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# General model for spin-order induced polarization in multiferroics

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# *General Model for the Spin-Order Induced Polarization in Multiferroics*

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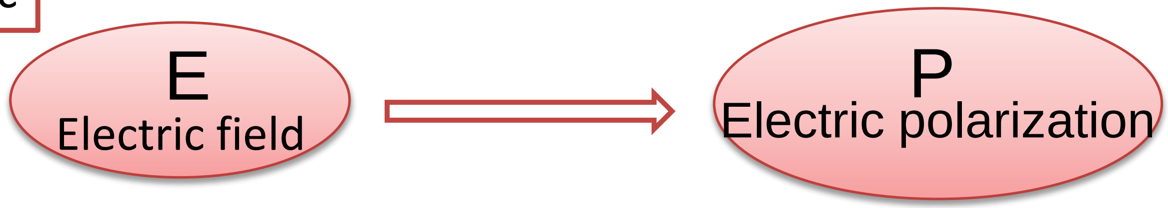
# Outline

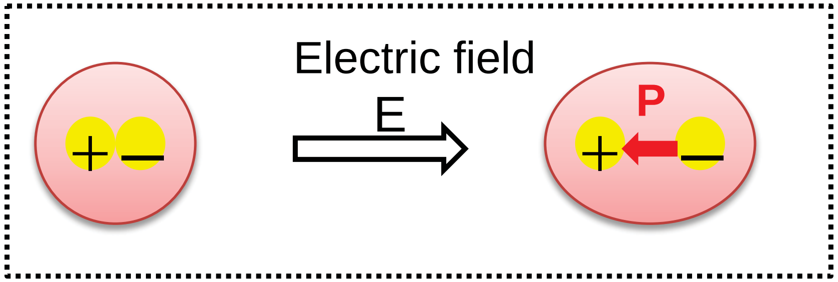
- Introduction for multiferroics
- General theory for ferroelectricity induced by spin order
- Triangular systems: Generalized spin current model
- $CaMn_7O_{12}$: Combined effect of Dzyaloshinskii-Moriya interaction and exchange striction
- $Cu_2OSeO_3$: Single-site term
- Summary

# Introduction: ferroelectricity and ferromagnetism

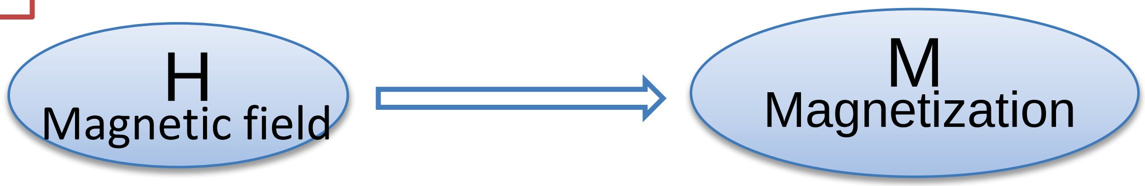
Ferroelectric



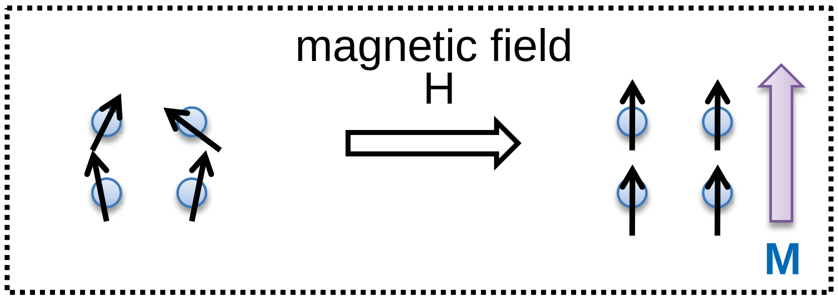

Electric polarization is controlled by the electric field.




Ferromagnetic




Magnetization is controlled by the magnetic field.

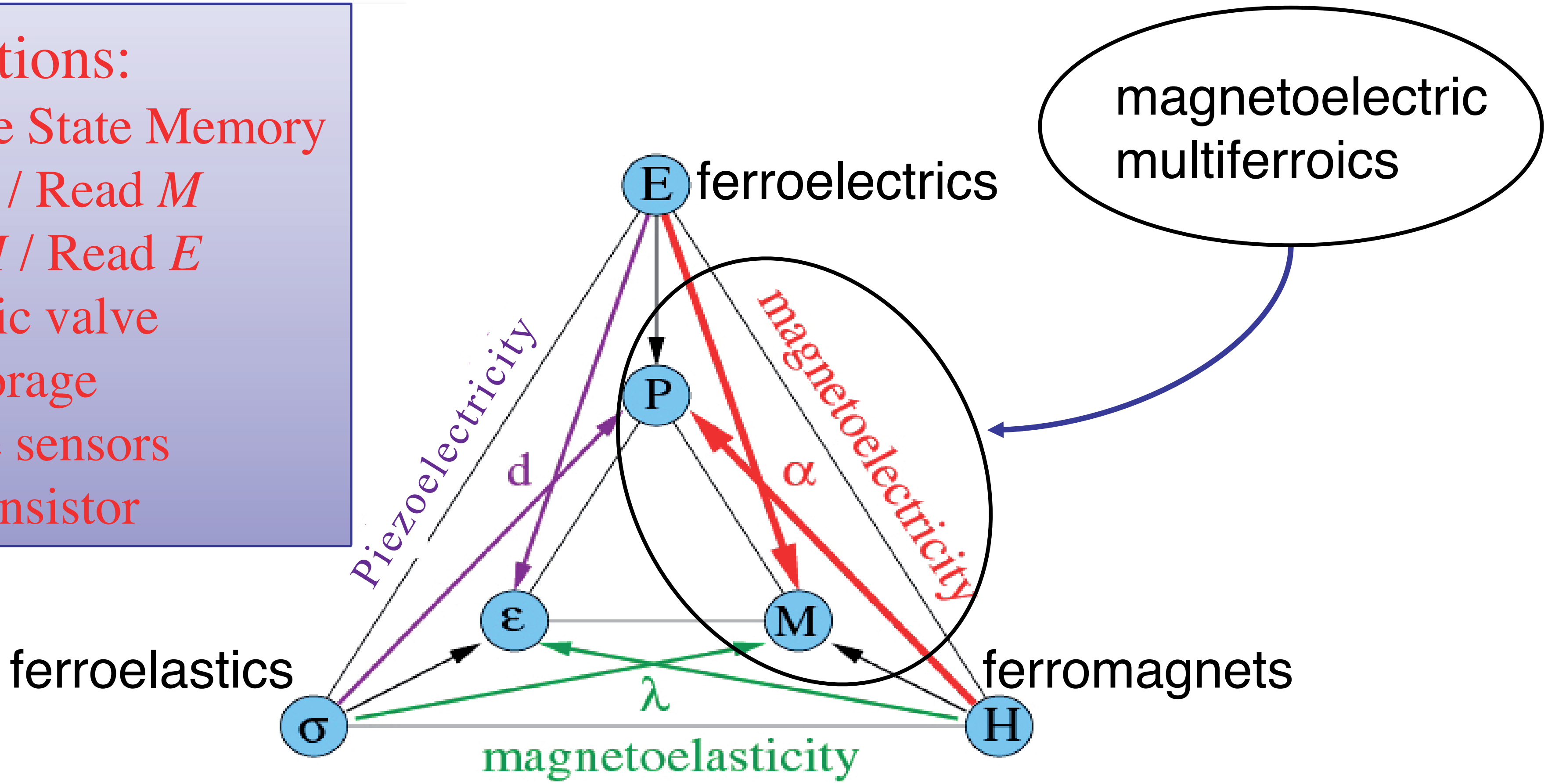


# Introduction: multiferroics

- two or more ferroic properties
- coupling between ferroic properties

**Applications:**

- Multiple State Memory
- Write *E* / Read *M*
- Write *M* / Read *E*
- Magnetic valve
- Data storage
- Tunable sensors
- Spin transistor

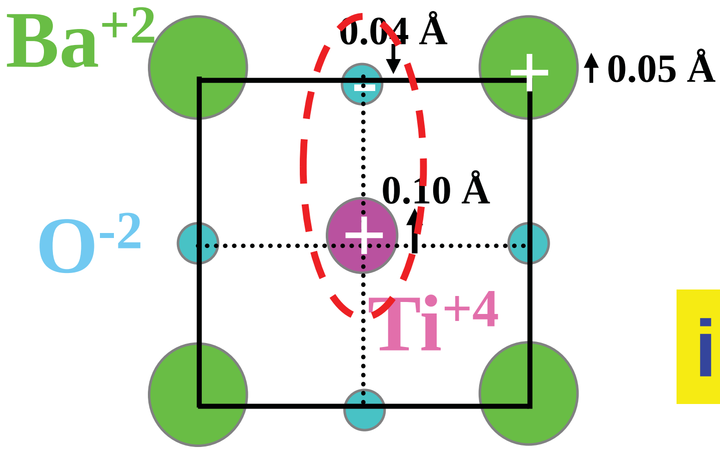


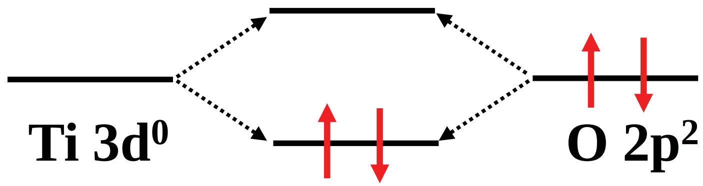

Spaldin *et al.*, Science 309, 391 (2005)

# Conventional ferroelectricity

**Covalent bond formation**

**$BaTiO_3$: polarization from cation/anion paired diploes**



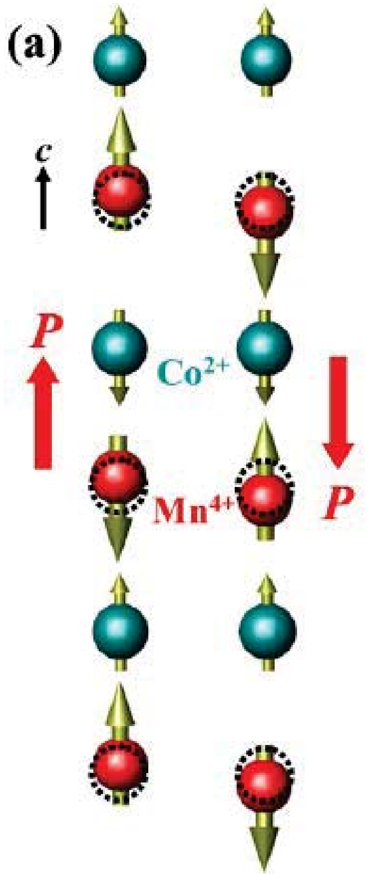




**inversion symmetry broken**

**Empty *d* band, no room for magnetism!**

N. A. Hill: "***Why Are There so Few Magnetic Ferroelectrics?***"
J. Phys. Chem. B **104**, 6694 (2000)

# Spin-order induced ferroelectricity: Exchange striction




$Ca_3CoMnO_6$

$$H = \sum_{<i,j>} J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

Minimize the symmetric spin exchange interaction energy

**Usually collinear order: e.g., E-type order in $HoMnO_3$**

Choi *et al*., PRL **100**, 047601 (2008)
Picozzi *et al*., PRL 99, 227201, (2007)
Wu *et al*., PRL **102**, 026404 (2009)

# Spin-order induced ferroelectricity: KNB model

$$\vec{P}_{12} \propto -\vec{e}_{12} \times (\vec{S}_1 \times \vec{S}_2)$$

**Noncollinear order**

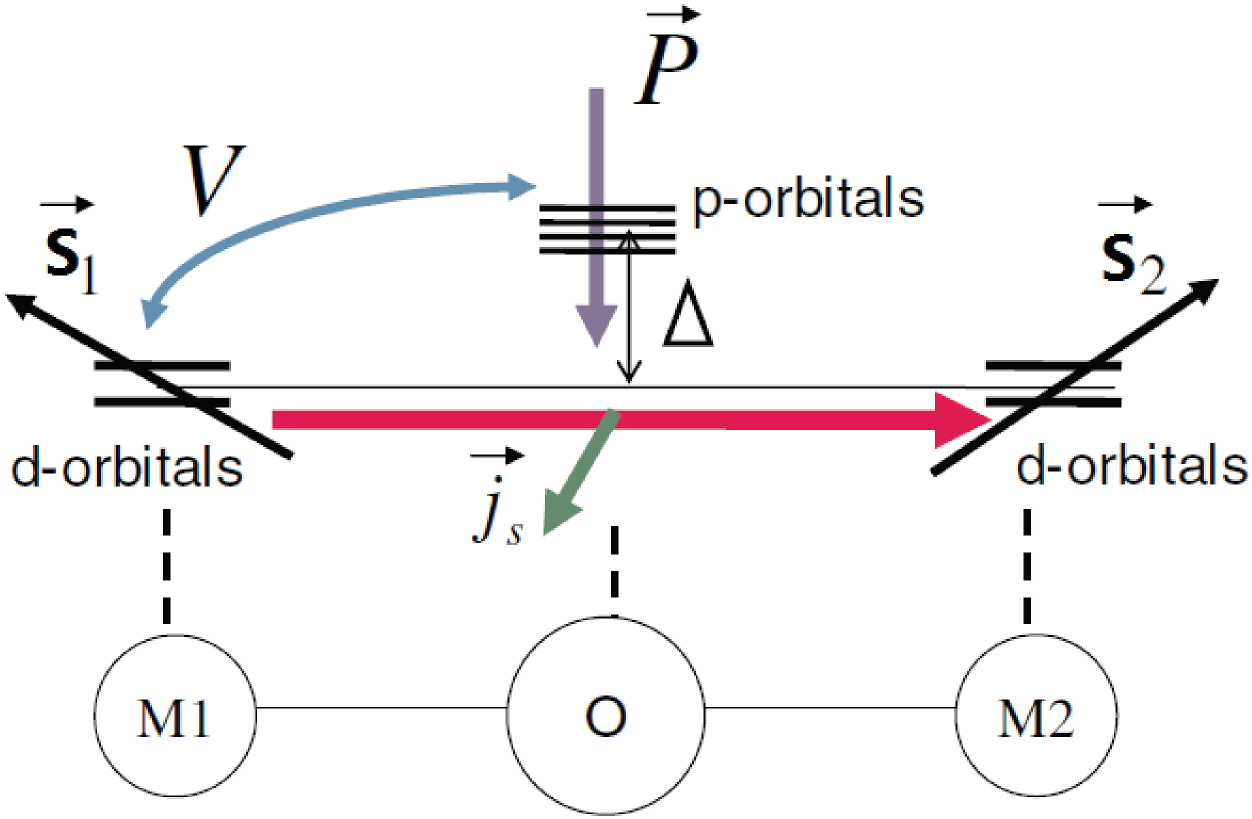
$$\vec{j}_s \propto \vec{S}_1 \times \vec{S}_2$$

**Spin current model**

Pure electronic model
(ions fixed)

Spin-orbit coupling

L-M-L line model

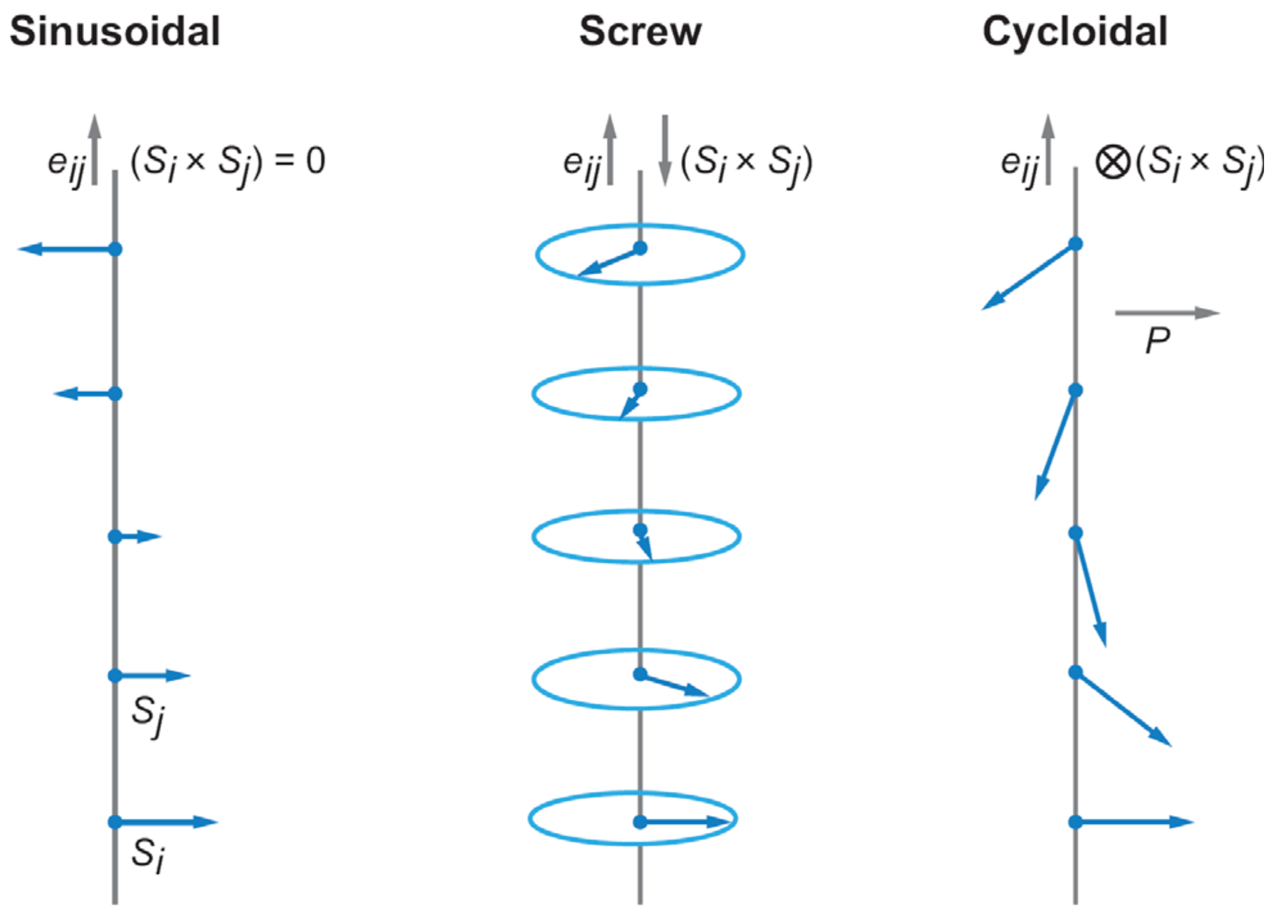



Katsura, Nagaosa, and Balatsky, Phys. Rev. Lett. **95**, 057205 (2005)

# Ferroelectricity induced by spin spiral: KNB model

$$\vec{P}_{ij} \propto -\vec{e}_{ij} \times (\vec{S}_i \times \vec{S}_j)$$

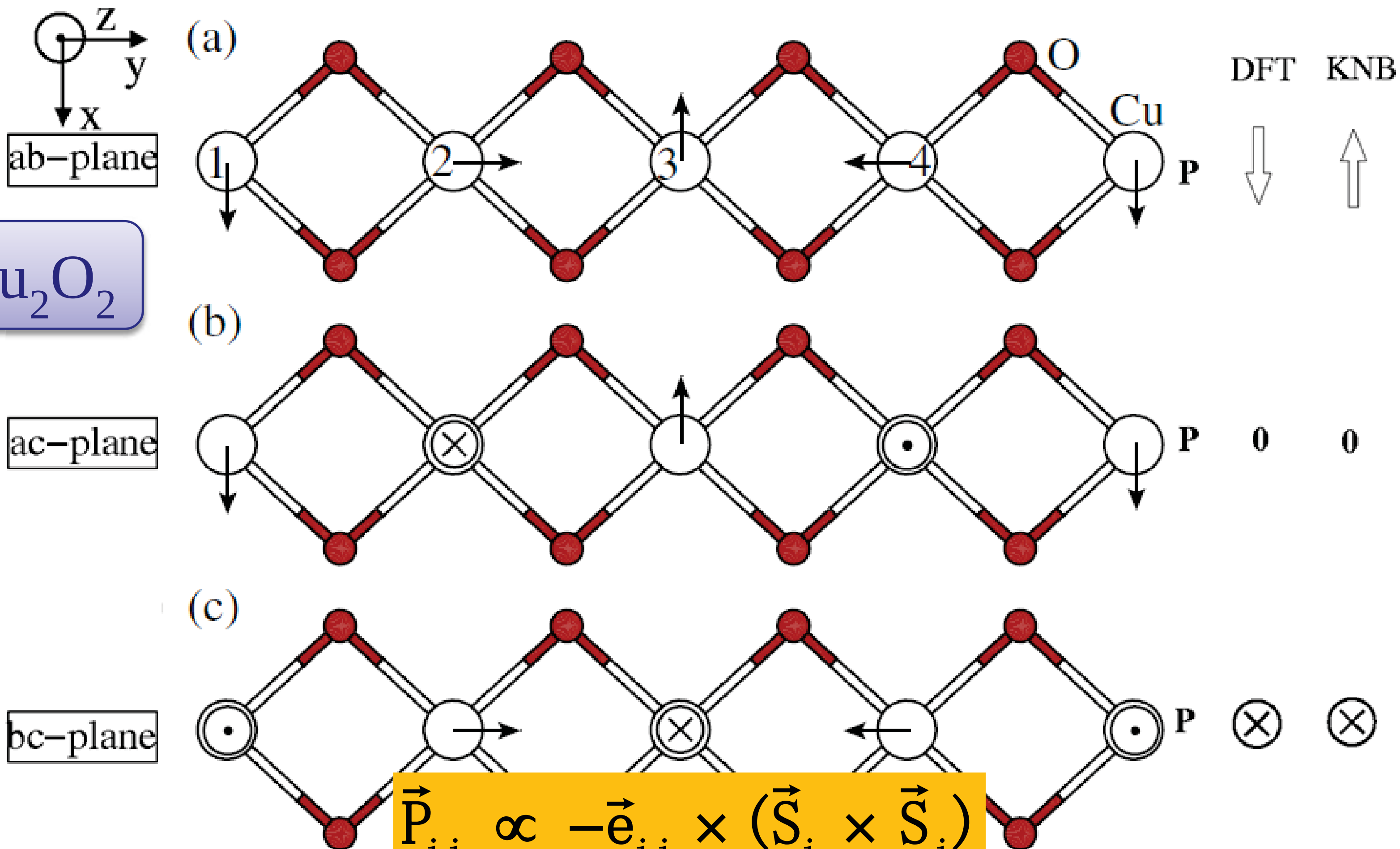
**P = 0** **P = 0** **P ≠ 0**




Katsura, Nagaosa, and Balatsky, Phys. Rev. Lett. **95**, 057205 (2005)

# Ferroelectricity induced by spin spiral: DFT

$LiCu_2O_2$

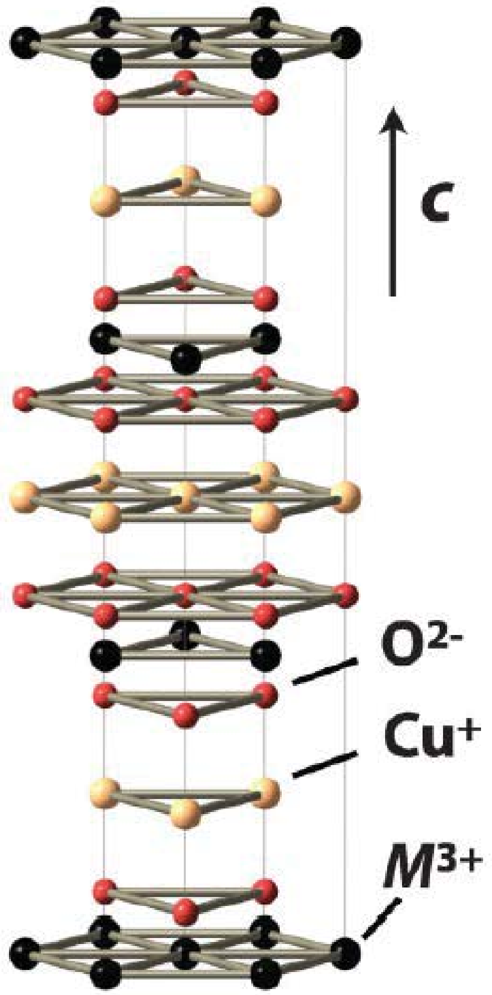


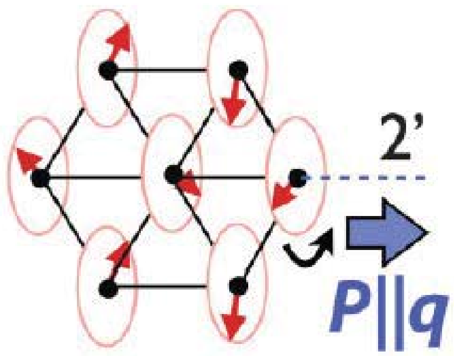

$$\vec{P}_{ij} \propto -\vec{e}_{ij} \times (\vec{S}_i \times \vec{S}_j)$$

- KNB: correct symmetry, wrong absolute direction
- Anisotropy in polarization: |P(ab)| > |P(bc)| ($TbMnO_3$: similar)

H. J. Xiang *et al.*, PRL **99**, 257203 (2007); PRL **101**, 037209 (2008)
Malashevich and Vanderbilt, PRL 101, 037210 (2008)

# Ferroelectricity induced by screw spin spiral




- $CuFeO_2$ and $CuCrO_2$
- ***P***//***q***
- ***P***(**q**)=-***P***(-***q***)
- KNB model and exchange striction: ***P*** = 0

New model needed!

T. Kimura *et al.*, Phys. Rev. B **73**, 220401(R) (2006).
S. Seki *et al.*, Phys. Rev. Lett. **101**, 067204 (2008).

# Reminder: Symmetry of moments

Time Reversal

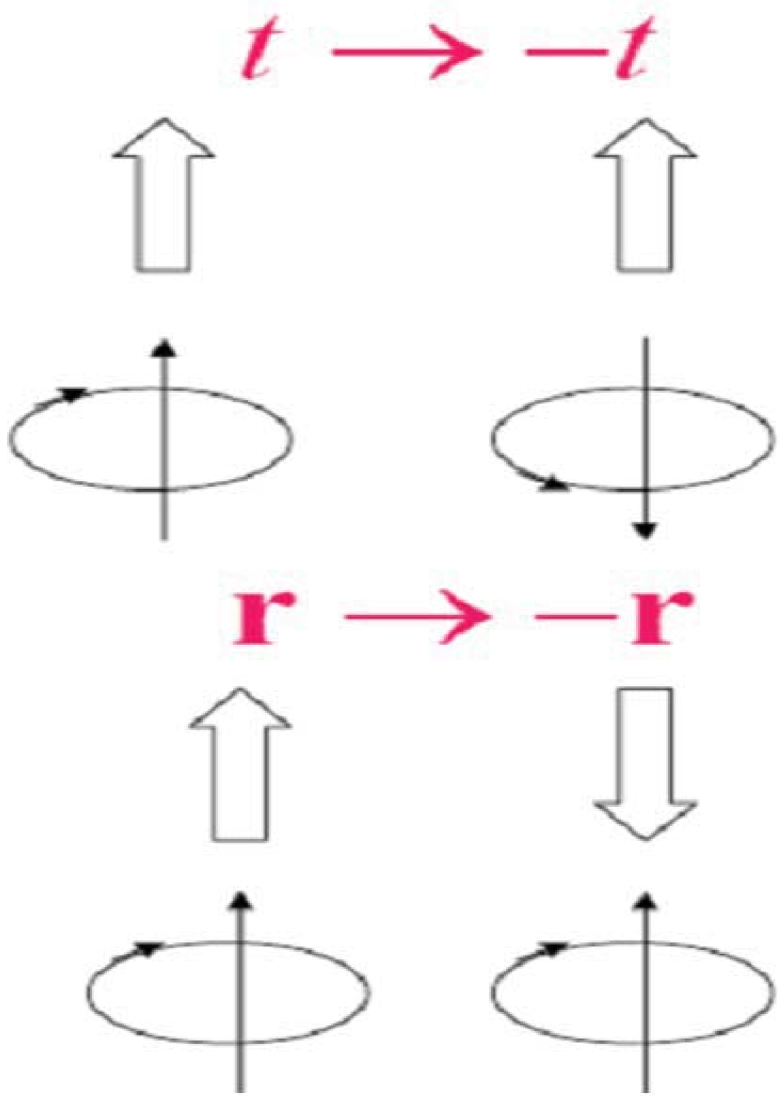
$$\vec{P} \rightarrow +\vec{P}$$

$$\vec{M} \rightarrow -\vec{M}$$

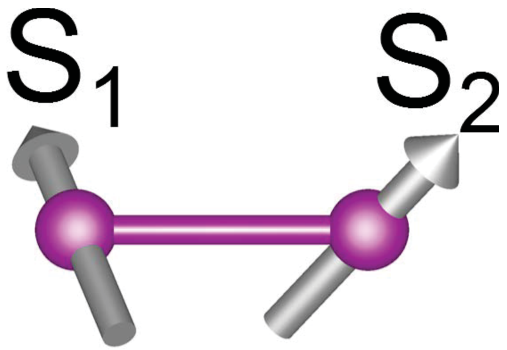
Spatial Inversion

$$\vec{P} \rightarrow -\vec{P}$$

$$\vec{M} \rightarrow +\vec{M}$$



# Theory of spin order induced ferroelectricity




$$\mathbf{P} = \mathbf{P}(\mathbf{S}_1, \mathbf{S}_2) = \mathbf{P}(S_{1x}, S_{1y}, S_{1z}, S_{2x}, S_{2y}, S_{2z})$$

time-reversal symmetry:
only even order

$$\mathbf{P} = \mathbf{P}_1(\mathbf{S}_1) + \mathbf{P}_2(\mathbf{S}_2) + \mathbf{P}_{12}(\mathbf{S}_1, \mathbf{S}_2)$$

2nd order

**Single-site**

$$\mathbf{P}_i(\mathbf{S}_i) = \sum_{\alpha\beta} \mathbf{P}_i^{\alpha\beta} S_{i\alpha} S_{i\beta}$$

**Intersite**

$$\mathbf{P}_{12}(\mathbf{S}_1, \mathbf{S}_2) = \sum_{\alpha\beta} \mathbf{P}_{12}^{\alpha\beta} S_{1\alpha} S_{2\beta}$$

H. J. Xiang*, E. Kan, Y. Zhang, M.-H. Whangbo*, and X. G. Gong*, Phys. Rev. Lett. **107**, 157202 (2011);
X. Z. Lu, M.-H. Whangbo, S. Dong, X. G. Gong, and H. J. Xiang*, Phys. Rev. Lett. **108**, 187204 (2012) (Editors' suggestion)

# Single-site term

$$\boldsymbol{P}_s(\boldsymbol{S}) = \boldsymbol{P}_{xx}S_x^2 + \boldsymbol{P}_{yy}S_y^2 + \boldsymbol{P}_{zz}S_z^2 + 2\boldsymbol{P}_{xy}S_xS_y + 2\boldsymbol{P}_{xz}S_xS_z + 2\boldsymbol{P}_{yz}S_yS_z$$

$$= (S_x, S_y, S_z)\begin{pmatrix} \boldsymbol{P}_{xx} & \boldsymbol{P}_{xy} & \boldsymbol{P}_{xz} \\ \boldsymbol{P}_{yx} & \boldsymbol{P}_{yy} & \boldsymbol{P}_{yz} \\ \boldsymbol{P}_{zx} & \boldsymbol{P}_{zy} & \boldsymbol{P}_{zz} \end{pmatrix}\begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix} = S^t P_M S.$$

- Spin orbit coupling effect
- If magnetic ion is at the center of the spatial inversion symmetry, single site term is zero

Chenglong Jia *et al.*, Phys. Rev. B **74**, 224444 (2006)

# Decomposition of the intersite term

$$\mathbf{P}_{12}(\mathbf{S}_1,\mathbf{S}_2)=\sum_{\alpha\beta}\mathbf{P}_{12}^{\alpha\beta}S_{1\alpha}S_{2\beta}$$

$$=(S_{1x},S_{1y},S_{1z})\begin{pmatrix}\mathbf{P}_{12}^{xx} & \mathbf{P}_{12}^{xy} & \mathbf{P}_{12}^{xz}\\ \mathbf{P}_{12}^{yx} & \mathbf{P}_{12}^{yy} & \mathbf{P}_{12}^{yz}\\ \mathbf{P}_{12}^{zx} & \mathbf{P}_{12}^{zy} & \mathbf{P}_{12}^{zz}\end{pmatrix}\begin{pmatrix}S_{2x}\\ S_{2y}\\ S_{2z}\end{pmatrix}$$

$$=\mathbf{S}_1^{T}\mathbf{P}_{\mathbf{int}}\mathbf{S}_2$$

$$\boldsymbol{P}_{int}=\boldsymbol{P}_J+\boldsymbol{P}_D+\boldsymbol{P}_\Gamma$$

Similar to exchange interaction

$\boldsymbol{P}_J$: Isotropic symmetric diagonal matrix

$\boldsymbol{P}_D$: Antisymmetric matrix

$\boldsymbol{P}_\Gamma$: Anisotropic symmetric matrix

# Spin orbit coupling: the intersite term

No spin-orbit coupling
rotating all spins do not change the state and **P**

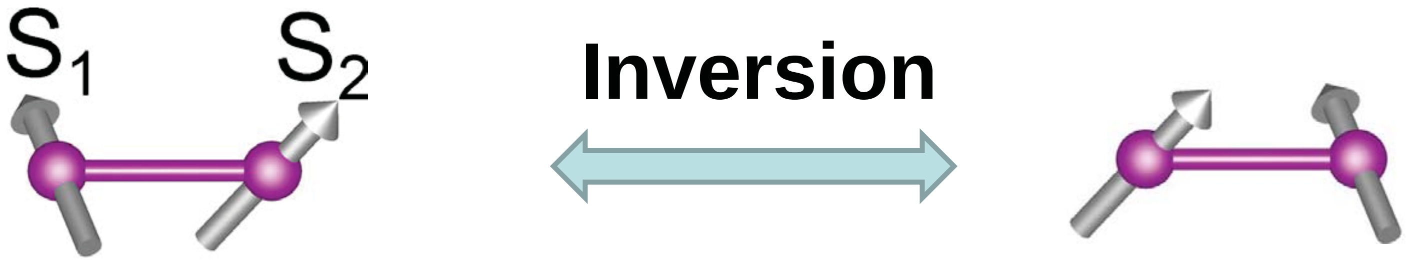
$$\mathbf{P}_{12}^{xx} = \mathbf{P}_{12}^{yy} = \mathbf{P}_{12}^{zz}, \text{ other} = 0$$

Only exchange striction

$$\mathbf{P}_{12}(\mathbf{S}_1, \mathbf{S}_2) = \mathbf{P}_{\text{es}}(\mathbf{S}_1 \cdot \mathbf{S}_2) \qquad \mathbf{P}_{\text{es}} = \mathbf{P}_{\mathbf{J}}^{\mathbf{11}} = \mathbf{P}_{\mathbf{J}}^{\mathbf{22}} = \mathbf{P}_{\mathbf{J}}^{\mathbf{33}}$$

Spin-orbit coupling: nonzero $\boldsymbol{P}_D$ and $\boldsymbol{P}_\Gamma$
Usually $\boldsymbol{P}_J$ is much larger than $\boldsymbol{P}_D$ and $\boldsymbol{P}_\Gamma$,
and $\boldsymbol{P}_D \gg \boldsymbol{P}_\Gamma$

# Spatial inversion: the intersite term




I: $\mathbf{S}_1 = \mathbf{S}$ and $\mathbf{S}_2 = \mathbf{S}'$

II: $\mathbf{S}_1 = \mathbf{S}'$ and $\mathbf{S}_2 = \mathbf{S}$

$$\boldsymbol{P}_I = -\boldsymbol{P}_{II}$$

$$\sum_{\alpha\beta} \mathbf{P}_{12}^{\alpha\beta} S_\alpha S'_\beta = -\sum_{\alpha\beta} \mathbf{P}_{12}^{\alpha\beta} S'_\alpha S_\beta$$

$$\mathbf{P}_{12}^{\alpha\beta} = -\mathbf{P}_{12}^{\beta\alpha};\ \mathbf{P}_{12}^{\alpha\alpha} = 0$$

**No exchange striction!**

$$\mathbf{P}_{12} = \mathbf{P}_{12}^{yz}\ (\mathbf{S}_1 \times \mathbf{S}_2)_x + \mathbf{P}_{12}^{zx}\ (\mathbf{S}_1 \times \mathbf{S}_2)_y + \mathbf{P}_{12}^{xy}\ (\mathbf{S}_1 \times \mathbf{S}_2)_z$$

**Generalized KNB**

$$\mathbf{P}_{12} = \mathbf{S}_1^T \mathbf{P}_D \mathbf{S}_2 = \mathbf{M}\ (\mathbf{S}_1 \times \mathbf{S}_2)$$

$$\mathbf{M} = \begin{bmatrix} (\mathbf{P}_{12}^{yz})_x & (\mathbf{P}_{12}^{zx})_x & (\mathbf{P}_{12}^{xy})_x \\ (\mathbf{P}_{12}^{yz})_y & (\mathbf{P}_{12}^{zx})_y & (\mathbf{P}_{12}^{xy})_y \\ (\mathbf{P}_{12}^{yz})_z & (\mathbf{P}_{12}^{zx})_z & (\mathbf{P}_{12}^{xy})_z \end{bmatrix}$$

# General polarization model

Single-site term | Exchange striction | GKNB

$$P_t = \sum_{i,\alpha\beta} \mathbf{P}_{i,\alpha\beta} S_{i\alpha} S_{i\beta} + \sum_{<i,j>} \mathbf{P}_{es}^{i,j} \vec{S}_i \cdot \vec{S}_j + \sum_{<i,j>} \boldsymbol{M}_{ij} (\vec{S}_i \times \vec{S}_j)$$

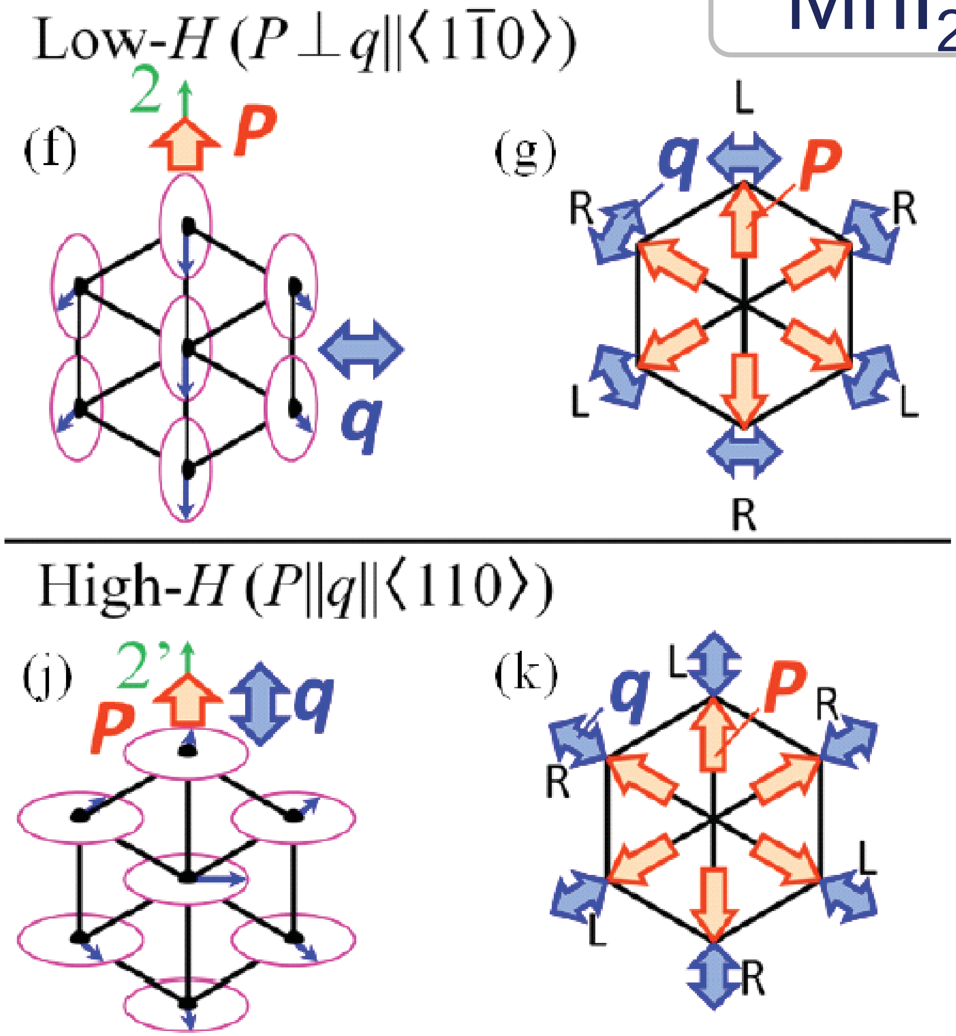
$$\hat{H}_{spin} = \sum_{i,\alpha\beta} A_{i,\alpha\beta} S_{i\alpha} S_{i\beta} + \sum_{<i,j>} J_{ij} \vec{S}_i \cdot \vec{S}_j + \sum_{<i,j>} \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$$

Single-ion anisotropy | Symmetric exchange | DM interaction

# Spin Hamiltonian

# Application: FE induced by screw spiral

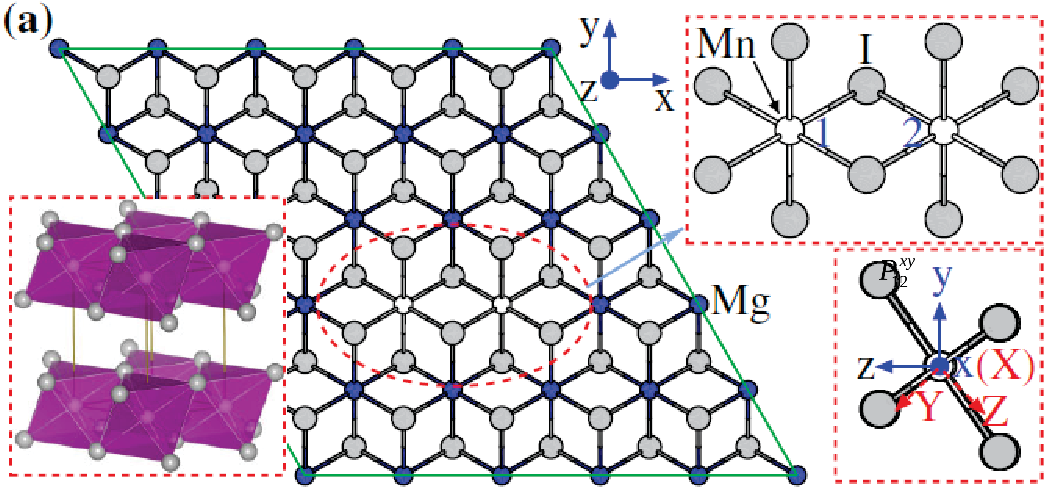
$MnI_2$




- Low-H: ***P*⊥*q***
- High-H:***P*//*q*,** ***P*(q)=-*P*(-*q*)**
- Same as $CuFeO_2$ and $CuCrO_2$
- Not explained by KNB model and exchange striction

T. Kurumaji *et al.*, Phys. Rev. Lett. **106**, 167206 (2011)

# Estimate the coefficients: method-I




- 5x5x1 supercell
- DFT+U+SOC
- **P:** Berry phase

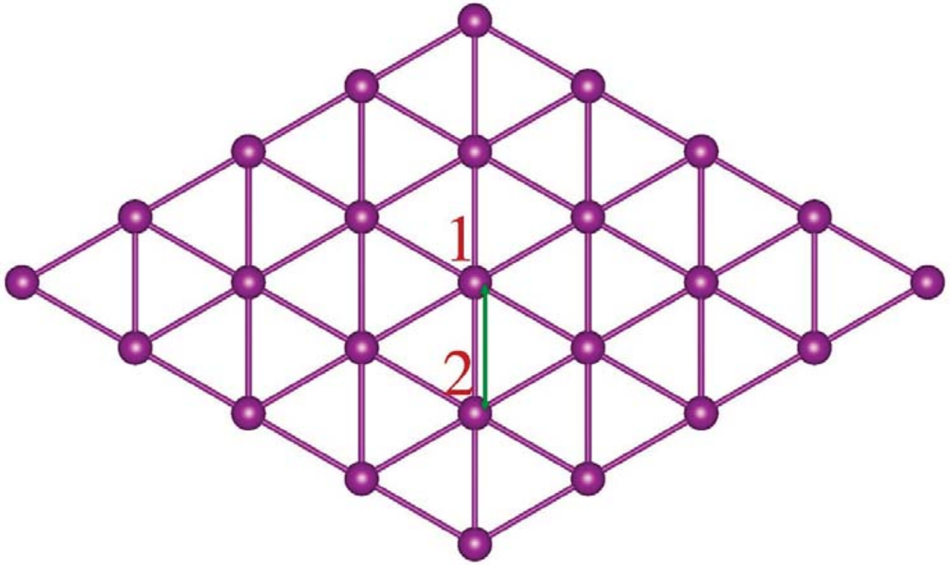
King-Smith and Vanderbilt, PRB **47**, 1651 (1993); Resta, RMP **66**, 899 (1994).

**Nonmagnetic substitution**

| | $Mn_1$ spin | $Mn_2$ spin | other Mn ions |
|---|---|---|---|
| **A** | (1, 0, 0) | (0, 1, 0) | Mg |
| **B** | (1, 0, 0) | (0, -1, 0) | Mg |

$$P_{12}^{xy} = (\mathbf{P}_A - \mathbf{P}_B)/2$$

# Estimate the coefficients: method-II




**Four-state mapping**

- Non-substitution
- More accurate
- General
- Give $\frac{\partial J}{\partial \vec{u}}, \frac{\partial \vec{D}}{\partial \vec{u}}$ etc

| | $S_1$ | $S_2$ | other spins |
|---|---|---|---|
| **A** | (1, 0, 0) | (0, 1, 0) | (0, 0, 1) |
| **B** | (1, 0, 0) | (0, -1, 0) | (0, 0, 1) |
| **C** | (-1, 0, 0) | (0, 1, 0) | (0, 0, 1) |
| **D** | (-1, 0, 0) | (0, -1, 0) | (0, 0, 1) |

$$H_{DM} = \vec{D} \cdot (\vec{S}_i \times \vec{S}_j): \quad D_z^{12} = [E(A) + E(D) - E(B) - E(C)]/(4S^2)$$

$$P_{12}^{xy} = (\mathbf{P}_A + \mathbf{P}_D - \mathbf{P}_B - \mathbf{P}_C)/4$$

H. J. Xiang *et al.*, Phys. Rev. B **84**, 224429 (2011)

# Coefficients from DFT

**Single-site** $\mathbf{P}_1^{xx} = (0, 0, 0)$ $\mathbf{P}_1^{yy} = (2.5, 0, 0)$ $\mathbf{P}_1^{zz} = (-2.5, 0, 0)$

in units of $10^{-6}$ eÅ.

**Intersite**

$$\mathbf{M} = \begin{bmatrix} M_{11} & 0 & 0 \\ 0 & M_{22} & M_{23} \\ 0 & M_{32} & M_{33} \end{bmatrix}$$

Intersite polarization: much larger than single-site polarization

$M_{11}$ = -4.8, $M_{22}$ = 39.5
$M_{23}$ = 49.0, $M_{32}$ = -44.5
$M_{33}$ = -26.0

in units of $10^{-5}$ eÅ

# Intersite term vs. KNB model

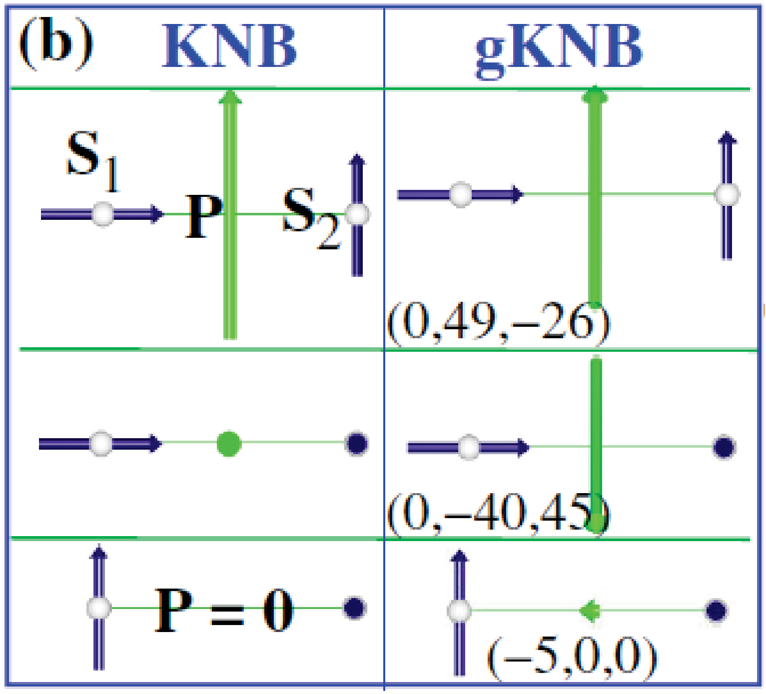
## Intersite term from DFT

$$\mathbf{M} = \begin{bmatrix} M_{11} & 0 & 0 \\ 0 & M_{22} & M_{23} \\ 0 & M_{32} & M_{33} \end{bmatrix}$$

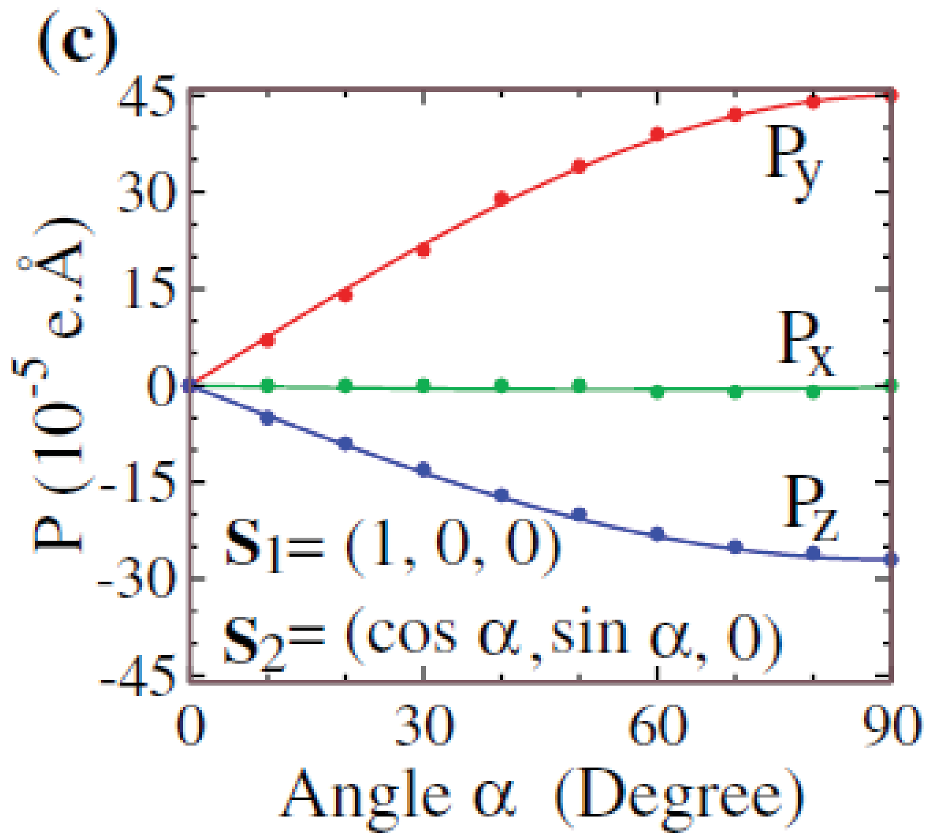
## KNB model

$$\mathbf{M} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & M_{23} = -C \\ 0 & M_{32} = C & 0 \end{bmatrix}$$

**gKNB**

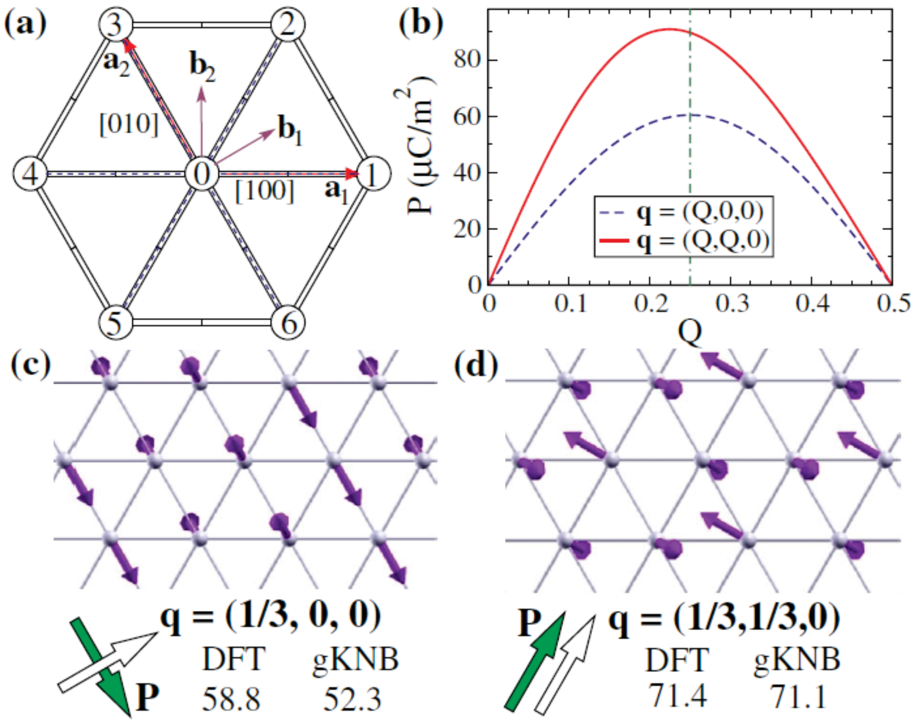


# Compare gKNB to DFT




- **P** as a function of the angle between the spins $\mathbf{S}_1$ and $\mathbf{S}_2$. Points: direct DFT Solid: gKNB model.
- Agreement, so higher order terms are unimportant

# Screw spin spiral: gKNB model




- gKNB: **P** ⊥ **q** for **q**=(Q,0,0), but **P** || **q** for **q**=(Q,Q,0)
- **P(-q) = -P(q)**
- $M_{11}$ and $M_{22}$ is zero in KNB
- gKNB agrees with DFT for proper screw spiral

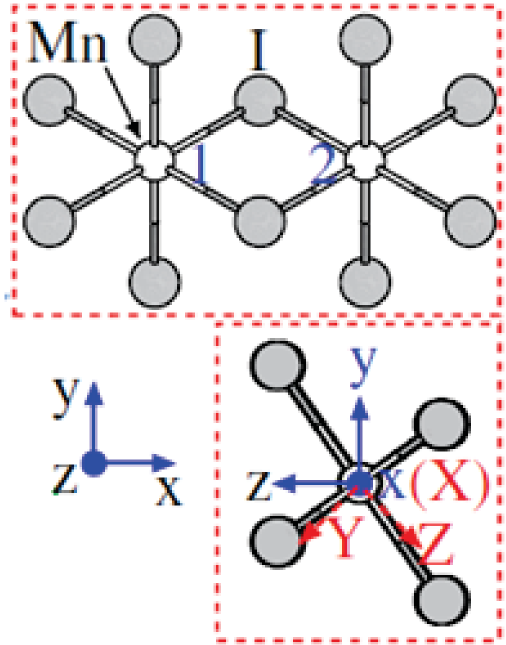
$$\mathbf{P}_i^{tot} = \sum_{k=1}^{6} \mathbf{P}_{0k} = \sum_{k=1}^{6} \mathbf{M}^{0k} (\mathbf{S}_0 \times \mathbf{S}_k)$$

$$\mathbf{q} = (\mathrm{Q},\ 0,\ 0): \quad \mathbf{P}_0^{tot} = (\tfrac{\sqrt{3}}{2}\mathrm{A},\ -\tfrac{3}{2}\mathrm{A},\ 0)$$

$$\mathbf{q} = (\mathrm{Q},\ \mathrm{Q},\ 0): \quad \mathbf{P}_0^{tot} = (\tfrac{1}{2}\mathrm{B},\ \tfrac{\sqrt{3}}{2}\mathrm{B},\ 0)$$

# Tight-binding v.s. DFT

**DFT**



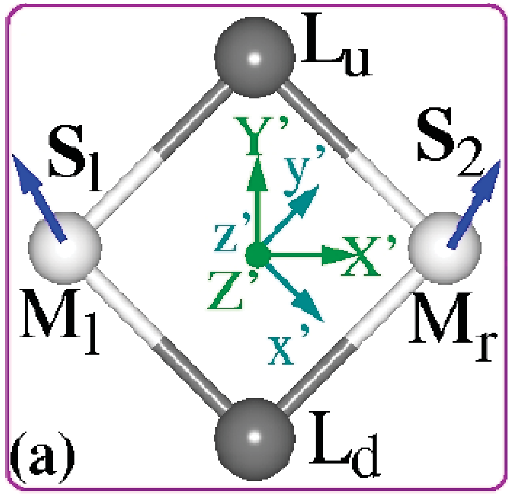

KNB model:

$$(P_{12}^{X'Y'})_{Y'} = -(P_{12}^{Z'X'})_{Z'}$$

SOC: essential

Polarization anisotropy:
Structural anisotropy

**TB**




$D_{2h}$ symmetry

X'Y'Z'

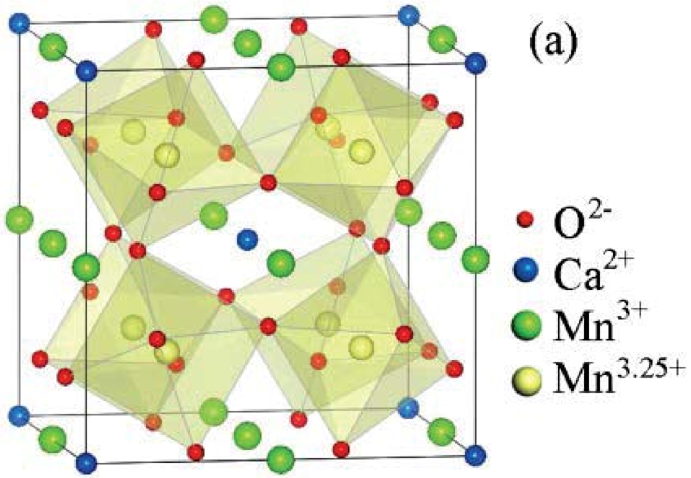
XYZ

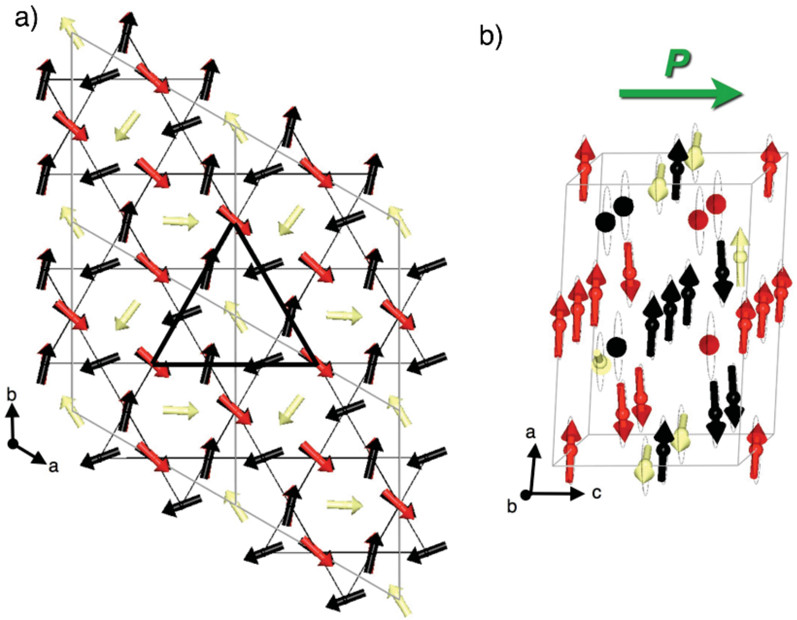
$$\mathbf{M} = \begin{bmatrix} -4.8 & 0 & 0 \\ 0 & 6.8 & 79.6 \\ 0 & -13.9 & 6.8 \end{bmatrix}$$

$$\mathbf{M} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & (\mathbf{P}_{12}^{X'Y'})_{Y'} \\ 0 & (\mathbf{P}_{12}^{Z'X'})_{Z'} & 0 \end{bmatrix}$$

$$(P_{12}^{X'Y'})_{Y'} \gg -(P_{12}^{Z'X'})_{Z'}$$

# Application: Giant polarization in $CaMn_7O_{12}$

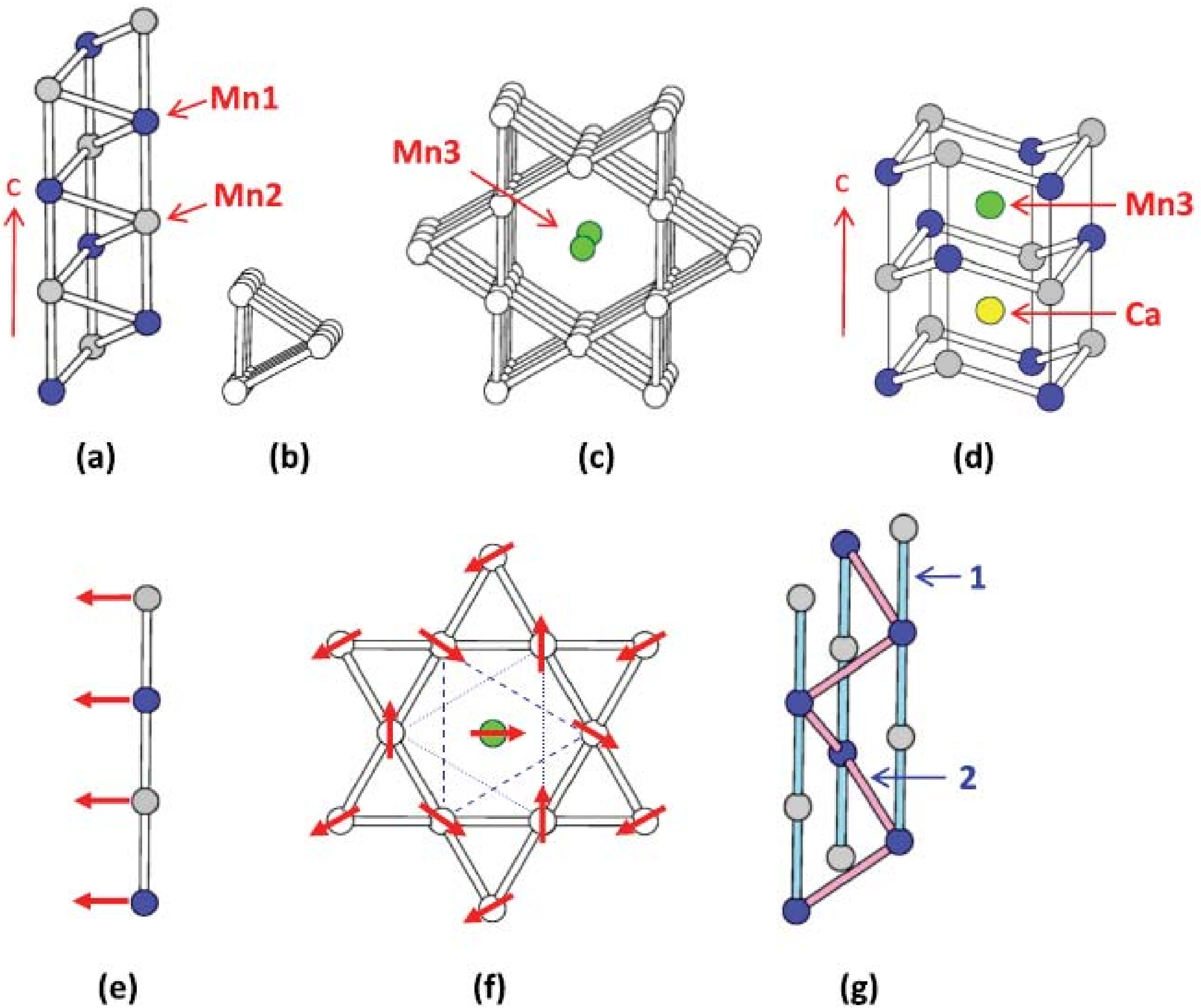


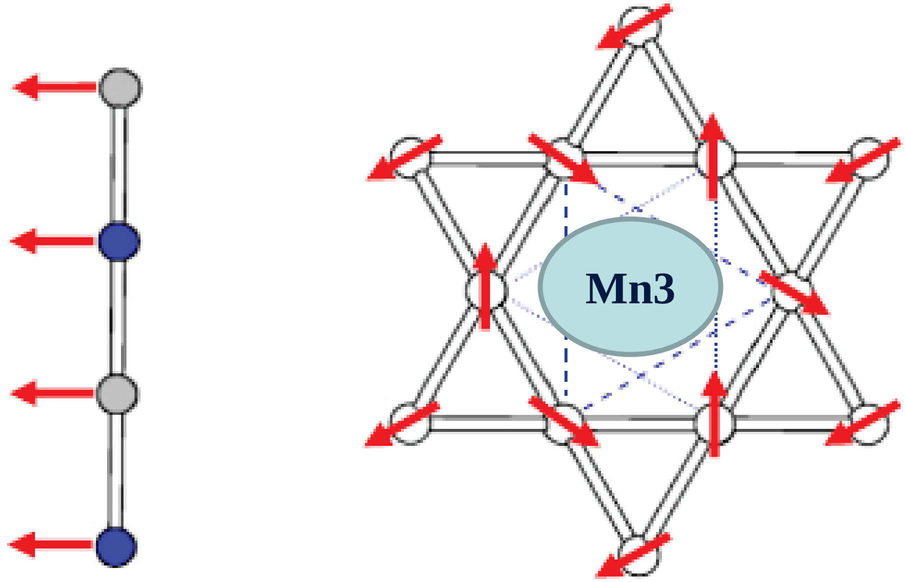

- Mn1(+3): A sites
- B site CO below 440 K: $4Mn^{3.25+} \rightarrow 3Mn^{3+}(Mn2) + Mn^{4+}(Mn3)$
- $T_N = 90$ K, FM chains along c and frustration between them
- Giant polarization: 2870 μC/m²

Zhang *et al*., Phys. Rev. B **84**, 174413 (2011);
Johnson *et al*., Phys. Rev. Lett. **108**, 067201 (2012).

# Detailed experimental magnetic structures



# Spin arrangements of Mn1 and Mn2 ions

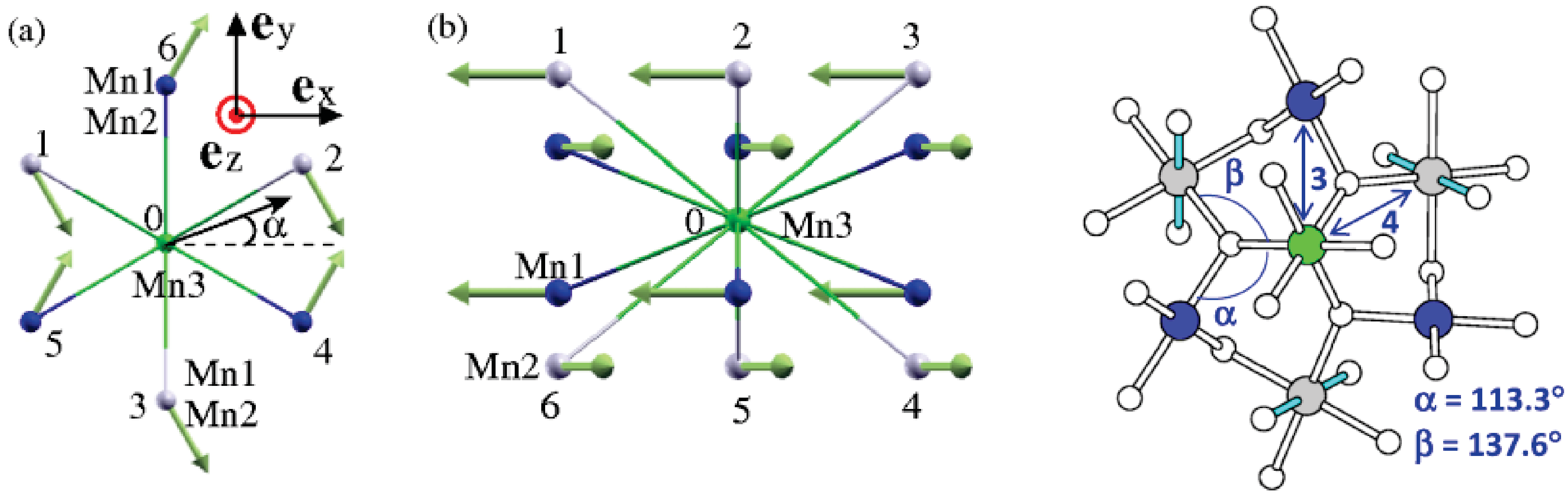



- Intrachain exchange $J_1$ between Mn1 and Mn2: strongly FM (-5.57 meV)

- Inter-chain exchange $J_2$ between Mn1 ions: strongly AFM (6.37 meV) due to Mn-O…O-Mn super-SE

- Explain the FM chain and frustration between them

# Preferred spin direction of Mn3 ion

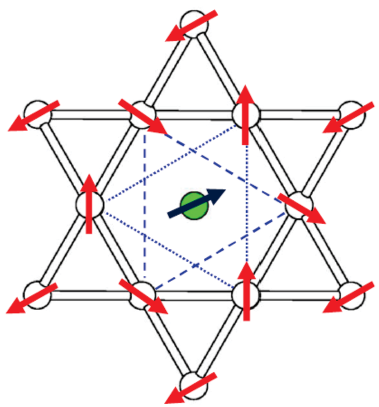



(c) $J_3$ and $J_4$

Spin exchange interaction energy:

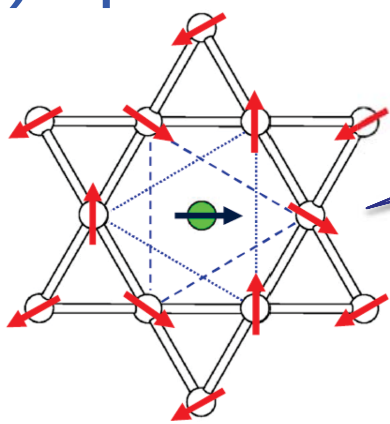
$$E_{SE} = 3(J_3 + J_4)\cos\alpha, \qquad \alpha = 0 \qquad (J_3 + J_4 < 0)$$

(60,60) spin arrangement



?

(30,90) spin arrangement

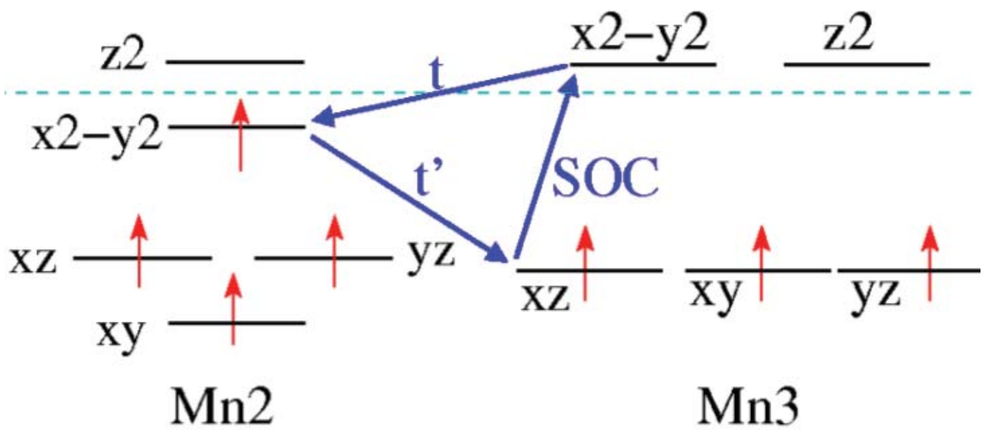


EXP

# Extraordinarily large DM interaction

| Path | Pair | J (meV) | D (meV) | \|D/J\| |
|---|---|---|---|---|
| **4** | Mn2-Mn3 | -2.961 | (-0.064, 0.862, 1.361) | 54% |

H. J. Xiang *et al.*, Phys. Rev. B **84**, 224429 (2011)




$$E_{tot} = E_{SE} + E_{DM}$$
$$= 3(J_3 + J_4)\cos\alpha + 3D_4^z \sin\alpha$$

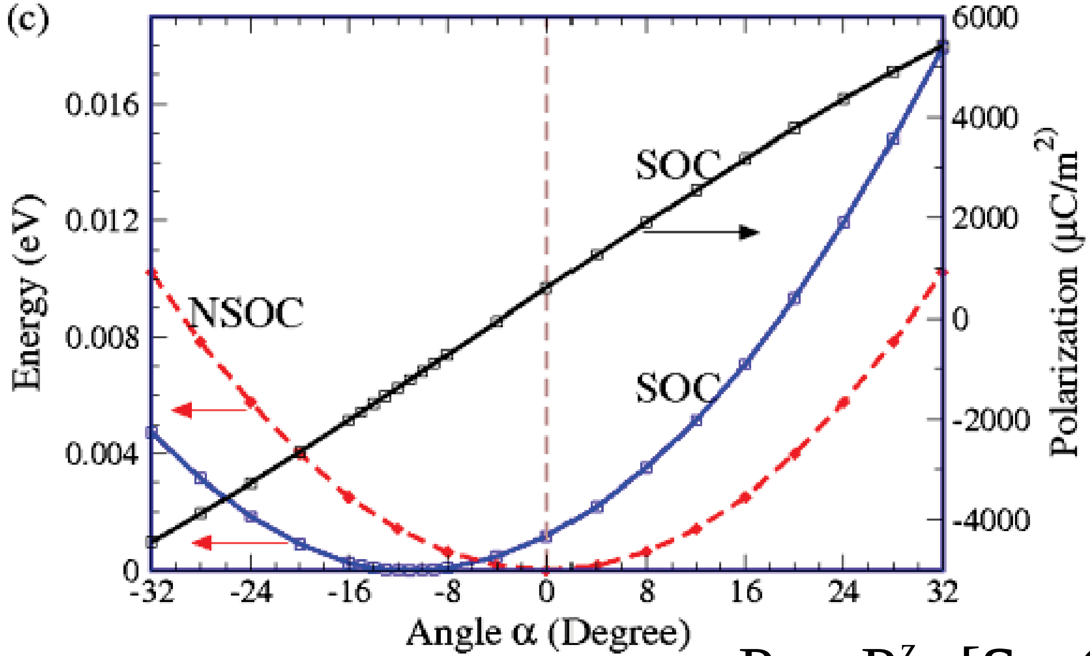
$$\alpha_m = \arctan[D_4^z / (J_3 + J_4)]$$

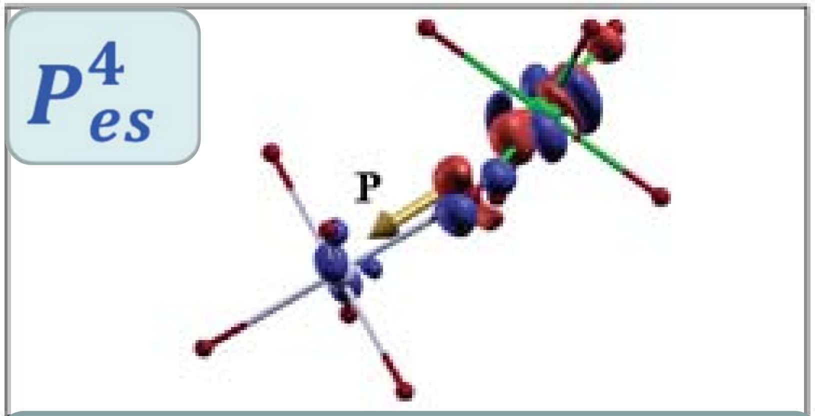
DFT+SOC+U (U= 3 eV): $\alpha_m$ = -11°

DFT+SOC+U (U= 2 eV): $\alpha_m$ = -36°

EXP: $\alpha_m \approx$ -30°

# Origin of giant polarization

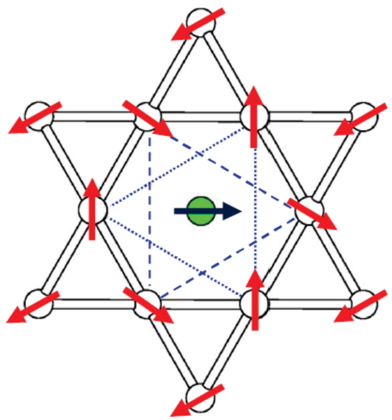



Electron density difference between FM and AFM ($J_4$)

$$P_z = P^z_{es,4}[\mathbf{S}_0 \cdot (\mathbf{S}_1 - \mathbf{S}_4 + \mathbf{S}_2 - \mathbf{S}_5 + \mathbf{S}_3 - \mathbf{S}_6)]$$

$$= 3\sqrt{3}P^z_{es,4}(\sin\alpha)\,\mathrm{sign}[\mathbf{e_z} \cdot (\mathbf{S_4} \times \mathbf{S_1})]$$

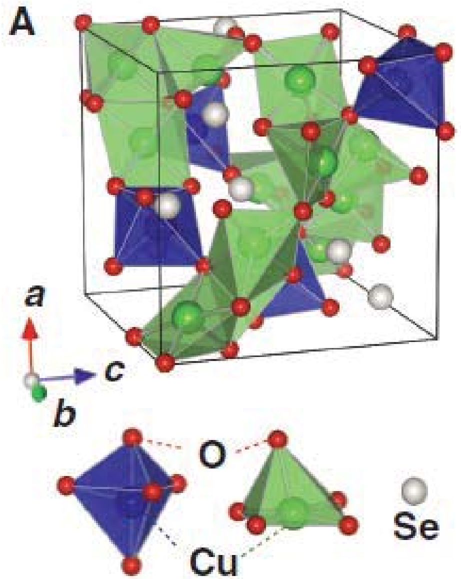


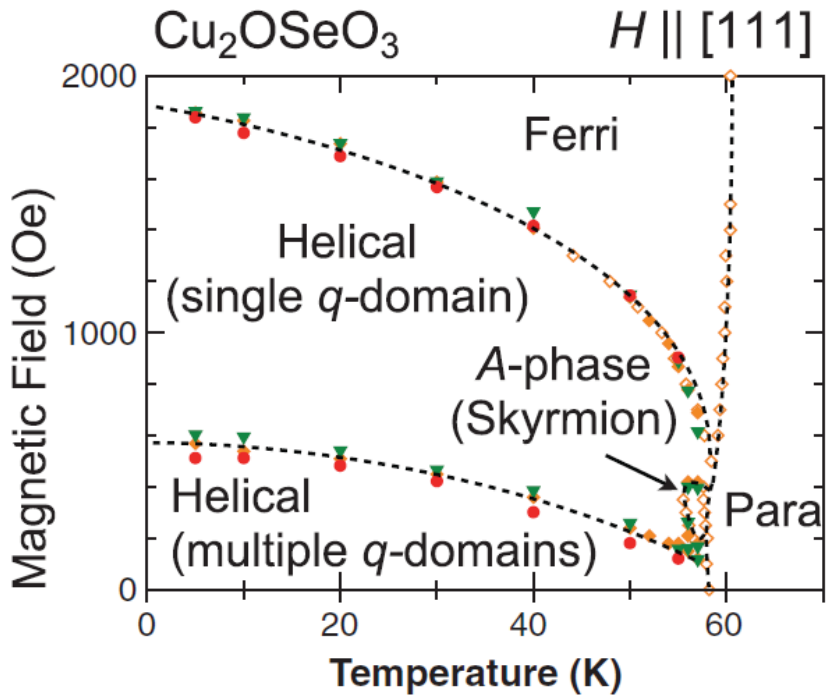
**DM** induces canting of Mn3 spins, resulting in polarization due to **exchange stricition**

**Not ferroaxial mechanism!**

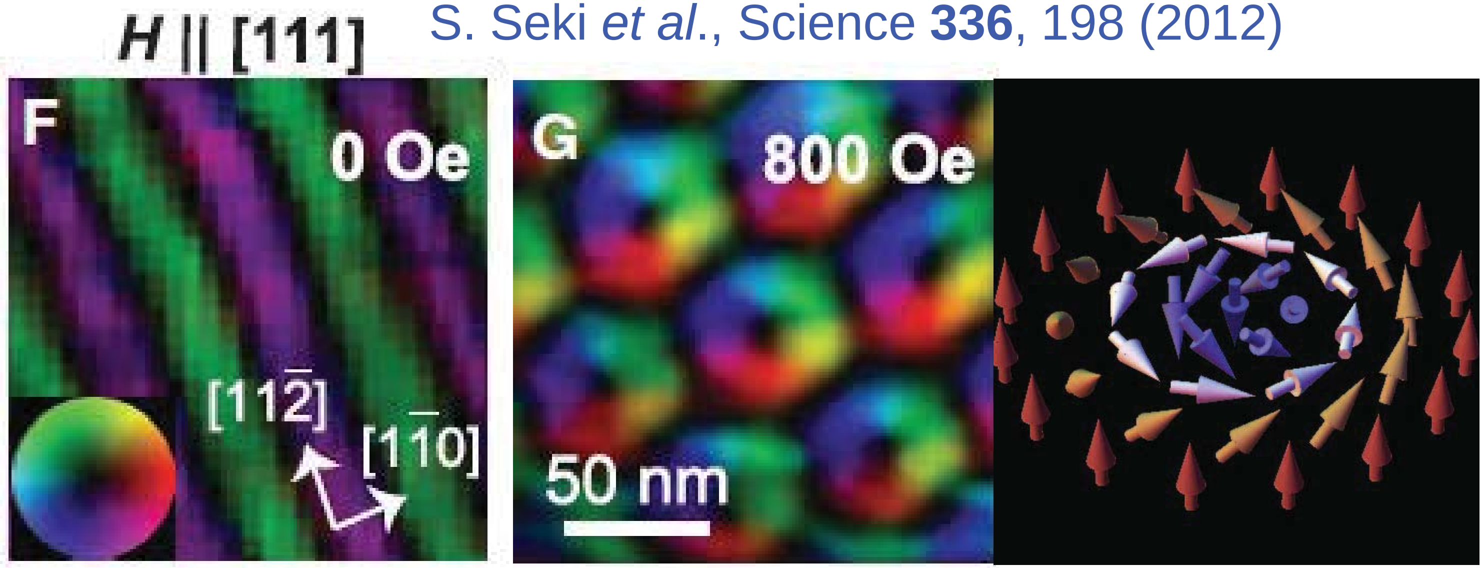
Lu, Xiang *et al.*, Phys. Rev. Lett. 108, 187204 (2012)

# Application: Insulating skyrmion crystal $Cu_2OSeO_3$

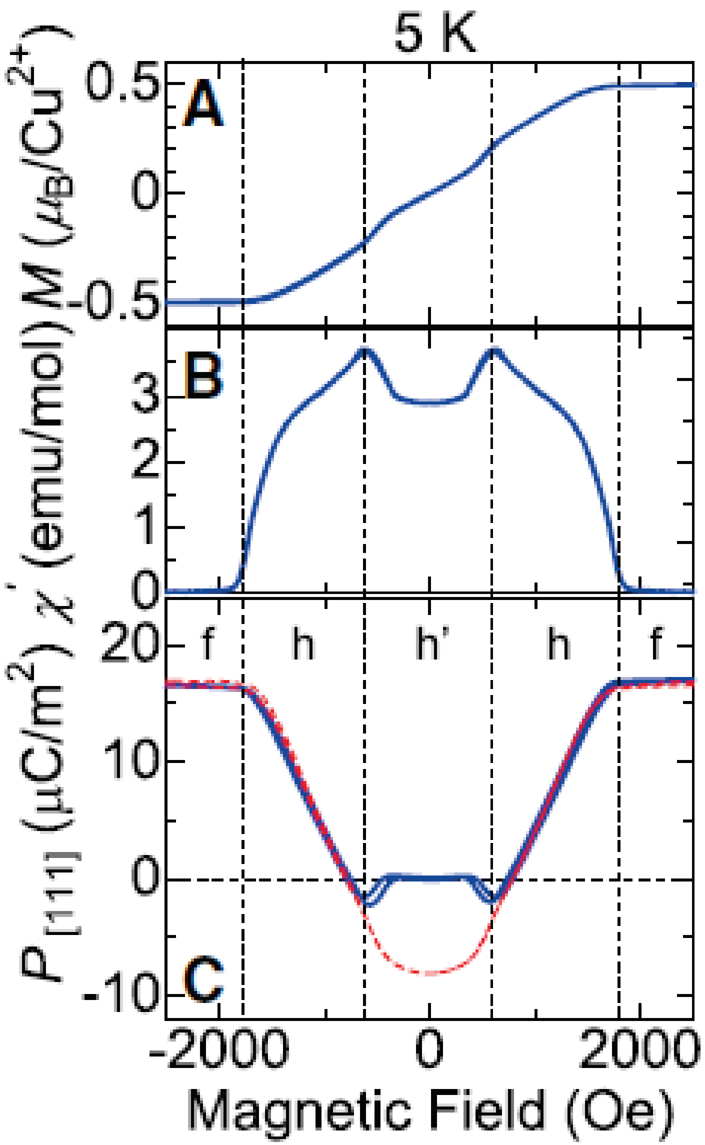


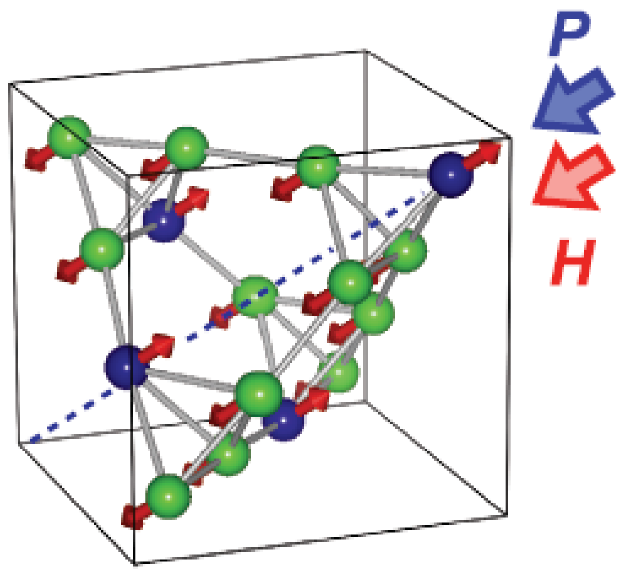

S. Seki *et al*., Science **336**, 198 (2012)



# Ferroelectric properties of $Cu_2OSeO_3$



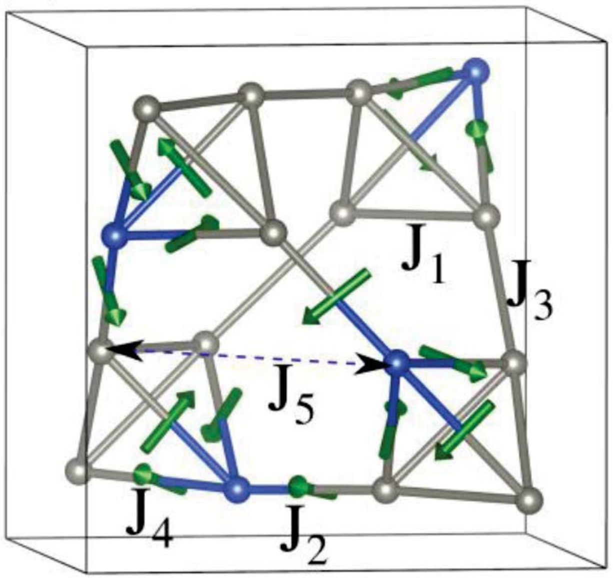

H || [111]

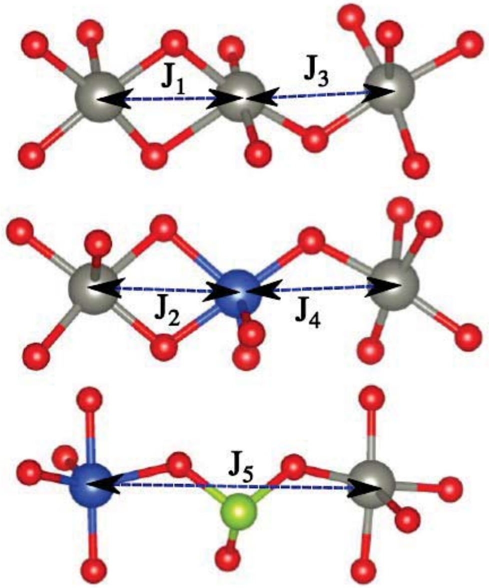



- Helical, skyrmion-crystal, and ferrimagnetic states are ferroelectric

- $P_{[111]} = P_0 + \beta M^2$

# Magnetic properties: Theory




- Ferrimagnetism
- Strong DM interaction: helical and skyrmion-crystal

| Path | Distance (Å) | J (meV) | D (meV) | \|D/J\| |
|---|---|---|---|---|
| 1: II-II | 3.039 | -1.132 | (0.289,-0.325,-0.051) | 0.39 |
| 2: I-II | 3.057 | 6.534 | (-1.12,1.376,-0.300) | 0.28 |
| 3: II-II | 3.220 | -3.693 | (-0.263,0.167,-0.407) | 0.14 |
| 4: I-II | 3.300 | 0.900 | (0.490,-1.238,-1.144) | 1.95 |
| 5: I-II | 6.352 | 0.984 | (0.059,0.045,0.087) | 0.12 |

# Ferroelectricity of the ferrimagnetic state

$$\boldsymbol{P}_t = \sum_i \boldsymbol{P}_{\boldsymbol{s}}(\boldsymbol{S}_i) + \sum_{<ij>} \boldsymbol{P}_p(\boldsymbol{S}_i, \boldsymbol{S}_j) + \cdots$$

Collinear ferrimagnetic state

- Intersite terms (exchange striction and GKNB) are zero
- Single-site term might be non-zero by symmetry

$$\boldsymbol{P}_s(\boldsymbol{S}) = (S_x, S_y, S_z) \begin{pmatrix} \boldsymbol{P}_{xx} & \boldsymbol{P}_{xy} & \boldsymbol{P}_{xz} \\ \boldsymbol{P}_{yx} & \boldsymbol{P}_{yy} & \boldsymbol{P}_{yz} \\ \boldsymbol{P}_{zx} & \boldsymbol{P}_{zy} & \boldsymbol{P}_{zz} \end{pmatrix} \begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix}$$

$$= S^t P_M S$$

Yang, Gong, Xiang *et al.*, Phys. Rev. Lett. **109**, 107203 (2012)

# Single-site term from DFT

Single-site term for Cu-I is much larger than Cu-II

$$P_M = \begin{bmatrix} (126,38,148) & (38,-126,0) & (3,7,0) \\ (38,-126,0) & (-126,-38,148) & (-8,3,0) \\ (3,7,0) & (-8,3,0) & (0,0,-296) \end{bmatrix} \times 10^{-6}\ \text{eÅ}$$

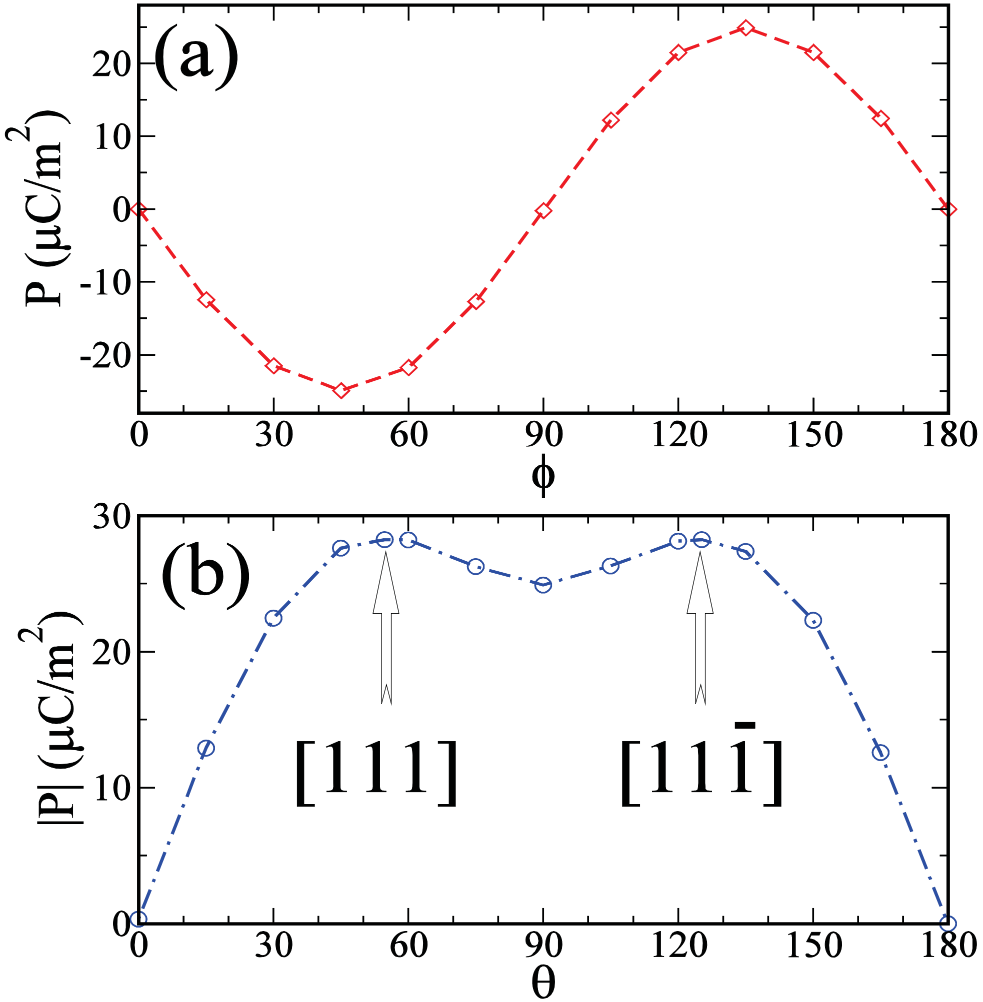
Ferrimagnetic state with spin axis $\boldsymbol{h} = (h_x, h_y, h_z)$

$$\boldsymbol{P}_t = \sum_i \boldsymbol{P}_s(\boldsymbol{S}_i)$$

$$= A(h_y h_z \boldsymbol{e}_x + h_x h_z \boldsymbol{e}_y + h_x h_y \boldsymbol{e}_z)$$

$(A = -40.00\ \mu\text{C/m}^2)$

# Compare model to DFT results




J. H. Yang, Gong*, Xiang* *et al.*,
Phys. Rev. Lett. **109**, 107203 (2012)

(a) $P_z$ as a function of the angle (φ) between the spin axis and the x axis (spins in the *ab*-plane).
$P_z \propto -\sin(2\varphi)$

(b) |P| as a function of the angle (θ) between the spin axis and the z axis [spins in (110) plane]. |P| is the largest when S || <111>.

**Agreement!**

# Summary

**Spin Hamiltonian**

$$\hat{H}_{spin} = \sum_{i,\alpha\beta} A_{i,\alpha\beta} S_{i\alpha} S_{i\beta} + \sum_{<i,j>} J_{ij} \vec{S}_i \cdot \vec{S}_j + \sum_{<i,j>} \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$$

| Single-ion anisotropy | Symmetric exchange | DM interaction |
|---|---|---|
| ⇕ | ⇕ | ⇕ |
| Single-site term | Exchange striction | GKNB |

$$P_t = \sum_{i,\alpha\beta} \mathbf{P}_{i,\alpha\beta} S_{i\alpha} S_{i\beta} + \sum_{<i,j>} \mathbf{P}_{es}^{i,j} \vec{S}_i \cdot \vec{S}_j + \sum_{<i,j>} \boldsymbol{M}_{ij} (\vec{S}_i \times \vec{S}_j)$$

**General polarization model**

Applications: triangular systems, $CaMn_7O_{12}$ and $Cu_2OSeO_3$

PRL **107**, 157202 (2011); PRL **108**, 187204 (2012);
PRL **109**, 107203 (2012)

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