Transformations of Correlated Electronic States by Electric or Optical Impacts.
International research school and workshop IMPACT – 2024
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Photoinduced Symmetry Breaking, Off-Diagonal Conductivity and Magnetism

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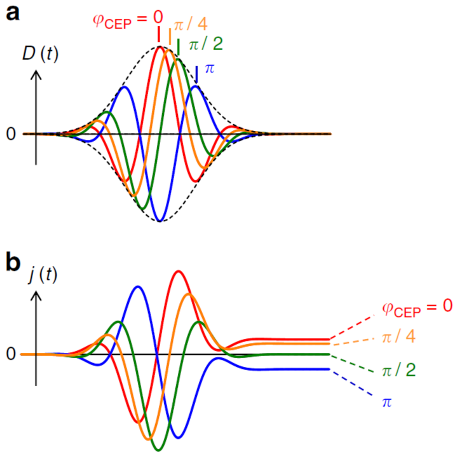
From photoinduced phase transitions
To manipulation of states directly by light

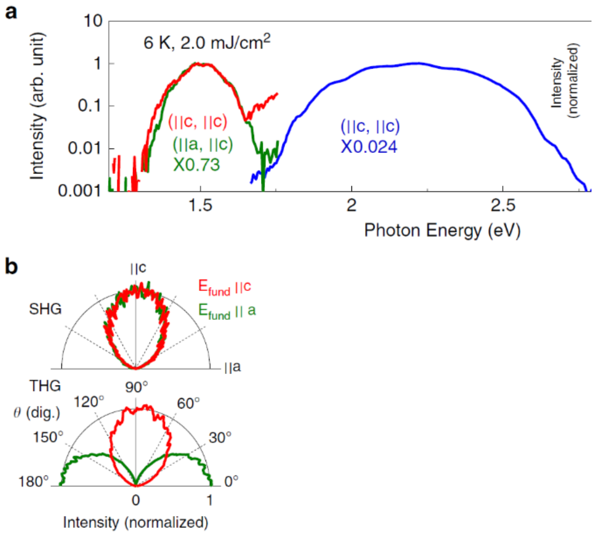
- Symmetry of electronic states modified by light
- Using linearly polarized (LP), circularly polarized (CP), or bicircular (BC) light
- In this talk, states during the photoirradiation are the main topic.
  - Before the main topic
- One example where the symmetry is modified just after irradiation

-- SHG in an organic SC caused by transient breaking of inversion symmetry through CEP controlled linearly polarized light

Y. Kawakami et al., Nat. Commun. 11, 4138 (2020).

-- This SHG is transient.





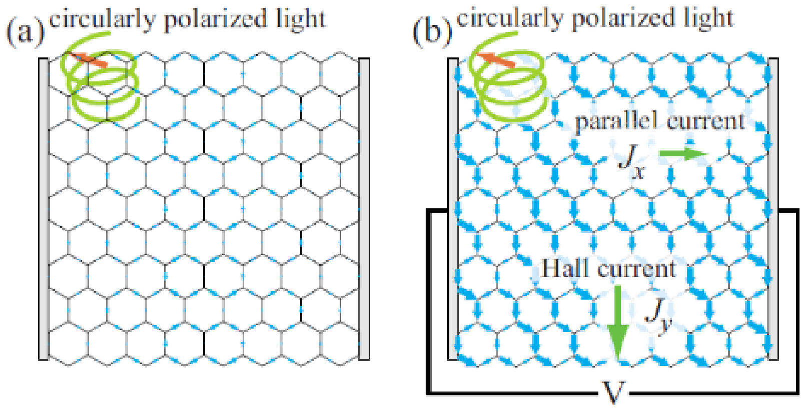
Laser light can break time-reversal/ space-inversion/ mirror symmetry by choosing CP, BC, or LP light.

- Let us see the consequence by calculating σ_{yx}^{C} or σ_{yx}^{S}, which shows how a charge or spin current flows in $\parallel y$ when the probe field is applied in $\parallel x$.

- CP light causes the anomalous Hall effect ($\sigma_{yx}^{C} = -\sigma_{xy}^{C} \neq 0$).

T. Oka and H. Aoki, Phys. Rev. B 79, 081406(R) (2009).

When the helicity of CP light is changed, the sign of σ_{yx}^{C} is reversed: time-reversal symmetry is broken.



Onsager reciprocal relations

$$\sigma_{yx}^{C}(\boldsymbol{H}) = \sigma_{xy}^{C}(-\boldsymbol{H})$$
$$\sigma_{xy}^{C}(-\boldsymbol{H}) = -\sigma_{xy}^{C}(\boldsymbol{H})$$
$$\therefore\ \sigma_{yx}^{C}(\boldsymbol{H}) = -\sigma_{xy}^{C}(\boldsymbol{H})$$

$$\sigma_{yx}^{C}[\boldsymbol{A}_{\mathrm{LCP}}(t)] = \sigma_{xy}^{C}[\boldsymbol{A}_{\mathrm{LCP}}(-t)]$$
$$= \sigma_{xy}^{C}[\boldsymbol{A}_{\mathrm{RCP}}(t)]$$
$$= -\sigma_{xy}^{C}[\boldsymbol{A}_{\mathrm{LCP}}(t)]$$
$$\therefore\ \sigma_{yx}^{C}[\boldsymbol{A}_{\mathrm{LCP}}(t)] = -\sigma_{xy}^{C}[\boldsymbol{A}_{\mathrm{LCP}}(t)]$$

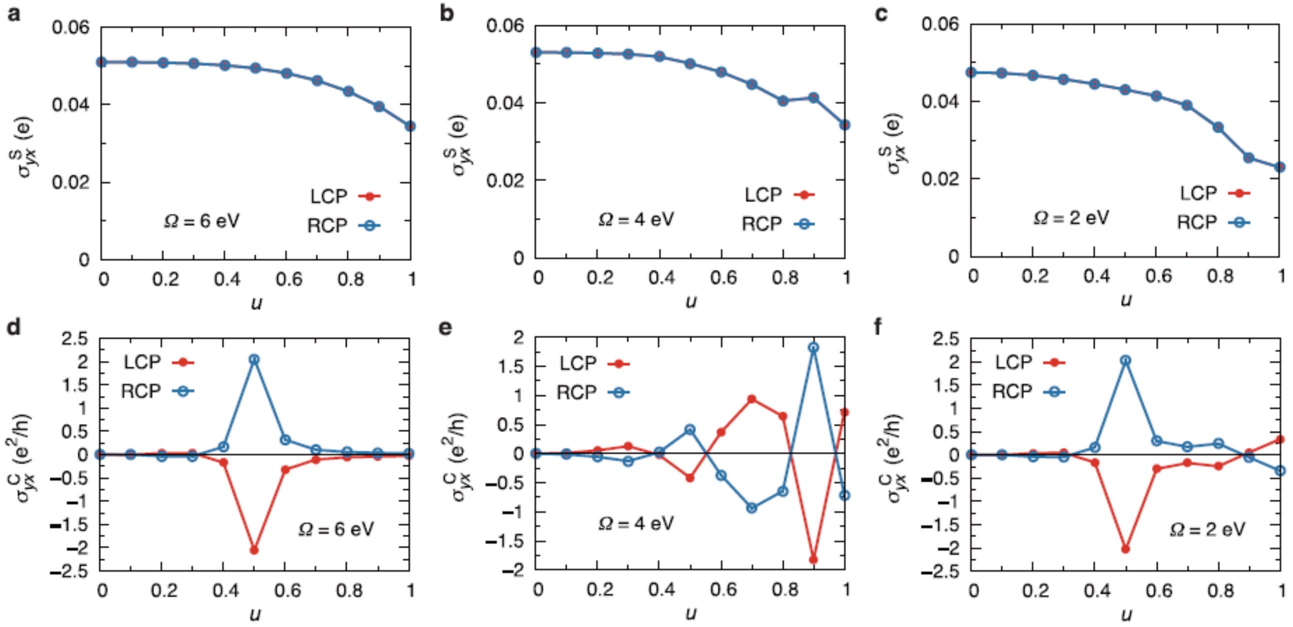
CP light:
It breaks time-reversal symmetry.

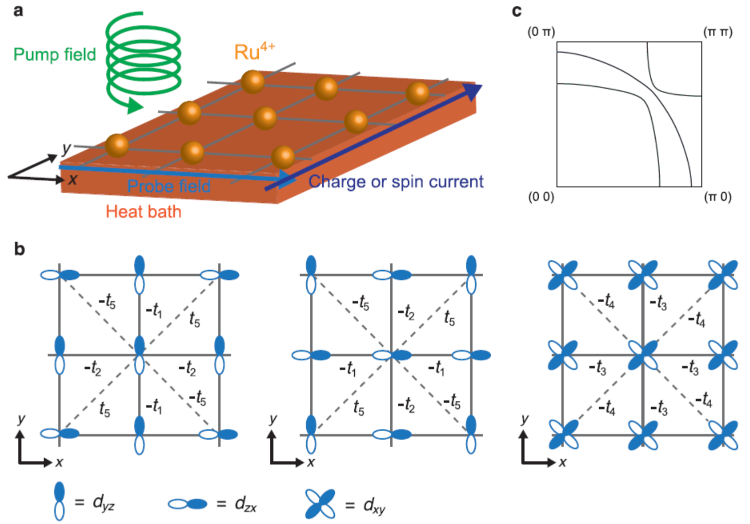
- How about the spin Hall effect (in multi-orbital Sr_2RuO_4 with strong SOC)

N. Arakawa and KY, Commun. Phys. 6, 43 (2023).

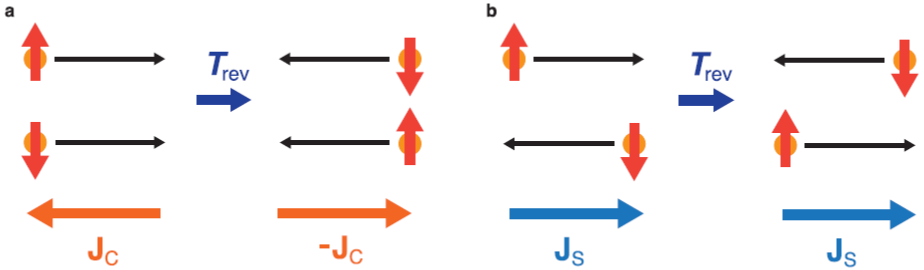
Spin Hall σ_{yx}^{S}

Anomalous Hall σ_{yx}^{C}





$$u = eA_0 = \frac{eE_0 a}{\hbar\Omega}$$



σ_{yx}^{S} is modulated by CP light.
The modulation is independent of the helicity of CP light because the spin current is unchanged by the time-reversal operation.

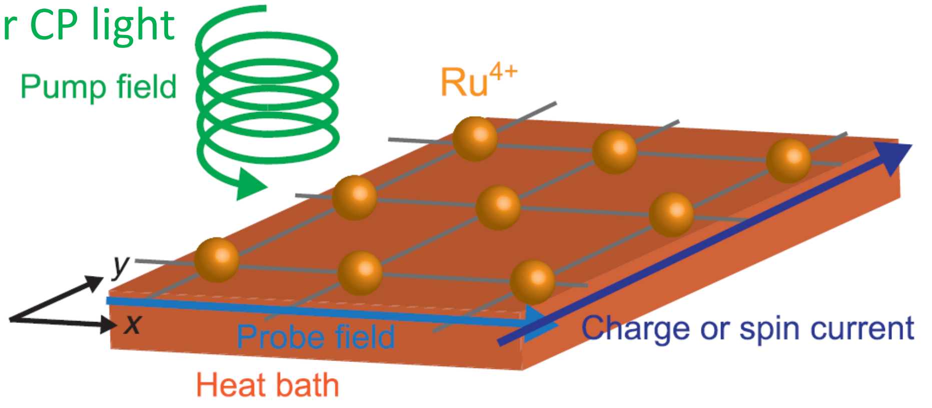
Model

- Pump-probe measurements of SHE or AHE

$$\boldsymbol{A}(t) \;=\; \boldsymbol{A}_{\rm pump}(t) + \boldsymbol{A}_{\rm prob}(t)$$

$$\boldsymbol{A}_{\rm pump}(t) = {}^t(A_0 \cos\Omega t \;\; A_0 \sin(\Omega t + \delta))$$

for CP light



$$\sigma^{\rm Q}_{yx}(t,t') = \frac{1}{i\omega}\frac{\delta\langle j^y_{\rm Q}(t)\rangle}{\delta A^x_{\rm prob}(t')}$$

$$J^y_{\rm Q}(t) = \sum_{\boldsymbol{k}}\sum_{a,b}\sum_{\sigma} v^{({\rm Q})y}_{ab\sigma}(\boldsymbol{k},t)c^{\dagger}_{\boldsymbol{k}a\sigma}(t)c_{\boldsymbol{k}b\sigma}(t),$$

$$v^{({\rm C})y}_{ab\sigma}(\boldsymbol{k},t) = (-e)\frac{\partial\epsilon_{ab}(\boldsymbol{k},t)}{\partial k_y}, \quad v^{({\rm S})y}_{ab\sigma}(\boldsymbol{k},t) = \frac{1}{2}{\rm sgn}(\sigma)\frac{\partial\epsilon_{ab}(\boldsymbol{k},t)}{\partial k_y}$$

Units: $\hbar = 1,\ k_{\rm B} = 1,$ and $a = 1$

N. Arakawa and KY, Commun. Phys. 6, 43 (2023).

Some detail 2/3

Method

- Keldysh Green's function + Floquet representation

$$G = G_0 + G_0 \Sigma G$$ Dyson's eq. (matrix form)

$$G = \begin{pmatrix} G^{\mathrm{R}} & G^{\mathrm{K}} \\ 0 & G^{\mathrm{A}} \end{pmatrix}, G_0 = \begin{pmatrix} G_0^{\mathrm{R}} & G_0^{\mathrm{K}} \\ 0 & G_0^{\mathrm{A}} \end{pmatrix}, \Sigma = \begin{pmatrix} \Sigma^{\mathrm{R}} & \Sigma^{\mathrm{K}} \\ 0 & \Sigma^{\mathrm{A}} \end{pmatrix}$$

Coupling to heat bath

$$G^r_{a\sigma b\sigma';n}(\boldsymbol{k};\omega) = \int_{-\infty}^{\infty} dt_{\mathrm{rel}} e^{i\omega t_{\mathrm{rel}}} \int_0^T \frac{dt_{\mathrm{av}}}{T} e^{in\Omega t_{\mathrm{av}}} G^r_{a\sigma b\sigma'}(\boldsymbol{k};t,t')$$

Wigner rep.

$$t_{\mathrm{rel}} = t - t',\ t_{\mathrm{av}} = \frac{t+t'}{2}$$

$$[G^r_{a\sigma b\sigma'}(\boldsymbol{k};\omega)]_{mn} = G^r_{a\sigma b\sigma';m-n}(\boldsymbol{k};\omega + \frac{m+n}{2}\Omega)$$ Floquet rep.

Some detail 3/3

Analytic results

- Time-averaged ~~Hall~~ off-diagonal conductivities

Hall if $\sigma_{yx}^{C} = -\sigma_{xy}^{C}$
OK for CPL

$$\sigma_{yx}^{\mathrm{Q}} = \lim_{\omega\to 0} \mathrm{Re} \int_0^T \frac{dt_{\mathrm{av}}}{T} \int_{-\infty}^{\infty} dt_{\mathrm{rel}} e^{i\omega t_{\mathrm{rel}}} \sigma_{yx}^{\mathrm{Q}}(t,t')$$

$$= \frac{1}{N} \sum_{\boldsymbol{k}} \sum_{a,b,c,d} \sum_{\sigma,\sigma'} \int_{-\Omega/2}^{\Omega/2} \frac{d\omega'}{2\pi} \sum_{m,l,n,q=-\infty}^{\infty} [v_{ab\sigma}^{(\mathrm{Q})y}(\boldsymbol{k})]_{ml}$$

$$\times [v_{cd\sigma'}^{(\mathrm{C})x}(\boldsymbol{k})]_{nq} \Big\{ \frac{\partial [G_{b\sigma c\sigma'}^{\mathrm{R}}(\boldsymbol{k},\omega')]_{ln}}{\partial\omega'} [G_{d\sigma' a\sigma}^{<}(\boldsymbol{k},\omega')]_{qm}$$

$$- [G_{b\sigma c\sigma'}^{<}(\boldsymbol{k},\omega')]_{ln} \frac{\partial [G_{d\sigma' a\sigma}^{\mathrm{A}}(\boldsymbol{k},\omega')]_{qm}}{\partial\omega'} \Big\}$$

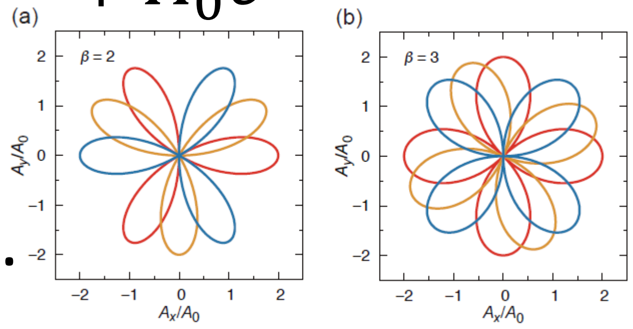
$$G^{<} = \frac{1}{2}(G^{\mathrm{K}} - G^{\mathrm{R}} + G^{\mathrm{A}})$$

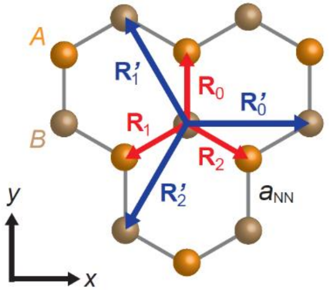
$$[v_{ab\sigma}^{(\mathrm{Q})\nu}(\boldsymbol{k})]_{mn} = \int_0^T \frac{dt}{T} e^{i(m-n)\Omega t} v_{ab\sigma}^{(\mathrm{Q})\nu}(\boldsymbol{k},t)$$

N. Arakawa and KY, Commun. Phys. 6, 43 (2023).

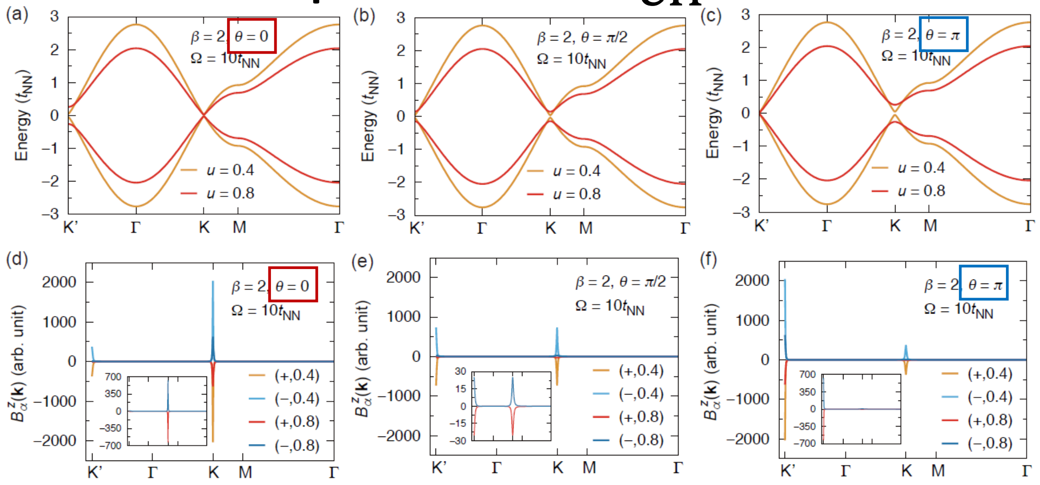
Bicircular (BC) light:

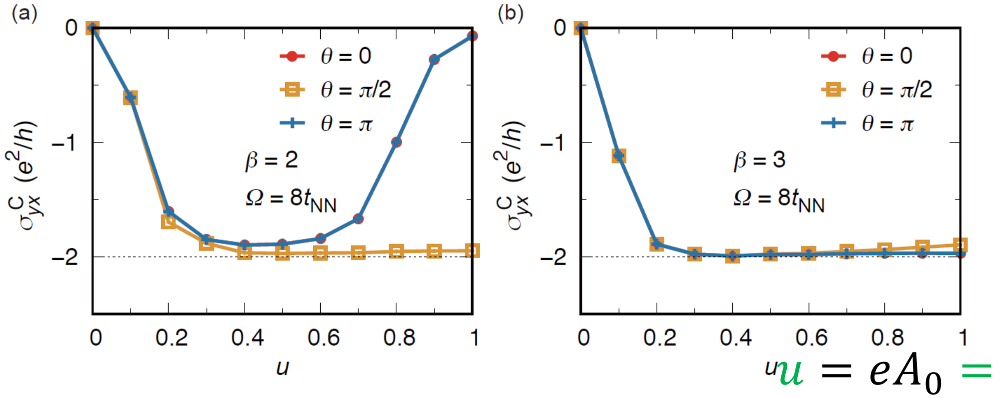
it consists of L&R-handed CP light w/ freq. Ω & $\beta\Omega$ and phase diff. θ, $A_x(t) + iA_y(t) = A_0 e^{i\Omega t} + A_0 e^{-i(\beta\Omega t - \theta)}$





- BC light breaks time-rev. sym. at least.
- It also breaks inversion sym. dep. on β&θ.
- $\beta = 2$ has a trefoil field. (even β has odd # of leaves in the trajectory)
- When graphene is irradiated by BC light, the valley degeneracy at K and K' is lifted for $\beta = 2$ and $\theta = 0, \pi$, leading to disproportionate Berry curvatures at K and K' points in H_{eff} and a valley-selective Hall effect.



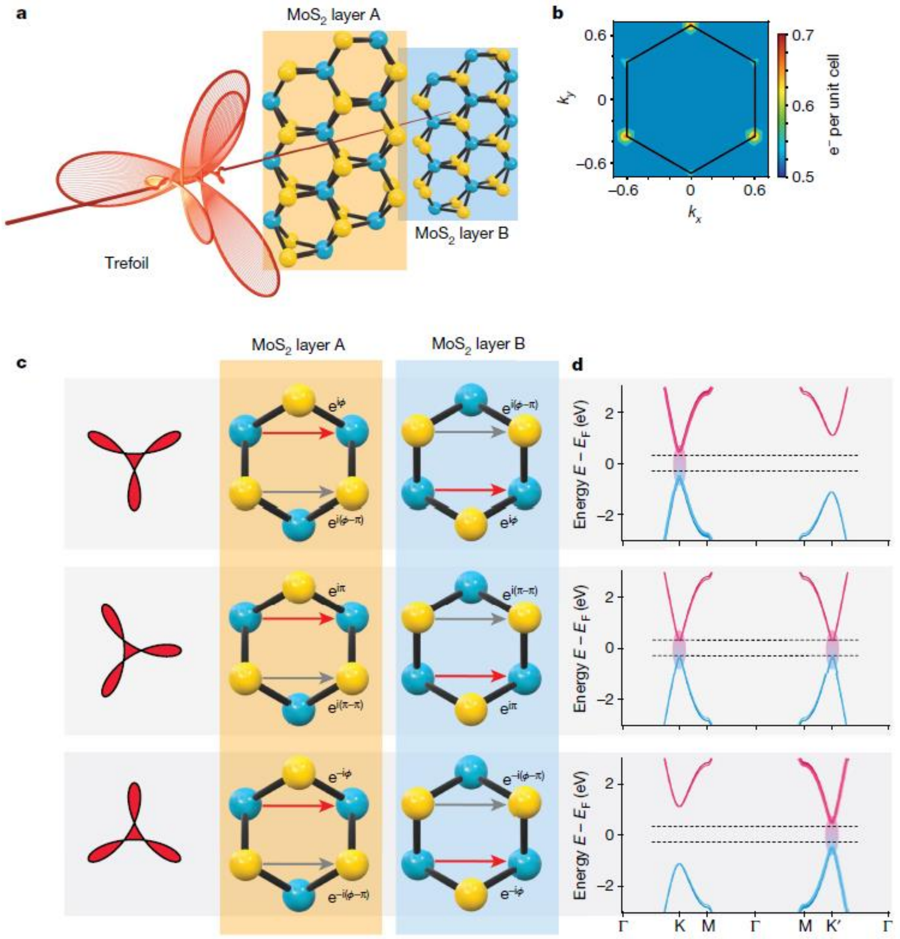


$u = eA_0 = \dfrac{eE_0 a}{\hbar\Omega}$

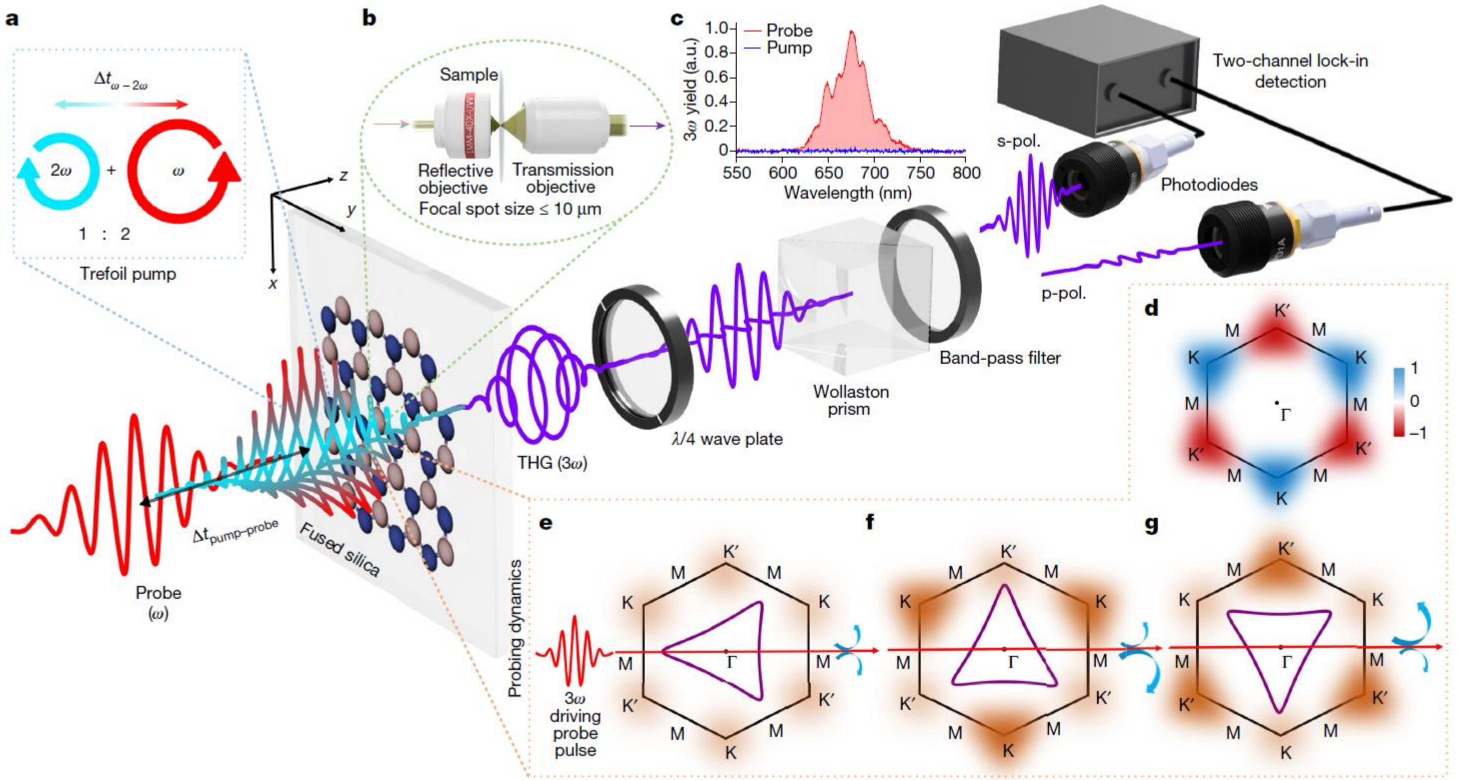
N. Arakawa and KY, Phys. Rev. B **109**, L241201 (2024).

Recently, trefoil $(\beta = 2)$ fields shown to induce valley pol.

Igor Tyulnev et al.,
"Valleytronics in bulk MoS_2 with a topologic optical field,"
Nature 628, 746 (2024).

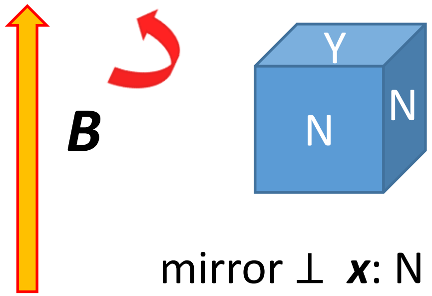


Sambit Mitra et al.,
"Light-wave-controlled Haldane model in monolayer hexagonal boron nitride"
Nature 628, 752 (2024).



So far, breaking of time-reversal & inversion symmetry is treated. Then, how about that of mirror (reflection) symmetry?

- Mirror symmetry is relevant to off-diagonal (transverse) conductivity including the Hall and anomalous Hall effects.



mirror ⊥ ***x***: N (sym. broken)
mirror ⊥ ***y***: N (sym. broken)
B* // *z mirror ⊥ ***z***: Y (sym. preserved)

M. Naka, Y. Motome, and H. Seo, PRB 106, 195149 (2022).
"Anomalous Hall effect in antiferromagnetic perovskites"

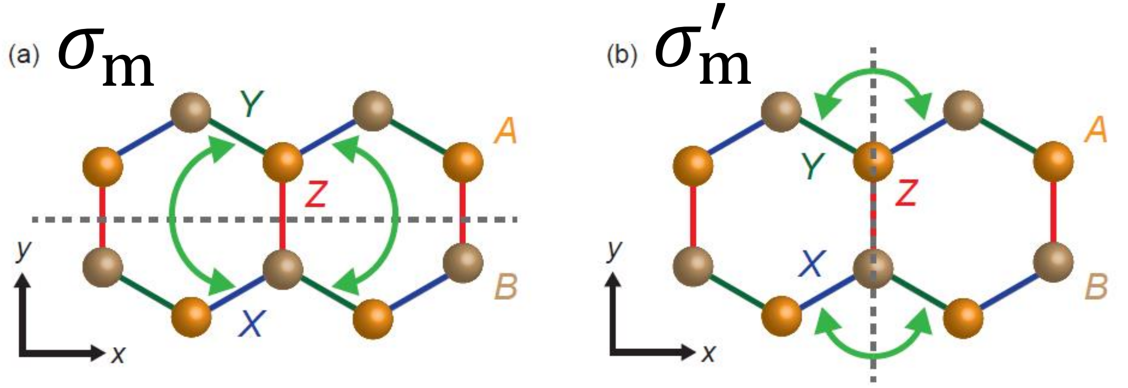
TABLE I. Relation between the AHE and the symmetries of the AFM patterns on *Pbnm* structure. Y (N) represents the presence (absence) of the symmetry.

AFM	mirror ⊥ z	b-glide ⊥ x	n-glide ⊥ y	active AHE
$C_xF_yA_z$	N	N	Y	σ_{xz}
$F_xC_yG_z$	N	Y	N	σ_{yz}
$G_xA_yF_z$	Y	N	N	σ_{xy}
$A_xG_yC_z$	Y	Y	Y	N/A

glide (=reflection + translation) ~ mirror (=reflection)

Which mirror (glide) symmetry is broken is important for $\sigma_{ij} \neq 0$ with particular ij.

In addition to CP, we consider the breaking of mirror symmetry, keeping time-reversal sym., i.e., LP light w/ polarization not ∥, ⊥ the mirror plane.

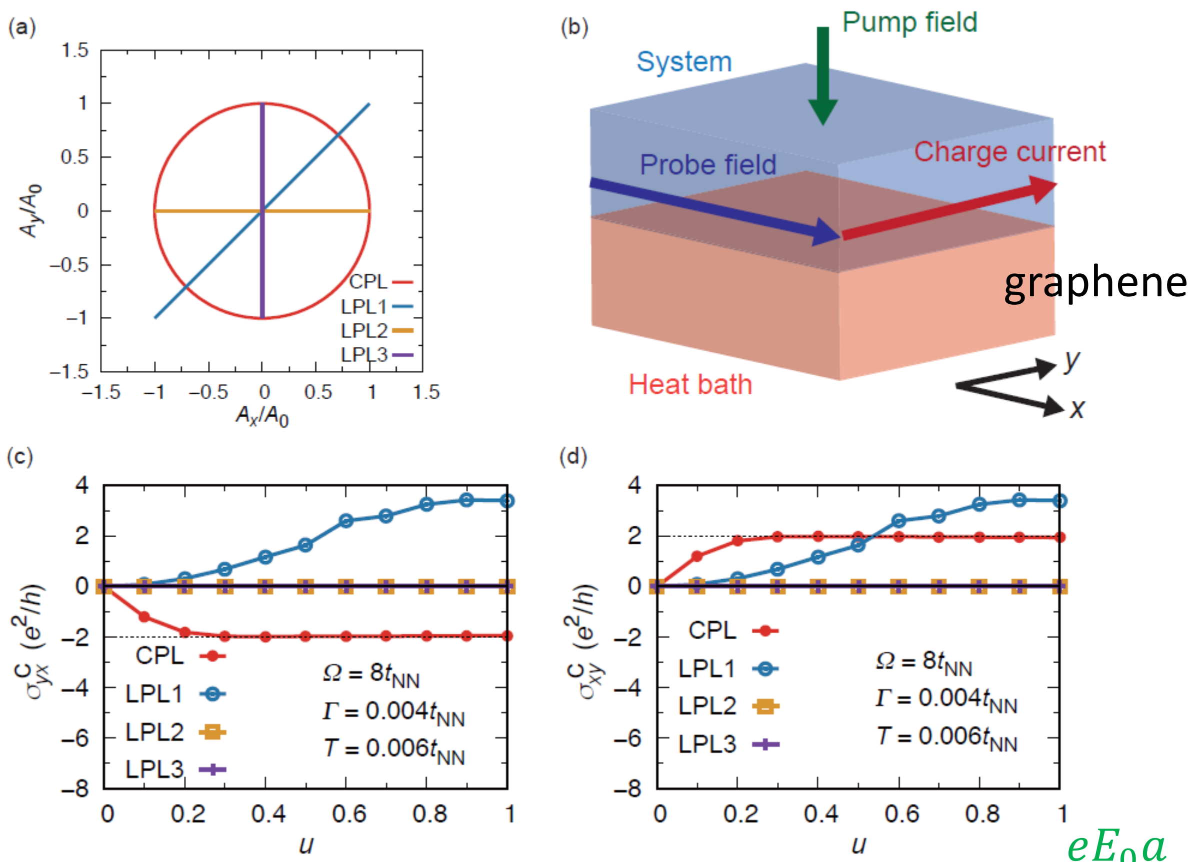


	CPL	LPL1	LPL2	LPL3
T_rev	Broken	Preserved	Preserved	Preserved
σ_m	Broken	Broken	Preserved	Broken
$\sigma_\mathrm{m} T_t$	Broken	Broken	Preserved	Preserved
σ'_m	Broken	Broken	Broken	Preserved
$\sigma'_\mathrm{m} T_t$	Broken	Broken	Preserved	Preserved
C_3	Broken	Broken	Broken	Broken
$C_3 T_t$	Preserved	Broken	Broken	Broken
σ^C_{yx}	Antisymmetric	Symmetric	Vanishing	Vanishing

$T_t : t \to t - \frac{\pi}{\Omega}$ (for $\sigma_\mathrm{m} T_t$)

$T_t : t \to t - \frac{\pi}{\Omega}$ (for $\sigma'_\mathrm{m} T_t$)

$T_t : t \to t + \frac{2\pi}{3\Omega}$ (for $C_3 T_t$)



$u = eA_0 = \frac{eE_0 a}{\hbar\Omega}$

N. Arakawa and KY, J. Phys. Soc. Jpn. 93, 084701 (2024).

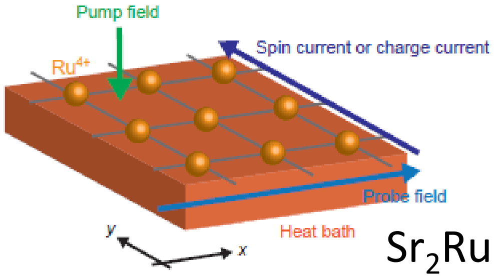
- LP light can break mirror symmetry, depending on its polarization, and lead to nonzero σ^C_{yx}, which is not a Hall effect since $\sigma^C_{yx} = \sigma^C_{xy}$ (time-reversal symmetry is preserved).

Bicircular (BC) light again:

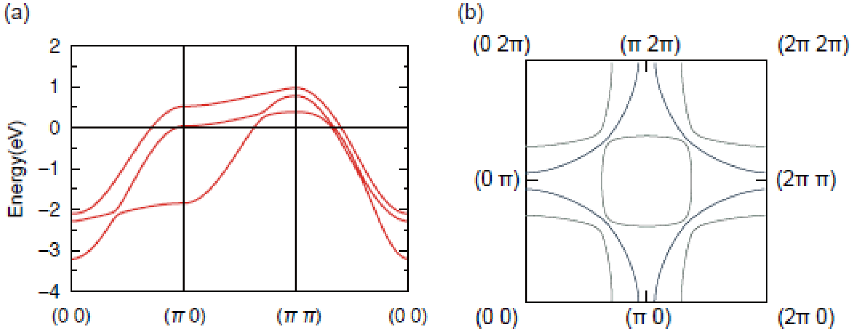
$$A_x(t) + iA_y(t) = A_0 e^{i\Omega t} + A_0 e^{-i(\beta\Omega t - \theta)}$$

- The trajectory of $\boldsymbol{A}(t)$ can break mirror symmetry, depending on θ.

N. Arakawa and KY, in preparation

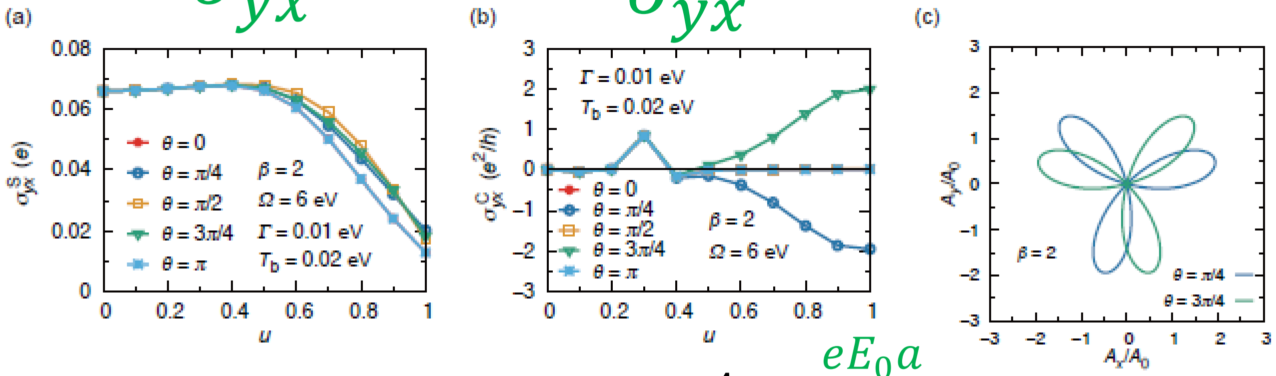


Sr_2RuO_4



Spin Hall σ^{S}_{yx}

Anomalous Hall σ^{C}_{yx}



$$u = eA_0 = \frac{eE_0 a}{\hbar\Omega}$$

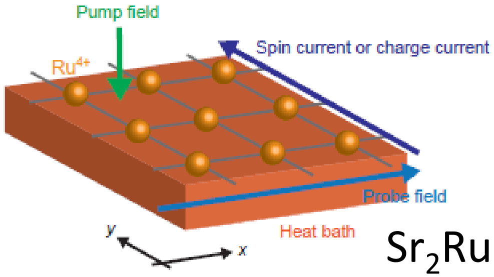
If a mirror operation for θ gives the trajectory for θ_{M}, σ^{C}_{yx} for θ and σ^{C}_{yx} for θ_{M} are of opposite signs and almost the same magnitude for moderately large field strengths u.

Bicircular (BC) light again:

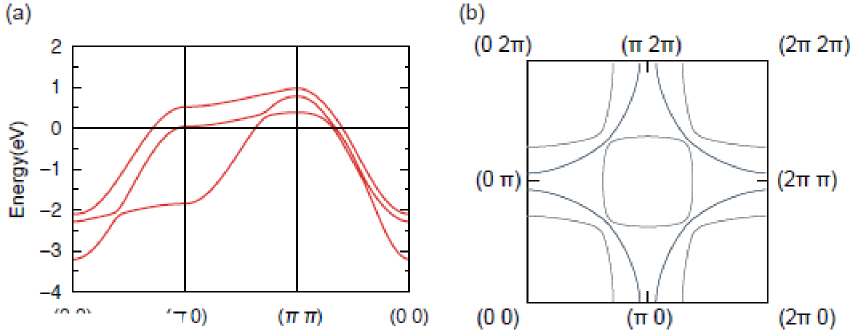
$$A_x(t) + iA_y(t) = A_0 e^{i\Omega t} + A_0 e^{-i(\beta\Omega t - \theta)}$$

- The trajectory of $\boldsymbol{A}(t)$ can break mirror symmetry, depending on θ.

N. Arakawa and KY, in preparation

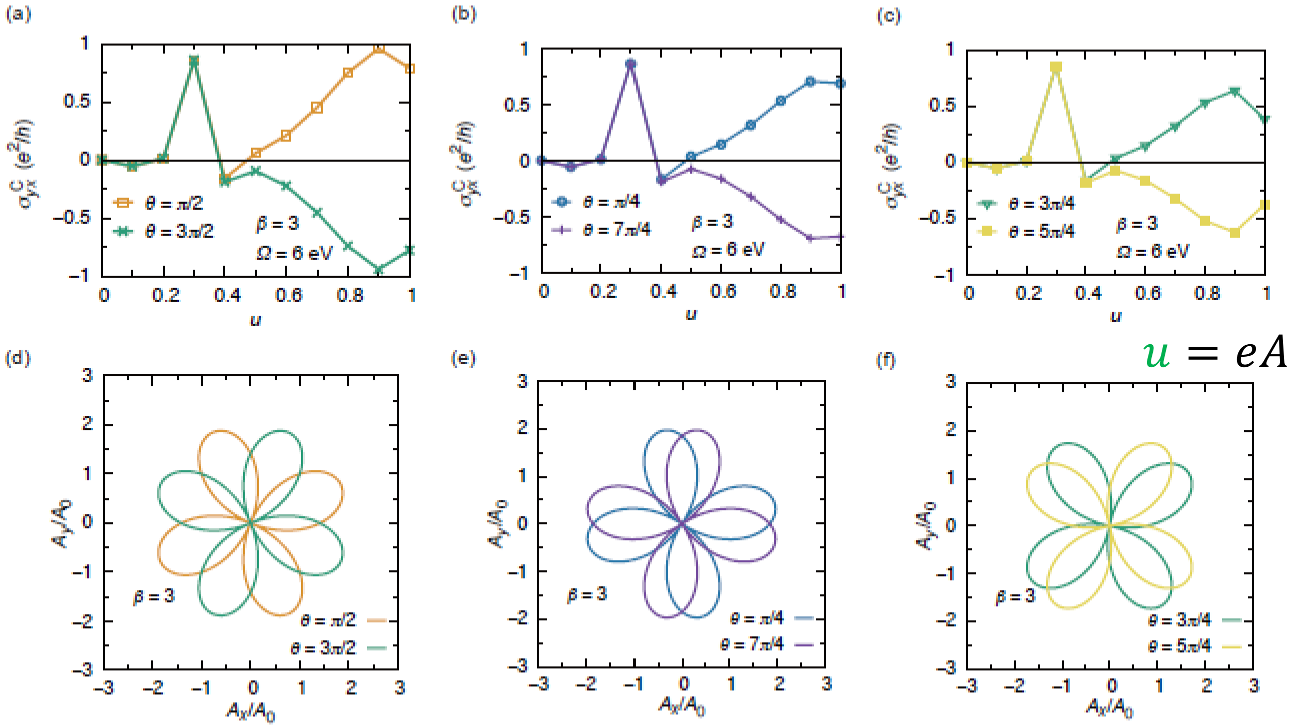


Sr_2RuO_4



Anomalous Hall

σ_{yx}^{C}



$$u = eA_0 = \frac{eE_0 a}{\hbar\Omega}$$

Summary 1:

- Linearly pol. (LP) light can break mirror sym.: $\sigma_{yx}^{C} = \sigma_{xy}^{C} \neq 0$ (not Hall)

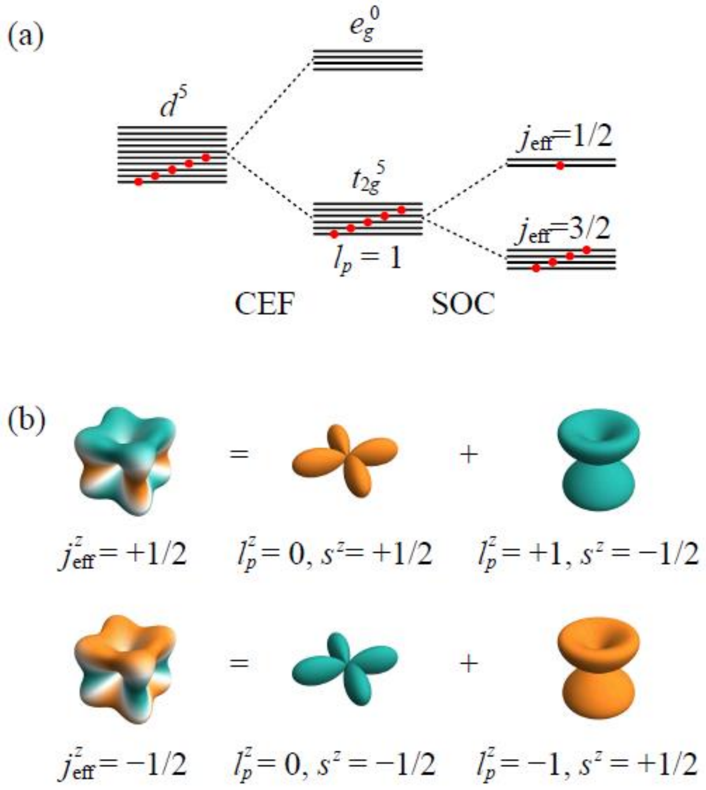
- Circularly pol. (CP) light breaks time-rev. sym.: $\sigma_{yx}^{C} = -\sigma_{xy}^{C} \neq 0$, inverse Faraday, etc.

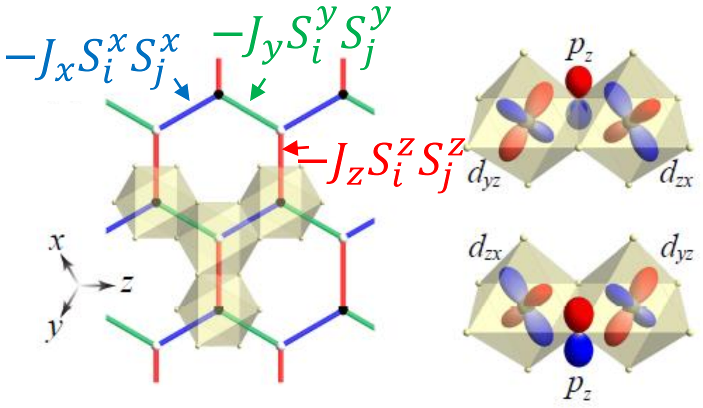
- Bicircular (BC) light can break time-rev. and inv. sym.: valley-selective Hall, valley pol. etc.

or

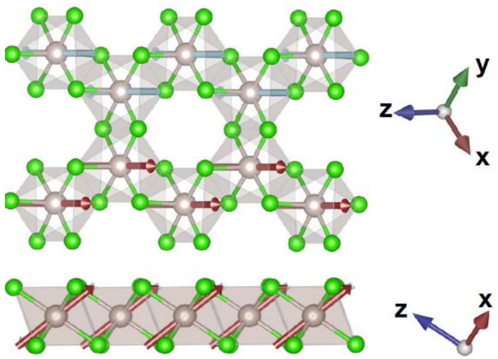
time-rev. and mirror sym.: θ-sensitive σ_{yx}^{C} for $\frac{eE_0 a}{\hbar\Omega} \sim 1$

Photoinduced magnetization in quantum spin-liquid α-$RuCl_3$

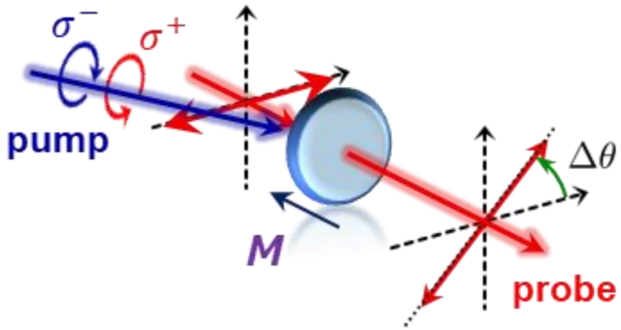




Y. Motome and J. Nasu, JPSJ 2020



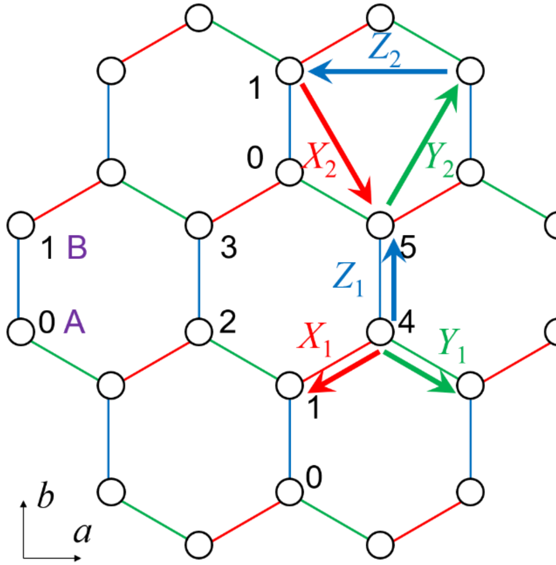
S. M. Winter et al., PRB 2016



T. Amano, KY, S. Iwai *et al.*,
Phys. Rev. Res. 4, L032032 (2022).

- Theories
  - A. Kitaev, Ann. Phys. 321, 2 (2006)
  - G. Jackeli and G. Khaliullin, PRL 102, 017205 (2009)
  - H.-S. Kim and H.-Y. Kee, PRB 93, 155143 (2016).
  - S. M. Winter *et al.*, PRB 93, 214431 (2016).
- Experiments
  - L. J. Sandilands *et al.*, PRB 93, 075144 (2016).
  - L. J. Sandilands *et al.*, PRB 94, 195156 (2016).
  - P. Warzanowski *et al.*, PRR 2, 042007(R) (2020).

Three-orbital Hubbard model for Kitaev materials



p.b.c., 6 sites (3 unit cells)

S. M. Winter, Y. Li, H. O. Jeschke, and Roser Valentí, PRB2016

$$H_{\rm tot} = H_{\rm hop} + H_{\rm CF} + H_{\rm SO} + H_U$$

$$\vec{c}_i^\dagger = \left(c_{i,yz,\uparrow}^\dagger\ c_{i,yz,\downarrow}^\dagger\ c_{i,xz,\uparrow}^\dagger\ c_{i,xz,\downarrow}^\dagger\ c_{i,xy,\uparrow}^\dagger\ c_{i,xy,\downarrow}^\dagger\right)$$

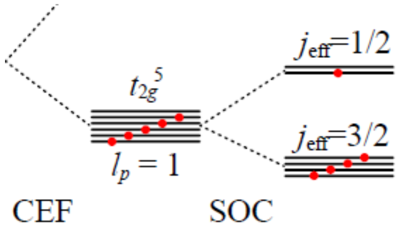
(hole picture)

$$H_{\rm hop} = -\sum_{ij} \vec{c}_i^\dagger \{\mathbf{T}_{ij} \otimes \mathbb{I}_{2\times 2}\} \vec{c}_j$$

$$\mathbf{T}_1^{\rm X} = \begin{pmatrix} t_3 & t_4 & t_4 \\ t_4 & t_1 & t_2 \\ t_4 & t_2 & t_1 \end{pmatrix} \qquad \mathbf{T}_1^{\rm Y} = \begin{pmatrix} t_1 & t_4 & t_2 \\ t_4 & t_3 & t_4 \\ t_2 & t_4 & t_1 \end{pmatrix} \qquad \mathbf{T}_1^{\rm Z} = \begin{pmatrix} t_1 & t_2 & t_4 \\ t_2 & t_1 & t_4 \\ t_4 & t_4 & t_3 \end{pmatrix}$$

$$H_{\rm SO} = \frac{\lambda}{2} \sum_i \vec{c}_i^\dagger \begin{pmatrix} 0 & -i\sigma_z & i\sigma_y \\ i\sigma_z & 0 & -i\sigma_x \\ -i\sigma_y & i\sigma_x & 0 \end{pmatrix} \vec{c}_i$$

$$H_U = U, U', J_{\rm H} \text{ terms} \qquad U' = U - 2J_{\rm H}$$



Y. Motome and J. Nasu, JPSJ 2020

$$J_{\rm eff} = \frac{1}{2} : \text{half filled}$$

$$\left|\frac{1}{2}, \frac{1}{2}\right\rangle = p_\uparrow^\dagger |0\rangle \equiv \frac{1}{\sqrt{3}}\left(-c_{xy,\uparrow}^\dagger - i c_{xz,\downarrow}^\dagger - c_{yz,\downarrow}^\dagger\right)|0\rangle$$

$$\left|\frac{1}{2}, -\frac{1}{2}\right\rangle = p_\downarrow^\dagger |0\rangle \equiv \frac{1}{\sqrt{3}}\left(c_{xy,\downarrow}^\dagger + i c_{xz,\uparrow}^\dagger - c_{yz,\uparrow}^\dagger\right)|0\rangle$$

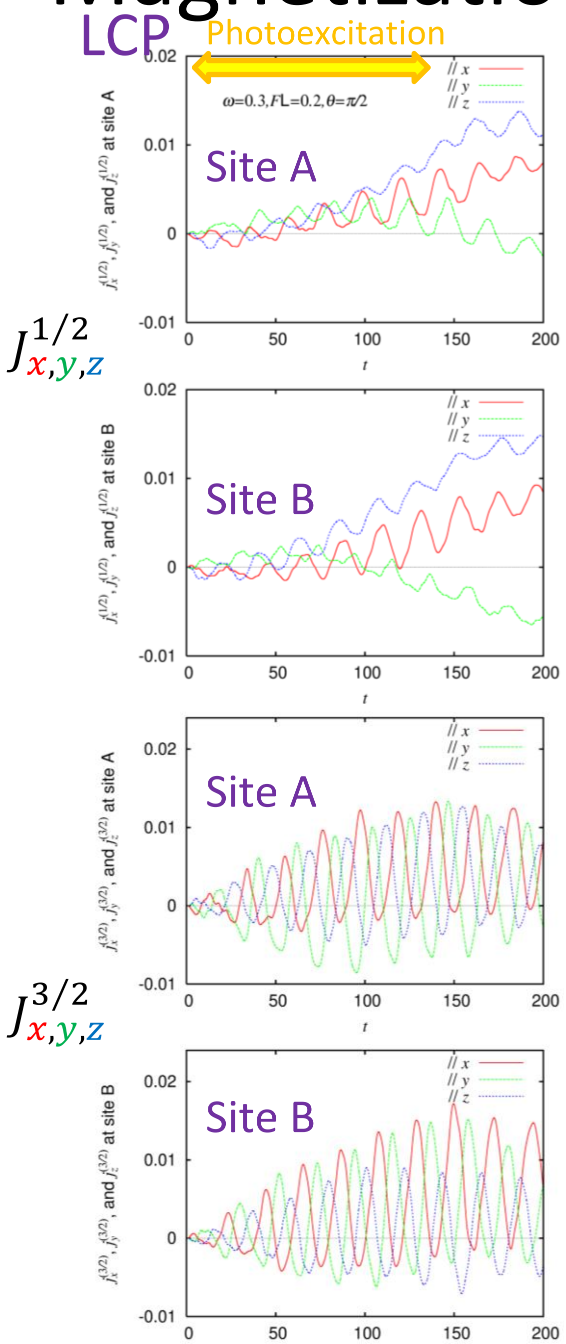
$$m_x^{(1/2)} \equiv \frac{1}{2}\left\langle p_\uparrow^\dagger p_\downarrow + p_\downarrow^\dagger p_\uparrow \right\rangle$$

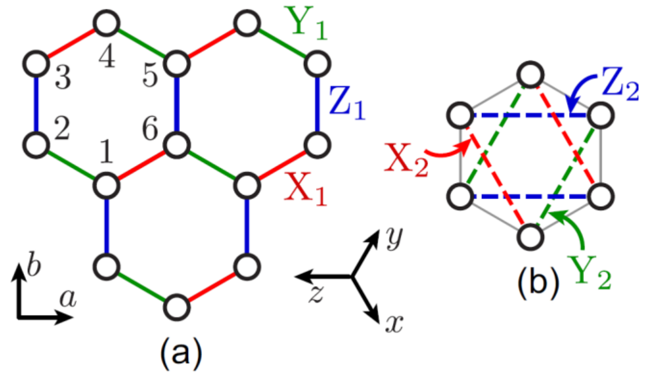
$$m_y^{(1/2)} \equiv \frac{1}{2}\left\langle -i p_\uparrow^\dagger p_\downarrow + i p_\downarrow^\dagger p_\uparrow \right\rangle$$

$$m_z^{(1/2)} \equiv \frac{1}{2}\left\langle p_\uparrow^\dagger p_\uparrow - p_\downarrow^\dagger p_\downarrow \right\rangle$$

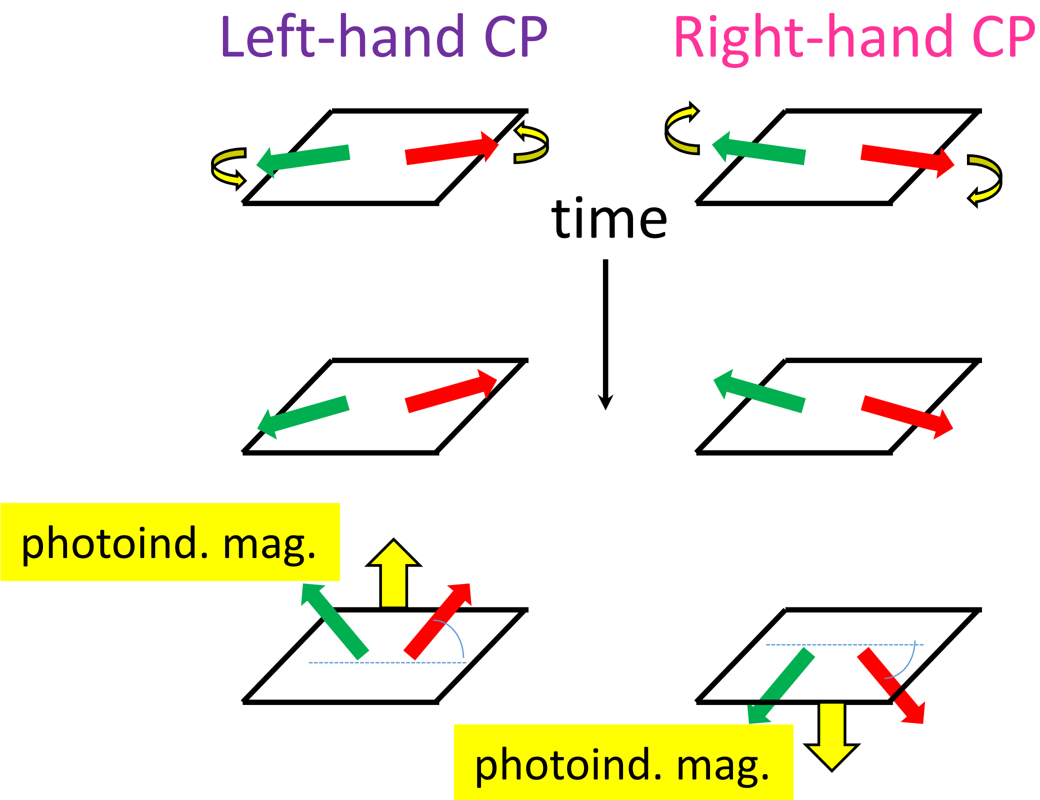
$$m_\perp^{(1/2)} \equiv \frac{1}{\sqrt{3}}\left(m_x^{(1/2)} + m_y^{(1/2)} + m_z^{(1/2)}\right)$$

Magnetization dynamics induced by circularly polarized light



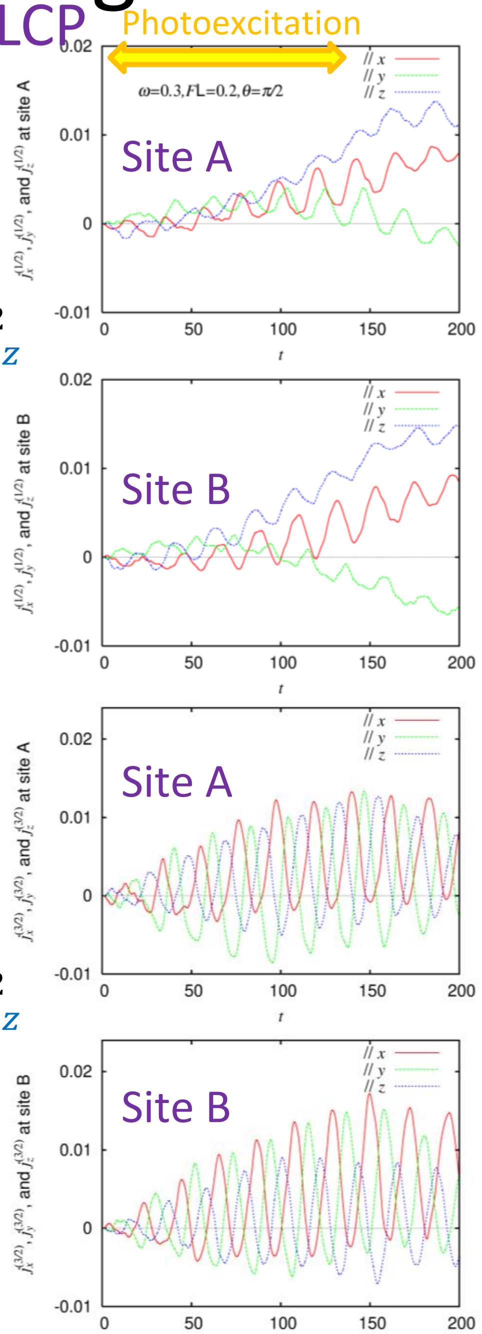


KY, JPSJ 91, 104702 (2022)



Magnetization dynamics induced by circularly polarized light

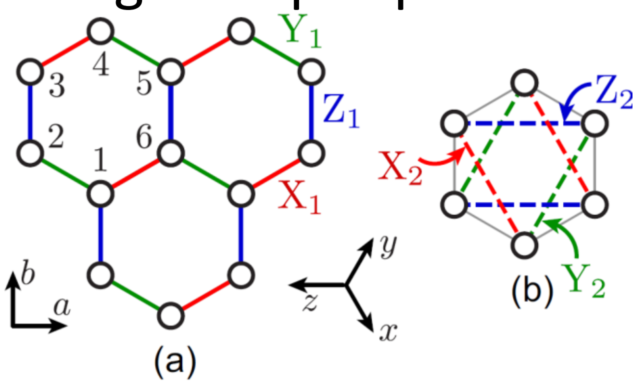
LCP



$J^{1/2}_{x,y,z}$

$J^{3/2}_{x,y,z}$

High-freq. expansion in Floquet theory: $H_F^{(2)}$



$$H_F^{(2)} = \sum_{m>0} \frac{[H_m, H_{-m}]}{m\hbar\omega}$$

KY, JPSJ 91, 104702 (2022)

$$H_F^{(2)} \simeq \frac{1}{\hbar\omega} \sum_{iab\sigma} J_1^2\left(\frac{eaF_{\mathrm{L}}}{\hbar\omega}\right)(2i)\sin\frac{2\pi}{3}\left([\mathrm{T}_1^{\mathrm{Y}}, \mathrm{T}_1^{\mathrm{Z}}] + [\mathrm{T}_1^{\mathrm{Z}}, \mathrm{T}_1^{\mathrm{X}}] + [\mathrm{T}_1^{\mathrm{X}}, \mathrm{T}_1^{\mathrm{Y}}]\right)_{ab} c^\dagger_{i,a,\sigma} c_{i,b,\sigma}$$

$$\simeq \underline{\frac{1}{\hbar\omega} J_1^2\left(\frac{eaF_{\mathrm{L}}}{\hbar\omega}\right)\sqrt{3}(t_2 - t_4)[t_2 - t_4 + 2(t_3 - t_1)]}$$

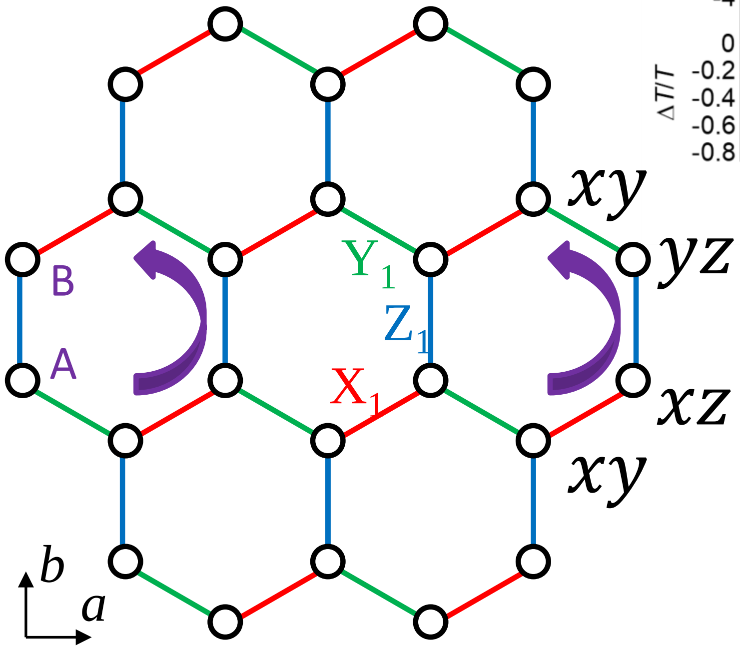
< 0 for L(, > 0 for R)

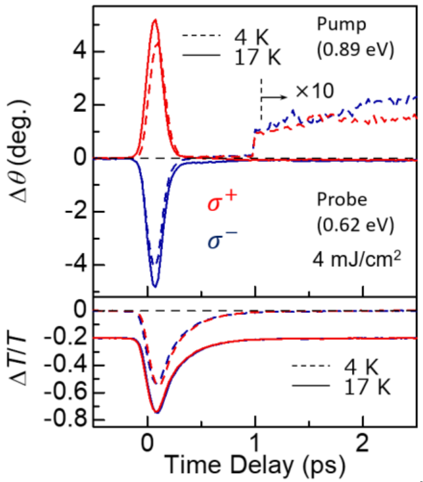
$$\times \sum_{i\sigma} \begin{pmatrix} c^\dagger_{i,yz,\sigma} & c^\dagger_{i,xz,\sigma} & c^\dagger_{i,xy,\sigma} \end{pmatrix} \begin{pmatrix} 0 & -i & i \\ i & 0 & -i \\ -i & i & 0 \end{pmatrix} \begin{pmatrix} c_{i,yz,\sigma} \\ c_{i,xz,\sigma} \\ c_{i,xy,\sigma} \end{pmatrix}$$

$$l_x = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix},\ l_y = \begin{pmatrix} 0 & 0 & i \\ 0 & 0 & 0 \\ -i & 0 & 0 \end{pmatrix},\ l_z = \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Emergence of angular momentum in multi-orbital systems

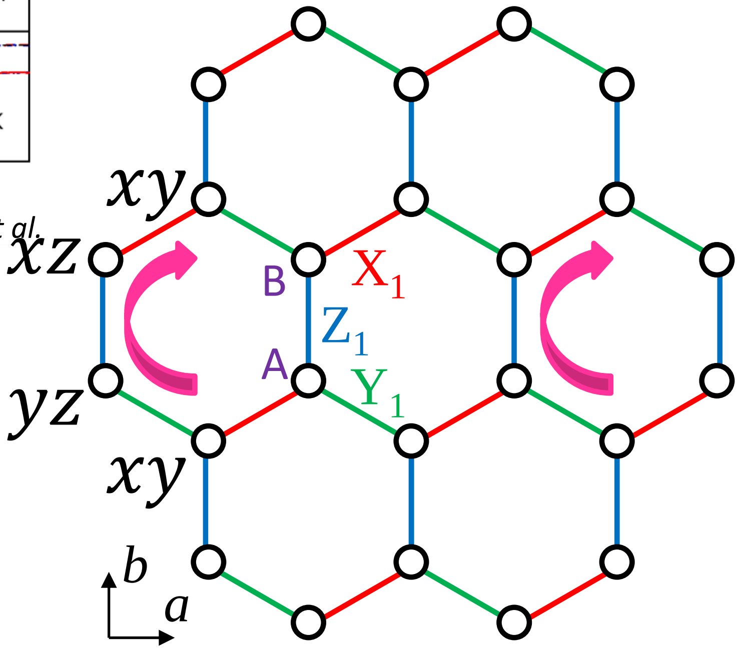
- Left-hand circular pol.





T. Amano, KY, S. Iwai *et al.* Phys. Rev. Res. (2022).

- Right-hand circular pol.



- Main transitions when (interorbital) t_2 processes are dominant.
- Quantitatively, (intraorbital) t_3 processes are also important. $\mathbf{T}_1^Z = \begin{pmatrix} t_1 & t_2 & t_4 \\ t_2 & t_1 & t_4 \\ t_4 & t_4 & t_3 \end{pmatrix}$

Summary 2:

CP → effective mag. fields in multi-orb. systems

- Photoinduced magnetization in α-$RuCl_3$: explained by Floquet theory.
- $H_F^{(2)} = \sum_{m>0} \frac{[H_m, H_{-m}]}{m\hbar\omega}$ gives a helicity-dependent effective magnetic field on l_{eff} ⊥ honeycomb plane.

T. Amano, KY, S. Iwai *et al.* Phys. Rev. Research 4, L032032 (2022).

KY, JPSJ 91, 104702 (2022).

Intersite interorbital hopping processes are essential.

Charge DOF in frustrating spin systems for emergent magnetization

in the spin-orbit assisted Mott insulator.

⇔ effective magnetic fields deep in the insulating phase.

A. Sriram and M. Claassen, Phys. Rev. Research 4, L032036 (2022),

S. Banerjee et al., Phys. Rev. B 105, L180414 (2022).