Optical Manipulation of Magnetism in a Correlated Electron System

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New Frontier of Strongly Correlated Electron Material,
August 6-24, 2018 Kavli ITS Beijing, China

Outline

[1] Excitonic insulating state in a correlated material as an orbital physics

J. Nasu (Tokyo Tech.), M. Naka (Waseda Univ.)
T. Tatsuno (Tohoku Univ.), T. Watanabe (Chiba Tech.)

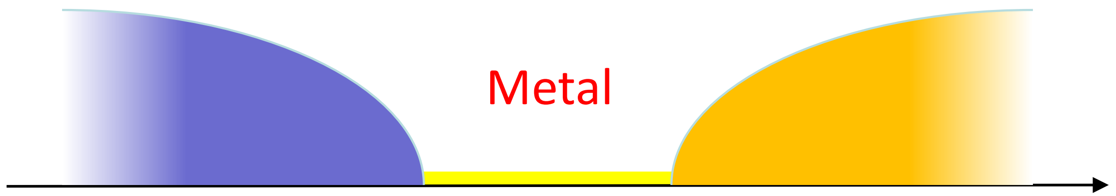
Phys. Rev. B **93**, 205136 (2016)
J. Phys. Soc. Jpn. 85, 083706 (2016)

[2] Double exchange interaction in non-equilibrium state

A. Ono (Tohoku Univ.) J. Ohara (Hokkaido Univ.),
Y Kanamori (Tohoku Univ.)

Phys. Rev. Lett. 119, 207202 (2017) (Editors' suggestion)
Phys. Rev. B 88, 085107 (2013)

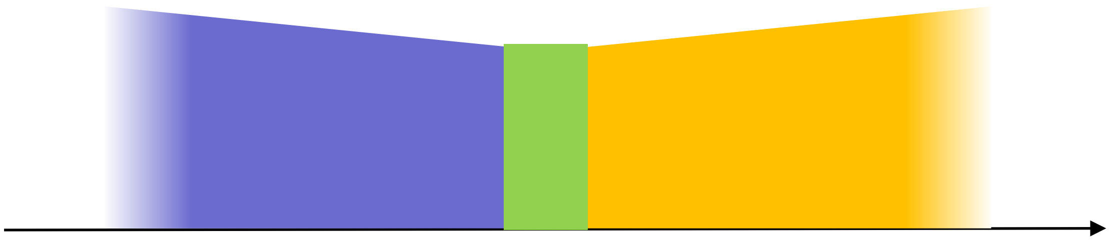
Band insulator v.s. Mott insulator



Band Insulator

Mott Insulator

Metal



Band Insulator

Mott Insulator

Another type
of insulator

Excitonic insulator (EI)

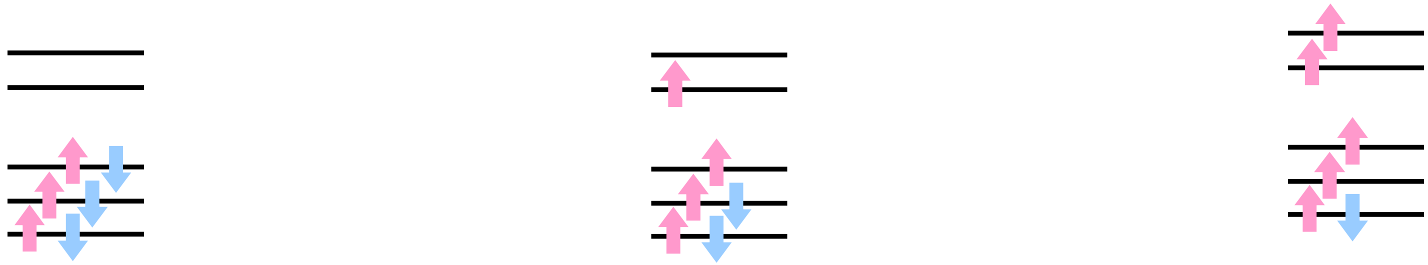
Perovskite cobaltites

$LaCoO_3$

Spin state degree of freedom in Co ion

Co^{3+}

(d^6)

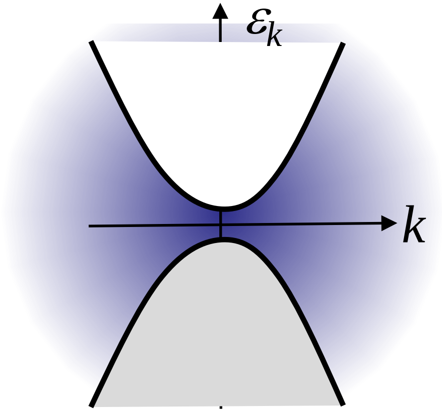


Low spin
LS
(S=0)

Intermediate spin
(IS) (S=1)

High spin
(HS)
(S=2)

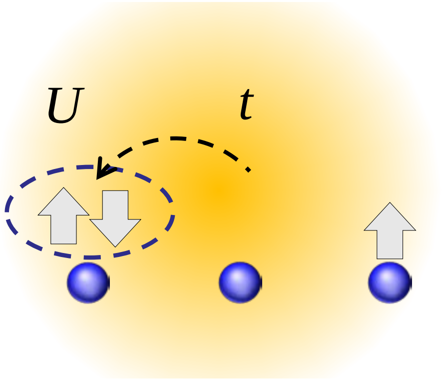
Band Insulator



Level splitting
Δ

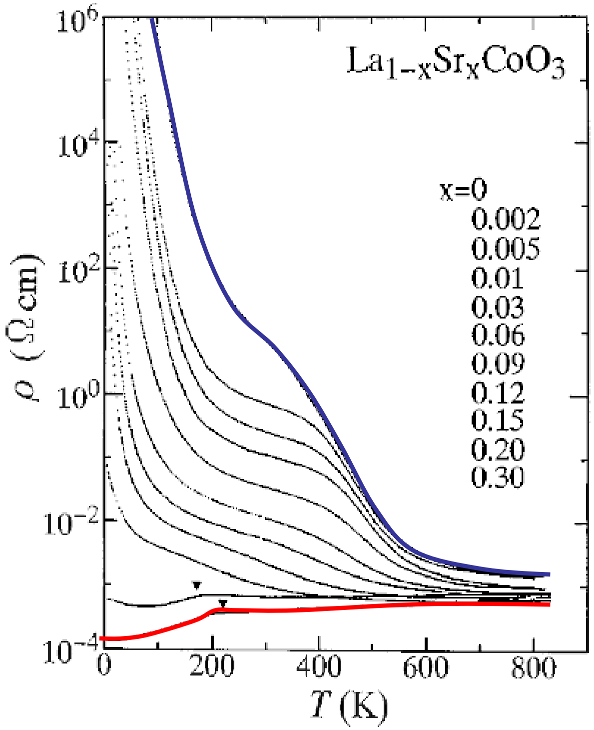
Hund coupling
J

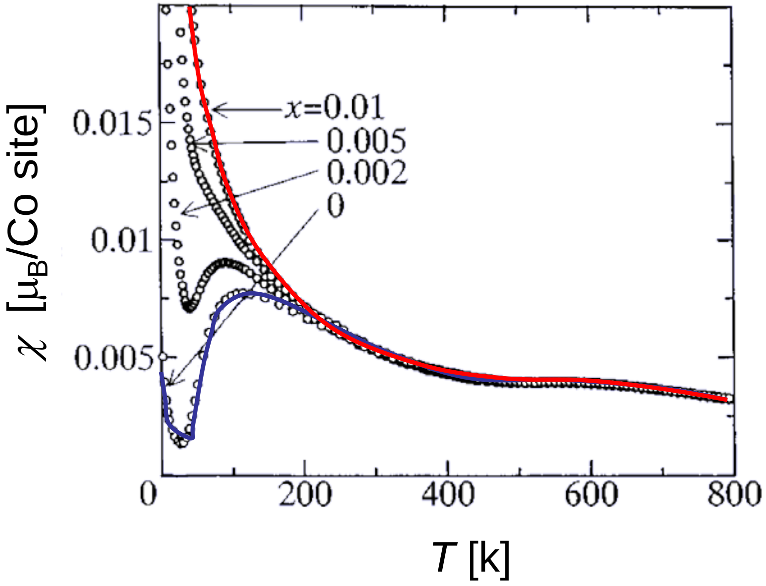
Mott Insulator



Perovskite cobaltites

$La_{1-x}Sr_xCoO_3$



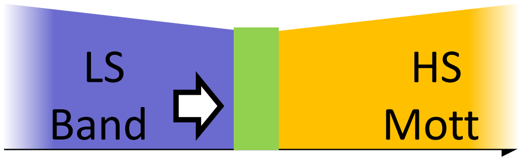


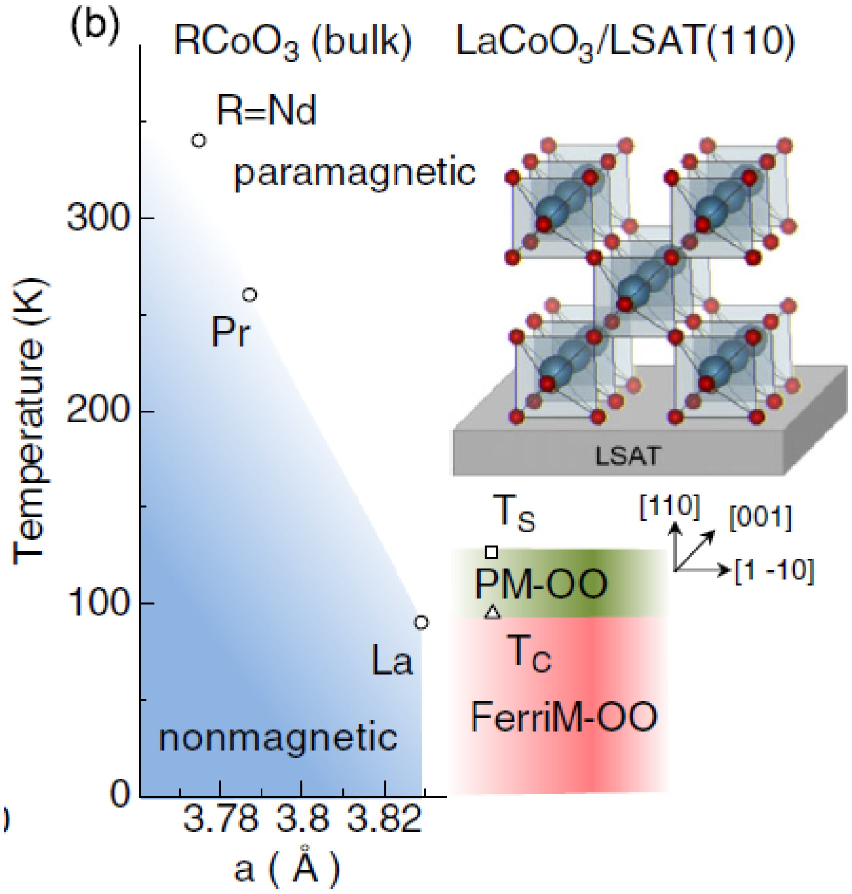
Tokura *et al.* PRB **58** R1699 (1998)

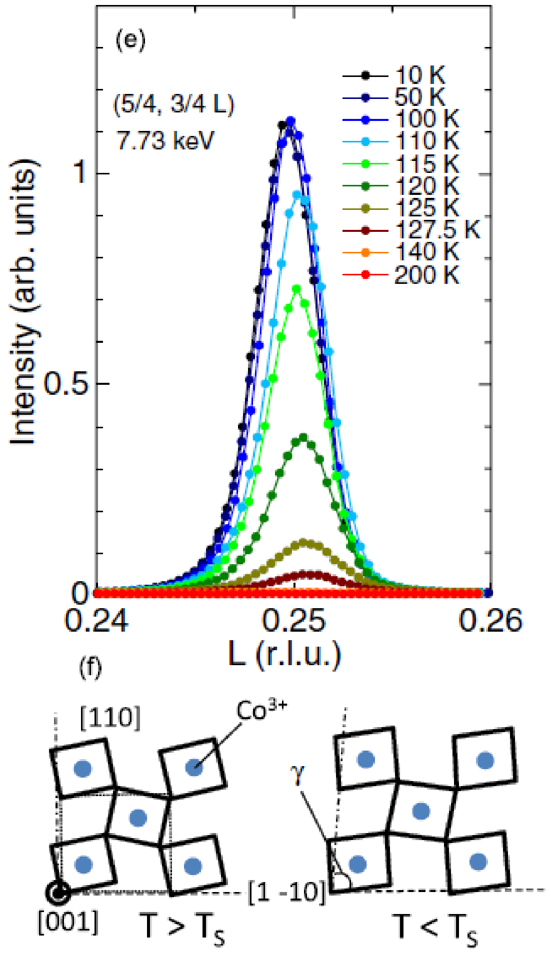
- $LaCoO_3$: LS Insulator to HS (IS) metal with increasing T
- LS Insulator to FM metal with x

Strain on thin film

J. Fujioka et al.
PRL 111, 027206 (2013)



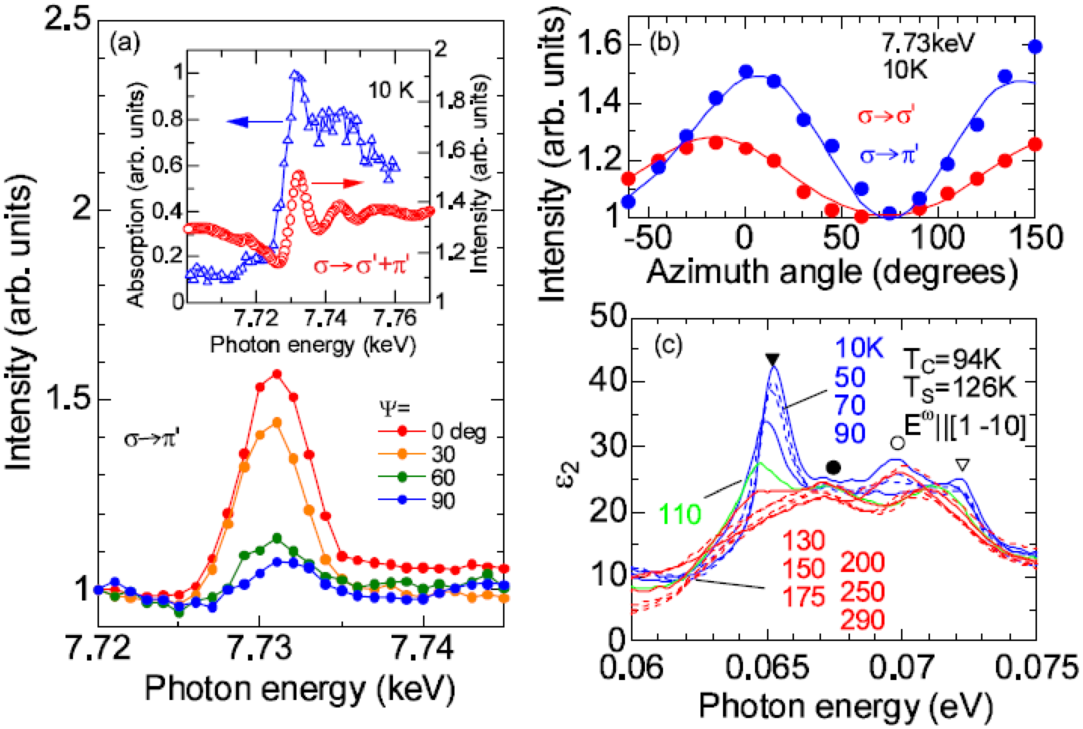


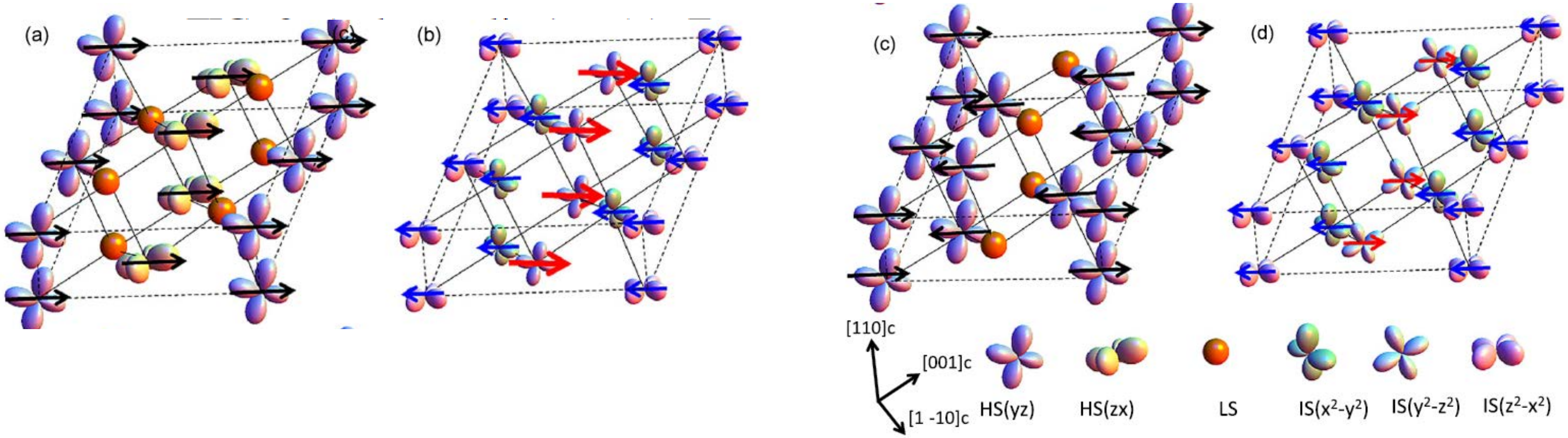


Strain on thin film

RXS @ Co K

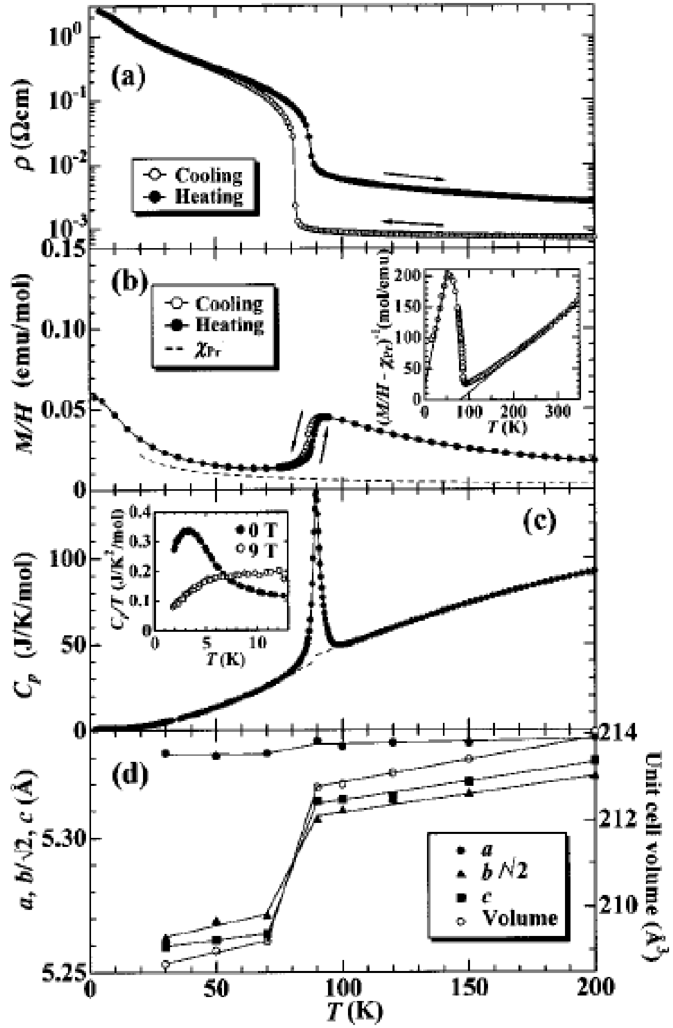
J. Fujioka et al.
PRL 111, 027206 (2013)





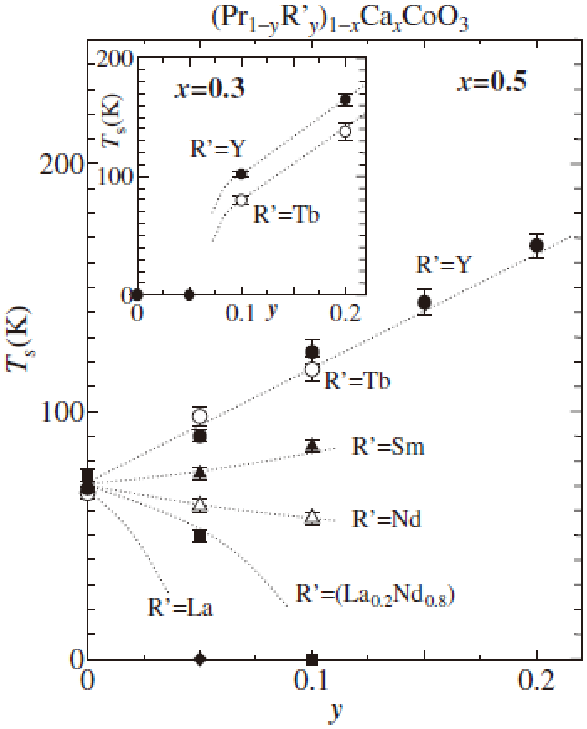
Ion substitution (II)

$R_{1-x}A_xCoO_3$ (R: Pr A: Ca, Sr, Ba)



Tsubouchi-Itoh et al. Phys. Rev. B **66**, 052418 (2002)

Fujita-Satoh et al. J. Phys. Soc. Jpn. 73, 1987(2004)



Probably

Pr^{4+}

Co^{3+}

J. Kuneš and P. Augustinský PRB 89, 115134 (2014)

J. Kuneš and P. Augustinský PRB 90, 235112 (2014)

a candidate of excitonic insulator (EI)

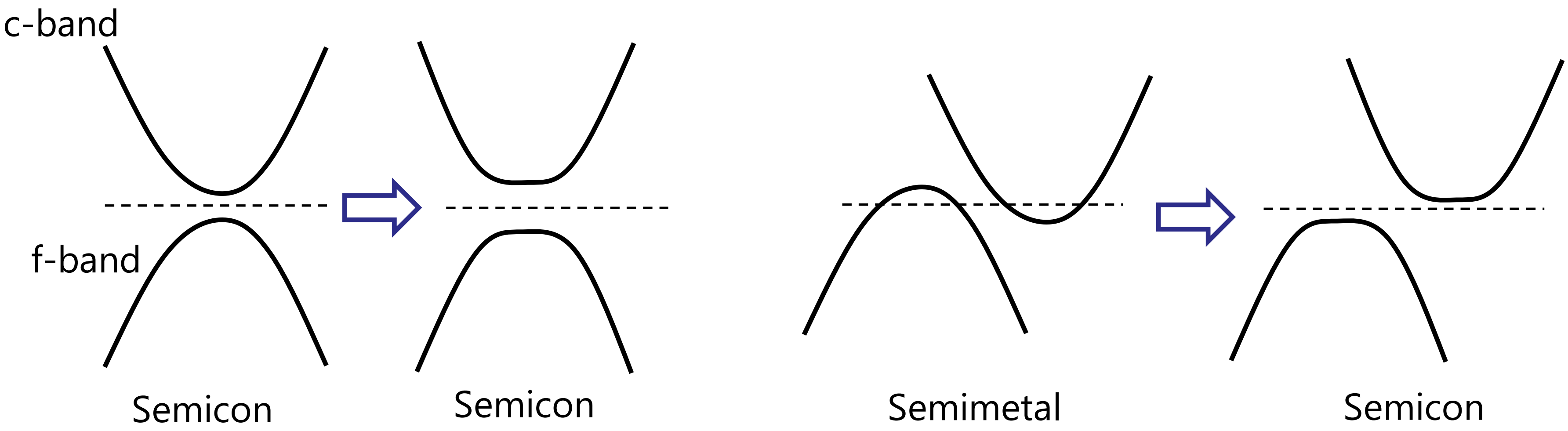
Excitonic Insulators

Semiconductor, Semimetal

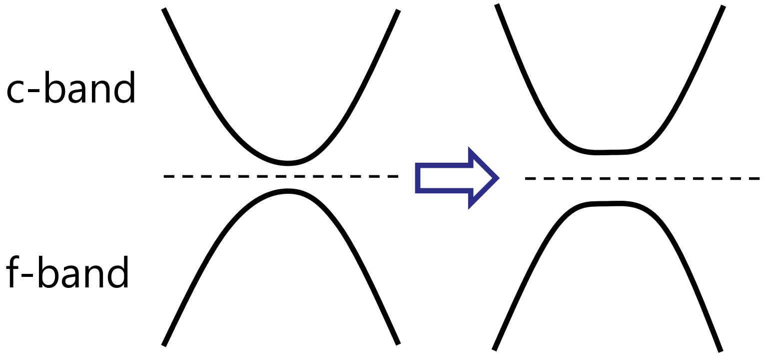
Electron-Hole binding energy > band gap

Condensation of macroscopic number of excitons

Mott(61) Knox (63) Keldysh(65), Jerome-Rice-Khon (1967)
Halperin, Rice, Solid State Physics, 21 (1968)
Fukuyama (1971), Kuramoto(1978)



Excitonic Insulators



Different symmetries in c & f bands No direct hybridization

$$\cancel{\mathcal{H}_t = -\sum_{<ij>\sigma} t\left(c^\dagger_{i\sigma} f_{j\sigma} + H.c.\right)}$$

Order parameter

$$N^{-1}\langle \sum_i c_i^\dagger f_i \rangle e^{iQr_i}$$

$$|EI\rangle \sim N^{-1}\sum_i \left(u + v c_i^\dagger f_i\right)|0\rangle$$

Spontaneous symmetry breaking

$$c^\dagger c f^\dagger f \rightarrow -c^\dagger f \langle f^\dagger c\rangle$$

Analogy with Superconductivity

Non-conserved

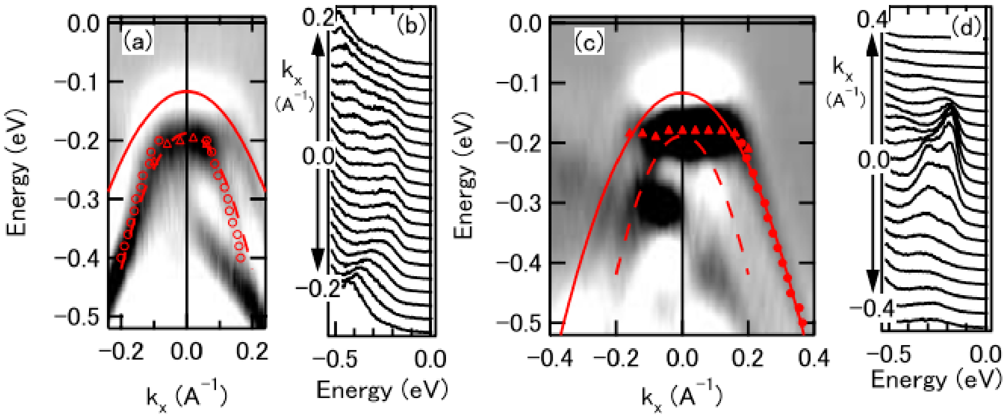
$\langle f^\dagger f\rangle - \langle c^\dagger c\rangle$ (EI)

$\langle f^\dagger f\rangle + \langle c^\dagger c\rangle$ (SC)

Excitonic Insulators

Ta_2NiSe_5

- Flat dispersion observed in ARPES

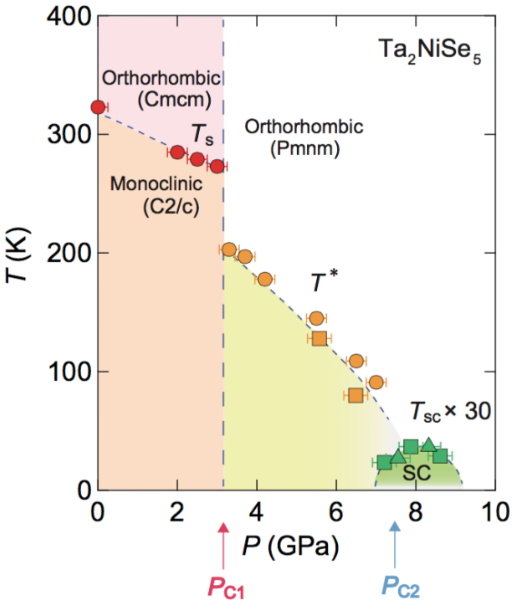


Y. Wakisaka et al., PRL 103, 026402 (2009).

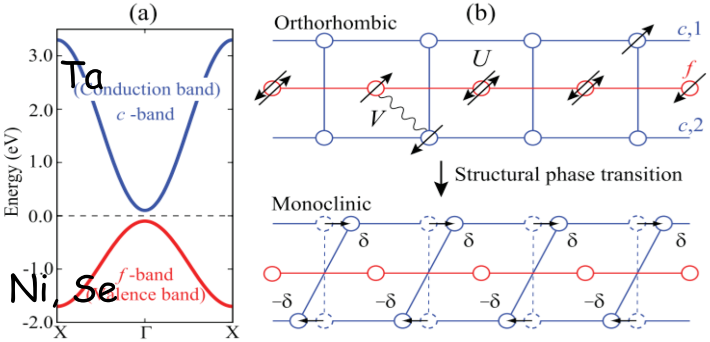
Y. Wakisaka et al., J. Supercond. Nov. Magn. 25, 1231 (2012).

T. Kaneko, T. Toriyama, T. Konishi, and Y. Ohta, PRB 87, 035121 (2013).

T. Kaneko and Y. Ohta, PRB 90, 245144 (2014).



図：Ta_2NiSe_5の圧力–温度相図



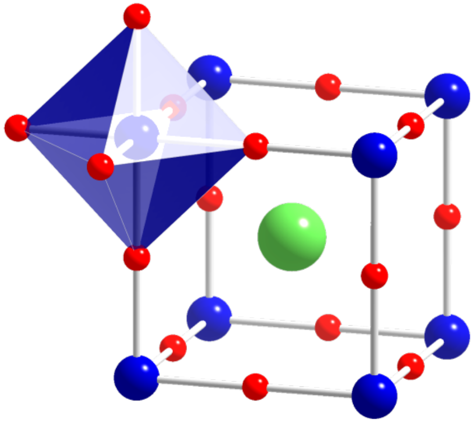
Approach from Band Ins.

1T−$TiSe_2$

J. Ishioka et al, PRL. 105, 176401 (2010).
H. Watanabe, K. Seki, and S. Yunoki, PRB 91, 205135 (2015).

Mott physics / Mottness (?)

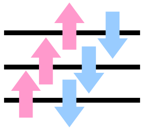
Perovskite cobaltites



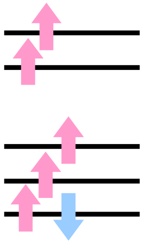
Spin state degree of freedom

Co^{3+}

(d^6)



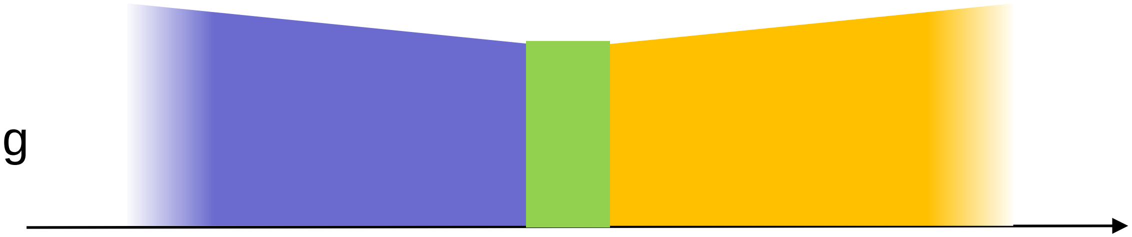
Low spin
LS
(S=0)



High spin
(HS)
(S=2)

Band Insulator

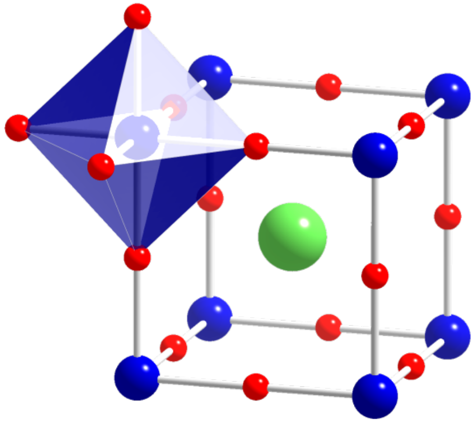
Mott Insulator

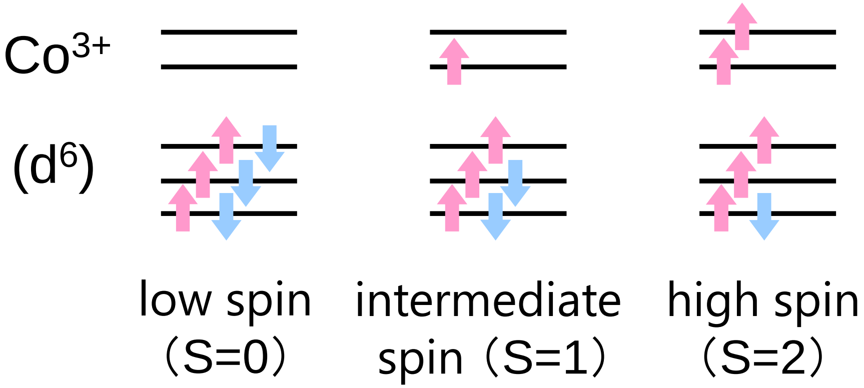


Level splitting
Δ

Hund coupling
J

Theoretical approaches





5 orbital Hubbard model

Weak coupling approach

Hartree-Fock
Phase diagram
Collective mode

2 orbital Hubbard model

Strong coupling approach

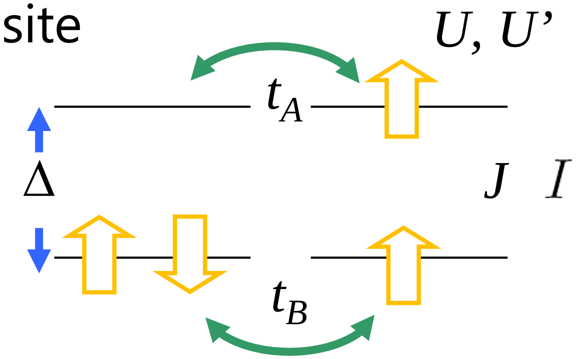
Low energy effective model

Phase diagram
Collective mode

Two band Hubbard with energy difference

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2$$

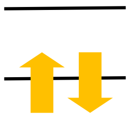
2 electrons/ site



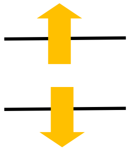
Energy difference

Transfer

$$\mathcal{H}_1 = \Delta \sum_{i,m=\{e_g\}\sigma} c^\dagger_{im\sigma} c_{im\sigma} + \sum_{\langle i,j\rangle,m,\sigma} t_{mm'} \left(c^\dagger_{im'\sigma} c_{jm\sigma} + \mathrm{H.c.} \right)$$

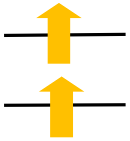


Intra/inter band Coulomb



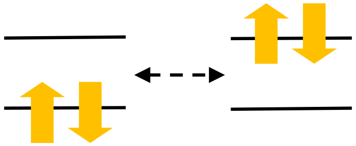
$$\mathcal{H}_2 = U \sum_{i,m} c^\dagger_{im\uparrow} c_{im\uparrow} c^\dagger_{im\downarrow} c_{im\downarrow} + U' \sum_{i,m\neq m',\sigma\sigma'} c^\dagger_{im\sigma} c_{im\sigma} c^\dagger_{im'\sigma'} c_{im'\sigma'}$$

$$+ J \sum_{i,m\neq m',\sigma\sigma'} c^\dagger_{im\sigma} c_{im'\sigma} c^\dagger_{im'\sigma'} c_{im\sigma'} + I \sum_{i,m\neq m'} c^\dagger_{im\uparrow} c_{im'\uparrow} c^\dagger_{im\downarrow} c_{im'\downarrow}$$



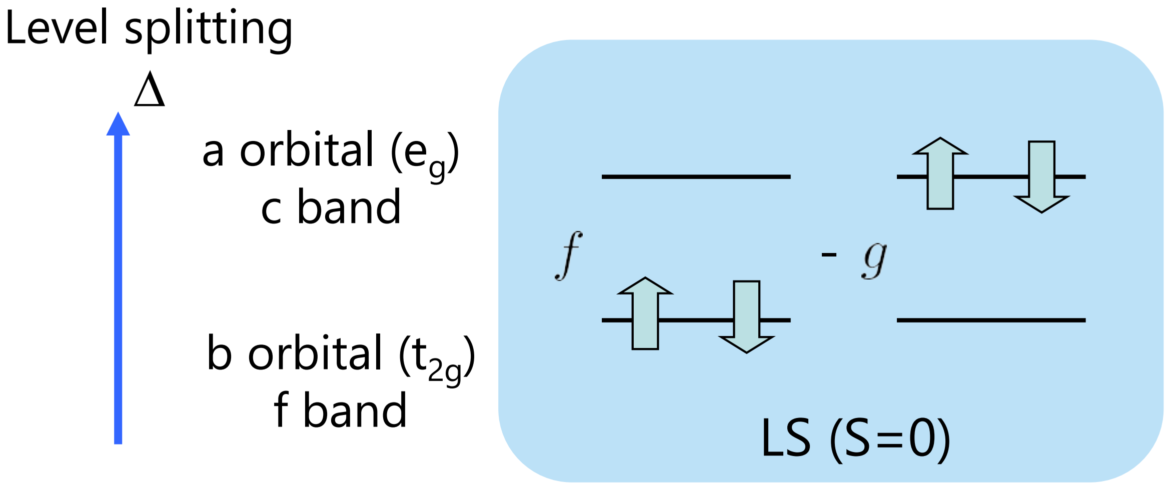
Hund coupling

Pair hopping



(sama order of magnitudes)

Local states



Level splitting

Δ

a orbital (e_g)
c band

b orbital (t_{2g})
f band

f - g

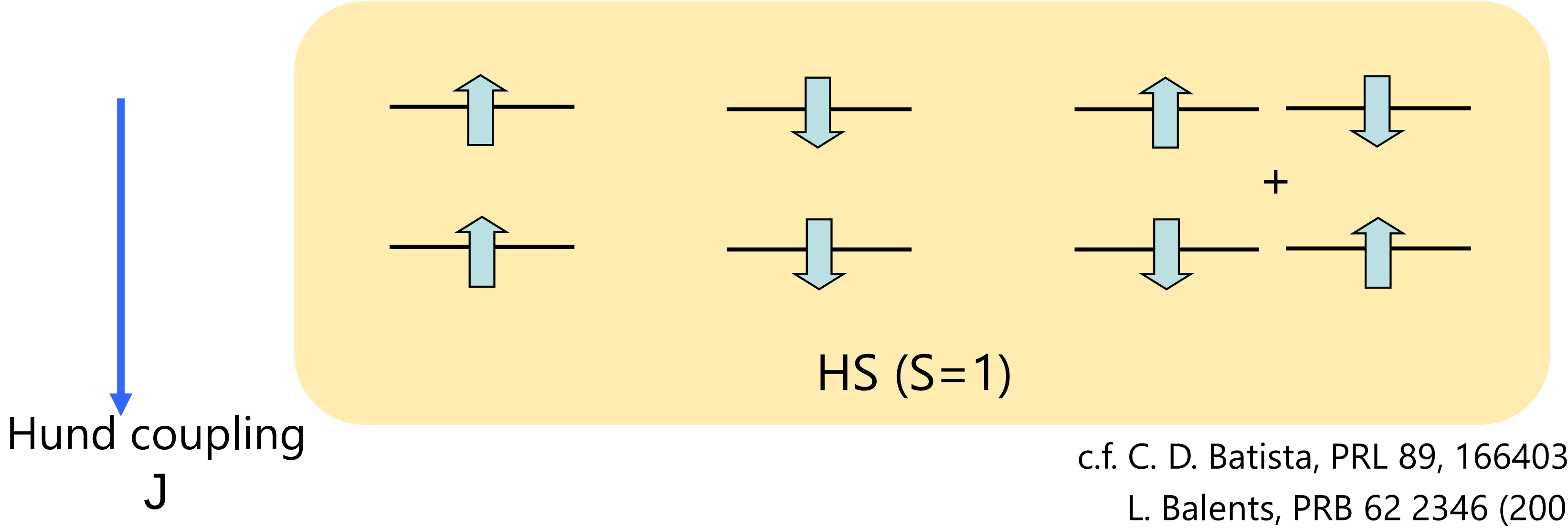
LS (S=0)

Strong coupling approach

$$\Delta E \sim \Delta, J >> t$$

$$f \sim 1 \ g \sim I/\Delta$$

If (pair hopping)I=0,
then g=0



+

HS (S=1)

Hund coupling
J

c.f. C. D. Batista, PRL 89, 166403 (2002)
L. Balents, PRB 62 2346 (2000)

Psudo-spins for excitonic state

Pseudo-spin operator

EI order parameter

orbital

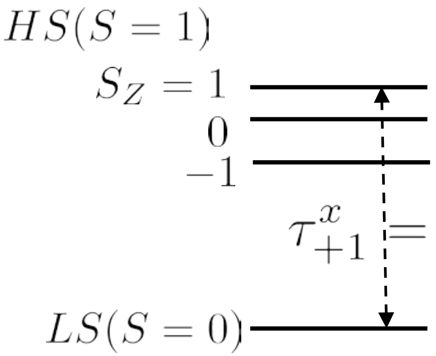
spin

$$\tau^{x}_{S_z} = \left(|HS(S_z)\rangle\langle LS| + |LS\rangle\langle HS(S_z)|\right) \qquad \sim f^{\dagger}c + c^{\dagger}f$$

$$\tau^{y}_{S_z} = -i\left(|HS(S_z)\rangle\langle LS| - |LS\rangle\langle HS(S_z)|\right) \qquad \sim f^{\dagger}c - c^{\dagger}f$$

$$\tau^{z}_{S_z} = |HS(S_z)\rangle\langle HS(S_z)| - |LS\rangle\langle LS| \qquad \sim f^{\dagger}f - c^{\dagger}c$$

$$\tau^{x}_{-1} = \frac{1}{\sqrt{2}} \begin{array}{c} \begin{array}{cccc} & HS & & LS \\ S_Z=1 & 0 & -1 & \end{array} \\ \left(\begin{array}{ccc|c} & & & \\ & & & \\ & & & 1 \\ \hline & & 1 & \end{array}\right) \end{array} \qquad \tau^{y}_{X} = \frac{1}{\sqrt{2}} \left(\begin{array}{ccc|c} & & & i \\ & & & \\ & & & -i \\ \hline -i & & i & \end{array}\right)$$



$$\tau^{x}_{+1} = c^{\dagger}_{\uparrow} f_{\downarrow} + H.c.$$

Low energy model

$$\mathcal{H}_{eff} = E_0 - h_z \sum_i \tau_i^z + J_s \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j + J_z \sum_{<ij>} \tau_i^z \tau_j^z$$

Band gap

LS-HS int.

$$+ J_x \sum_{<ij>\Gamma} \tau_{i\Gamma}^x \tau_{j\Gamma}^x + J_y \sum_{<ij>\Gamma} \tau_{i\Gamma}^y \tau_{j\Gamma}^y$$

$$\tau^x = \sum_\Gamma \tau_\Gamma^x$$

Exciton-exciton interaction

XYZ-like model with transverse field

If no pair-hopping, then $J_x = J_y$ XXZ-like model with transverse field

Y. Kanamori, H. Matsueda and S. Ishihara
Phys. Rev. Lett. 107, 167403 (2011), Phys. Rev. B 86, 045137 (2012)

J. Kuneš and P. Augustinský PRB 89, 115134 (2014), PRB 90, 235112 (2014)

C. D. Batista, PRL 89, 166403 (2002)
L. Balents, PRB 62 2346 (2000)
G. Khalliuline, PRL 111 197201(2013)

Symmetry

$$\mathcal{H}_{eff} = E_0 - h_z \sum_i \tau_i^z + J_s \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j + J_z \sum_{<ij>} \tau_i^z \tau_j^z$$
$$+J_x \sum_{<ij>\Gamma} \tau_{i\Gamma}^x \tau_{j\Gamma}^x + J_y \sum_{<ij>\Gamma} \tau_{i\Gamma}^y \tau_{j\Gamma}^y$$

Symmetry & Conservation

- ☐ $S^x,\ S^y,\ S^z$ Total spin angular momentum $O(3)$
- ☐ $\sum_i (n_i^a + n_i^b)$ Total electron number $U(1)$
- ☐ If no pair-hopping $J_x = J_y$

  $\sum_i \tau_i^z \sim \sum_i (n_i^a - n_i^b)$

  Electron number difference between c/f bands $U(1)$

  Relative phase $a|HS\rangle + e^{i\theta} b|LS\rangle$
- ☐ $\tau_\Gamma^x \to -\tau_\Gamma^x$ $\tau_\Gamma^y \to -\tau_\Gamma^y$ Z_2 Relative sign $a|HS\rangle \pm b|LS\rangle$

Symmetry of EI order parameter

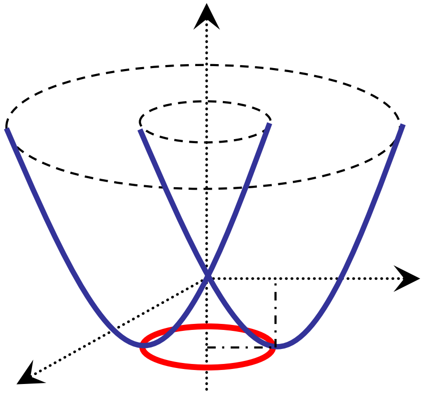
Collective mode and symmetry

☐ If no pair-hopping $J_x = J_y$

$$\sum_i \tau_i^z \sim \sum_i (n_i^a - n_i^b)$$

Electron number difference between c/f bands $U(1)$

Relative phase $a|HS\rangle + e^{i\theta} b|LS\rangle$



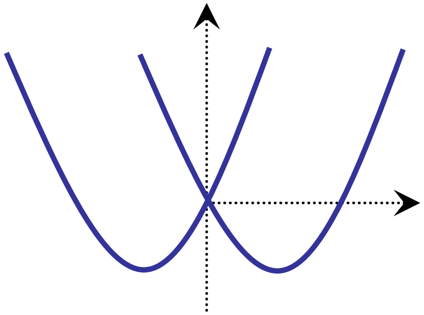
Amplitude (Higgs) mode

Phase mode : Goldstone mode

(similar to SC)

☐ If pair-hopping

$\tau_\Gamma^x \to -\tau_\Gamma^x$ $\tau_\Gamma^y \to -\tau_\Gamma^y$ Z_2 Relative sign $a|HS\rangle \pm b|LS\rangle$

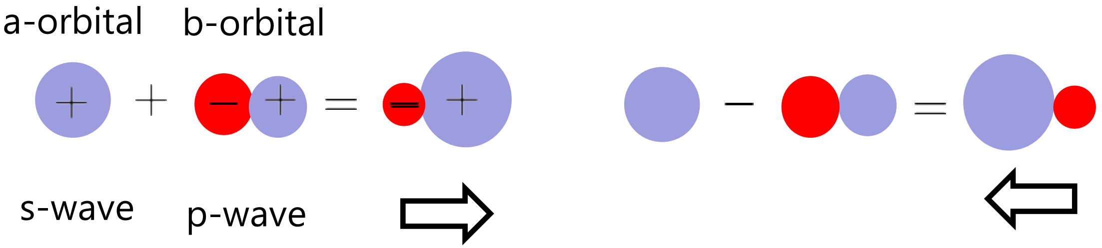


Amplitude (Higgs) mode

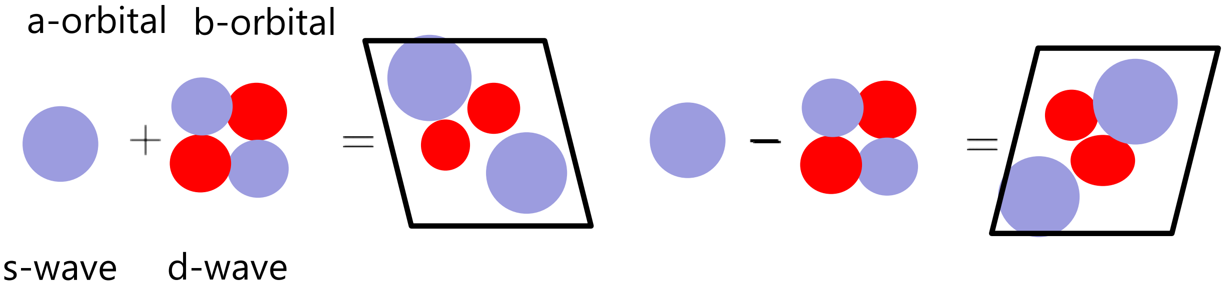
Meaning of sign degree of freedom

From more general point of view

- $\tau_\Gamma^x \to -\tau_\Gamma^x$ $\tau_\Gamma^y \to -\tau_\Gamma^y$ Z_2 Relative sign $a|HS\rangle \pm b|LS\rangle$



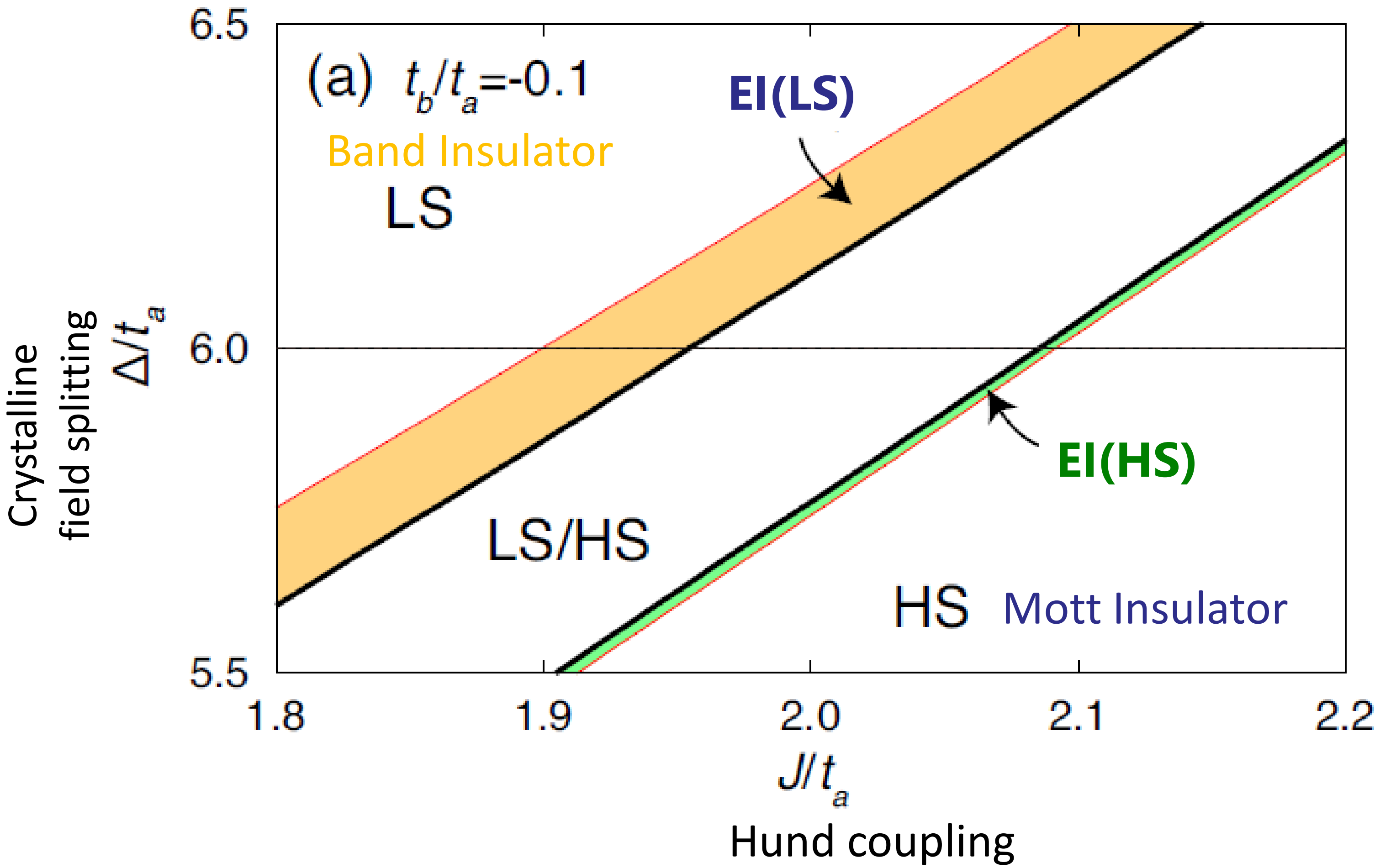
c.f. electronic ferroelectricity



Ferroelastic
Cubic-monoclinic

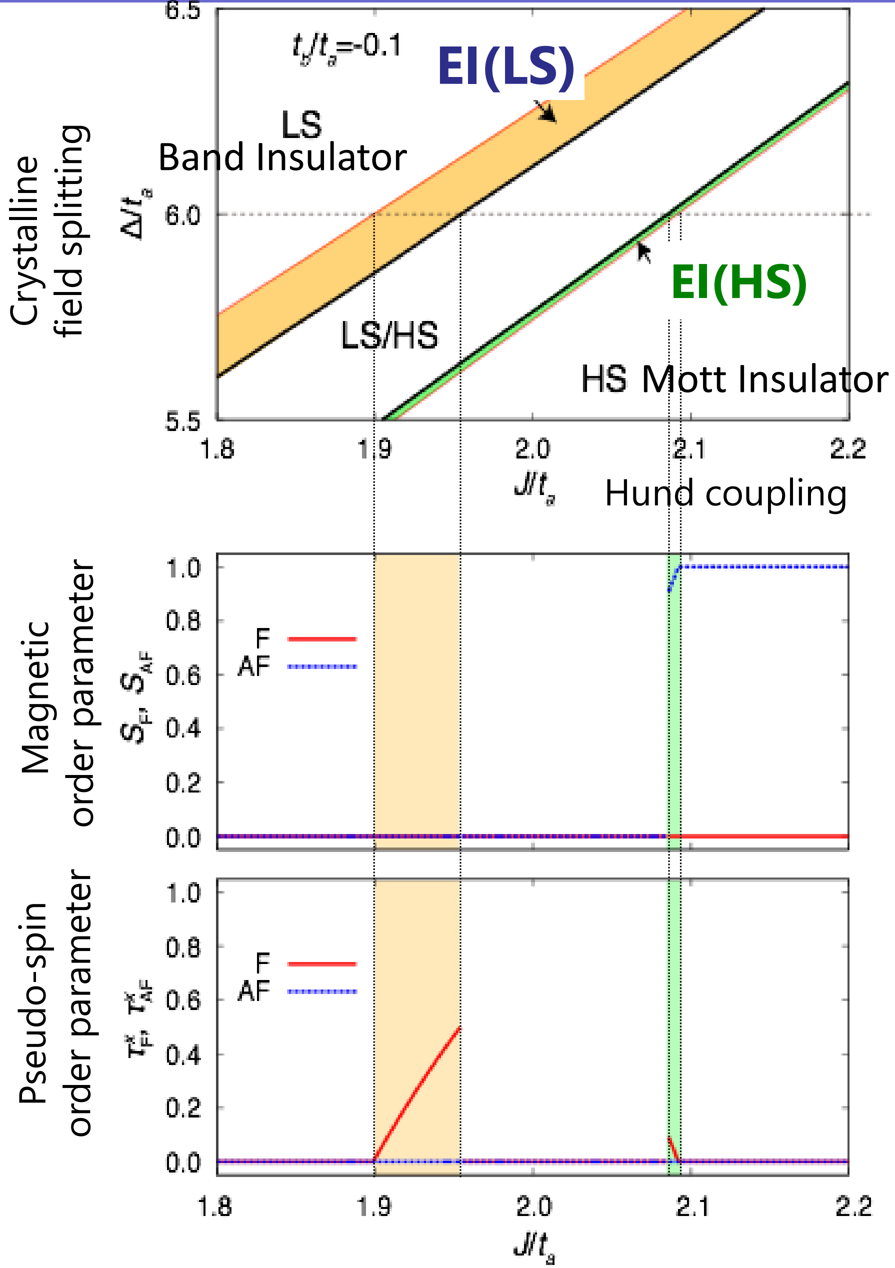
Phase diagram at T=0

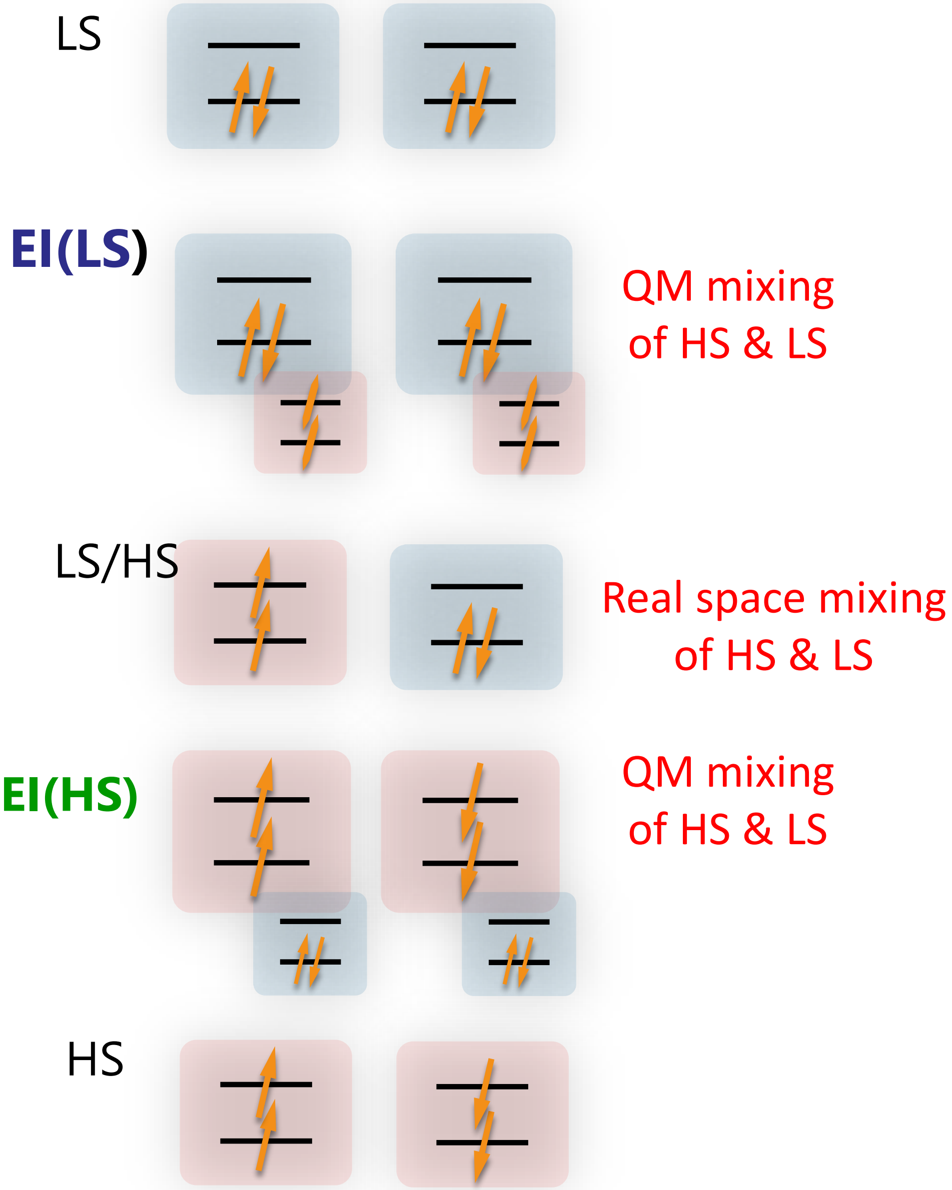
Mean field approximation
2dim square lattice



Phase diagram

Mean field approximation
2dim square lattice





LS

EI(LS)

QM mixing
of HS & LS

LS/HS

Real space mixing
of HS & LS

EI(HS)

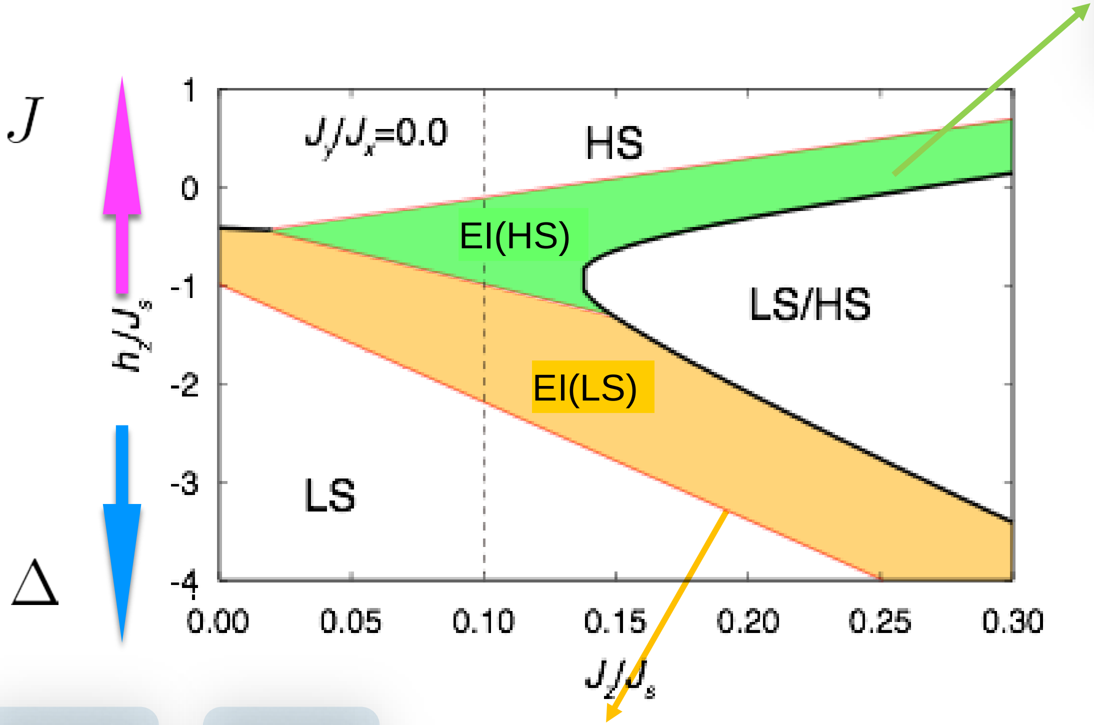
QM mixing
of HS & LS

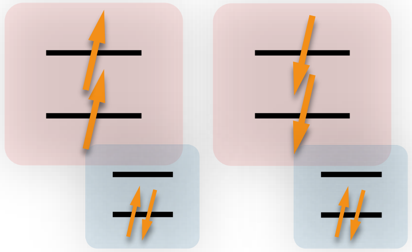
HS

Two EI phases

$$\mathcal{H}_{eff} = E_0 - h_z \sum_i \tau_i^z + J_s \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j + J_z \sum_{<ij>} \tau_i^z \tau_j^z + J_x \sum_{<ij>\Gamma} \tau_{i\Gamma}^x \tau_{j\Gamma}^x + J_y \sum_{<ij>\Gamma} \tau_{i\Gamma}^y \tau_{j\Gamma}^y$$

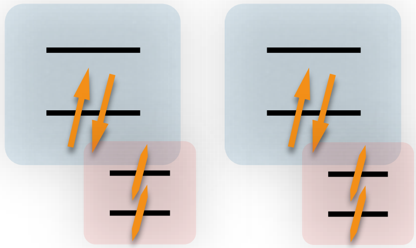
$|J_x|/J_s = 0.5$ $J_x < 0,\ J_y < 0$





$\langle \tau^x \rangle \neq 0$ Pseudo spin: F

$\langle S^z \rangle \neq 0$ Spin: AF

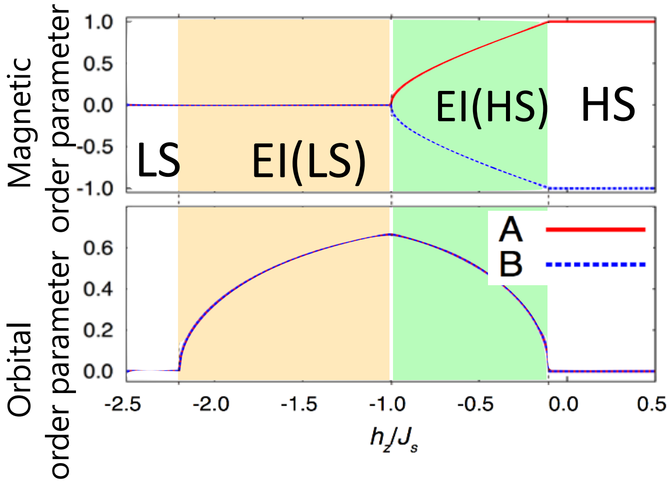


$\langle \tau^x \rangle \neq 0$ $\langle S^z \rangle = 0$

$\langle Q^{x^2-y^2} \rangle = \langle (S^x)^2 - (S^y)^2 \rangle \neq 0$

Pseudo spin: F

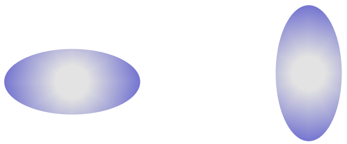
Spin: quadrupole (nematic)



Spin nematic order

$\langle S^\gamma \rangle = 0$

$\langle S^\alpha S^\beta \rangle \neq 0$

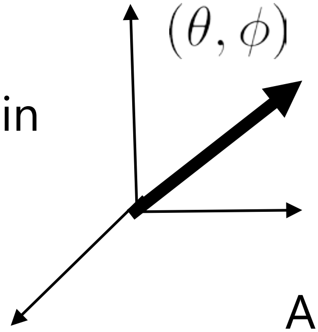


$S = 1$

$\Psi\rangle = a|1\rangle + b|0\rangle + c|-1\rangle$

6-2=4 degrees of freedom

Classical vector for spin



2 degrees of freedom

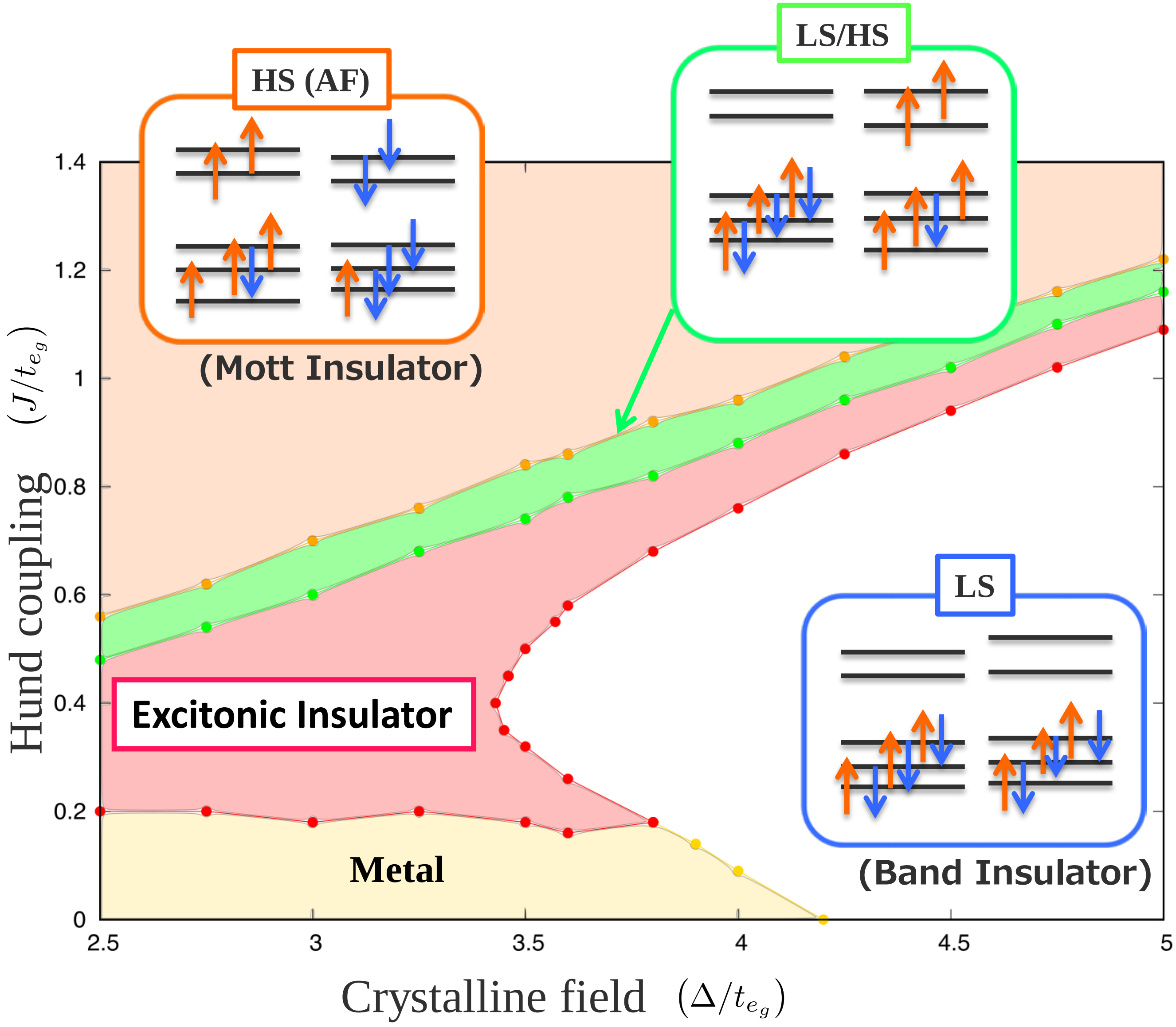
Additional 2 degrees of freedom exit

$NiGa_2S_4$

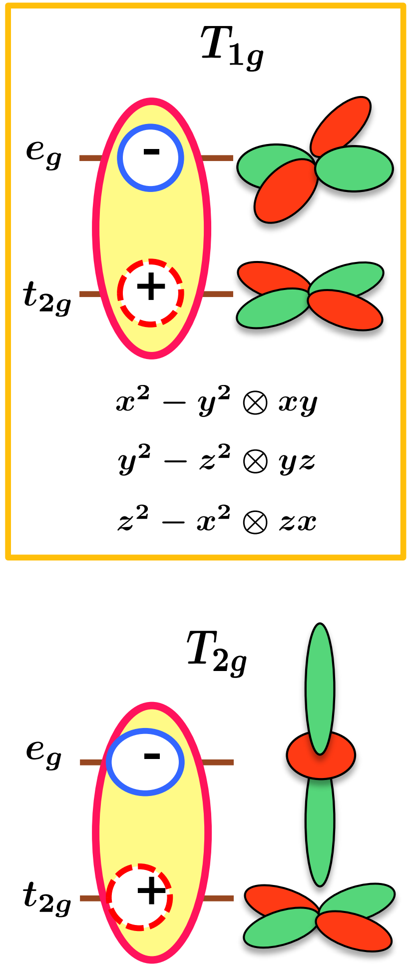
A. Läuchli, F. Mila, and K. Penc, PRL 97, 087205 (2006).

H. Tsunetsugu and M. Arikawa, JPSJ 75, 083701 (2006).

5 orbital model

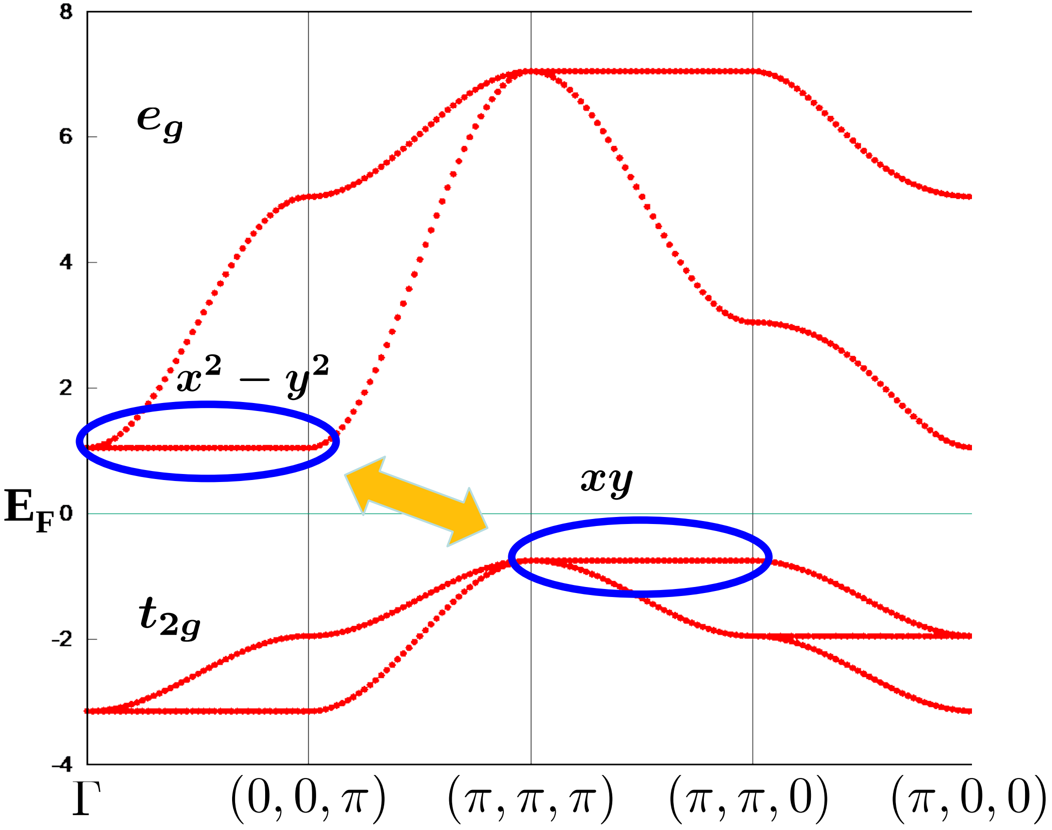


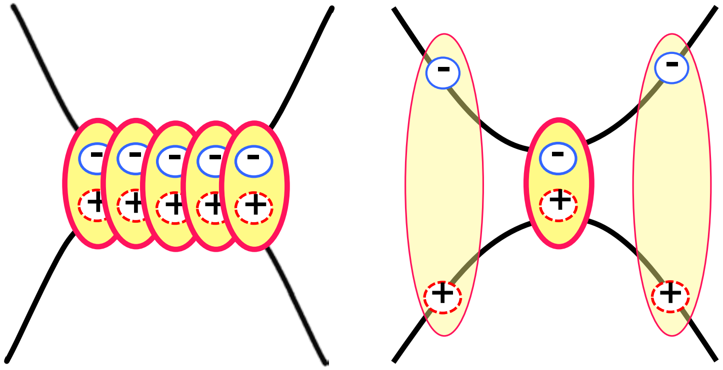
$e_g \otimes t_{2g}$

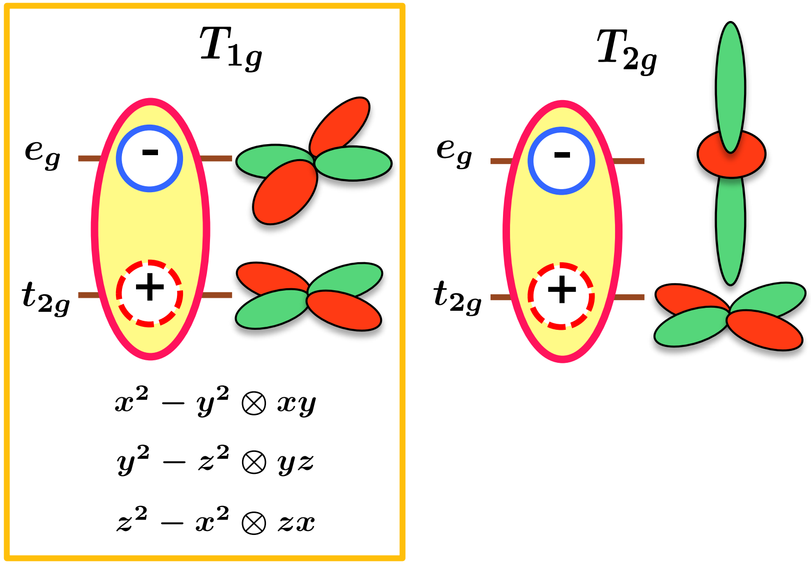


5 orbital model

Non-interacting
electron band





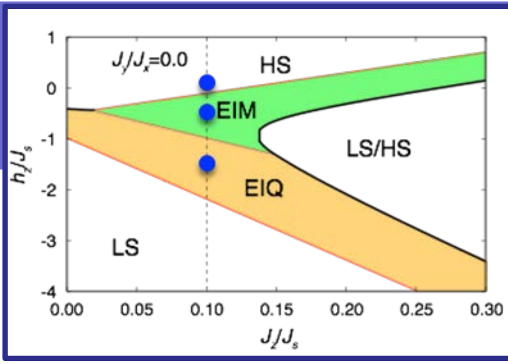


$x^2-y^2 \otimes xy$

$y^2-z^2 \otimes yz$

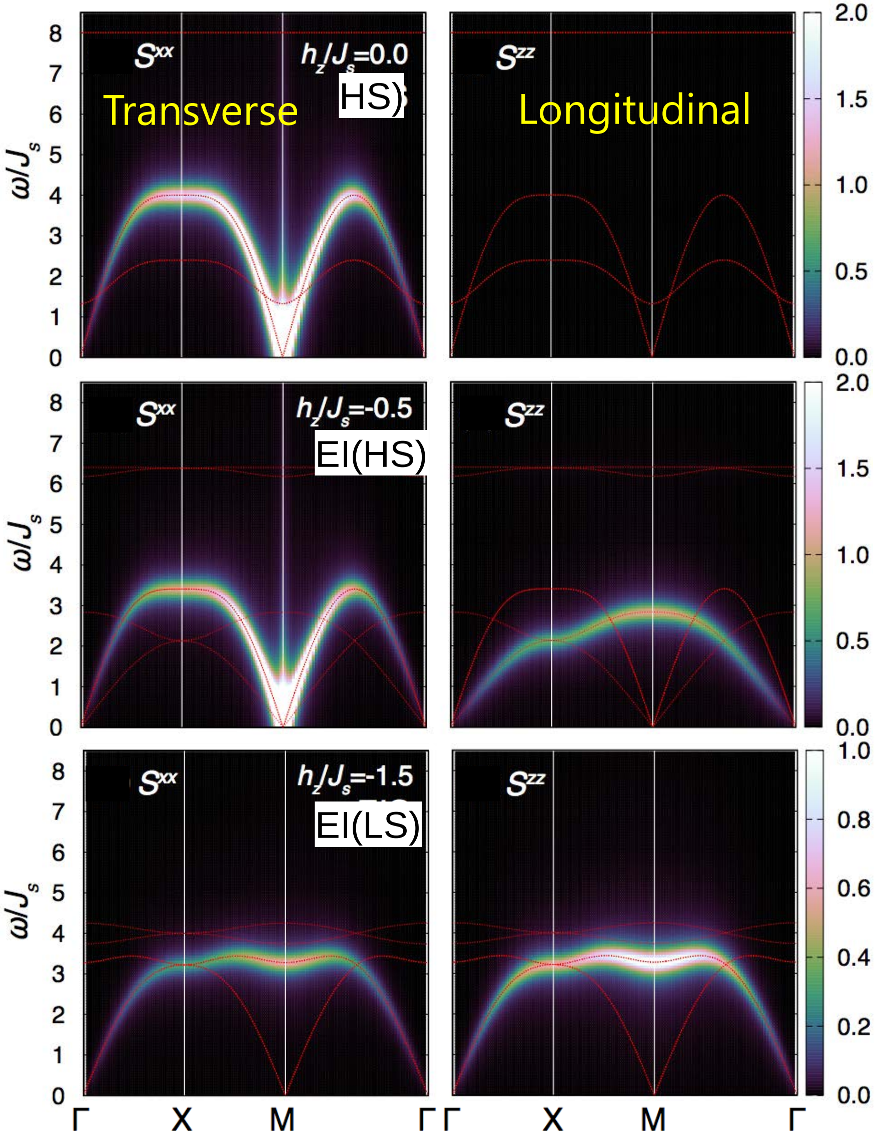
$z^2-x^2 \otimes zx$

Magnetic Excitation



2 orbital model

Phys. Rev. B **93**, 205136 (2016)



Dynamical spin correlation function

$$S^{ll}(\boldsymbol{q},\omega) = \frac{1}{N}\sum_{ij}\int_{-\infty}^{\infty} dt\langle S_i^l(t)S_j^l\rangle e^{i\omega t - i\boldsymbol{q}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)}$$

$$S^{xx}(\boldsymbol{q},\omega) = S^{yy}(\boldsymbol{q},\omega)$$

AFM Spin wave in Sxx (Transverse)

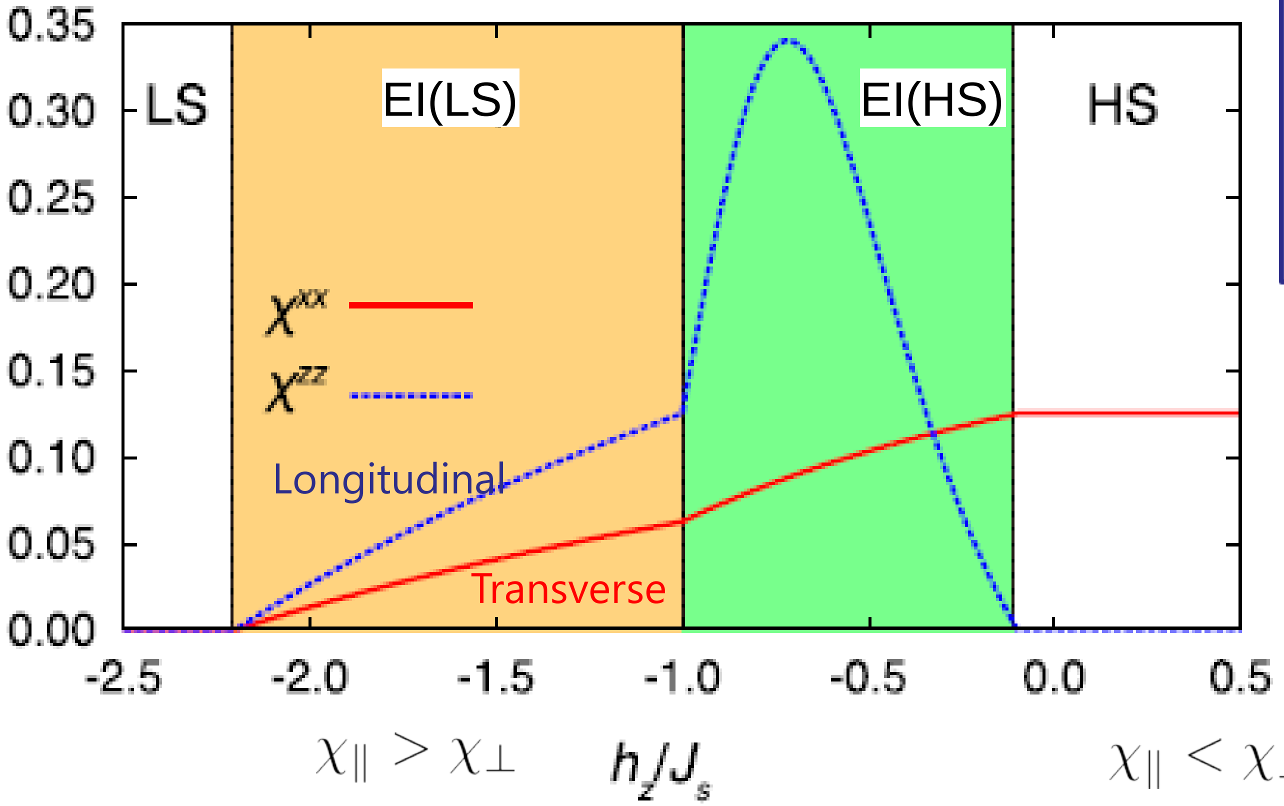
AFM Spin wave
S^{xx}(Transverse) and
S^{zz}(Longitudinal) (due to LS-HS mixing))

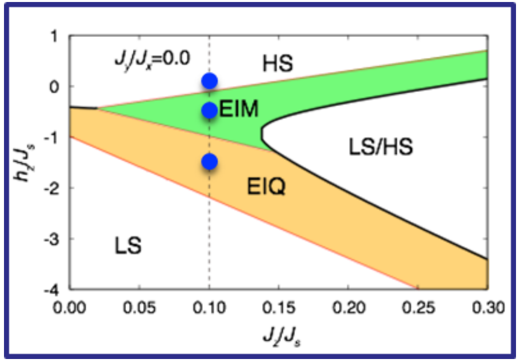
$$a|HS\rangle + b|LS\rangle$$

Spin wave in spin nematic order

c.f G. Khalliuline, PRL 111 197201(2013)

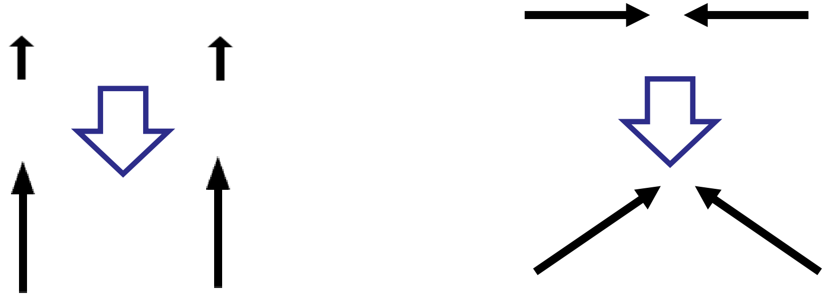
Magnetic susceptibility (T=0)





$\chi_\| > \chi_\perp$

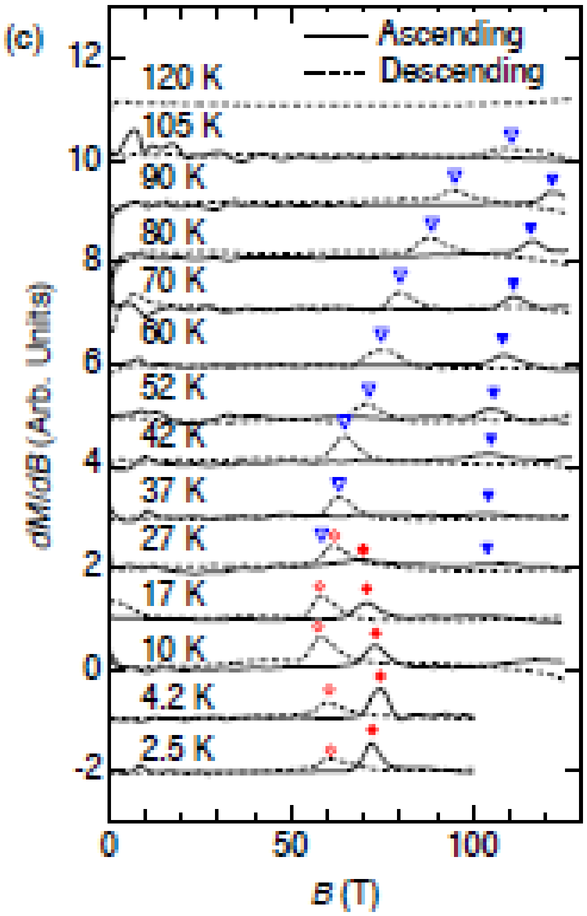
$\chi_\| < \chi_\perp$

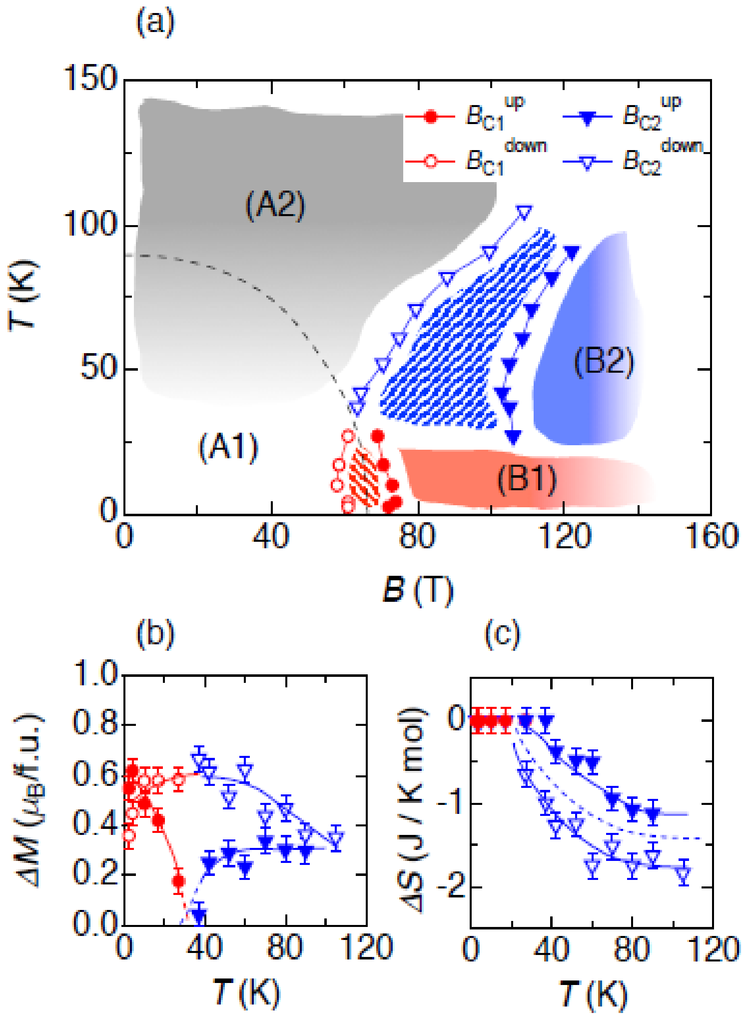


$a|HS\rangle + b|LS\rangle$

Magnetic field effect

$LaCoO_3$





See also

J. Kuneš et al. (Sci. Rep. 2016)

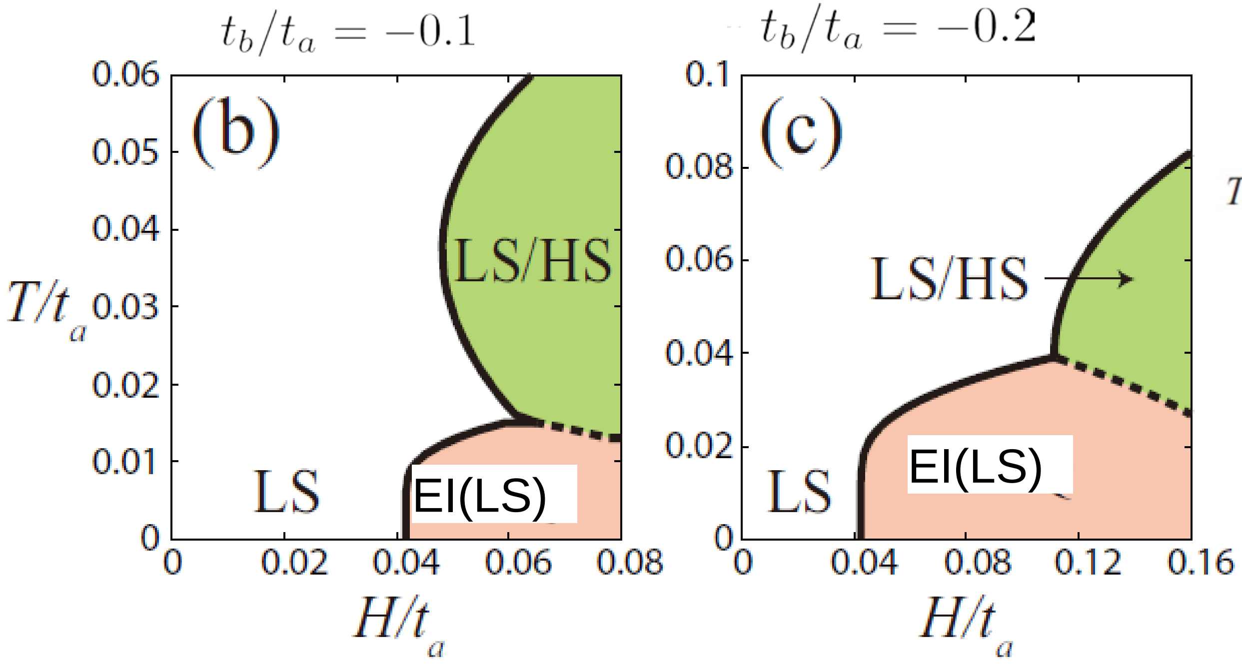
Phys. Rev. B 93, 220401 (2016)

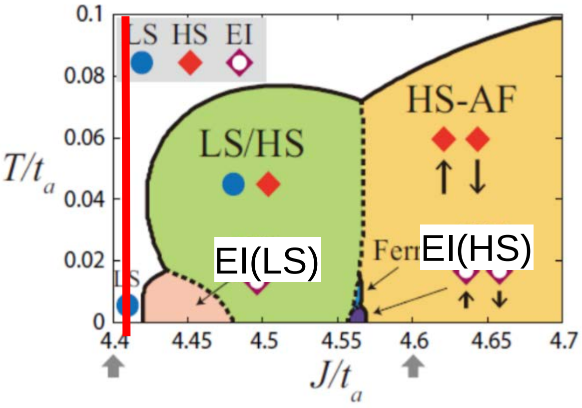
A Ikeda, T Nomura, Y. H. Matsuda, A. Matsuo, K. Kindo, and K. Sato

Magnetic field induced EI

T. Tatsuno, E. Mizoguchi, J. Nasu, M. Naka, and SI,
J. Phys. Soc. Jpn. 85, 083706 (2016)

LS GS

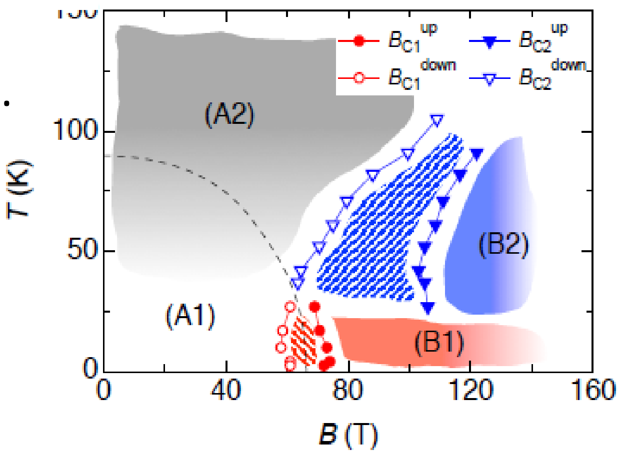




Magnetic field induced
EI & LS/HS

Exp. Ikeda et al.

(a)



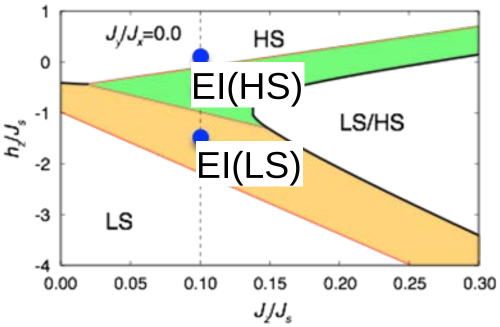
See also J. Kuneš et al. (Sci. Rep. 2016)

Summary

Mott Insulator vs. Band Insulator: EI is a possible candidate

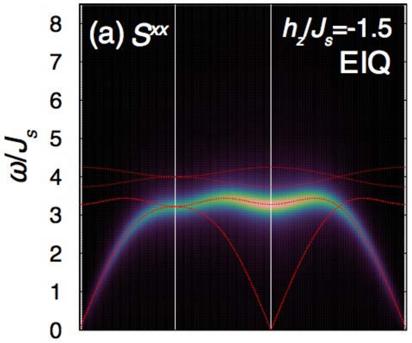
Ground state

- Two EI phases
- Breaking Z2 symmetry in EI phase
  (In no-pair hopping, U(1))
- Nematic spin order in EI(LS)



Collective excitations

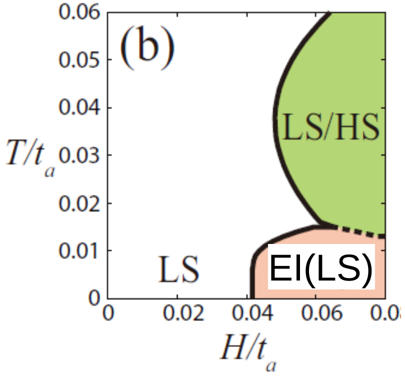
- Magnons : Longitudinal excitation
- Excitonic mode (Higgs mode)



Good targets for X-ray / Neutron spectroscopies

Magnetic field effect

- Transverse v.s longitudinal susceptibilities
- H induced EI



Phys. Rev. B **93**, 205136 (2016)
J. Phys. Soc. Jpn. 85, 083706 (2016)

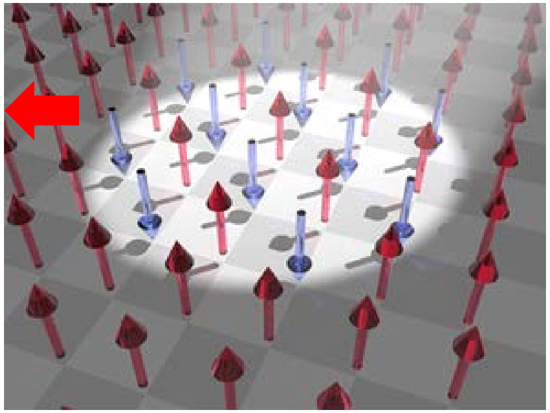
Outline

[1] Excitonic insulating state in a correlated material

J. Nasu (Tokyo Tech.), M. Naka (Waseda Univ.)
T. Tatsuno (Tohoku Univ.), T. Watanabe (Chiba Tech.)

J. Nasu, T.Watanabe, M.Naka, and SI, Phys. Rev. B **93**, 205136 (2016)
T. Tatsuno, E. Mizoguchi, J. Nasu, M. Naka, and SI,
J. Phys. Soc. Jpn. 85, 083706 (2016)

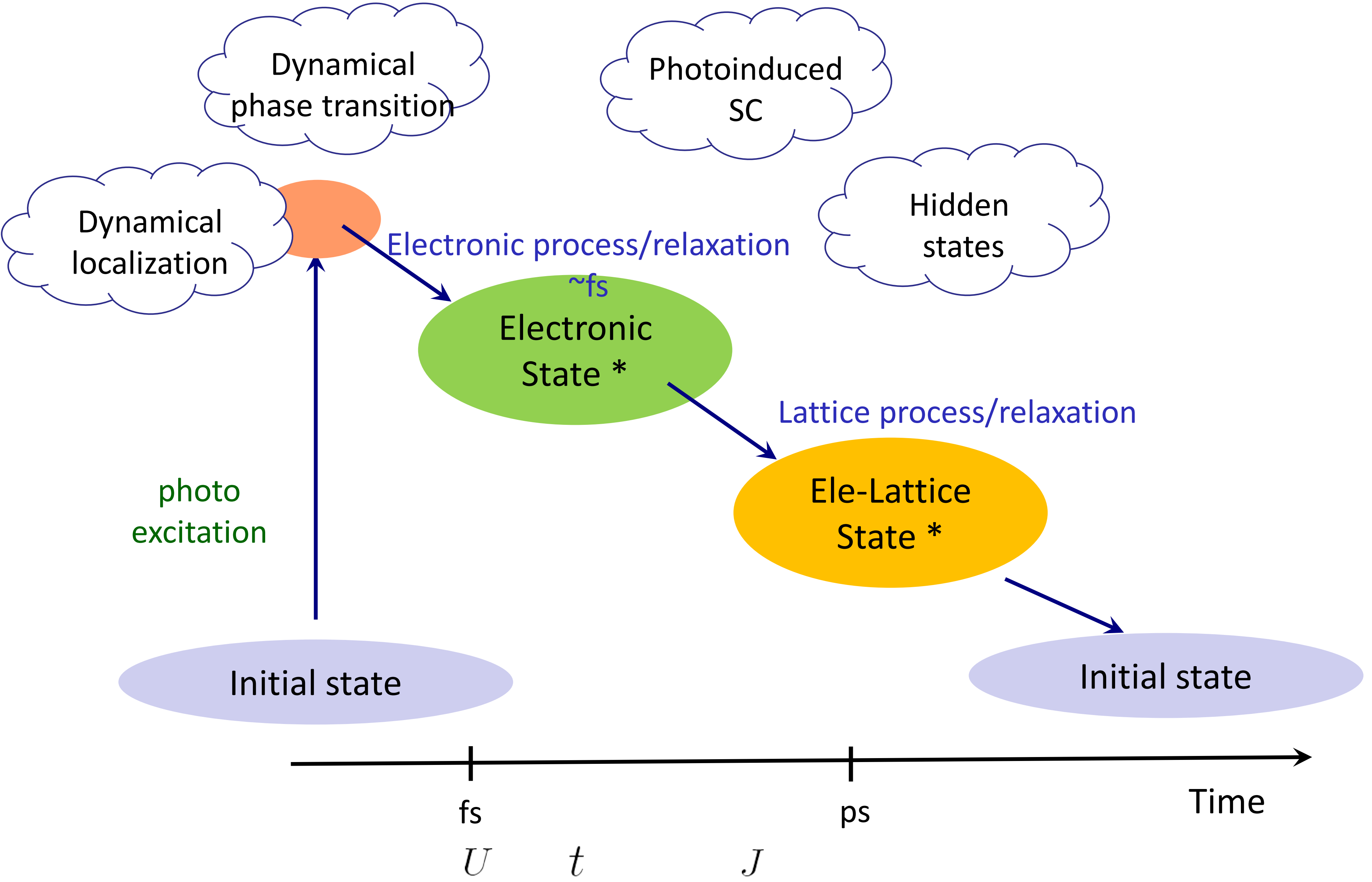
[2] Double exchange interaction in non-equilibrium state



A. Ono (Tohoku Univ.) J. Ohara (Hokkaido Univ.),
Y Kanamori (Tohoku Univ.)

A. Ono and SI, Phys. Rev. Lett. 119, 207202 (2017)
(Editors’ suggestion)
J. Ohara, Y. Kanamori and SI, Phys. Rev. B 88, 085107 (2013)

Non-eq. dynamics in correlated materials



Dynamical phase transition

Photoinduced SC

Dynamical localization

Hidden states

Electronic process/relaxation ~fs

Electronic State *

Lattice process/relaxation

Ele-Lattice State *

photo excitation

Initial state

Initial state

fs

ps

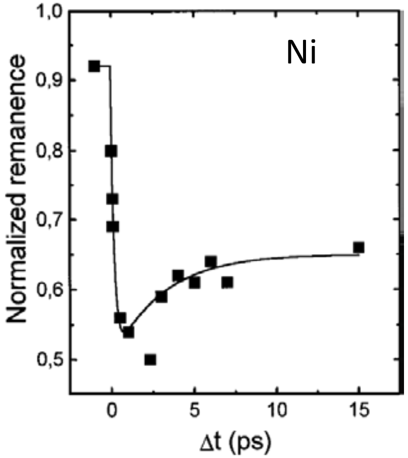
Time

U t J

Optical manipulation of magnetism

Ultrafast demagnetization

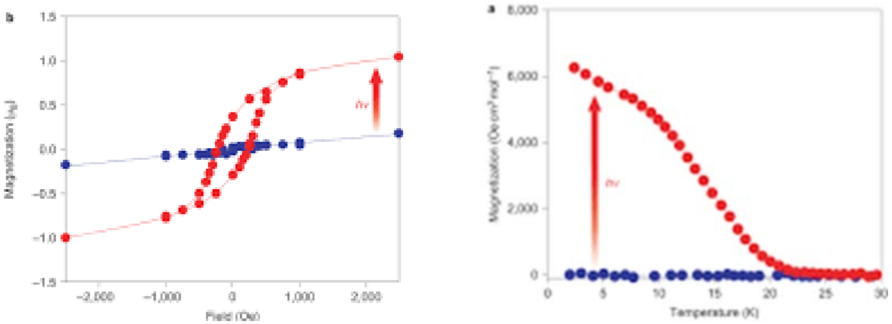
E. Beaurepaire, J. Merle, et al. PRL (1996)



Light induced spin crossover

S. Ohkoshi, et al. Nat. Chem. (2010)

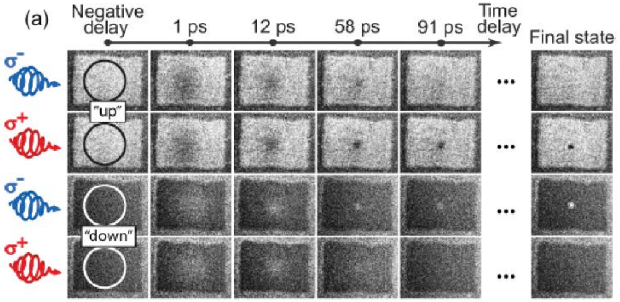
$Fe_2[Nb(CN)_8]\cdot(4\text{-pyridinealdoxime})_8\cdot 2H_2O$



Ultrafast magnetization reverse

K. Vahaplar, et al. PRL (2009)

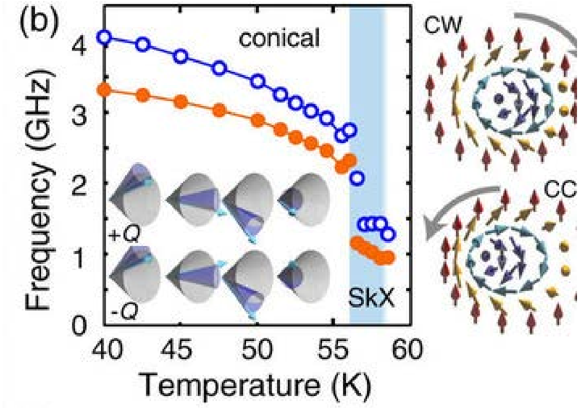
$Gd_{22}Fe_{68.3}Co_{9.8}$



Optical excitation of skyrmion

N. Ogawa, et al. Sci. Rep. (2015)

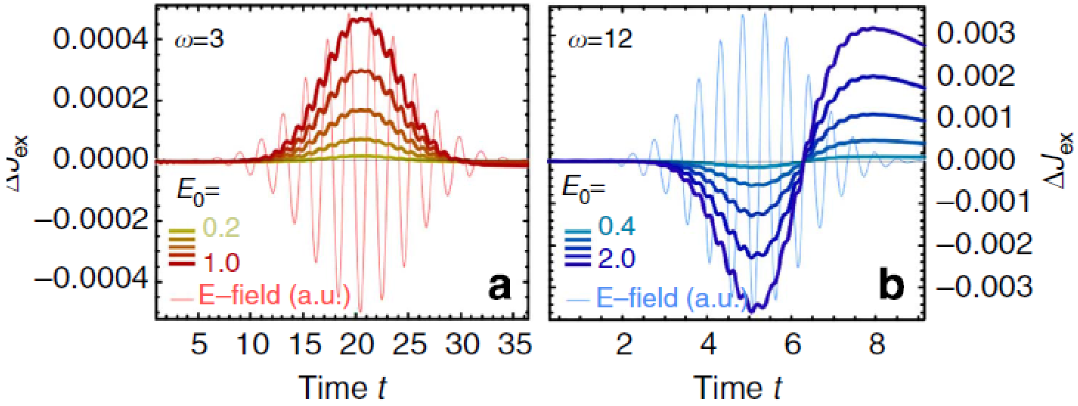
Cu_2OSeO_3



Manipulation of exchange interaction

Superexchange interaction in Mott insulator

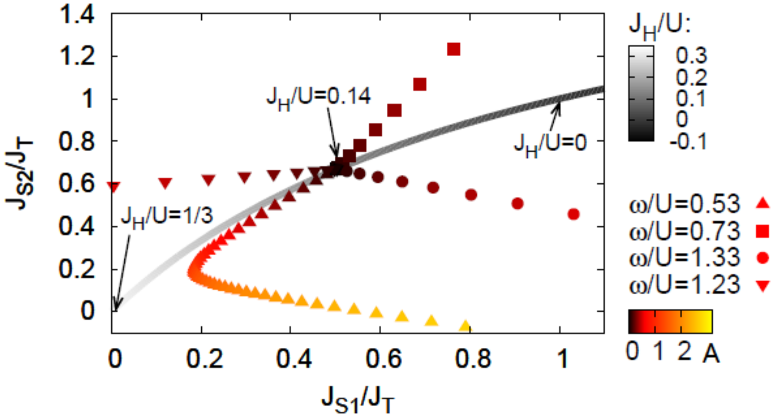
J. H. Mentink, K. Balzer, and M. Eckstein, Nat. Commun. (2015).



JS_iS_j

Spin-orbital exchange interaction in orbital degenerate Mott insulator

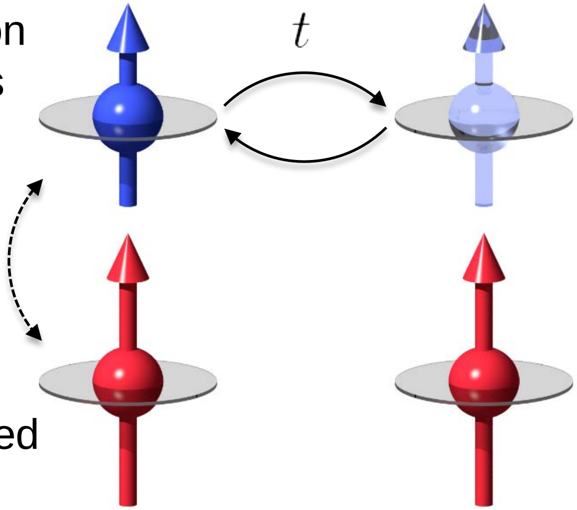
M. Eckstein, J. H. Mentink, and P. Werner, arXiv:1703.03269v1



$J(T_iT_j)(S_iS_j)$

Double exchange interaction

Zener ('51), Anderson-Hasegawa ('55), de Gennes ('59)
Metallic magnet



Conduction electrons

Transfer t

Hund coupling J_H

Localized spins

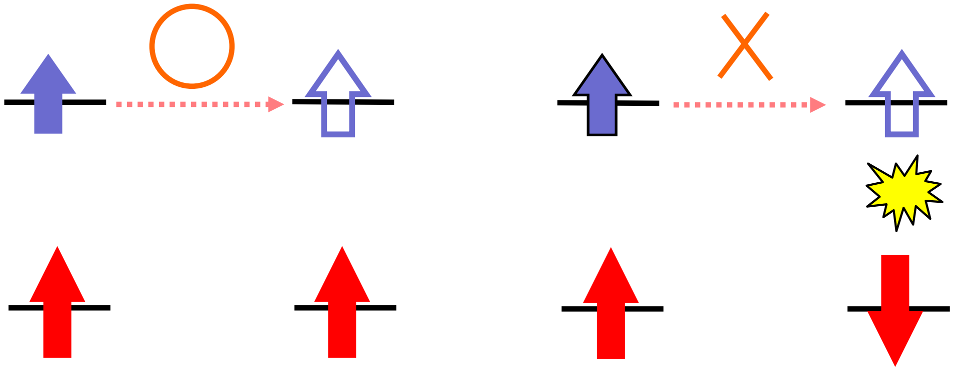
$$J_H >> t$$

$$t_{eff} = t \cos \frac{\theta}{2}$$

⇕

$$J S_i S_j \propto \cos \theta$$

Go & No-go rule



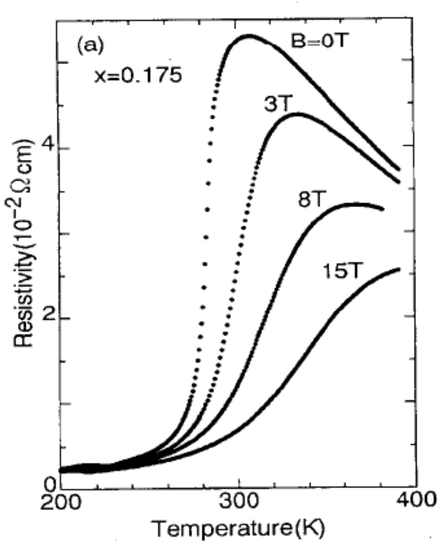
Magnetism (Spin)

⇕

Conduction (Electron)

DEx interaction in solids

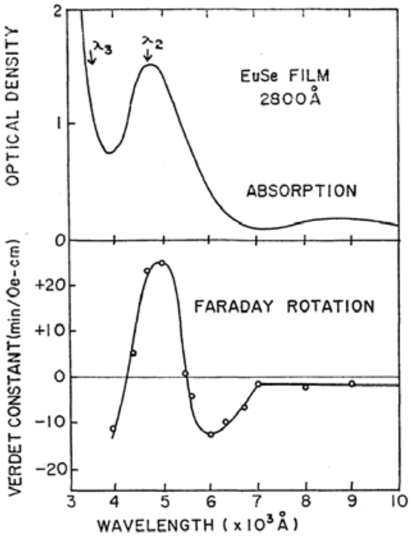
Colossal Magneto Resistance



$La_{1-x}Sr_xMnO_3$

Urushibara et al. JPSJ

Magnetic semiconductor



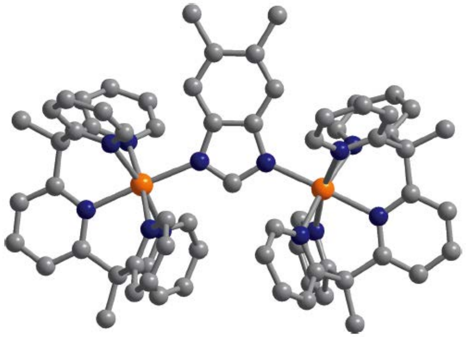
EuSe

From
A. Yanase, and T. Kasuya,
J. Phys. Soc. Jpn. 25,(1968).

And more

Molecular magnet

[(PY5Me2)2V2(m-5,6-dmbzim)]31 in 14.3.5MeCN.Et2O



B. Bechlars, et al. Nat. Chem. 2, 362 (2010).

Anomalous Hall effect

Y. Taguchi, et al. 2001 Science 291

$Nd_2Mo_2O_7$

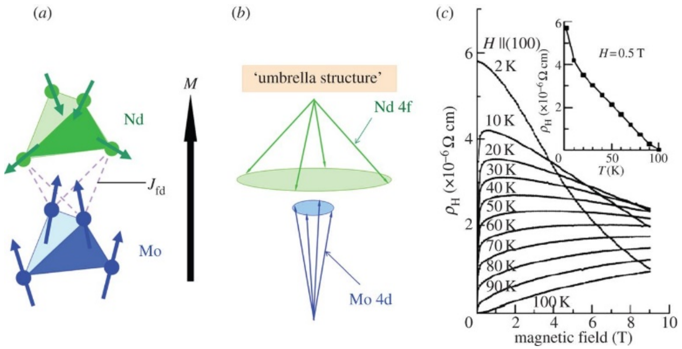
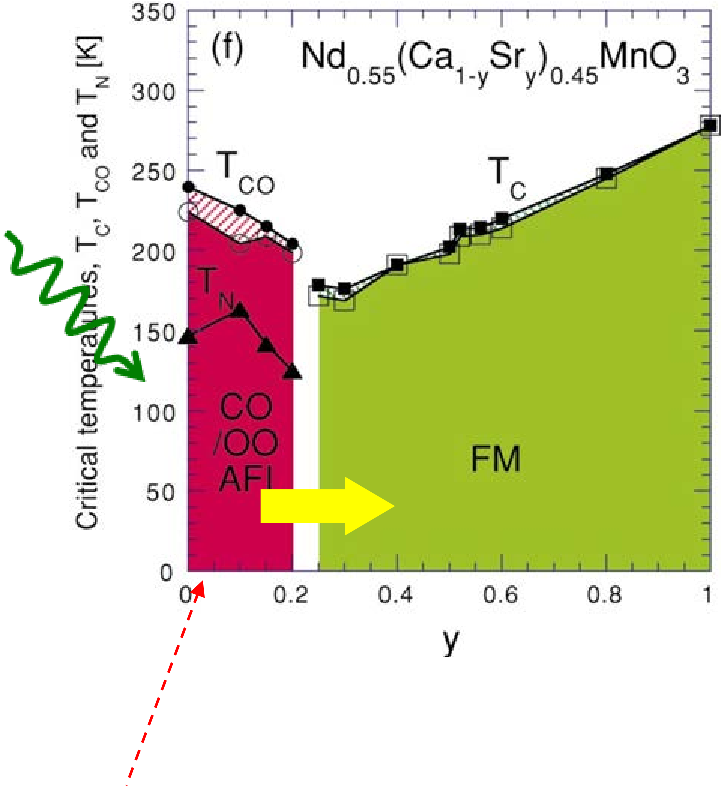


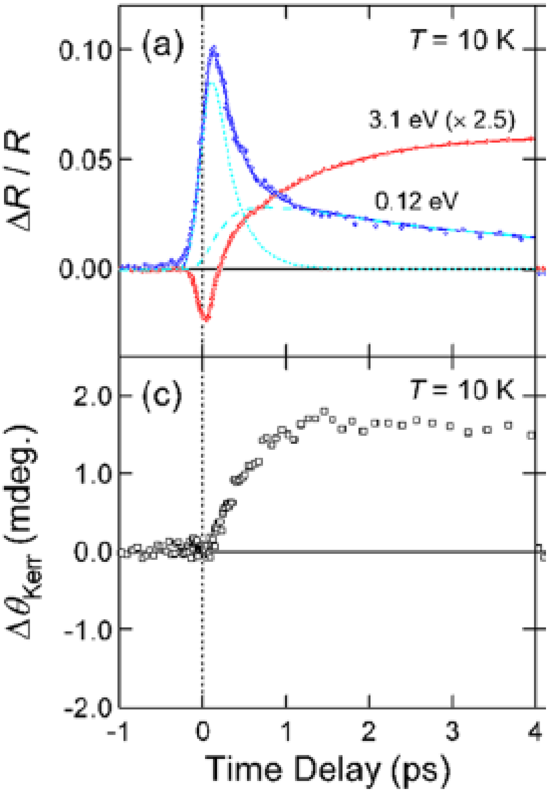
Photo irradiation in DEx system

Tomioka-Tokura et al. PRB (‘04)



AFM exchange interaction
Coulomb interaction
in addition to original DEx interaction

Optical pump-probe



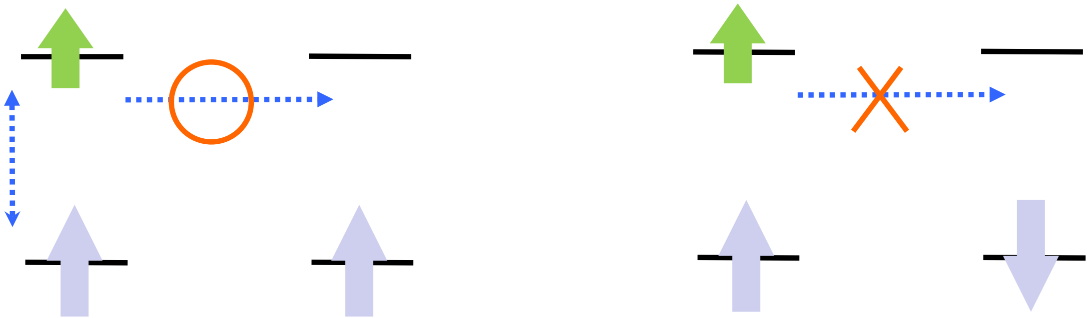
Fiebig, Miyano,
Tokura, Okamoto,
Koshihara
and many

$Gd_{0.55}Sr_{0.45}MnO_3$, Matsubara et al.

also $Nd_{0.5}Sr_{0.5}MnO_3$, Miyasaka et al
Ogasawara et al. (’05)

Photo-induced
AFM/CO to metallic FM

Photo irradiation as a carrier doping

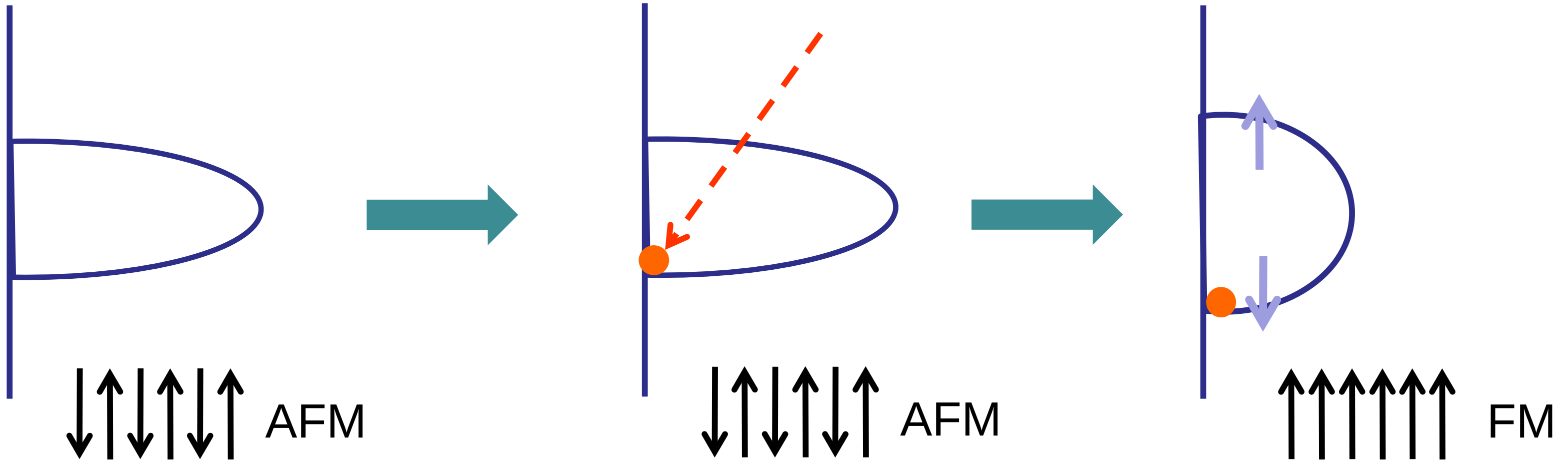


Conduction electron

Hund coupling

Local spin

Carrier doping

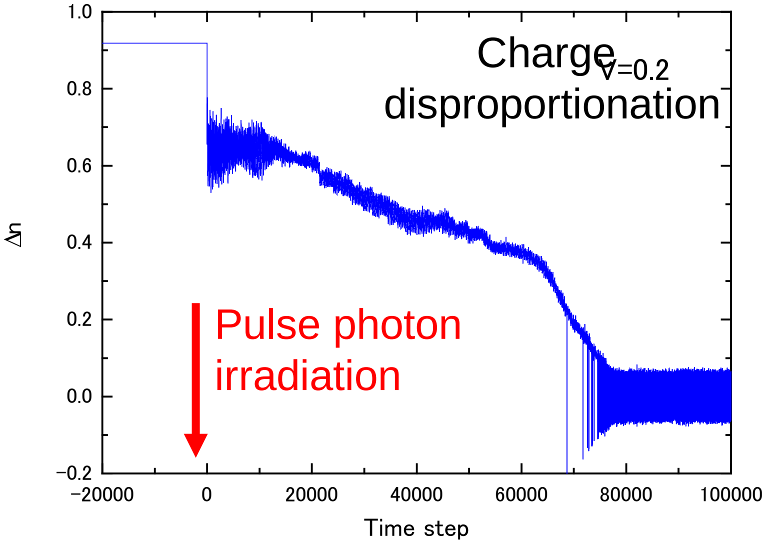


Theoretical demonstration

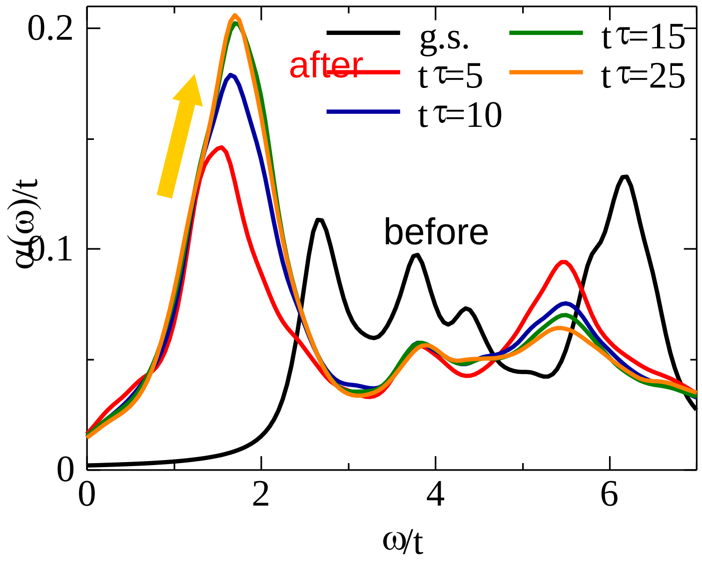
K. Satoh and SI
JMMM 130, 798-800 (2007)

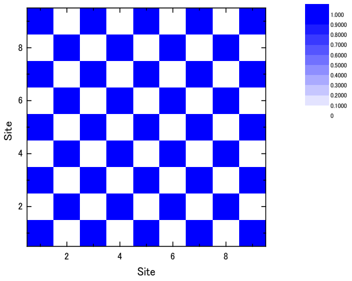
H. Matsueda & SI, JPSJ76, 083703, ('07)
Y. Kanamori, H. Matsueda and SI PRL 103, 26740 ('09)
Y. Kanamori, H. Matsueda and SI, PRB 82, 115101 ('10)

Real time simulation

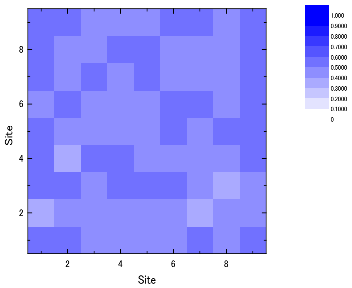


Pump-probe spectra





AFM-CO insulator

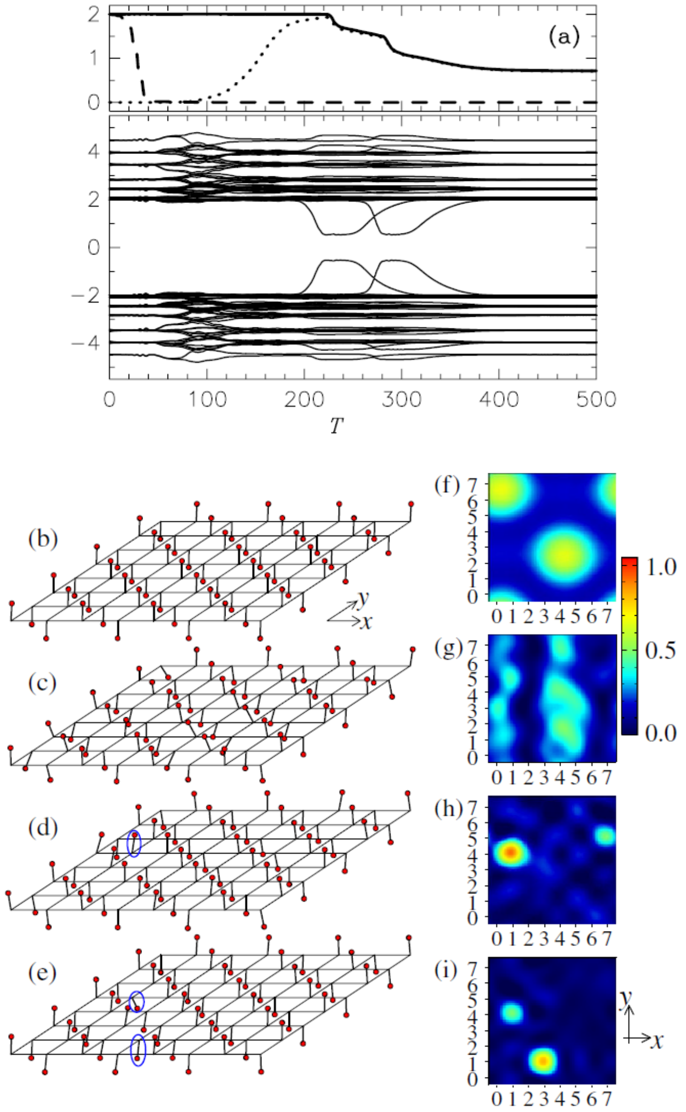


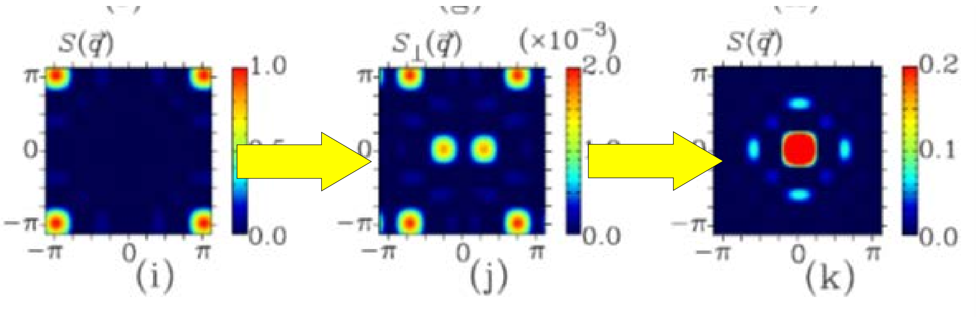
FM metal

Photo-induced
AFM/CO to metallic FM

Theoretical demonstration

Koshibae-Furukawa-Nagaosa PRL 03, 266402 (2009)
EPL 94, 27003 (2011)





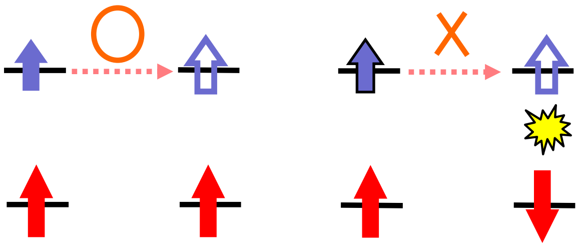
AFM

FM

Weak excitation (~1 photon/100sites)

AFM to FM

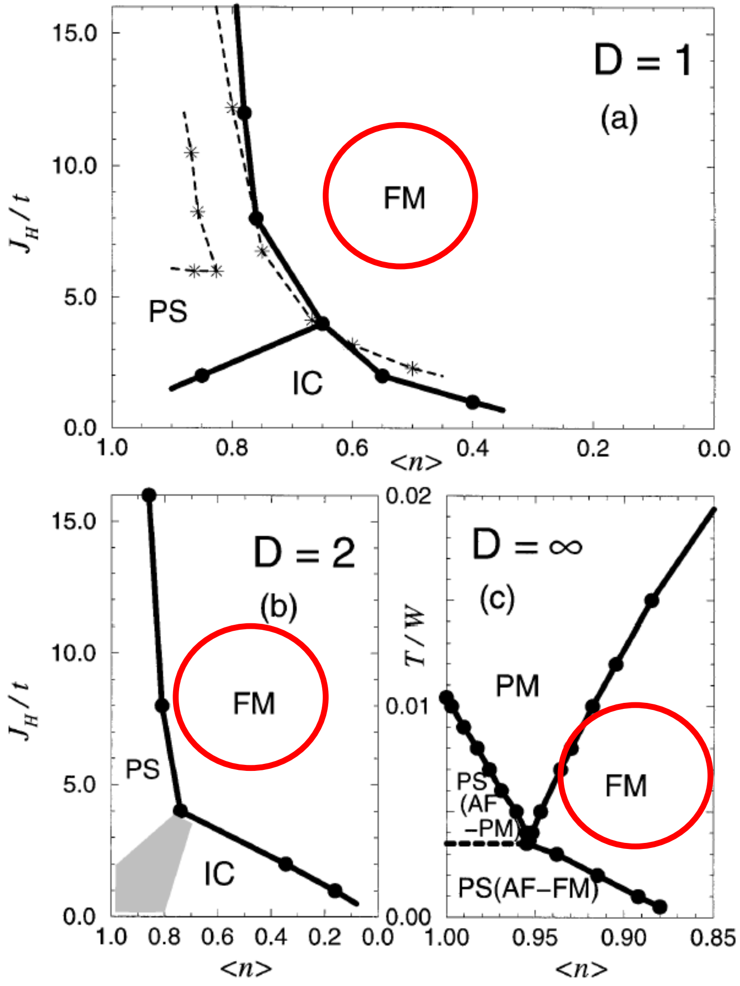
Photodoped carrier motion → FM



Ground state in DEx model

Yunoki et al. PRL (1998)

DEx model



$$H = -t \sum_{\langle i,j\rangle\sigma} c^{\dagger}_{i\sigma} c_{j\sigma} - 2J_{\mathrm{H}} \sum_i \boldsymbol{S}_i \cdot \boldsymbol{s}_i$$

What is happen by
strong excitation in FM phase
?

$$n = 1 - x$$

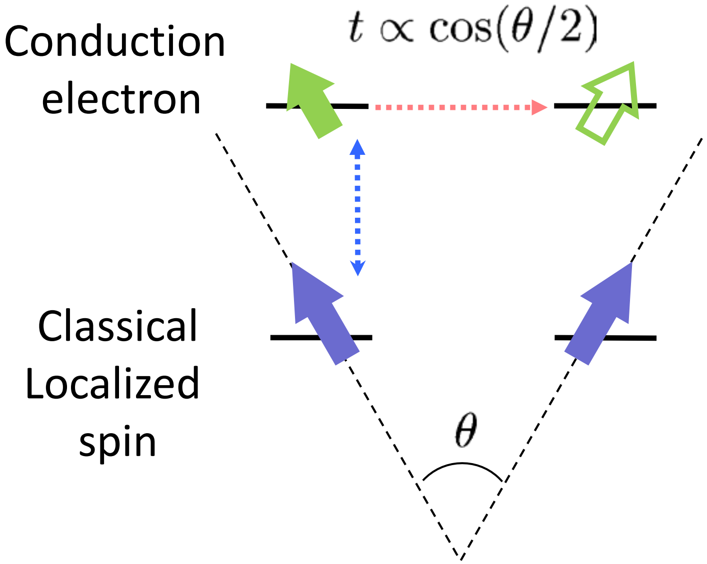
Electron # / site

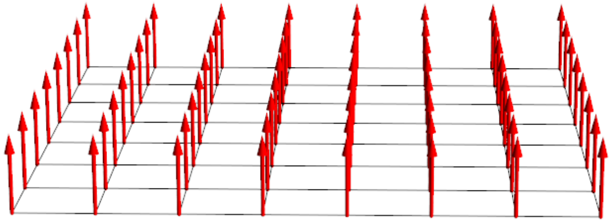
DEx interaction revisit

(pure) Double Exchange Model

$$H = -t \sum_{\langle i,j \rangle \sigma} c^\dagger_{i\sigma} c_{j\sigma} - 2J_{\mathrm{H}} \sum_i \boldsymbol{S}_i \cdot \boldsymbol{s}_i$$

- No AF interaction
- Classical localized spin
- FM metallic GS (mainly 1/4 filling)





Model & Method

Conduction electrons

Wave function

$$|\Psi(\tau)\rangle = \prod_{\nu=1}^{N_e} \psi^{\dagger}_{\nu}(\tau)|0\rangle \qquad H(\tau) = \sum_{\nu} \varepsilon_{\nu}(\tau)\tilde{c}^{\dagger}_{\nu}(\tau)\tilde{c}_{\nu}(\tau)$$

Time evolution

$$\psi^{\dagger}_{\nu}(\tau+\delta\tau) = e^{iH(\delta\tau)\delta\tau}\psi^{\dagger}_{\nu}(\tau)e^{-iH(\delta\tau)\delta\tau}$$

Localized classical spins

Landau–Lifshitz–Gilbert (LLG) equation

$$\frac{d\boldsymbol{S}_i}{d\tau} = \boldsymbol{h}_i^{\mathrm{eff}} \times \boldsymbol{S}_i + \alpha \boldsymbol{S}_i \times \frac{d\boldsymbol{S}_i}{d\tau}$$

α : Gilbert damping factor

Randomness in initial spins

2-dimensional square

N = 8×8-12×12 sites (PBC/APBC)

Vector potential

$$t \to t e^{iA(\tau)}$$

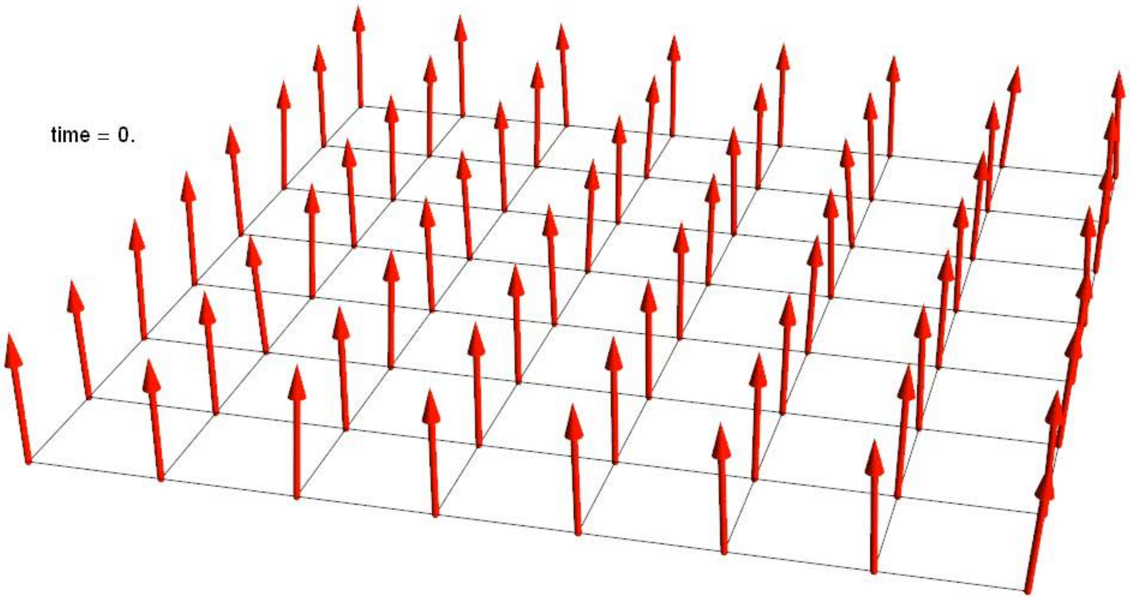
Linearly polarized CW / Pulse field

$$A \sim \mathrm{MV/cm}$$
$$\omega \sim t$$

Koshibae-Furukawa-Nagaosa PRL(09)

Animation

CW field: A_0/t = 2.0, ω/t = 1.0



Ferromagnetic metal → Antiferromagnet

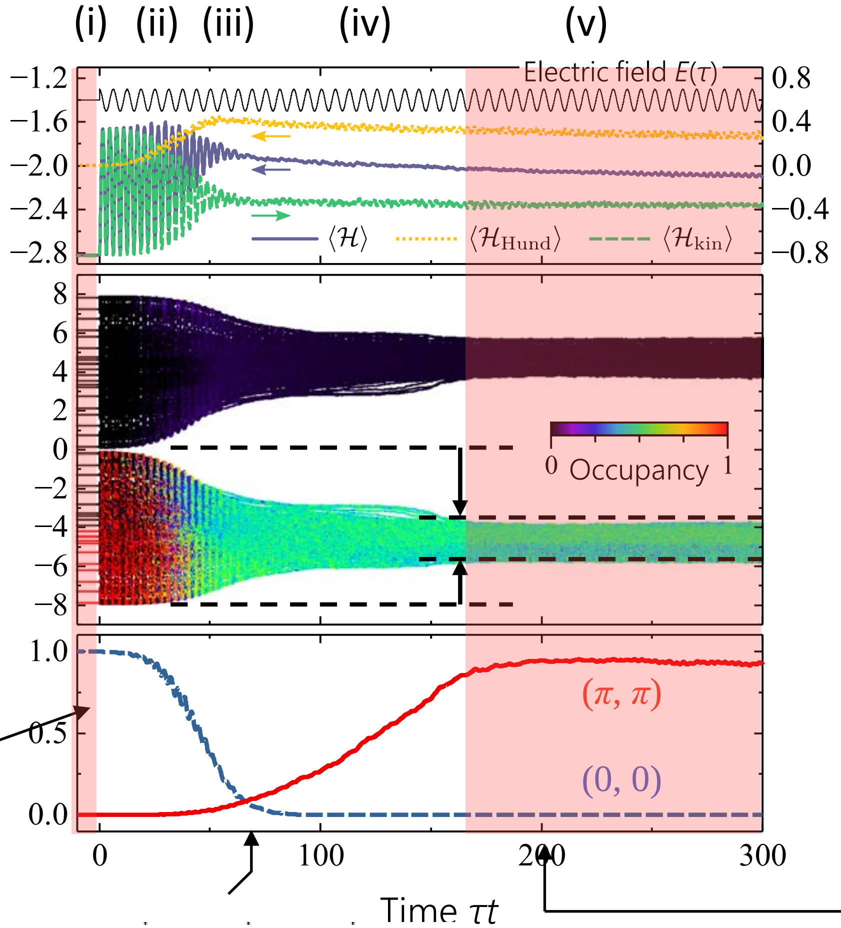
Time profiles

CW

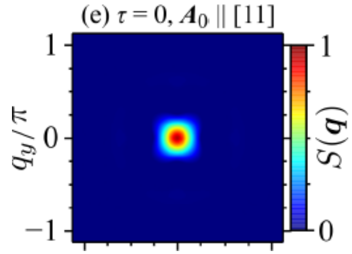
(Energy)/t

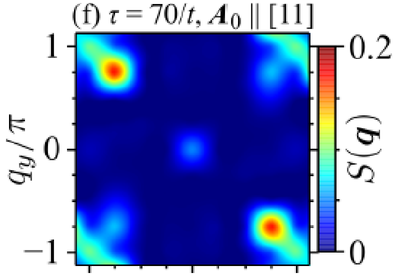
(Energy level)/t

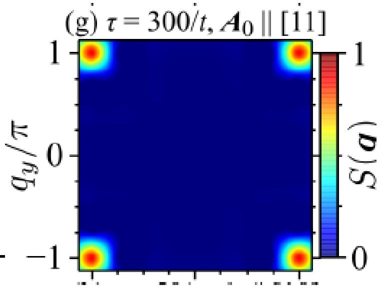
Spin structure factor



Almost perfect AFM steady state







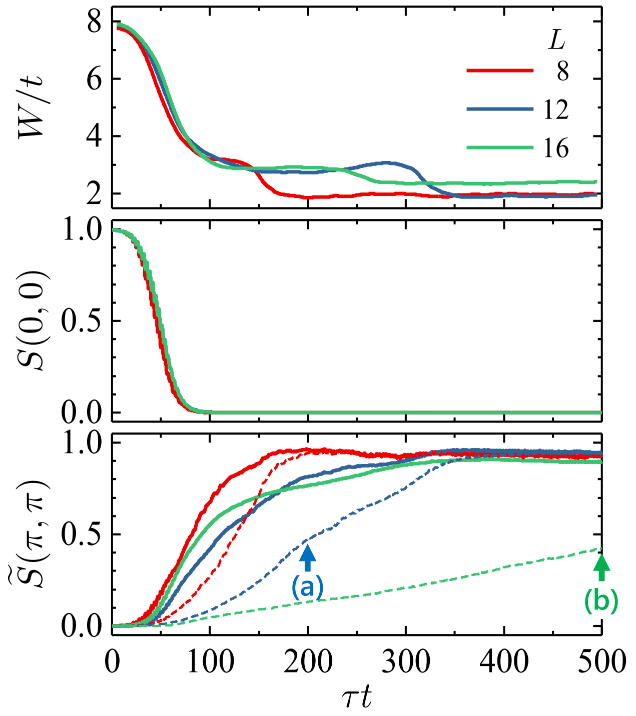
$$\tau t = 1 :\sim 8fs$$

Half metallic FM

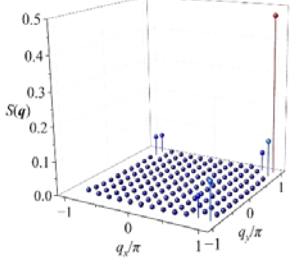
A. Ono, and SI, Phys. Rev. Lett. (2017) (Editor suggestion) arXiv:1705.00240v1

Cluster Size & Light Polarization dependences

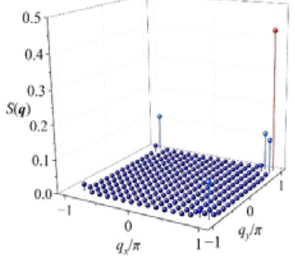
Cluster size



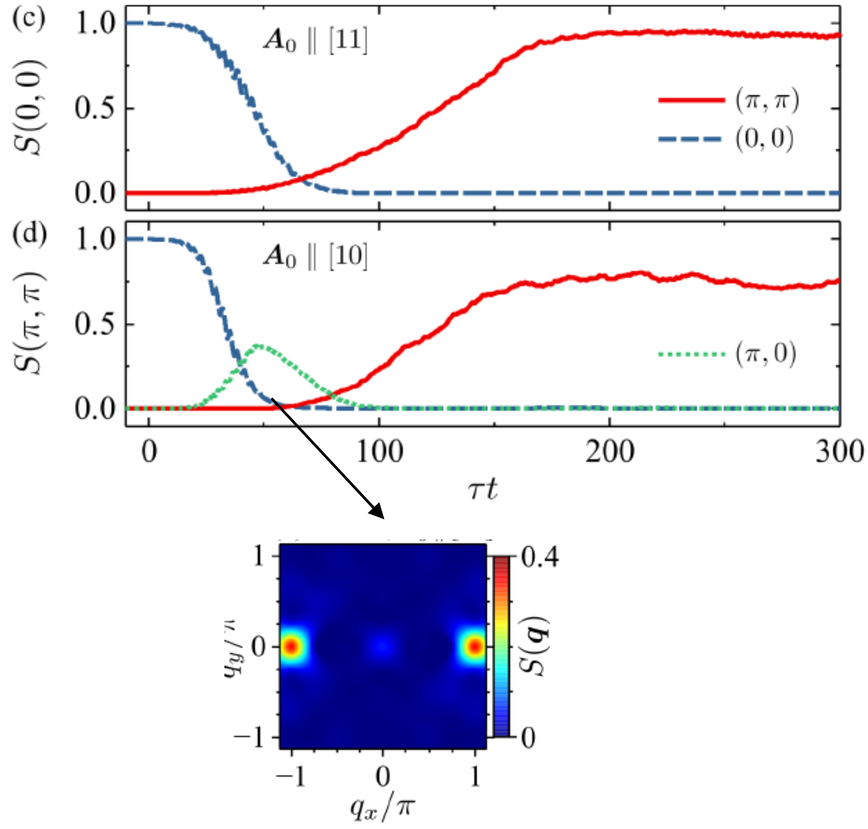
(a) *L* = 12



(b) *L* = 16



Light polarization

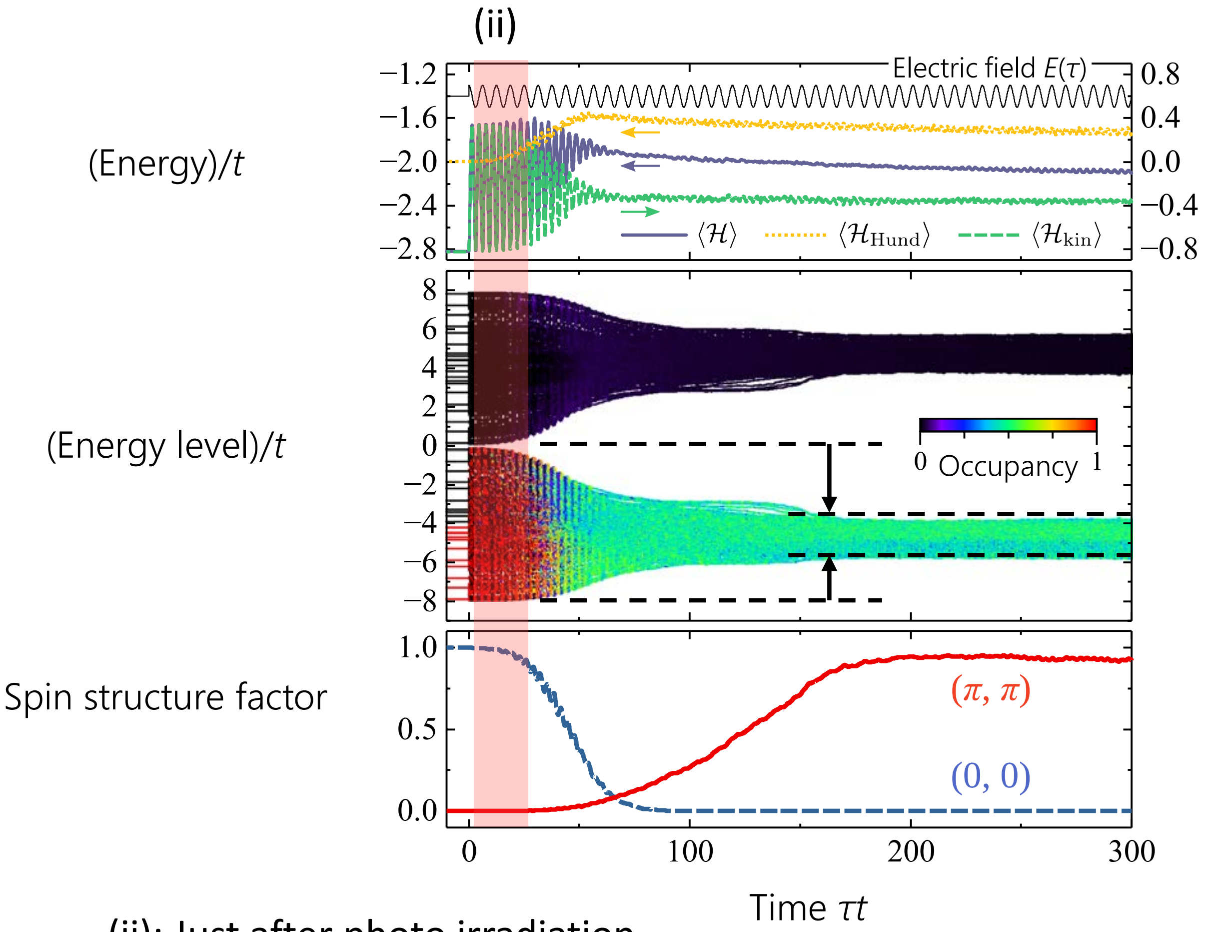


$$\begin{aligned}\widetilde{S}(\pi,\pi) &= S(\pi,\pi)\\ &\quad + 2S(\pi-\Delta q,\pi) + 2S(\pi,\pi-\Delta q)\\ &\quad + 2S(\pi-\Delta q,\pi-\Delta q),\end{aligned}$$

$(\delta q = 2\pi/L)$

At early time domain

CW



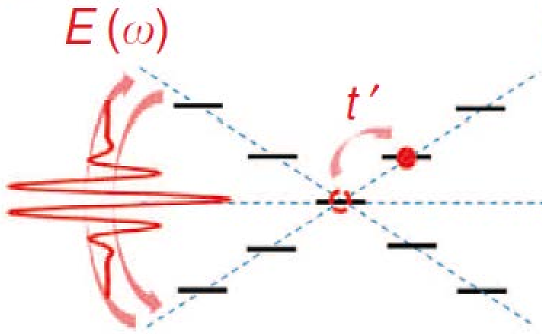
(ii): Just after photo irradiation

Excitation inside of the lower band : $\langle n \rangle = 1$ and 0 are intermingled.

Band width reduction : Dynamical localization

A. Ono, and SI, Phys. Rev. Lett. (2017) (Editor suggestion) arXiv:1705.00240v1

Dynamical localization at early time domain

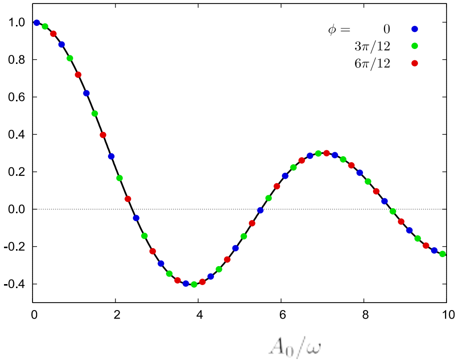


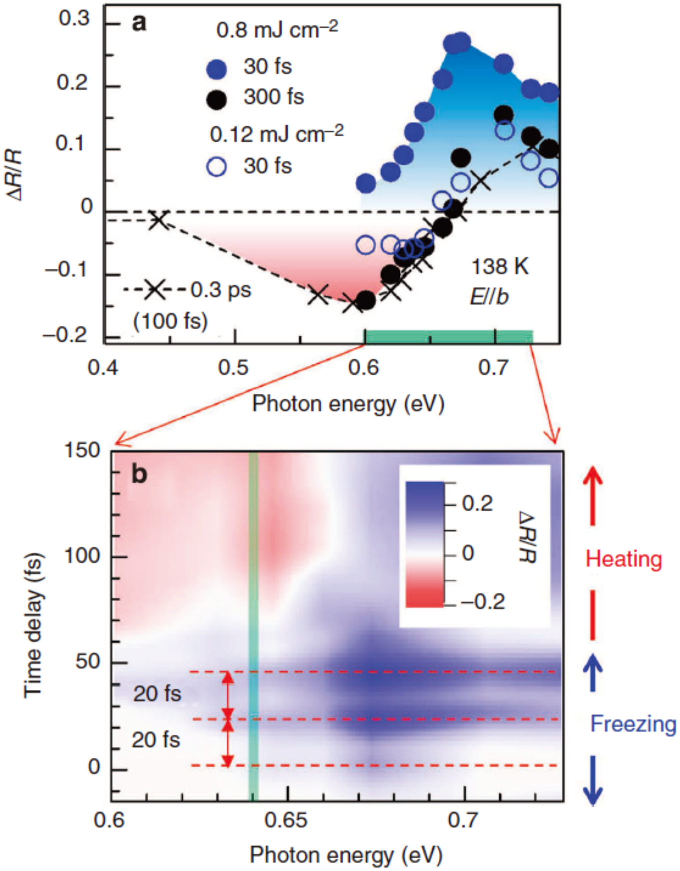
D. H. Dunlap and V. M. Kenkre, PRB **34**, 3625 (1986)
Y. Kayanuma, Phys. Rev. A 50, 843 (1994).
N. Tsuji, T. Oka, H. Aoki, and P. Werner, PRB **85**, 155124 (2012).
K. Yonemitsu and K. Nishioka, JPSJ **84**, 054702 (2015).
Ishikawa, S. Iwai et al.Nature commun. **5**, 5528(2014)
A. Ono and SI Phys. Rev. B 95, 085123 (2017)
and more

Effective electron transfer

$$t_{\mathrm{eff}} = tJ_0(A_0/\omega)$$

0-th order Bessel function





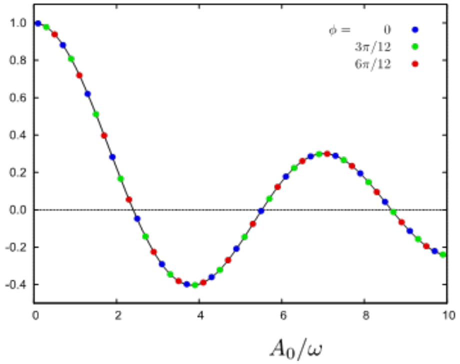
T. Ishikawa, SI, K. Yonemitsu, S. Iwai et al.
Nature commun. 5, 5528(2014)

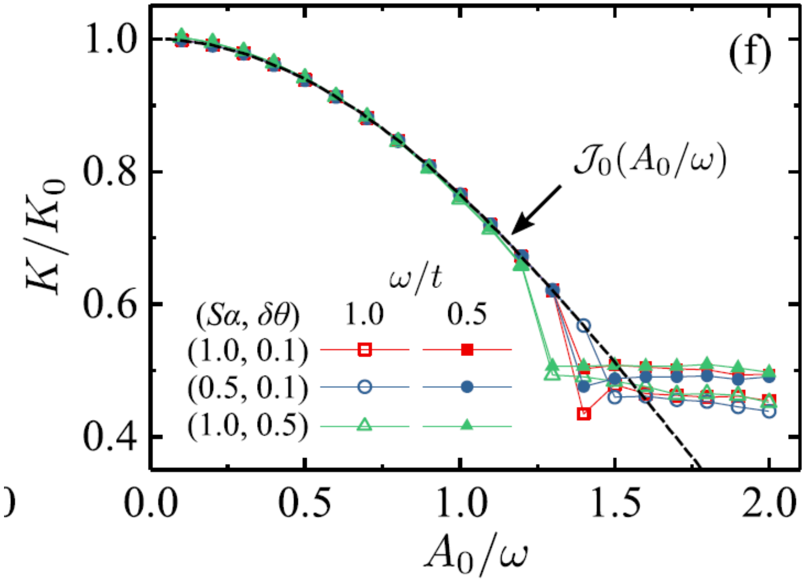
Dynamical localization at early time domain

Time average of the kinetic energy in early time domain

$$K \equiv (\Delta T)^{-1} \int_{\Delta T} d\tau \langle \mathcal{H}_t \rangle$$

$$t_{\mathrm{eff}} = t J_0(A_0/\omega)$$





Dynamical localization scenario
works well at early stage

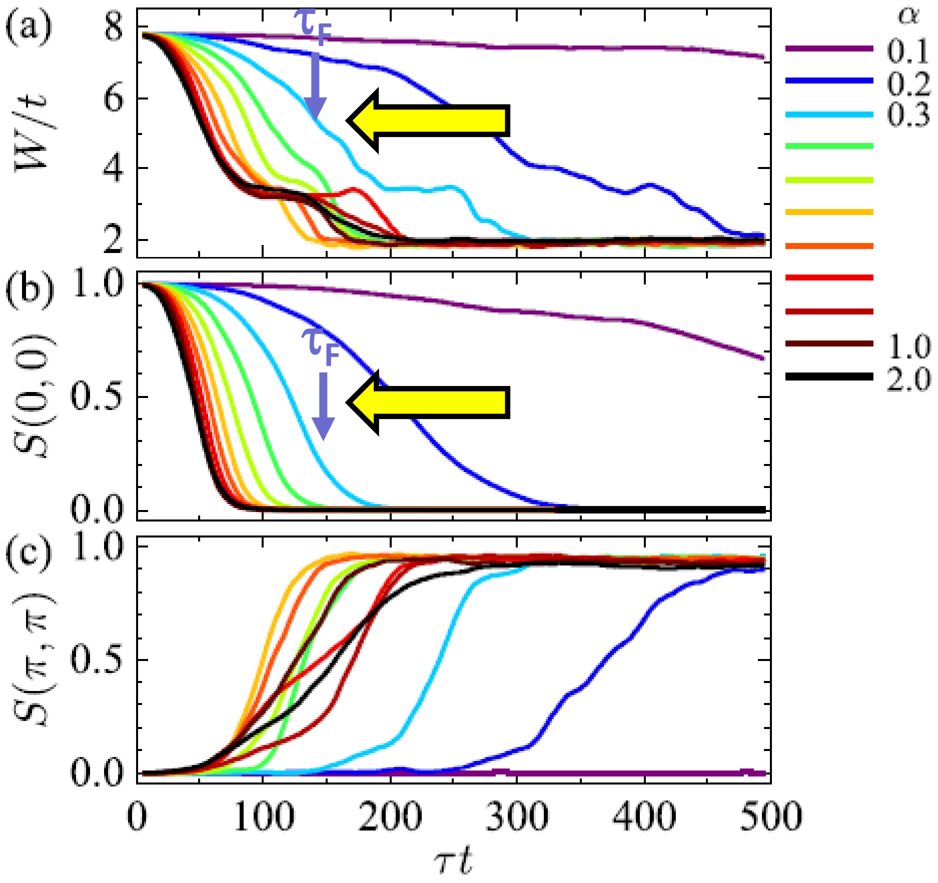
Key parameters for the FM-to-AFM conversion

Gilbert damping α dependence

Band width

FM

AFM



Key parameters for the FM-to-AFM conversion

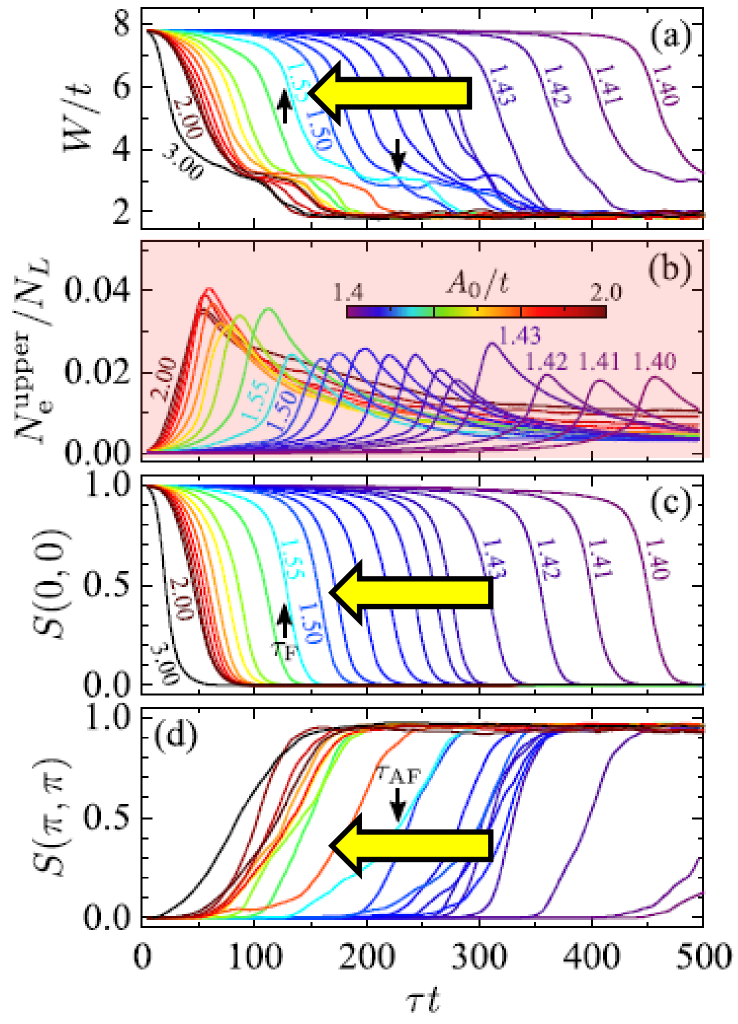
A (pump fluence) dependence

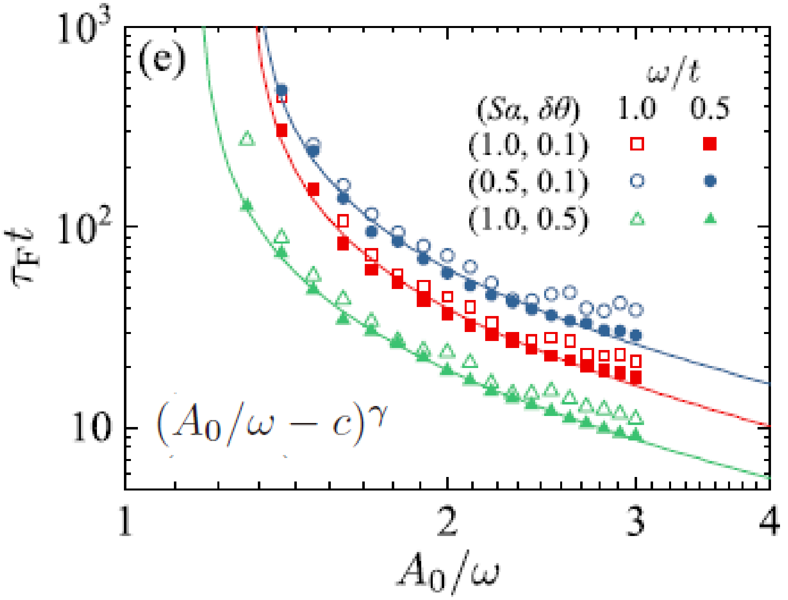
Band width

Electron # in upper band

FM

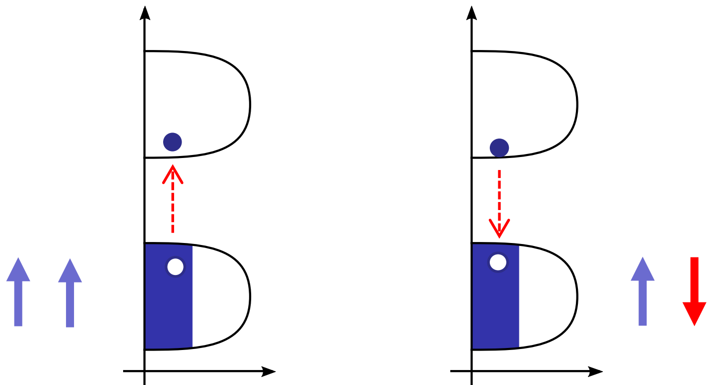
AFM





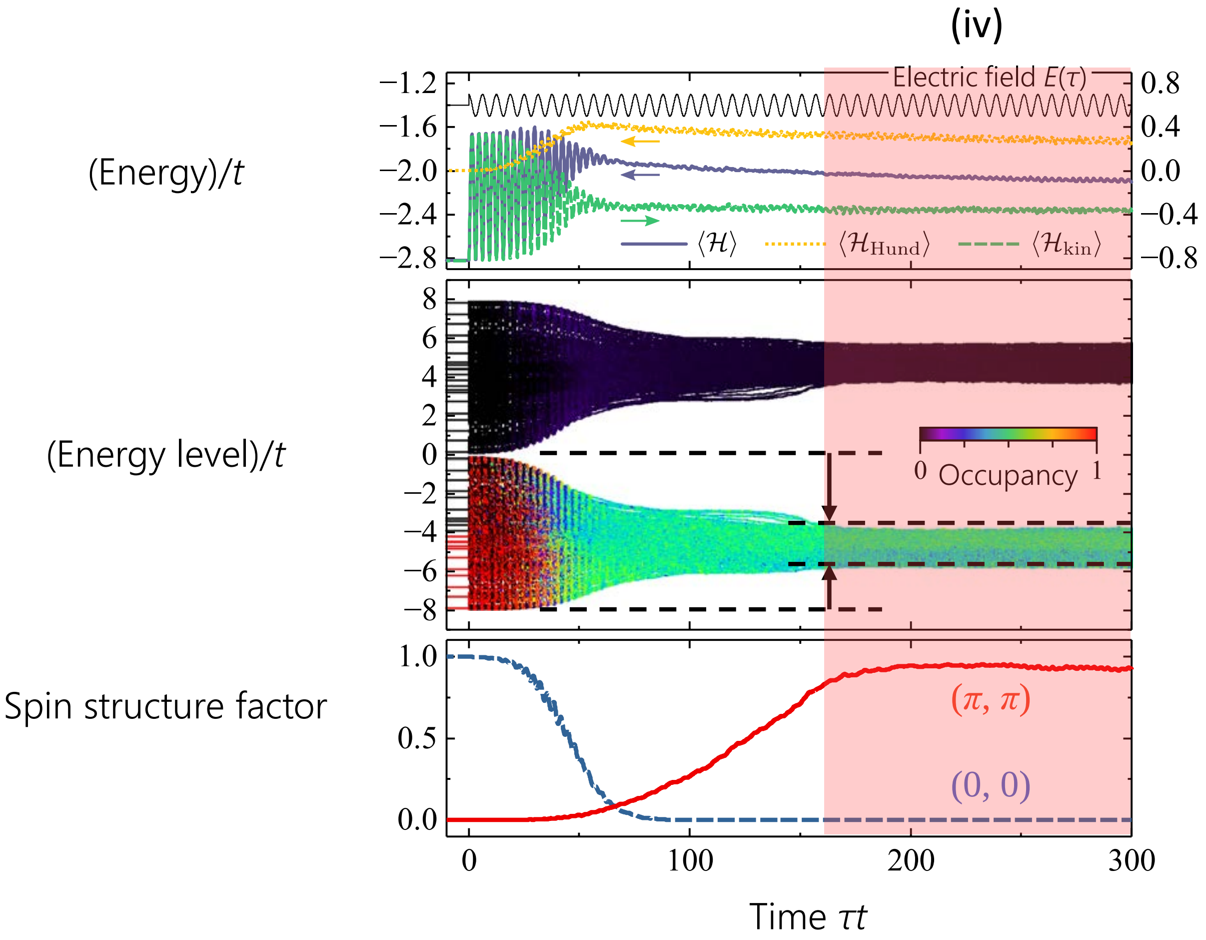
τ_F : scaled by A_0/ω

Auger-like process



Steady NEq AFM state

CW

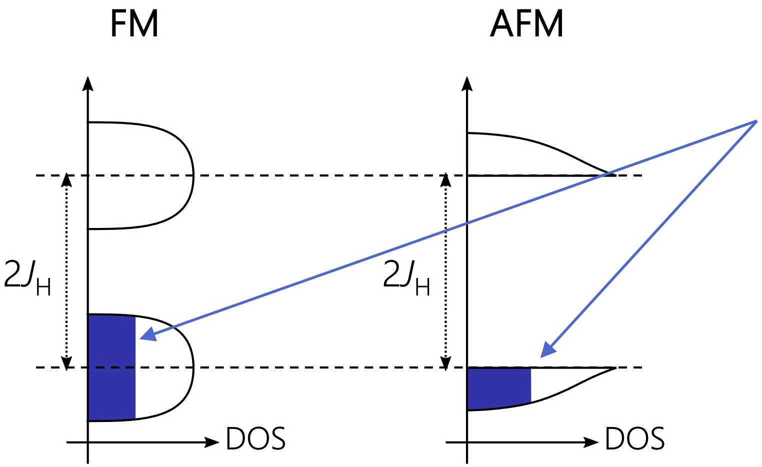


(iv): Steady AFM state

Electron distribution is almost uniform in the lower band

Steady NEq AFM state

Equilibrium cal.



Assumption:

Uniform electron distribution (≠ Fermi-Dirac)

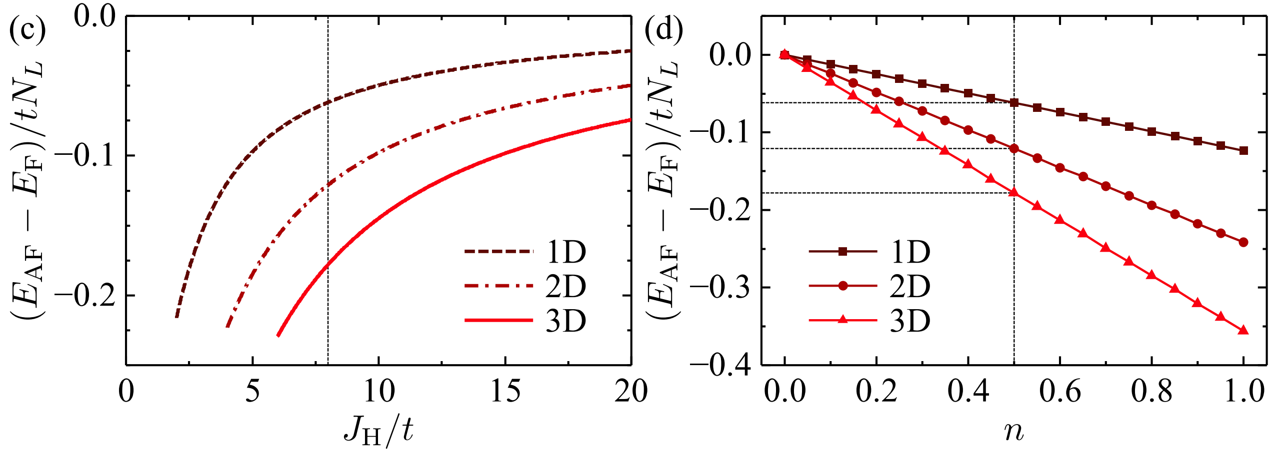
$$\langle n_\nu \rangle = \mathrm{const.}$$

⬇

Total energy $$E_\alpha = \frac{1}{N}\int_{-\infty}^{0} d\varepsilon\, \varepsilon D_\alpha(\varepsilon) f(\varepsilon)$$

$$(\alpha = \mathrm{F},\ \mathrm{AF})$$

Energy difference *E*(AF)-*E*(F) with uniform electron distribution



AFM steady state gives lower energy in wide range

Beyond the CW light

Pulse

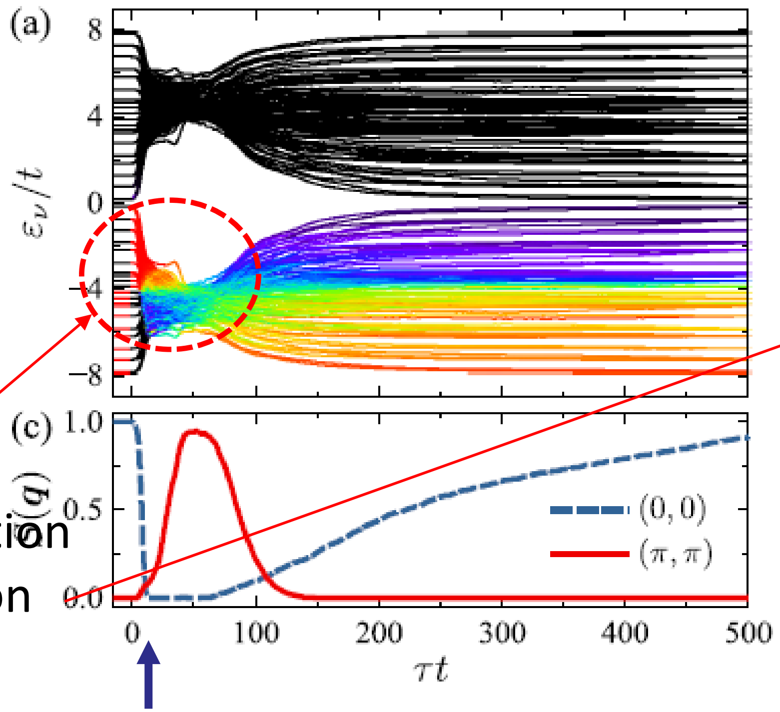
$$\boldsymbol{A}(\tau) = \boldsymbol{A}_1 \theta(\tau)$$

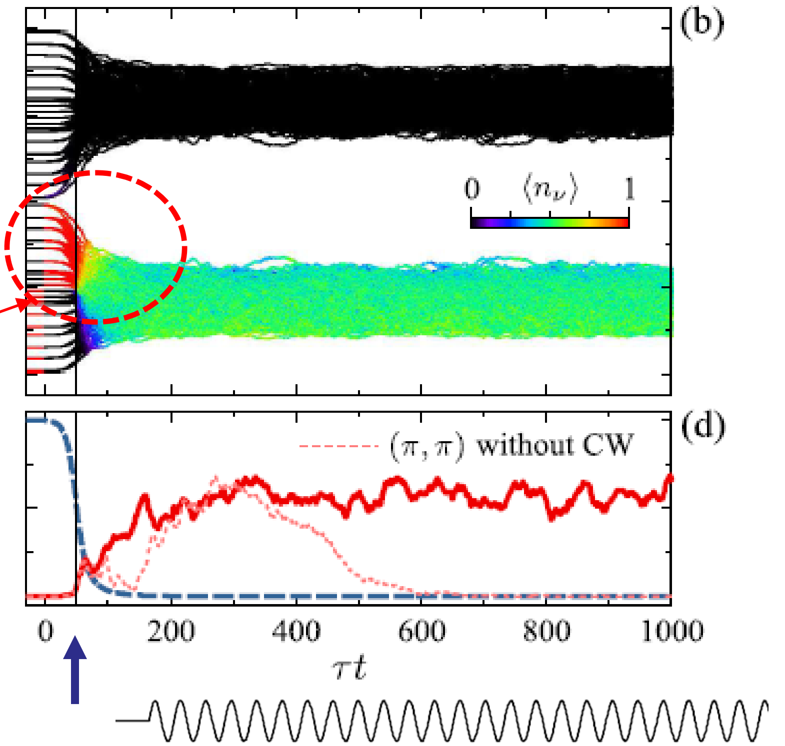
$$\boldsymbol{E}(\tau) = -\boldsymbol{A}_1 \delta(\tau)$$

Pulse + CW

$$\boldsymbol{A}(\tau) = \boldsymbol{A}_1 \theta(\tau) + (\boldsymbol{A}_0/\omega) \sin[\omega(\tau - \tau_0)]\theta(\tau - \tau_0)$$

$$\boldsymbol{E}(\tau) = -\boldsymbol{A}_1 \delta(\tau) - \boldsymbol{A}_0 \cos[\omega(\tau - \tau_0)]\theta(\tau - \tau_0)$$



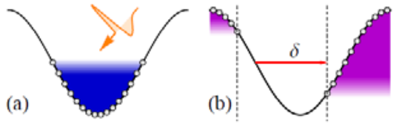


Population inversion

π-shift

$$\delta k = (\pi, \pi)$$

$$\delta \boldsymbol{k} = \int d\tau \boldsymbol{E}(\tau)$$



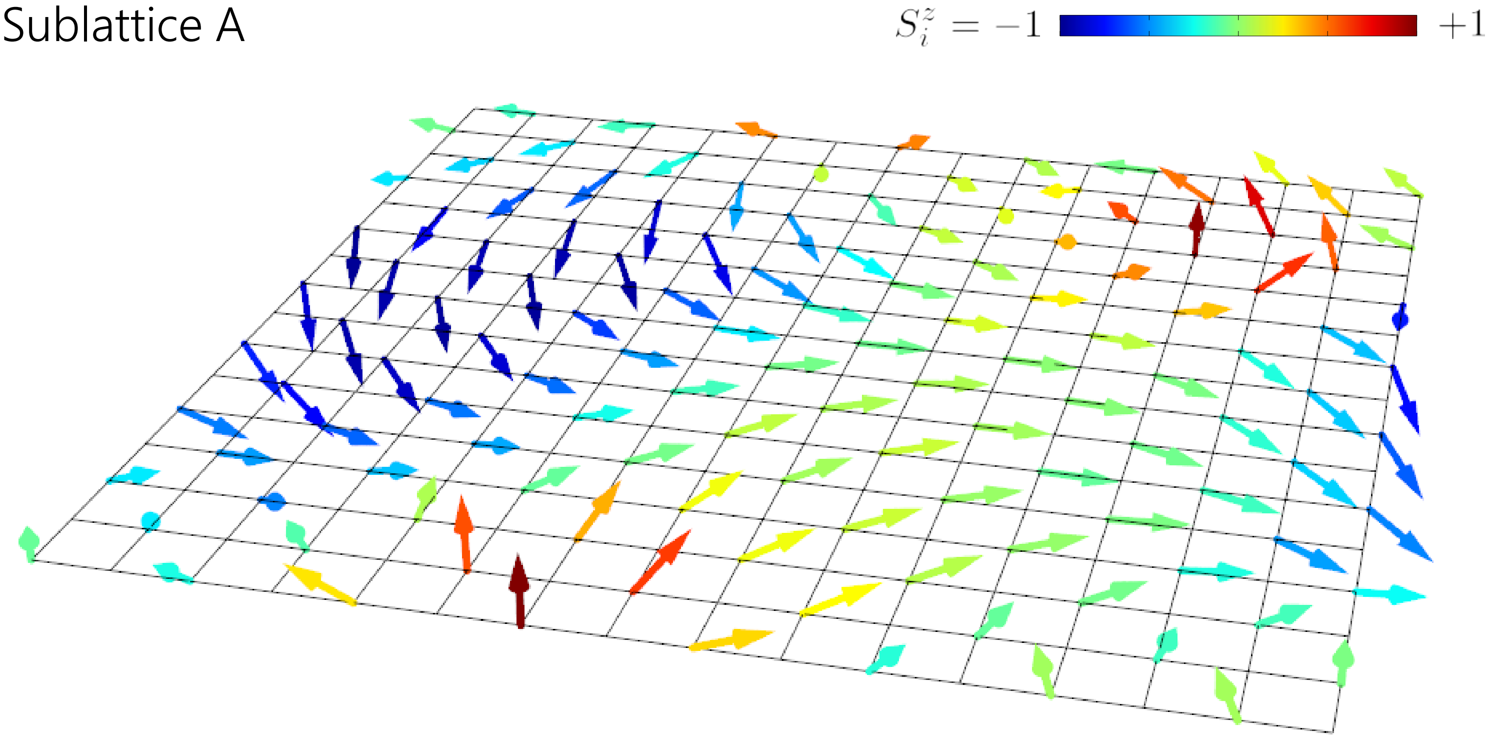
c.f. N. Tuji, T. Oka, H. Aoki, P. Werner, PRB 85, 155124 ('12)

Transient spin structure

Intermediate time domain (τ = 200/t)
Larger cluster (L = 16)

CW

Sublattice A

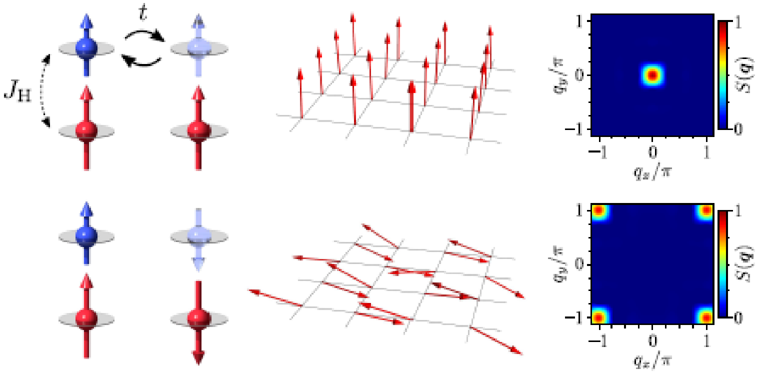


Vortex-like magnetic structure

Summary

Double exchange interaction in non-eq. state revisited

FM to AFM conversion by strong light field



Non-eq. electron distribution

Topological texture in transient state

Experimental confirmation

- Candidates: cubic/layered manganites
- Pulse + CW method : more realistic
- transient optical spectra
- tr. magnetic x-ray diffraction
- tr. ARPES (BZ folding)
- tr. Ramman (AFM magnon)

A. Ono and SI, Phys. Rev. Lett. 119, 207202 (2017) (Editor suggestion)
A. Ono and SI, Phys. Rev. B 95, 085123 (2017)