

# Constrained non-collinear density functional theory implemented in OpenMX

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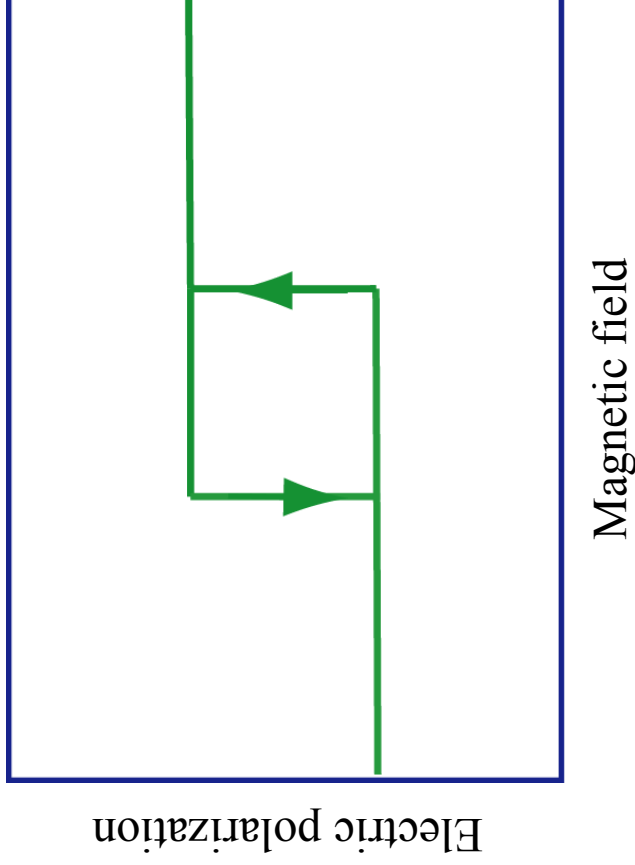
# Issues related to NC-DFT

- **Non-collinear magnetic structures**
- **Spin-orbit interactions**
- **Orbital magnetism**
- **Magneto-electric effects**
- **LDA+U**
- **etc.,**

# Motivation of the development

The magnetoelectric effect (ME) effect is one of cross-correlation phenomena, which bridges the magnetic field and electric polarization. Even with the lattice inversion symmetry, it appears. This may be due to spin orbit interaction.

Magnetoelectric effect (ME) effect



Materials	Electric Polarization ( $\mu\text{C}/\text{cm}^2$ )	
	$P_S$	$\Delta P$
TbMnO <sub>3</sub>	0.06-0.08( $P_c$ )	0.04( $P_a$ )
DyMnO <sub>3</sub>	0.06( $P_c$ )	0.2( $P_a$ , 1T), 0.26( $P_a$ , 4T)
GdMnO <sub>3</sub>	0	0.05 ( $P_a$ )
(Eu, Y)MnO <sub>3</sub>	0.12( $P_a$ )	0.03 ( $P_c$ , 8T)
TbMn <sub>2</sub> O <sub>5</sub>	0.04( $P_b$ )	0.08( $P_b$ )
DyMn <sub>2</sub> O <sub>5</sub>	0.02-0.04( $P_b$ )	0.05( $P_b$ , 8T)
HoMn <sub>2</sub> O <sub>5</sub>	0.04( $P_b$ )	
YMn <sub>2</sub> O <sub>5</sub>	0.1	
CoCr <sub>2</sub> O <sub>4</sub>	0.0002	0.0004
MnWO <sub>4</sub>	0.005	0.004
CuFeO <sub>2</sub>	0	0.04( $P_c$ , 14T)
Ni <sub>3</sub> V <sub>2</sub> O <sub>8</sub>	0.01( $P_a$ ), 0.01( $P_c$ )	0.005( $P_a$ , 5T)
Ga <sub>2-x</sub> Fe <sub>x</sub> O <sub>3</sub>	0	0.006( $P_b$ , 6T)

# Possible origin

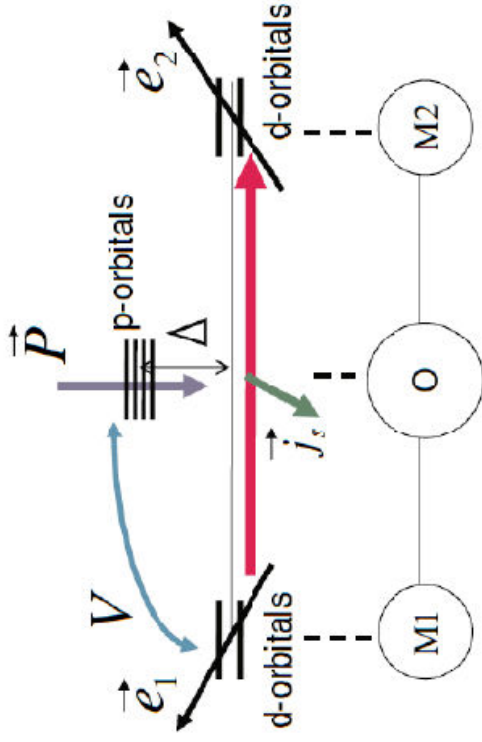


FIG. 1 (color online). The cluster model with two transition metal ions M1, M2 with the oxygen atom O between them. With the noncollinear spin directions  $\vec{e}_1$  and  $\vec{e}_2$ , there arises the spin current  $\vec{J}_s \propto \vec{e}_1 \times \vec{e}_2$  between M1 and M2. Here the direction of the vector  $\vec{J}_s$  (denoted by the short arrow near the middle of the diagram) is that of the spin polarization carried by the spin current. The direction of the electric polarization  $\vec{P}$  is given by  $\vec{P} \propto \vec{e}_{12} \times \vec{J}_s$  where  $\vec{e}_{12}$  is the unit vector connecting M1 and M2.

Katsura, Nagaosa, and Balatsky, PRL 95,057205 (2005)

Lattice with inversion symmetry leads to **paraelectric**.

Non-collinear spin order may break the inversion symmetry of the charge distribution via the spin-orbit coupling.

$$\begin{aligned} \mathbf{J} &\propto \mathbf{e}_1 \times \mathbf{e}_2 \\ \mathbf{P} &\propto \mathbf{e}_{12} \times \mathbf{J} \end{aligned}$$

## **A minimal scheme to treat the systems from the first-principles**

- Non-collinear (NC) DFT with two-component spinor
- Spin-orbit interaction
- LDA+U within NC-DFT
- Constrained schemes for spin and orbital moments
- Macroscopic polarization by Berry phase
- Orbital magnetic moment

# Dirac equation

$$\begin{aligned}
 (\varepsilon - eV - mc^2) \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} &= c\boldsymbol{\sigma} \cdot (\mathbf{p} + e\mathbf{A}) \begin{pmatrix} \varphi_3 \\ \varphi_4 \end{pmatrix} && \text{Large components} && \text{Small components} \\
 (\varepsilon - eV + mc^2) \begin{pmatrix} \varphi_3 \\ \varphi_4 \end{pmatrix} &= c\boldsymbol{\sigma} \cdot (\mathbf{p} + e\mathbf{A}) \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} && \varphi_1 && \varphi_3 \\
 &&& \varphi_2 && \varphi_4
 \end{aligned}$$

## Pauli matrices

$$\sigma_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \sigma_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

- Under the Lorentz transformation, the equation is invariant.

e.g., in case two coordinate systems move with a relative velocity  $v$  along x-direction

$$x = \frac{x' + vt'}{\sqrt{1 - \frac{v^2}{c^2}}} \quad y = y' \quad z = z' \quad t = \frac{t' + \frac{v}{c^2}x'}{\sqrt{1 - \frac{v^2}{c^2}}}$$

- It contains the first order derivatives with respect to space and time.
- It includes spin automatically without ad-hoc treatments.

# Simplification of Dirac eq.

$$\begin{aligned}(\varepsilon - eV - mc^2) \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} &= c\boldsymbol{\sigma} \cdot (\mathbf{p} + e\mathbf{A}) \begin{pmatrix} \varphi_3 \\ \varphi_4 \end{pmatrix} \\(\varepsilon - eV + mc^2) \begin{pmatrix} \varphi_3 \\ \varphi_4 \end{pmatrix} &= c\boldsymbol{\sigma} \cdot (\mathbf{p} + e\mathbf{A}) \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} \\ \varepsilon &= mc^2 + \varepsilon'\end{aligned}$$

Assuming that

$$|\varepsilon'| \ll mc^2 \quad \longrightarrow \quad \begin{pmatrix} \varphi_3 \\ \varphi_4 \end{pmatrix} \approx \frac{v}{c} \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix}$$

With the assumption, the Dirac eq. can be simplified as

$$\left\{ \frac{1}{2m}(\mathbf{p} + e\mathbf{A})^2 + \frac{e\hbar}{2m}\boldsymbol{\sigma} \cdot \mathbf{B} + V \right\} \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} = \varepsilon' \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix}$$

It looks Schrodinger eq., but the wave function is a two-component spinor.

# Eq. used in OpenMX

By expanding explicitly the simplified eq., we obtain

$$\left\{ -\frac{1}{2m}\nabla^2 + \frac{e}{2m}\mathbf{B} \cdot \mathbf{l} + \frac{e}{m}\mathbf{B} \cdot \frac{1}{2}\hbar\sigma + \text{Diamagnetic term} + V \right\} \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} = \varepsilon' \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix}$$

This has the Zeeman and diamagnetic terms, but unfortunately does not take account of the spin-orbit interaction.

By ignoring the diamagnetic term, and giving j-dependence of V, we get the following eq:

$$\left\{ -\frac{1}{2m}\nabla^2 + \frac{e}{2m}\mathbf{B} \cdot \mathbf{l} + \frac{e}{m}\mathbf{B} \cdot \frac{1}{2}\hbar\sigma + V_j \right\} \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix} = \varepsilon' \begin{pmatrix} \varphi_1 \\ \varphi_2 \end{pmatrix}$$

**This is the eq. used in the NC-DFT calculations of OpenMX**



# Spin-orbit interaction

Radial Dirac eq. for the majority component

$$\left[ \frac{1}{2M(r)} \left( \frac{d^2}{dr^2} + \frac{a^2}{2M(r)} \frac{dV}{dr} \frac{d}{dr} + \frac{a^2}{2M(r)} \frac{\kappa dV}{r dr} - \frac{\kappa(\kappa+1)}{r^2} \right) + \varepsilon_{nlj} - V \right] G_{nlj} = 0$$

$$M(r) = 1 + \frac{a^2(\varepsilon_{nlj} - V)}{2} \quad \kappa = l \text{ and } \kappa = -(l+1) \text{ for } j = l - \frac{1}{2} \text{ and } j = l + \frac{1}{2}$$

For each quantum number  $j$ , the dirac eq. is solved numerically, and its norm-conserving pseudopotential is constructed by the TM scheme.

**The unified pseudopotential is given by**

$$V_{\text{ps}} = \sum_{lm} \left[ |\Phi_J^M\rangle V_{\text{ps}}^{l+\frac{1}{2}} \langle \Phi_J^M| + |\Phi_{J'}^{M'}\rangle V_{\text{ps}}^{l-\frac{1}{2}} \langle \Phi_{J'}^{M'}| \right]$$

with the analytic solution for spherical coordinate:

where for  $J = l + \frac{1}{2}$  and  $M = m + \frac{1}{2}$

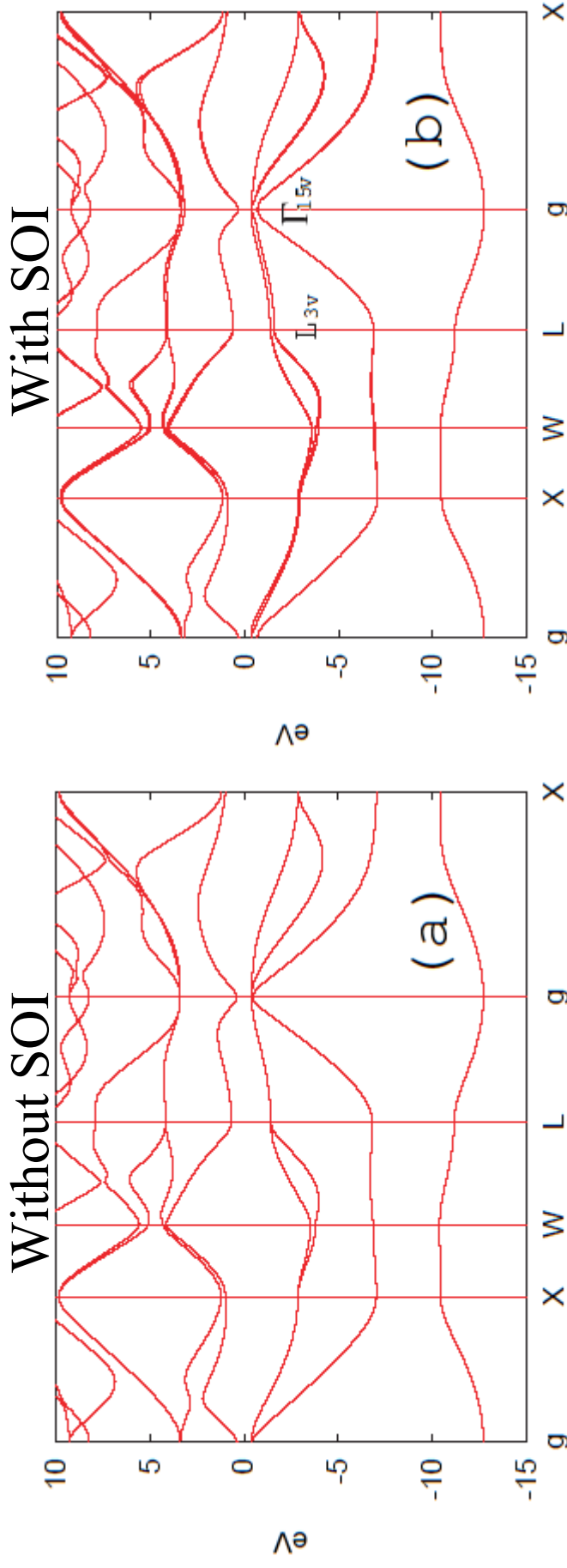
$$|\Phi_J^M\rangle = \left( \frac{l+m+1}{2l+1} \right)^{\frac{1}{2}} |Y_l^m\rangle |\alpha\rangle + \left( \frac{l-m}{2l+1} \right)^{\frac{1}{2}} |Y_l^{m+1}\rangle |\beta\rangle,$$

and for  $J' = l - \frac{1}{2}$  and  $M' = m - \frac{1}{2}$

$$|\Phi_{J'}^{M'}\rangle = \left( \frac{l-m+1}{2l+1} \right)^{\frac{1}{2}} |Y_l^{m-1}\rangle |\alpha\rangle - \left( \frac{l+m}{2l+1} \right)^{\frac{1}{2}} |Y_l^m\rangle |\beta\rangle.$$

# Spin-orbit splitting

e.g., GaAs



Level	OpenMX	LMTO <sup>(a)</sup>	PP <sup>(b)</sup>	Expt.
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$\Gamma_{15v}$	0.348	0.351	0.35	0.34
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$L_{3v}$	0.218	0.213	0.22	
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(a) M. Cardona, N. E. Christensen, and G. Gasol, Phys. Rev. B **38**, 1806 (1988).

(b) G. Theurich and N. A. Hill, Phys. Rev. B **64**, 073106 (2001).

# Non-collinear DFT No.1

Two-component spinor  $|\psi_\nu\rangle = |\varphi_\nu^\alpha\rangle + |\varphi_\nu^\beta\rangle$ ,

The charge density operator is defined by

$$\hat{n} = \sum f_\nu |\psi_\nu\rangle \langle \psi_\nu|, \quad \begin{pmatrix} n'_\uparrow & 0 \\ 0 & n'_\downarrow \end{pmatrix} = U n U^\dagger, \\ = U \begin{pmatrix} n_{\alpha\alpha} & n_{\alpha\beta} \\ n_{\beta\alpha} & n_{\beta\beta} \end{pmatrix} U^\dagger.$$

The total energy is a simple extension of the collinear case.

$$E_{\text{tot}} = \sum_{\sigma=\alpha,\beta} \sum_\nu f_\nu \langle \varphi_\nu^\sigma | \hat{T} | \varphi_\nu^\sigma \rangle + \sum_{\sigma\sigma'} w_{\sigma\sigma'} n_{\sigma'\sigma} + \frac{1}{2} \iint \frac{n'(\mathbf{r})n'(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{v}d\mathbf{v}' + E_{\text{xc}} \{n_{\sigma\sigma'}\},$$

The variation of wave functions leads to

$$\left. \begin{aligned} \frac{\delta F}{\delta \varphi_\mu^{\alpha,*}} = 0 \\ \frac{\delta F}{\delta \varphi_\mu^{\beta,*}} = 0 \end{aligned} \right\} \Rightarrow \begin{pmatrix} \hat{T} + w_{\alpha\alpha} + V_{\text{H}} + V_{\text{xc}}^{\alpha\alpha} & w_{\alpha\beta} + V_{\text{xc}}^{\alpha\beta} \\ w_{\beta\alpha} + V_{\text{xc}}^{\beta\alpha} & \hat{T} + w_{\beta\beta} + V_{\text{H}} + V_{\text{xc}}^{\beta\beta} \end{pmatrix} \begin{pmatrix} \varphi_\mu^\alpha \\ \varphi_\mu^\beta \end{pmatrix} = \varepsilon_\mu \begin{pmatrix} \varphi_\mu^\alpha \\ \varphi_\mu^\beta \end{pmatrix}$$

# Non-collinear DFT No.2

The spin-1/2 matrix gives us the relation between the spin direction in real space and spinor.

$$D \equiv \exp\left(\frac{-i\hat{\sigma} \cdot \mathbf{h}\phi}{2}\right)$$

- First, rotate  $\theta$  on the y-axis  $\rightarrow \exp\left(-i\frac{\sigma_2\theta}{2}\right)$
- Second, rotate  $\phi$  on the z-axis  $\rightarrow \exp\left(-i\frac{\sigma_3\phi}{2}\right)$

$$\begin{pmatrix} 1 \\ 0 \end{pmatrix} \Rightarrow \exp\left(-i\frac{\sigma_3\phi}{2}\right) \exp\left(-i\frac{\sigma_2\theta}{2}\right) \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

$U$

$$\text{Condition} \quad UnU^\dagger = \begin{pmatrix} n'_\uparrow & 0 \\ 0 & n'_\downarrow \end{pmatrix}.$$

We would like to find  $U$  which diagonalizes the matrix  $n$ , after algebra, it is given by

$$\begin{aligned} \phi &= -\arctan \frac{\text{Im } n_{\alpha\beta}}{\text{Re } n_{\alpha\beta}} \\ \theta &= \arctan \left( \frac{2(\text{Re } n_{\alpha\beta} \cos(\phi) - \text{Im } n_{\alpha\beta} \sin(\phi))}{n_{\alpha\alpha} - n_{\beta\beta}} \right) \\ n'_\uparrow &= \frac{1}{2}(n_{\alpha\alpha} + n_{\beta\beta}) + \frac{1}{2}(n_{\alpha\alpha} - n_{\beta\beta}) \cos(\theta) + (\text{Re } n_{\alpha\beta} \cos(\phi) - \text{Im } n_{\alpha\beta} \sin(\phi)) \sin(\theta) \\ n'_\downarrow &= \frac{1}{2}(n_{\alpha\alpha} + n_{\beta\beta}) - \frac{1}{2}(n_{\alpha\alpha} - n_{\beta\beta}) \cos(\theta) - (\text{Re } n_{\alpha\beta} \cos(\phi) - \text{Im } n_{\alpha\beta} \sin(\phi)) \sin(\theta) \end{aligned}$$

# Constrained NC-DFT: a harmonic constraint

1) On each site,  $N_{\uparrow}$  and  $N_{\downarrow}$  can be found by diagonalizing  $N$

$$N = \begin{pmatrix} N_{\alpha\alpha} & N_{\alpha\beta} \\ N_{\beta\alpha} & N_{\beta\beta} \end{pmatrix}$$

2) The density matrix with a given  $\theta_{0,i}$  and  $\phi_{0,i}$ , and the calculated  $N_{\uparrow}$  and  $N_{\downarrow}$  can be constructed by

$$N^{(0)} = U_0^\dagger \begin{pmatrix} N_{\uparrow} & 0 \\ 0 & N_{\downarrow} \end{pmatrix} U_0$$

3) If  $N$  and  $N_0$  are different, the penalty functional is given by

$$E_{cs} = v \sum_i \text{Tr} \left( (N_i - N_i^{(0)})^2 \right)$$

- Without largely changing the magnitude of spin moment, it is possible to control the spin direction self-consistently.
- The scheme can be applied to only the spin magnetic moment. For the orbital magnetic moment, it does not work since there is no driving force in pinning the direction in the LDA and GGA.

# Constrained NC-DFT: a Zeeman constraint

It is assumed that the magnetic field can vary from one atomic site to the others.

Then, the energy contributions arising the Zeeman terms can be written by

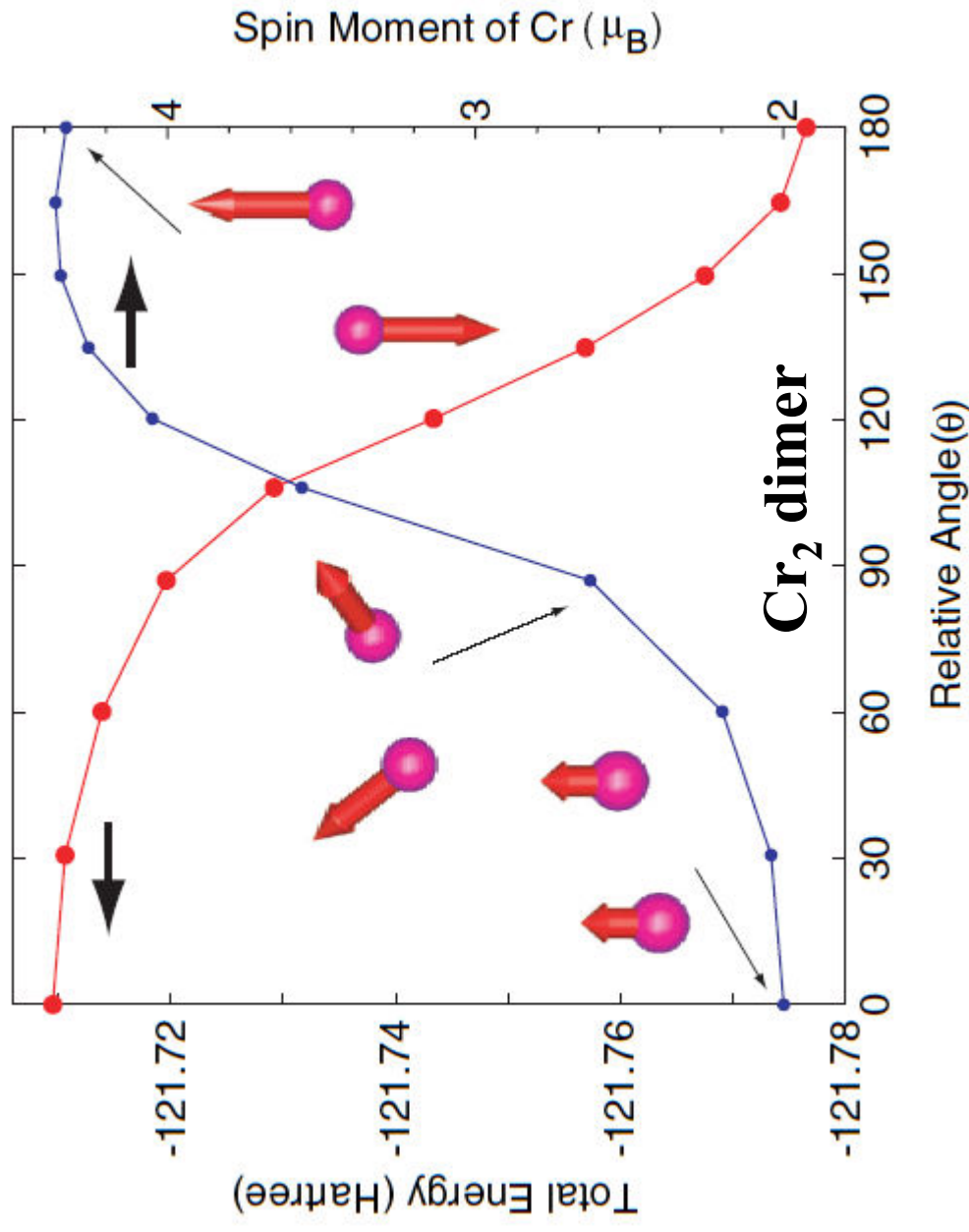
$$E_z = E_{zs} + E_{zo},$$

$$E_{zs} = \sum_i \mathbf{B}_i^s \cdot \mathbf{s}_i = \sum_i (B_{ix}^s s_{ix} + B_{iy}^s s_{iy} + B_{iz}^s s_{iz}),$$

$$E_{zo} = \frac{1}{2} \sum_i \mathbf{B}_i^o \cdot \mathbf{l}_i = \frac{1}{2} \sum_i (B_{ix}^o l_{ix} + B_{iy}^o l_{iy} + B_{iz}^o l_{iz}).$$

- Both the direction spin and orbital magnetic moments can be controlled.
- The magnitude of moments are enlarged depending on the magnetic field.

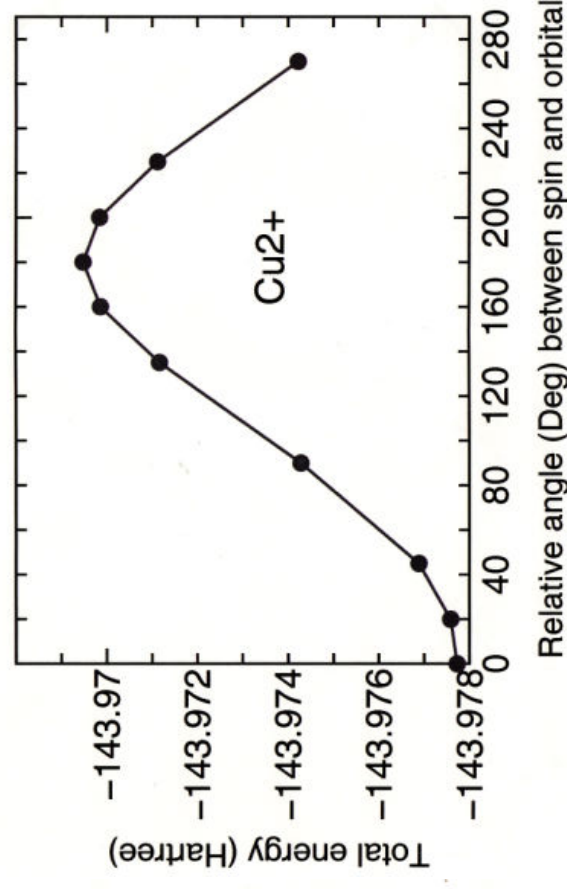
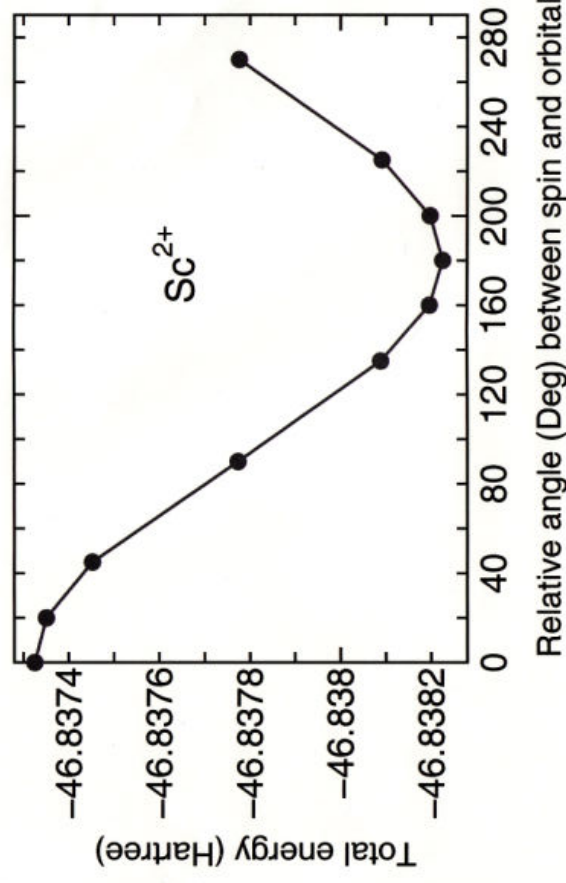
# Example: a harmonic constraint



The spin direction is controlled by the harmonic constraint, and the spin moment is also determined self-consistently.

# Example: a Zeeman constraint

Since the magnetic field can be applied to spin and orbital moments independently, **it enables us to directly evaluate Hund's third rule.**



Less than half in the shell structure → the anti-parallel is favored  
More than half in the shell structure → the parallel is favored



# Notes in the constraint schemes

There are **many degree of freedom in the occupation number space**. This brings serious problems such that the calculated result is a **meta stable state**. In order to find the ground state, the following procedure is recommended.

1. Using relatively large  $\nu$  and  $\mathbf{B}$ , try to get a polarized state.
2. With the restart file generated by the procedure 1, perform the SCF calculation with small  $\nu$  and  $\mathbf{B}$ .
3. Redo the procedure 2 with the restart file one-step before by gradually decreasing  $\nu$  and  $\mathbf{B}$  until the values you want

$$E_{cs} = \nu \sum_i \text{Tr} \left( (N_i - N_i^{(0)})^2 \right)$$

$$E_{zs} = \sum_i \mathbf{B}_i^s \cdot \mathbf{s}_i$$

$$E_{zo} = \frac{1}{2} \sum_i \mathbf{B}_i^o \cdot \mathbf{l}_i$$

# LDA+U within NC-DFT

In conjunction with unrestricted Hartree-Fock theory, we introduce a Hubbard term.

$$E_{\text{LDA+U}} = E_{\text{LDA}} + E_{\text{U}}$$

Starting from the diagonal occupation matrix, a rotational invariant Formula can be obtained even for the NC case.

$$\begin{aligned} E_{\text{U}} &= \frac{1}{2} \sum_s U_s \left[ \text{Tr}(A_s N_s A_s^\dagger) - \text{Tr}(A_s N_s A_s^\dagger A_s N_s A_s^\dagger) \right], \\ &= \frac{1}{2} \sum_s U_s \left[ \text{Tr}(n_s) - \text{Tr}(n_s n_s) \right], \\ &= \frac{1}{2} \sum_s U_s \left[ \sum_{\sigma m} n_{s,mm}^{\sigma\sigma} - \sum_{\sigma m, \sigma' m'} n_{s,mm'}^{\sigma\sigma'} \right], \end{aligned}$$

The occupation number operator is given by

$$\hat{n}_{smm'}^{\sigma\sigma'} = \frac{1}{2} \left( |\tilde{sm}\sigma\rangle \langle sm'\sigma'| + |sm\sigma\rangle \langle \tilde{sm}'\sigma'| \right),$$

Then, the effective potential operator becomes

$$\hat{v}_{\text{U}} = \frac{1}{2} \sum_{\sigma\sigma'} \sum_{smm'} \left[ |\tilde{sm}\sigma\rangle v_{\text{U},smm'}^{\sigma\sigma'} \langle sm'\sigma'| + |sm\sigma\rangle v_{\text{U},smm'}^{\sigma\sigma'} \langle \tilde{sm}'\sigma'| \right].$$

## The effective Hamiltonian due to the constraints and LDA+U

The effective Hamiltonian due to the constraints  
and LDA+U take **the same form**

$$\hat{v}_{\text{eff}}^{\sigma\sigma'} = \frac{1}{2} \sum_{\sigma\sigma'} \sum_{smm'} \left[ |s\tilde{m}\sigma\rangle v_{\text{eff},smm'}^{\sigma\sigma'} \langle sm'\sigma'| + |sm\sigma\rangle v_{\text{eff},smm'}^{\sigma\sigma'} \langle s\tilde{m}'\sigma'| \right].$$

$$v_{\text{eff},smm'}^{\sigma\sigma'} = v_{\text{U},smm'}^{\sigma\sigma'} + v_{\text{constraint},smm'}^{\sigma\sigma'}$$

Thus, **we only have to add each contribution**,  
leading to that the implementation makes easier.

# Orbital magnetic moment

The orbital moment for **localized electrons** can be calculated by projecting wave functions onto the local angular momentum operator on each site as follows:

$$\begin{aligned}
 l_{iv} &= \int dE \sum_{\mathbf{k}} \sum_{\nu} f(E) \langle \psi_{\mathbf{k}\nu} | \hat{l}_v | \psi_{\mathbf{k}\nu} \rangle \delta(E - \varepsilon_{\mathbf{k}\nu}), \\
 &= \int dE \sum_{\mathbf{k}} \sum_{\nu} f(E) \left[ \langle \varphi_{\mathbf{k}\nu}^{\alpha} | \hat{l}_v | \varphi_{\mathbf{k}\nu}^{\alpha} \rangle + \langle \varphi_{\mathbf{k}\nu}^{\beta} | \hat{l}_v | \varphi_{\mathbf{k}\nu}^{\beta} \rangle \right] \delta(E - \varepsilon_{\mathbf{k}\nu}), \\
 &= \sum_{\mathbf{k}} \sum_{\nu} f(\varepsilon_{\mathbf{k}\nu}) \left[ \sum_{\kappa, \kappa'} c_{\mathbf{k}\nu, i\kappa}^{\alpha, *} c_{\mathbf{k}\nu, i\kappa'}^{\alpha} \langle \phi_{i\kappa}^{\alpha} | \hat{l}_v | \phi_{i\kappa'}^{\alpha} \rangle + c_{\mathbf{k}\nu, i\kappa}^{\beta, *} c_{\mathbf{k}\nu, i\kappa'}^{\beta} \langle \phi_{i\kappa}^{\beta} | \hat{l}_v | \phi_{i\kappa'}^{\beta} \rangle \right], \\
 &= \sum \rho_{i\kappa, i\kappa'}^{\alpha\alpha} \langle \phi_{i\kappa}^{\alpha} | \hat{l}_v | \phi_{i\kappa'}^{\alpha} \rangle + \rho_{i\kappa, i\kappa'}^{\beta\beta} \langle \phi_{i\kappa}^{\beta} | \hat{l}_v | \phi_{i\kappa'}^{\beta} \rangle,
 \end{aligned}$$

Compound	$M_s$		$M_o$		Expt. in total
	OpenMX	Other calc.	OpenMX	Other calc.	
MnO	4.560	4.49 <sup>[1]</sup>	0.001	0.00 <sup>[1]</sup>	4.79, 4.58
FeO	3.586	3.54 <sup>[1]</sup>	1.010	1.01 <sup>[1]</sup>	3.32
CoO	2.685	2.53 <sup>[1]</sup>	1.137	1.19 <sup>[1]</sup>	3.35, 3.8
NiO	1.603	1.53 <sup>[1]</sup>	0.171	0.27 <sup>[1]</sup>	1.77, 1.64, 1.90
Fe <sub>2</sub> O <sub>3</sub>	4.014		0.034		
GaFeO <sub>3</sub> (GGFF)	3.950-3.956		0.020-0.021		
GaFeO <sub>3</sub> (GFFF)	3.923-4.001		0.020-0.022		

[1] A. Svane and O. Gunnarsson, Phys. Rev. Lett. **65**, 1148 (1990).

# Macroscopic polarization by the Berry phase within NC-DFT

The Berry phase formula for macroscopic electric polarization is given by

$$2\pi P_i = \mathbf{G}_i \cdot \mathbf{P} = -\frac{e}{(2\pi)^3} \sum_{\sigma} \int_{\mathbf{B}} d\mathbf{k}^3 \mathbf{G}_i \cdot \left( \frac{\partial}{\partial \mathbf{k}'} \eta_{\sigma}(\mathbf{k}, \mathbf{k}') \right)_{\mathbf{k}'=\mathbf{k}},$$

The quantum phase is defined by

$$\eta_{\sigma}(\mathbf{k}, \mathbf{k}') = \text{Im} \left\{ \ln \left( \det \langle u_{\sigma\mu}^{(\mathbf{k})} | u_{\sigma\nu}^{(\mathbf{k}')} \rangle \right) \right\},$$

The overlap matrix can be easily evaluated by

$$\langle u_{\sigma\mu}^{(\mathbf{k})} | u_{\sigma\nu}^{(\mathbf{k}+\Delta\mathbf{k})} \rangle = \langle \psi_{\alpha,\sigma\mu}^{(\mathbf{k})} | e^{-i\Delta\mathbf{k}\cdot\mathbf{r}} | \psi_{\alpha,\sigma\mu}^{(\mathbf{k}+\Delta\mathbf{k})} \rangle + \langle \psi_{\beta,\sigma\mu}^{(\mathbf{k})} | e^{-i\Delta\mathbf{k}\cdot\mathbf{r}} | \psi_{\beta,\sigma\mu}^{(\mathbf{k}+\Delta\mathbf{k})} \rangle,$$

since the eigenfunctions  $\alpha$  and  $\beta$  are orthogonal in spin space.

# Summary

**The theory and implementation to OpenMX related to non-collinear DFT were summarized**

**The method can be very useful to treat many kind of interesting phenomena.**