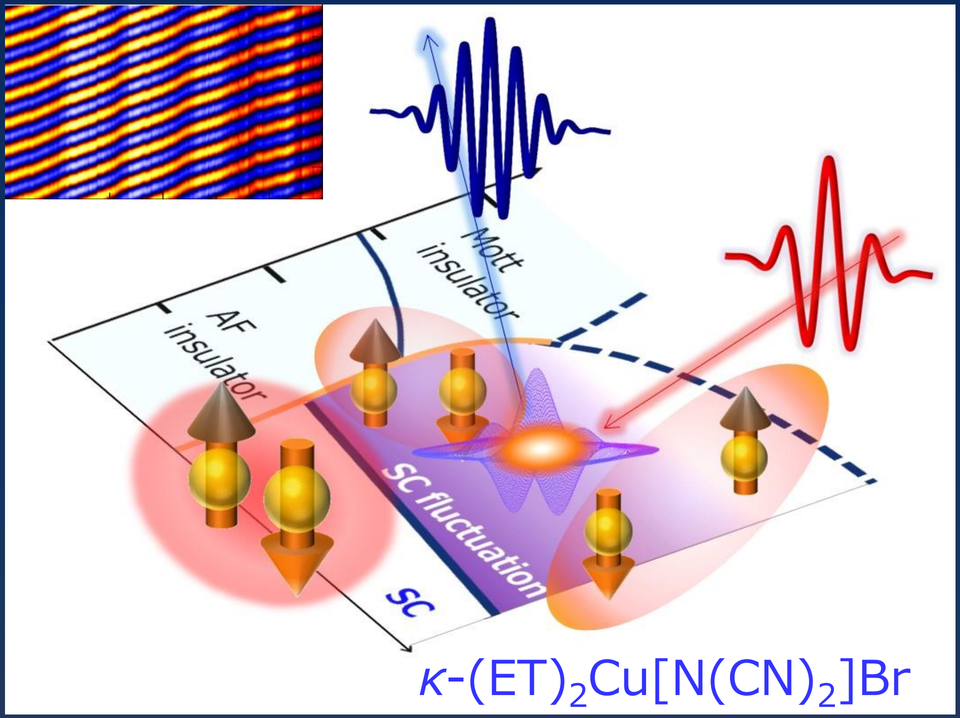
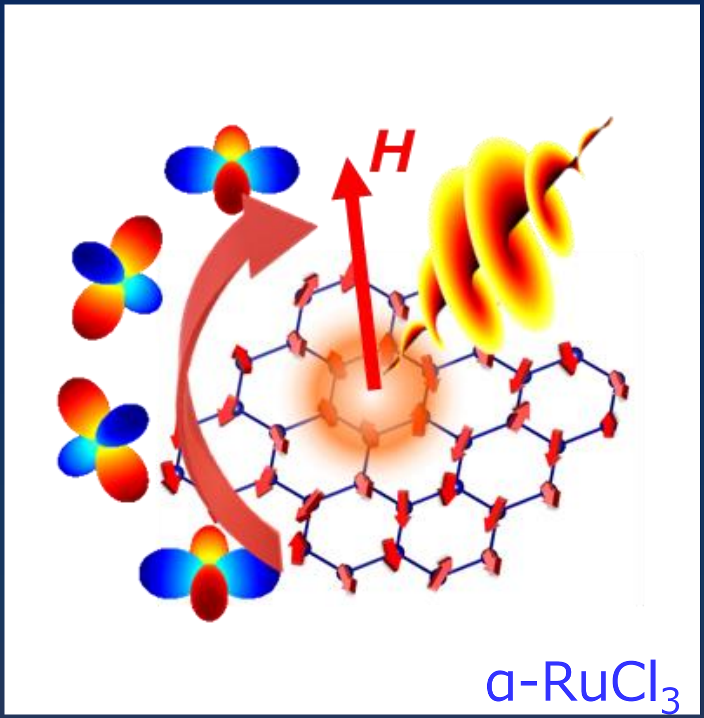
Ultrafast breaking of spatial/time reversal symmetry by a single-cycle light-field in strongly correlated chargers/spins

Tohoku University

Shinichiro Iwai



Nat. Commun.10, 1038 (2020)



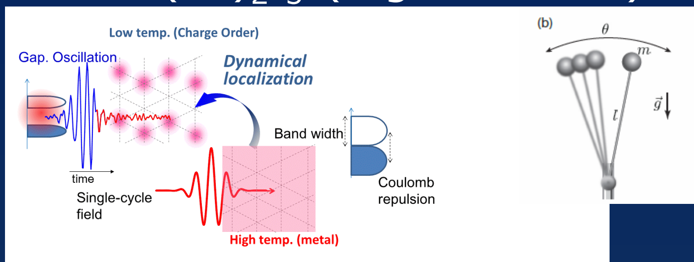
Phys. Rev. Res. 4, L032032 (2022)

Supported by ***JST-CREST*** **JP19198318**
Q-leap **JPMXS0118067426**

New pathway toward ultrafast control of correlated electrons

✓ Coherent modulation of electronic states (within scattering time window)

α-$(ET)_2I_3$ (organic metal)

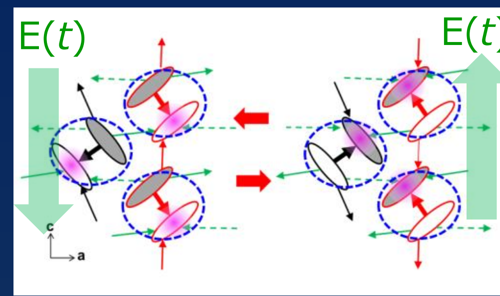


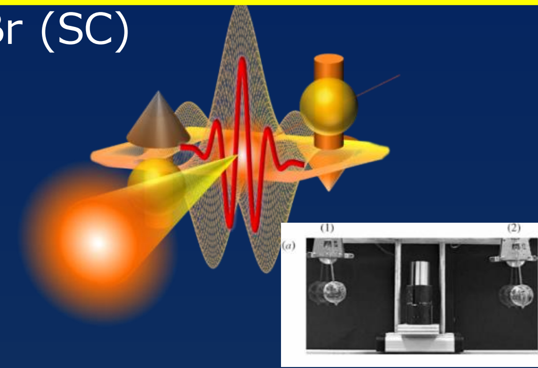
Dynamical localization

Nature commun. 5, 5528(2014)

PRB 2016, *PRB*(R)2017

κ-$(ET)_2Cu[N(CN)_2]Br$ (SC)



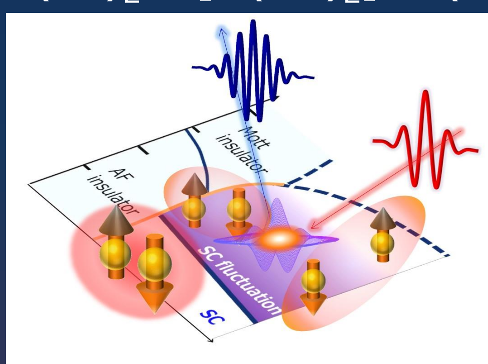


Charge Synchronization

Nature Photon.12, 474 (2018)

✓ Spatial/ time reversal symmetry breaking

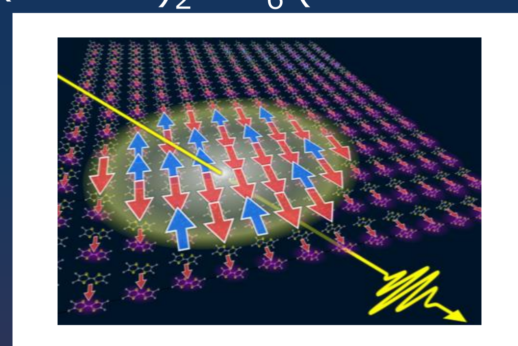
κ-$(ET)_2Cu[N(CN)_2]Br$ (SC)



Nonlinear current in organics SC

Nat. Commun.10, 1038 (2020)

$(TMTTF)_2AsF_6$ (Ferroelectric)

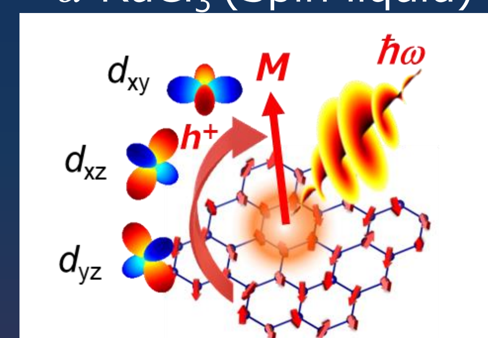


Enhance of correlation

Phys. Rev. Res.

3, L032043(2021)

α-$RuCl_3$ (Spin liquid)



Ultrafast magnetization

Phys. Rev. Res.

4, L032032 (2022)

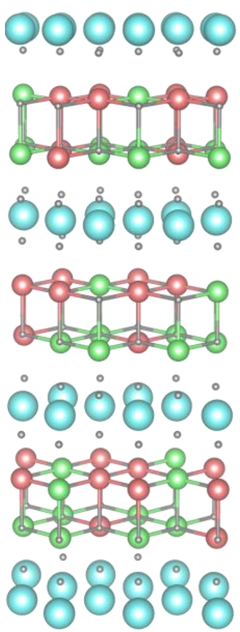
Ultrafast & efficient control of ferroelectrics

$LuFe_2O_4$

with Yu、Okimoto, Koshihara (TITec)
Fujiwara、Ikeda (Okayama)



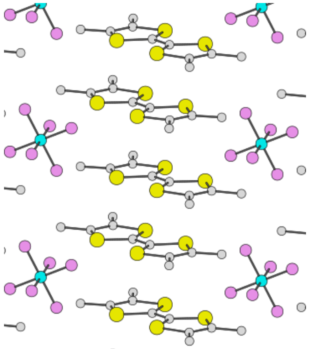
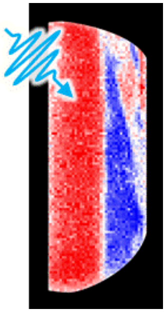




Polarization control by THz field

$(TMTTF)_2BF_4$

with M. Dressel
(Stuttgart)

Outline

i) Introduction

- ✓ Optical responses of strongly correlated system
- ✓ *Time/energy scales of strongly correlated system*
- ✓ Coherent charge motion in correlated syatem
  (Coherent chare motion, Dynamical localization)
- ✓ *6-fs NIR pulse, CEP control/detection*

*PRL*2010
Nat. commun. 2014
PRB 2016
PRB 2017(R)
J. Phys. B 2018
(review)

ii) Stimulated emission in organic SC κ-ET salt

- ✓ Ultrafast stimulated emission (SE) driven by strong field
- ✓ Quantum mechanical analysis (charge synchronization)
- ✓ Temperature dependence (anomaly around T_{SC})

Nat. Photon 2018

iii) SHG in κ-ET salt

- ✓ SHG induced by Petahertz no-scattering current (CEP sensitive)
- ✓ Enhancement of SHG near SC fluctuation
- ✓ Quantum mechanical analysis of unconventional SHG

Nat. Commun. 2020

Outline (continued)

iv) Ultrafast magnetization in Kitaev spin-liquid α-$RuCl_3$

- ✓ Kitaev spin-liquid candidate α-$RuCl_3$
- ✓ Ultrafast mqgnetization (larger for $T>T_N$)
- ✓ Resonant effect to spin-orbit excitons
- ✓ Coherent carrier dynamics & theory

Phys. Rev. Res. 4, L032032 (2022)
arXiv: 2207.03877

v) Summary & Future problems

- ✓ High-T_c Cuprates, Correlated Dirac semimetal (Iridates)···

Collaborators

Ultrafast & THz spectroscopy

Tohoku U. H. Itoh, Y. Kawakami
T. Amano, Y. Akamine, H. Ohashi


Itoh


Kawakami


Amano


Ohashi

Samples and basic measurements

(organic)

Okayama Univ. Sci. K. Yamamoto
IMR T. Sasaki
Nagoya Univ. Y. Nakamura, H. Kishida
IMS H. Yamamoto
Univ. Stuttgurt M. Dressel


T. Sasaki


H. Kishida


H. Yamamoto


M. Dressel

(High-T_c superconducting cuprates)

Kyusyu sangyo University T. Nishizaki
Tohoku University K. Ohgushi


T. Nishizaki


T. Ogushi

Theory

Chuo Univ. N. Arakawa, K. Yonemitsu
TITEC Y. Murakami
Tohoku. Univ. S. Ishihara


K. Yonemitsu


S. Ishihara

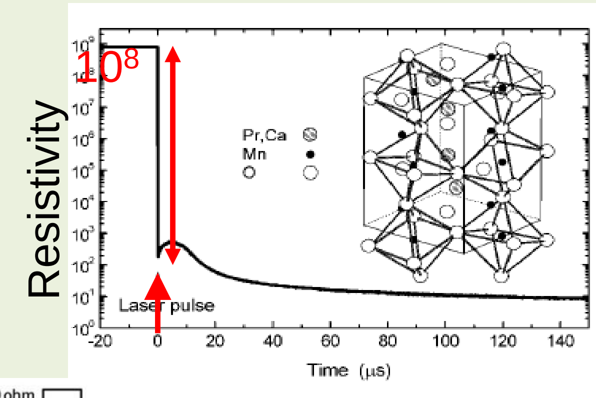
Supported by
JST-CREST JP19198318
Q-leap JPMXS0118067426

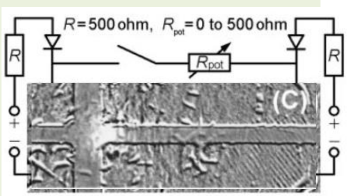
Photoinduced Insulator –Metal transition

25 years ago

Miyano, Tokura *et al*. PRL1997.

(Mn oxides)



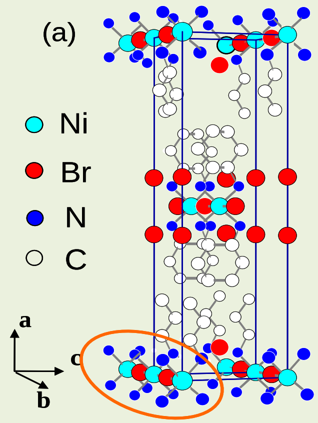


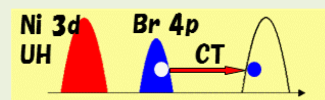
Fiebig *et al*. Science 280, 1925, 1998

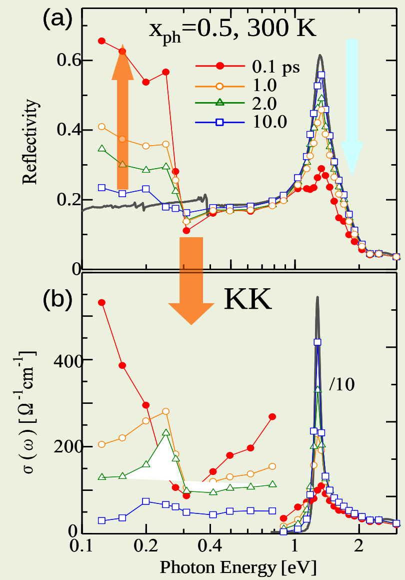
Melting of Charge/Orbital order PM→FM

Iwai, Okamoto, Tokura *et al*. PRL 2003

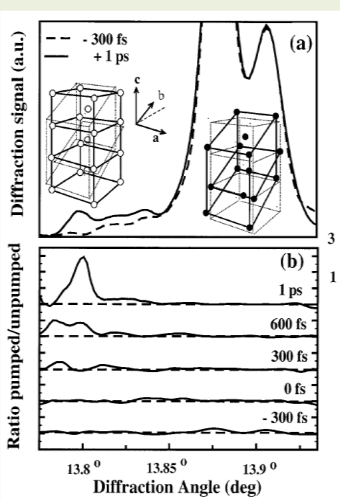
(Ni complex)







Photoinduced Mott transition



Cavalleri et al. PRL 2001

Structural dynamics (XRD)

(VO_2)

Gonokami, Koshihara eds. *JPSJ* 2006

Yonemitsu, Nasu *Phys. Rep*. 2008

Bosov, Averitt, Dressel et al., *RMP* 2011

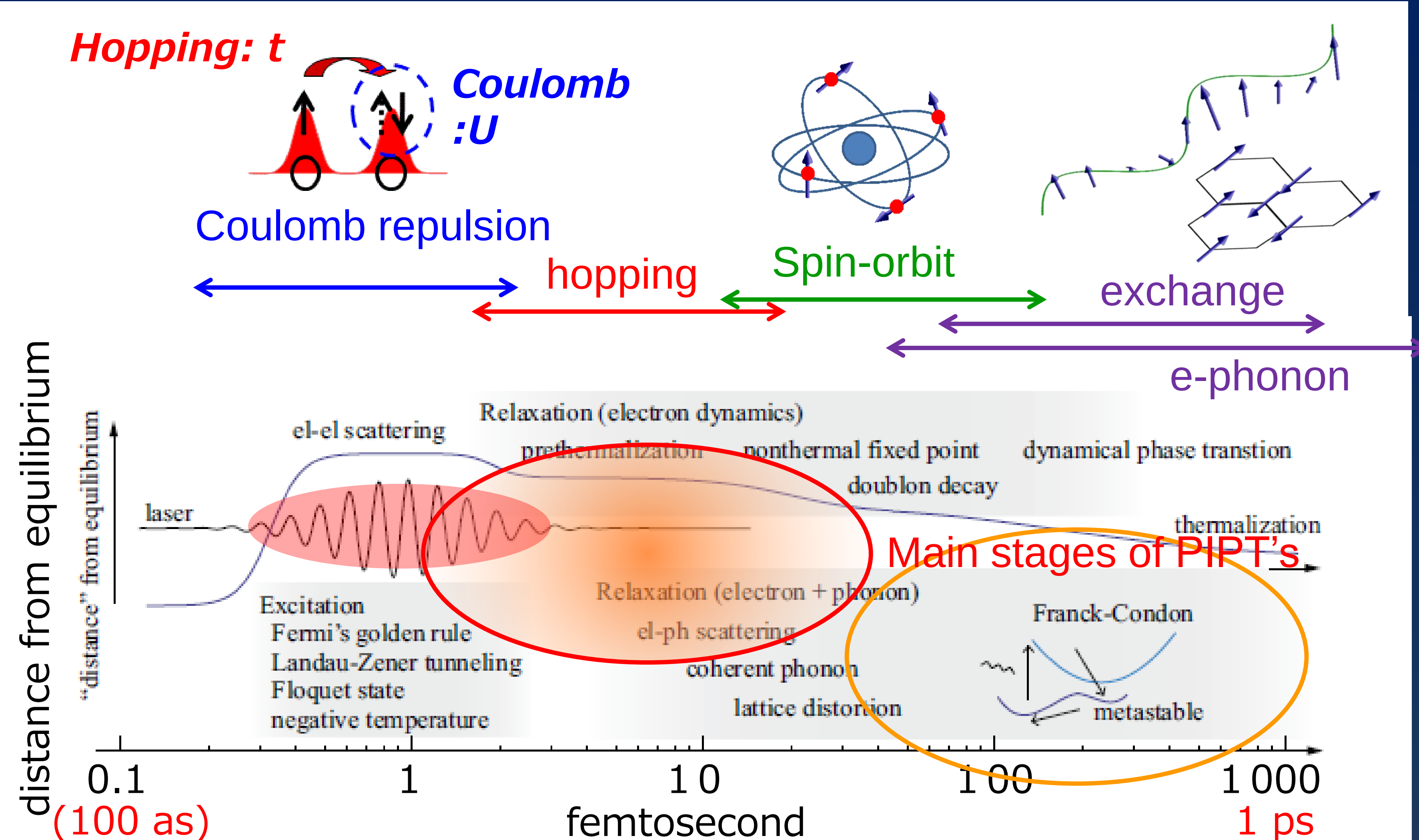
Mihailovic et al. , *Advances in Physics*, 2016

Cavalleri et al. *Adv. in Opt. Photon.* 2016

Bosov, Averitt, Hsieh, *Nat. Matter* 2017

Koshihara et al. *Phys. Rep*. 2021

Non equilibrium in correlated system

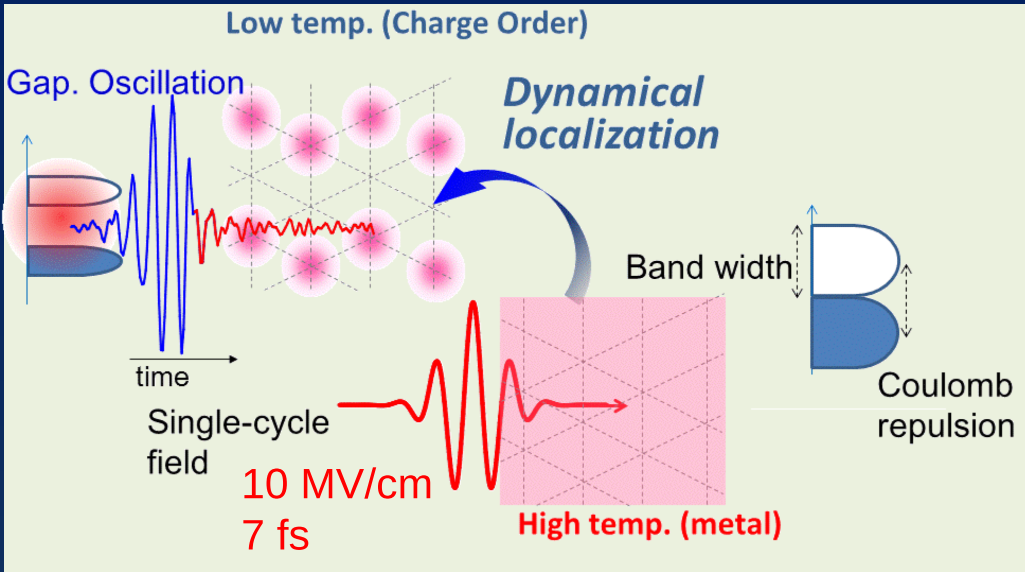


Aoki, Tsuji, Eckstein, Oka, Werner *Rev. Mod. Phys*. 86, 779 (2014).

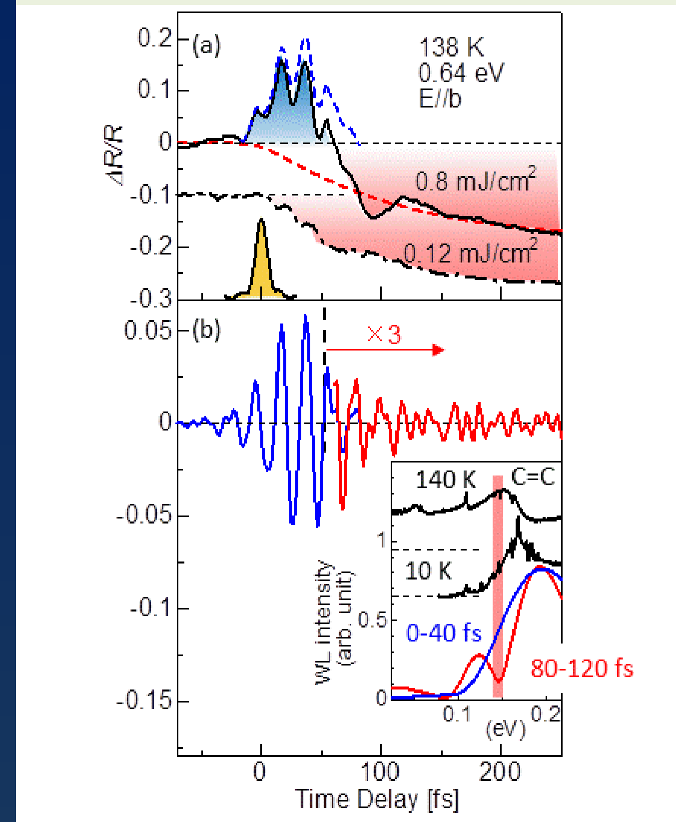
✓ Limited degrees of freedom can work

✓ Electronic coherence survives (depends on material)

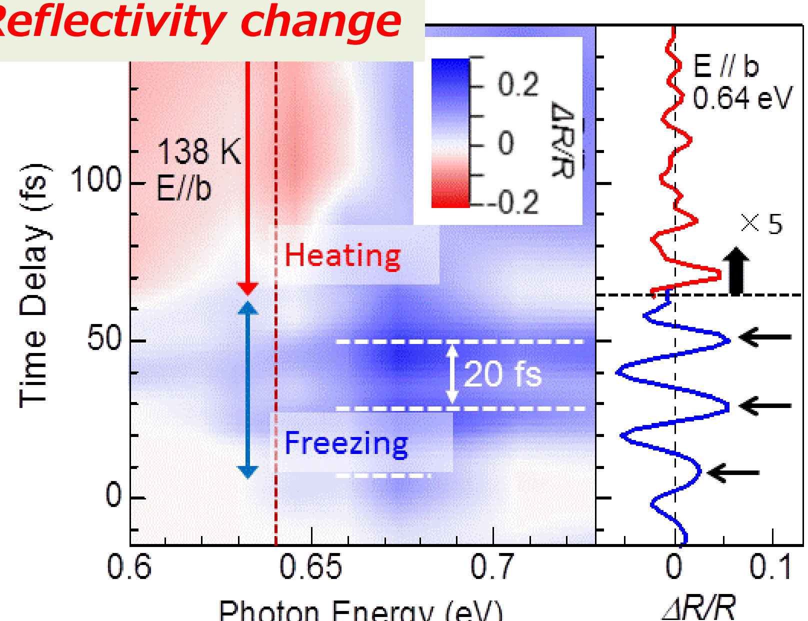
Optical freezing of charge motion



Insulating gap in time axis



Reflectivity change



Ishikawa, Iwai *et al., Nat. commun*. 2014

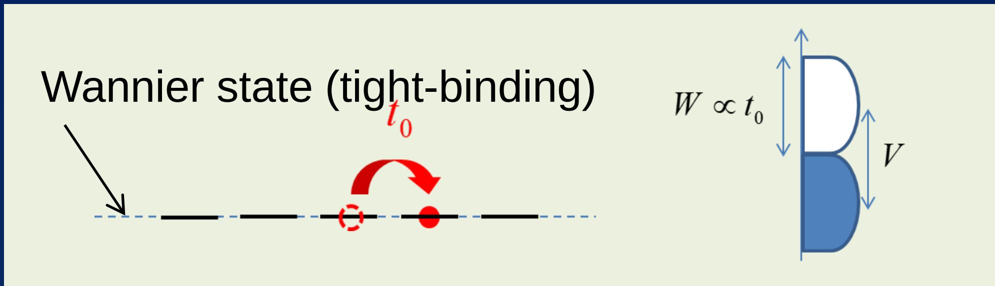
Nature commun. 5, 5528(2014)

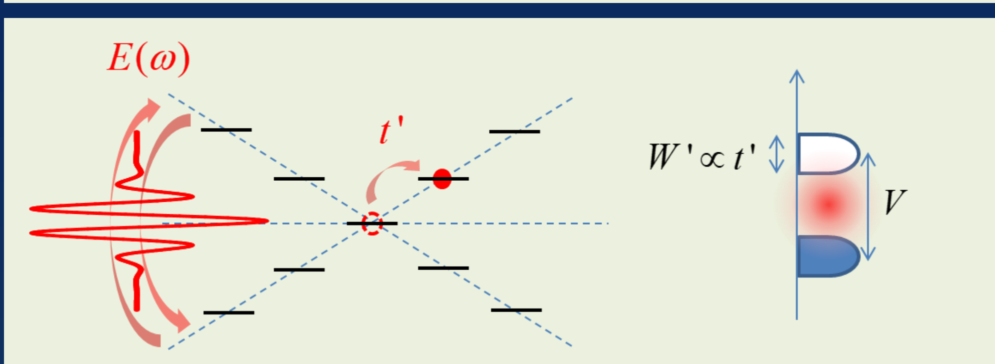
PRB 95, 201106(R) (2017)

PRB 93, 165126 (2016)

J. Phys. B51, 174005(2018) (Review)

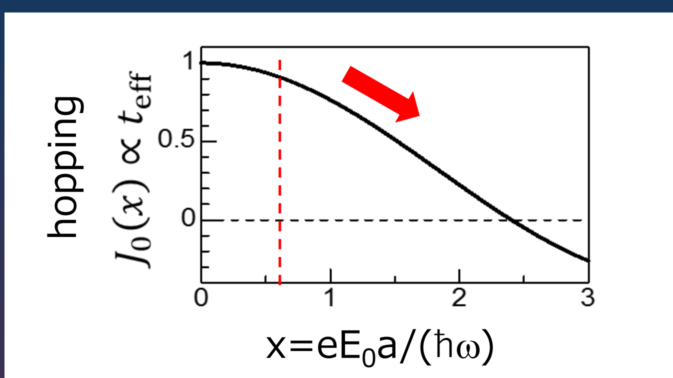
Dynamical localization

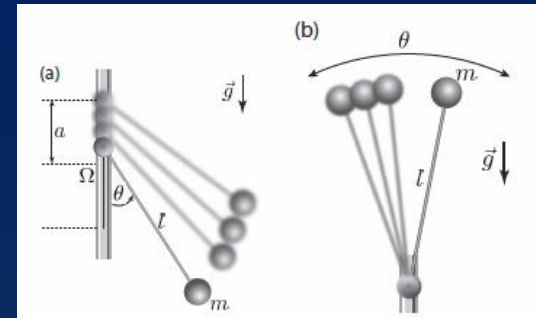




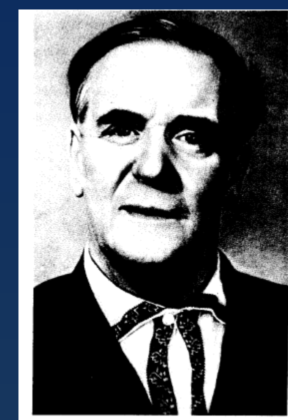
High-frequency strong field
disturbs hopping (ω=0；stark ladders)

$$H_1 = -eE(t)a\sum_i m\left(|m\rangle\langle m|\right) + t_0 \sum_{i,j}\left(|i\rangle\langle j| - |j\rangle\langle i|\right)$$





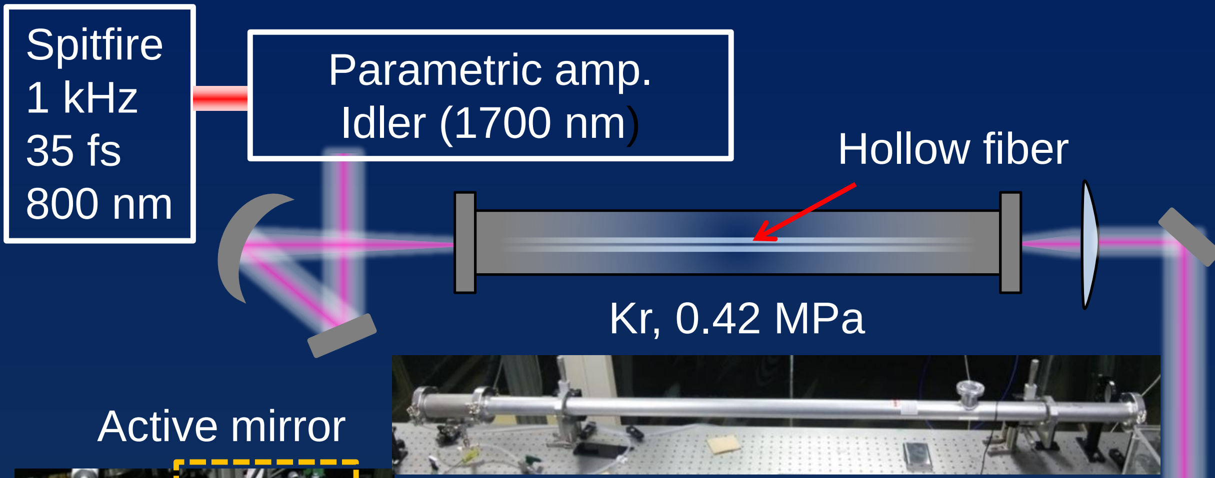
Dynamical stabilization
M. Bukovet al.,
Advances in Physics, **64**, 139(20

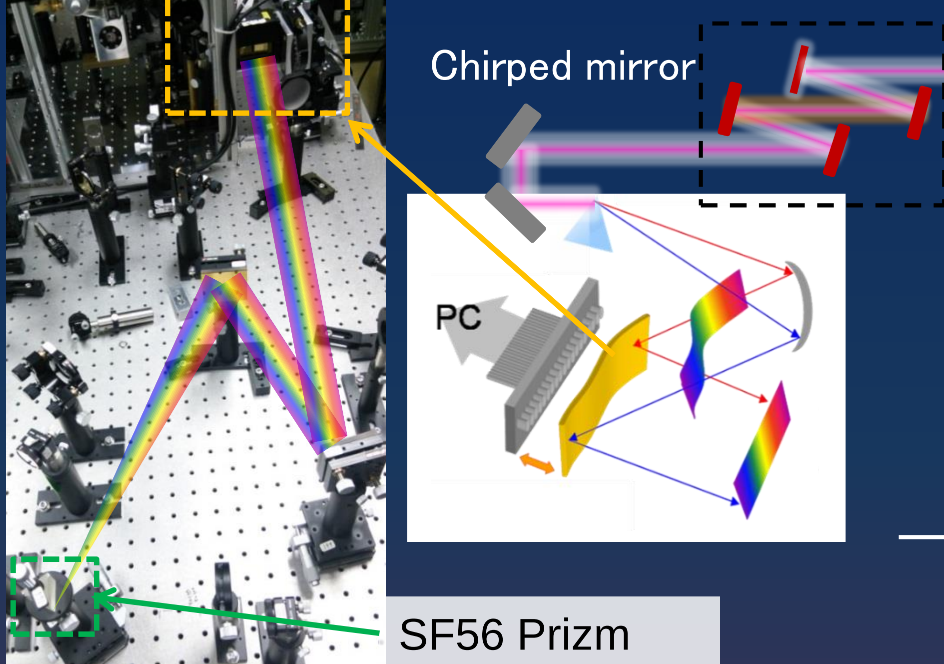

Petr Leonidovich Kapitza

P. L. Kapitza, Soviet Phys. JETP, 21, 588(1951)

Dunlap, Kenkre, PRB(1986)
Grossmann,Hanggi, PRL(1991)
Kayanuma, Saito, PRA(2008)

6 fs NIR pulse (1.3 cycle, CEP stabilized)





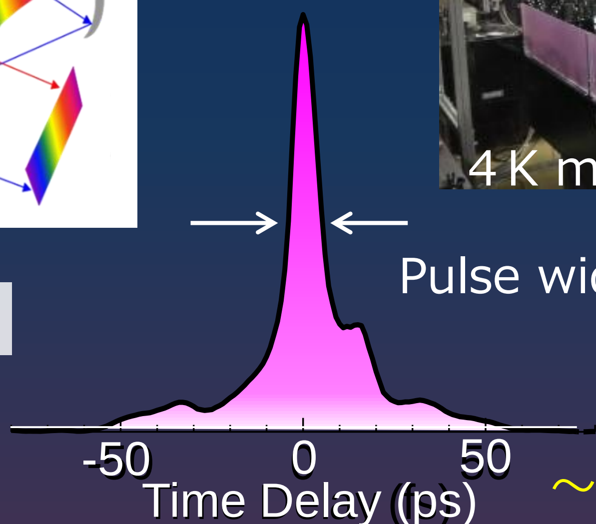
✔ **OPA Idler ($\chi^{(2)}$)**

$$\varphi_{Idler} = -\frac{\pi}{2} + \varphi_{Pump} - \varphi_{Signal}$$

✔ **SPM($\chi^{(3)}$)**

$$\varphi_{SPM} = \frac{\pi}{2} + \varphi_1 + \varphi_2 - \varphi_3$$

→ CEP stable



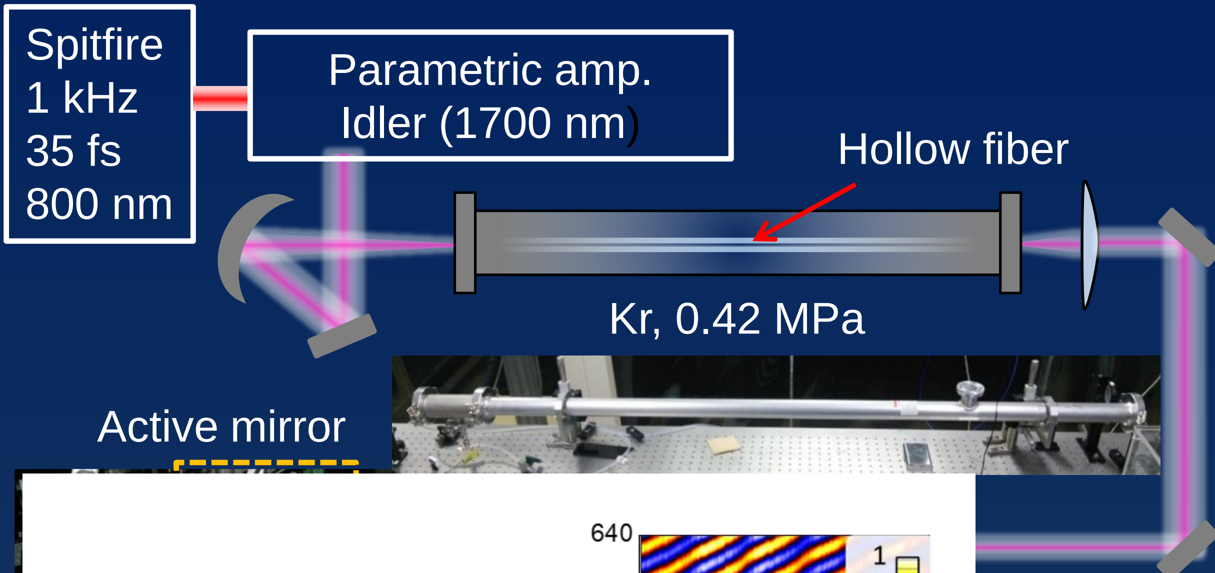
Pulse width =6 fs

30 μJ/pulse
Φ200 μm

Max >100 MV/cm !
~20 MV/cm for organics

J. Phys. B51, 174005(2018)
(Review)

6 fs NIR pulse (1.3 cycle, CEP stabilized)



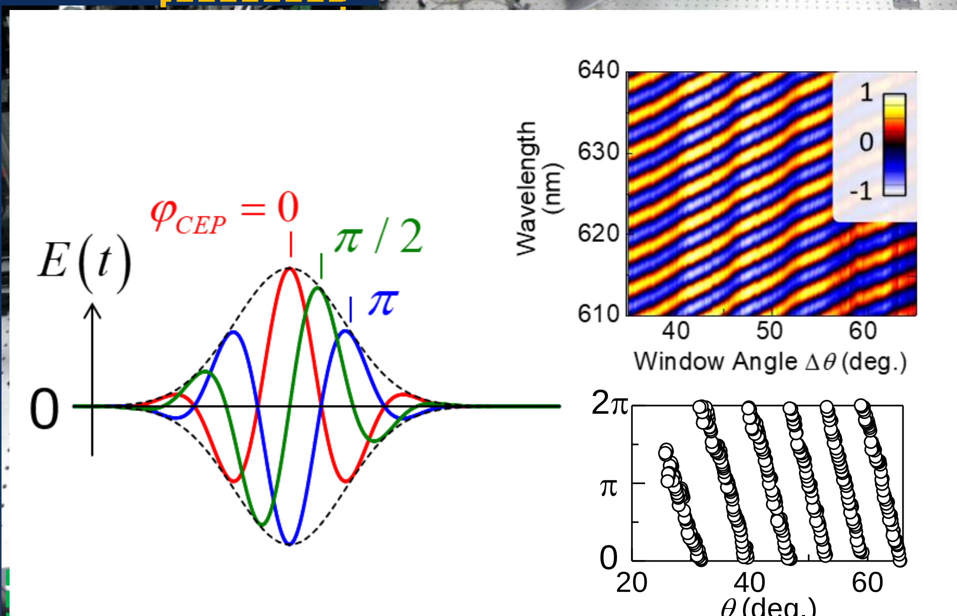
✔ OPA Idler ($\chi^{(2)}$)

$$\varphi_{Idler} = -\frac{\pi}{2} + \varphi_{Pump} - \varphi_{Signal}$$

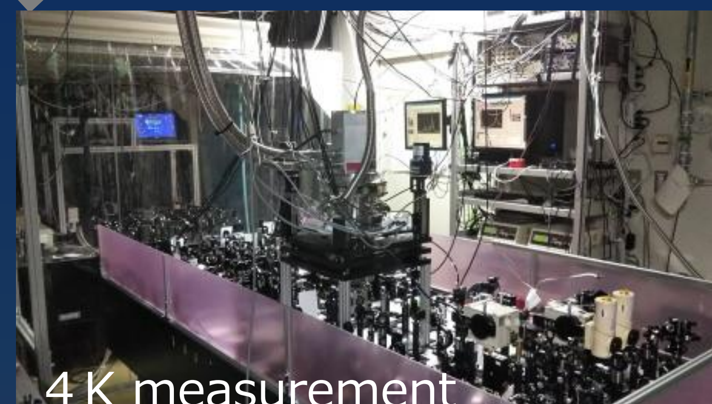
✔ SPM($\chi^{(3)}$)

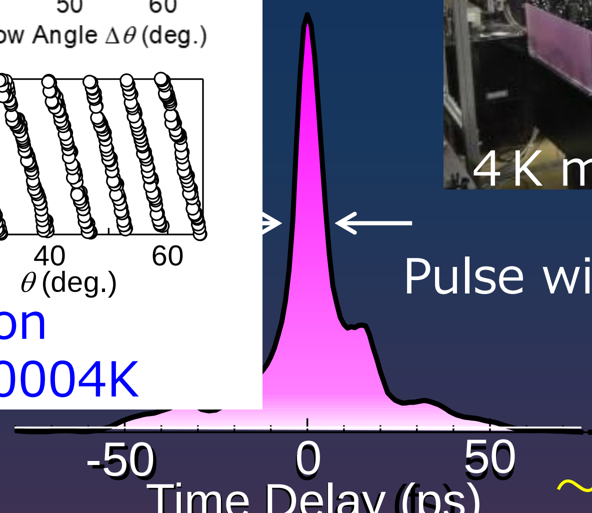
$$\varphi_{SPM} = \frac{\pi}{2} + \varphi_1 + \varphi_2 - \varphi_3$$

→ CEP stable



Iwai et al. , Faraday Discussion 2022 doi.org/10.1039/D2FD0004K





Pulse width =6 fs

30 μJ/pulse
Φ200 μm

Max >100 MV/cm !
～20 MV/cm for organics

Outline

i) Introduction

- ✓ Optical responses of strongly correlated system
- ✓ *Time/energy scales of strongly correlated system*
- ✓ Coherent charge motion in correlated syatem
  (Coherent chare motion, Dynamical localization)
- ✓ *6-fs NIR pulse, CEP control/detection*

*PRL*2010
Nat. commun. 2014
PRB 2016
PRB 2017(R)
J. Phys. B 2018
(review)

ii) Stimulated emission in organic SC κ-ET salt

- ✓ Ultrafast stimulated emission (SE) driven by strong field
- ✓ Quantum mechanical analysis (charge synchronization)
- ✓ Temperature dependence (anomaly around T_{SC})

Nat. Photon 2018

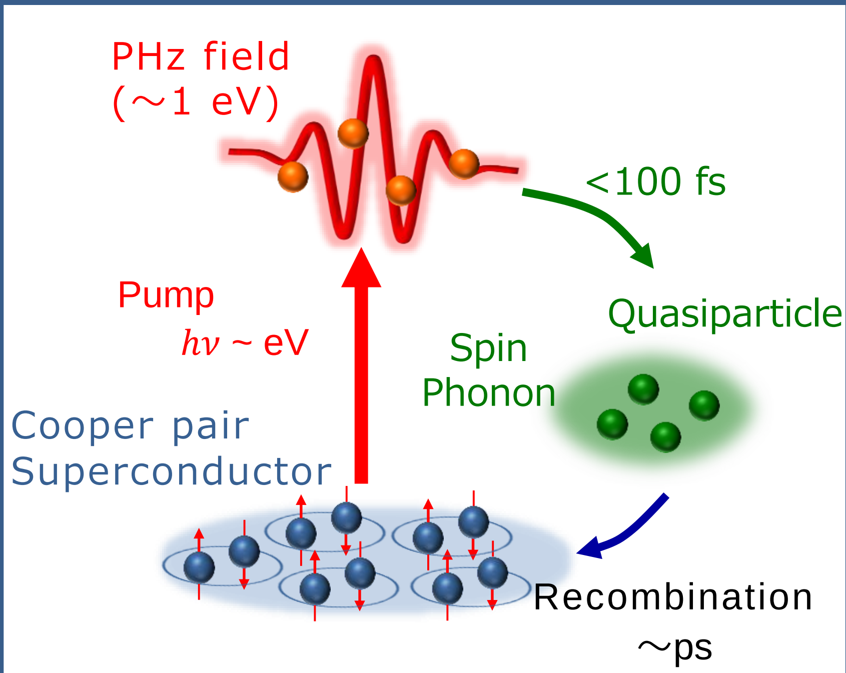
iii) SHG in κ-ET salt

- ✓ SHG induced by Petahertz no-scattering current (CEP sensitive)
- ✓ Enhancement of SHG near SC fluctuation
- ✓ Quantum mechanical analysis of unconventional SHG

Nat. Commun. 2020

Excitation of superconductors

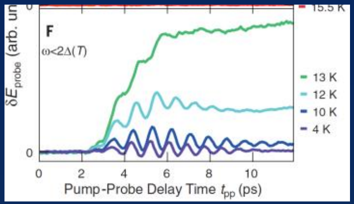
Excitation by light (NIR-Visible)
→ rise of electron temp. (scattering)

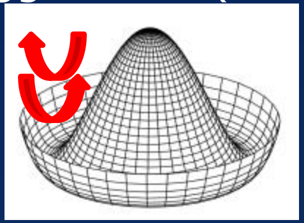


Giannetti, Mihailovic et al., Advances in Physics, **65**, 58-238(2016).

✓ Avoid increasing temp
→low energy excitations

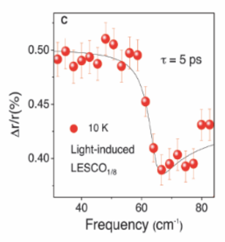
- Coherent exc. of Higgs mode (BCS)

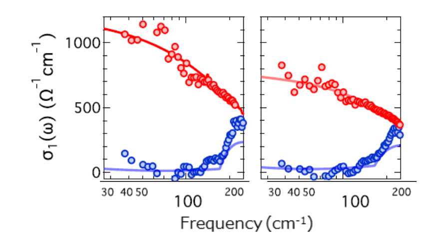




Matsunaga, Shimano et al. Science 2014.

- Photoinduced SC Cuprates, Organics





D. Faustii et al., Science 2011

M. Buzzi et al., PRX 10, 031028 (2020)

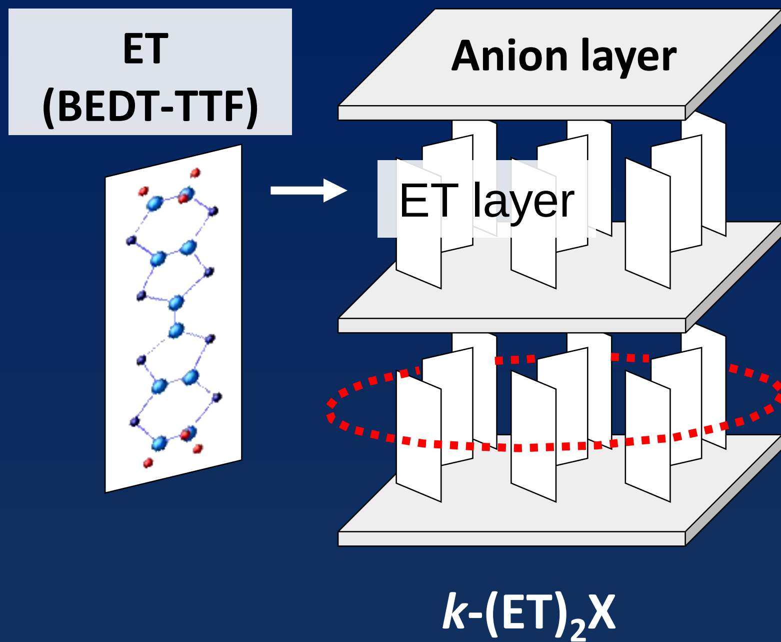
✓ Another approach

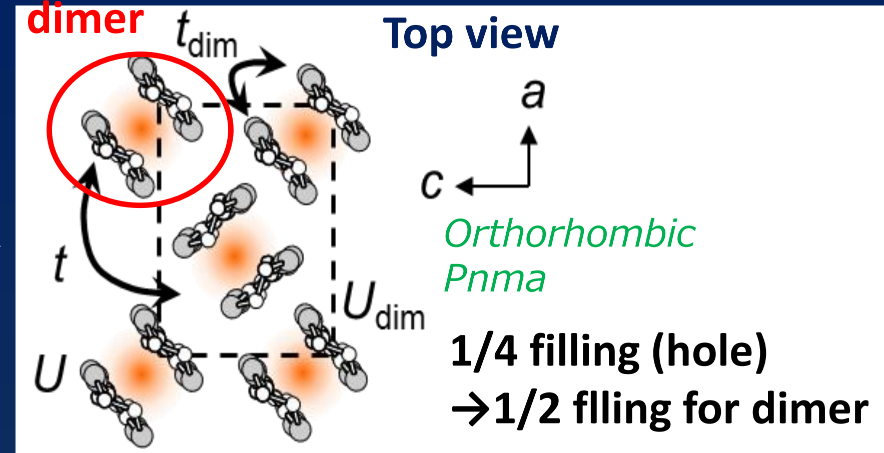
e-e scattering time

- **～40 fs h/(0.1 eV) organic SC**
- ～ 4 fs or shorter ? High-T_c Cuprates

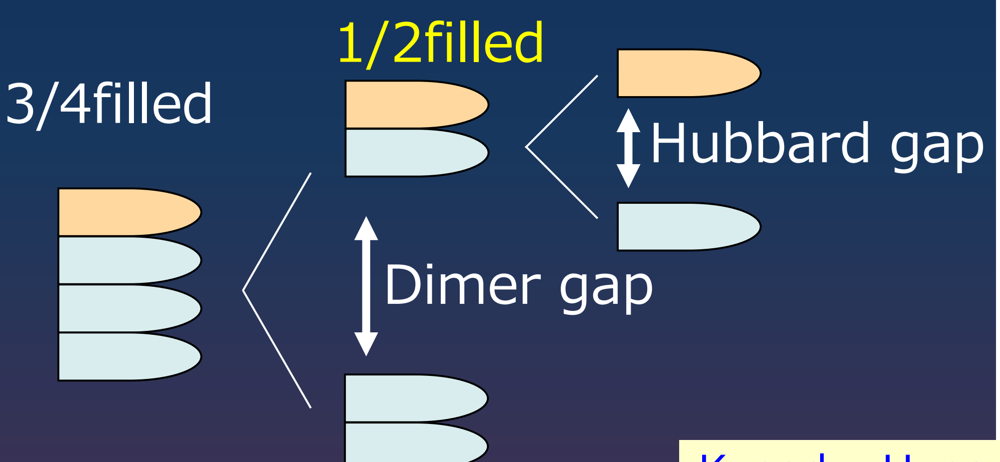
- **6 fs pulse can control electrons in no-scattering time window ?**

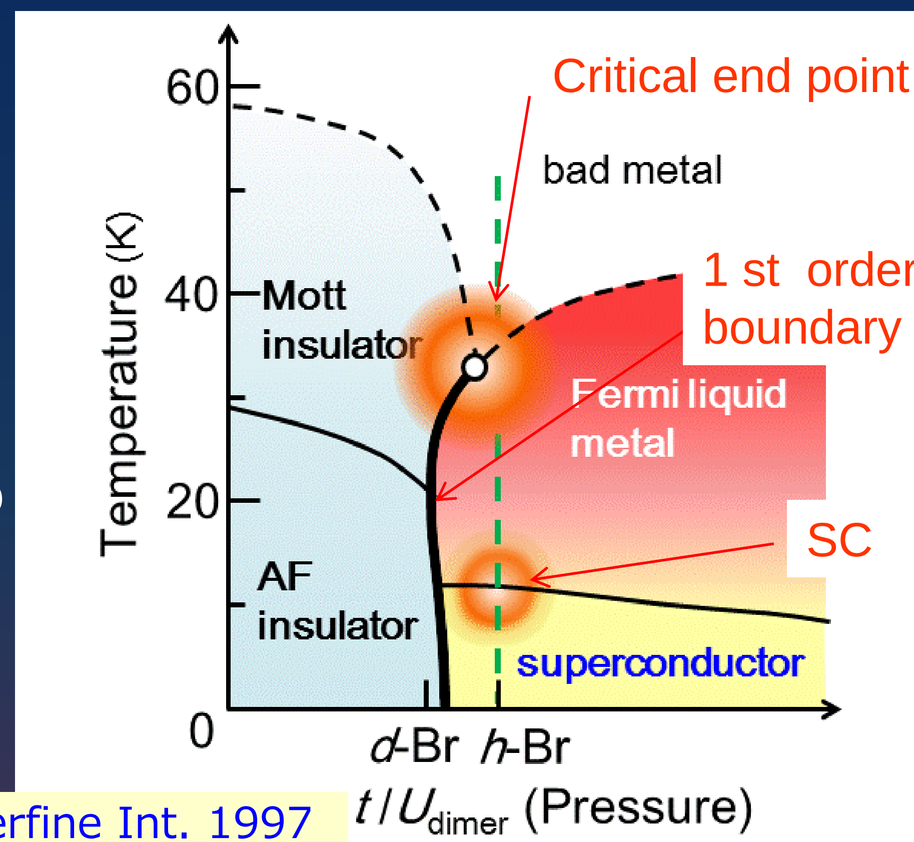
Organic superconductor κ-$(ET)_2Cu[N(CN)_2]Br$





Dimer Mott insulator



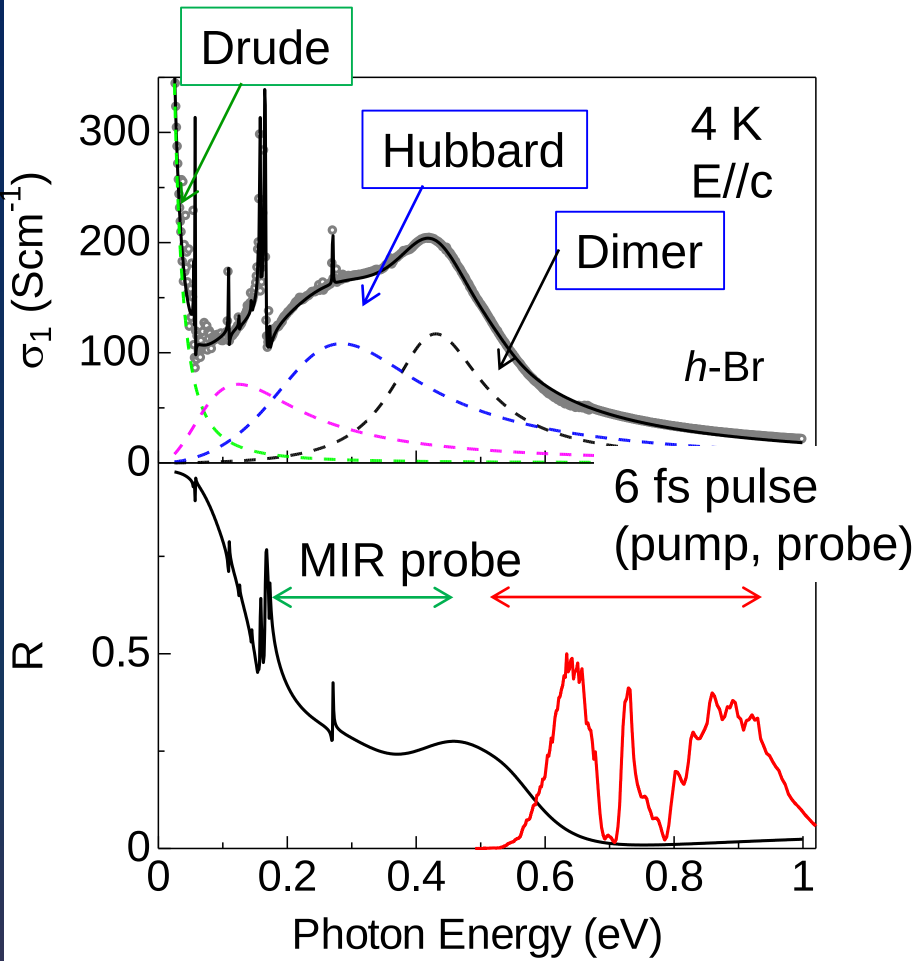


Kanoda, Hyperfine Int. 1997

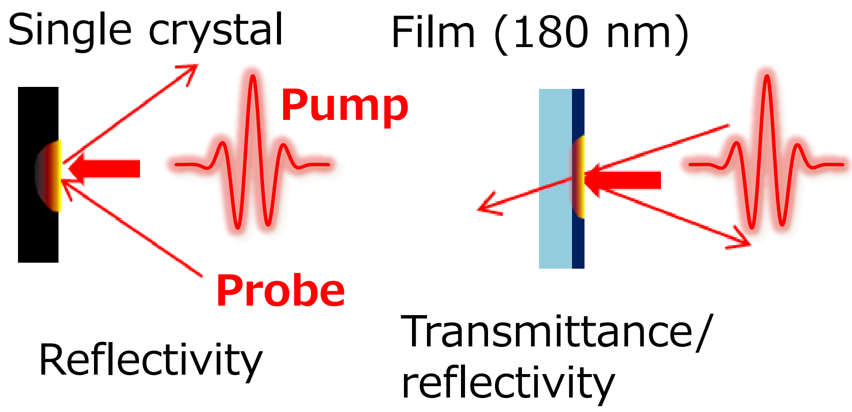
κ-(*h*-ET)$_2$Cu[N(CN)$_2$]Br (*h*-Br)

Reflectivity/Optical conductivity (*h*-Br)

c.f. Faltermeier, Dressel *et al.*, PRB 76, 165113 (2007).

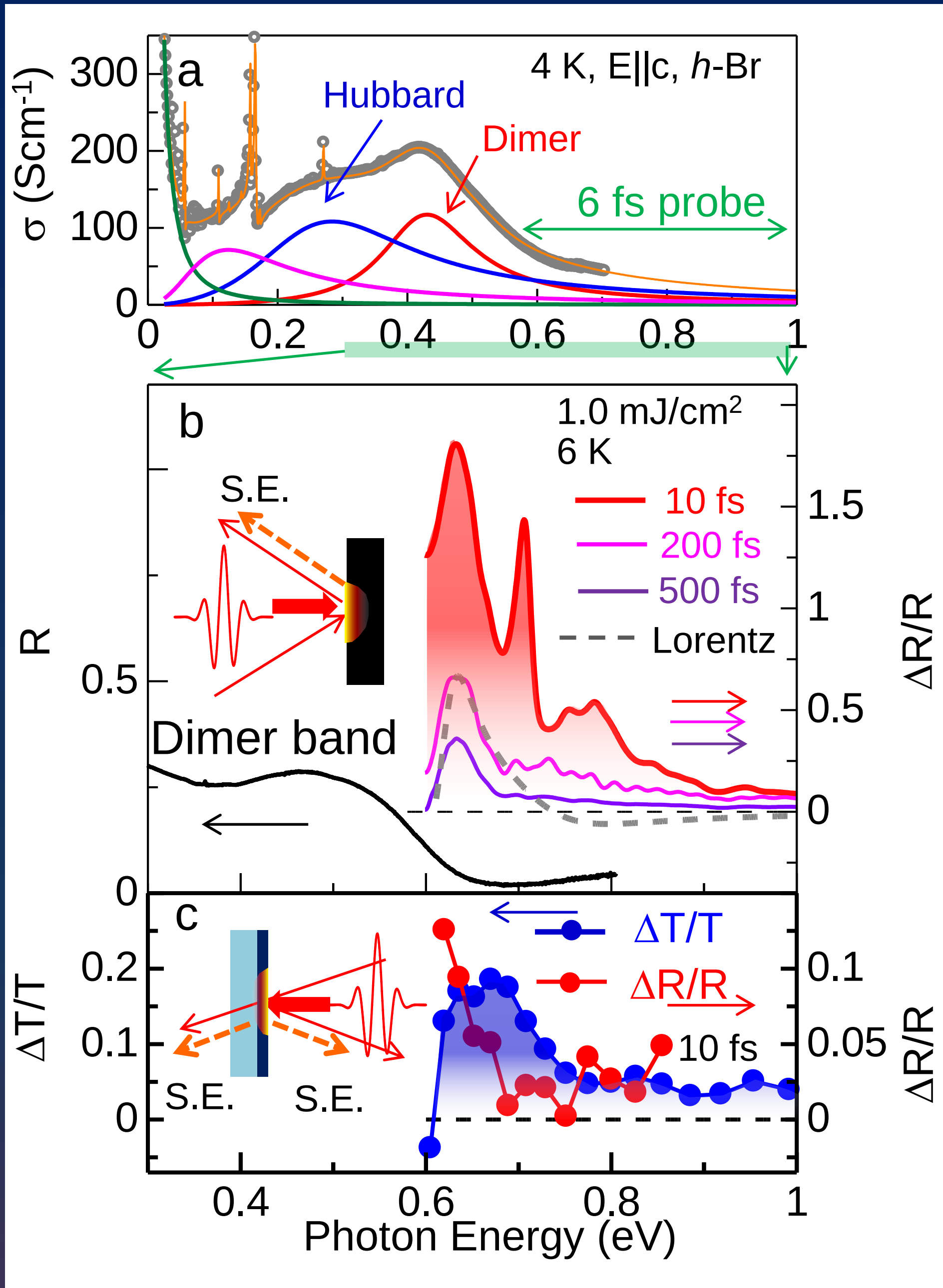


Pump-probe(2-or 3- beam)



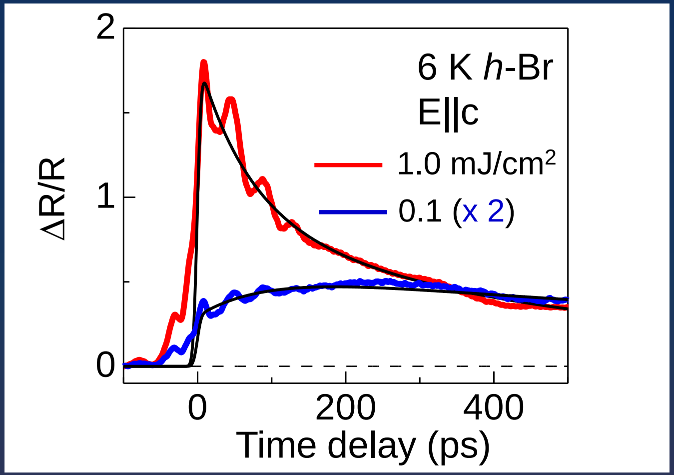
Pump/Probe～ 1000/1

Tr. reflectivity(ΔR/R) & transmittance(ΔT/T)



- ✓ Same-color pump & probe
- ✓ New reflectivity peak
  0.63 eV, at 10 fs (FID at 0.7 eV)
  ~200 % reflectivity increase !
- ✓ Lorentz analysis

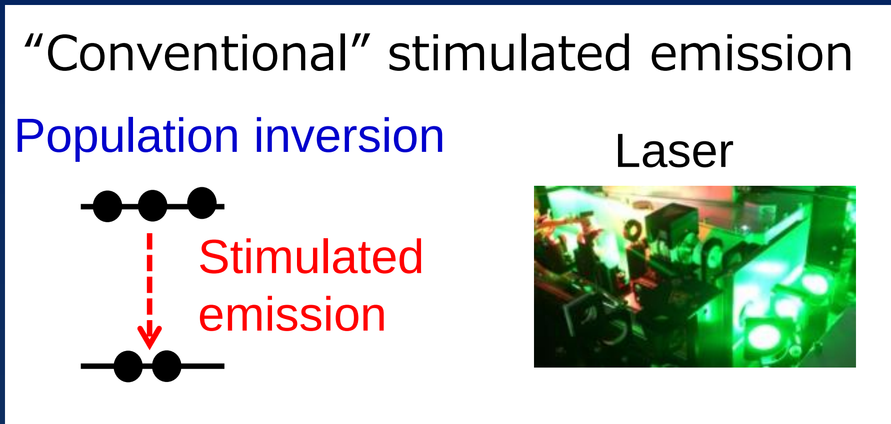
  Additional oscillator
  frequency: 0.61 eV
  damping: 0.04 eV

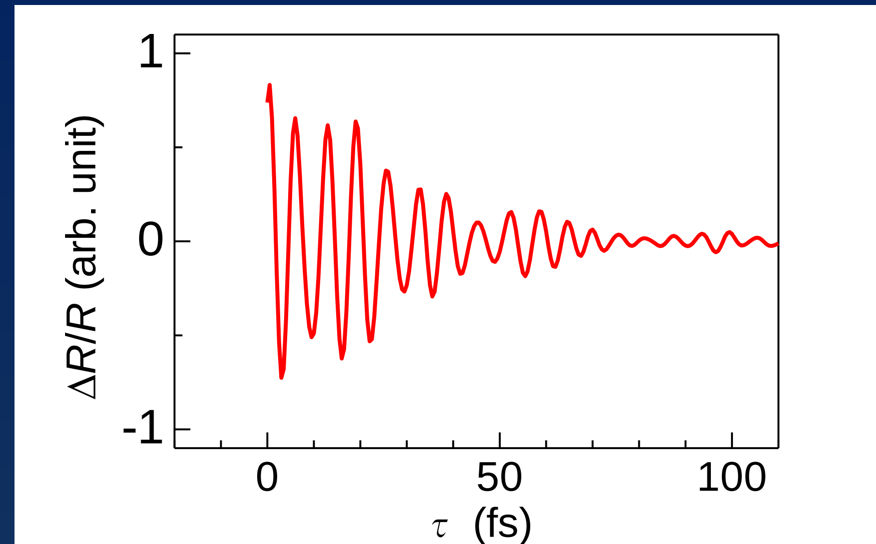


Fast (65%) rise <10 fs, decay 70 fs

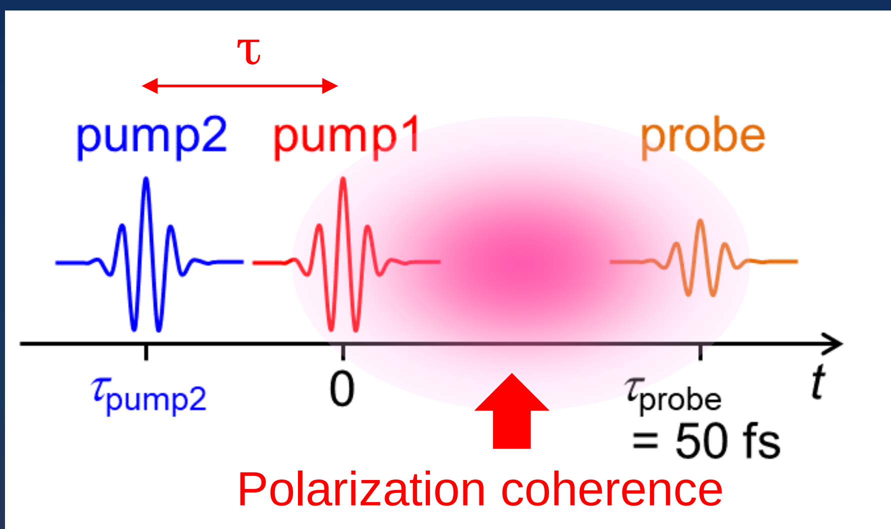
Slow(35%) rise 90 fs, decay 360 fs, 4.2 ps, > 100 ps

Double –pump & probe
to detect coherence of the nonlinear charge osc.





Difference between pump1 and pump2



Supplementary
Nat. Commun. 10, 1038 (2020)

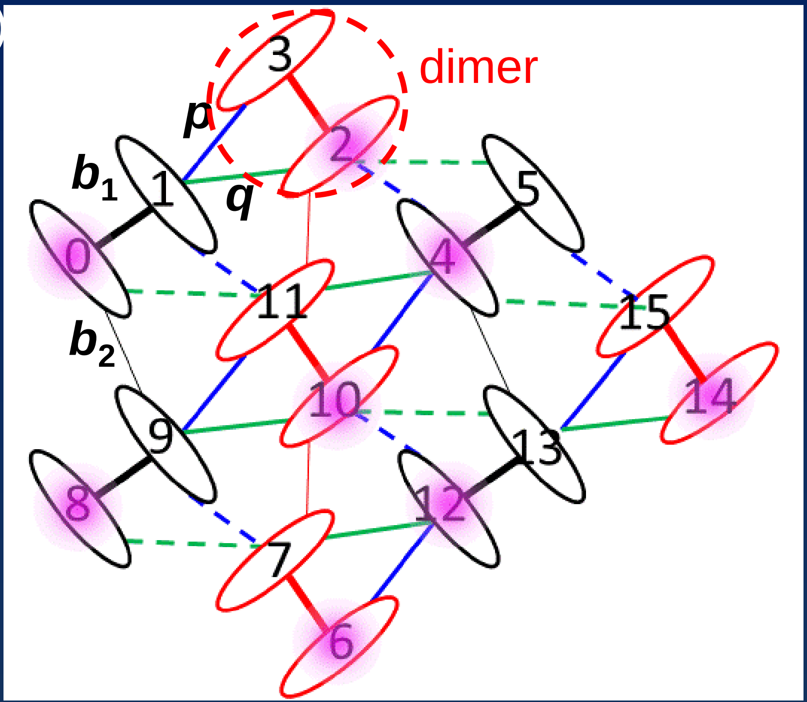
✓ Decay time of SE (70 fs) is approximately equal to the electronic coherence time

Matsubara, Itatani, Yonemitsu, Koshihara, Onda et al., PRB89, 161102(R)(2014)
Kimata, Kayanuma, Nakamura et al., PRB101, 174301(2020)

Origin of the SE is coherent charge motion ?

Time dependent Schröedinger eq. (Yonemitsu)

2D ext. Hubbard model (16-site, ¾ filling)
(Exact diagonalization)



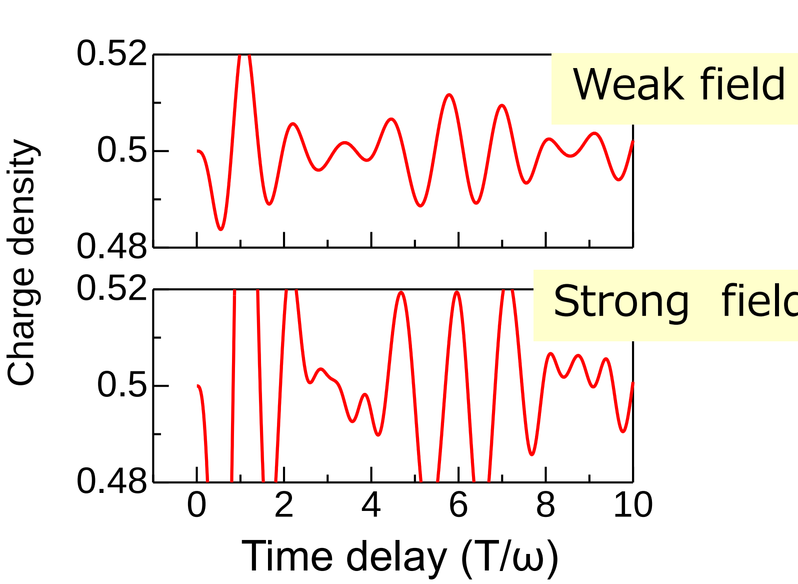
$$H_{2D} = \sum_{\langle i,j \rangle \sigma} t_{ij} \left(c_{i,\sigma}^{+} c_{j,\sigma} + c_{j,\sigma}^{+} c_{i,\sigma} \right) + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \sum_{\langle i,j \rangle} V_{ij} n_i n_j$$

Photoexcitation (Peierls phase)

$$c_{i,\sigma}^{+} c_{j,\sigma} \rightarrow \exp\left[\frac{ie}{\hbar c} \boldsymbol{r}_{ij} \cdot \boldsymbol{A}(t) \right] c_{i,\sigma}^{+} c_{j,\sigma}$$

Single-cycle pulse

$$\boldsymbol{A}(t) = \frac{c\boldsymbol{F}}{\omega} \left[\cos(\omega t) - 1 \right] \theta(t) \theta\left(\frac{2\pi}{\omega} - t \right)$$



Time evolution of charge density on a molecule

Yonemitsu, JPSJ87, 044708(2018).

Synchronization (entrainment)

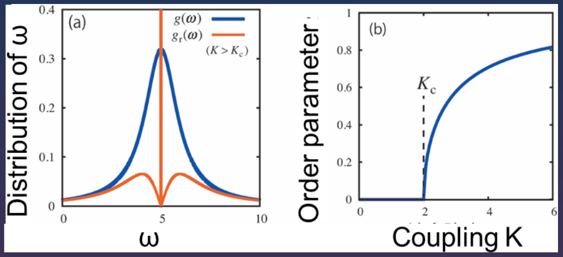
Huygens's clock
(cycloidal pendulum clock)

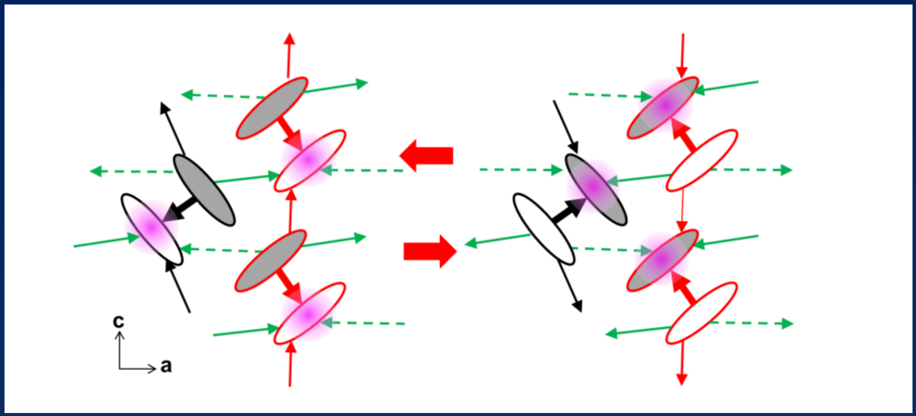
M. Bennet et al.,
Proc. R. Soc. Lond. A 458, 563(2002)

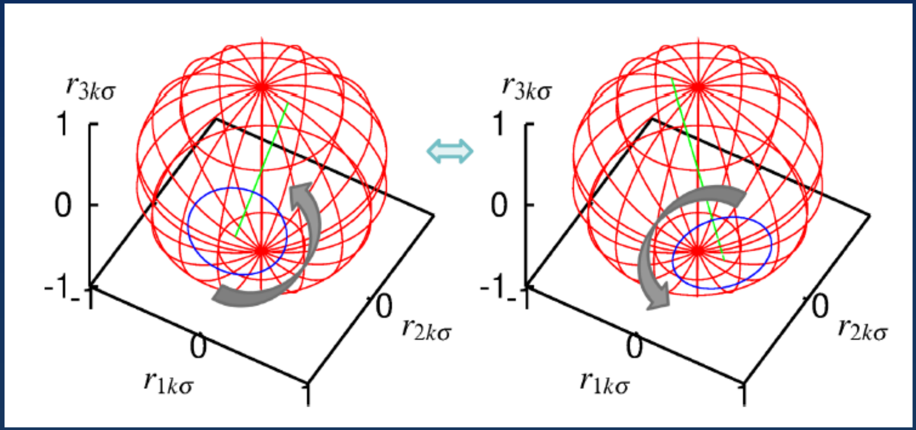
Huygens 1669

Kuramoto model (1975)

$$\frac{d\phi_i}{dt} = \omega_i + \frac{K}{N}\sum_{j=1}^{N} h\left(\phi_i - \phi_j\right)$$







非線形同期振動の一般的理解
✓Bipartite lattice
✓Bloch eq. (pseudospin)
電荷密度の差、電流密度、結合密度
✓Hubbard model, spinless Fermion

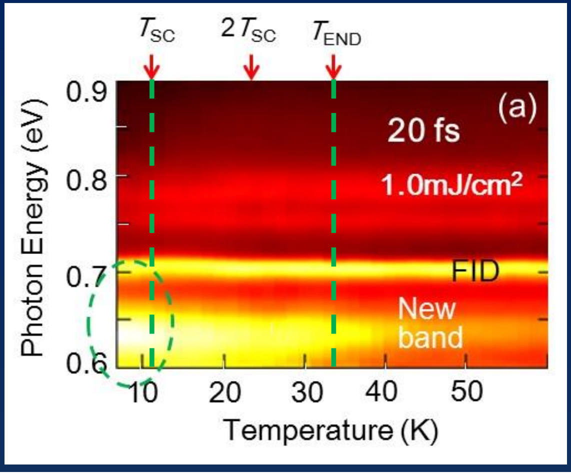
Yonemitsu, Werner JPSJ89, 084701(2021)

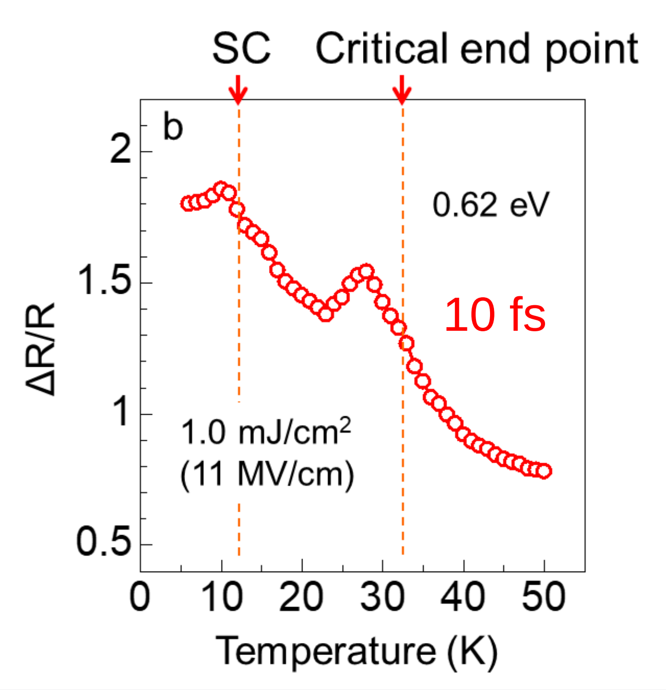
Anomaly at T_{SC}

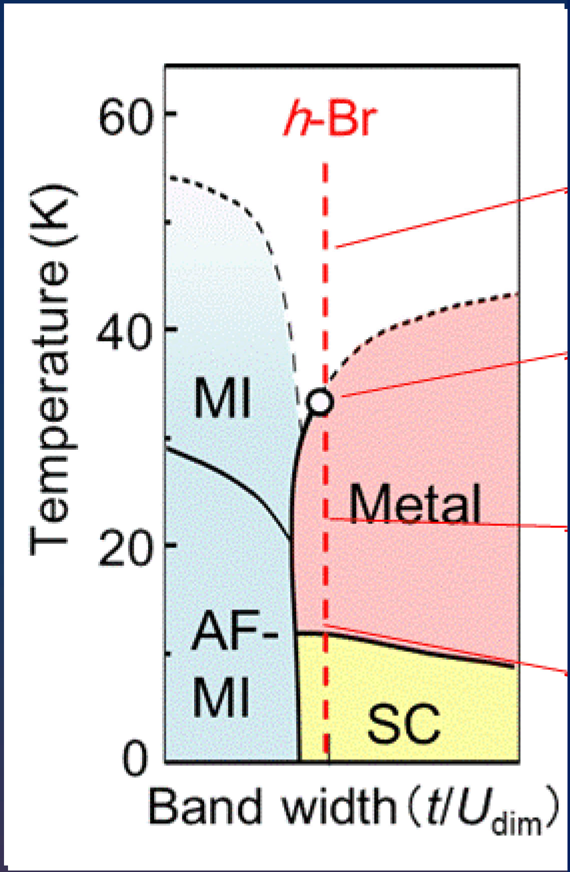
✓ Gradual increase from 20 K ∼2 T_{SC}

✓ Anomaly at 10 K ∼T_{SC} (heating effect ∼3 K)

Nonlinear charge osc. (transient current) amplified by SC fluctuation







10 fs (time const. 5 fs) response (strong field)

High energy (> 0.4 eV) Interaction

⇔ U 0.8 eV

V 0.2∼0.3 eV

t 0.1∼0.3 eV

Outline

i) Introduction

- ✓ Optical responses of strongly correlated system
- ✓ *Time/energy scales of strongly correlated system*
- ✓ Coherent charge motion in correlated syatem
  (Coherent chare motion, Dynamical localization)
- ✓ *6-fs NIR pulse, CEP control/detection*

*PRL*2010
Nat. commun. 2014
PRB 2016
PRB 2017(R)
J. Phys. B 2018
(review)

ii) Stimulated emission in organic SC κ-ET salt

- ✓ Ultrafast stimulated emission (SE) driven by strong field
- ✓ Quantum mechanical analysis (charge synchronization)
- ✓ Temperature dependence (anomaly around T_{SC})

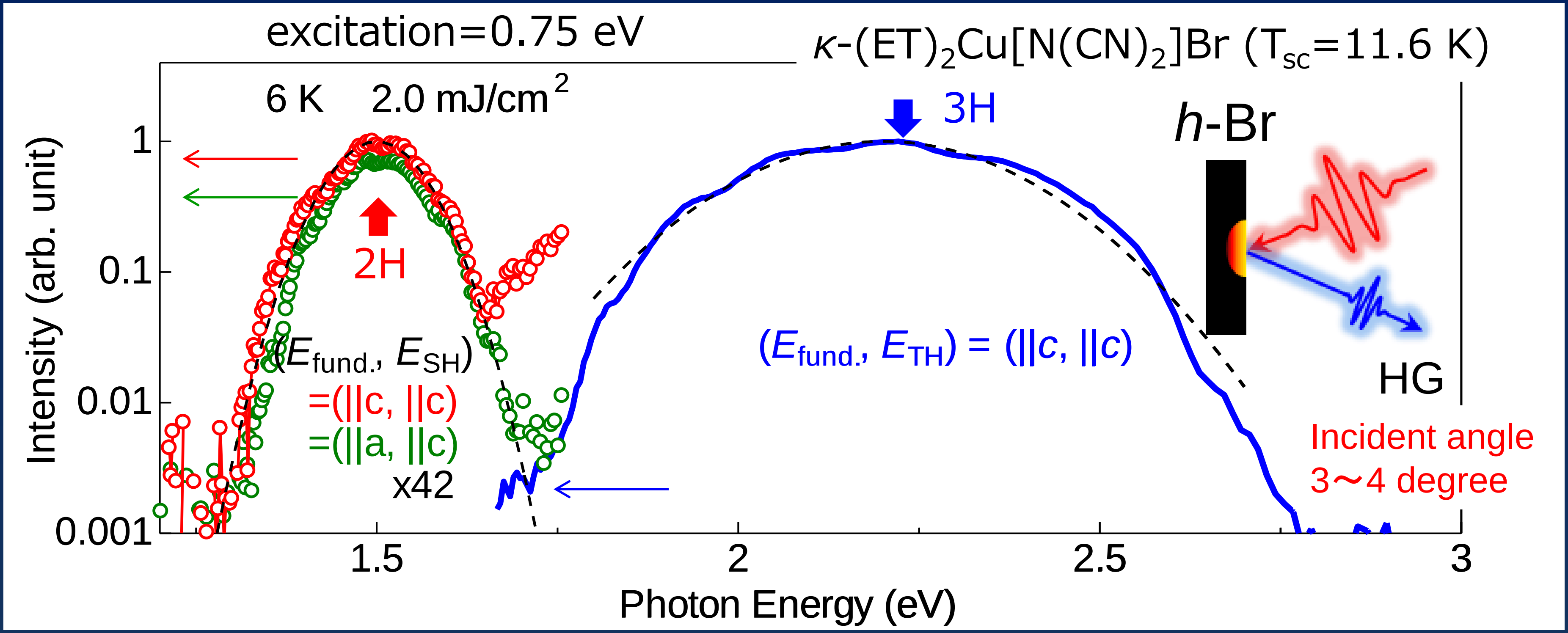
Nat. Photon 2018

iii) SHG in κ-ET salt

- ✓ SHG induced by Petahertz no-scattering current (CEP sensitive)
- ✓ Enhancement of SHG near SC fluctuation
- ✓ Quantum mechanical analysis of unconventional SHG

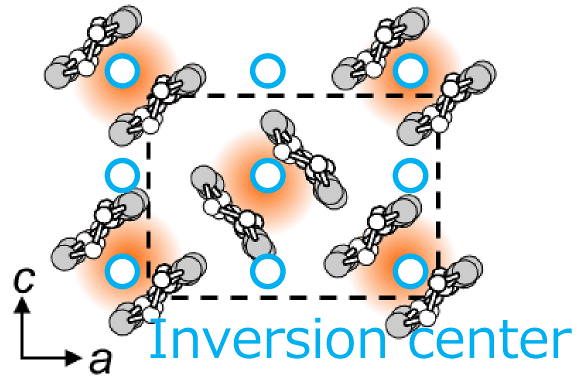
Nat. Commun. 2020

SHG &THG in organic superconductor



- ✓ SHG is not active (in perturbation)
- ✓ CEP (carrier-envelope phase) dependence
- ✓ Temperature dependence
- ✓ Polarization dependence

→ unconventional SHG

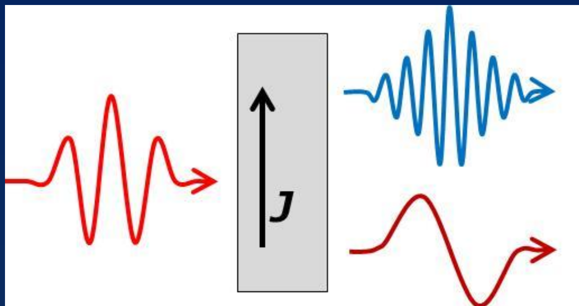


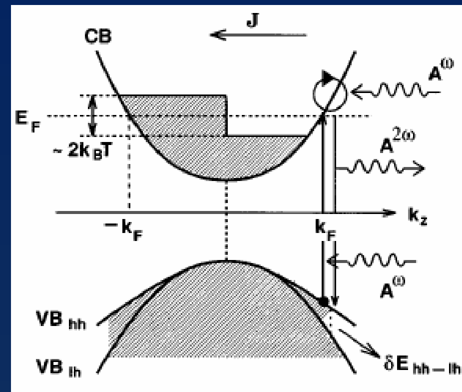
Orthorhombic

Pnma → $\chi^{(2)} = 0$

Current induced SHG

N-doped (direct gap) semiconductor (Density matrix theory)





$$\mathbf{J} = -\sum_{\mathbf{k}} e\mathbf{v}(\mathbf{k})f(\mathbf{k}) = -\frac{e\hbar}{m_c}\sum_{\mathbf{k}} \mathbf{k}f(\mathbf{k})$$

$$\chi_J^{(2)}(2\omega;\omega,\omega) = \frac{e^3}{4\epsilon_0 m_0^2 \mu\omega^4 (k_B T + i\hbar T_2^{-1})} \times \sum_{\mathbf{k}} \mathbf{P}_{vc,\mathbf{k}}(\mathbf{k}\cdot\hat{\mathbf{e}})(\mathbf{P}_{cv,\mathbf{k}}\cdot\hat{\mathbf{e}})f(\mathbf{k})$$

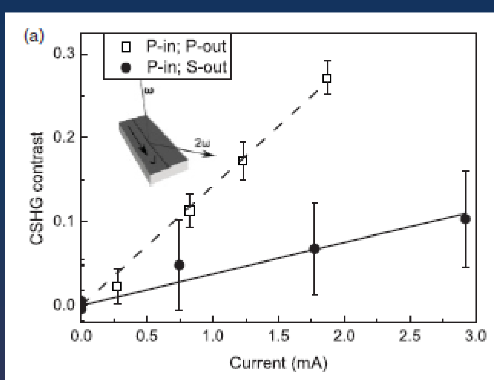
Khurgin,
APL67, 1113 (1995)

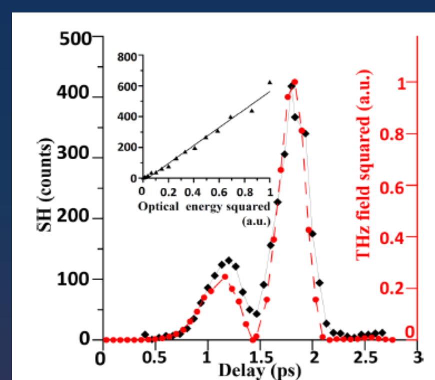
*Breaking of spatial symmetry
in the sense that the induced current can't be described as odd function of E(t)

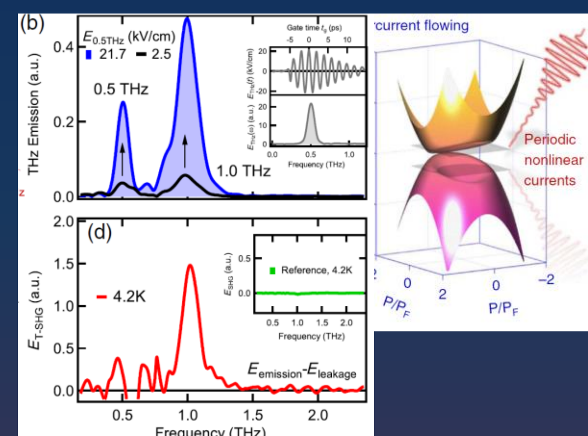
Graphene (DC)

Graphene (THz)

Nb_3Sn (THz)







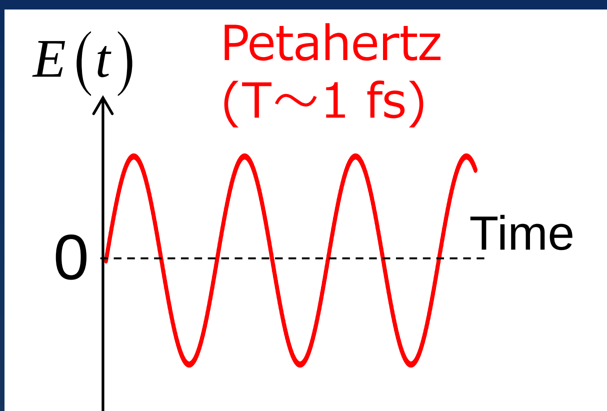
Bykov *et al*.,
PRB85, 121413 (2012)

M. Tokman et al.,
PRL. 99, 155411(2019)

Vaswani et al.
PRL124, 207003(2020)

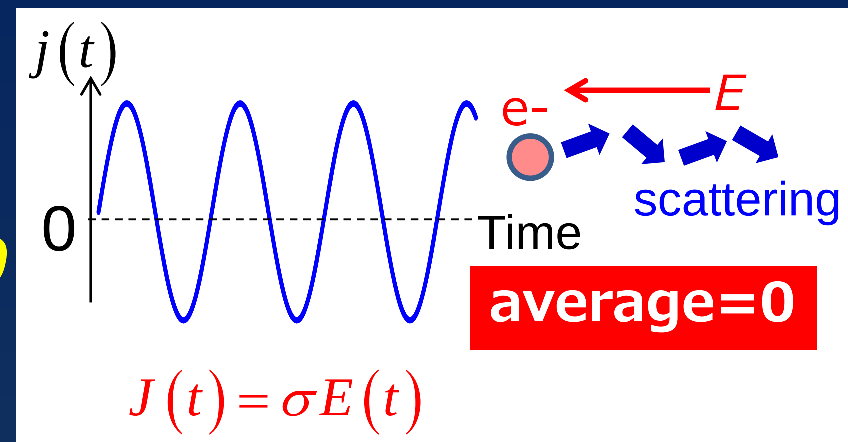
Does osc. light field induce current ?

No ! If ohm's low works



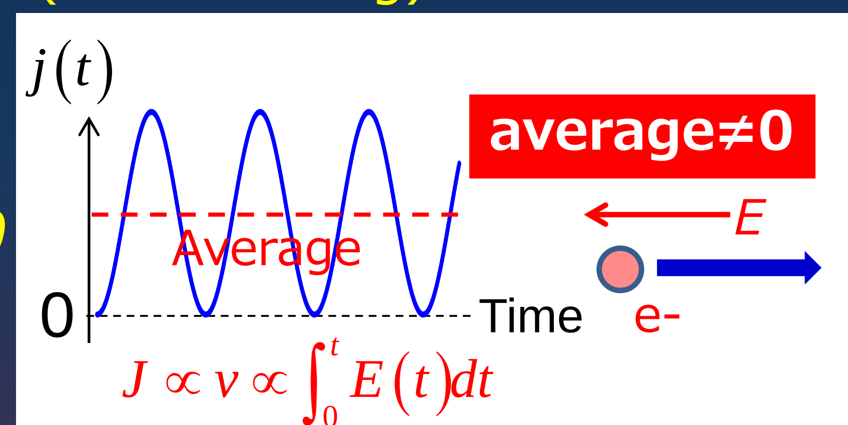
Net current= 0

Ohm's low (with scattering)



Net current≠ 0

Electron acceleration
(no-scattering)

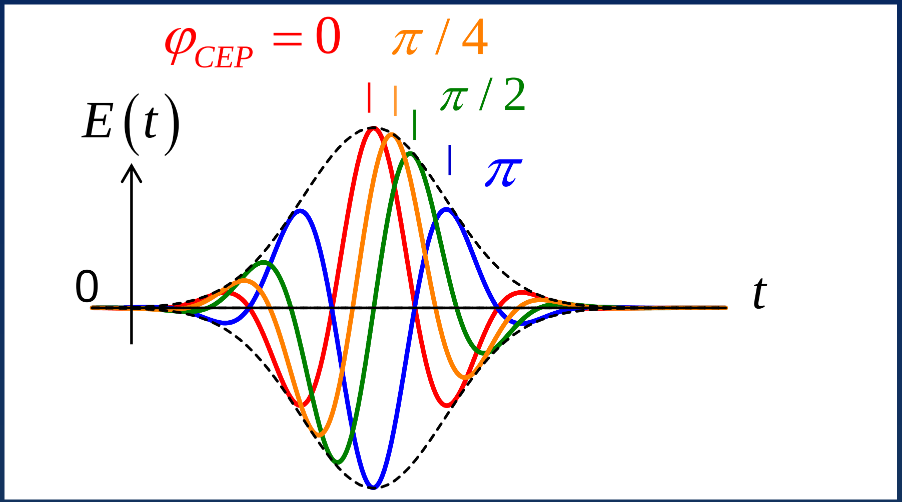


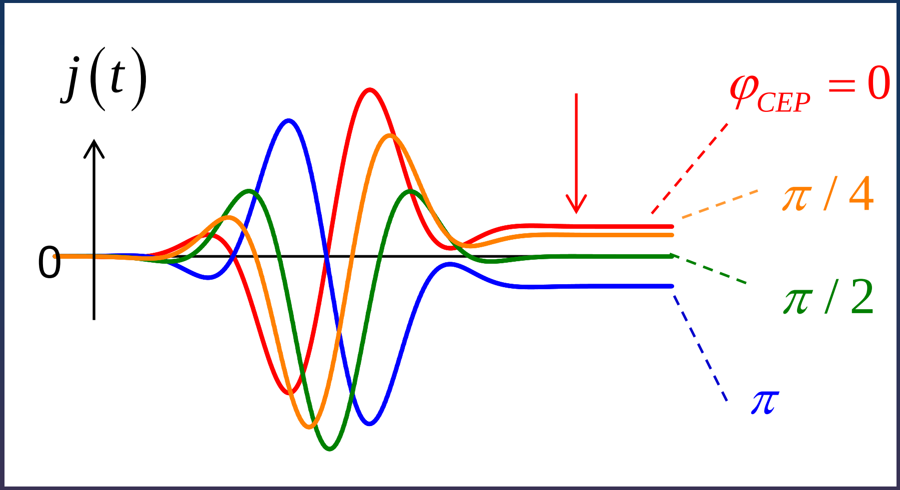
If, we have no scattering, averaged current ≠0

Carrier Envelope Phase (CEP)

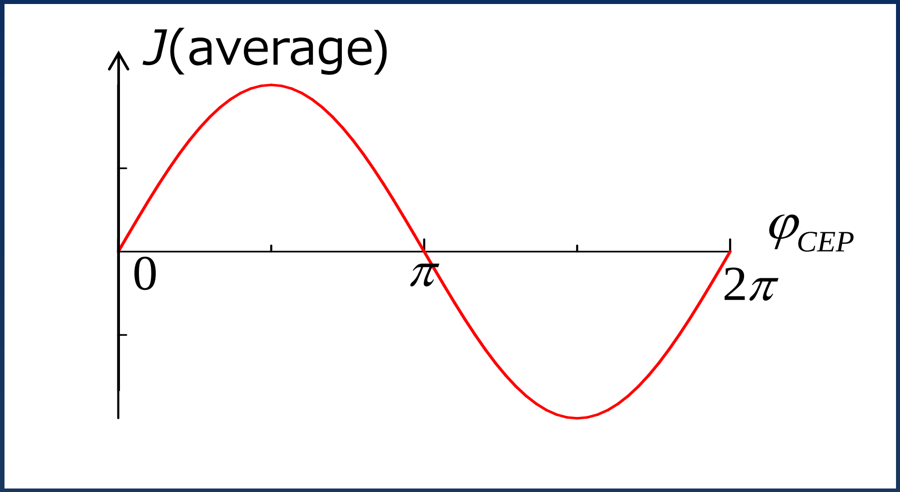
$$E(t) = E_0(t)\sin(\omega t - \varphi_{CEP}) \qquad \varphi_{CEP} : \text{CEP}$$

$$J \propto v \propto \int_0^t E(t)dt$$



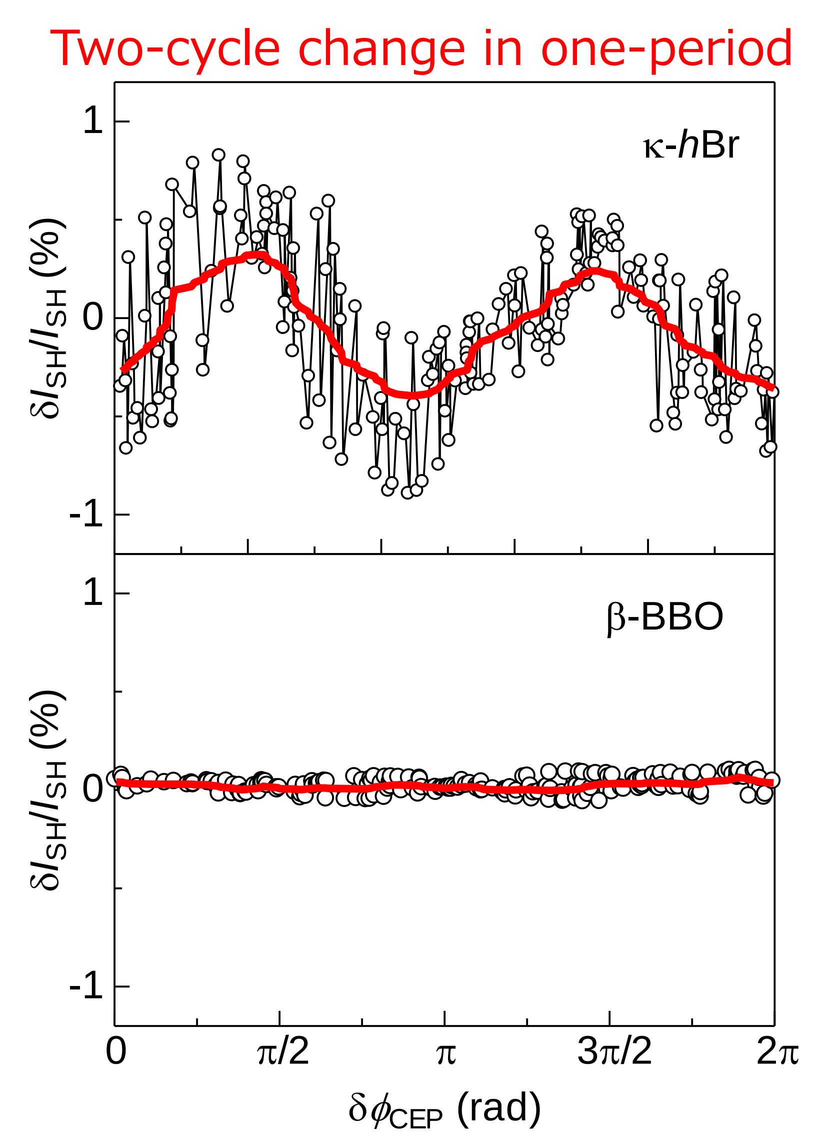


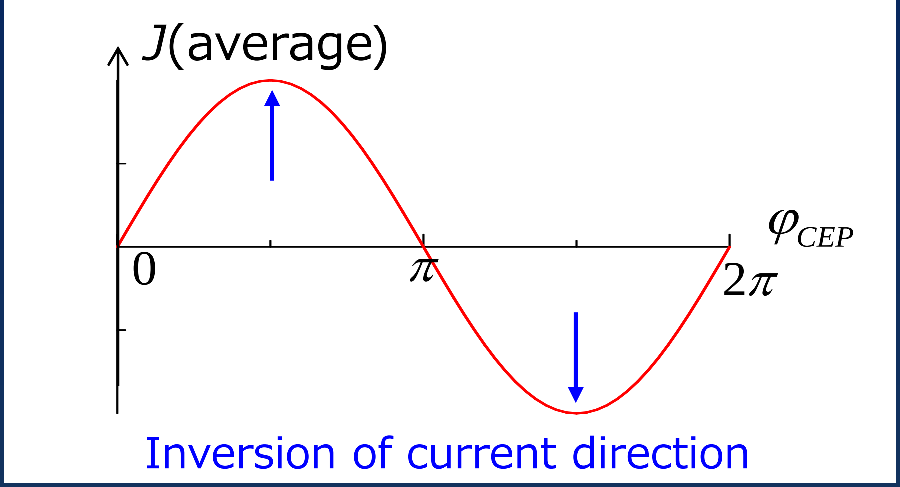
✓ *J* survives after the pulse (during scattering time window)



✓ Non-dissipative *J* is CEP sensitive
One-cycle change in one-period

CEP dependence of SHG

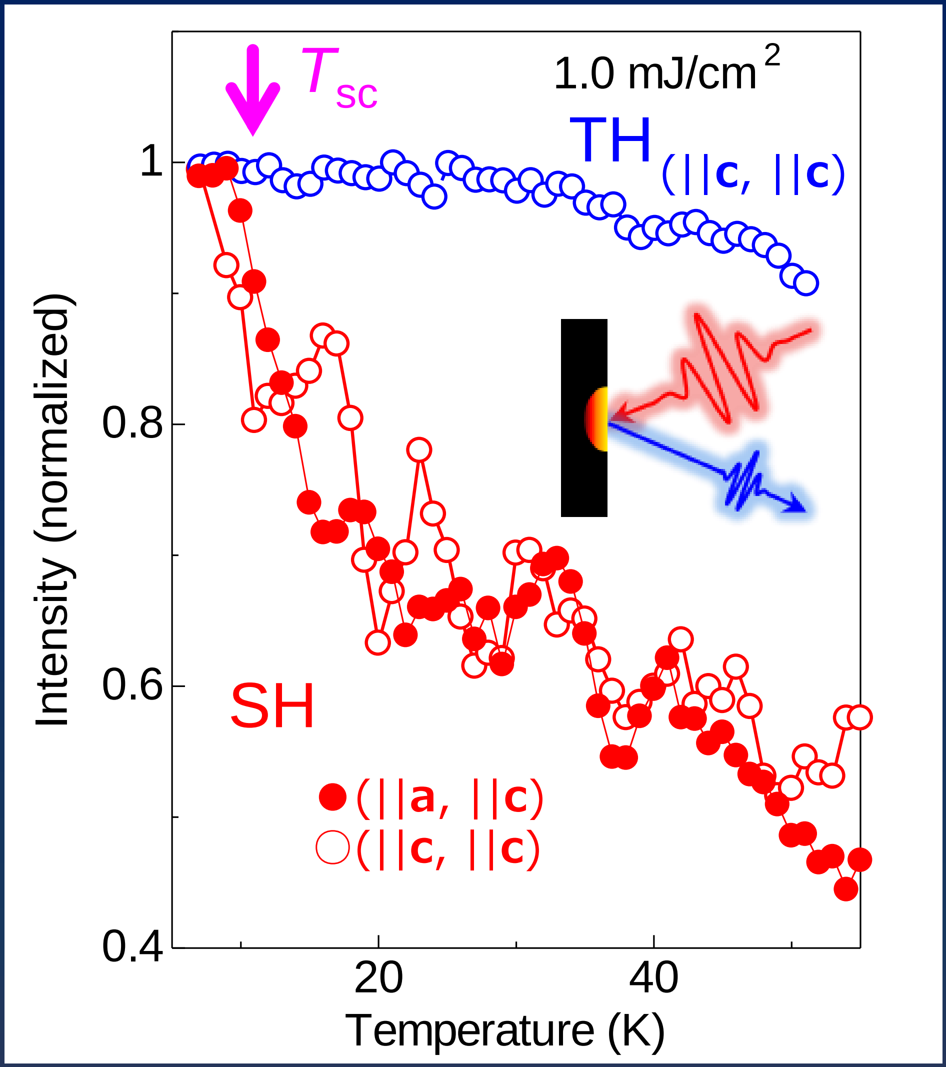


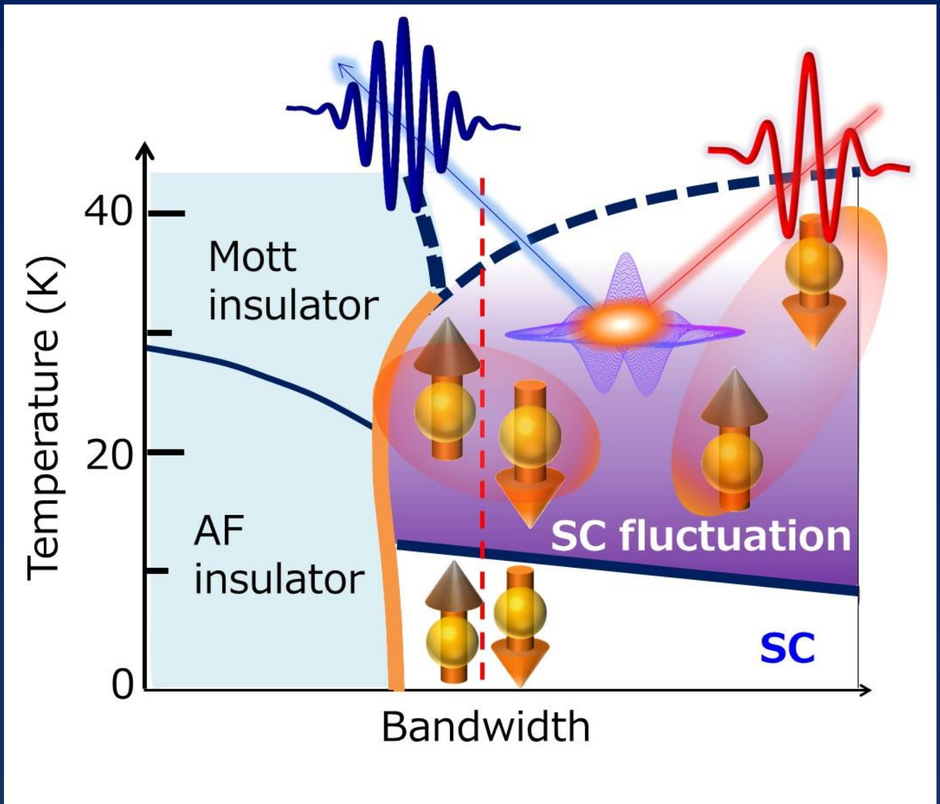


Current is modulated by *f* .
But, SHG can't distinguish direction of current
(SHG is modulated by 2 *f*)

SHG is described by non-scattering current

Temperature dependences of SHG





✓ SHG increases toward T_{SC}

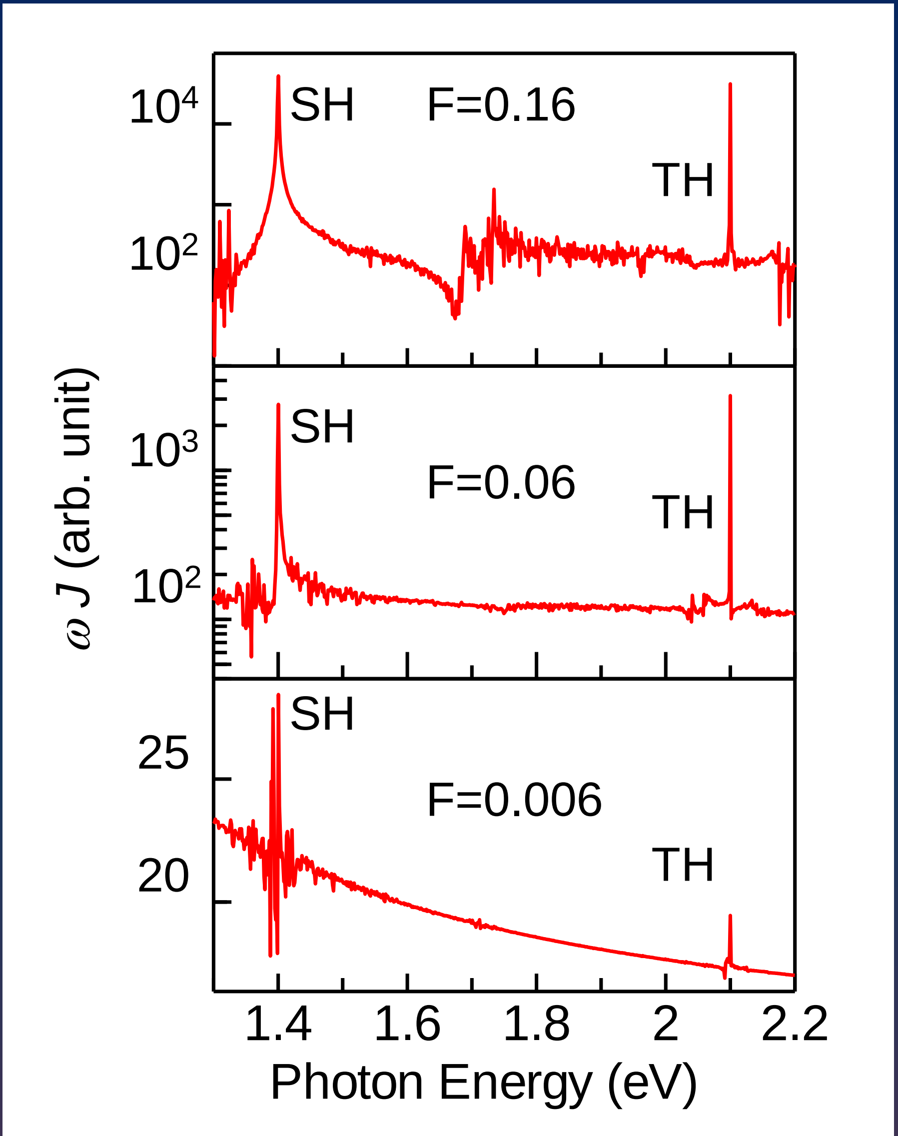
SC fluctuation ($T>T_{SC}$)

- Lang et al., PRB49, 15227(1994)
- Kobayashi et al., PRB89, 165141(2014)
- Mckenzie., Science (1997)

✓ SHG is sensitive to SC fluctuation
(reflecting the small working distance of non-scattering current?)

Mean field theory （Prof. Yonemitsu ）

✓Hartree Fock (98x98), U=0.8, V=0, triangular lattice,
✓Hubbard model, Peierls phase ω=0.7 eV, E//c



Hubbard model (3/4 filled)

$$H = \sum_{\langle i,j\rangle\sigma} t_{ij}\left(c_{i\sigma}^{\dagger}c_{j\sigma} + c_{j\sigma}^{\dagger}c_{i\sigma}\right) + U\sum_i n_{i\uparrow}n_{i\downarrow}$$

Electric field of light: substitution

$$\mathrm{S}c_{i\sigma}^{\dagger}c_{j\sigma} \to \exp\left[\frac{ie}{\hbar c}\boldsymbol{r}_{ij}\cdot\boldsymbol{A}(t)\right]c_{i\sigma}^{\dagger}c_{j\sigma}$$

with vector potential

$$\boldsymbol{A}(t) = \theta(t)\frac{\boldsymbol{F}}{\omega_{\mathrm{fund}}}\left[\cos(\omega_{\mathrm{fund}}t-\phi)-\cos\phi\right]$$

current density $\boldsymbol{j}(t) = -\langle\frac{\partial H}{N\partial \boldsymbol{A}}\rangle$

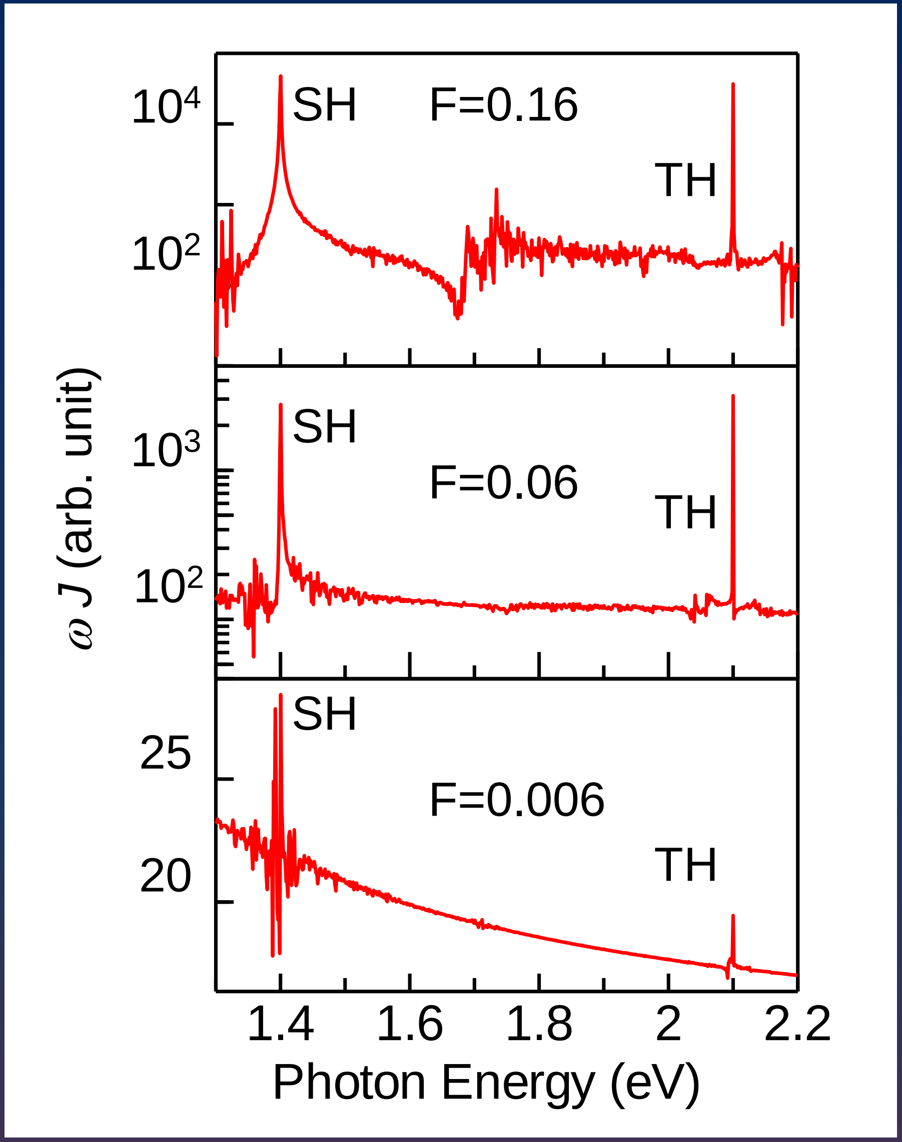
SHG and THG are evaluated as

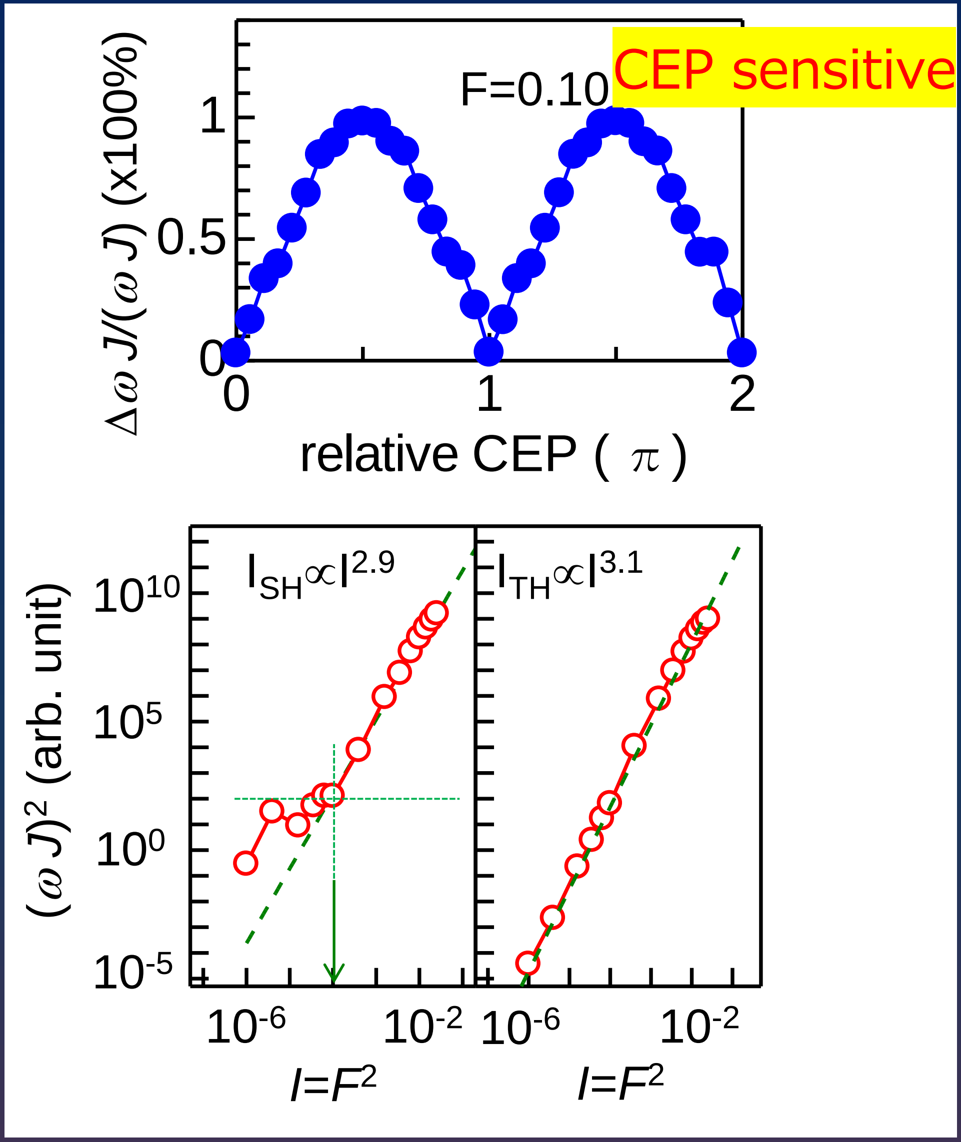
ωJ : absolute value of

Fourier transform of $\mathrm{d}\boldsymbol{j}/\mathrm{d}t)$
(500 cycle)

Mean field theory （Prof. Yonemitsu ）

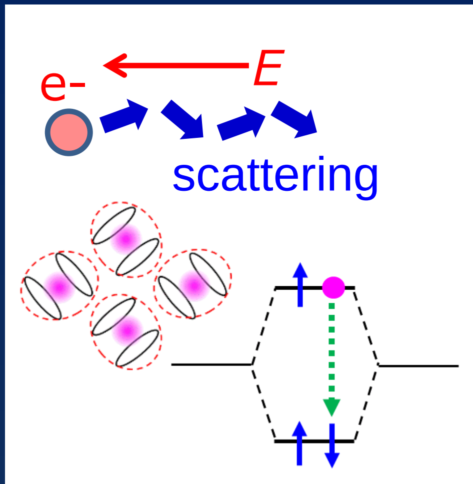
- ✓Hartree Fock (98x98), U=0.8, V=0, triangular lattice,
- ✓Hubbard model, Peierls phase ω=0.7 eV, E//c





https://doi.org/10.1039/D2FD00004K

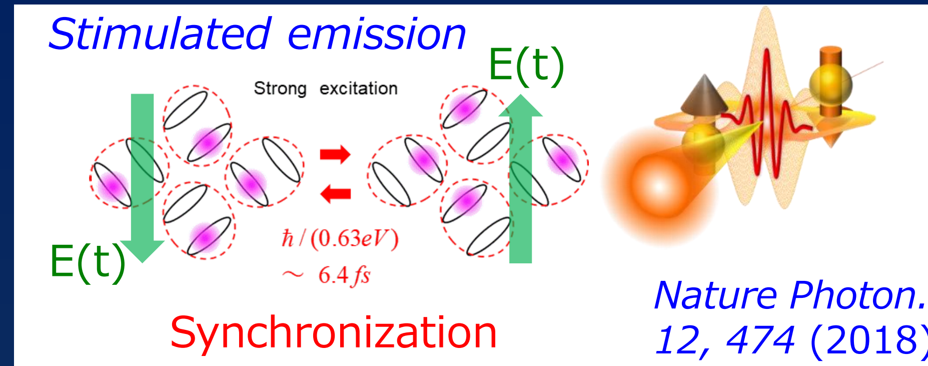
Summary Organic superconductor *h*-Br

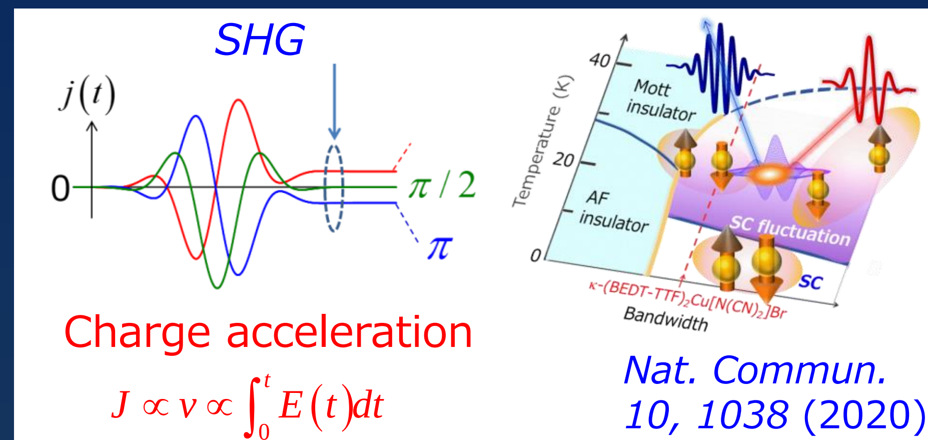


Linear response

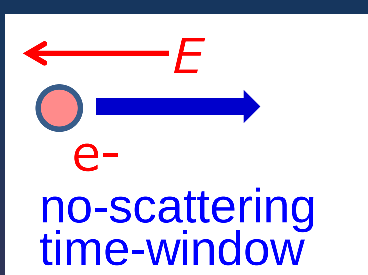
Correlated charge motion

Current induced SHG





No-scattering current is sensitive to SC fluctuation ?



$$J \propto v \propto \int_0^t E(t)dt$$

(One-electron approx.)

?

$$j = \frac{n_s e^2}{m} A$$

(London eq.)

(Many electron system)

Open problem between ultrafast and SC ?

Iwai et al. , Faraday Discussion 2022 doi.org/10.1039/D2FD0004K

Outline (continued)

iv) Ultrafast magnetization in Kitaev spin-liquid α-RuC

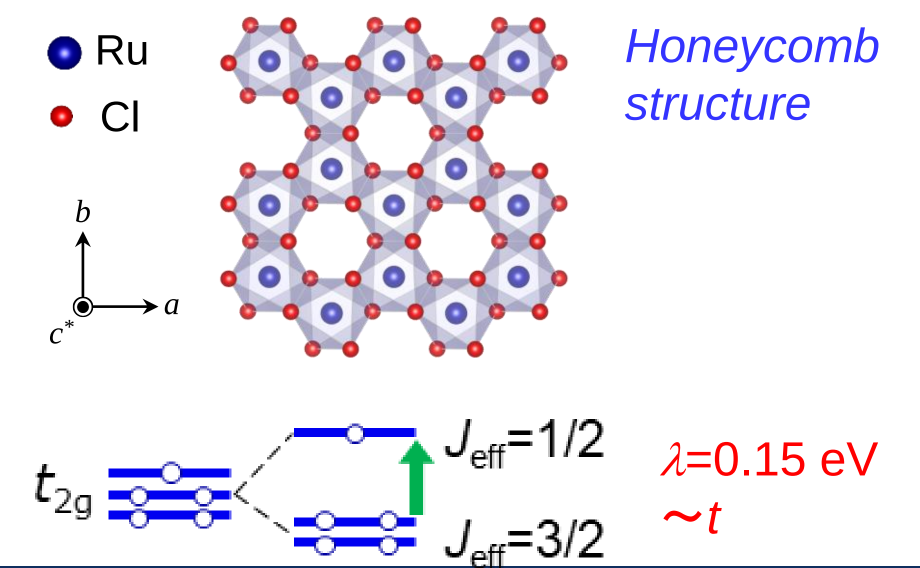
✓ Kitaev spin-liquid candidate α-$RuCl_3$
✓ Ultrafast mqgnetization (larger for $T>T_N$)
✓ Resonant effect to spin-orbit excitons
✓ Coherent carrier dynamics & theory

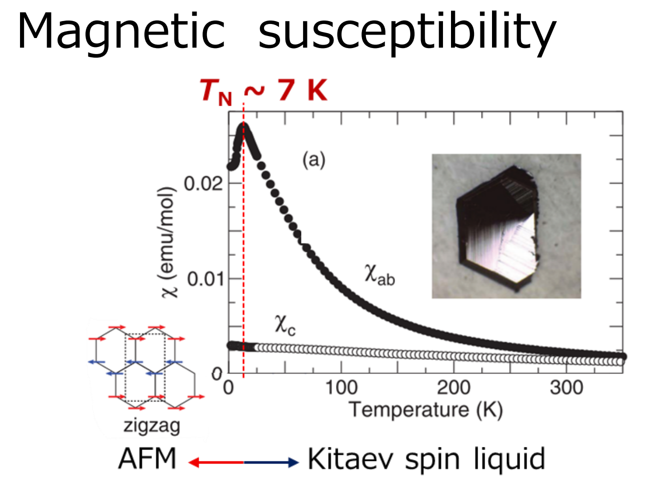
Phys. Rev. Res. 4, L032032 (2022)
arXiv: 2207.03877

v) Summary & Future problems

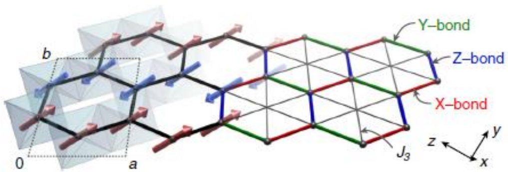
✓ High-T_c Cuprates, Correlated Dirac semimetal (Iridates)···

α-$RuCl_3$: Spin-orbit assisted Mott insulator





Sears *et al*., PRB 2015



Zig-zag AFM ($T<T_N$)
Winter, Valenti et al., Nat. Commu 2017

✓ No magnetic order ($T>T_N$=7K)

- described as Kitaev spin liquid
- excited : Majorana Fermion

A. Kitaev, Ann. Phys. 2006

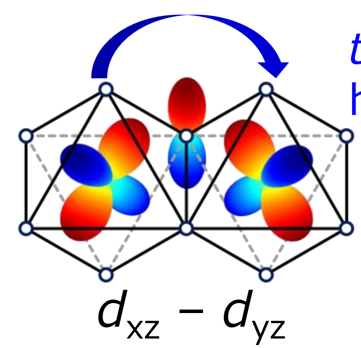
✓ Spin-orbit assisted Mott Insulator

- 1/2 filling is realized by SOI (λ~0.15 eV)

Jackeli, Khaliullin PRL 2009

✓ Inter-site hopping *t*
(between different t_{2g} orbitals (such as d_{xz} – d_{yz})

Winter al. Phys. Rev. B93, 214431(2016)



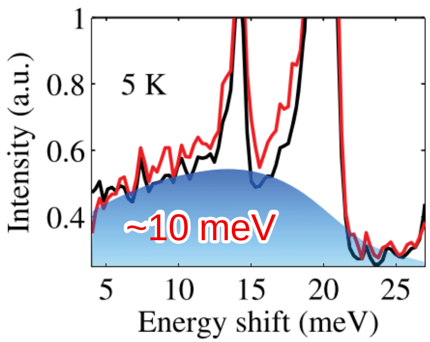
t: intersite hopping

Orbital moment?

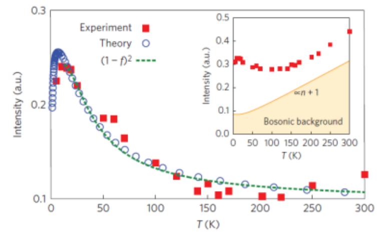
Optical properties of α-$RuCl_3$

✓ Raman

Sandilands *et al.*, PRL114, 147201(2015)



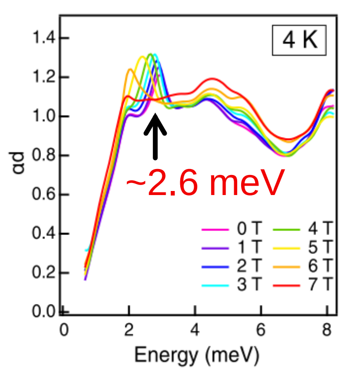
Nasu *et al.*, Nat. Phys. 12, 912 (2016)



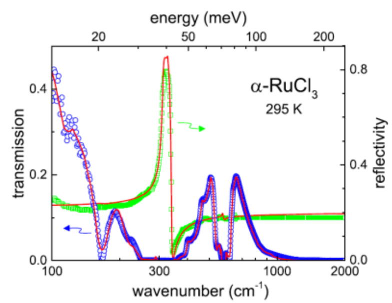
Excitation of QSL

✓ IR

Little *et al.*, PRL119, 227201(2017)



Reschke *et al.*, PRB96,165120(2017)

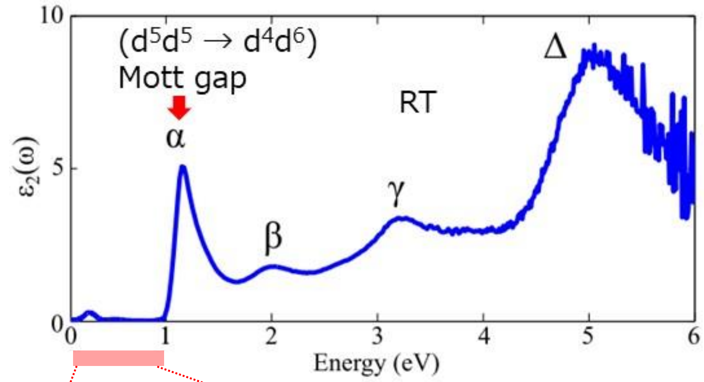


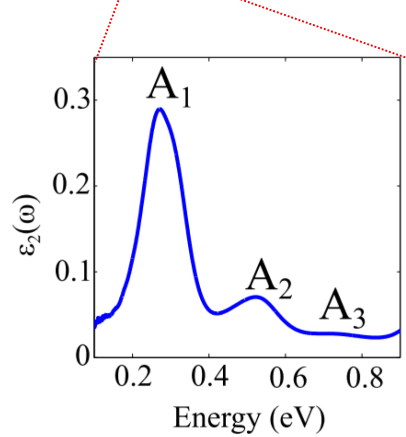
Glamazda *et al.*, PRB95,174429(2017)

Magnon

Phonon

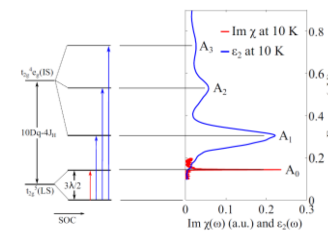
✓ Mid-IR～UV (> 0.1 eV)



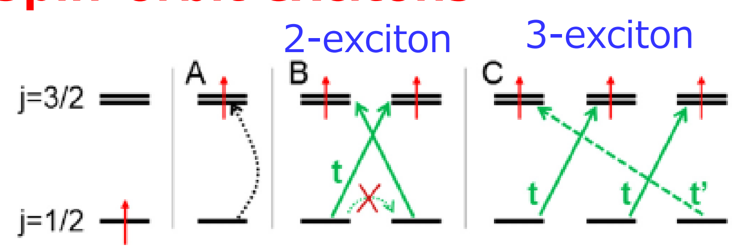


Sandilands *et al.*,
PRB **93**, 075144 (2016)
PRB **94**, 195156 (2016)

・Intra site d-d ?



・**Spin-orbit excitons**

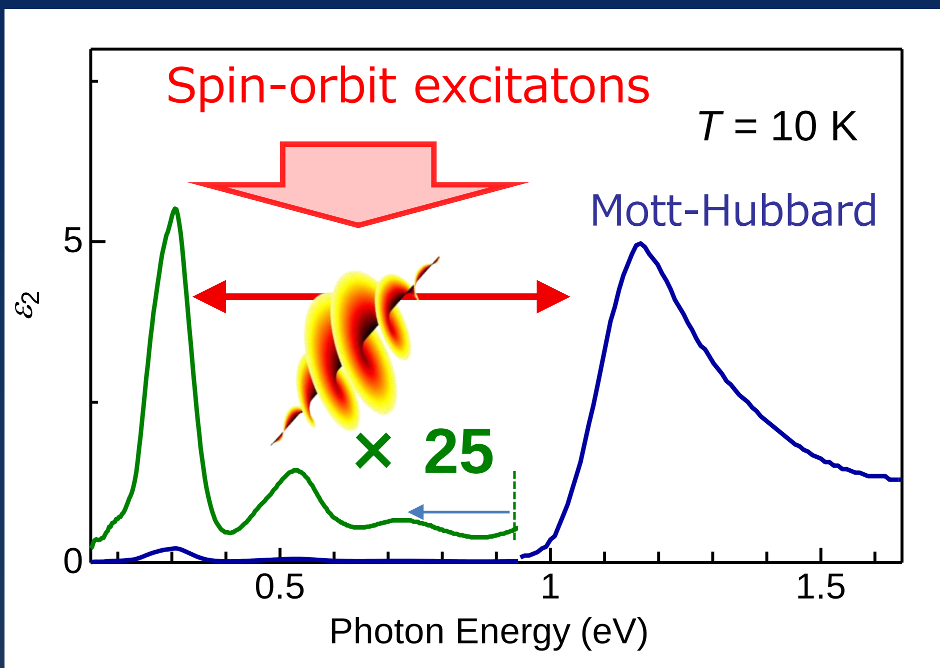


Warzanowski et al., PRR2,042007R(2020)

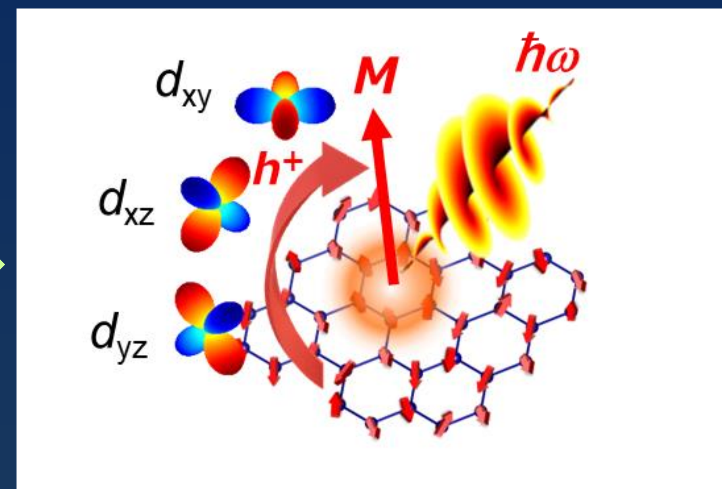
Objective

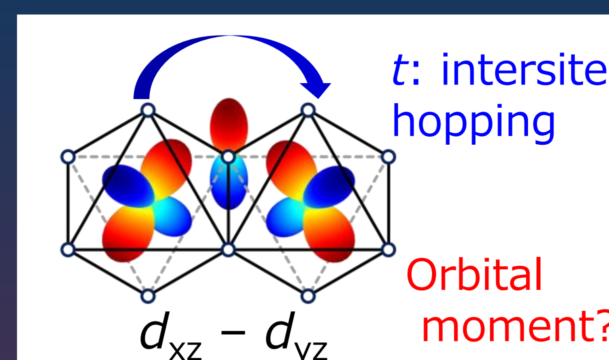
Modulating magnetic properties
by excitation of spin-orbit excitations

✓ Resonant excitation
by circular polarized light



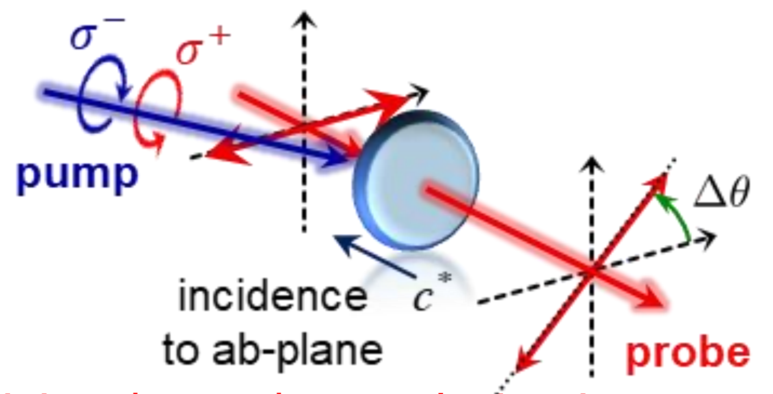
✓ We expect that
orbital moment is induced
by inter-site charge hopping
(bwteen different t_{2g} orbitals) ?





Query
Light-induced magnetization
is possible via orbital moment ?

Experimental



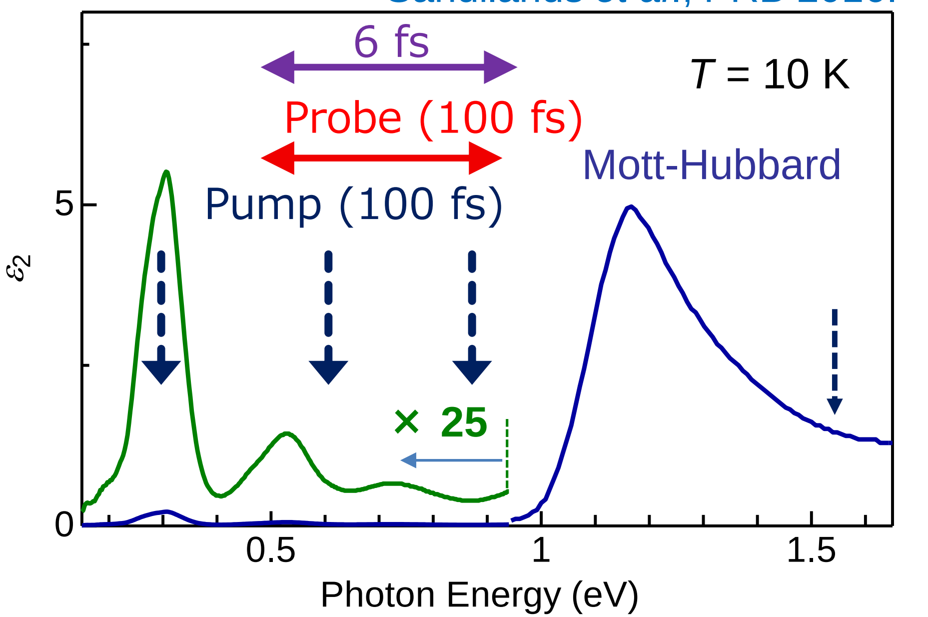
Helicity dependent polarization rotation

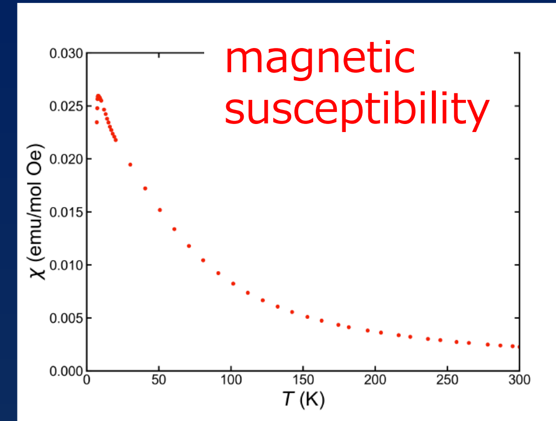
Inverse Faraday effect

WFM Kimel *et al.*, Nature 2005

AFM Satoh et al., PRL 2010

Sandilands *et al.*, PRB 2016.





Sample : α-$RuCl_3$(single crystal)

E || *ab* plane, thickness ~ 50 μm

Temperature :

4 K - 20 K, 300 K **(T_N = 7-8 K)**

○100 fs pulse (spot size 100 μm)

Pump : 0.30, 0.62, 0.89 eV, (1.55 eV)

(0.1-4.0 mJ/cm^2)

✓ Circular (σ^+, σ^-) polarization

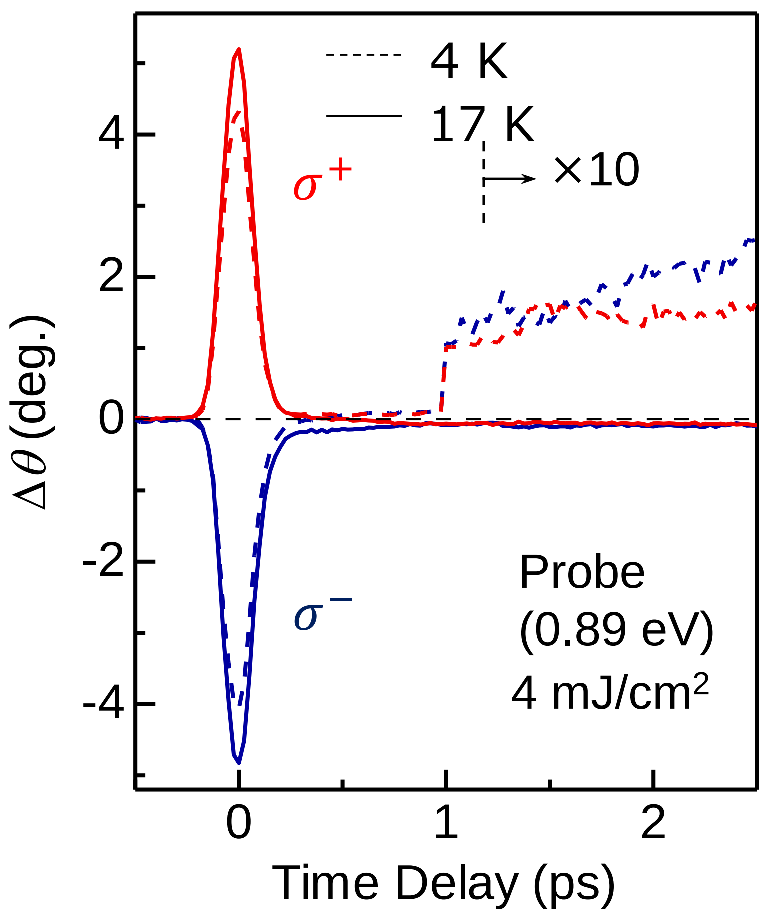
Probe : 0.54-1.03 eV

✓ Libear polarization

○ 6 fs pulse ΔR/R (charge dynamics)

0.55 eV- 1 eV, 1mJ/cm^2

Polarization rotation Δθ



✓ **Ultrafast(~100 fs) & large response (helicity sensitive)**

$\Delta\theta = 5°$, t~50 μm, 4mJ/cm²

AFM(NiO) ~1/20
$\Delta\theta = 1.14°$, t=0.1mm, 10mJ/cm²
Satoh et al., et al., *PRL* 2010

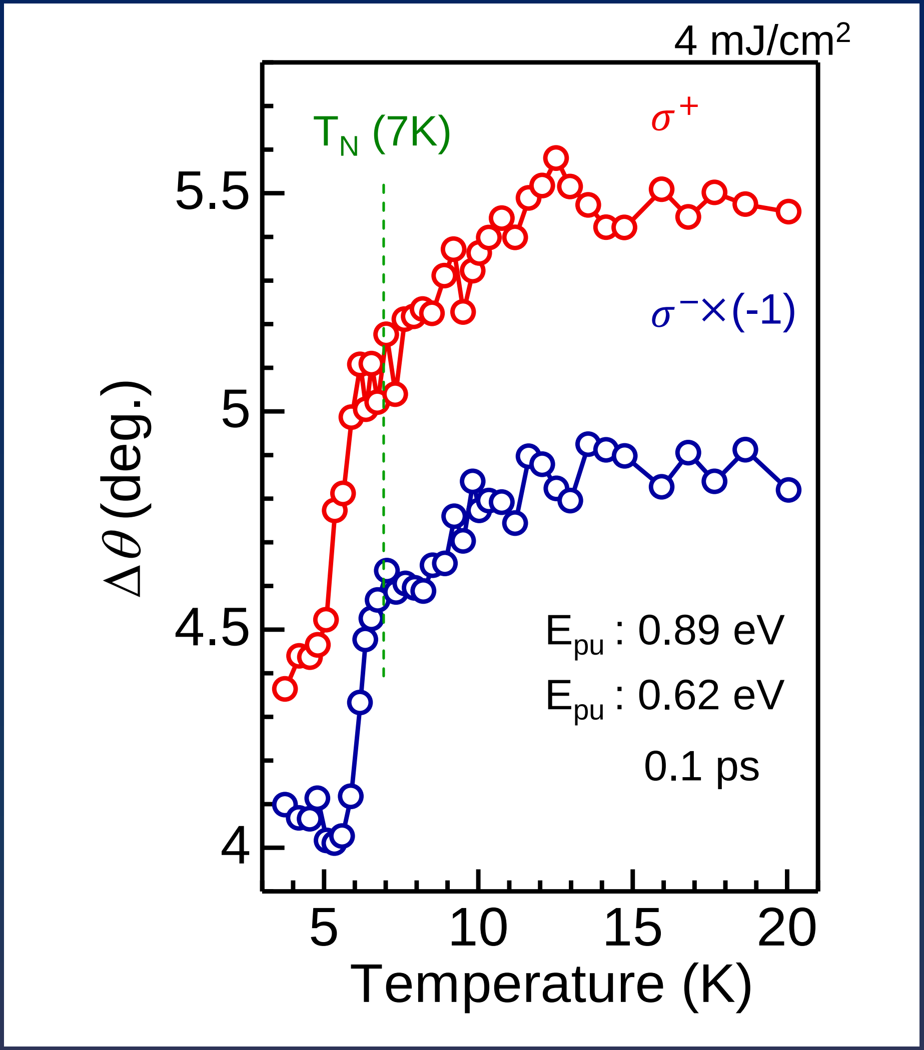
Paramagnet (TGG) ~1/400
$\Delta\theta = 0.15°$, t=1mm, 3mJ/cm²
Mikhaylovskiy et al., *PRB* 2012

✓ Temperature dependence
$\Delta\theta$ for $T > T_N$ is larger
(→next page)

✓Helicity-independent slow component
(> 10 ps,)
→ melting of AFM order

Light induced ultrafast magnetization (⊥ plane)

Temperature dependence (fast component)



✓ Reduction at $T < T_N$?

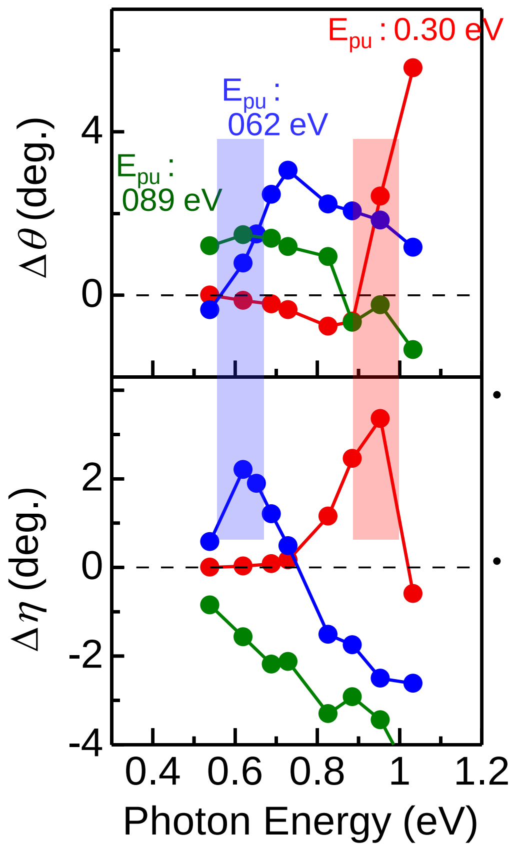
opposit to the expected tendency in typical IFE in AFM & WFM (increase in Δθ below T_N)

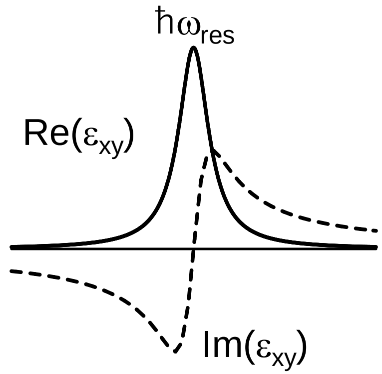
New mechanism of light-induced magnetization ?

Excitation & detection energy dependence

Rotation angle (Δθ) and Ellipticity (Δη)

Pump (i) 0.30 eV, (ii) 0.62 eV, (iii) 0.89 eV,



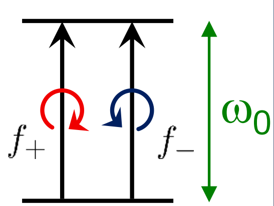


- **Rotation angle**

$$\theta = \frac{\omega l}{2cn}\mathrm{Im}(\varepsilon_{xy})$$

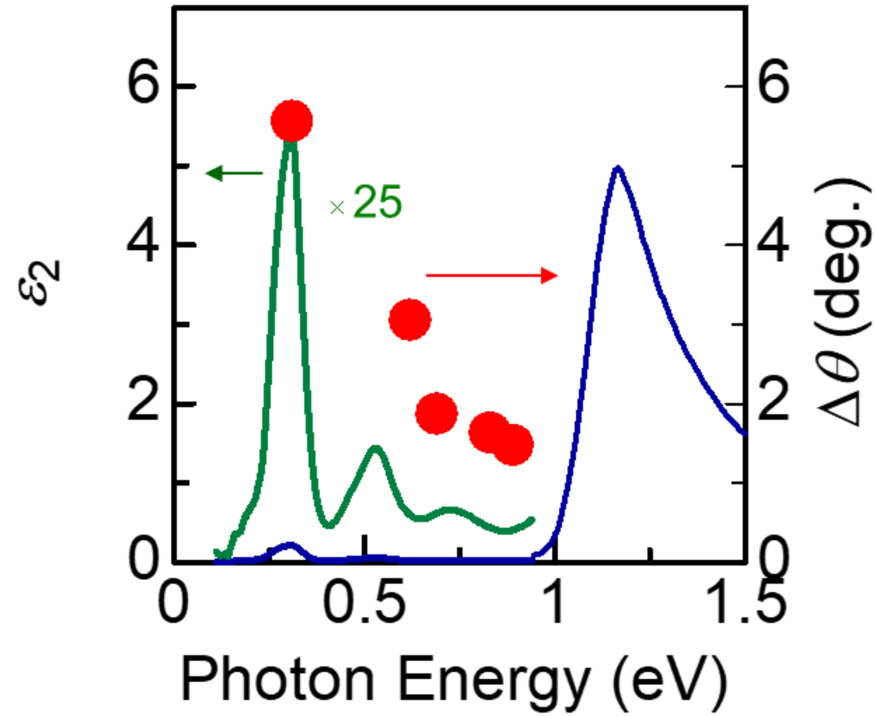
- **Ellipticity**

$$\eta = -\frac{\omega l}{2cn}\mathrm{Re}(\varepsilon_{xy})$$



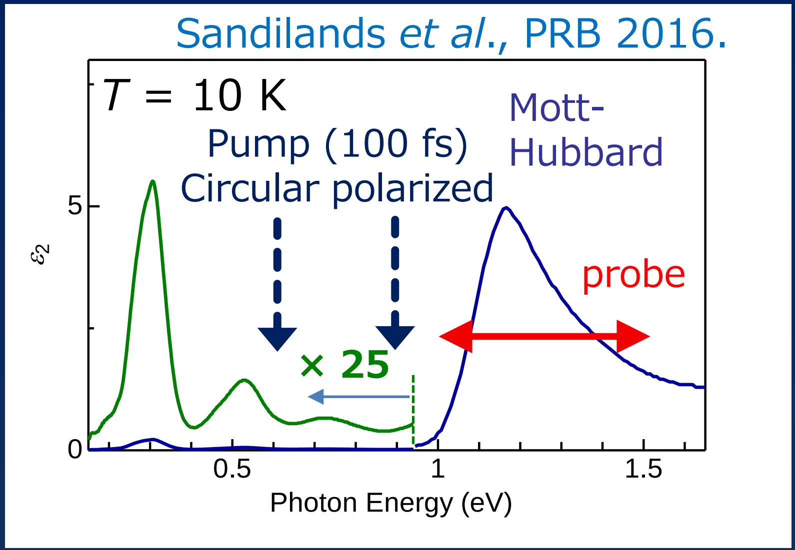
Helicity dependent change of oscillator strength

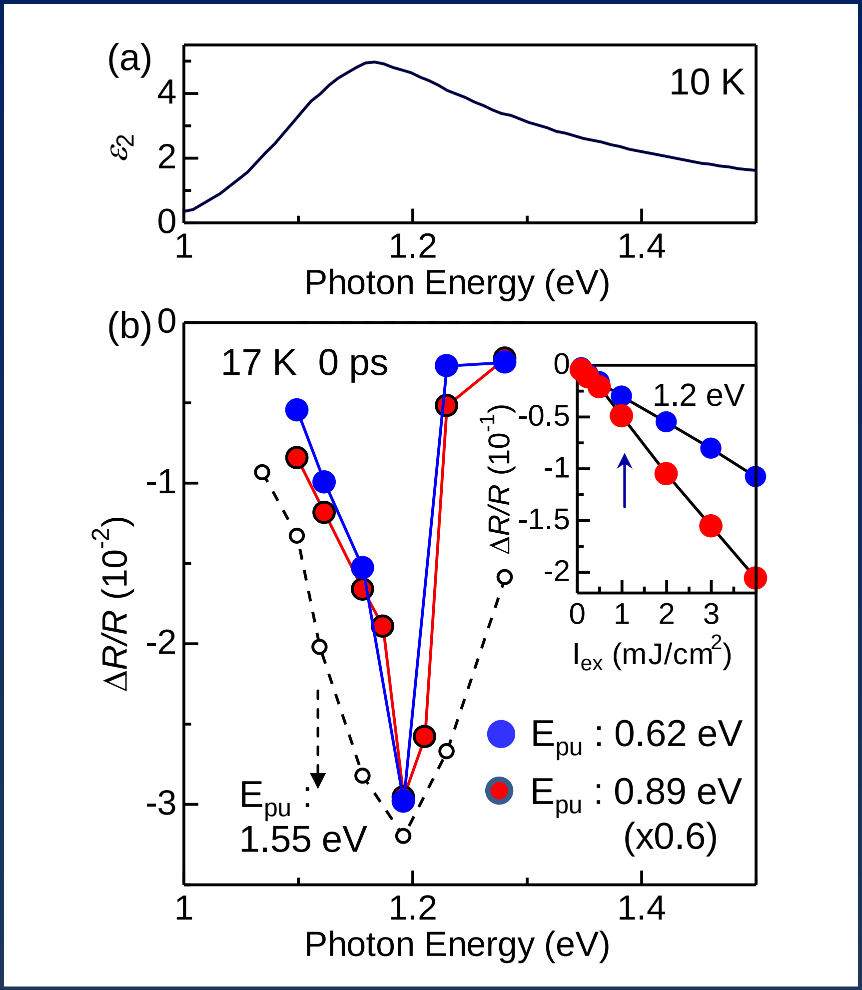
Excitation energy dependence (Excitation spectrum of Δθ)



Resonance effect to spin-orbit excitons

ΔR/R under Spin-orbit excitation





✓ **Pump : S-O excitation (0.3, 0.62 eV)**

✓ **Probe : Mott-Hubbard transition**

- Bleaching of Mott-Hubbard transition under in-gap excitation
- Linear response
  →2-photon abs. is ruled out

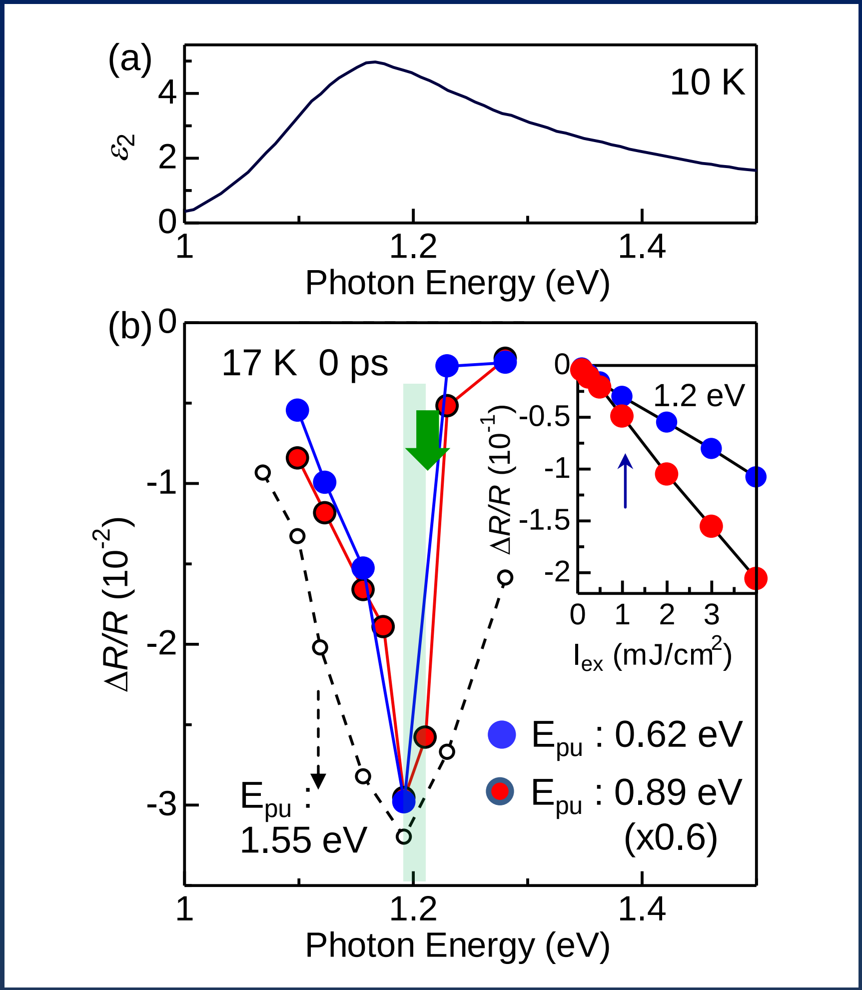
"Spin-orbit assisted Mott insulator"

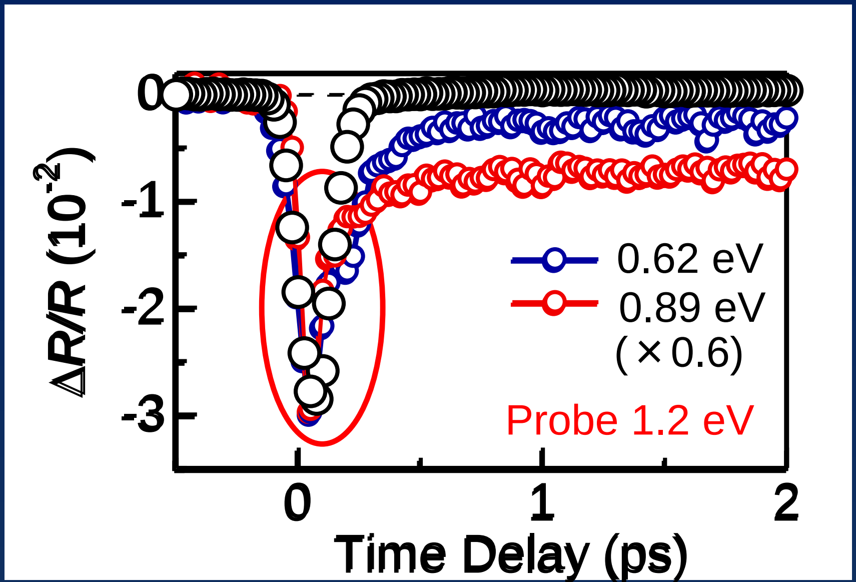
Plumb *et al*., PRB 90, 041112 (R) (2014)

Kim *et al*., PRL 117, 187201 (2016)

Mott gap is modulated even for in-gap excitation

Time profile of ΔR/R





✓ **Relaxation dynamics**

i) < 100 fs

→ Ultrafast magnetization ?

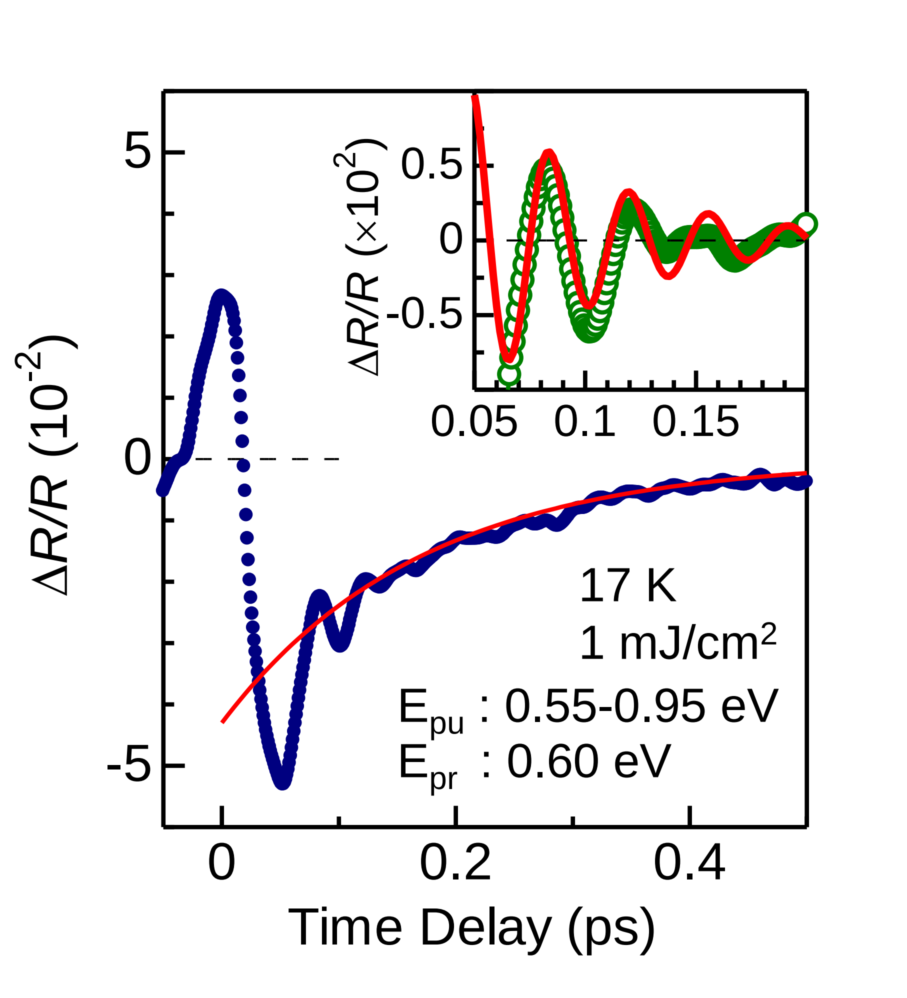
ii) ~ 400 fs

→ phonon, spin ?

✓ Ultrafast magnetization is related to the fast charge dynamics ?

Charge dynamics captured by 6 fs pulse

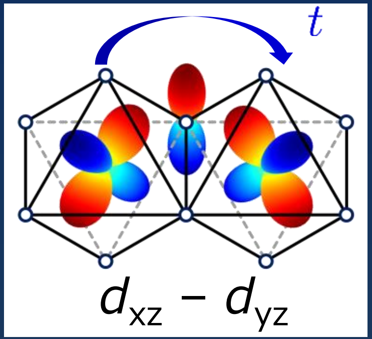
ΔR/R measurement using 6 fs pulse (CEP locked)



✓ oscillation period ～ 40 fs
(<shortest phonon period 100 fs)

⇔ *charge hopping t*
(0.1 eV=h/(40 fs))

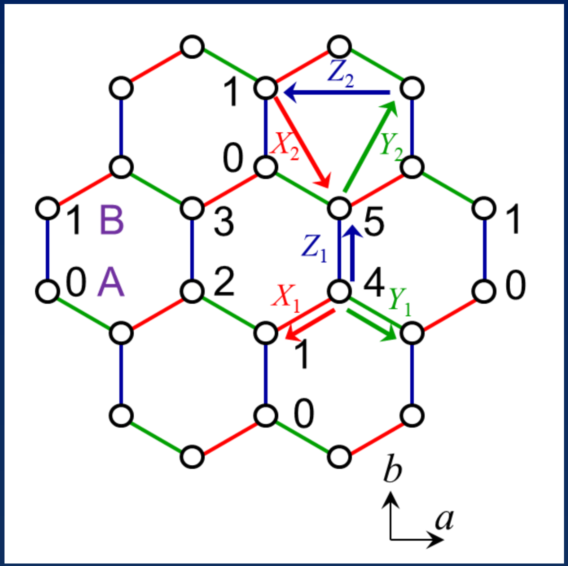
→**Coherent charge hopping between different t_{2g} orbitals?**



dephasing time 60 fs
～ lifetime of magnetization

Magnetization is induced by the coherent charge motion between different t_{2g} (lifting orbital moment)

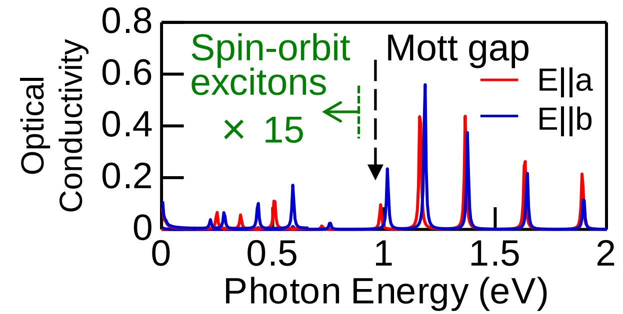
Opto-magneto effects in α-$RuCl_3$ (theory)



Quantum mechanical analysis (steady state)

J. G. Rau et al., PRL112, 077204 (2014)
H. –S. Kim et al., PRB93, 155143 (2016)
S. M. Winter ate al., PRB 93, 214431 (2016)

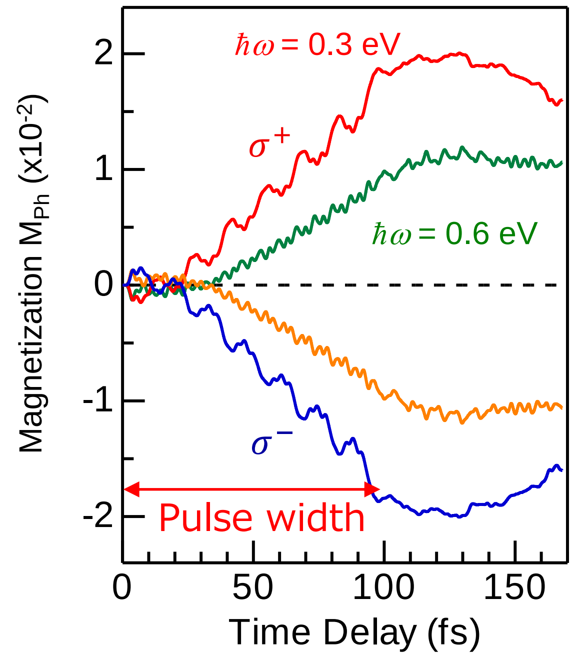
✓ The result of numerical calculation shows that in-gap excitation is essential



✓Exact diagonalization (6-site)
+time dependent Schrödinger equation

・3- orbital (d_{yz}, d_{xz}, d_{xy}) Hubbard model

$$H_U = U\sum_{i,a} n_{i,a,\uparrow}n_{i,a,\downarrow} + (U' - J_{\rm H})\sum_{i,a<b,\sigma} n_{i,a,\sigma}n_{i,b,\sigma} + U'\sum_{i,a\neq b} n_{i,a,\uparrow}n_{i,b,\downarrow}$$
$$-J_{\rm H}\sum_{i,a\neq b} c^\dagger_{i,a,\uparrow}c_{i,a,\downarrow}c^\dagger_{i,b,\downarrow}c_{i,b,\uparrow} + J_{\rm H}\sum_{i,a\neq b} c^\dagger_{i,a,\uparrow}c^\dagger_{i,a,\downarrow}c_{i,b,\downarrow}c_{i,b,\uparrow}$$



✓Peierls substitution

・ω=0.3 eV, 0.6 eV (pulse width =100 fs)

Calculation of magnetization ⊥ ab-plane

High-frequency expansion in Floquet theory

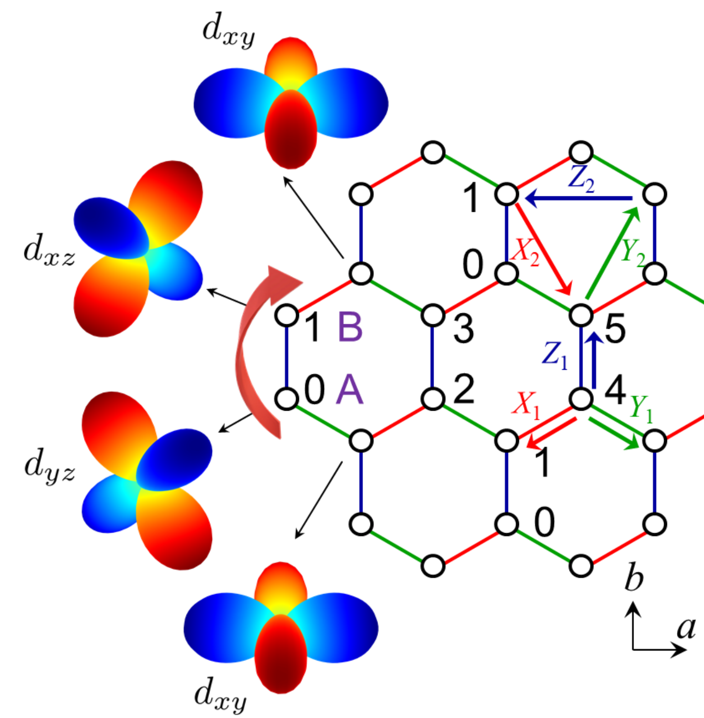
$$H_{\mathrm{F}}^{(2)} \cong \frac{1}{\omega} J_1^2 \left(\frac{F_{\mathrm{L(R)}}}{\omega}\right)\left(\pm\sqrt{3}\right)(t_2 - t_4)[t_2 - t_4 + 2(t_3 - t_1)]$$
$$\times \sum_{i\sigma} \begin{pmatrix} c_{i,yz,\sigma}^\dagger & c_{i,xz,\sigma}^\dagger & c_{i,xy,\sigma}^\dagger \end{pmatrix} \begin{pmatrix} 0 & -i & i \\ i & 0 & -i \\ -i & i & 0 \end{pmatrix} \begin{pmatrix} c_{i,yz,\sigma} \\ c_{i,xz,\sigma} \\ c_{i,xy,\sigma} \end{pmatrix}$$

the second-lowest order of the high-frequency expansion

orbital moment

✓ Effective magnetic field is induced by charge hopping between different t_{2g} orbitals

Spiral current drives magnetic moment



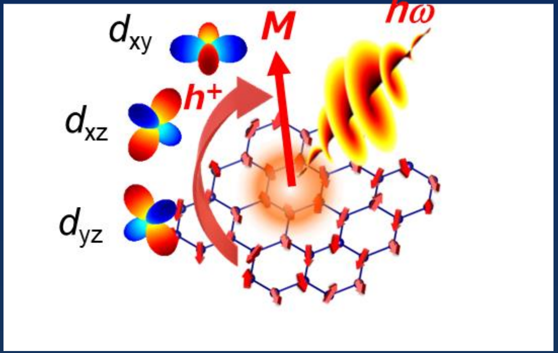
Summary

✓ Helicity dependent polarization rotation ($\Delta\theta$) in α-$RuCl_3$
→ 20 time larger than that of typical AF

✓ Increase of $\Delta\theta$ above T_N
→ opposite tendency from conventional IFE

✓ Resonant to spin-orbit excitons

✓ Possible scenario
→coherent charge motion between different t_{2g} orbitals such as d_{yz}-d_{xz}-d_{xy})

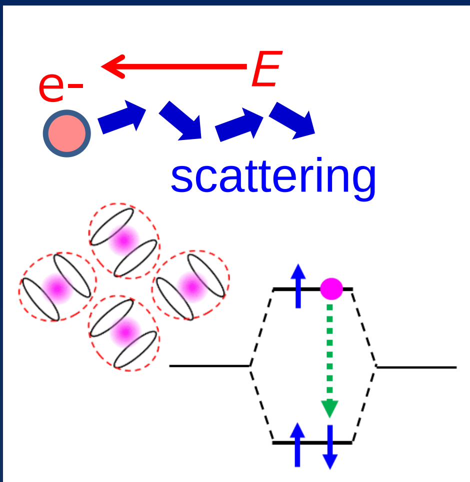


✓ Quantum mechanical analyses support the above mechanism

T. Amano et al., arXiv:2207.03877
***Phys. Rev. Research 4, L032032* (2022)**

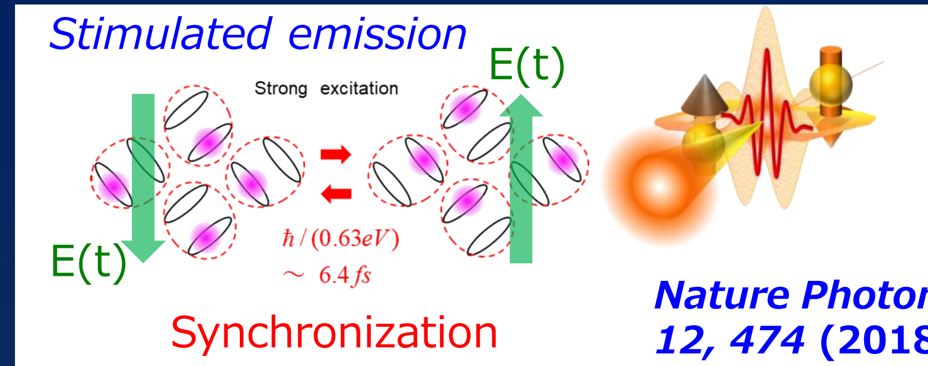
Summary

Organic SC κ-ET salt



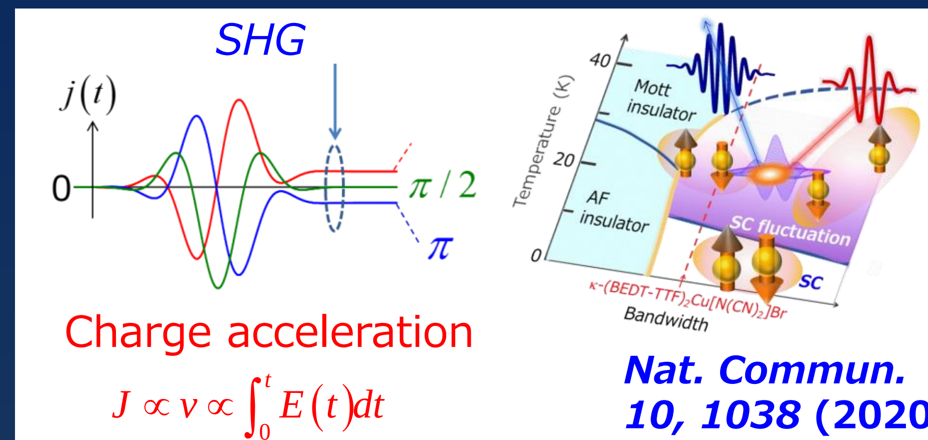
Linear response

Correlated charge motion



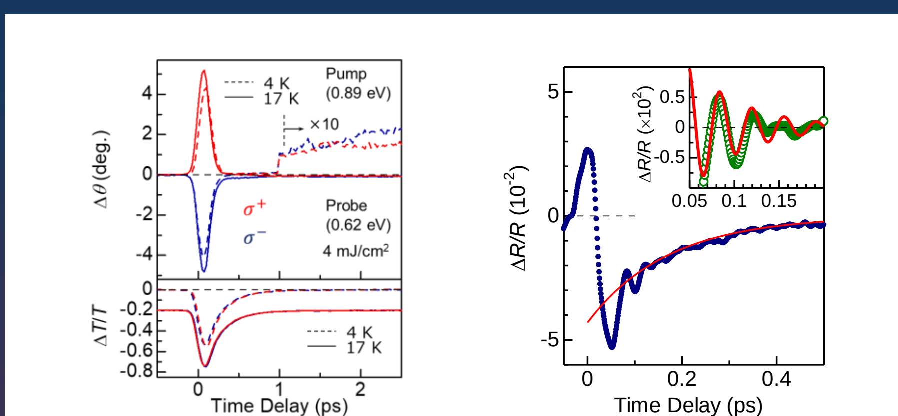
Nature Photon. **12, 474 (2018)**

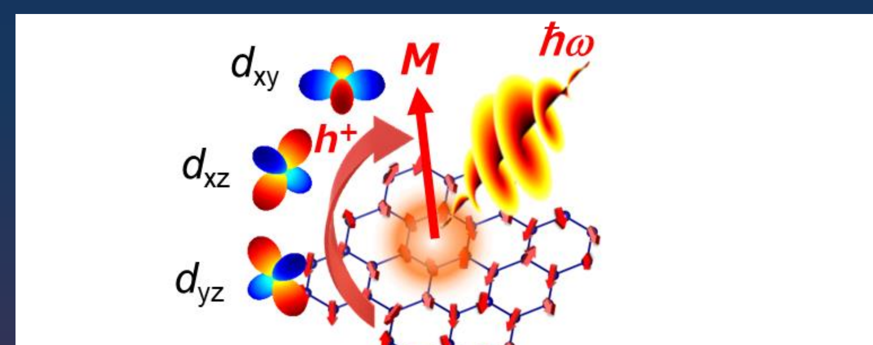
Current induced SHG



Nat. Commun. **10, 1038 (2020)**

Spin-orbit Mott insulator α-$RuCl_3$





Phys. Rev. Research 4, L032032 (2022)
arXiv:2207.03877