons**

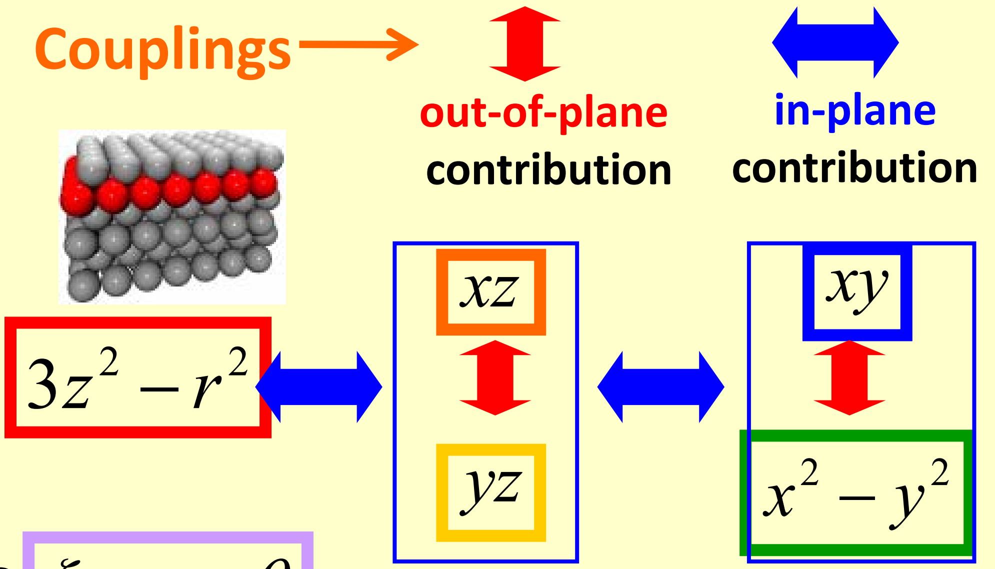
matrix element

z	$\langle xz \lvert \ell_z \rvert yz \rangle = 1$ $\langle x^2 - y^2 \lvert \ell_z \rvert xy \rangle = 2$
x	$\langle 3z^2 - r^2 \lvert \ell_x \rvert yz \rangle = \sqrt{3}$ $\langle xy \lvert \ell_x \rvert xz \rangle = 1$ $\langle x^2 - y^2 \lvert \ell_x \rvert yz \rangle = 1$
y	$\langle 3z^2 - r^2 \lvert \ell_y \rvert xz \rangle = \sqrt{3}$ $\langle xy \lvert \ell_y \rvert yz \rangle = 1$ $\langle x^2 - y^2 \lvert \ell_y \rvert xz \rangle = 1$

(A) 2nd perturbative contributions

$$\mathrm{MAE} = E_x - E_z \approx (\xi)^2 \sum_{o,u} \frac{\left|\langle o_\downarrow | \ell_z | u_\downarrow \rangle\right|^2 - \left|\langle o_\downarrow | \ell_x | u_\downarrow \rangle\right|^2}{\varepsilon_u^\downarrow - \varepsilon_o^\downarrow}$$





(B) Existence of partly-occupied degenerate levels

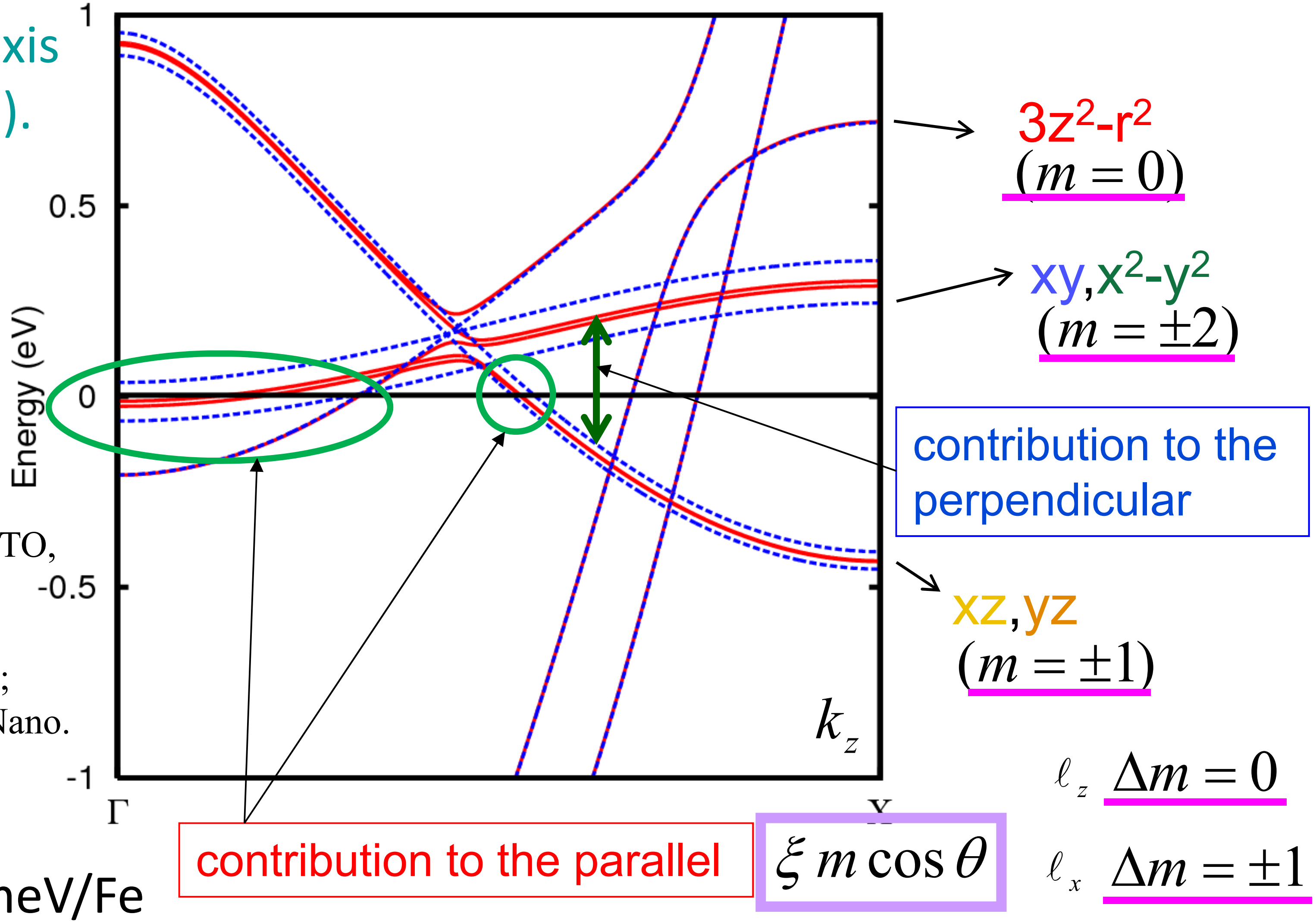
$\xi m \cos\theta$

Low dimentional system **D. S. Wang, R. Wu, and A. J. Freeman, Phys. Rev. B 47,14932 (1993).**

example: Isolated iron chain

- MAE=3.23 meV/Fe
- Easy axis: parallel to the chain-axis (z direction).

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



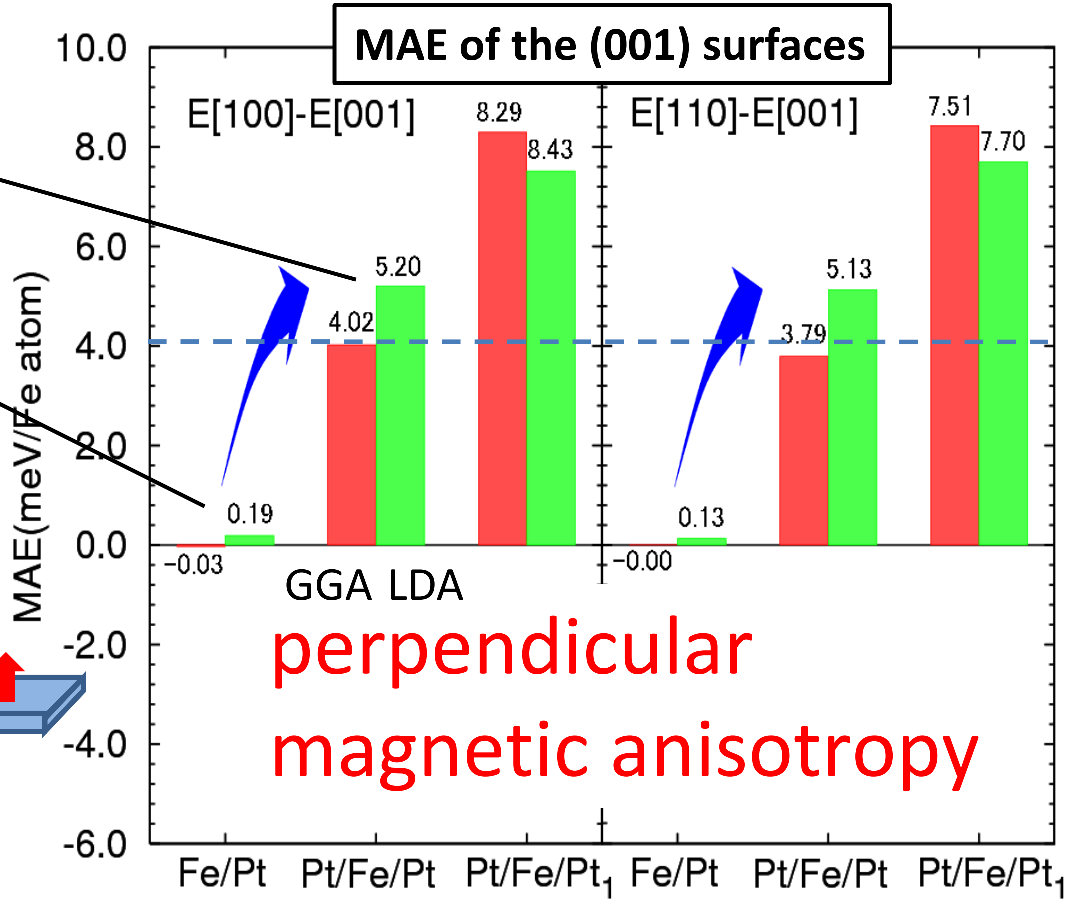
M. Tsujikawa and TO, J. Phys.: Condens. Matter, **21**, 064213 (2009); J. Compt. Theor. Nano. **6**, 2597 (2009).

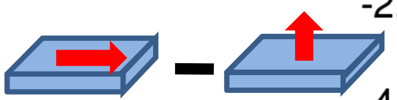
dipole: 0.05meV/Fe

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

11mJ/m^2

0.40mJ/m^2

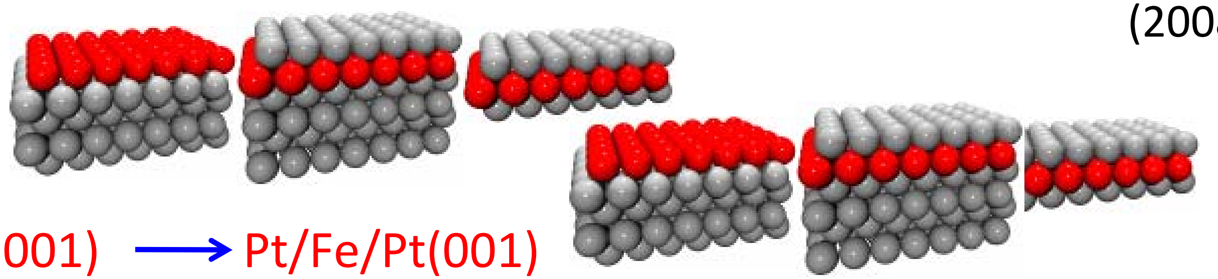




L1$_0$-FePt regular alloy(LDA)
MAE
2.61meV[a]、
2.724meV[b]

[a]P. Ravindran *et.al*, (2001).
[b]TO *et.al*, (2005).

Tsujikawa et al. (2008, 2009)



Fe/Pt(001) ⟶ Pt/Fe/Pt(001)

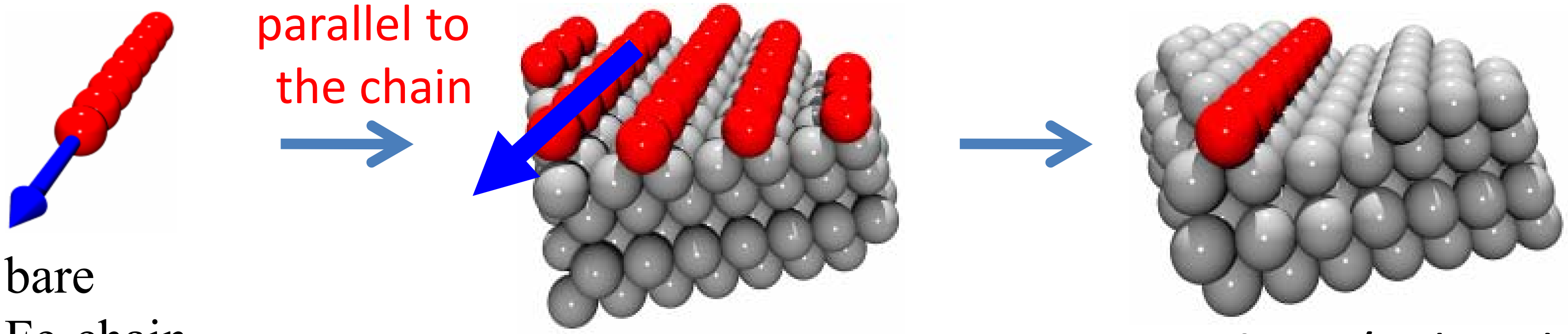
dipole: -0.32mJ/m^2

MAE of Fe-chain/Pt(664) surface

-3.23 meV/Fe

-2.25 meV/Fe

1.19 meV/Fe



bare
Fe-chain

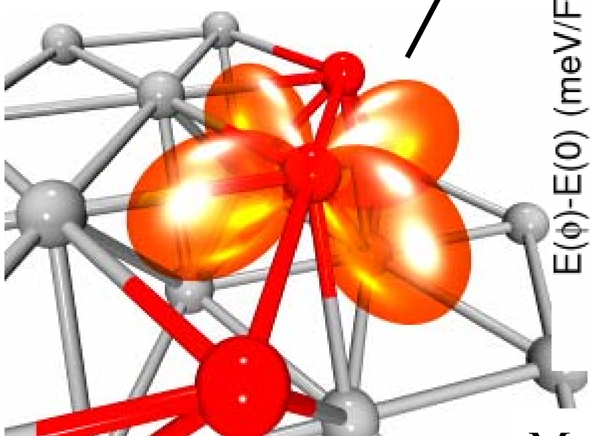
Fe-chain/Pt(111)

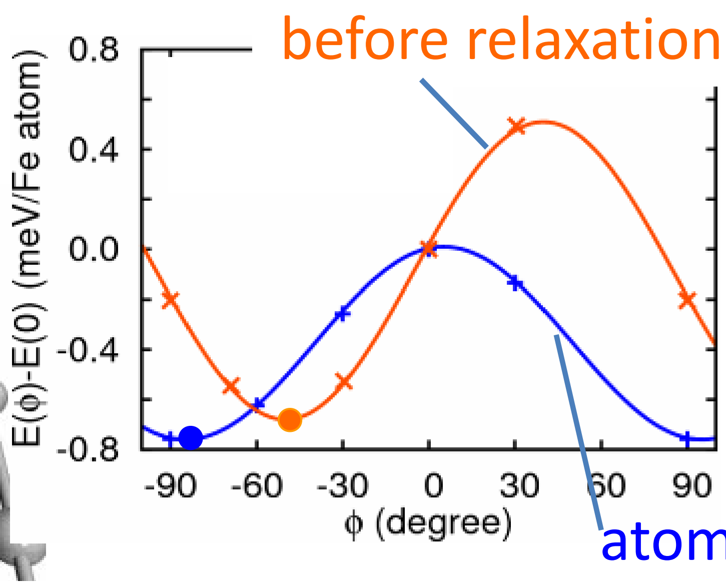
Fe-chain/Pt(664)

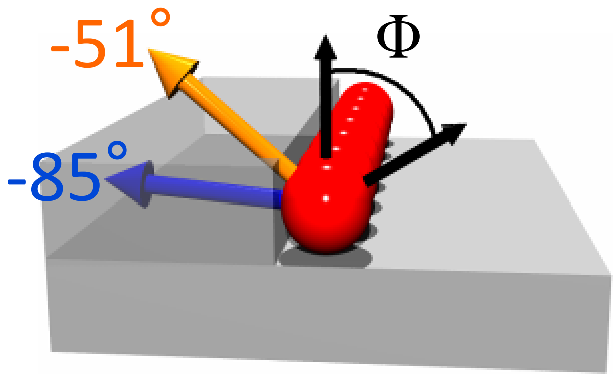
perpendicular to the chain

hybridization of 3*d* orbitals between Fe and Pt atoms

yz orbital



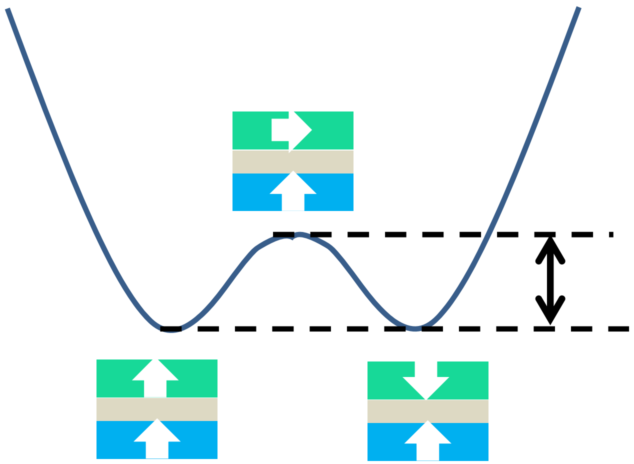




exp. -80°
(Repetto et al., 2006)

M. Tsujikawa A. Hosokawa, and TO, Phys. Rev. B, 77 (2008) 054413

Trade-off property in magnetic memory



$K_U V$

$K_U V$ becomes small, loosing memory

= small thermo-stability

Trade−off

$K_U V$ Becomes large, increasing the barrier for magnetization reverse

= large threshold of magnetization reverse

One of necessary condition

Non-volatile

$\boxed{\Delta \geq 60}$ (ten years)

$$\Delta = \frac{K_U V}{k_B T}$$

$K_U V$: Magnetic anisotropy energy of magnetic memory

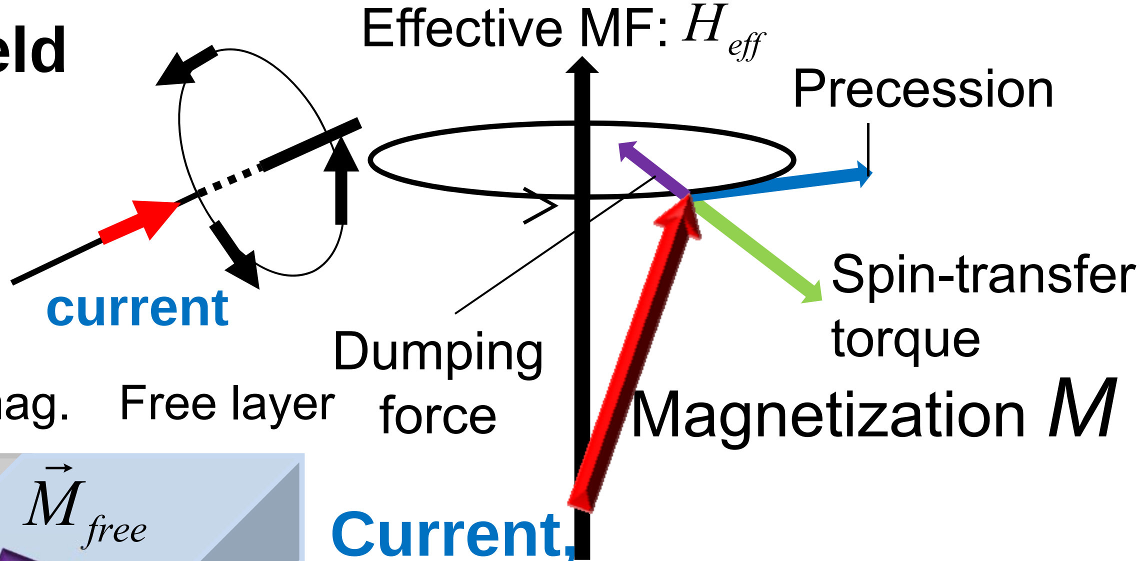
$k_B T$: Heat energy

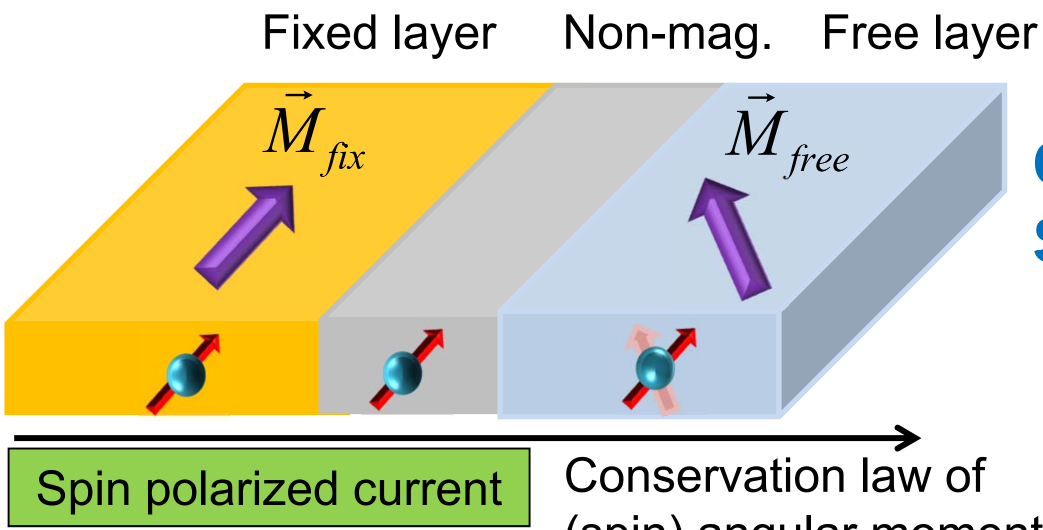
K_U Magnetic anisotropy energy per volume

V Volume of magnet

Driving forces in dynamical control of magnetization

- **External magnetic field (by current)**
- **Injection of spin polarized current**





Conservation law of (spin) angular momenta

Current,
Spin polarized current

Problem: Disturbing compactness,
energy consumption,
Joule heating, etc.

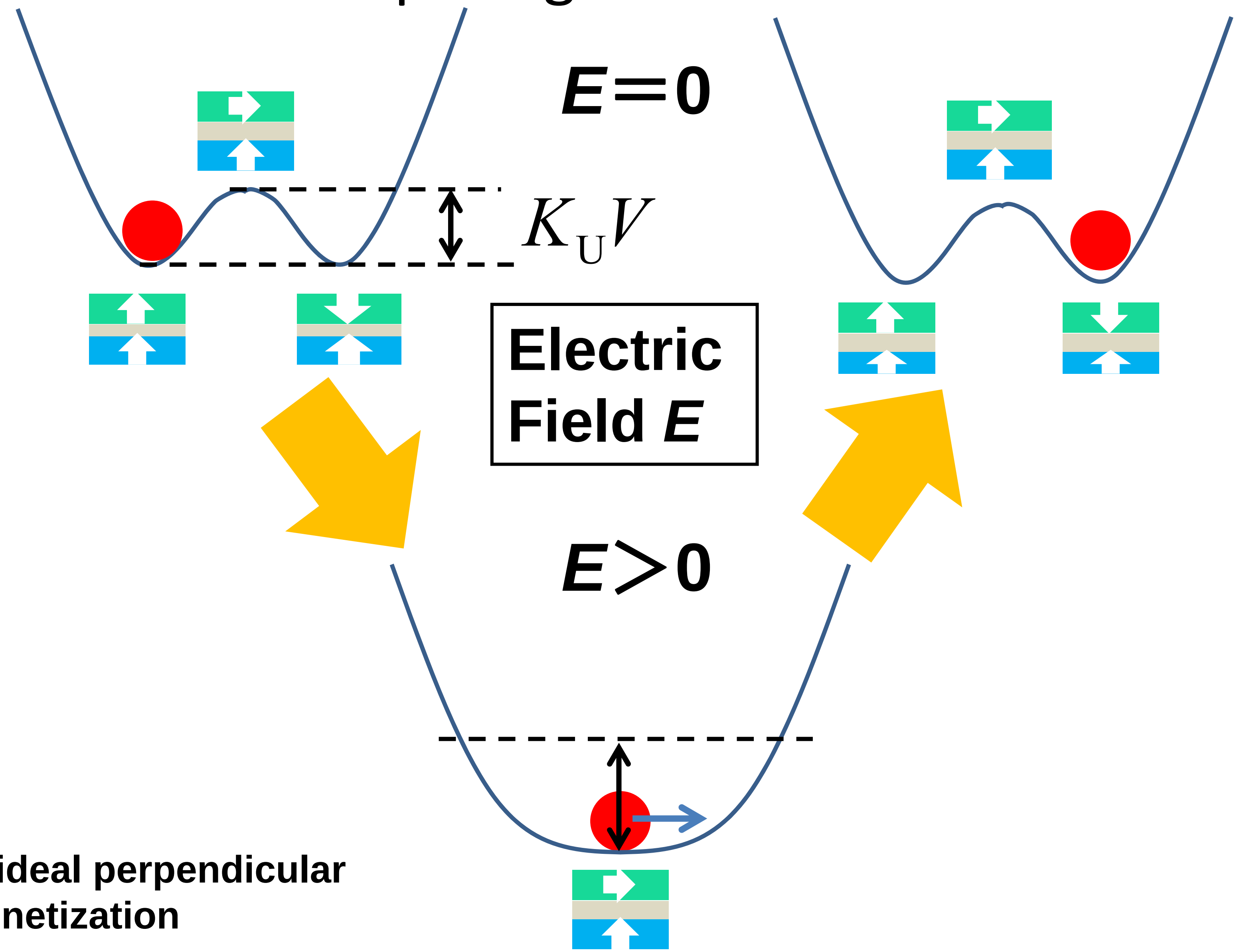
↓

Electric field (Voltage)

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

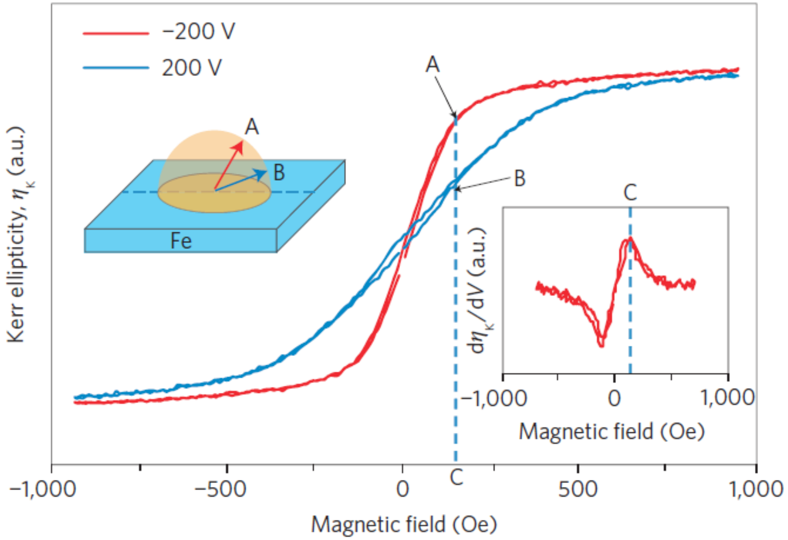
Expectation: ultra-low energy consumption, non-volatile property, compactness(high density memory), enough high speed in reading&writing

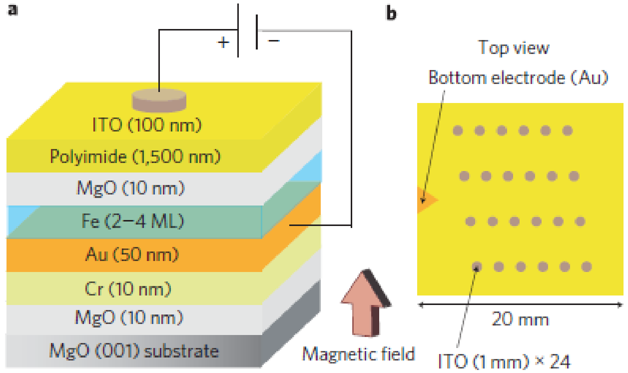
Schematic diagram of magnetization reversal in imposing electric field

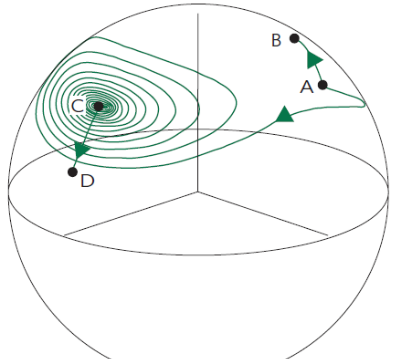


For ideal perpendicular magnetization

(5-9) Voltage-induced spin torque







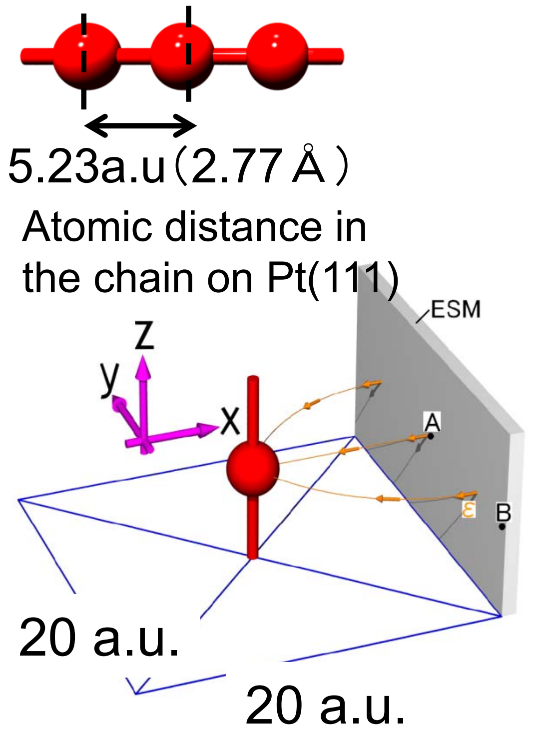
MgO/Fe/Au(001)

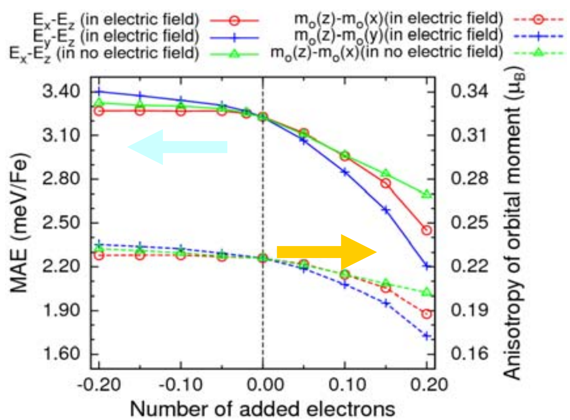
$$\frac{d\vec{M}}{dt} = \gamma\vec{M} \times \vec{H}_{eff} + \frac{\alpha\vec{M}}{M_s} \times \frac{d\vec{M}}{dt} \qquad \vec{H}_{eff} = -\frac{1}{\mu_0 M_s}\vec{\nabla}E_{mag}$$

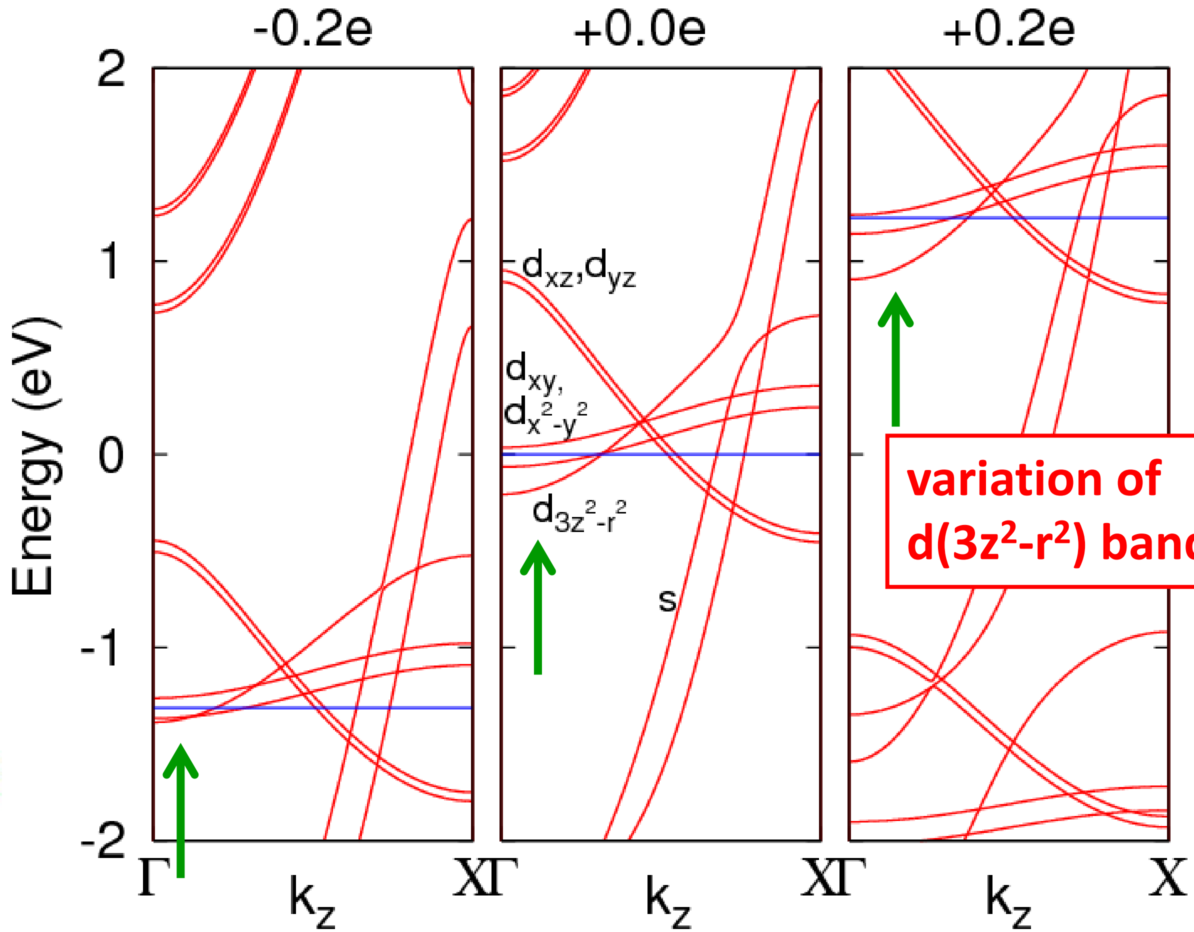
$$E_{mag} = -\mu_0 M_z H_{ext} + \frac{1}{2}\underline{K_u(V)}\left(\frac{M_z}{M_s}\right)^2 + \frac{1}{2}K_{//}\left(\frac{M_x}{M_s}\right)^2$$

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

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







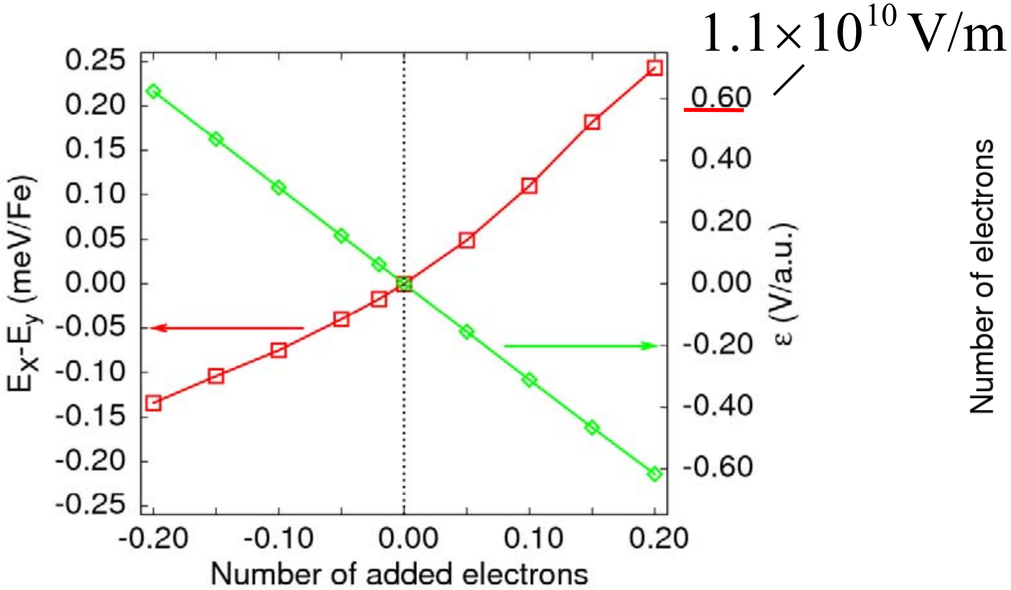
variation of $d(3z^2-r^2)$ band

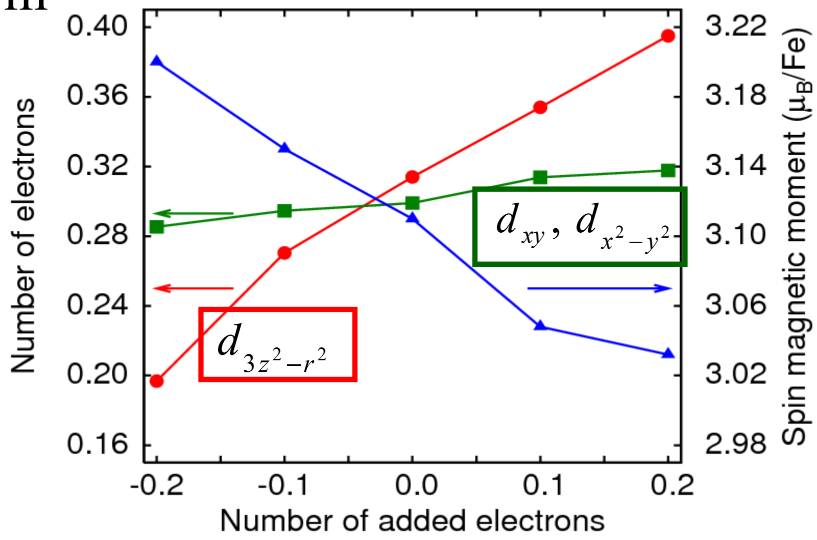
EF dependence depends on the orbital direction

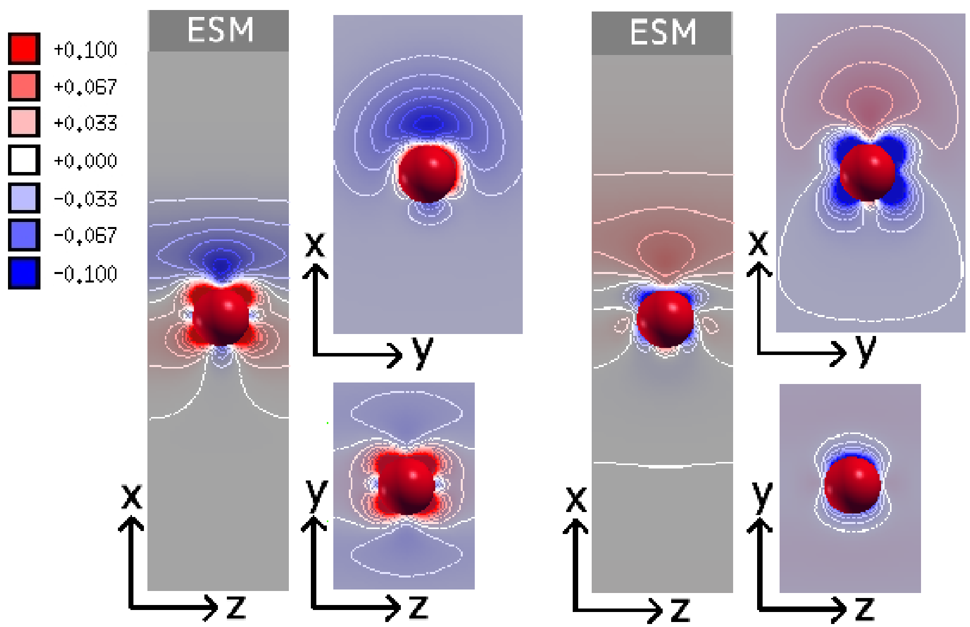
causes variations of MAE

M. Tsujikawa and TO, J. Phys.: Condens. Matter, **21**, 064213 (2009)

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

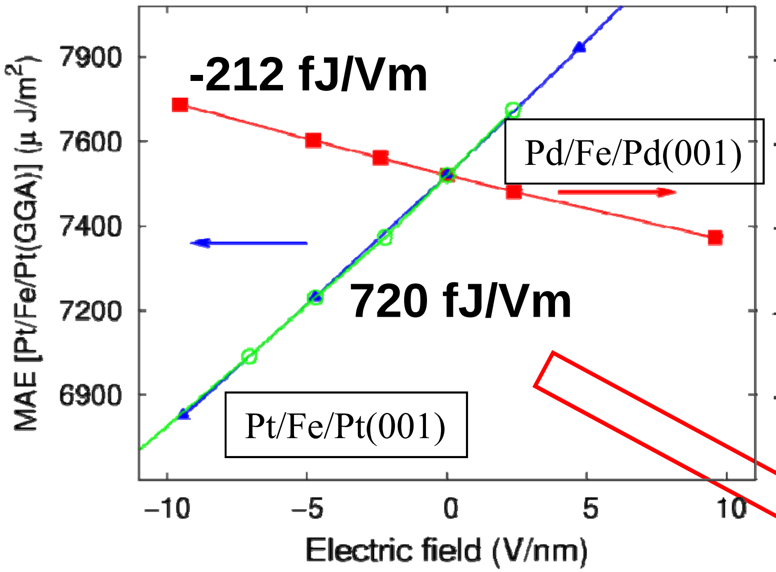




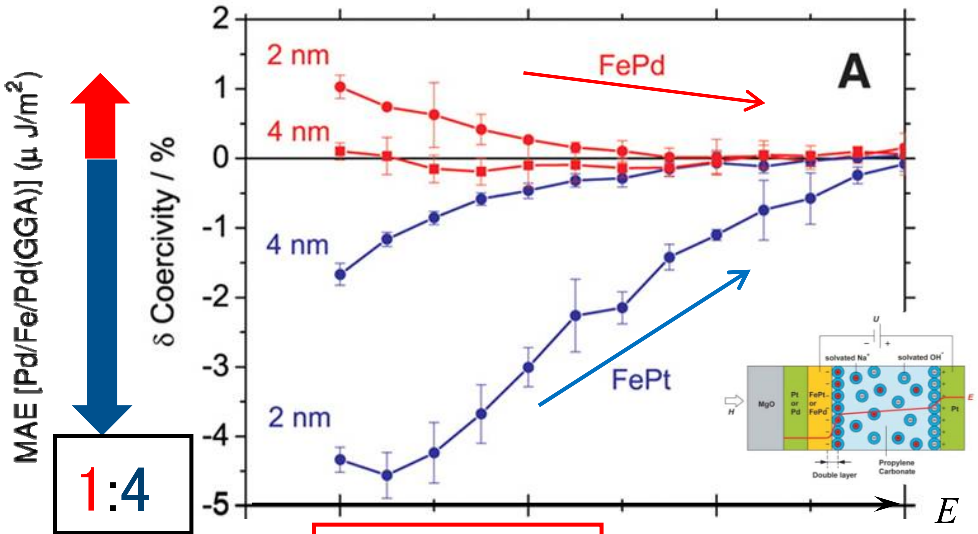


MAE and EF effects(comparison with exp.)

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



M. Weisheit et. al., Science **315**, 349 (2007).



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

- Pd/Fe/Pd(001)
  MAE: -425 μJ/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 $\varepsilon_r \approx 10$ (low frequency limit)

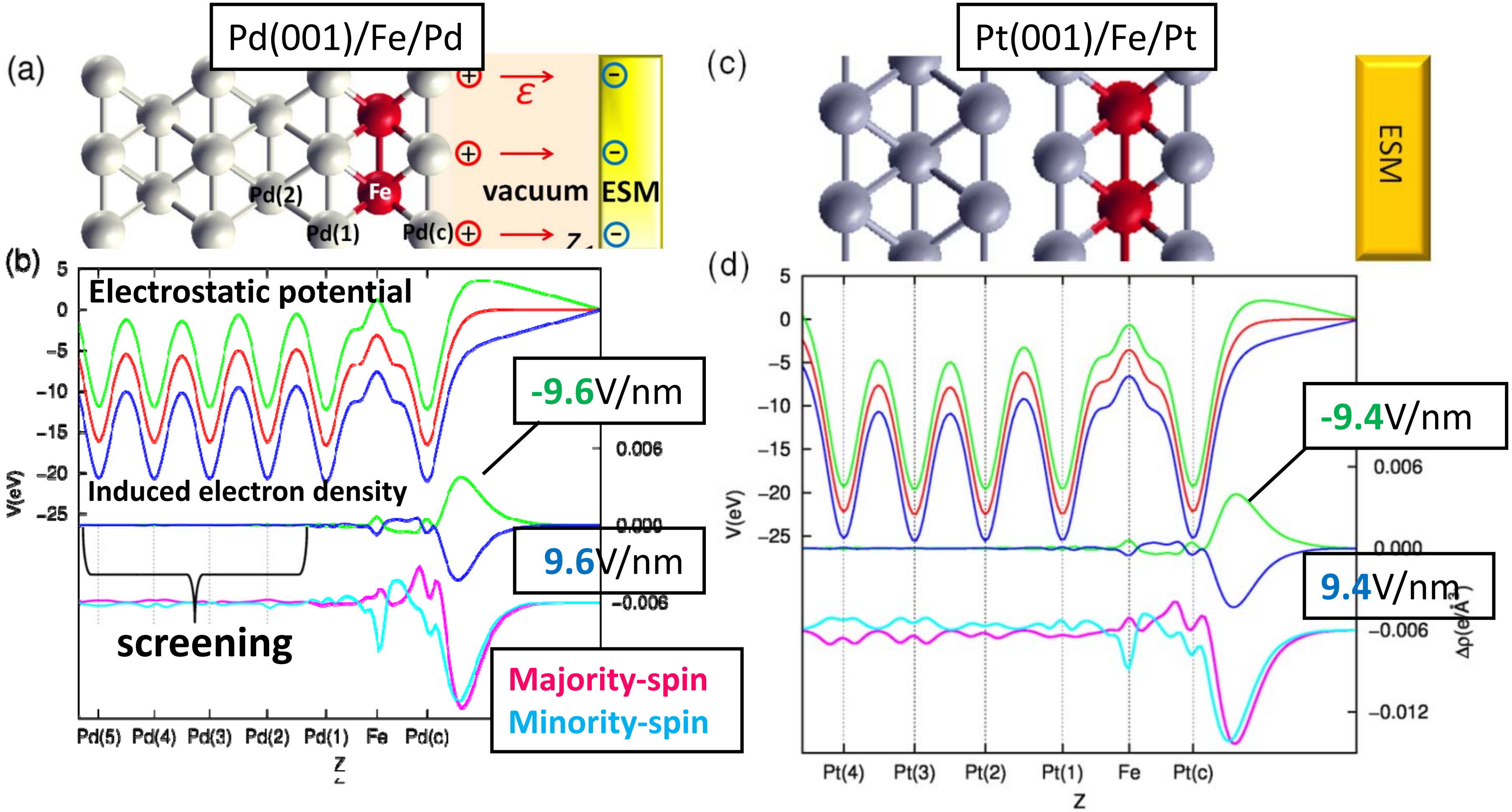
EF-effect=-21.2 × 10 fJ/Vm=-212 fJ/Vm

⇕ **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



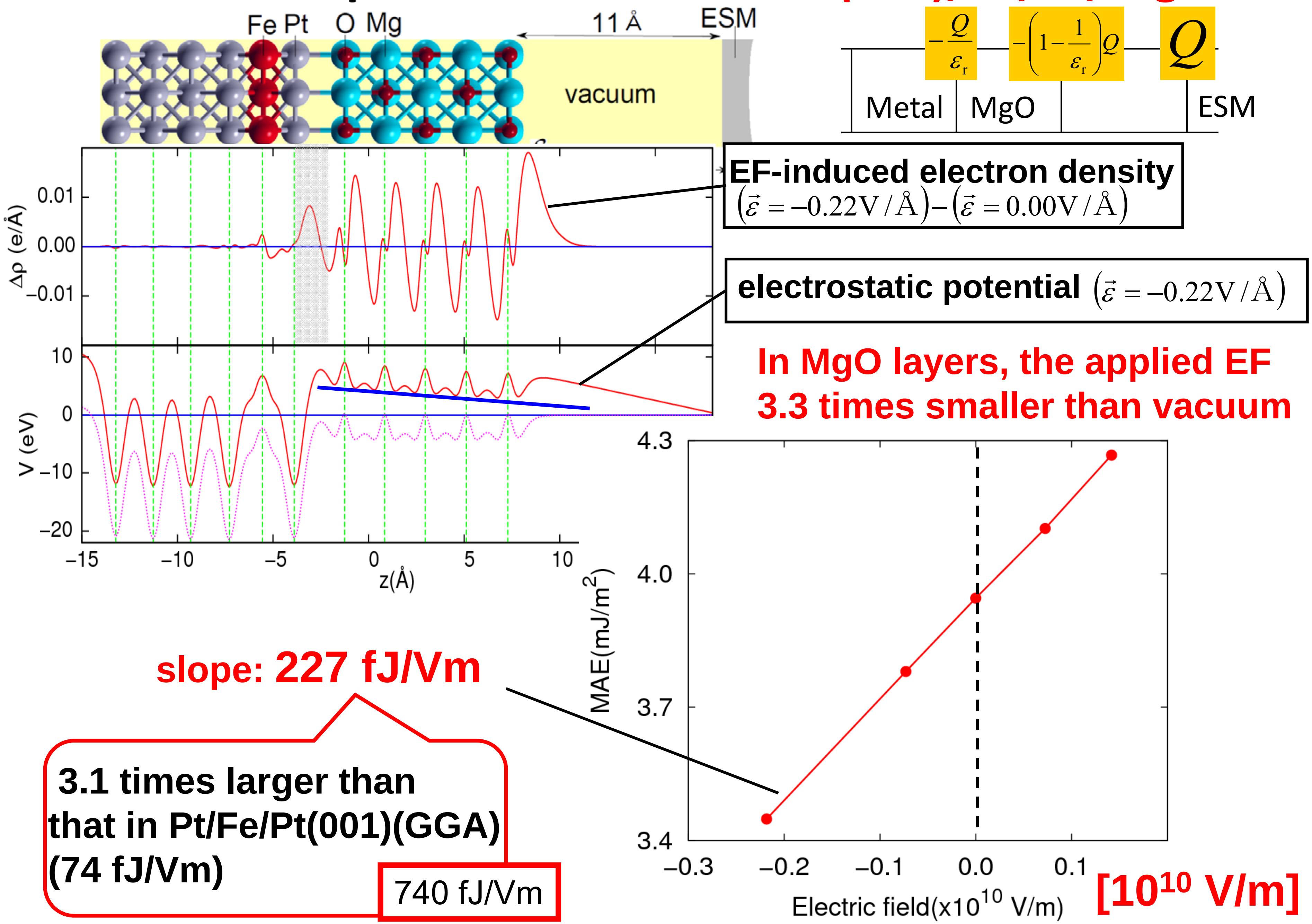
➢The electric field is screened in a few number of surface layers.

➢In these layers, EF effects are induced.

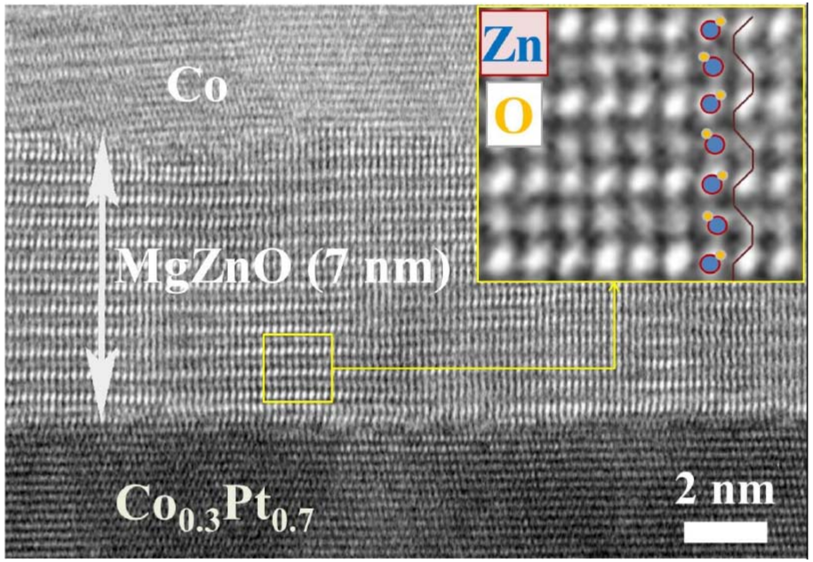
S. Haraguchi et. al., J. Phys. D: Appl. Phys. **44** (2011) 064005.

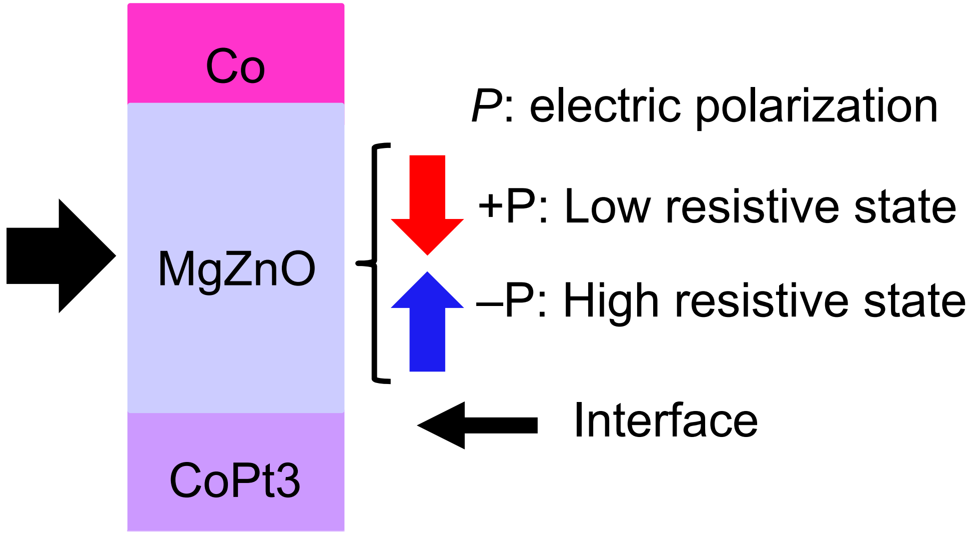
Effective Screening Medium(ESM method): Otani et al., PRB **73**,115407 (2006).

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

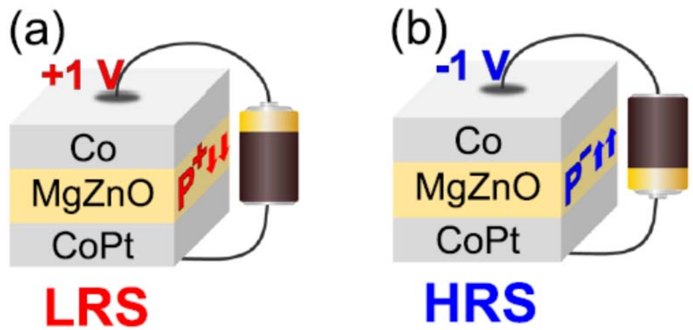


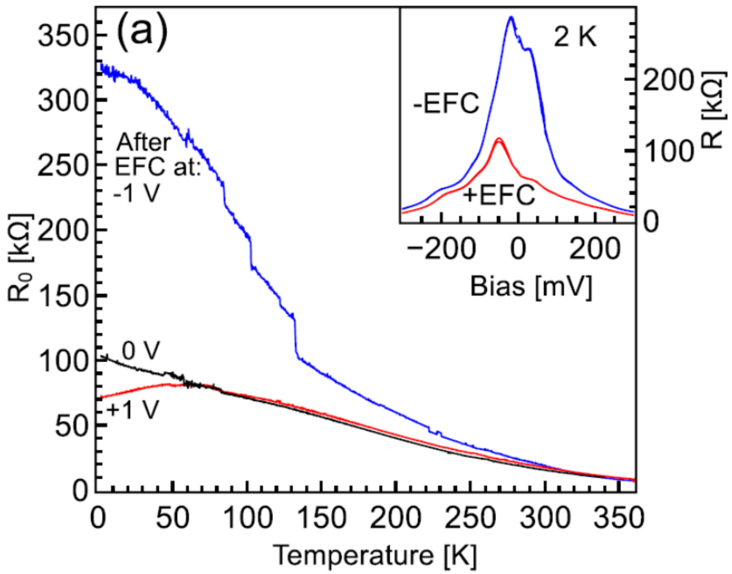
Interface: magnetic metal and dielectric insulator (Exp.)
magnetic anisotropy change by electric polarization switching





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

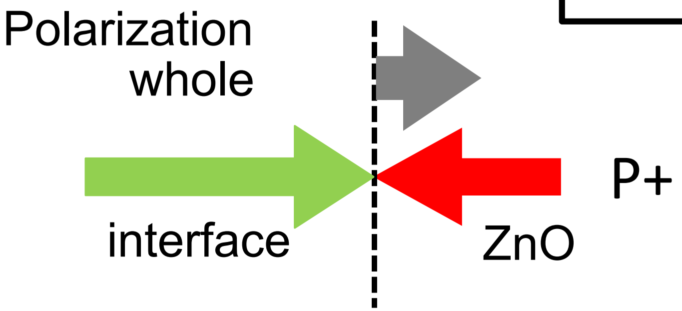




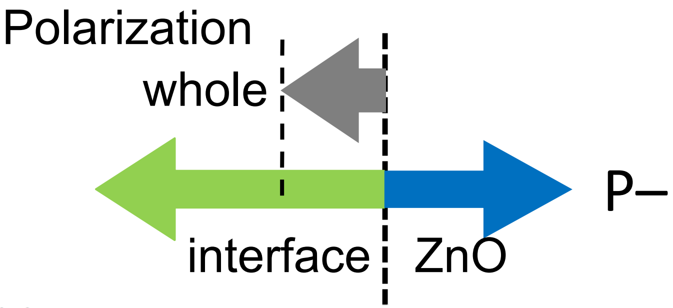
M. Belmoubarik et al., Appl. Phys. Lett.**109**, 054423(2016).

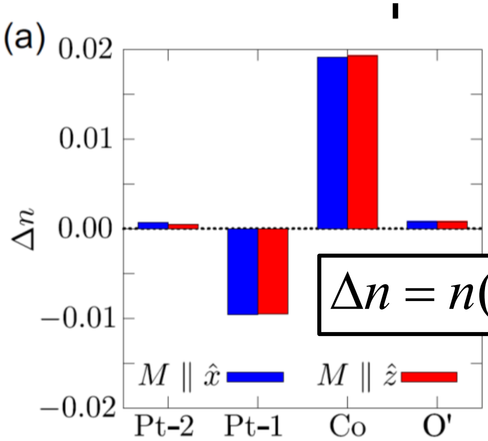
Interface: magnetic metal and dielectric insulator (DFT)

Low resistive state(LRS)



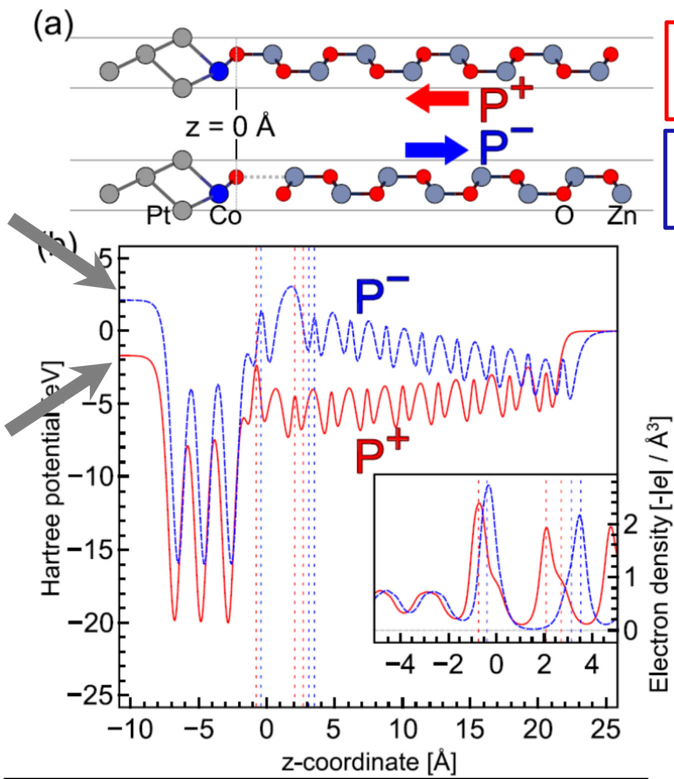
High resistive state(HRS)





$$\Delta n = n(\mathrm{P}-) - n(\mathrm{P}+)$$

PtCoO/(ZnO)n-lcZnO



$$\mathbf{D}(z) = \varepsilon_0 \mathbf{E}(z) + \mathbf{P}(z)$$

MAE(SOI, int)

0.25 meV/cell

$0.44\ \mathrm{mJ/m^2}$

$-2.16\ \mathrm{mJ/m^2}$

-1.23 meV/cell

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

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

$$\frac{d\vec{M}}{dt} = -\gamma\left(\vec{M} \times \vec{H}_{\mathrm{eff}}\right) + \frac{\alpha}{M_s}\vec{M} \times \frac{d\vec{M}}{dt} - \eta(\theta)\frac{\mu_{\mathrm{B}} I}{eV}\frac{\vec{M}}{M_s} \times \left(\frac{\vec{M}}{M_s} \times \frac{\vec{M}_{\mathrm{fix}}}{M_{\mathrm{fix}}}\right)$$

precessional term   dumping term   spin transfer torque

$\vec{M}$:Magnetization vector at free layer

α :Gilbert magnetic damping factor

γ :gyromagnetic factor

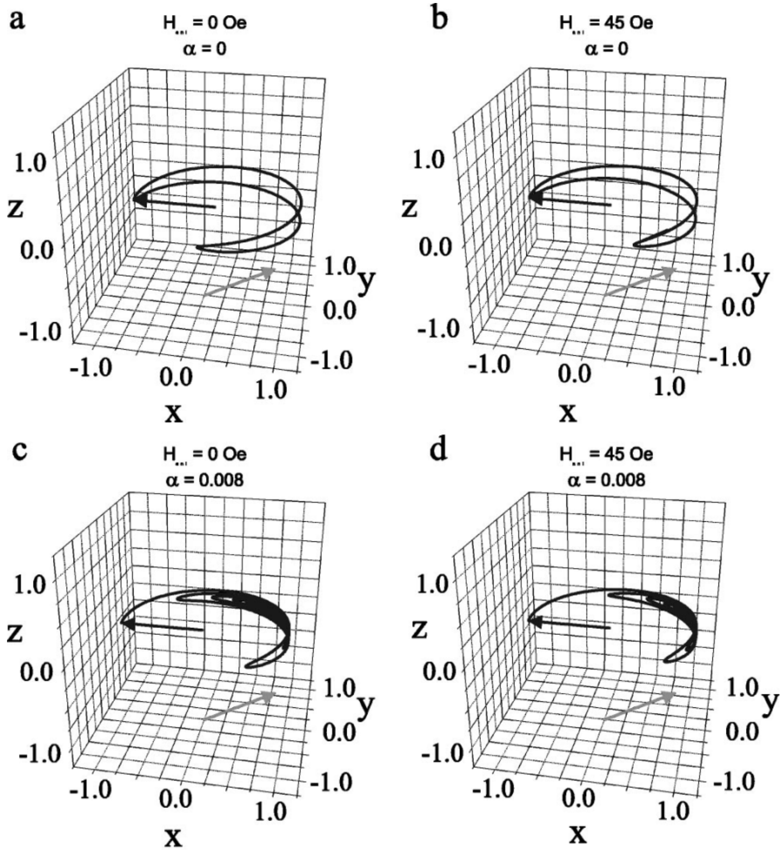
$\vec{M}_{fix}$:Magnetization vector at fixed layer

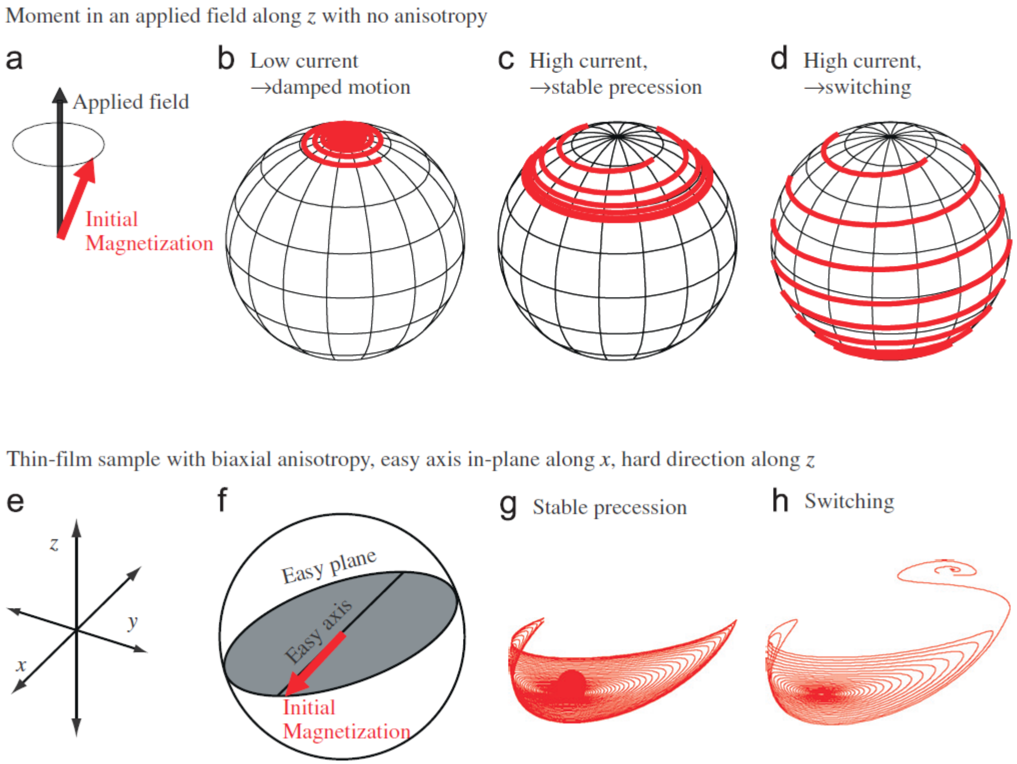
$\eta(\theta)$:spin transfer efficiency

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

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

Dynamics of the magnetization on the free layer





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

α about 0.01

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

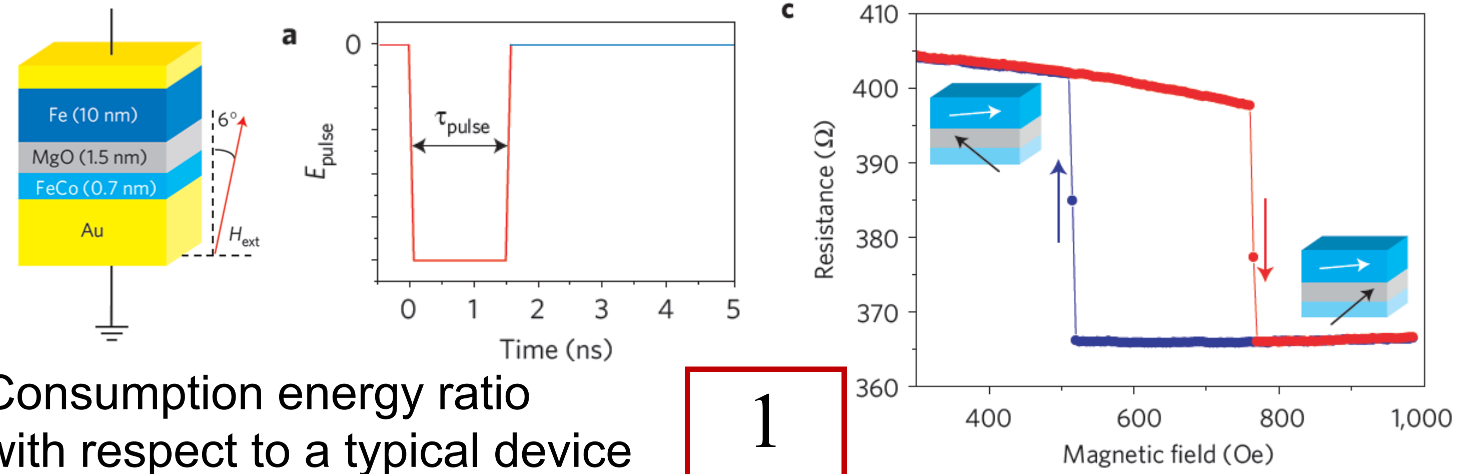
θ : the angle between the directions of spin current and fixed magnetization M_s

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

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

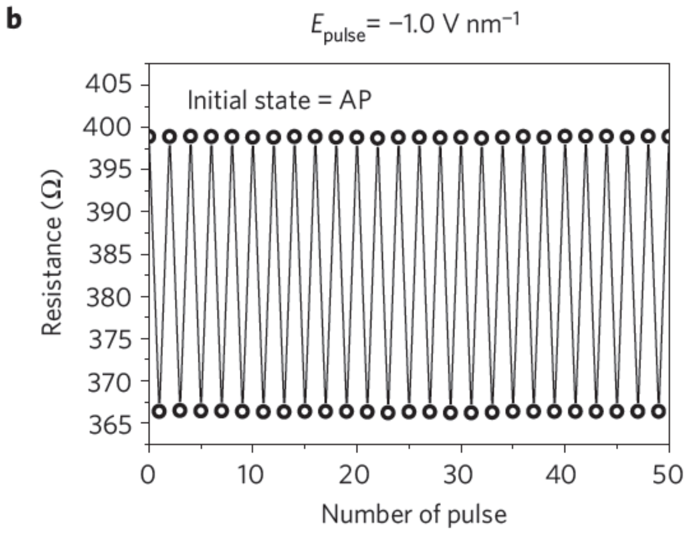
Magnetization switching using voltage pulse

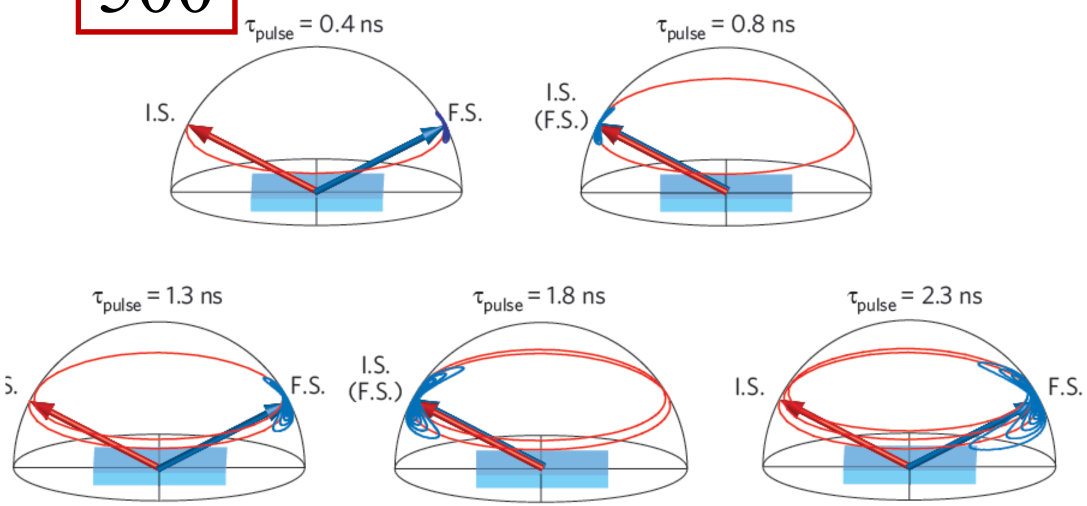
Y. Shiota, T. Nozaki, F. Bonell, S. Murakami, T. Shinio and Y. Suzuki. Nat. Mat.. **11**. 39(2012)



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

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

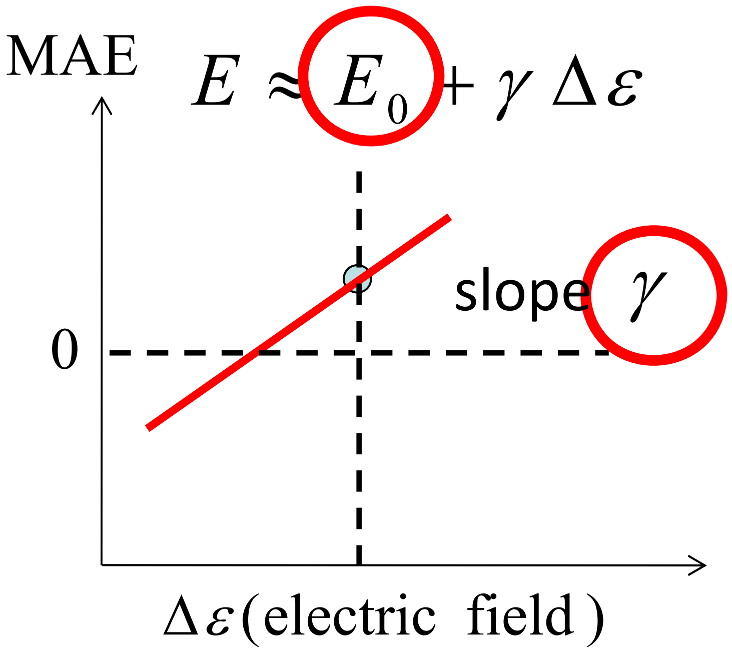


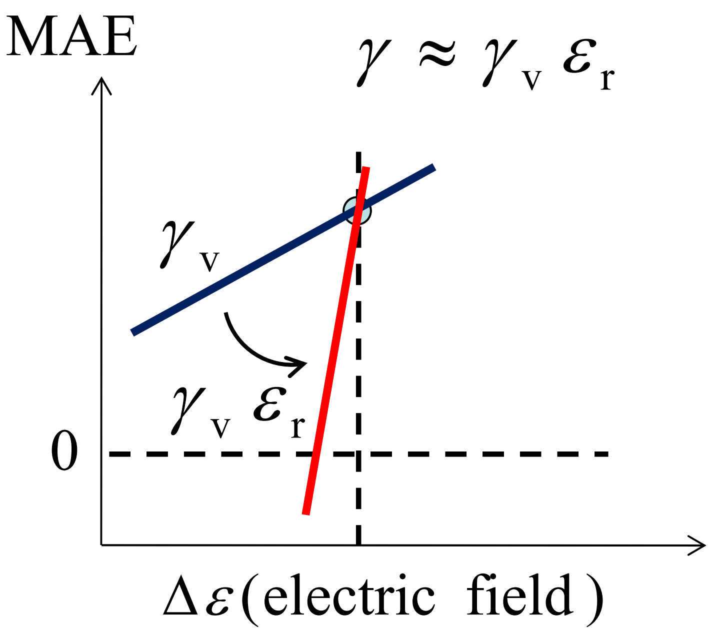


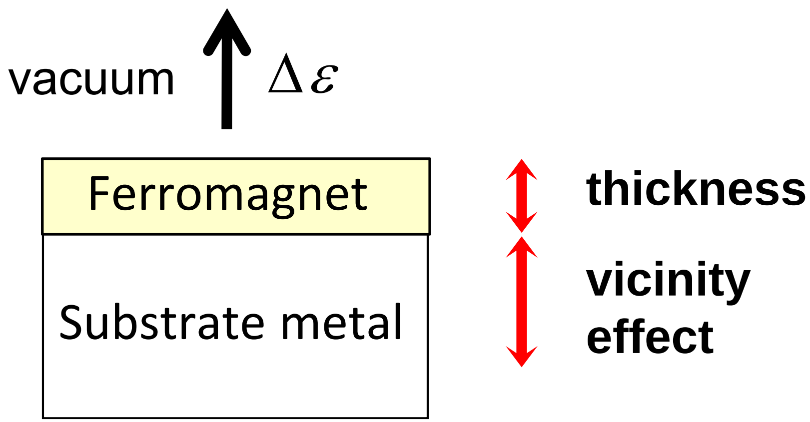
Macro-spin model simulation

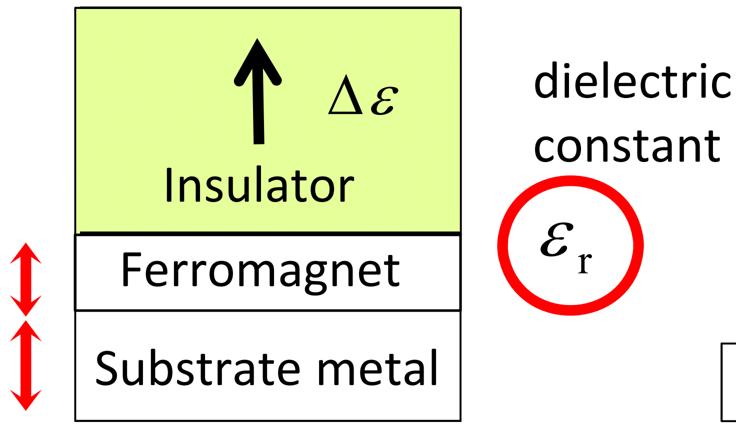
(5-11) Design on magnetic anisotropy in magnetic materials

➢ **Parameters for controlling MAE and its EF 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/Co2FeAl (Huesler alloy)

Mn3Ga　Mn3Ge

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

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

$\left(Fe_n/Ni_m\right)_\ell$

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

Insulating materials: larger dielectric constant : ε_r

insulator/ Fe

$\varepsilon_r(\mathrm{MgO}) = 9.8 \approx 10$

Magnetic interaction with neighboring layers.

Exchange bias between ferro- and antiferr-magnets.

Threshold to magnetic anisotropy transition

Films	E_b (mJ/m²)	E_s (mJ/m²)	E_b+E_s (mJ/m²)	MAE slope γ (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

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

$$\boxed{\varepsilon_c = -\left(E_b + E_s\right)/\,\gamma}$$

M. Tsujikawa et al., JAP, **111**, 083910 (2012).

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 effcts
(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.B**73**,115407,2006)

Energy functional

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

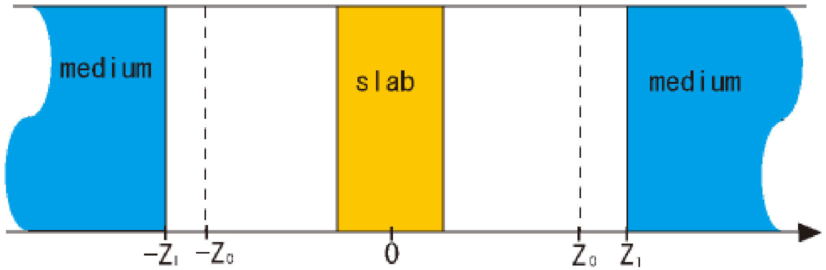
variation with the to static potential

variation with the orbital

Poisson equation

$$\nabla\cdot[\varepsilon(\vec{r})\nabla]V(\vec{r})=-4\pi n_{tot}(\vec{r})$$

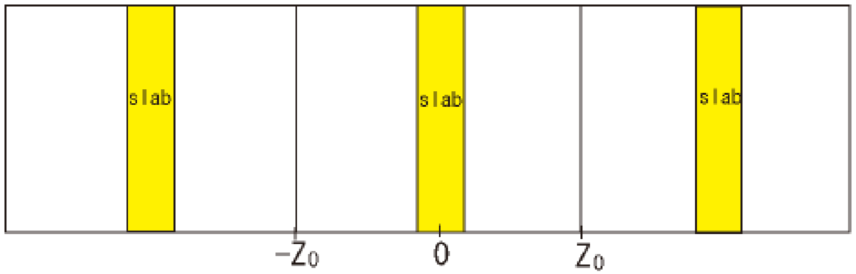
$$\nabla\cdot[\varepsilon(\vec{r})\nabla]G(\vec{r})=-4\pi\delta(\vec{r}-\vec{r}')$$



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

Kohn-Sham equation

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



periodic boundary condition

Total energy representation by the Green's function

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

$$+\iint d\vec{r}d\vec{r}'n_e(\vec{r})G(\vec{r},\vec{r}')n_I(\vec{r}')+\frac{1}{2}\iint n_I(\vec{r})G(\vec{r},\vec{r}')n_I(\vec{r}')$$

A1-2.
Usual first-principles approach

Electrostatic potential (solution of Poisson's equation) ;

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

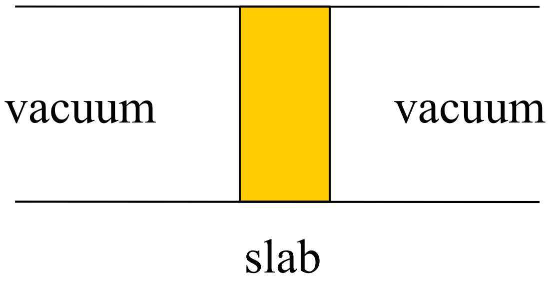
$$G(\vec{r},\vec{r}') = \frac{1}{|\vec{r}-\vec{r}'|}$$

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

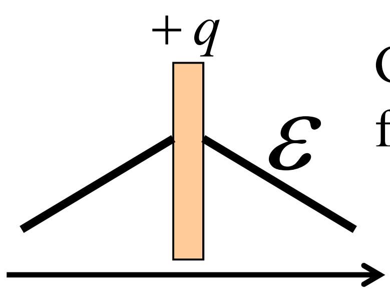


$$G(\vec{g}_{\|},z,z') = \frac{4\pi}{2g_{\|}} e^{-g_{\|}|z-z'|}$$

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

For small $g_{\|}$

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



Green's function for a charged sheet

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

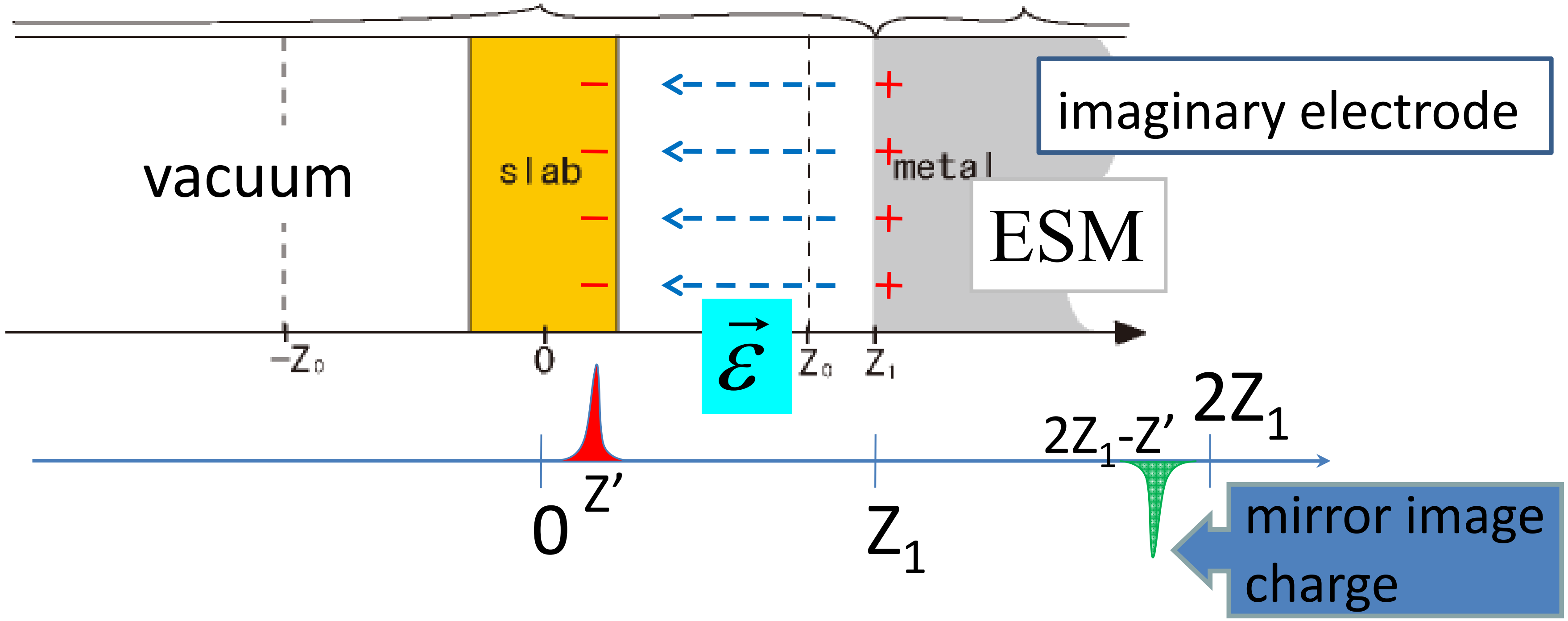
(M.Otani and O.Sugino,Phys.Rev.B73,115407,2006)

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

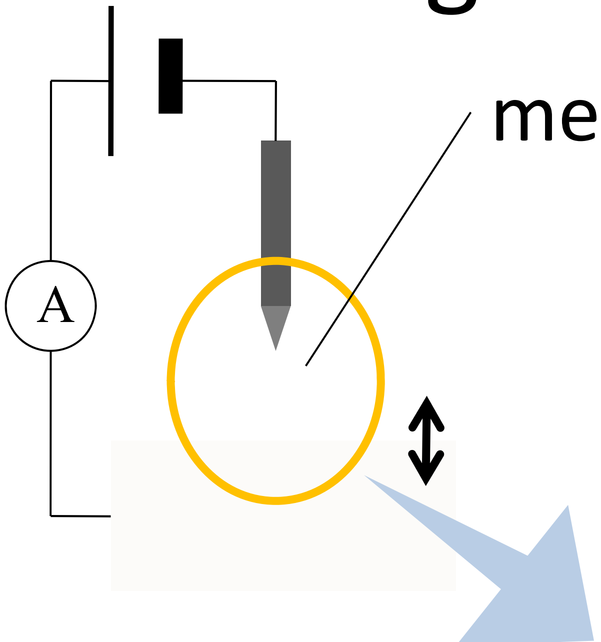
boundary condition $\begin{cases} V_H(\mathbf{g}_\parallel, z)\big|_{z=z_1} = 0 \\ \dfrac{\partial}{\partial z} V_H(\mathbf{g}_\parallel, z)\big|_{z=-\infty} = 0 \end{cases}$ $\varepsilon(z) = \begin{cases} 1 \text{ if } z \le z_1 \\ \infty \text{ if } z \ge z_1 \end{cases}$

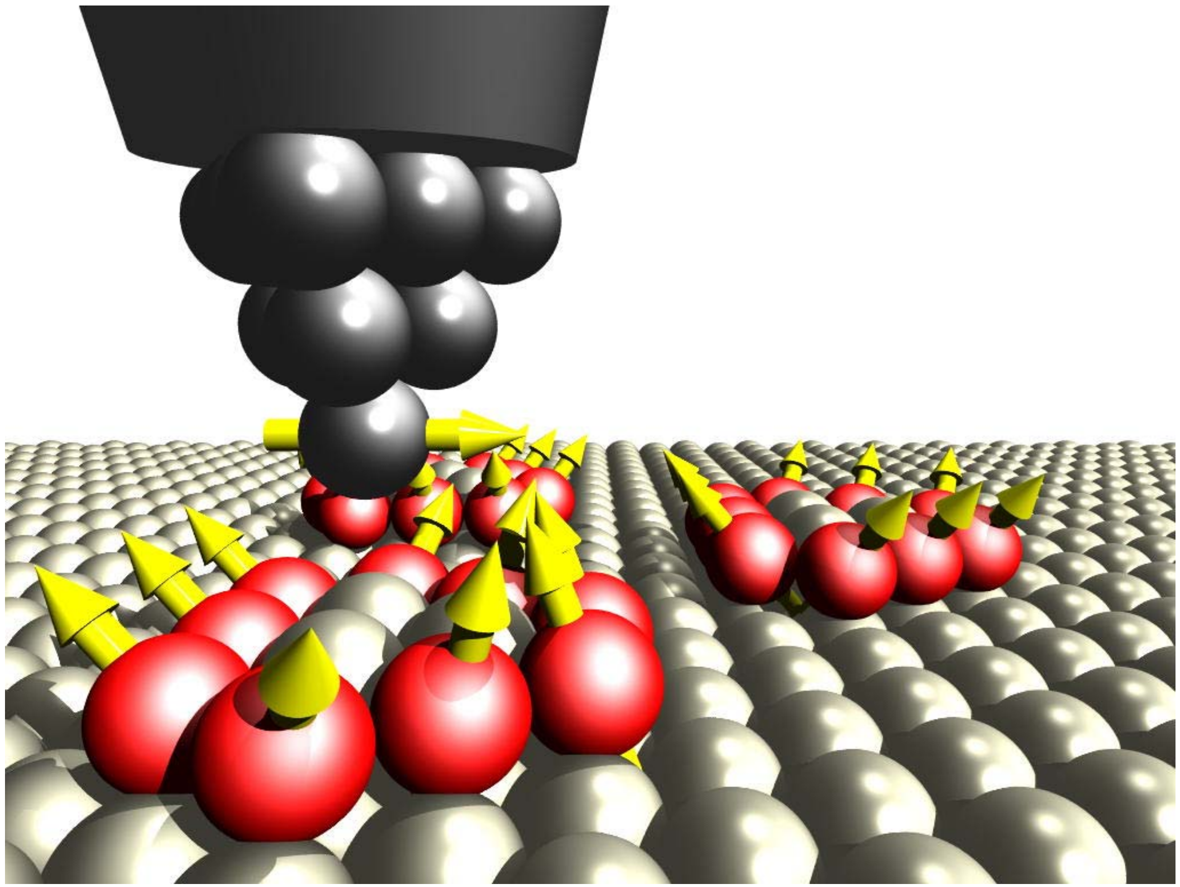
$$G(\mathbf{g}_\parallel, z, z') = \frac{4\pi}{2g_\parallel} e^{-g_\parallel |z-z'|} \boxed{- \frac{4\pi}{2g_\parallel} e^{-g_\parallel (2z_1 - z - z')}}$$

the contribution from mirror image charge



A1-4. Electric filed induced in scanning tunnel microscope (STM)





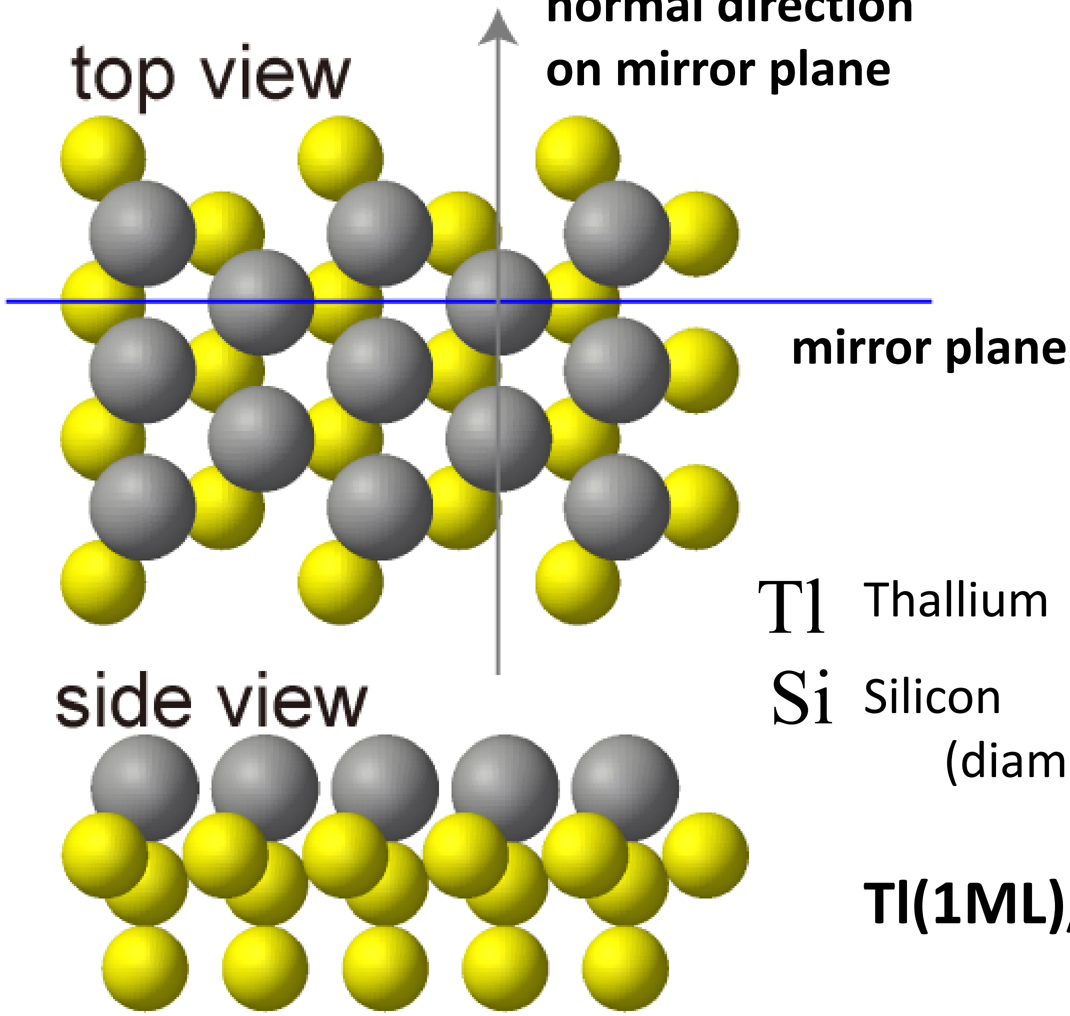
Scale of electric field

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

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

Tl/Si(111)-1×1 surface

C_{3V}



Tl Thallium (adsorption atom)

Si Silicon
(diamond structure)

Tl(1ML)/Si(24ML)/H(1ML)

Sakamoto, Oda, Kimura *et al*., Phys.Rev. Lett., 102 (2009) 096805.

Full spin-orbit interaction other than the Rashba term

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

$$\left(\varphi_{\vec{k}}\middle|H_{\mathrm{SOI}}\middle|\varphi_{\vec{k}}\right)$$

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

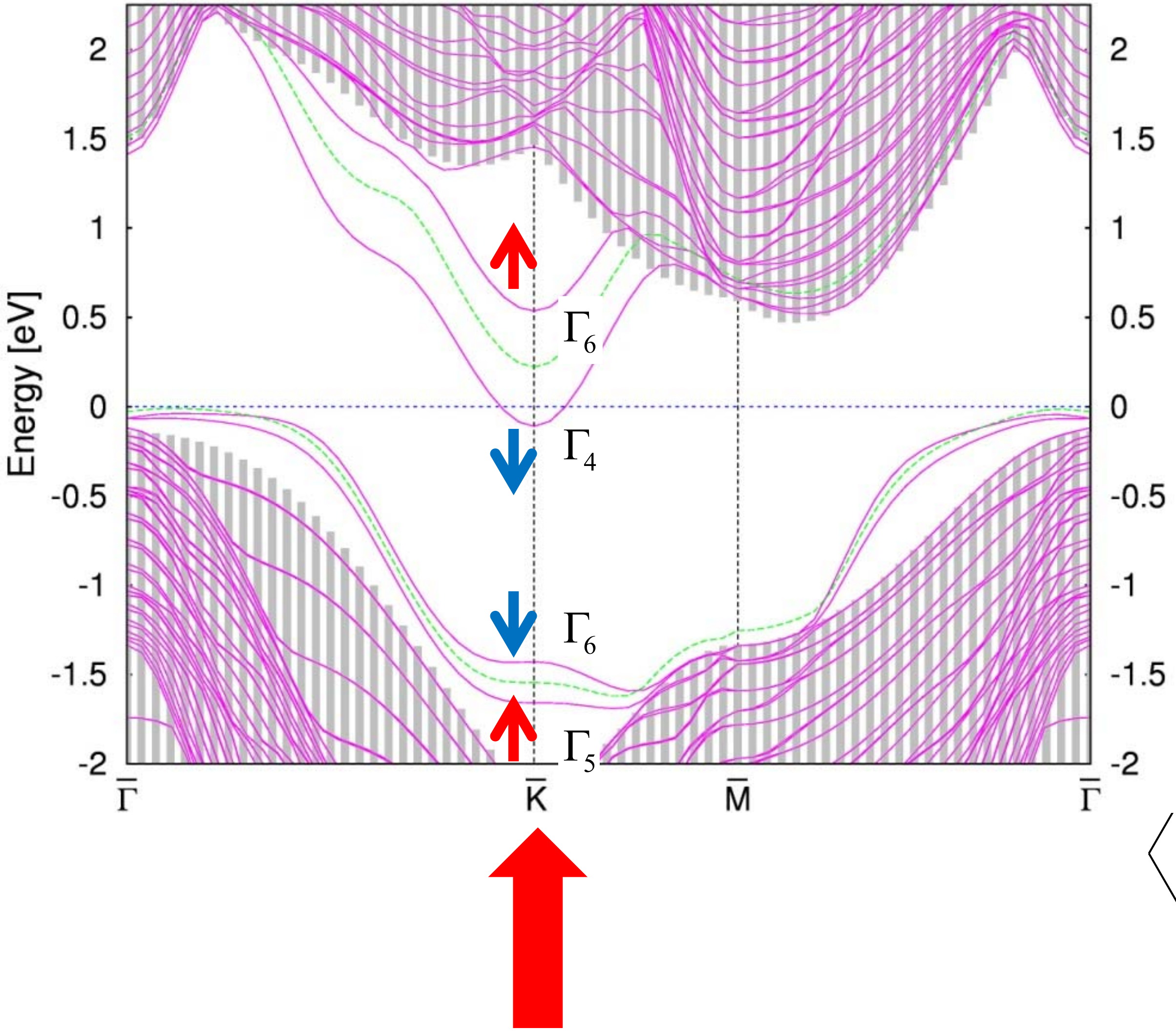
$$\varphi_{n\vec{k}}(\vec{r}) = \frac{1}{\sqrt{\Omega}}\exp(i\vec{k}\cdot\vec{r})\underline{u_{n\vec{k}}(\vec{r})}$$

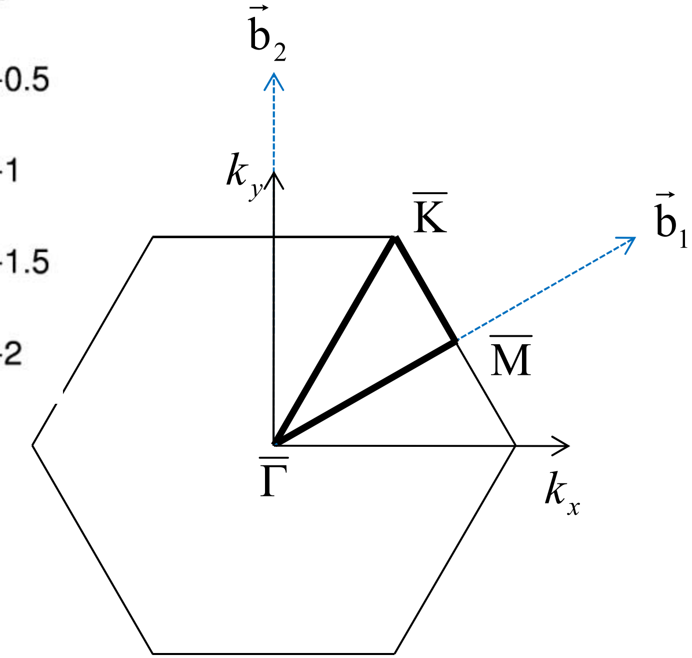
$$\vec{\alpha}_n = \frac{\hbar^2 N}{4m_e^2c^2\Omega}\int_{cell} d\vec{r}\left|u_{n\vec{k}}(\vec{r})\right|^2\vec{\nabla}V(\vec{r}) \longrightarrow \vec{\alpha}_n = \alpha\,\vec{e}_z$$

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

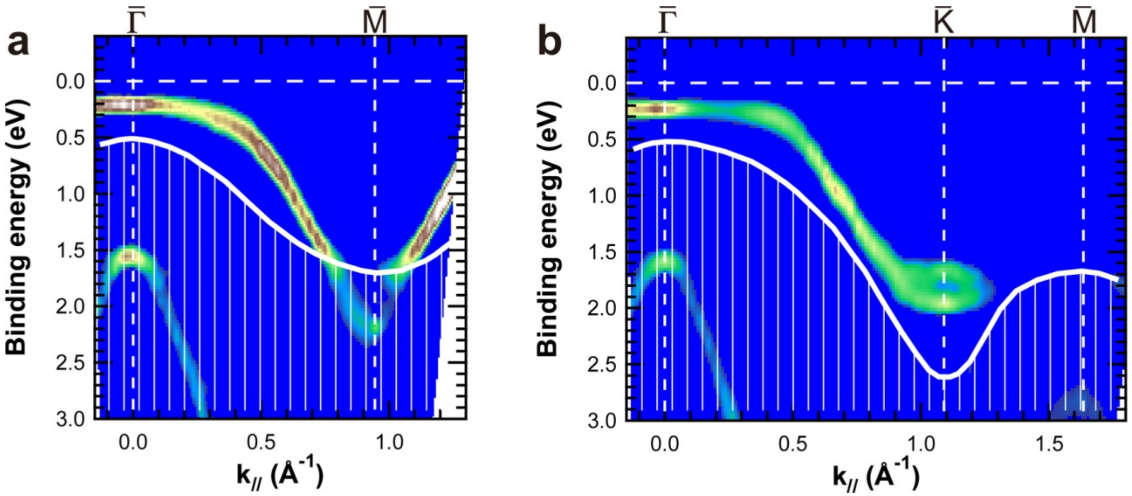
Sakamoto, Oda, Kimura *et al*., Phys.Rev. Lett., 102 (2009) 096805.

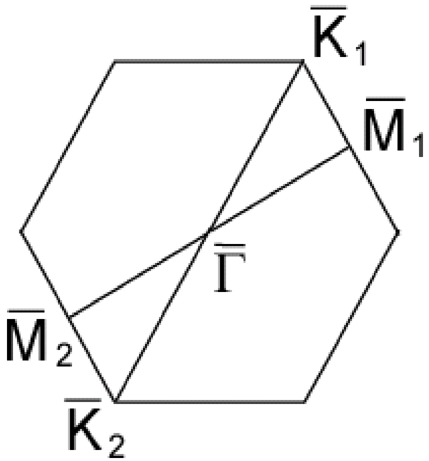
Surface band structure in Tl/Si(111)



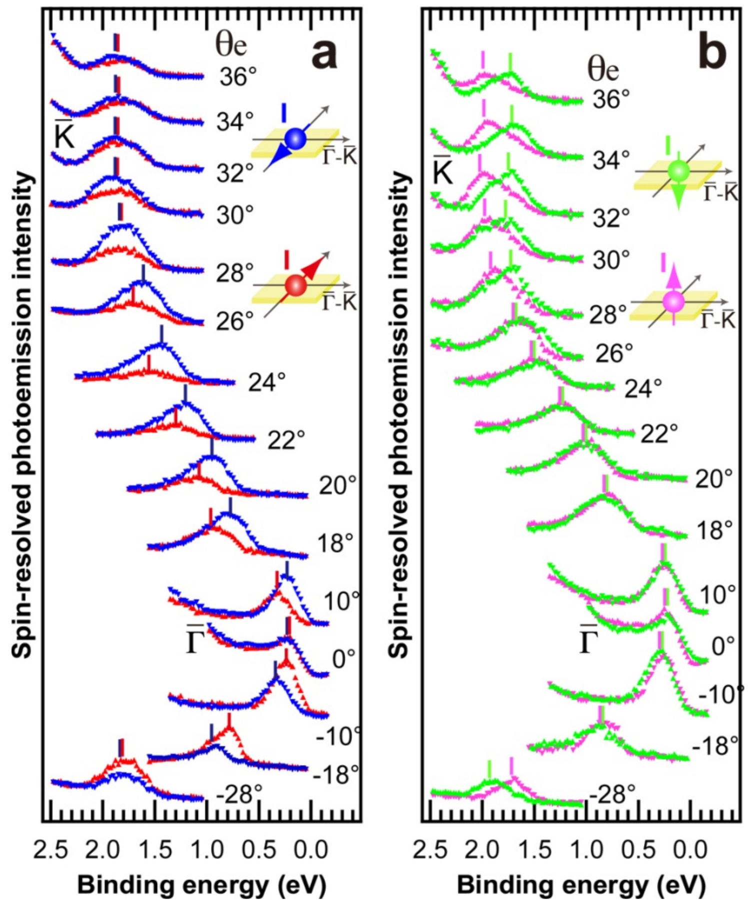


Spin splitting of Tl/Si（1 1 1）1 × 1 surface band





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

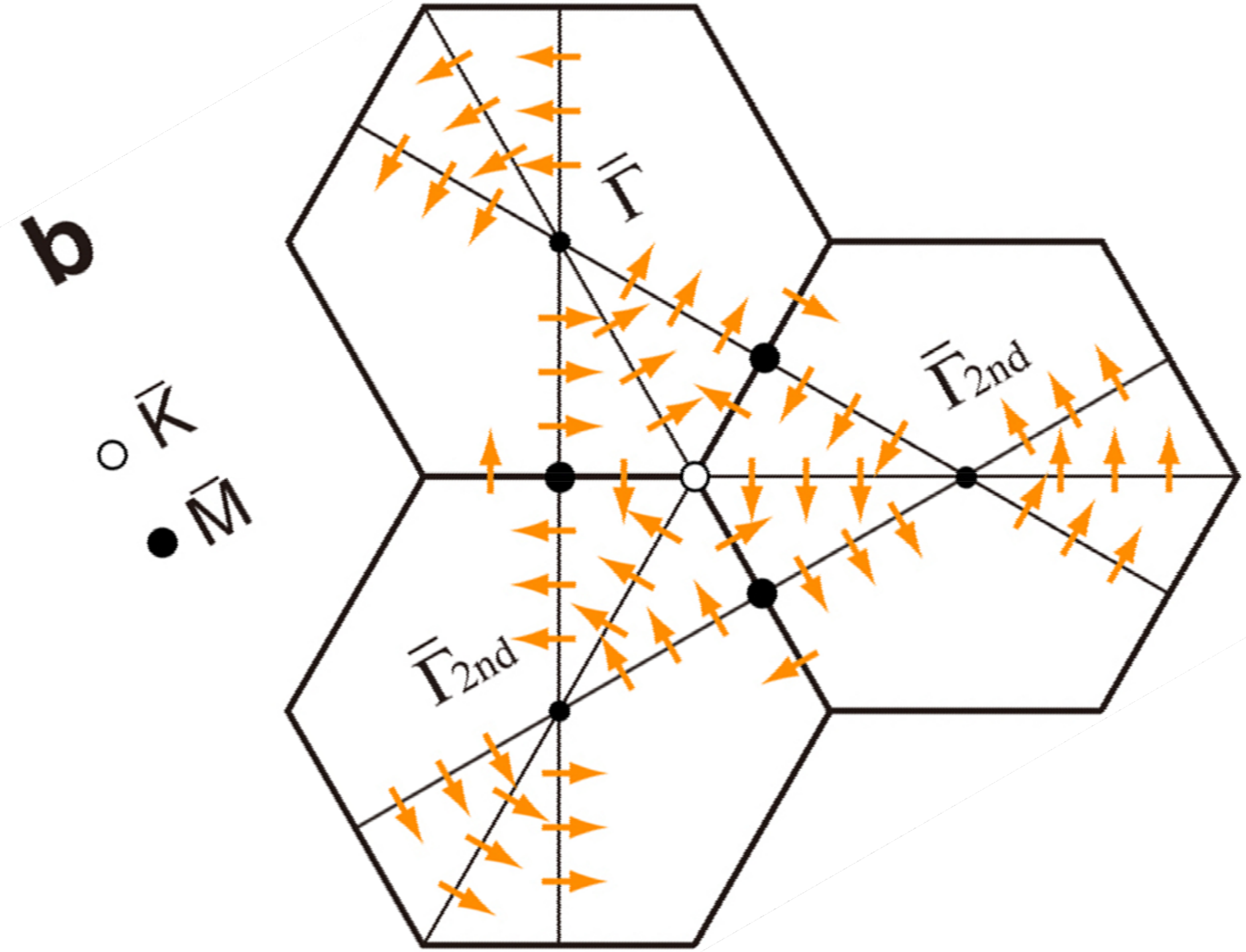


Spin splitting along Rashba direction

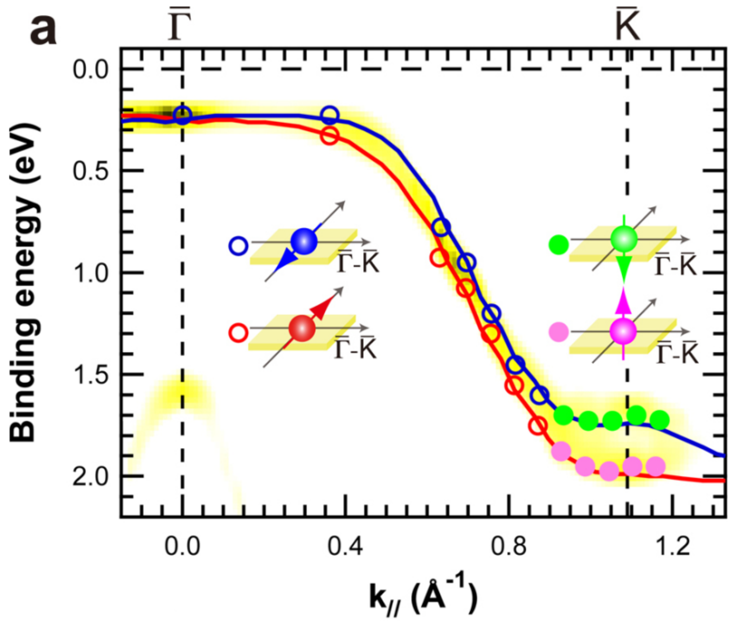
Normal to Rashba direction

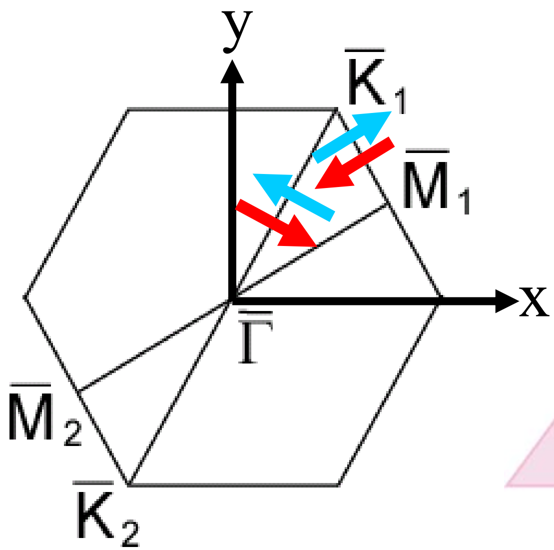
Sakamoto, Oda, Kimura *et al*., Phys.Rev. Lett., 102 (2009) 096805.

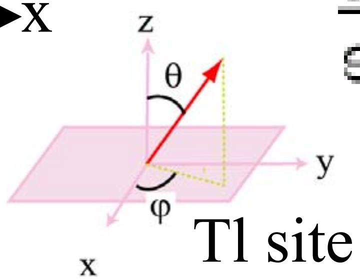
Wave vector dependence of spin direction : Voltical spin polarization

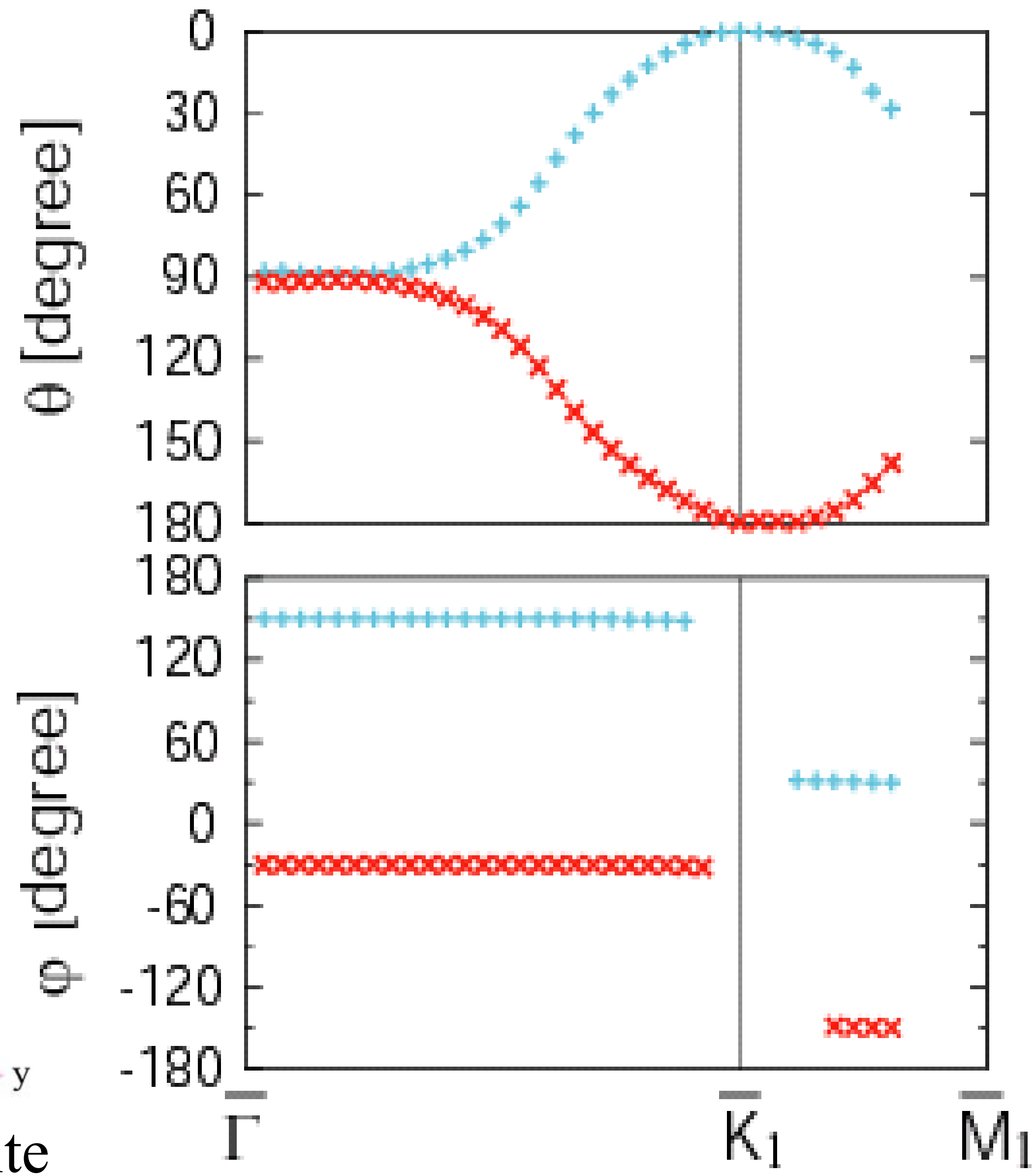


Wave vector dependence of electron spin direction : Tl site









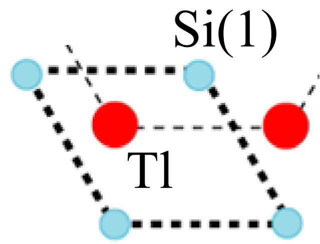
Atomic orbital components in the eigenstates at $\overline{\mathrm{K}}$

$\overline{\mathrm{K}}$ point	$\langle \ell_z \rangle_{\mathrm{Tl}}$	Tl(p_x,p_y)	(p_z)	Si(1)(p_x,p_y)	(p_z)	Si(2)(p_x,p_y)	(p_z)
Γ_6 -1.53eV	-0.047	↓0.060	---	---	↓0.105	↓0.028	---
Γ_5 -1.73eV	-0.068	↑0.090	↓0.002	---	↑0.090	↑0.024	↓0.002

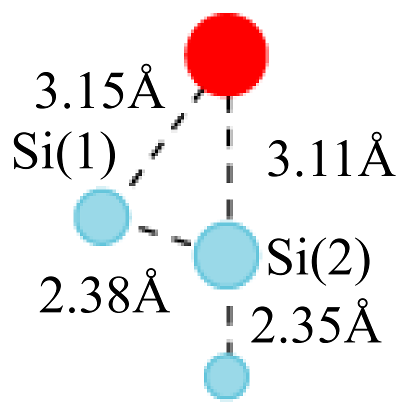
C_3 double group $\langle \ell_z \rangle_{\mathrm{Si(2)}} = -0.023$

Good quantum state of ℓ_z

$\boxed{\mathrm{Tl} : p_x - \mathrm{i}\, p_y}$



T_4 site



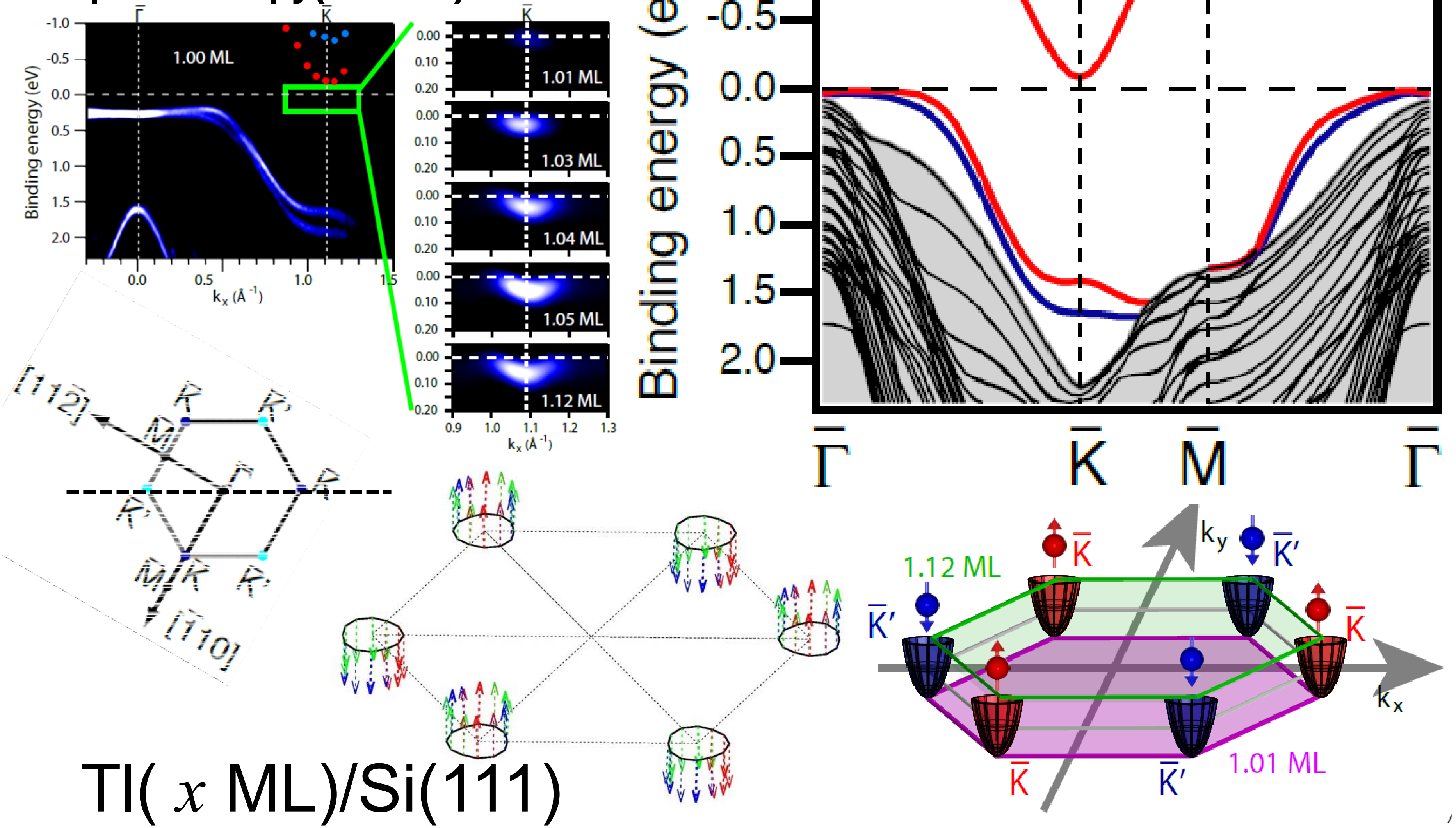
basis function of atomic orbital

	Tl site, Si(2)	Si(1) site
Γ_6 :	$\{(x+iy)\alpha,\ (x-iy)\beta\}$	$\{(x-iy)\alpha,\ z\beta\}$
Γ_5 :	$\{(x-iy)\alpha,\ z\beta\}$	$\{z\alpha,\ (x+iy)\beta\}$
Γ_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)

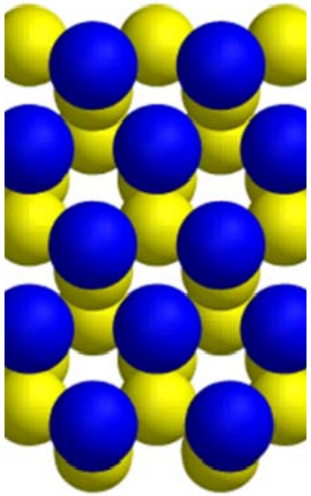


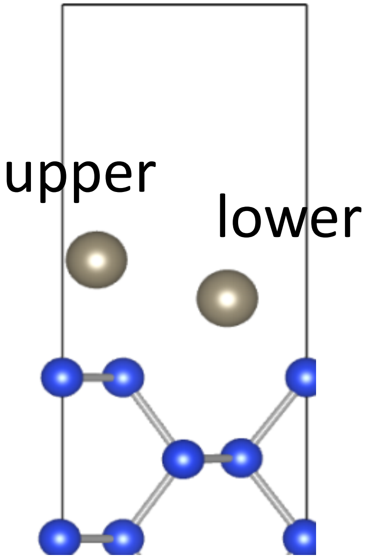
Tl(x ML)/Si(111)

K. Sakamoto et al., Nat. Commun., 4, 2073, doi: 10.1038/ncomms3073, (2013)

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

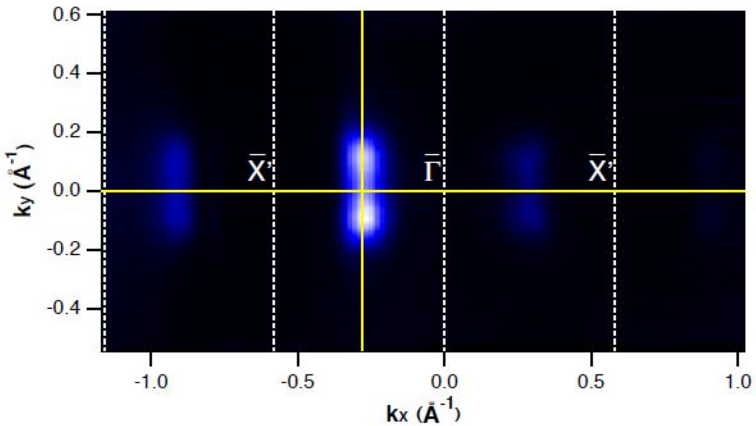
Structure



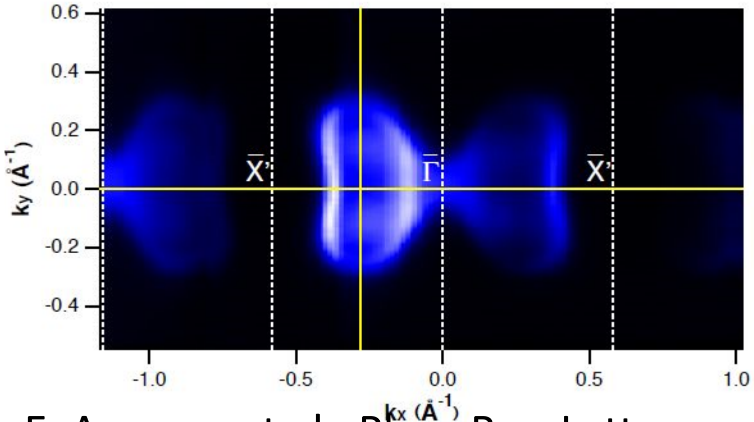


ARPES

Eκ=35.96 eV

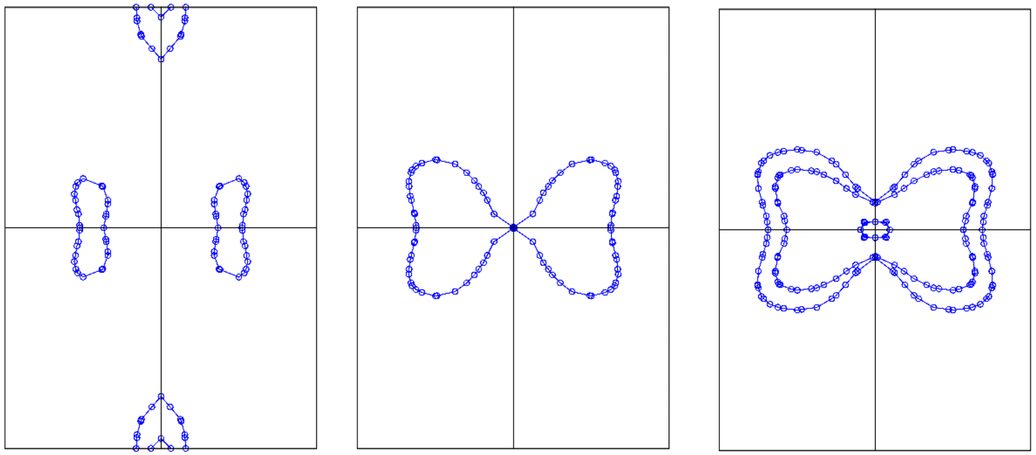


Eκ=35.78 eV



E-E_F

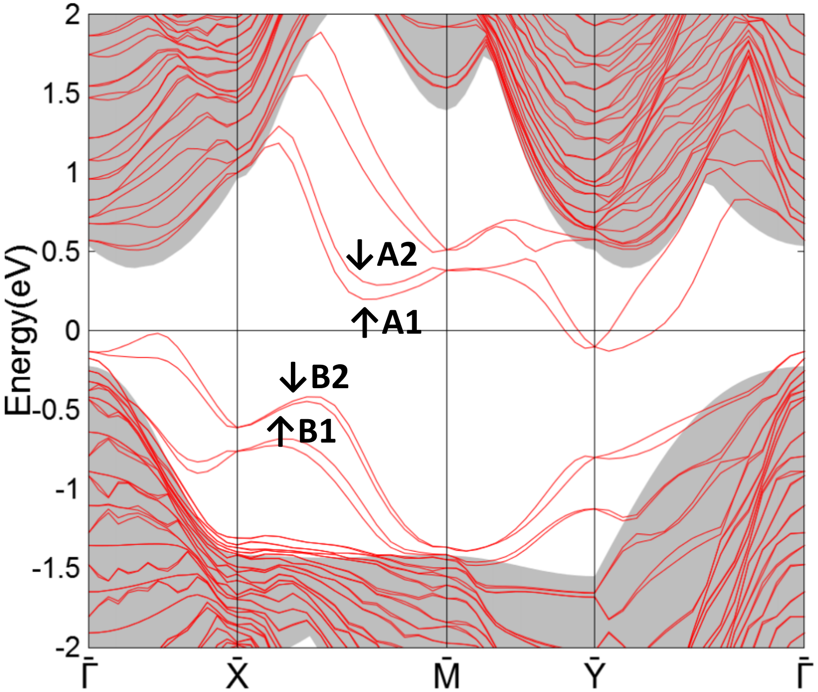
-0.04 eV **-0.13 eV** **-0.22 eV**

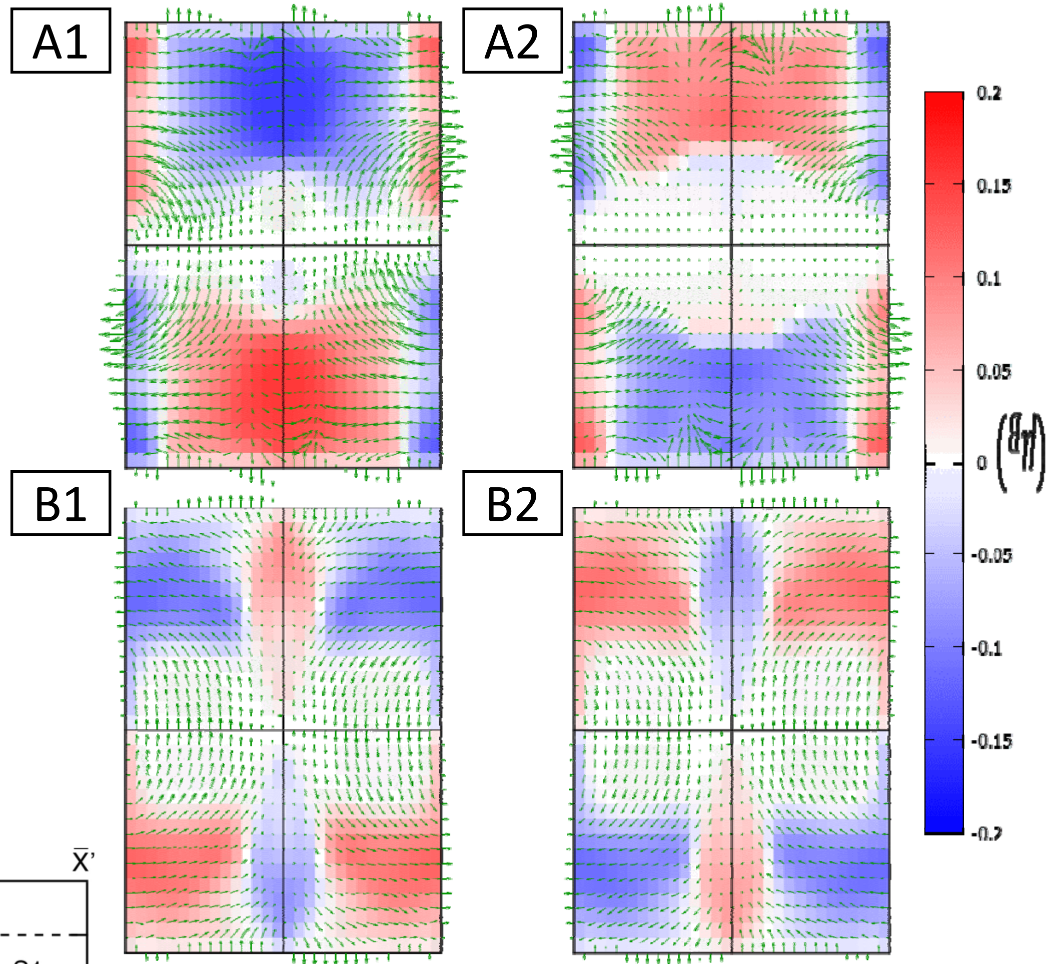


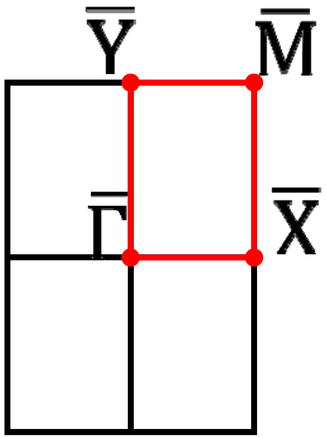
E. Annese, et al., Phys. Rev. Lett., 117, 16803(2016)

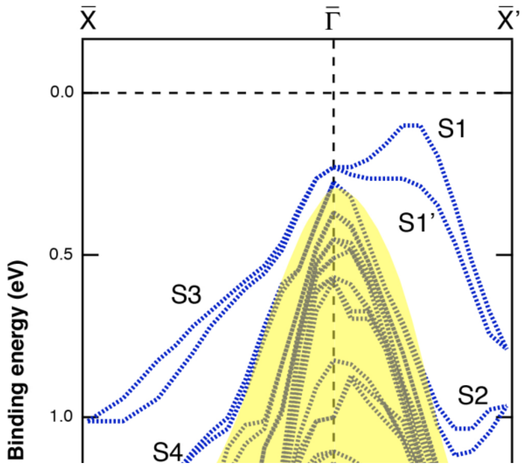
Band dispersion and Spin texture

Tl/Si(110)









- The electron spins at

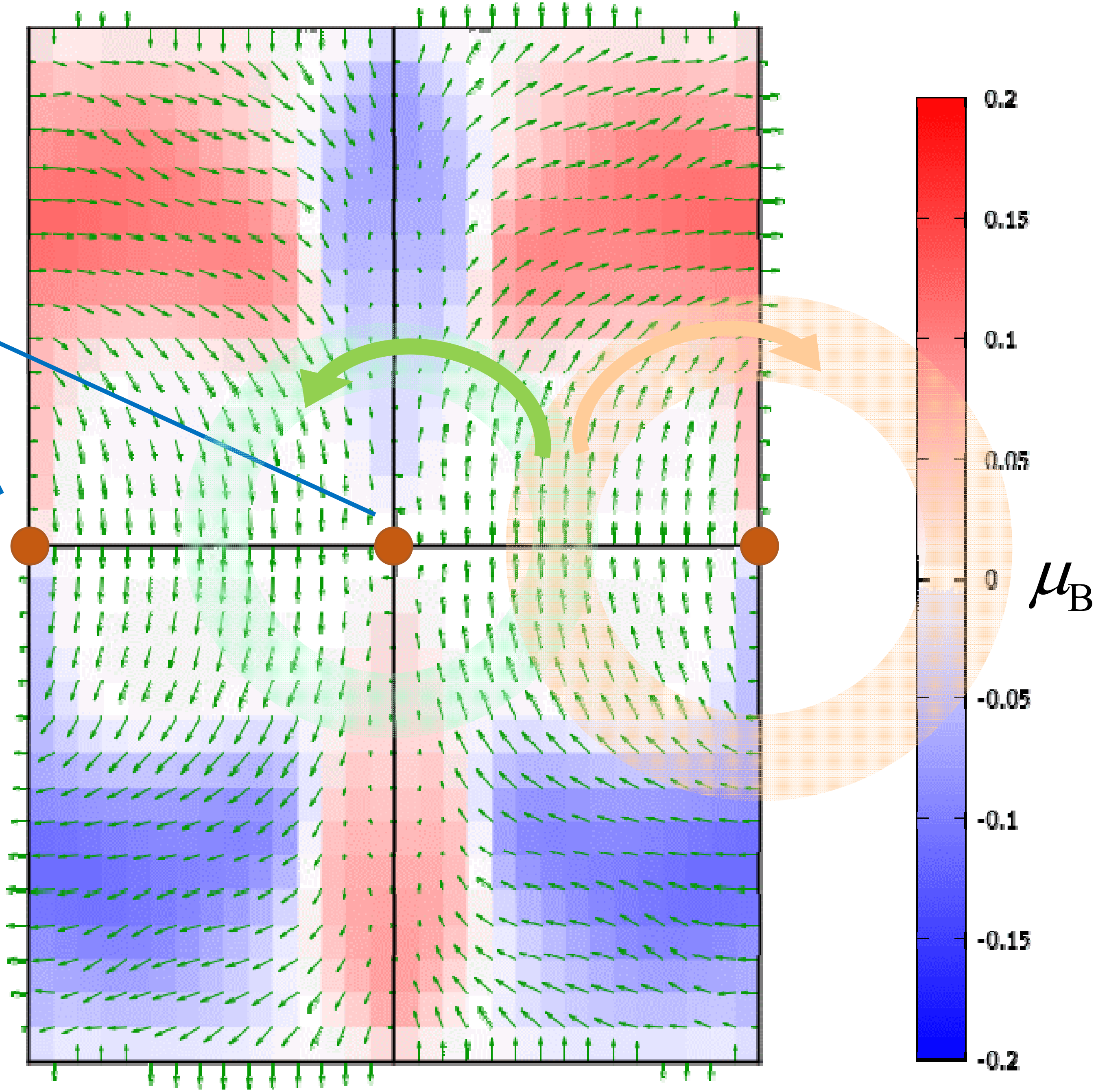
B2

$$\mathbf{K} = \alpha_{\mathbf{K}}\mathbf{K} + \mathbf{G}$$

Vortical

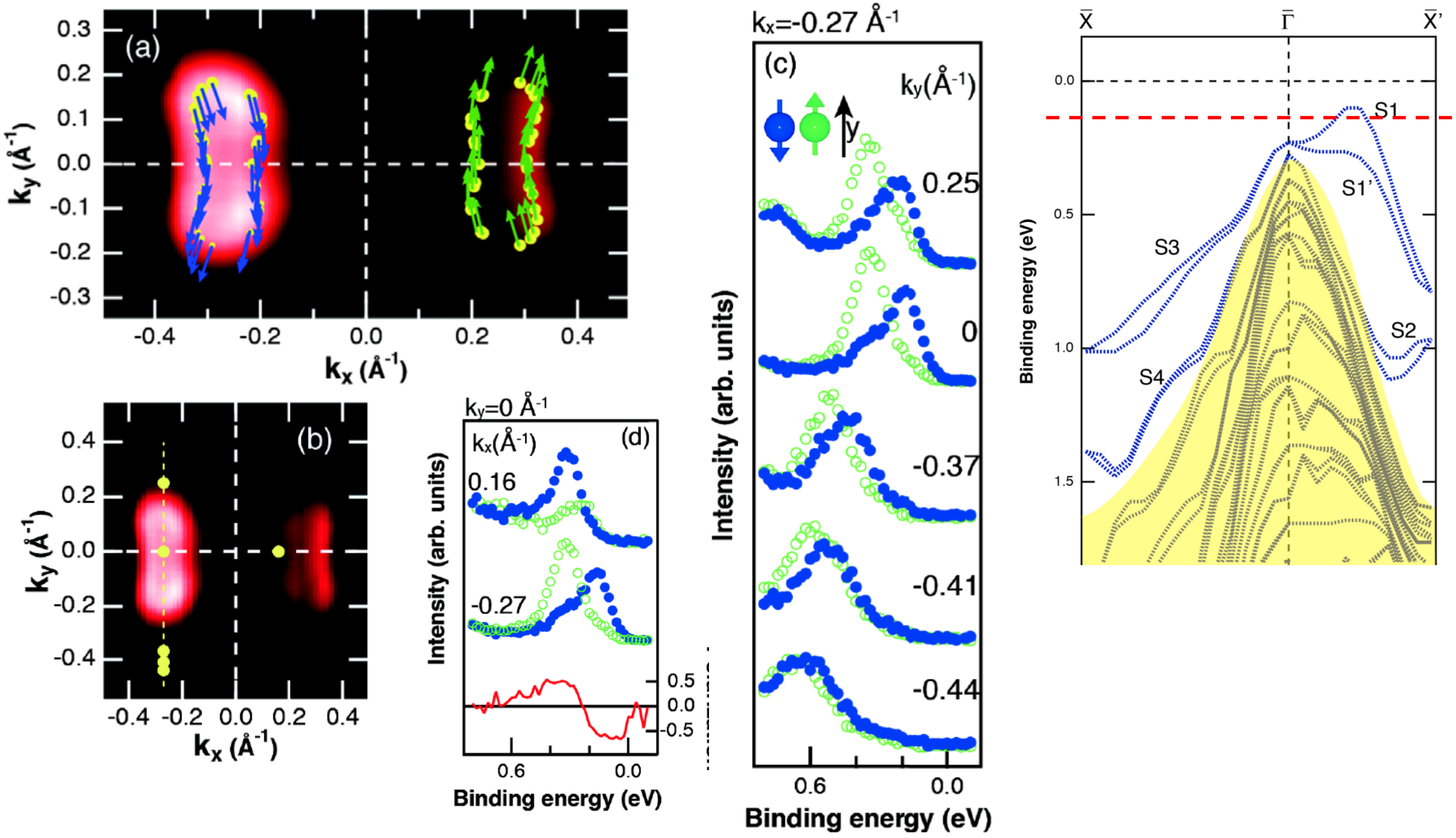
C_{1h}

The **mirror plane** requires the spin parallel to y-direction.



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)



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

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

Spin and orbital magnetic moments

spin magnetic moment

$$m^I_{spin,k} = -\mu_B \left\langle m_k(\vec{r}) \right\rangle_I$$

orbital magnetic moment

$$m^I_{orb,k} = -\mu_B \left\langle \ell_k \right\rangle_I$$

expansion with the local basis

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

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

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

$$\left\langle \ell_k \right\rangle_{I,VB} = \sum_{ipq} f_i \left\langle \Psi_i \middle| \beta^I_q \right\rangle \ell^I_{k,pq} \left\langle \beta^I_p \middle| \Psi_i \right\rangle$$

$$\ell^I_{k,pq} = \int_0^{r_c} \phi_p(r_I) \phi_q(r_I) r_I{}^2 dr_I \left\langle \mathrm{Y}^{\mathrm{sgn}(\kappa_q)}_{j_q \mu_q} \middle| \ell_k \middle| \mathrm{Y}^{\mathrm{sgn}(\kappa_p)}_{j_p \mu_p} \right\rangle$$

T. Oda and A. Hosokawa, Phys. Rev. B, 72, 224428 (2005)

Atomic magnetic moments in CoPt and FePt

r_c=2.5 a.u.		spin (μ_B)		orbital(μ_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;

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

Commute the parts with underline:

Dirac equation (part 3)

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

$$+ c^2 \underline{\left(\varepsilon + mc^2 + eA_0\right)}^{-2}\left(\vec{\sigma}, (-e\vec{p}A_0)\right)\left(\vec{\sigma}, \vec{p} + \frac{e}{c}\vec{A}\right)\varphi_L$$

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;

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

Using this approximation, the equation for the large component;

Formula 1

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

Using following general properties;

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

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

We obtain the following formula related with Zeeman' term;

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

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

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

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

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

Formula 2

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

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

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

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

Taking the leading terms in the approximation;

the approximation formula in (5-3).

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

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

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

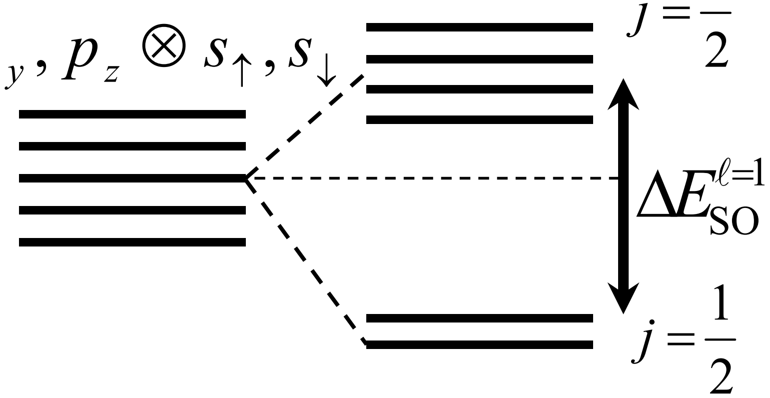
$$s = \frac{1}{2},\ \ s_z = \frac{1}{2}, -\frac{1}{2}$$

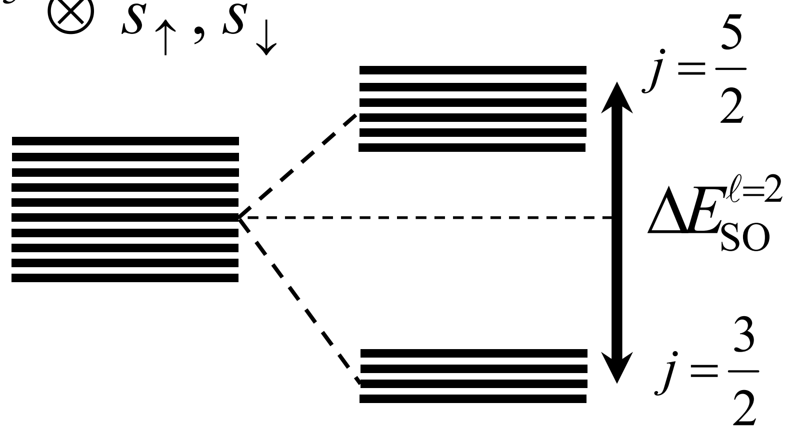
$$\ell = 1 \longrightarrow j = \frac{3}{2}, \frac{1}{2} \qquad\qquad \ell = 2 \longrightarrow j = \frac{5}{2}, \frac{3}{2}$$

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

$d^5 \otimes s_\uparrow, s_\downarrow$

$p_x, p_y, p_z \otimes s_\uparrow, s_\downarrow$





Eigenvalues of Pb atom

(in Ry energy unit)

Ref. State:
(Kr Core)$(5d)^{10}(6s)^2(6p)^2$

$n\ell$	j	all electron	pseudo (ΔE)
$5d$	3/2	-1.6734	-1.6733 (+0.0001)
$5d$	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)
$6p$	3/2	-0.2440	-0.2439 (+0.0001)

$\Delta E_{\mathrm{so}}(d)$
0.1912
2.601 eV

$\Delta E_{\mathrm{so}}(p)$
0.1106
1.505 eV

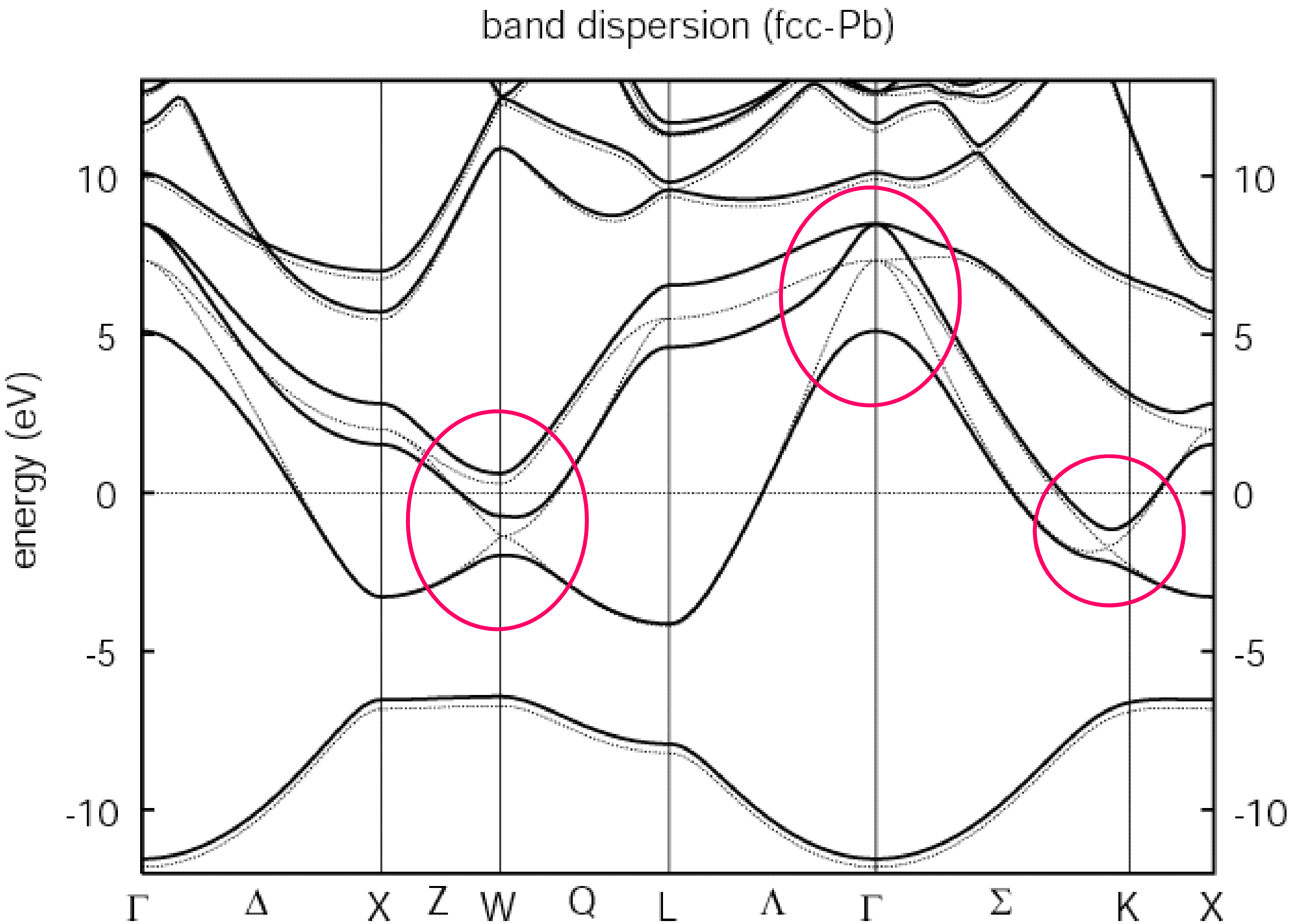
These values are in good agreement with the previous data.

$$\Delta E_{\mathrm{SO}}^{\ell} = E_{\mathrm{SO}}^{\ell+1/2} - E_{\mathrm{SO}}^{\ell-1/2} = \frac{\lambda}{2}(2\ell+1) \qquad \lambda = \frac{2}{2\ell+1}\Delta E_{SO}^{\ell}$$

$$\lambda_{\ell=2}^{\mathrm{Pb}} = 1.04\,\mathrm{eV}$$

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

Band dispersion of fcc Pb



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

a=9.35 a.u.(exp.)

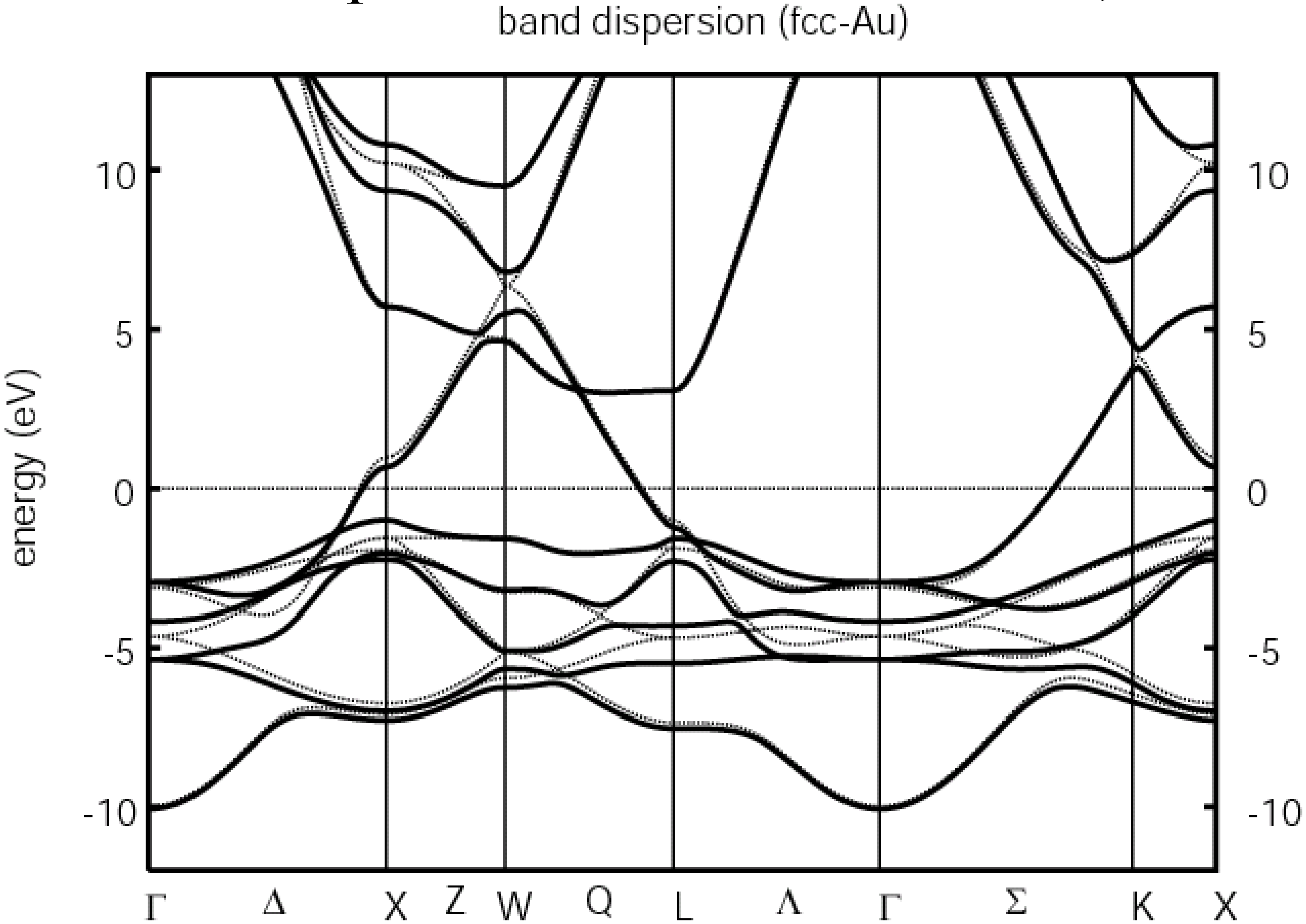
	USPP	(RAPW)
Γ:	-11.55	(-11.4)
L:	-7.92	(-8.2)
	-4.13	(-4.5)
K:	-6.61	(-6.7)
	-2.44	(-2.8)
	-0.93	(-1.2)
W:	-6.43	(-7.2)
X:	-6.52	(-6.7)
	-3.28	(-3.6)

At Γ point, ΔE_{so}

5*d*: 2.50 eV

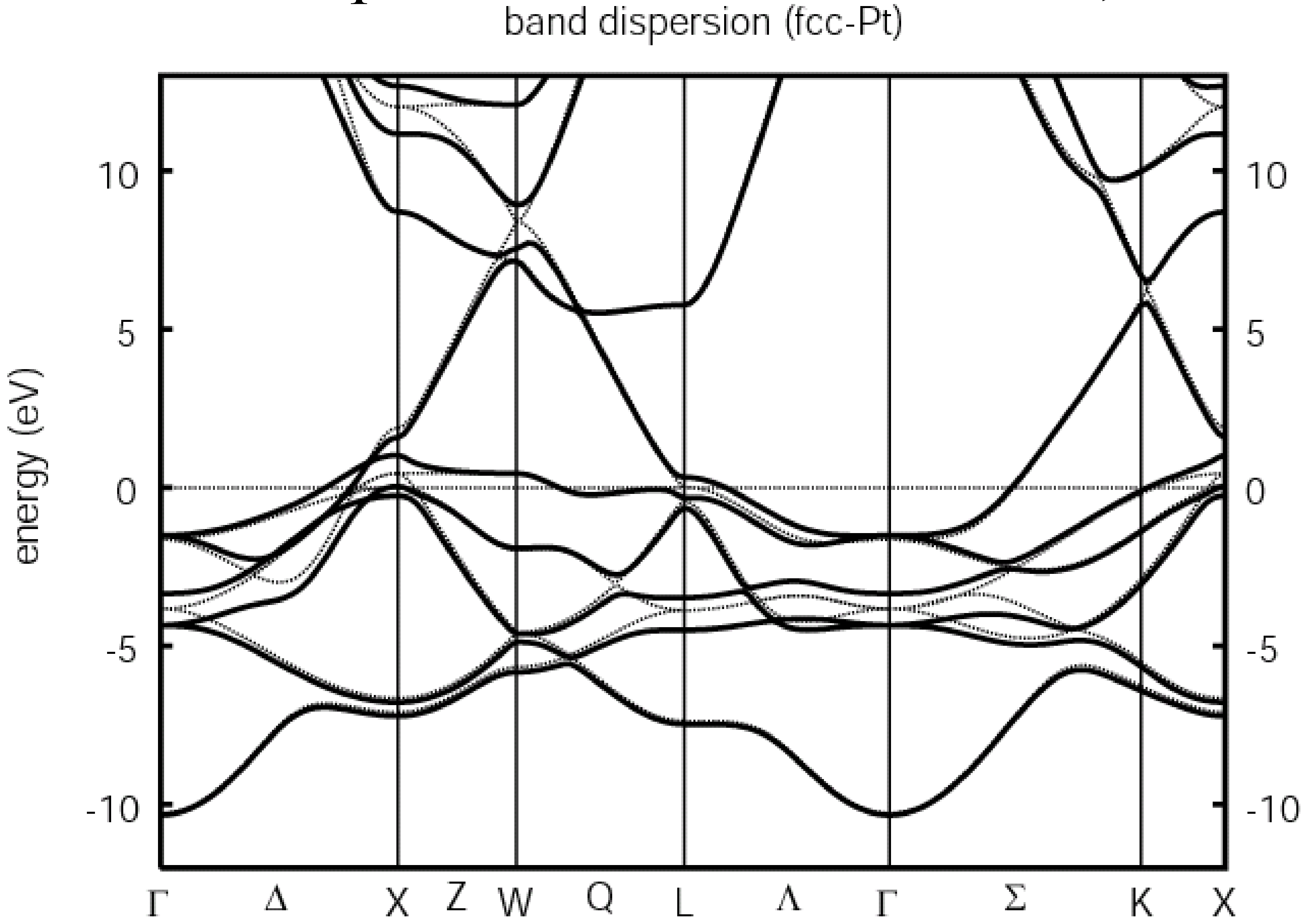
6*p*: 3.38 eV

Band dispersion of fcc noble metals, Au



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

Band dispersion of fcc noble metals, 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 eqution

Variational principles

$$\widetilde{E}[n(\mathbf{r})] = E[n(\mathbf{r})] - \mu\left(\int n(\mathbf{r})d\mathbf{r} - N_{\mathrm{e}}\right)$$

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

Kohn&Sham equation → Electron density

$$\left\{-\frac{1}{2}\nabla^2 + V_{\mathrm{eff}}(\mathbf{r})\right\}\Psi_i(\mathbf{r}) = \varepsilon_i \Psi_i(\mathbf{r})$$

$$n(\mathbf{r}) = \sum_i^{\mathrm{occ.}} \left|\Psi_i(\mathbf{r})\right|^2$$

Electron potential ← Poisson equation

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

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

$$\rho_{\alpha\beta}(r) = \sum_k f_k \{ \phi_{k\alpha}(r)\, \phi^*_{k\beta}(r) + \sum_{Inm} Q^I_{nm}(r) \langle \beta^I_n \mid \phi_{k\alpha} \rangle \langle \phi_{k\beta} \mid \beta^I_m \rangle \}$$

$$= \frac{1}{2} (n(r)\sigma_0 + m_x(r)\sigma_x + m_y(r)\sigma_y + m_z(r)\sigma_z)_{\alpha\beta}$$

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

Charge density $n(r)$

Spin density vector $m_\alpha(r)$

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

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

$$m_x(r) = 2\,\mathrm{Re}\,\rho_{12} \qquad m_y(r) = -2\,\mathrm{Im}\,\rho_{12} \qquad m_z(r) = \rho_{11} - \rho_{22}$$

- Total Energy(Energy Functional)

$$E_{tot}\left[\{\Phi_k\},\{R_I\}\right]=\sum_k f_k \langle \Phi_k \,|\, (-\frac{1}{2}\nabla^2 \;\; \sigma_0 + V_{NL}) \,|\, \Phi_k \rangle + \frac{1}{2}\iint \frac{n(r)n(r')}{|r-r'|}drdr'$$

$$+\int V_{loc}^{ion}\;(r)\,n(r)\,dr\; + \underline{E_{XC}\left[n(r),m(r)\right]} + U_{ion}\left[\{R_I\}\right]$$

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

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

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

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)

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

- Molecular Dynamics (Euler-Lagrange equation)

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

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

$$H = \frac{1}{f_k} \frac{\delta E_{tot}}{\delta \Phi_k}$$

$$\begin{pmatrix} \bar{\phi}_{k1}(r) \\ \bar{\phi}_{k2}(r) \end{pmatrix} = -\begin{pmatrix} H11 & H12 \\ H21 & H22 \end{pmatrix} \begin{pmatrix} \phi_{k1}(r) \\ \phi_{k2}(r) \end{pmatrix}$$

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

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

$$\rho_{\alpha\beta}(r) = \sum_k f_k \{\phi_{k\alpha}(r)\phi^*_{k\beta}(r) \quad + \sum_{Ipq} \underline{Q^I_{pq,\alpha\beta}(r)} \langle \beta^I_p | \Phi_k \rangle \langle \Phi_k | \beta^I_q \rangle \}$$

spinor type projector function

$$\beta^I_p(r) = b^I_{j\kappa\tau}(r) \mathrm{Y}^{\mathrm{sgn}(\kappa)}_{j\mu}(r_I) \qquad p = \{j\mu\kappa\tau\}$$

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

$$\kappa = -\ell - 1 < 0$$

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

$$\kappa = \ell > 0$$

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

Install to the plane wave method (II)

nonlocal potential

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

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

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

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

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

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

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

(Appendix 7)
Magnetic dipole-dipole interaction,
Shape magnetic anisotropy energy

Shape Magnetic Anisotropy Energy (SMAE)

From magnetic dipole interaction(MDI)

$$\mathbf{SMAE} = \boldsymbol{E}_{\mathbf{MDI}}^{[001]} - \boldsymbol{E}_{\mathbf{MDI}}^{[100]}$$

Continuum approach (CA)

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

M : Total magnetization

Ω : Volume of magnetic material

μ_0 : Permeability of vacuum

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

Discrete approach (DA) [1,2]

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

Spin density approach (SDA) [3]

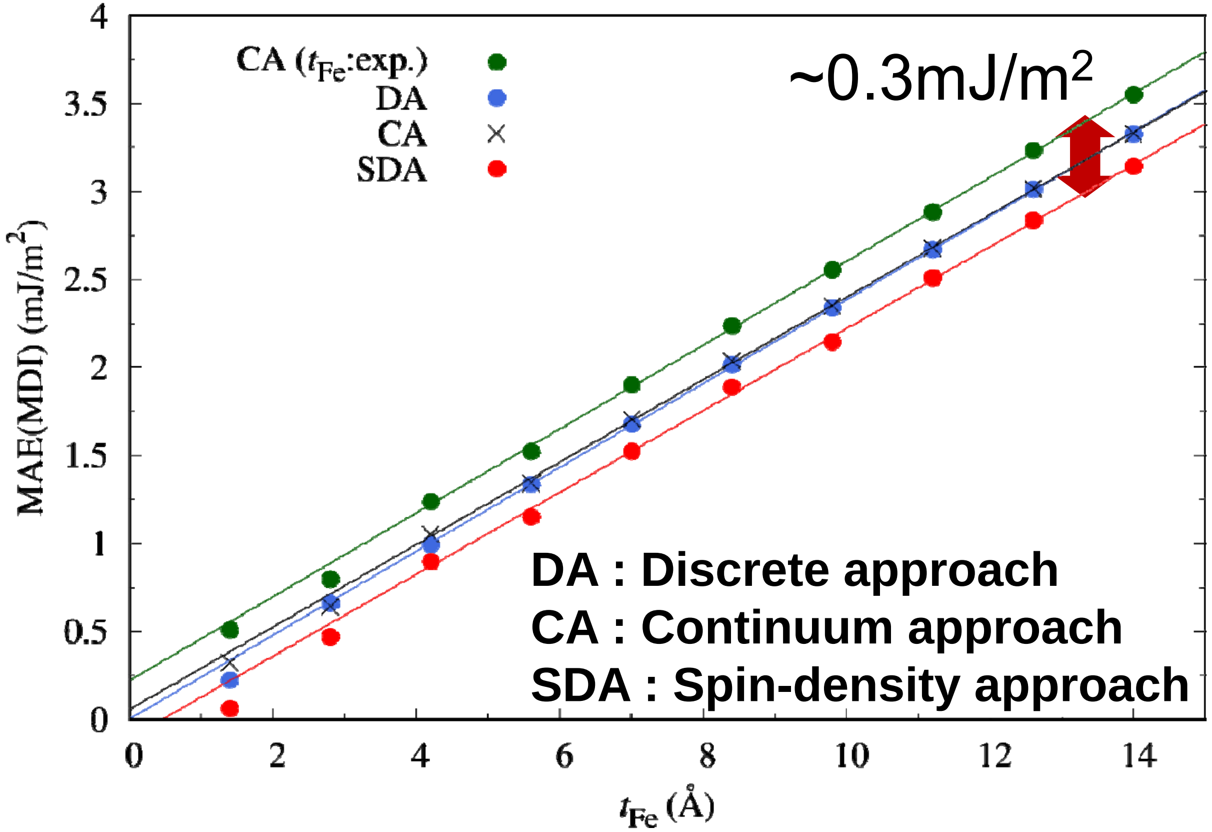
$$E_{MDI}^{\mathbf{m}} = \frac{e^2}{4m^2c^2} \iint d\mathbf{r}_1 d\mathbf{r}_2 \left[\frac{\mathbf{m}(\mathbf{r}_1) \cdot \mathbf{m}(\mathbf{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|^3} - 3\frac{\mathbf{m}(\mathbf{r}_1) \cdot (\mathbf{r}_1 - \mathbf{r}_2)\mathbf{m}(\mathbf{r}_2) \cdot (\mathbf{r}_1 - \mathbf{r}_2)}{|\mathbf{r}_1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

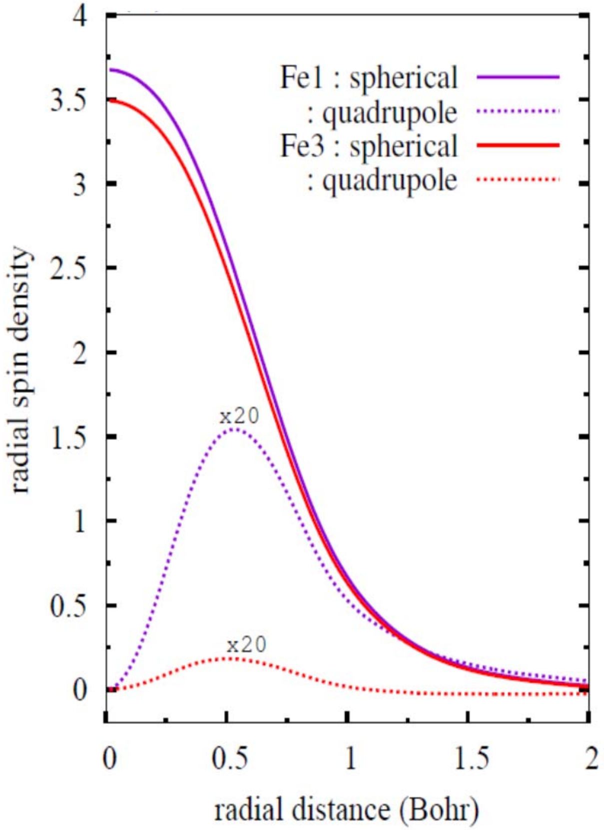
- High precision shape magnetic anisotropy from spin density distribution: magnetic interface/surface

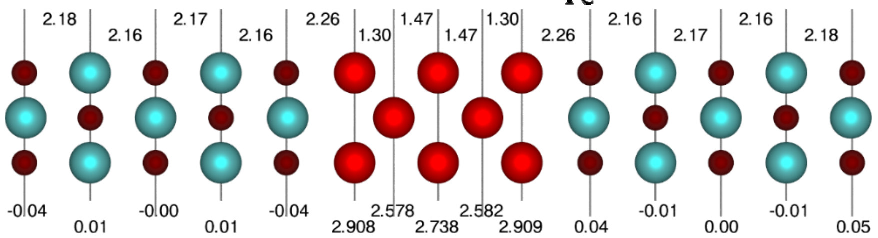
[1] H. J. G. Draaisma and W. J. M. de Jonge, J. Appl. Phys. 64, 1988.
[2] L. Szunyogh et al, Phys. Rev. B 51, 9552, 1995.
[3] T. Oda and M. Obata, J. Phys. Soc. Jpn. 87, 064803, 2018.

Quadrupole atomic spin density distribution at interface/surface

MgO(5ML)/Fe(xML)/MgO(5ML)-lcMgO(1x1)







Interface Fe/MgO supresses

Interface: quadrupole: Large

Inside : quadrupole: Small

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