Electric-field Control of Topological Spin Textures, Kitaev Spin Liquids, and Magnetic Orders

Masahiro Sato　(Chiba Univ.)

[1] K. Takasan and MS, PRB**100**, 060408(R) (2019).
[2] S. C. Furuya, K. Takasan and MS, PRR**3**, 033066 (2021).
[3] S. C. Furuya and MS, arXiv:2110.6503.

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Quantum Liquid Crystals

CHIBA UNIVERSITY FACULTY OF SCIENCE

科研費 KAKENHI

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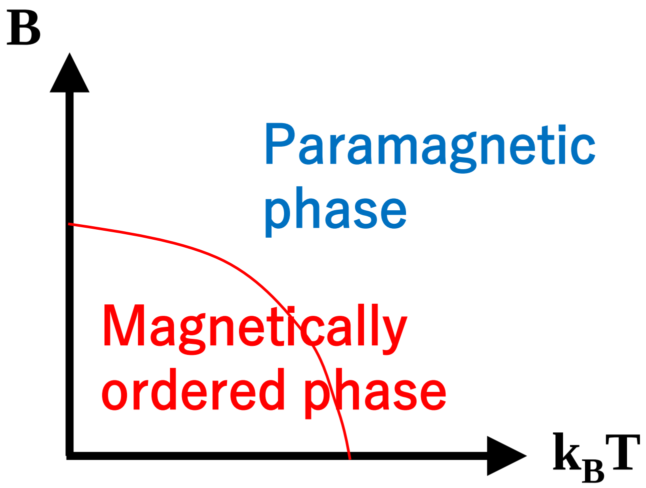
Shunsuke C. Furuya (Univ. Tokyo)

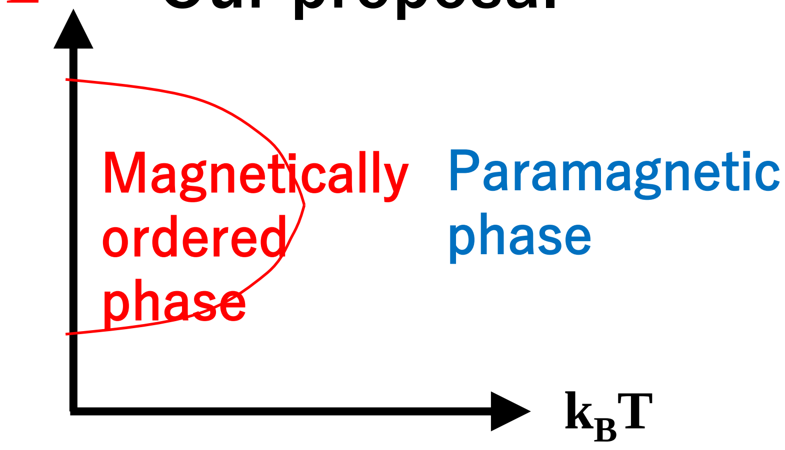
Kazuaki Takasan (Univ. Tokyo)

Introduction to strong DC electric fields in Mott insulators

Usual magnetic phase diagram



Our proposal



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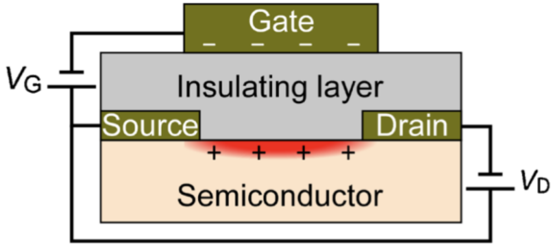
Strong DC electric field beyond 1MV/cm can be generated with current experimental techniques.

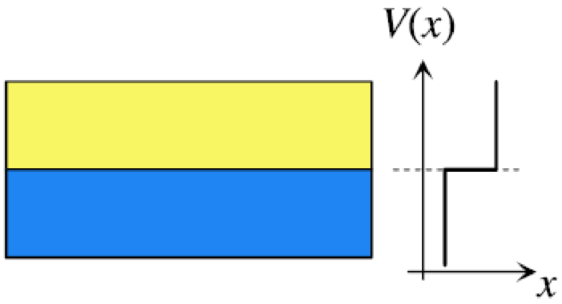
(1) Field-effect transistor (FET) or Electric double layer transistor (EDLT)

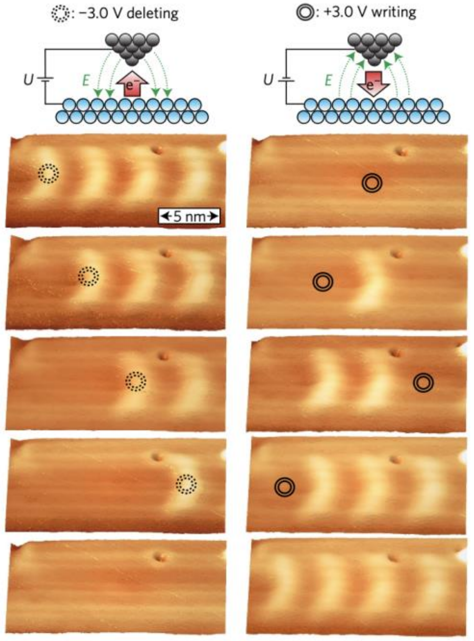


[Ueno *et al.*, JPSJ (2014)]

(2) Scanning Tunneling Microscope (STM)

(3) Interface between different crystals

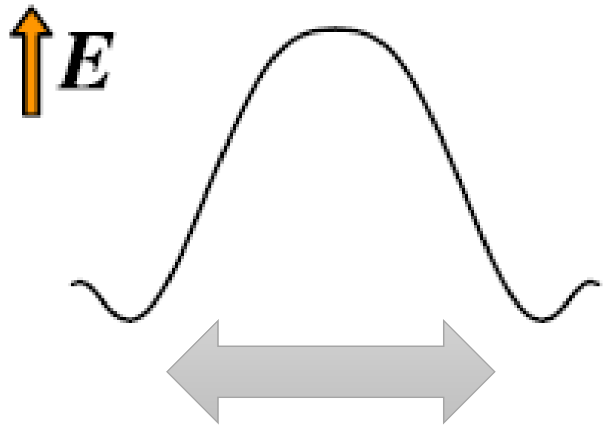




[Hsu *et al.*, Nat. Natnotech. (2016)]

(4) THz laser pulse may be also used as a quasi-static electric field

Half- or one-cycle
THz pulse



$\sim 1 - 10$ pico seconds (pico sec $= 10^{-12}$ sec)

Typical time scale of electron dynamics of solids : $\sim 1 - 100$ femto seconds
(femto sec $= 10^{-15}$ sec)

Time evolution of electric field of THz pulse is much slower than electron motion.

Electric field of THz pulse ~ nearly static for electrons

In insulating systems, effects of DC electric fields have long studied, especially, in the fields of multiferroics and spintronics.

H. Katsura, N. Nagaosa, and Balatsky, Phys. Rev. Lett. **95,** 057205 (2005),
M. Mostovoy, Phys. Rev. Lett. **96,** 067601 (2006),
I.A. Sergienkoand E. Dagotto, Phys. Rev. Lett. **96,** 067601 (2006),
F. Matsukura, Y. Tokura, and H. Ohno, Nature Nanotech. **10**, 209 (2015),
H. Katsura, MS, T. Furuta and N. Nagaosa, Phys. Rev. Lett. **103**, 177402 (2009),
,,,,,, many papers

Many theoretical studies are based on
the symmetry arguments and the effective low-energy theories.

Theories from the microscopic viewpoint
have been not developed enough.

We have tried to develop the theory
which can predict E-field induced phenomena in a quantitative way.

Our target
Effects of DC electric fields in Mott insulators (spin systems)

Outline

1, Electric field can change direct exchange interactions.

K. Takasan and MS, PRB**100**, 060408(R) (2019).

2, Electric field can change super exchange interactions.

S. C. Furuya, K. Takasan and MS, PRR**3**, 033066 (2021).

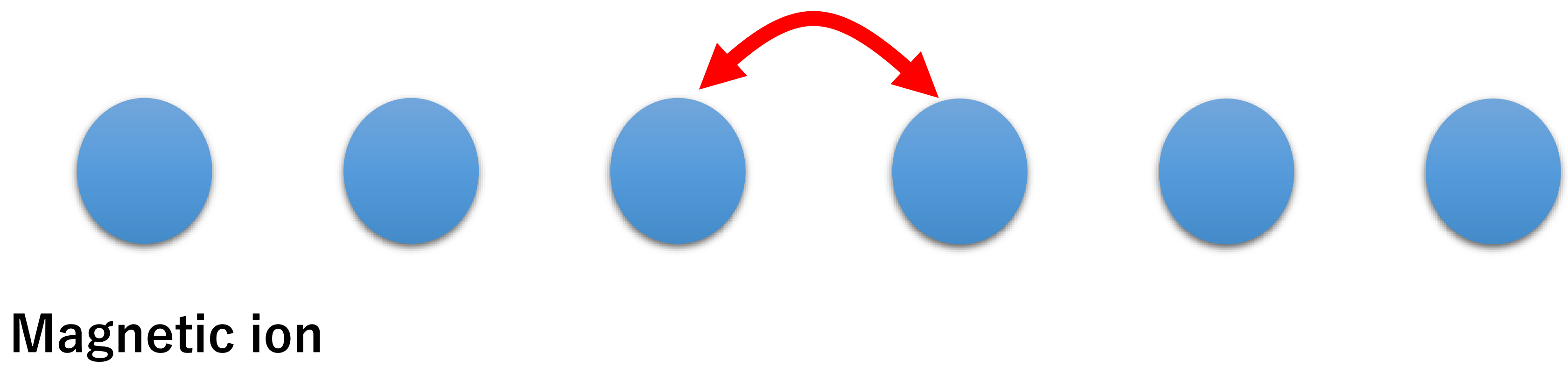
3, Electric field can change magnetic anisotropies.

S. C. Furuya and MS, arXiv:2110.6503.

Control of direct exchange interactions with DC electric fields

K. Takasan and MS, PRB**100**, 060408(R) (2019).

Direct exchange interaction



Definition of "direct" exchange interaction in our study

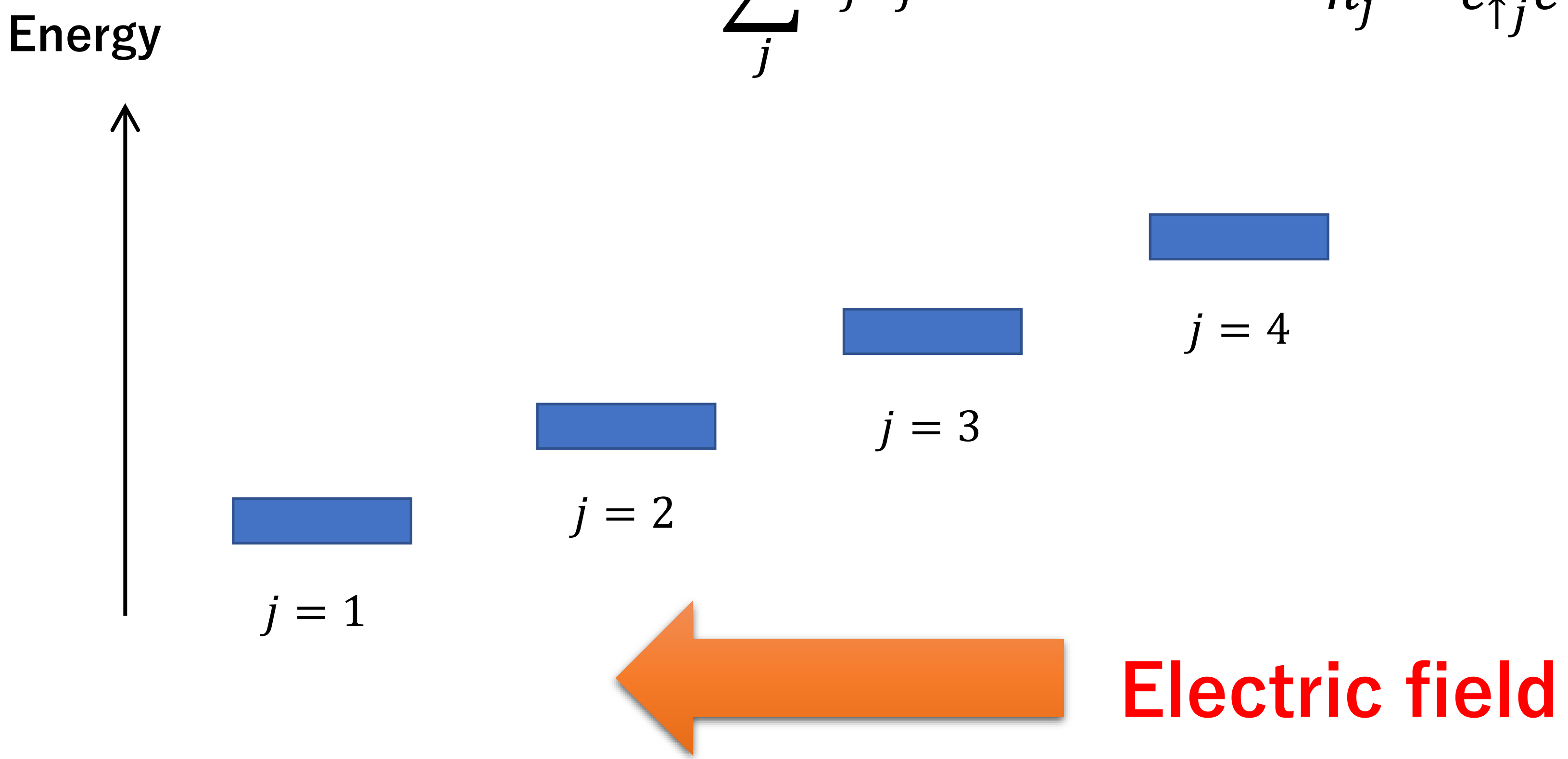
Exchange interaction generated from
direct hopping of electrons residing on neighboring magnetic ions

We theoretically treat the effect of DC electric field as the "voltage".

$$-e\sum_j V_j n_j$$

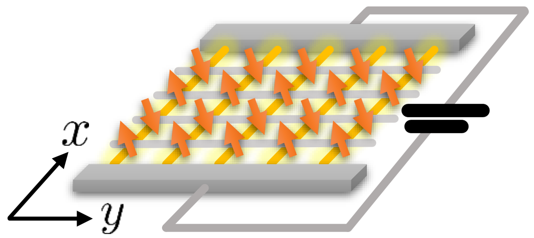
Electron number operator

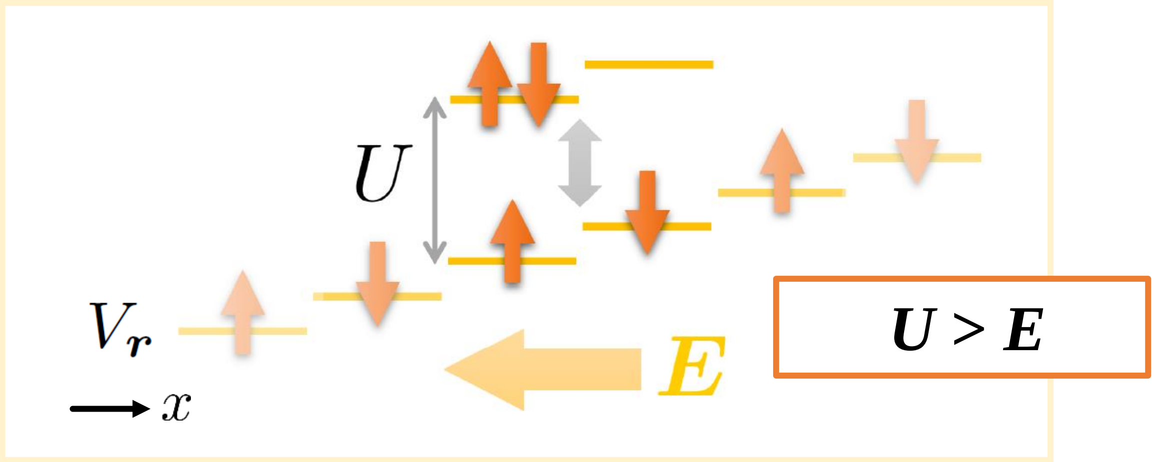
$$n_j = c^\dagger_{\uparrow j} c_{\uparrow j} + c^\dagger_{\downarrow j} c_{\downarrow j}$$



Setup : Hubbard model + E-field

DC Electric fields = **Spatial gradient of the on-site potential**





Hamiltonian

hopping | interaction | **on-site potential**

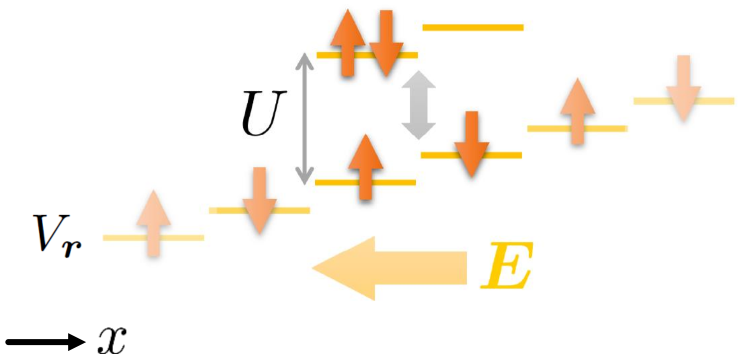
$$\mathcal{H} = \sum_{\boldsymbol{r}\boldsymbol{r}'\sigma} t_{\boldsymbol{r}\boldsymbol{r}'} c^{\dagger}_{\boldsymbol{r}\sigma} c_{\boldsymbol{r}'\sigma} + U \sum_{\boldsymbol{r}} n_{\boldsymbol{r}\uparrow} n_{\boldsymbol{r}\downarrow} + \sum_{\boldsymbol{r}\sigma} V_{\boldsymbol{r}} n_{\boldsymbol{r}\sigma},$$

In the strong coupling regime ($t \ll U$) and the half-filling case, **without** $V_{\boldsymbol{r}}$

$$\mathcal{H}_{\mathrm{eff}} = \sum_{\langle \boldsymbol{r}\boldsymbol{r}' \rangle} J_{\boldsymbol{r}\boldsymbol{r}'} \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'}$$

Heisenberg Model

Effective spin model by large-U expansion



With on-site potential $V_{\boldsymbol{r}}$

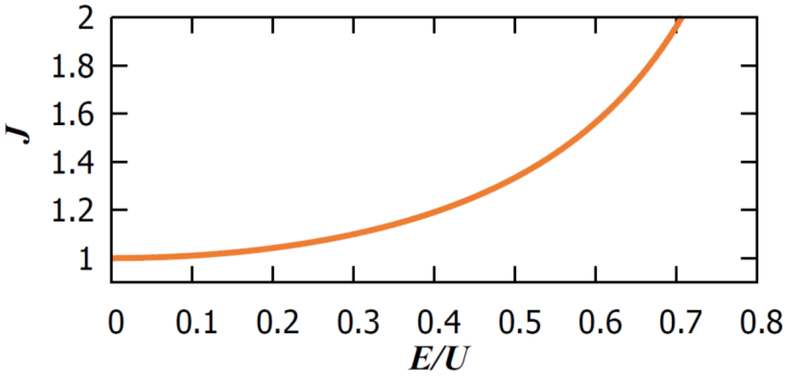
$$\mathcal{H}_{\text{eff}} = \sum_{\langle \boldsymbol{r}\boldsymbol{r}' \rangle} \frac{J_{\boldsymbol{r}\boldsymbol{r}'}}{1 - \left(\frac{\Delta V_{\boldsymbol{r}\boldsymbol{r}'}}{U} \right)^2} \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'}$$

$J_{\boldsymbol{r}\boldsymbol{r}'} = 4|t_{\boldsymbol{r}\boldsymbol{r}'}|^2/U$, $\Delta V_{\boldsymbol{r}\boldsymbol{r}'} = V_{\boldsymbol{r}} - V_{\boldsymbol{r}'}$

(Remark)
In 2-orbital case,
We obtain a
spin-1 model
with E-field dep.
Interaction.

→ Exchange coupling is enhanced !

For example, 1D case



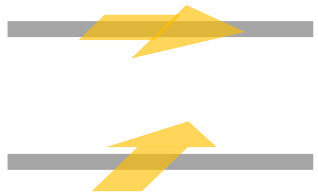
Effective exchange coupling

$$J_{\text{eff}}(E) = \frac{J_0}{1 - \left(\frac{E}{U}\right)^2}$$

At E~U, our treatment is not valid

What kinds of control is possible ?

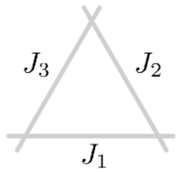
Parallel to the *E*-field ： **most** enhanced
Perpendicular **not**



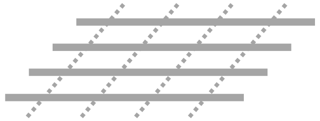
→ Control of **spatial anisotropy** of exchange interactions

Magnets where their spatial structure plays an important role should be nice for this scheme

1. Frustrated Magnets



2. Quasi-1D Magnets

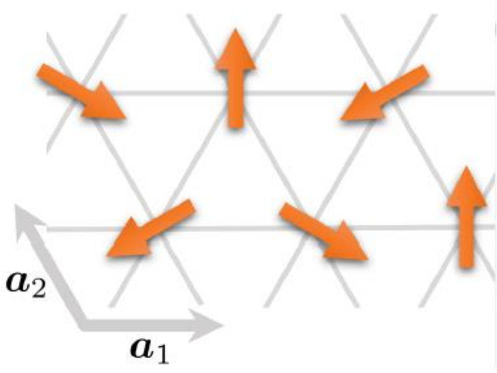


Application 1: frustrated systems

Triangular lattice antiferromagnets

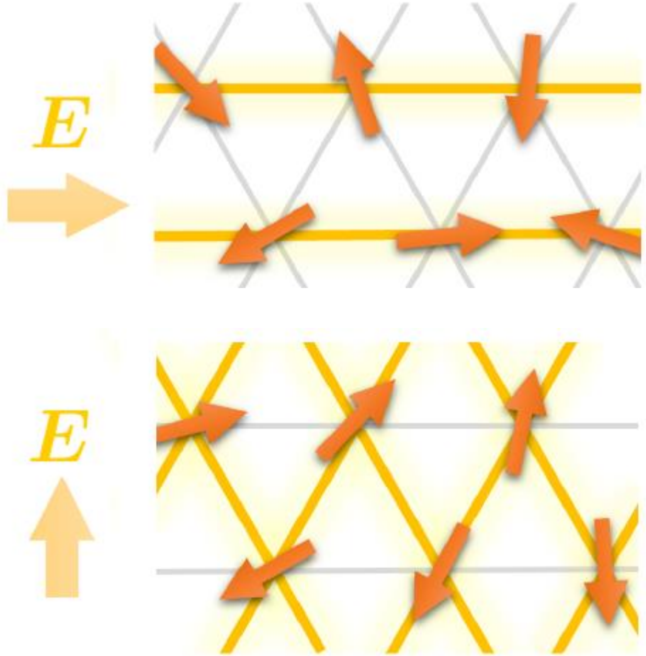
Spin Configuration (Classical G.S.)

E=0



120° structure

E>0



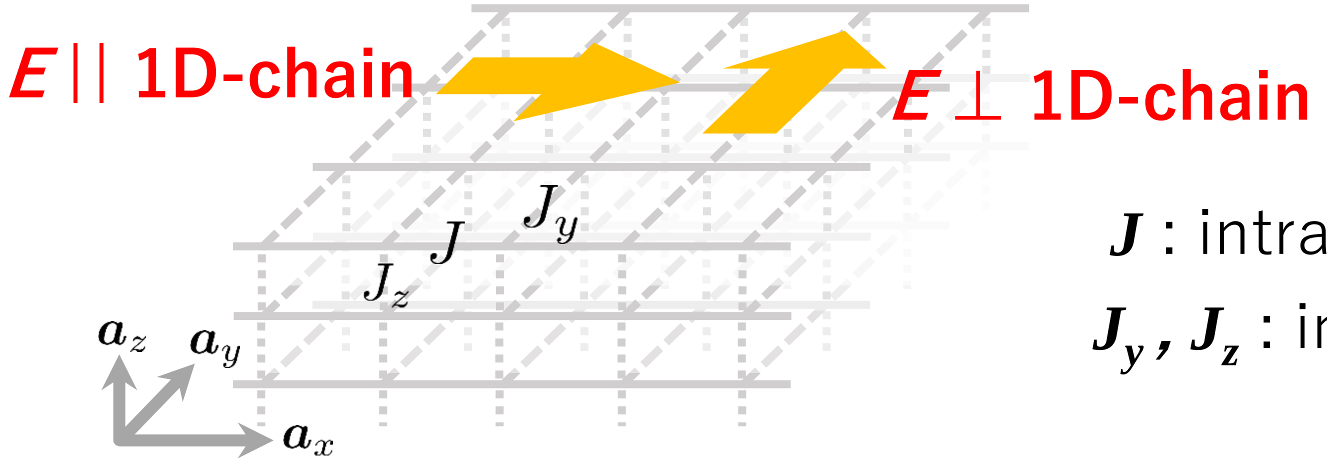
1D AFM-like spiral

2D AFM-like spiral

Control of spiral orders

Application 2: quasi-1D systems

Quasi-one-dimensional Magnets



J : intra-chain interaction

J_y , J_z : inter-chain interaction

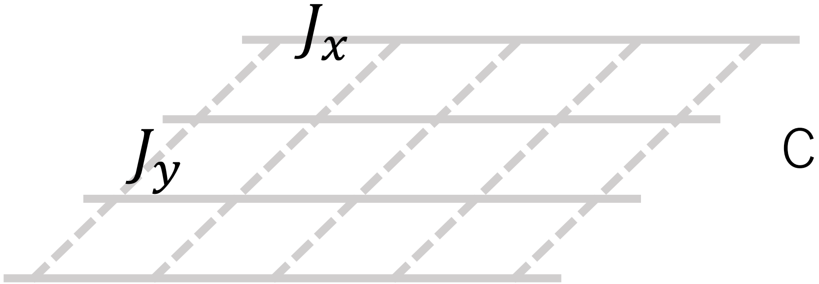
$J >> J_{y,z}$

- *E* || 1D-chain → 1D nature is enhanced
- *E* ⊥ 1D-chain → 1D nature is suppressed (2D-3D nature is enhanced)

"Dimensionality" can be controlled by changing the direction of the E-field

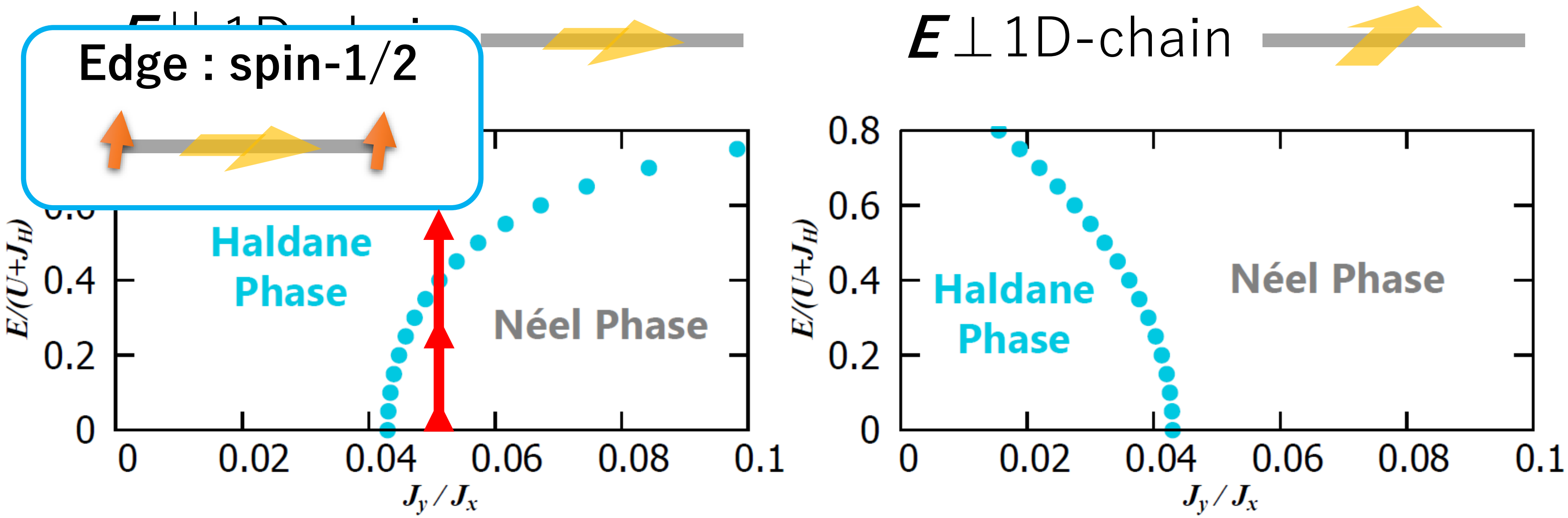
Phase diagram of spatially anisotropic 2D spin-1 system

(zero temperature, J_z=0)



Critical point between Haldane and Neel phases : $J_y = 0.043 J_x$

Matsumoto *et al.*, PRB **65**, 014407 (2001)



Haldane phase is **favored** (1D like)

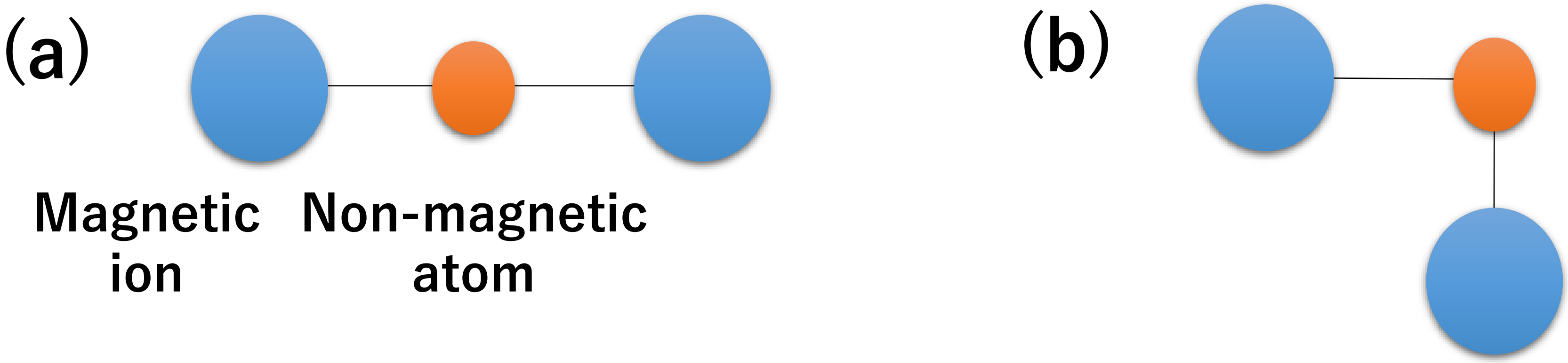
Haldane phase is **suppressed** (2D like)

→ **Field-induced Haldane-gap phase** (Control of topological order)

Control of super-exchange interactions with DC electric fields

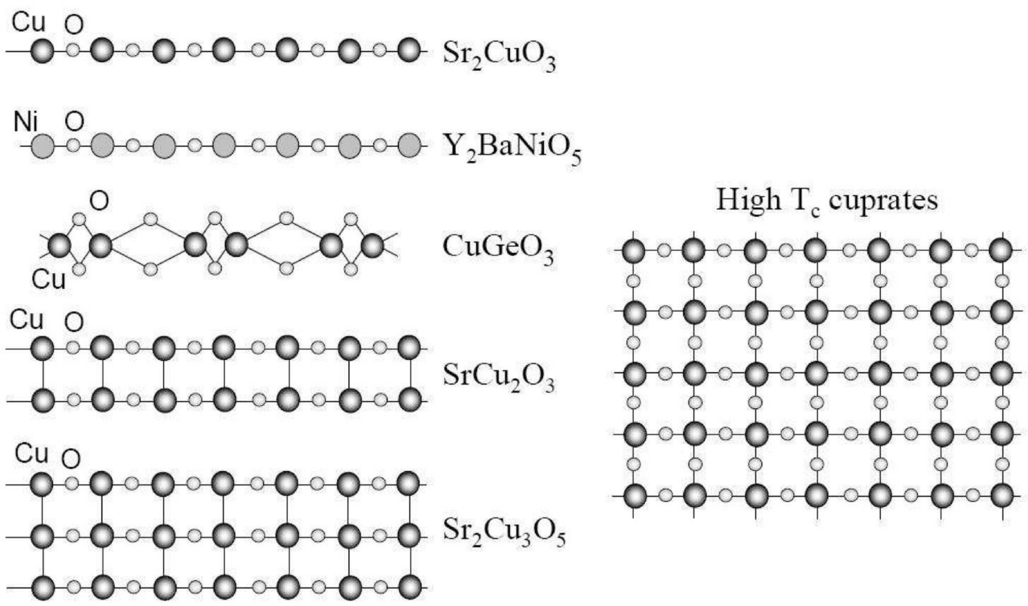
S. C. Furuya, K. Takasan and MS, PRR**3**, 033066 (2021).

Many magnetic materials have non-magnetic (ligand) atoms between magnetic ions



Super-exchange interaction :

exchange interaction between 2 magnetic ions induced by hopping processes with intermediate non-magnetic atoms



e.g) Alloul *et al.*, RMP (2009)

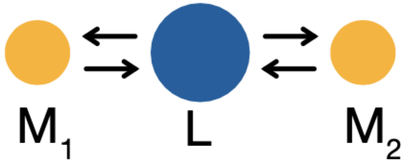
Set up:
Single-band Hubbard-like models including non-magnetic atoms

Spin-1/2 (one-electron in each magnetic ion)

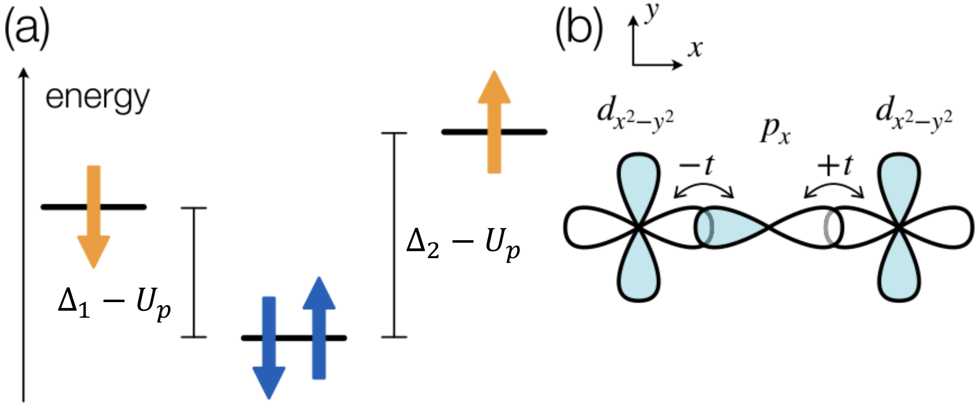
Electric potential (voltage)

$$H = -\sum_{i,j} t_{ij} \sum_{\sigma,\sigma'=\uparrow,\downarrow} (c^{\dagger}_{i,\sigma} c_{j,\sigma'} + \mathrm{H.c.}) + \sum_{i} U_i n_{i,\uparrow} n_{i,\downarrow} + \sum_{i}\sum_{\sigma} V_{i,\sigma} n_{i,\sigma} + \cdots$$

4^{th} order Large-U expansion ⇨ $H_{spin} =$



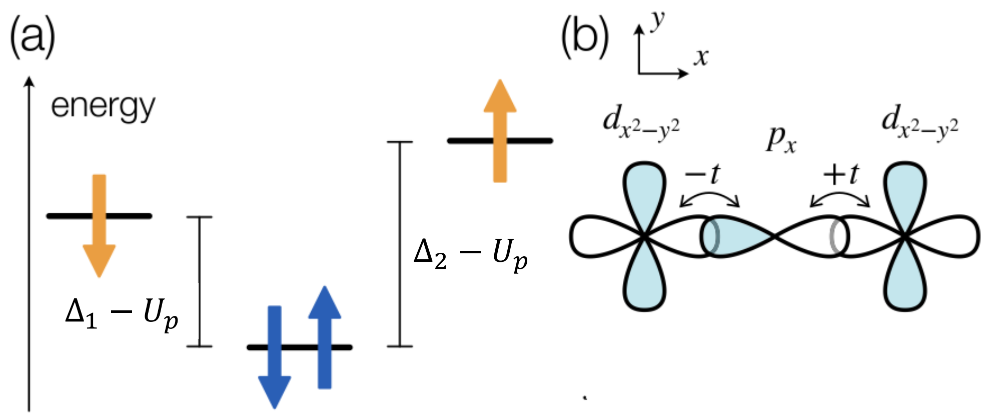
$= J\vec{S}_1 \cdot \vec{S}_2 + \cdots$

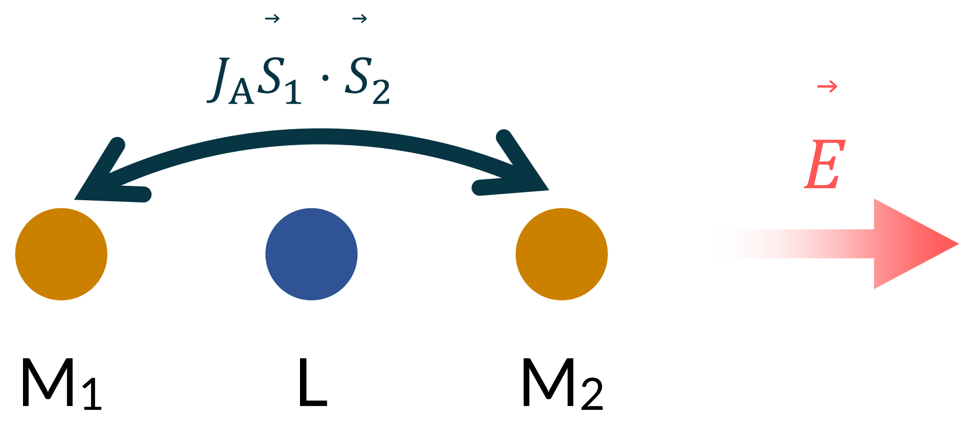


$M_{1,2}$: one electron in a d-orbit
(transition metal such as Cu)

L : two electrons in a p-orbit
(non-magnetic atom such as O)

Antiferromagnetic superexchange in the linear configuration



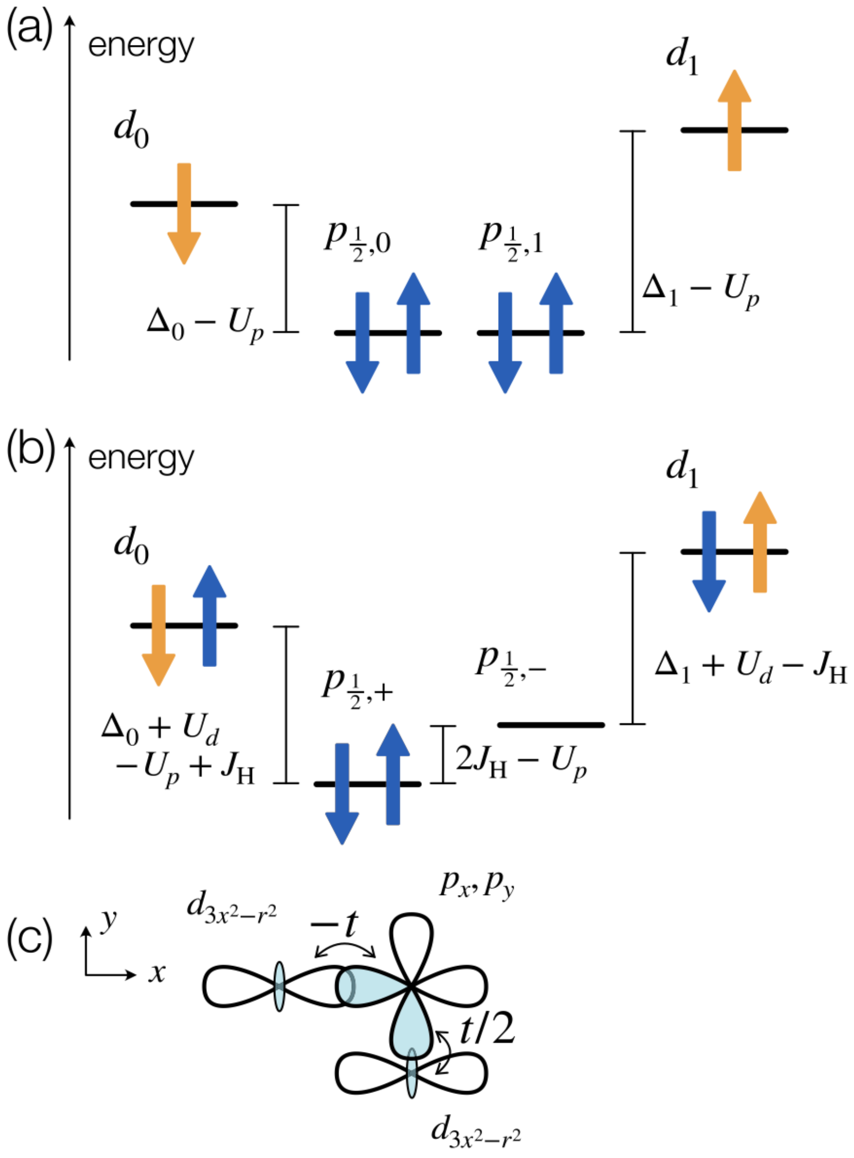


$$J_A = 2t^4\left(\frac{1}{\Delta_1(\vec{E}) + U_d - U_p} + \frac{1}{\Delta_2(\vec{E}) + U_d - U_p}\right)^2 \frac{1}{\Delta_1(\vec{E}) + \Delta_2(\vec{E}) + 2U_d - U_p}$$
$$+2t^4\left(\frac{1}{(\Delta_1(\vec{E})+U_d-U_p)^2}\frac{1}{\Delta_1(\vec{E})-\Delta_2(\vec{E})+U_d} + \frac{1}{(\Delta_2(\vec{E})+U_d-U_p)^2}\frac{1}{\Delta_2(\vec{E})-\Delta_1(\vec{E})+U_d}\right)$$

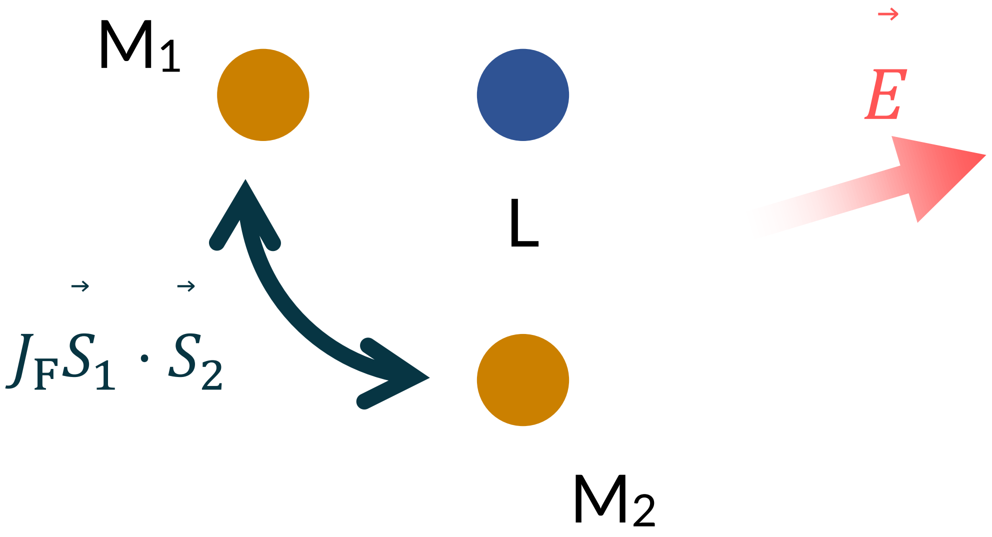
$\Delta_{1,2}$ **: potential energy at site 1 and 2**

$$\Delta_2(\vec{E}) - \Delta_1(\vec{E}) = -e\vec{E} \cdot \vec{r}_{12}$$

Ferromagnetic superexchange in the triangular configuration

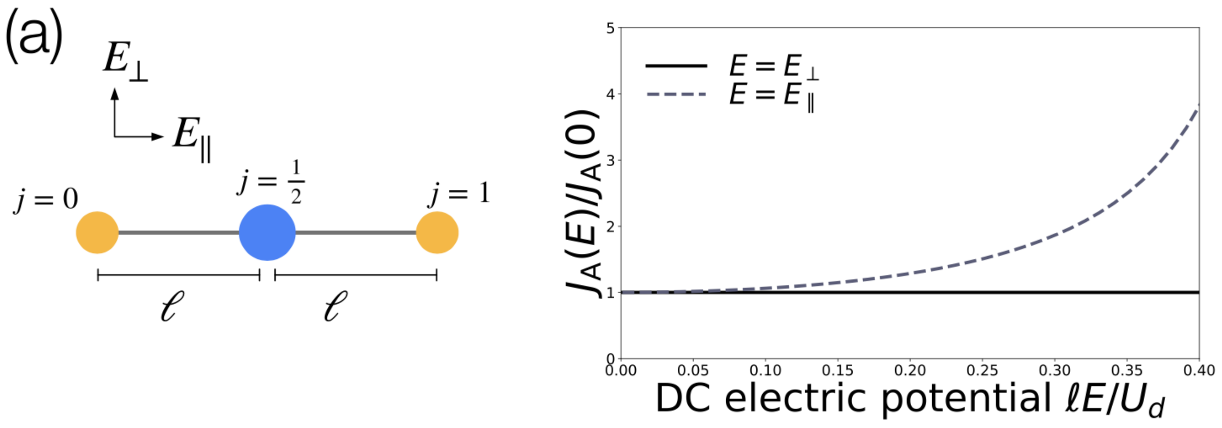


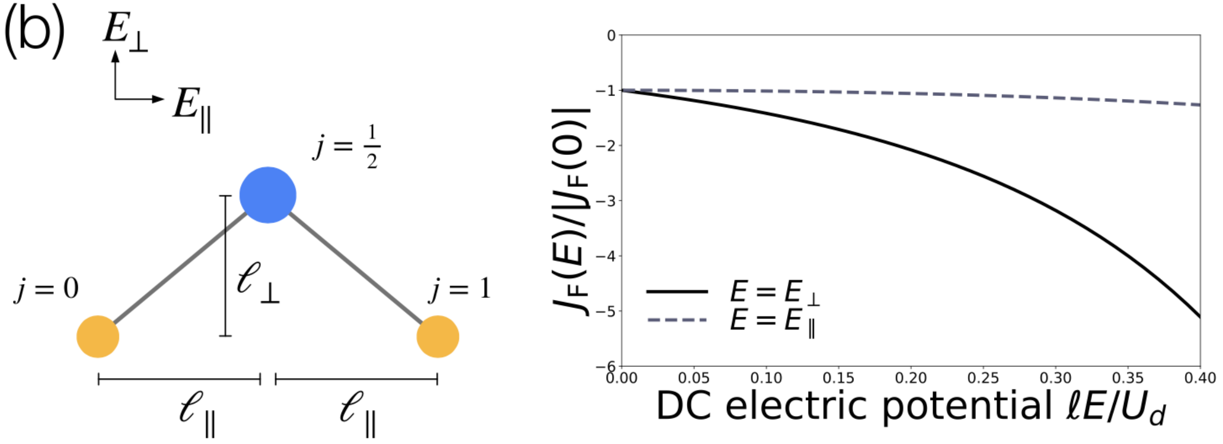
Kanamori-Goodenough rule
J_F is generally ferromagnetic ($J_F < 0$)



$$\Delta_2(\vec{E}) - \Delta_1(\vec{E}) = -e\vec{E} \cdot \vec{r}_{12}$$

DC electric-field controls of AFM/FM superexchanges





- Electric potential differences at three sites.

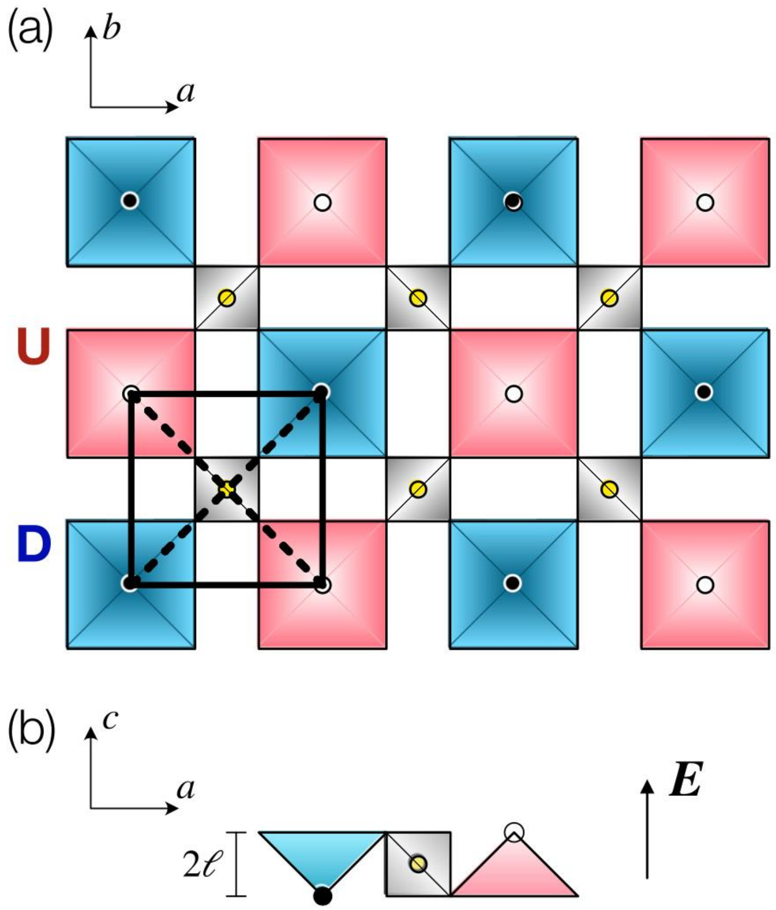
- For $U_d = 5\text{eV},\ U_p = 1\text{eV},$
  $\Delta_{dp}(0) = 2\text{eV},\ J_{\text{H}} = 1\text{eV},$
  $\ell = 5\text{Å}$, ...

  the O(1) MV/cm DC electric field is required to change the superexchange by O(1)%.

- The condition will be relaxed for thin films and organic compounds.

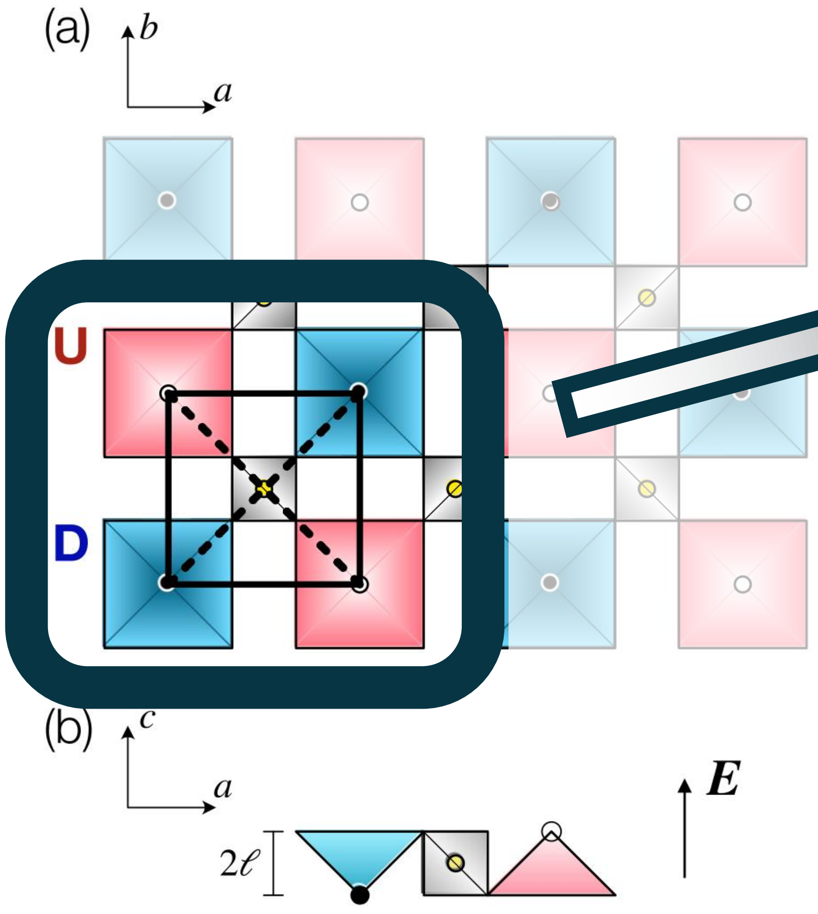
Application 1

$BaCdVO(PO_4)_2$

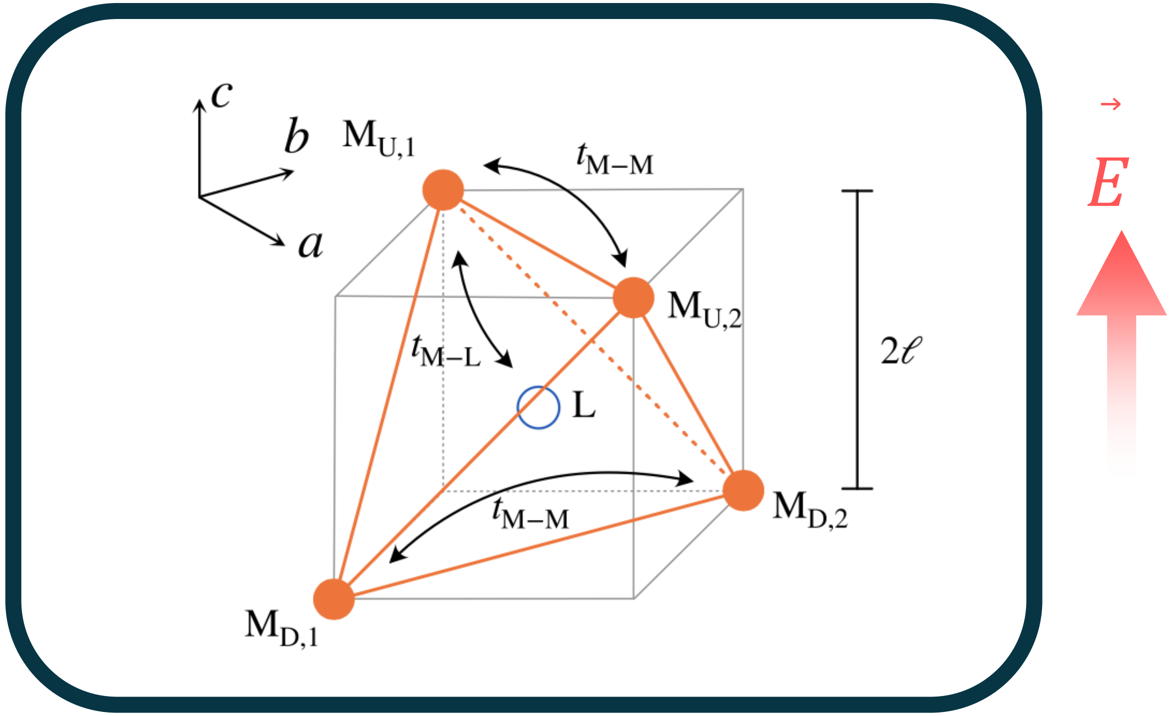


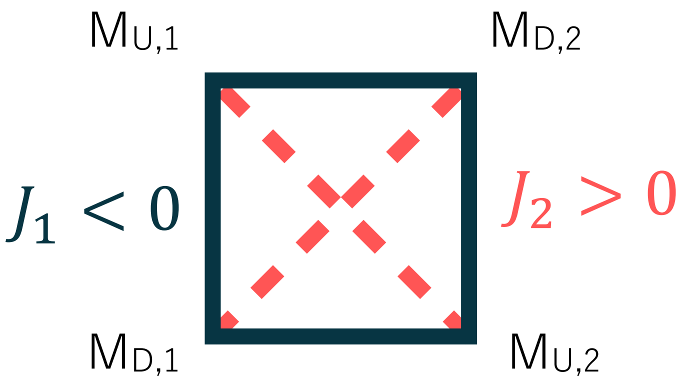
Nath *et al.*, PRB (2008)

Application 1



BaCdVO(PO4)2

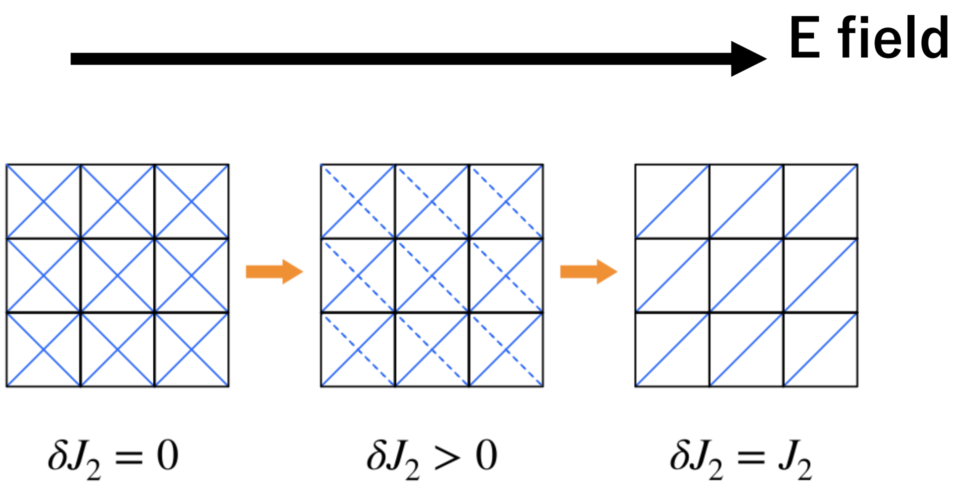


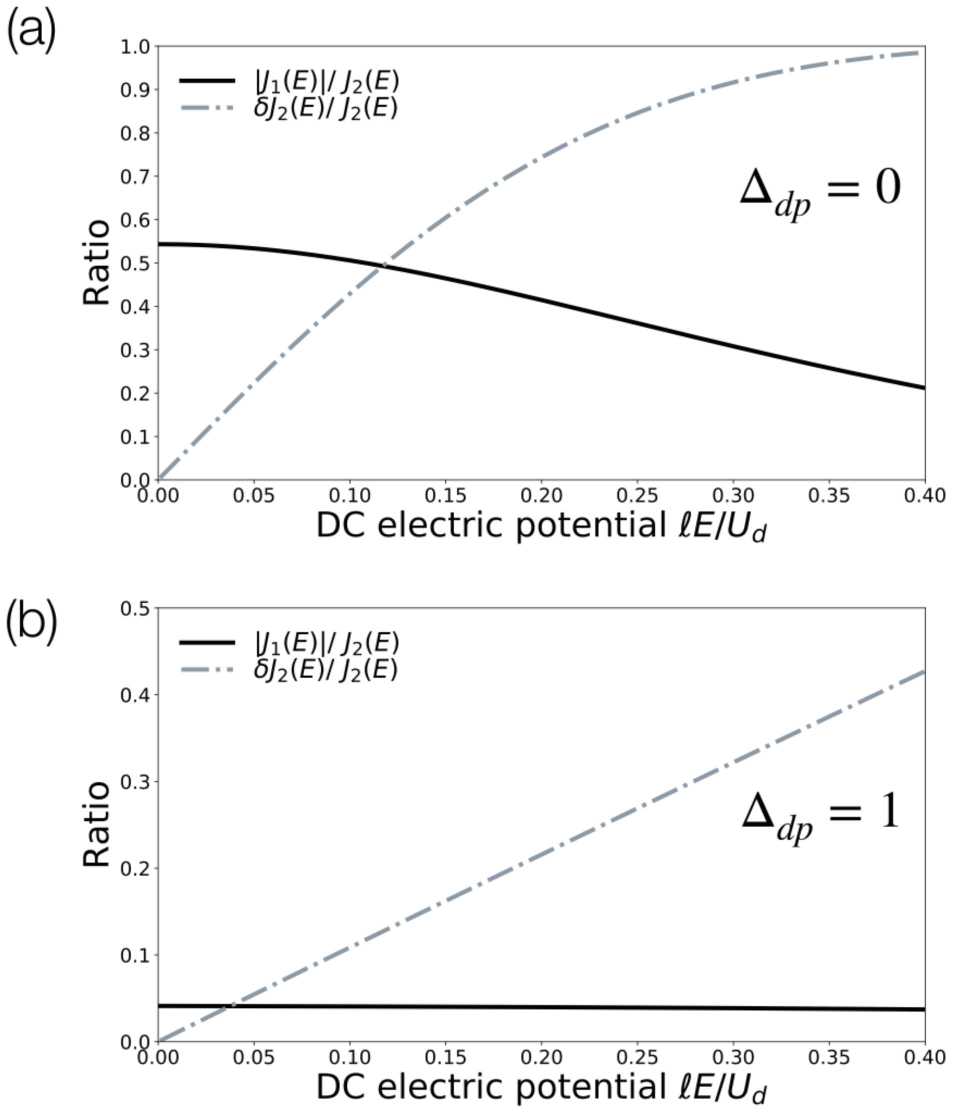


Nath *et al.*, PRB (2008)

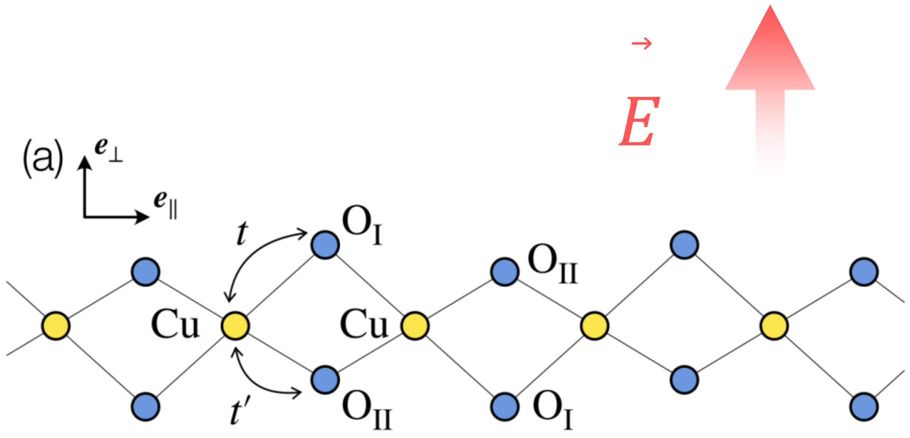
Application 1

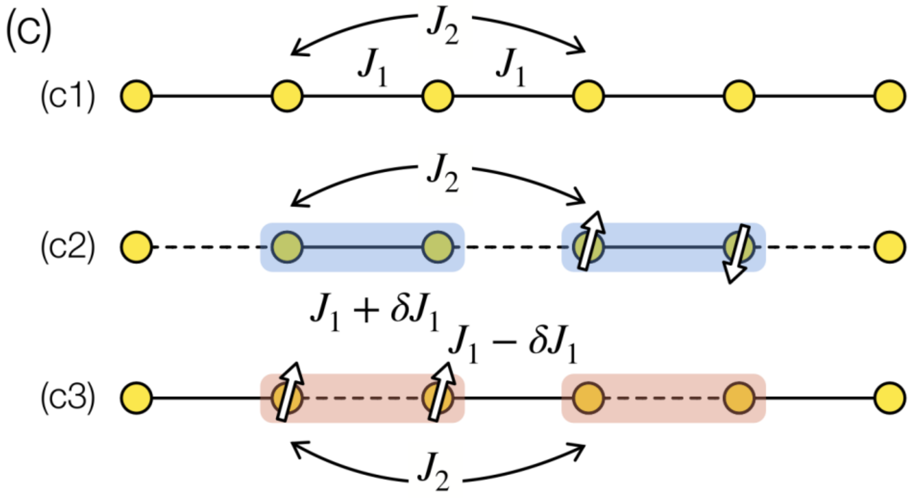
Out-of-plane DC electric field changes the model



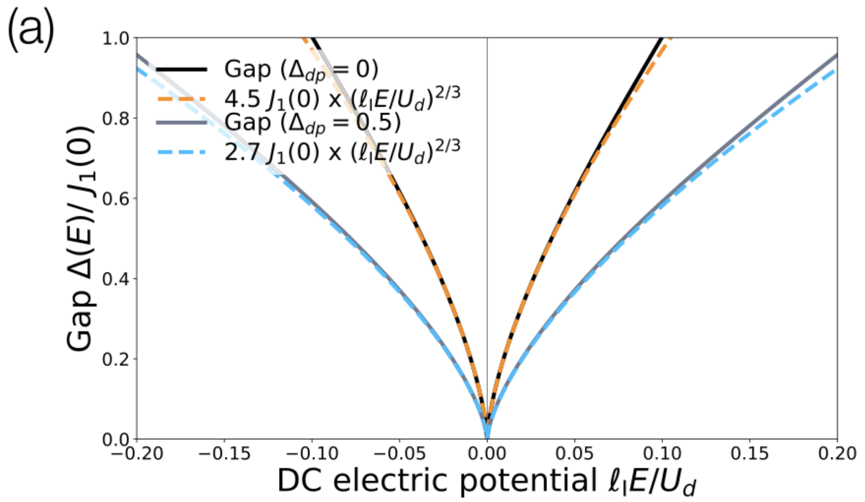


Application 2





- Spin-1/2 Heisenberg AFM chain.
  - GS = critical (Tomonaga-Luttinger liquid)
- DC electric-field drives dimerization of spins.
  - Spin gap opens.
  - Spin gap $\propto |E_\perp|^{2/3}$



Control of magnetic anisotropies and spin textures with DC electric fields

S. C. Furuya and MS, arXiv:2110.6503.

So far we considered exchange interaction modified by E-field.

isotropic in spin space

Electric field breaks inversion (parity) symmetry.

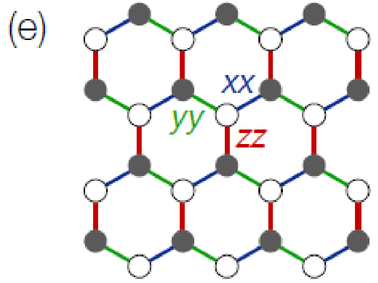
→ electron orbits are deformed (polarization appears).

→ Spin-orbit coupling is also changed/generated.

→ Magnetic anisotropy is generated by E field.

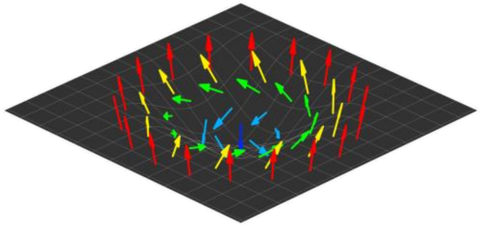
Case 1: Intra-atom SO coupling

Kitaev magnets

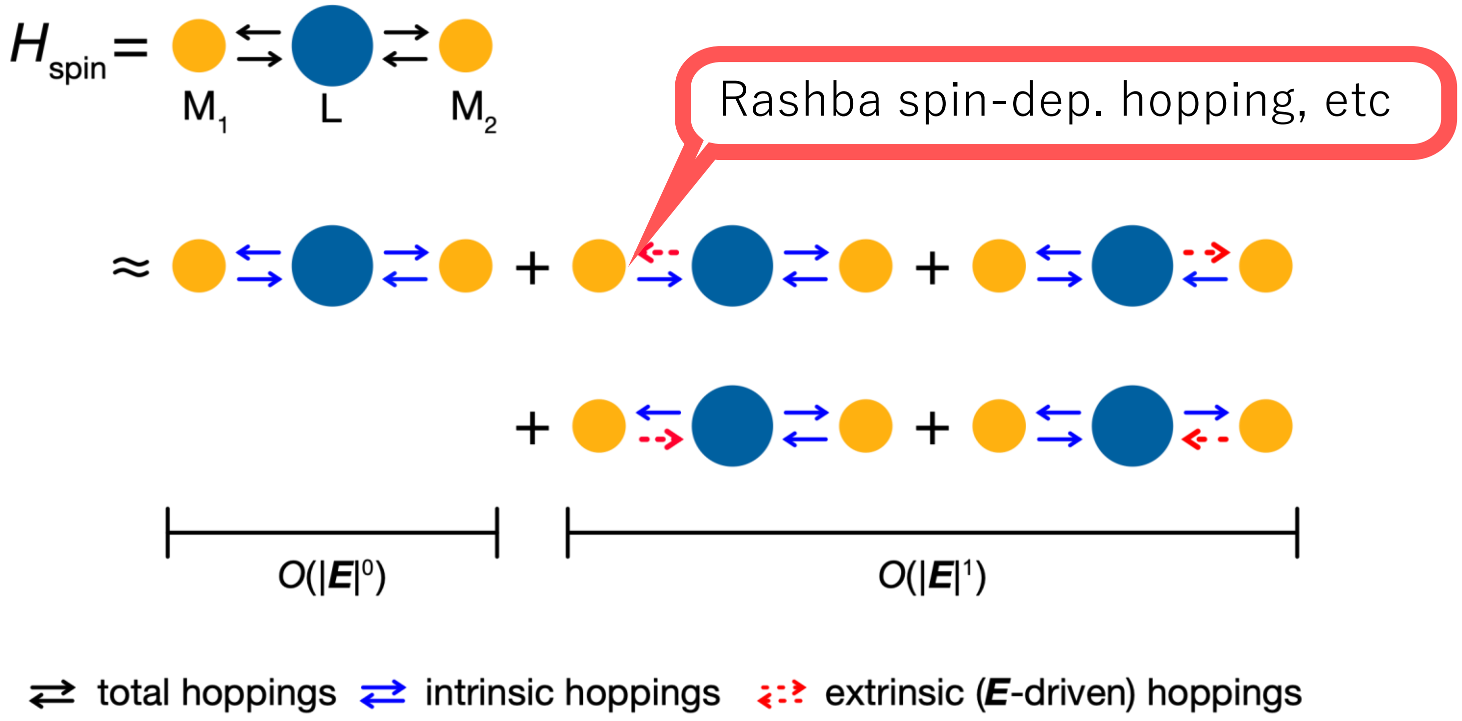


Case 2: Inter-atom SO coupling

E-field driven Rashba SO couplings

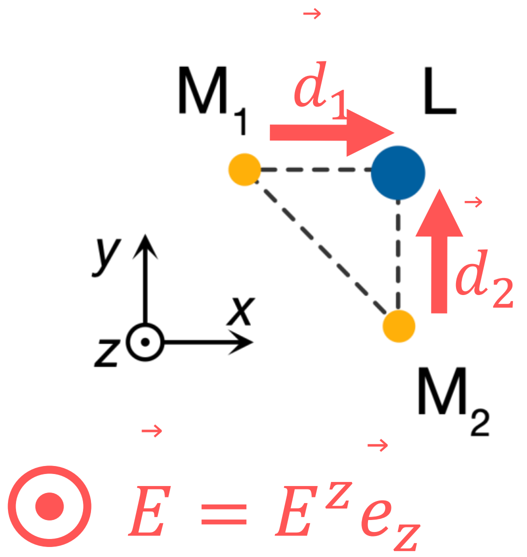


Large-U expansion leads to spin Hamiltonian with E-field induced magnetic anisotropy



Case 2 : Mott insulator with inter-atomic Rashba SOC

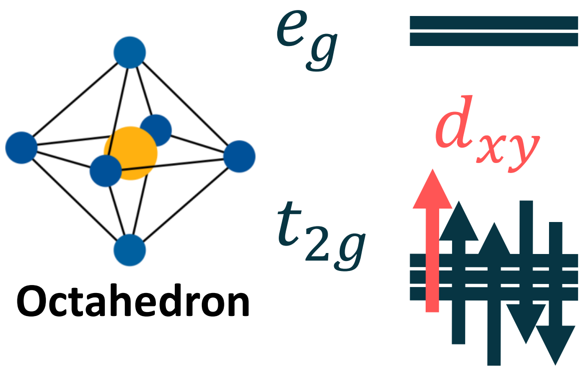
Building block (3 sites)



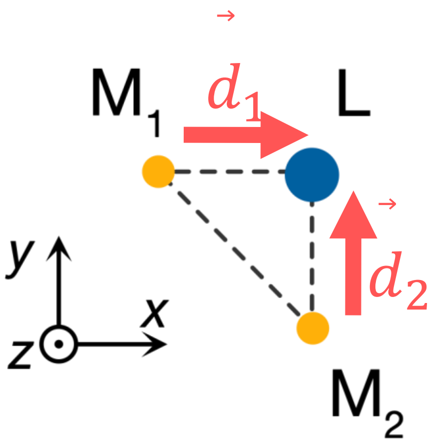
- Low-spin d^5 configuration.
  - One of the t_{2g} orbitals, say, the d_{xy} orbital, carries spin-1/2.
- Three-site model w/ an intra-atomic SOC, the Rashba SOC.
- Spin-dependent electron hoppings between magnetic ions and ligand (λ ∝ E electric field).

$$\lambda \sum_{s,s'} [i p_{y,s}^{\dagger} (\vec{\sigma}_{s,s'} \times \vec{d}_1)^z d_{1,xy,s'} + i p_{x,\sigma}^{\dagger} (\vec{\sigma}_{s,s'} \times \vec{d}_2)^z d_{2,xy,s'} + \mathrm{H.c.}]$$

- The out-of-plane electric field is assumed.



E-driven DM interaction



$\vec{E} = E^z \vec{e}_z$

- The spin Hamiltonian w/ $\vec{E} = E^z \vec{e}_z$:

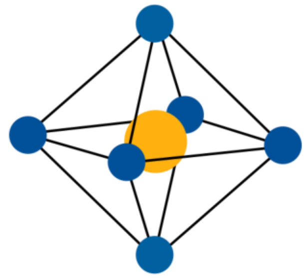
$$H_{\text{spin}} = -|J|\vec{S}_1 \cdot \vec{S}_2 - B\,(S_1^z + S_2^z) + \vec{D} \cdot \vec{S}_1 \times \vec{S}_2$$

- $\vec{E}$-driven DM interaction. $\vec{D} = D\,(\vec{e}_x + \vec{e}_y)$

$\lambda \propto E^z$

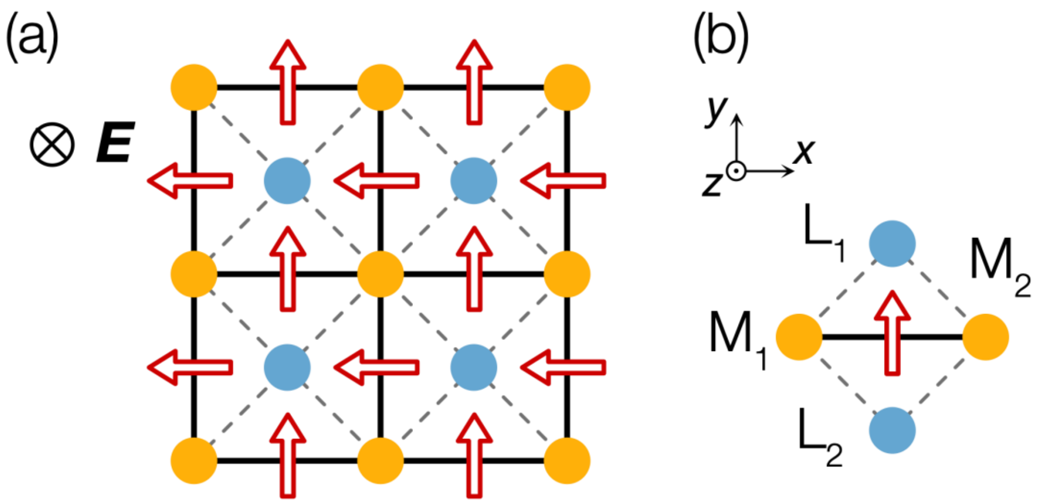
$$D = -4\lambda t^3 \frac{U_d - U_p + \Delta_{dp}}{4(U_d - U_p + \Delta_{dp})^2 - {J_{\text{H}}}^2}$$

Embedding to square lattice



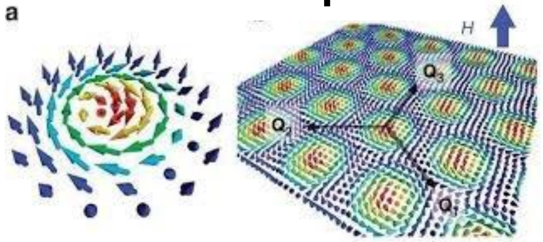
- Square lattice made of edge-sharing octahedra.
- Assume the Rashba SOC between M-L bonds.
- The resultant spin Hamiltonian is

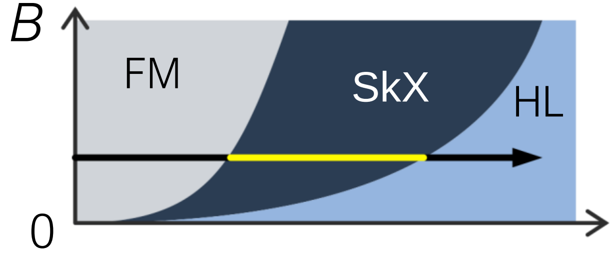
$$H_{\mathrm{spin}} = -|J|\sum_{\vec{r}}(\vec{S}_{\vec{r}} \cdot \vec{S}_{\vec{r}+\vec{e}_x} + \vec{S}_{\vec{r}} \cdot \vec{S}_{\vec{r}+\vec{e}_y})$$
$$-B\sum_{\vec{r}} S^z_{\vec{r}}$$
$$+D\sum_{\vec{r}}(\vec{e}_y \cdot \vec{S}_{\vec{r}} \times \vec{S}_{\vec{r}+\vec{e}_x} - \vec{e}_x \cdot \vec{S}_{\vec{r}} \times \vec{S}_{\vec{r}+\vec{e}_y}).$$



$$\odot \quad \vec{E} = E^z \vec{e}_z$$

SkX phase



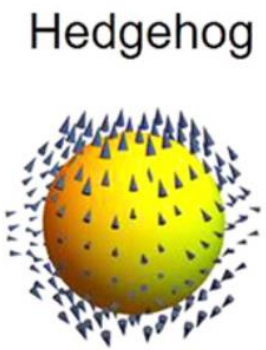


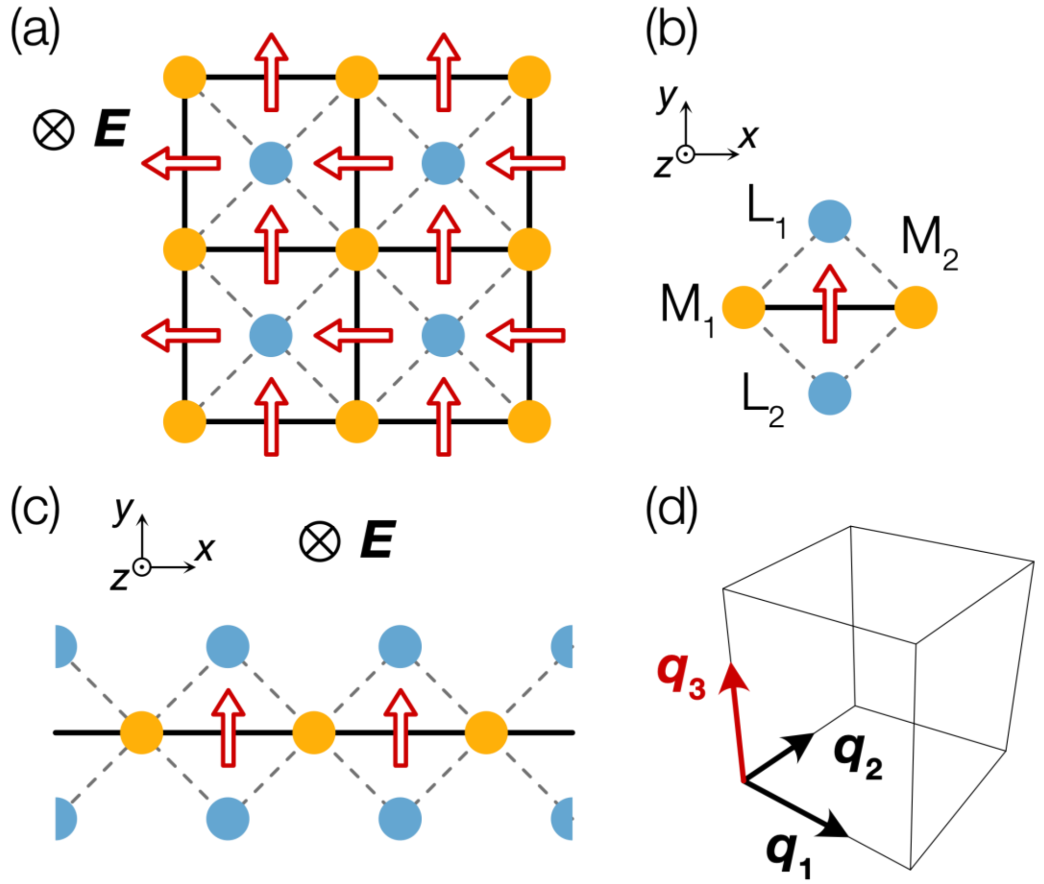
E-driven Néel-type skyrmion lattice expected.

Other applications

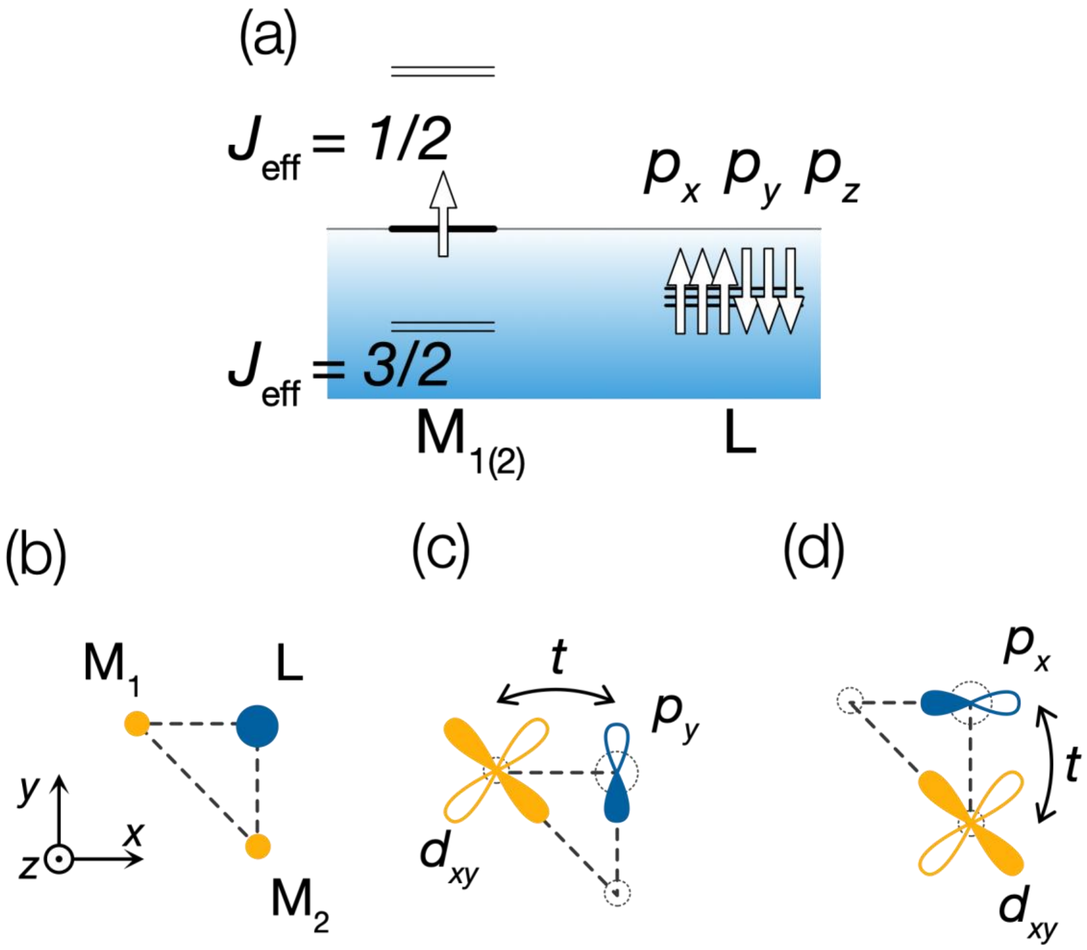
- Chiral soliton lattice.
  - 1D version of the square-lattice model.

- Magnetic hedgehogs.
  - A multiple-q state.
  - Suppose double-q states.
  - The electric field can drive a uniform DM interaction that leads to another q vector.





Case 1 : Kitaev magnet with intra-atom SO coupling



- Three-site models with two magnetic ions and one ligand.
- Low-spin d^5 configuration.
- $J_{\mathrm{eff}} = 1/2$ doublet carries a (pseudo)spin-1/2.
- Hubbard-like Hamiltonian with large on-site Coulomb repulsions.
- Our model is akin to the model used by Jackeli and Khaliullin [PRL (2009)] to realize the Kitaev model on the honeycomb lattice.

Large-U expansion : Kitaev-Heisenberg-Γ'-DM model

- W/o the DC electric field:

$$H_{\mathrm{spin}} = -|J|\vec{S}_1 \cdot \vec{S}_2 + K S_1^z S_2^z - \vec{h} \cdot \sum_j \vec{S}_j$$

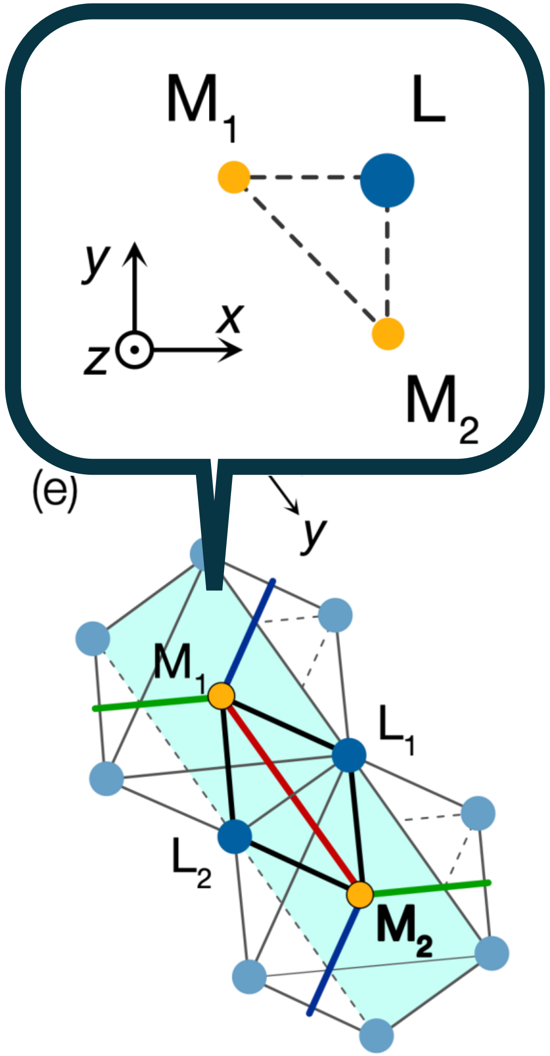
cf. G. Jackeli and G. Khaliullin, PRL (2009)

- W/ $\vec{E} = E^x \vec{e}_x + E^y \vec{e}_y$:

$$H_{\mathrm{spin}} = -|J|\vec{S}_1 \cdot \vec{S}_2 + K S_1^z S_2^z - \vec{h} \cdot \sum_j \vec{S}_j \quad + \vec{D} \cdot \vec{S}_1 \times \vec{S}_2$$

- W/ $\vec{E} = E^z \vec{e}_z$:

$$H_{\mathrm{spin}} = -|J|\vec{S}_1 \cdot \vec{S}_2 + K S_1^z S_2^z - \vec{h} \cdot \sum_j \vec{S}_j$$
$$+ \vec{D} \cdot \vec{S}_1 \times \vec{S}_2 + \Gamma' [S_1^z (S_2^x + S_2^y) + (S_1^x + S_1^y) S_2^z]$$

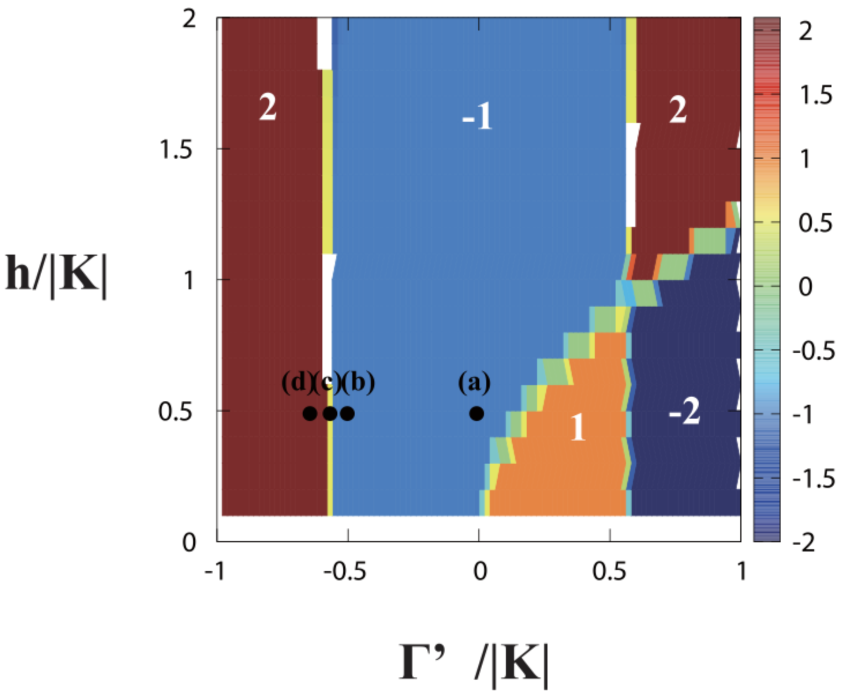


$\boldsymbol{\Gamma'}$ term and magnetic field cooperatively opens Majorana gap and induces QPTs.

- Γ' interaction: $$\Gamma' = \frac{32t^3}{9} I E^z \left(\frac{1}{U_d - U_p + \Delta_{dp}}\right)^2 \frac{U_d - U_p + \Delta_{dp}}{4(U_d - U_p + \Delta_{dp})^2 - {J_\mathrm{H}}^2}$$

- Perturbative expansion:
  - Second-order term: $-\frac{h^x\Gamma'}{K}\sum S_i^x S_j^y S_k^z$.
  - The Majorana gap $\frac{h^x\Gamma'}{K} \propto \frac{h^x E^z}{K}$

- $\vec{E}$ will change the Chern number of the chiral quantum spin liquid.

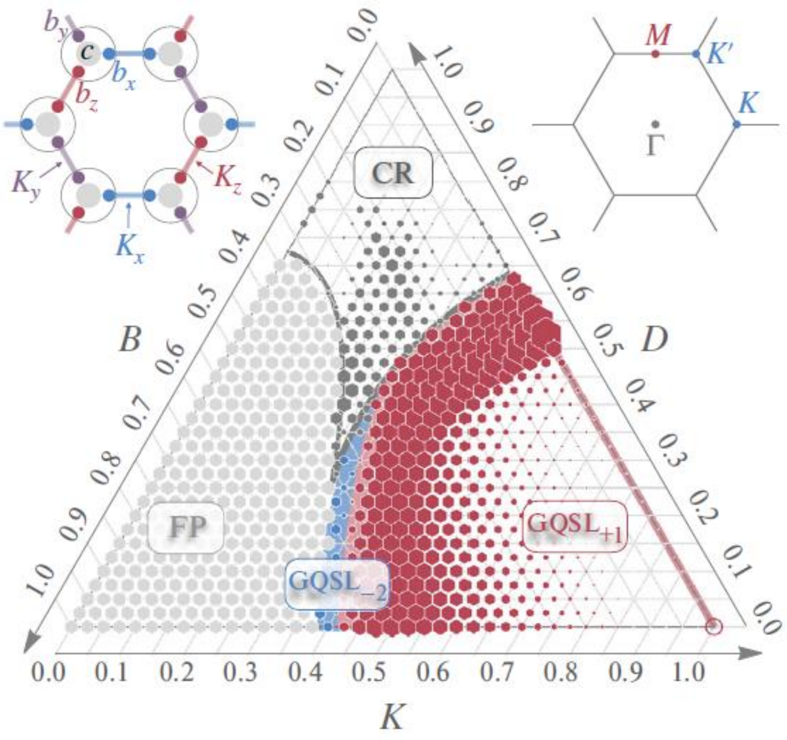


D. Takikawa and S. Fujimoto, PRB (2020)

DM interaction also leads to QPTs in Kitaev model

$$H = H_K + H_{\rm DM} + H_B$$

Kitaev interaction
+ DM interaction
+ Zeeman interaction



Ralko and Merino, PRL (2020)

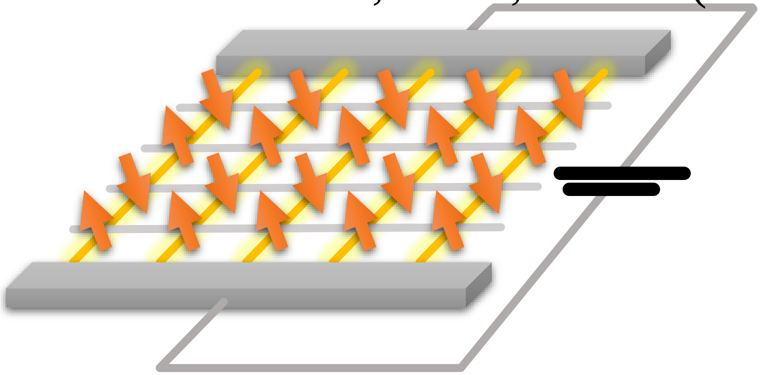
Summary

Summary 1

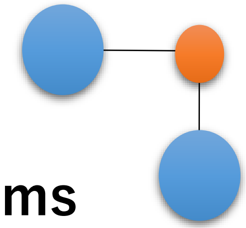
[1] K. Takasan and MS, PRB**100**, 060408(R) (2019).
[2] S. C. Furuya, K. Takasan and MS, PRR**3**, 033066 (2021).

Setup : **Mott insulator + DC Electric field**

(without current, No “Mott breakdown”)



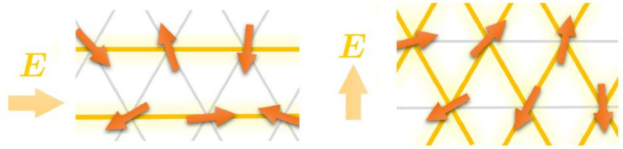
Control direct- and super-exchange interactions with a DC Electric field



(e.g.) Applications of Control of exchange interactions in many-spin systems

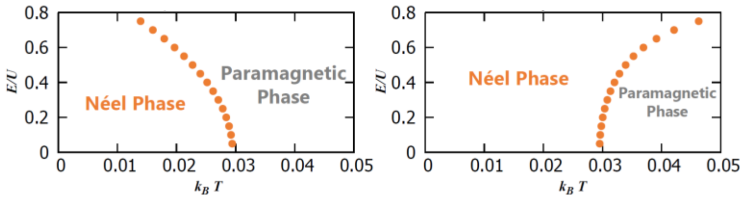
1. Frustrated Magnets

E-field-induced magnetic order and QSL



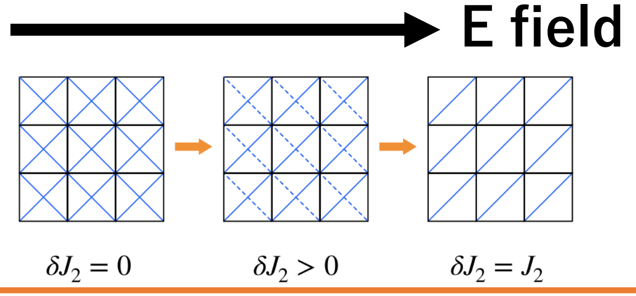
2. Quasi-1D Magnets

E-field-induced magnetic and top. order



3. Frustrated ferromagnets

E field changes models



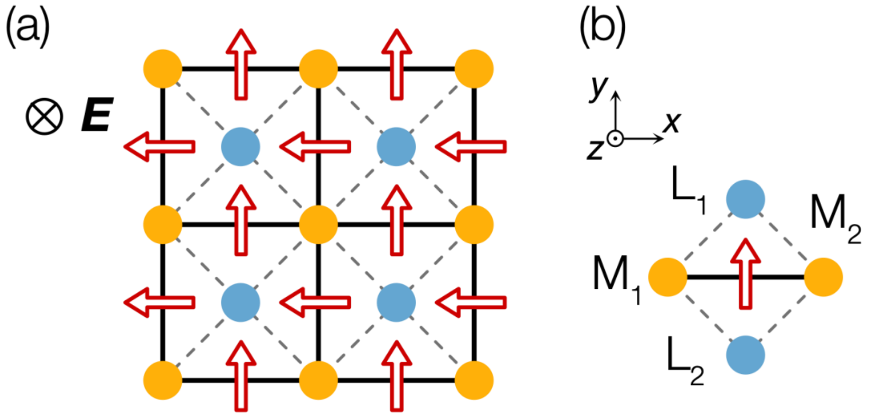
Summary 2

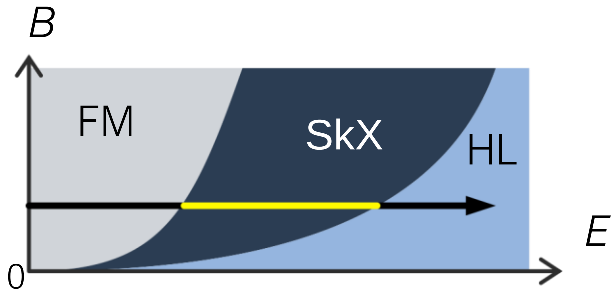
[3] S. C. Furuya and MS, arXiv:2110.6503.

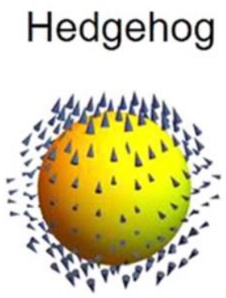
Control magnetic anisotropies with a DC Electric field

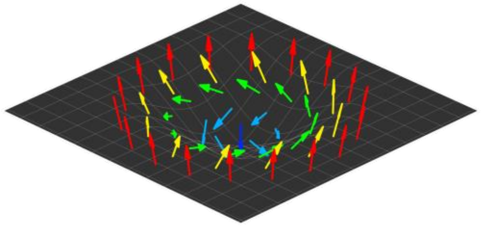
Case 2: Inter-atom Rashba SO coupling

- The DC electric field can yield DM interactions and drive quantum phase transitions from collinear phases to topological spin texture phases.









Case 1: Intra-atom SO coupling in Kitaev model

- The DC electric field can yield DM interactions and off-diagonal symmetric magnetic anisotropies $\Gamma'(S^aS^b + S^bS^a)$ to Kitaev materials such as α-$RuCl_3$.

$$\mathcal{H} = \sum_a \sum_{\langle i,j\rangle_a} (-K_a S_i^a S_j^a + J\vec{S}_i \cdot \vec{S}_j) + \Gamma' \sum_a \sum_{\langle i,j\rangle_a} \sum_{b \neq a} (S_i^a S_j^b + S_i^b S_j^a)$$