

Helical Spin Order in SrFeO_3 and BaFeO_3

Zhi Li

Yukawa Institute for Theoretical Physics
(YITP)

Collaborator:

Robert Laskowski (Vienna Univ.)

Toshiaki Iitaka (Riken)

Takami Tohyama (YITP)

Outline

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 - Helical spin in SrFeO_3 and BaFeO_3
- 2. Motivation and Purpose
- 3. Density functional theory (DFT) and model calculation
- 4. Conclusion

Introduction

- For random spin order, the position dependent spin moment can be expressed as :

$$S_i = S(\sin \theta_i \cos \phi_i, \sin \theta_i \sin \phi_i, \cos \theta_i)$$

For helical spin order, the constraint is:

$$\theta_i = \bar{q} \cdot \bar{r}_i \quad \phi_i = 0$$

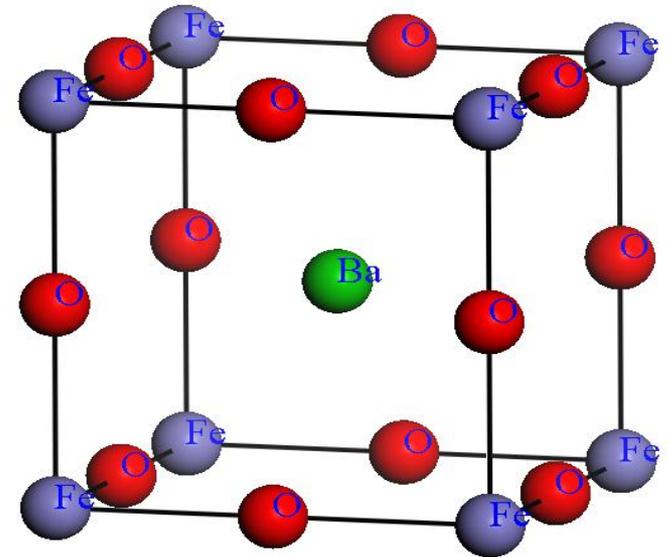
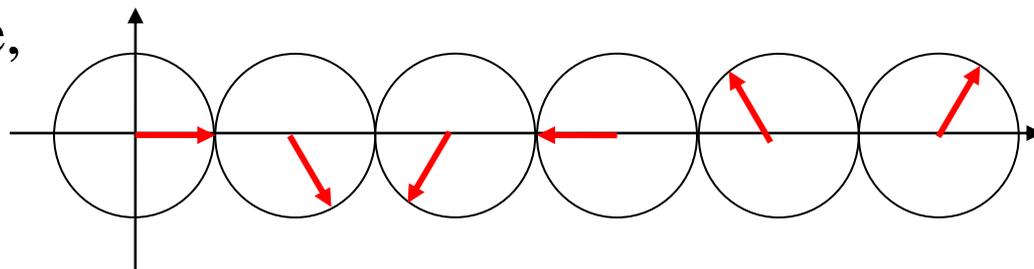
Propagating vector \bar{q} defined in reciprocal space

A-type helical spin $\bar{q} = \phi(1,0,0)$

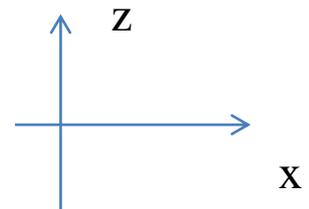
G-type helical spin $\bar{q} = \phi(1,1,1)$

For example,

A-type



crystal structure of BaFeO₃



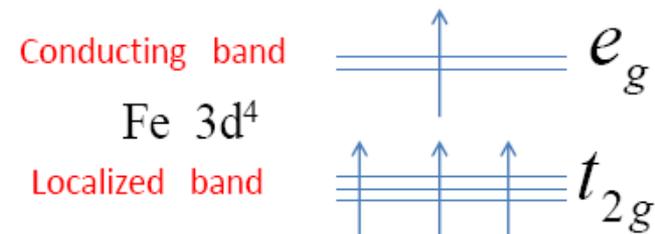
Helical spin order in AFeO₃

- Hallmark of AFeO₃ (A=Ca, Sr, Ba):

Helical spin and the energy of *p*-orbital of oxygen is lower than the energy of *d*-orbital of iron

	T _N (K)	q (2π/a)
SrFeO ₃	134	0.112(1,1,1)
BaFeO ₃	110	0.06(1,0,0)

- S. Kawasaki, et.al, **J. Phys. Soc. Jpn.** **67**, 1529 (1998)
- T. Takeda, et.al, **J. Phys. Soc. Jpn.** **33**, 967 (1972)
- N. Hayashi, et.al, **Angew. Chem. Int. Ed.** **50**, 12547 (2011)



Introduction-SrFeO₃

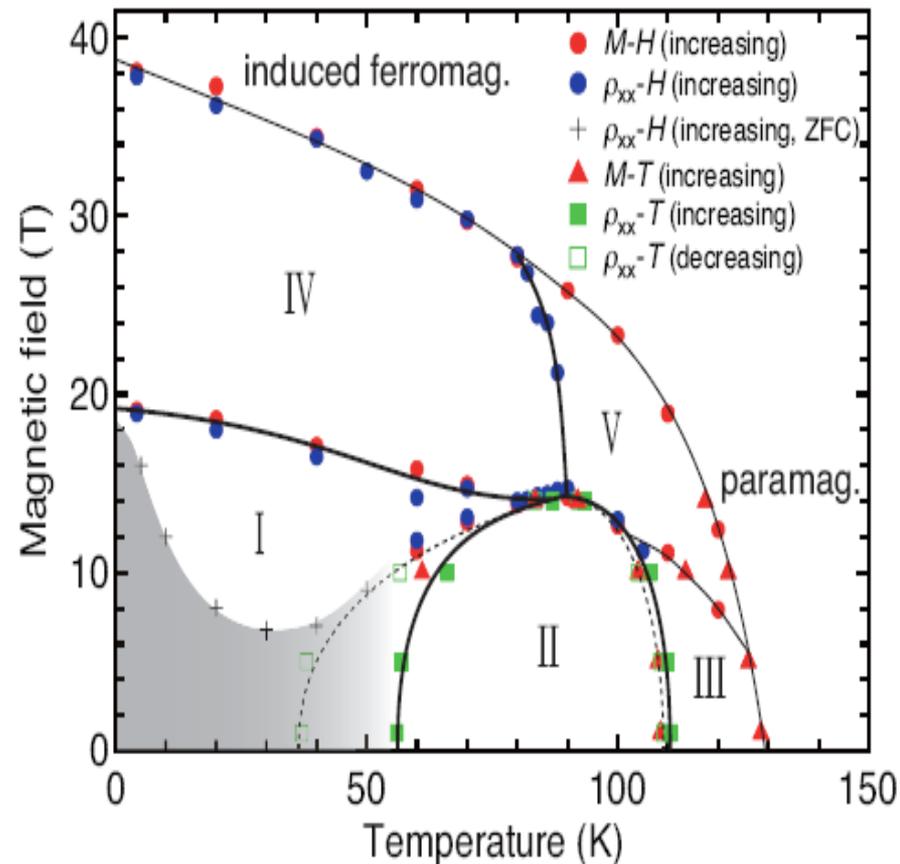
- Recent experimental result, SrFeO₃ presents more complicated phase diagram
- 40T magnetic field, ferromagnetism (FM)

- Unknown Phases

i , ii , iii , iv , v

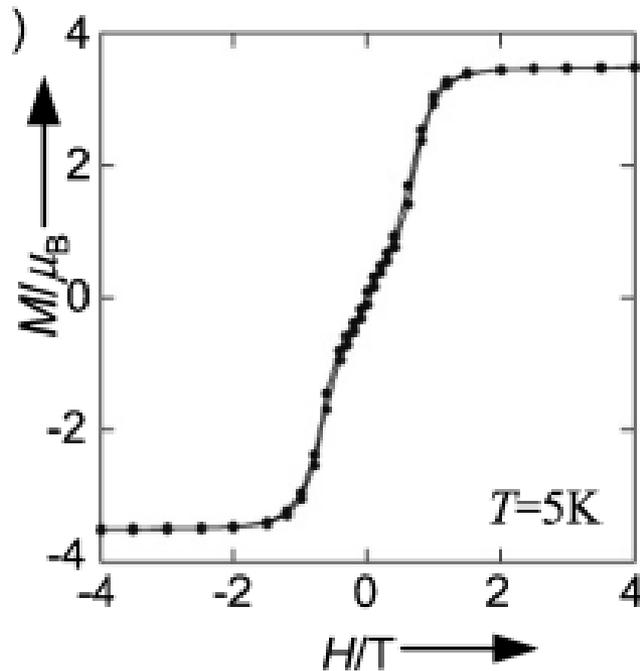
They are maybe related to the formation of domain wall, in which helical spin exists

S. Ishiwata et al, Phys. Rev. B **84**, 054427 (2011)



Introduction-BaFeO₃

- Lattice expansion.
- The size of Ba atom is larger than Sr.
- BaFeO₃ presents FM under **1T** magnetic field.



N. Hayashi, et.al, *Angew. Chem. Int. Ed.* **50**, 12547 (2011)

Model calculation

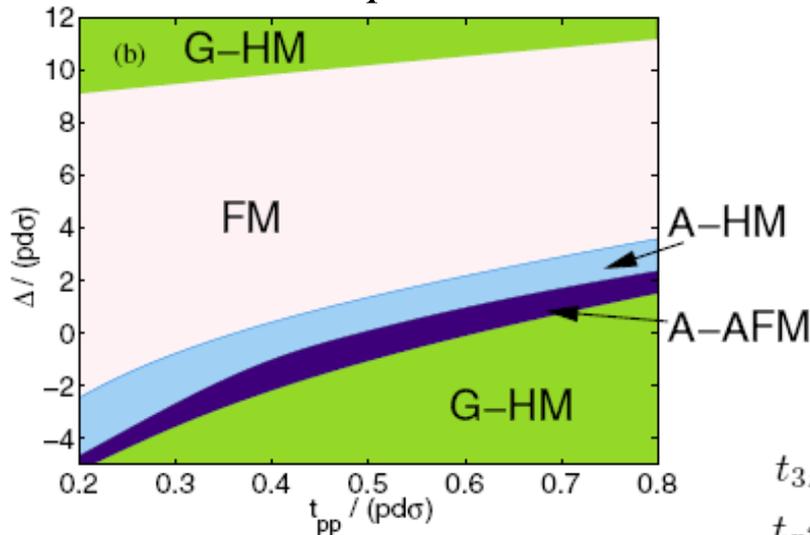
- Double exchange (favor FM) plus superexchange model (favor AFM)

$$H = H_{dp} + H_{SE}$$

$$H_{dp} = \sum_{jab} t_{ab} (d_{j\alpha}^\dagger u_j^\dagger P_{jb} + P_{jb}^\dagger u_j d_{j\alpha}) + t_{pp} \sum_{j,b \neq c} P_{jb}^\dagger P_{jc} + \Delta \sum_{jb} P_{j+b/2}^\dagger P_{j+b/2}$$

$$P_{jb} = P_{j+b/2} + P_{j-b/2}$$

$$H_{SE} = J_{SE} \sum_{\langle ij \rangle} S_i \cdot S_j \quad v_j = e^{i\frac{1}{2}\sigma_x(\mathbf{Q} \cdot \mathbf{x}_j)} \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$



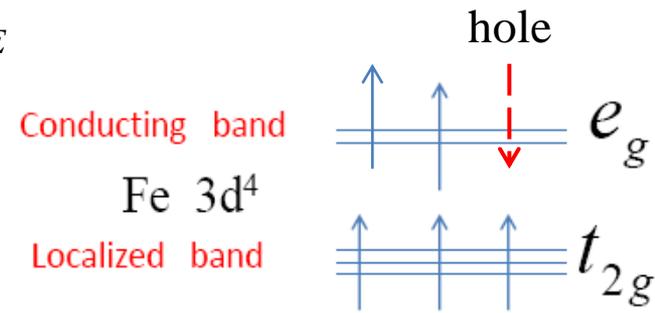
$$t_{3z^2-r^2,x} = t_{3z^2-r^2,y} = -(pd\sigma)/2, \quad t_{3z^2-r^2,z} = (pd\sigma)$$

$$t_{x^2-y^2,x} = -t_{x^2-y^2,y} = \sqrt{3}(pd\sigma)/2$$

- G-HM will concede to A-HM with decreasing J_{SE}
- Localized spin:

$$S_j = S (\hat{z} \cos \mathbf{Q} \cdot \mathbf{x}_j + \hat{y} \sin \mathbf{Q} \cdot \mathbf{x}_j)$$

- M. Mostovoy, Phys. Rev. Lett. **94**, 137205 (2005)



Motivation and purpose

1. What is the origin of different stable helical structures in SrFeO_3 and BaFeO_3 ? The most significant effect may come from lattice parameter. Since ionic radius of Ba^{2+} is larger than Sr^{2+} , BaFeO_3 is expanded as compared with SrFeO_3 .
2. The specialized double exchange model plus superexchange J_{SE} with several empirical parameters can describe SrFeO_3 and BaFeO_3 properly?
3. In order to include material information in theory, we need to perform the first principles calculation of magnetic structures in these compounds based on density functional theory (DFT).

Calculation method

- Density functional theory (DFT) calculation.
- For 3d electron in transition element, local spin density approximation (LSDA) is insufficient, we need additional on-site Hubbard interaction for correction.

$$E^{\text{LDA}+U}[\rho^\sigma(\mathbf{r}), \{n^\sigma\}] = E^{\text{LSDA}}[\rho^\sigma(\mathbf{r})] + E^U[\{n^\sigma\}] - E_{\text{dc}}[\{n^\sigma\}],$$

$$E_{\text{XC}}^{\text{LSDA}}[\rho] = \int dr \rho(r) \varepsilon_{\text{XC}}(\rho)$$

$$\Psi_{\mathbf{k}}(\mathbf{r}) = \begin{pmatrix} e^{i(\mathbf{k}-\mathbf{q}/2)\cdot\mathbf{r}} & 0 \\ 0 & e^{i(\mathbf{k}+\mathbf{q}/2)\cdot\mathbf{r}} \end{pmatrix} \begin{pmatrix} \mu_{\mathbf{k}}^\uparrow(\mathbf{r}) \\ \mu_{\mathbf{k}}^\downarrow(\mathbf{r}) \end{pmatrix}$$

A. I. Liechtenstein, V. I. Anisimov and J. Zaanen, Phys. Rev. B 52, R5467 (1995)

R. Laskowski, et al, Phys. Rev. B 69, 140408(R) (2004)

DFT calculation-LSDA

- Calculate the propagating vector-dependent total energy per primitive cell
- FM state predicted by LSDA

Total energy respect to FM state:

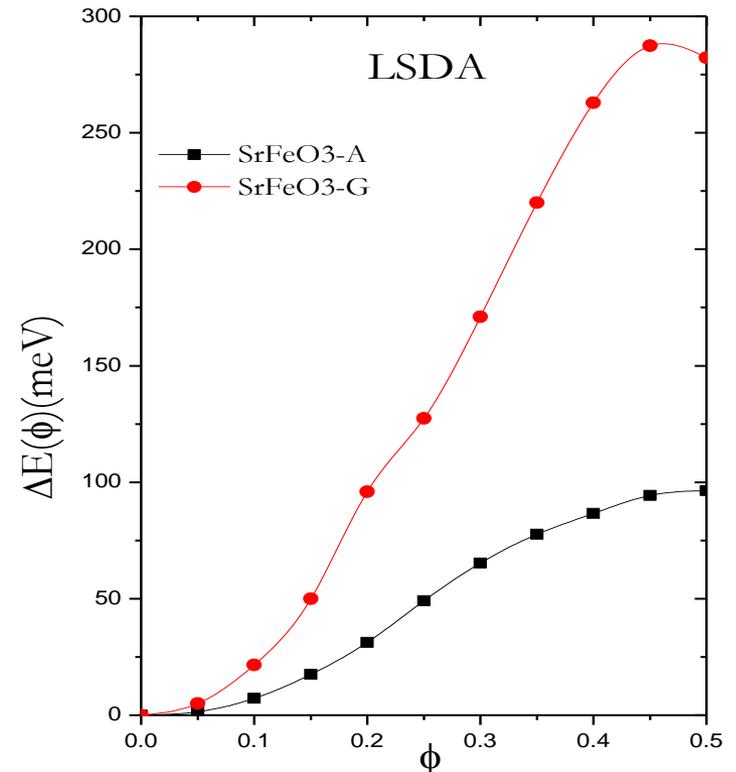
$$\Delta E(\phi) \equiv E(\phi) - E(\phi = 0)$$

$$\mathbf{q} = \phi(1, 1, 1) \times 2\pi/a_S$$

$$\mathbf{q} = \phi(1, 0, 0) \times 2\pi/a_S$$

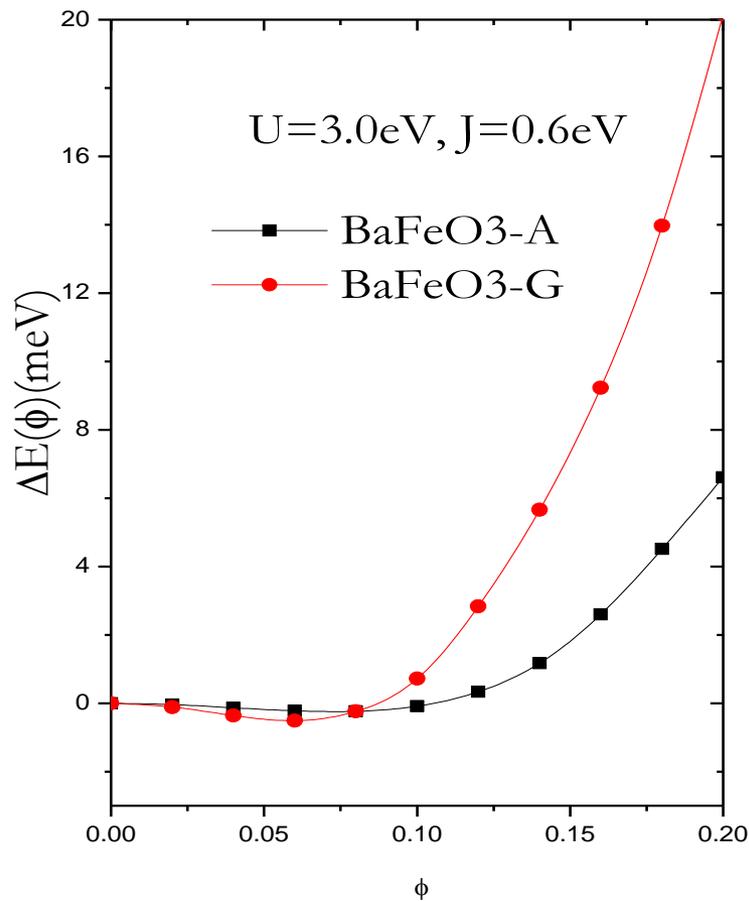
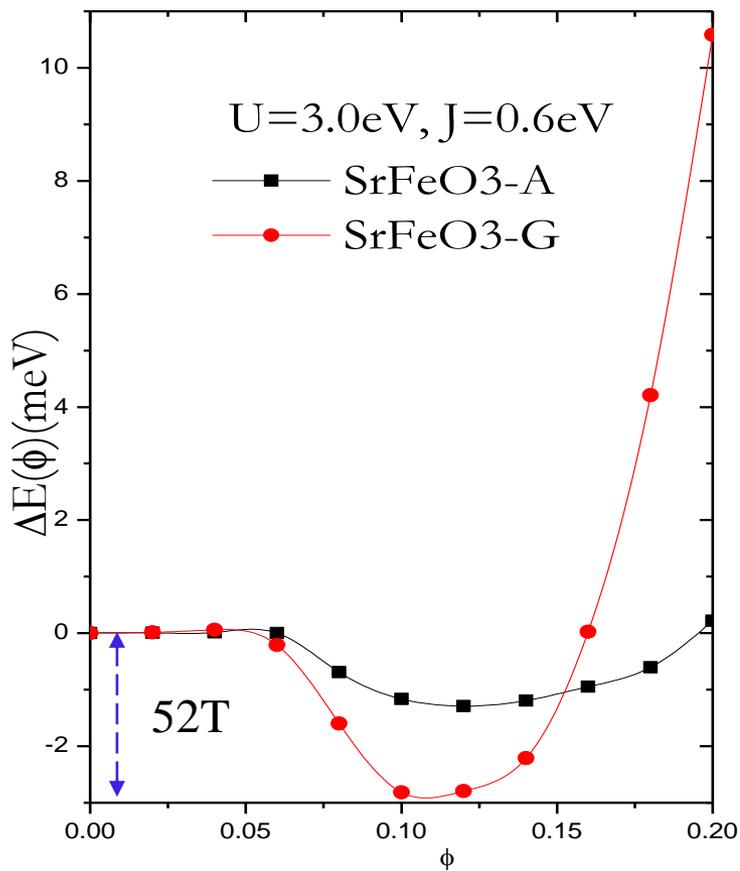
For SrFeO₃: $a_S = 3.85 \text{ \AA}$

For BaFeO₃: $a_B = 3.97106 \text{ \AA}$



DFT calculation-LSDA+U

Helical spin order predicted

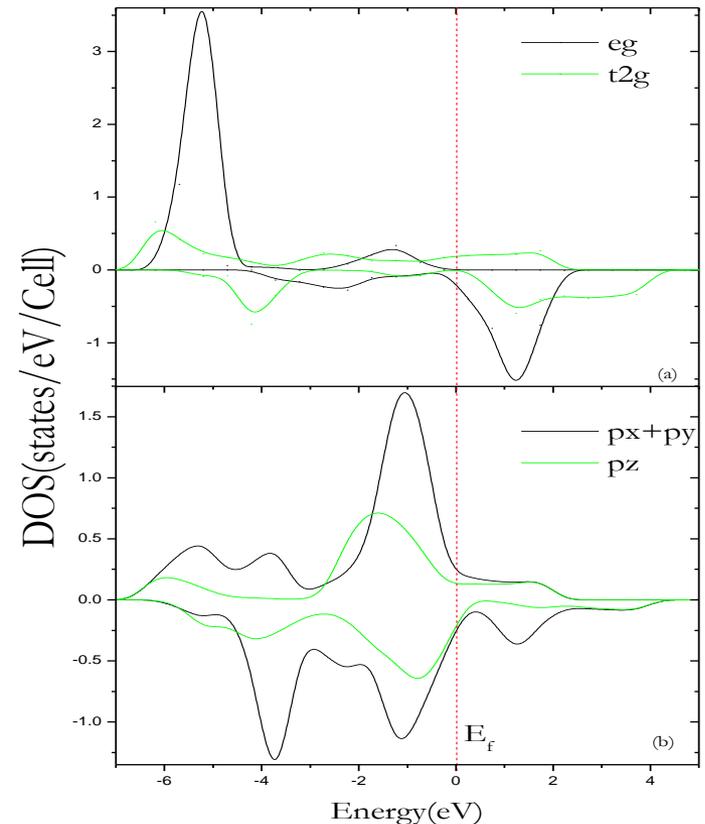


Density of State

- The density of state (DOS) of FM state in BaFeO_3 is calculated by LSDA+U, $U=3.0\text{eV}$ and $J=0.6\text{eV}$

1. The presence of O2p just below the Fermi level is an indirect evidence of small or negative charge transfer energy Δ .

2. Both e_g of iron and p orbital of oxygen makes contribution to the metallic conductivity

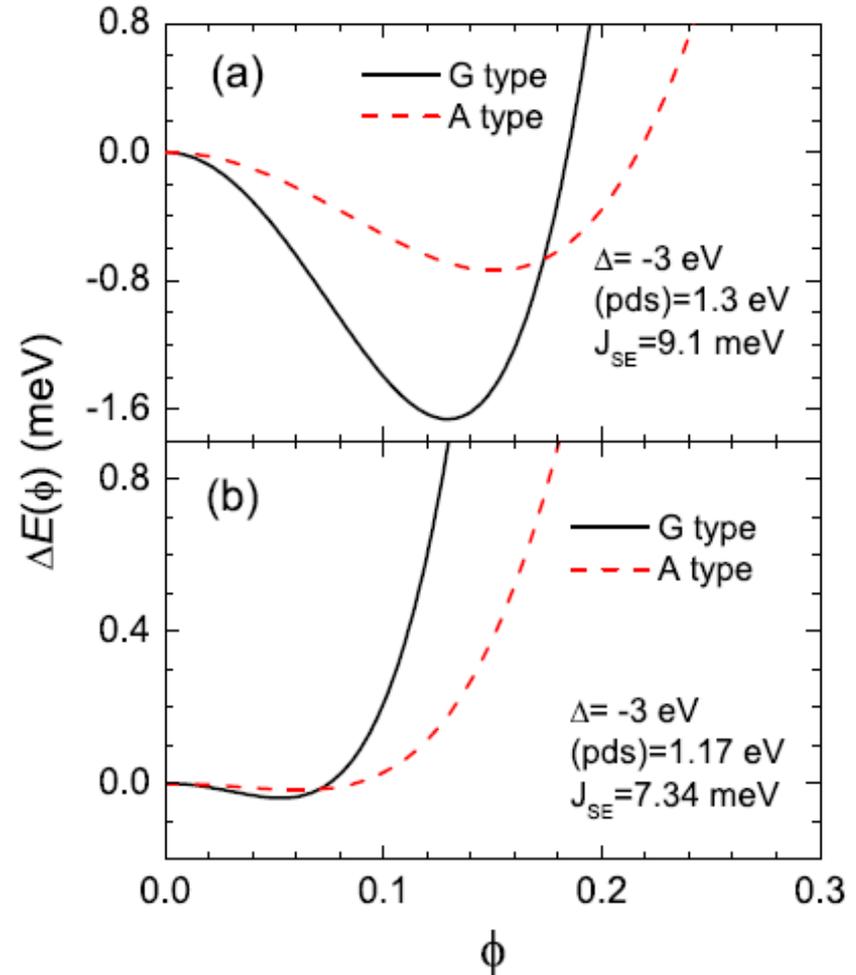


Model calculation

- Mostovoy's *dp*-model

Considering the different lattice parameter in SrFeO_3 and BaFeO_3 , the Slater-Koster and superexchange parameters is reduced in BaFeO_3 because of large lattice parameter.

- The results of the model calculation are qualitatively consistent with the $\text{LSDA}+U$.



Conclusion

1. The energy difference between the minimum-energy state and FM state is larger in SrFeO₃ than in BaFeO₃, consistent with experimental observation that SrFeO₃ requires larger magnetic field to achieve saturated FM state.
2. The difference of the two compounds is attributed to the difference of lattice constant.
3. We have shown clear correspondence between the first principles calculation based on LSAD+*U* and the double exchange model implemented by superexchange interaction, confirming the importance of the competitions between double exchange and superexchange interactions. The correspondence also implies that the characteristics of negative Δ is included in the first principles calculations.