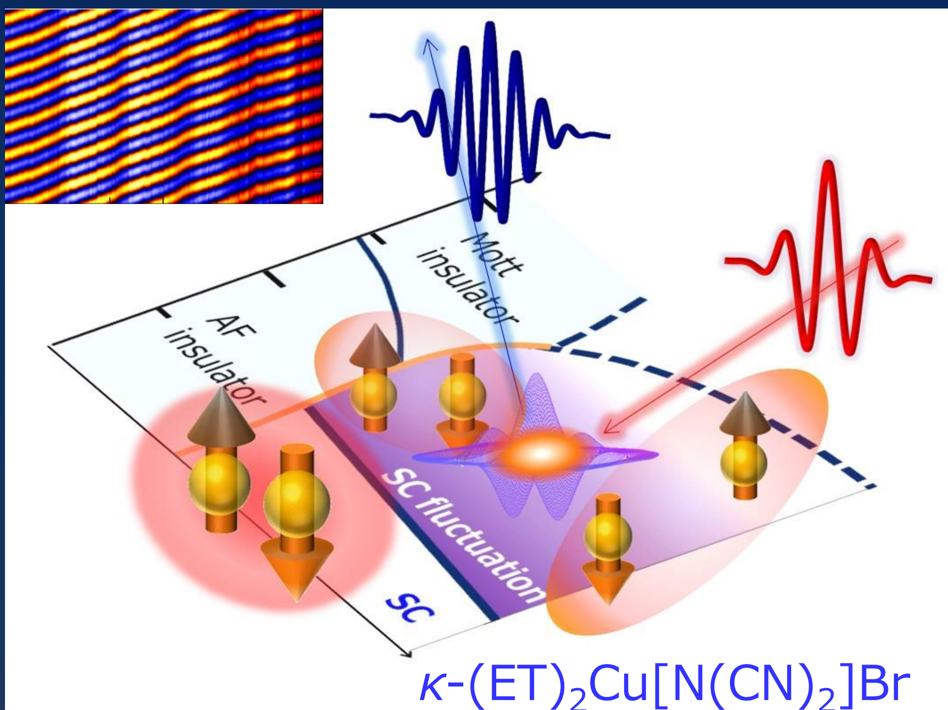
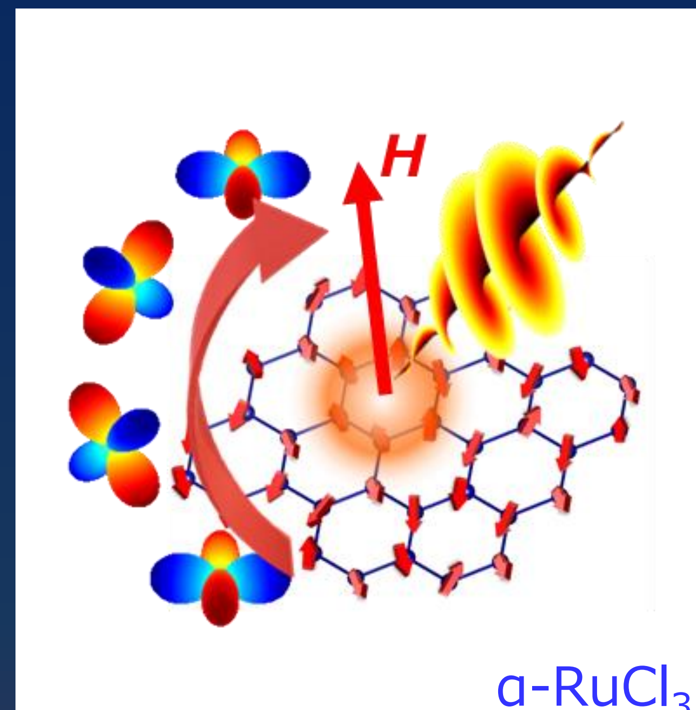
6 fs 単一サイクル光で駆動する強相関電子系、超伝導体の電荷コヒーレンス

東北大学大学院 理学研究科　岩井　伸一郎



Nat. Commun.10, 1038 (2020)



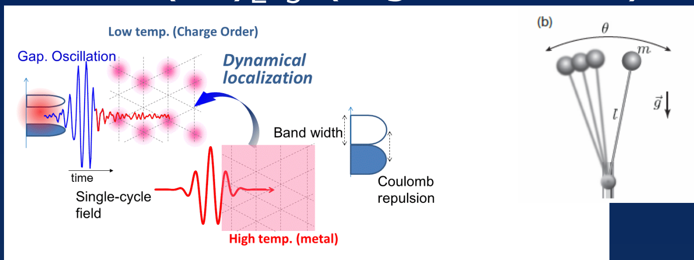
Phys. Rev. Research 4,
（*L*）*032032(2022)*

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

New pathway toward ultrafast control of correlated electrons

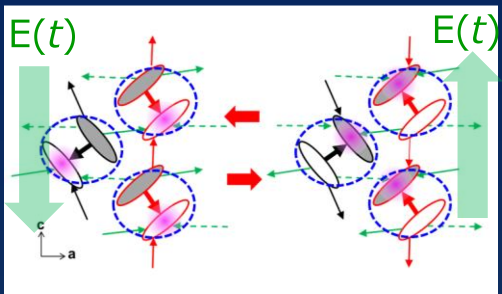
✓ Coherent modulation of electronic states (within scattering time window)

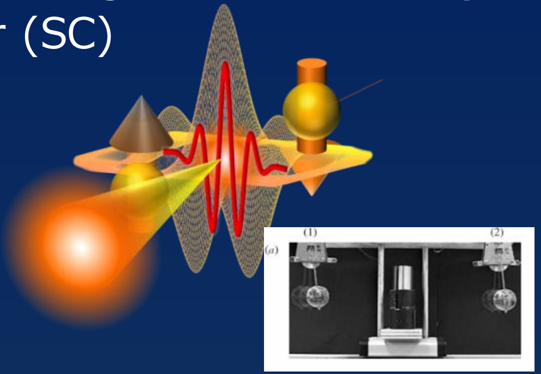
α-$(ET)_2I_3$ (organic metal)



Dynamical localization
Nature commun. 5, 5528(2014)
PRB 2016, *PRB*(R) (2017)
J. Phys. B51, 174005(2018)

κ-$(ET)_2Cu[N(CN)_2]Br$ (SC)

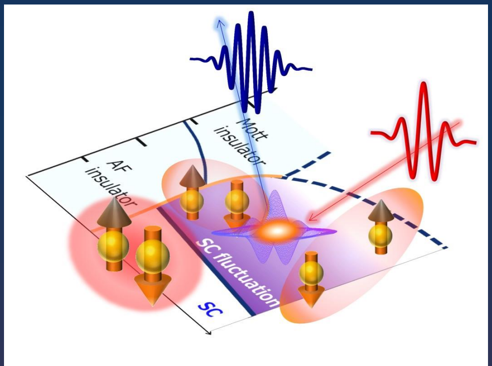




Charge Synchronization
Nature Photon.12, 474 (2018)
Faraday Discussion 237, 353(2022)

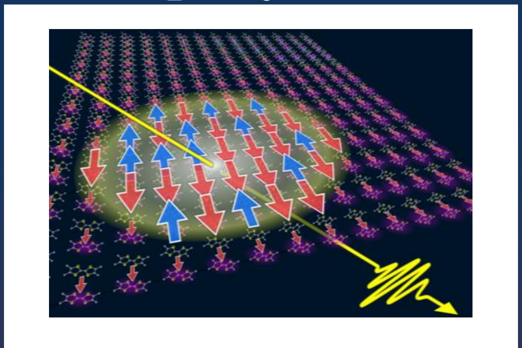
✓ Spatial/ time reversal symmetry breaking

κ-$(ET)_2Cu[N(CN)_2]Br$ (SC)



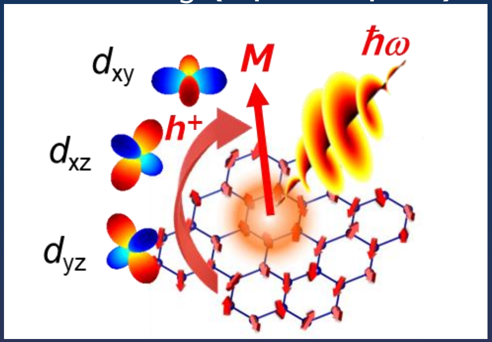
Nonlinear current in organics SC
Nat. Commun.10, 1038 (2020)

$(TMTTF)_2AsF_6$ (Ferroelectric)



Enhance of correlation
Phys. Rev. Res. 3,
L032043(2021)

α-$RuCl_3$ (Spin liquid)



Light-induced magnetization
Phys. Rev. Res. 4,
L032032(2022)

Outline

i) Introduction

- ✓ Coherent charge motion in correlated system
  (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
- ✓ Temperature dependence (anomaly around T_{SC})

Nat. *Photon* 2018

iii) Unconventional SHG in κ-ET salt

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

Nat. Commun. 2020

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

- ✓ Ultrafast magnetization (larger for $T>T_N$)
- ✓ Coherent carrier dynamics & theory

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

v) Summary & inprogress

- ✓ Correlated Dirac semimetal ($SrIrO_3$)
- ✓ Electronic ferroelectricity ,,,,,,,,,,,

Collaborators

Ultrafast & THz spectroscopy

Tohoku Univ.
H. Itoh, Y. Kawakami
T. Amano, Y. Minakami,
R. Kato

Samples and basic measurements

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

Tohoku University K. Ohgushi
Osaka University J. Matsuno
Kyusyu sangyo University T. Nishizaki

Theory

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

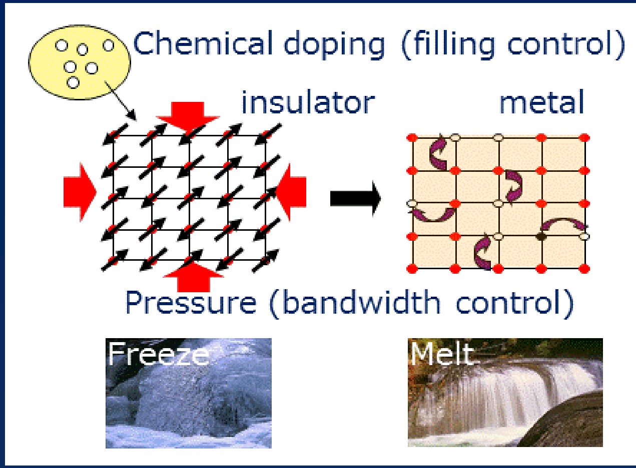
Metal-to-insulator transition

Extended Hubbard model

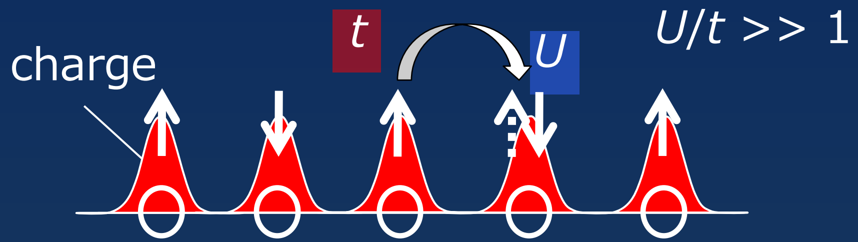
$$H_{\mathrm{EH}} = -\sum_{ij} t_{ij}\left(c_i^{+}c_j + c_j^{+}c_i\right) + \sum_i U_i n_{i\uparrow}n_{i\downarrow} + \sum_{ij} V_{ij} n_i n_j$$

Kinetic energy (transfer integral)

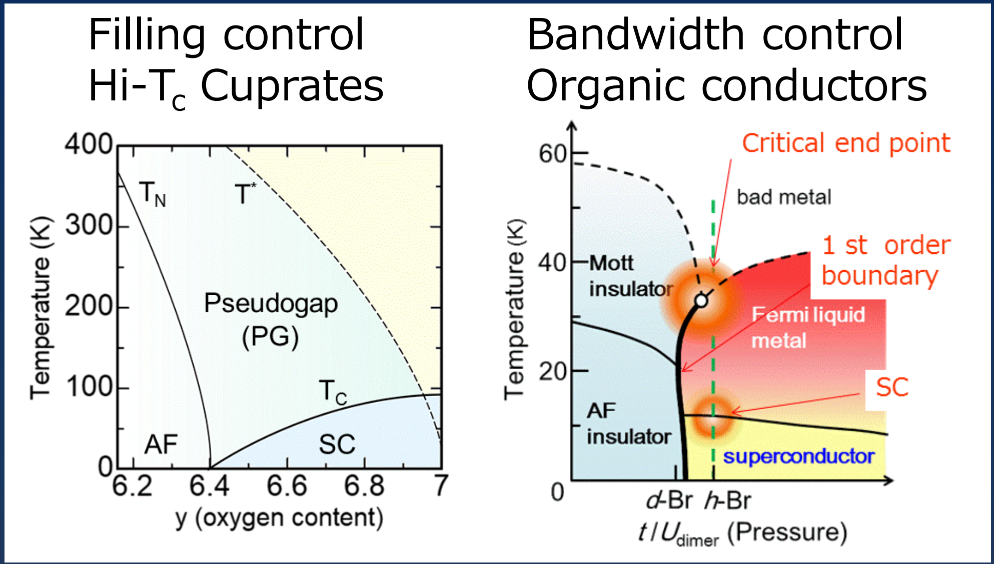
Coulomb repulsion (On-site、inter-site)



1/2 filled (averaged charge=1/site)



Mott insulator



✓ Not only I-M transition, but also FM, FE, SC can be accessed

"Metal-insulator transitions",

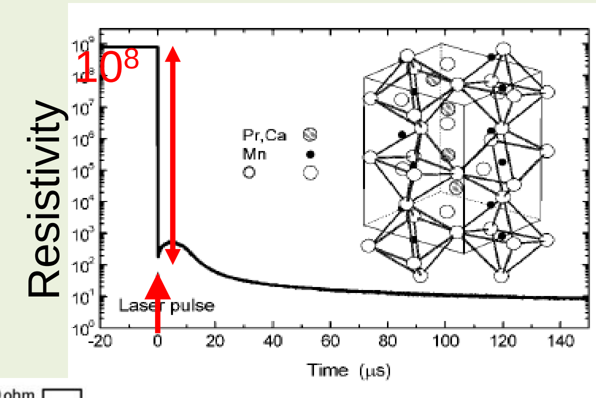
Rev. Mod. Phys. **70, 1039-1263 (1998).**

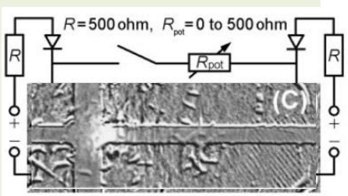
Photoinduced Insulator –Metal transition

25 years ago

Miyano, Tokura *et al*. PRL1997.

(Mn oxides)



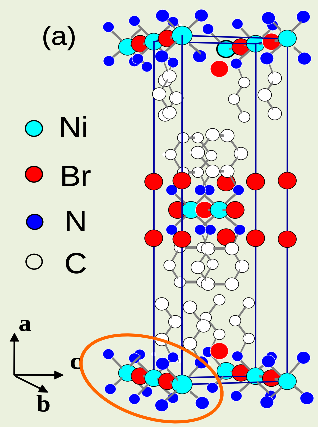


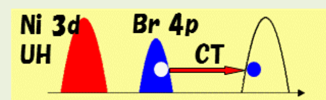
Fiebig *et al*.
Science 280, 1925, 1998

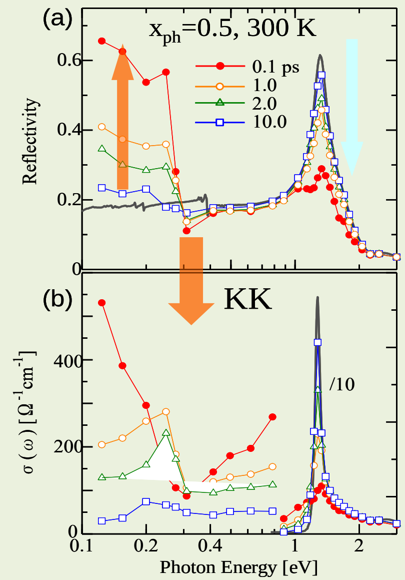
Melting of Charge/Orbital order PM→FM

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

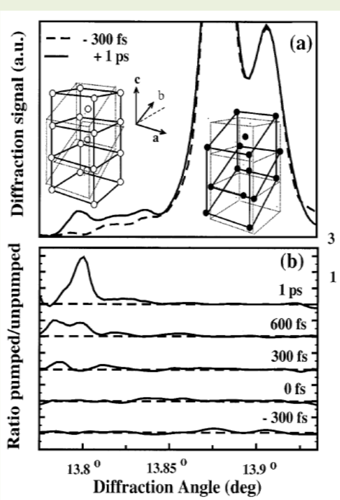
(Ni complex)







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

強相関系の電子コヒーレンスの観測（と操作）

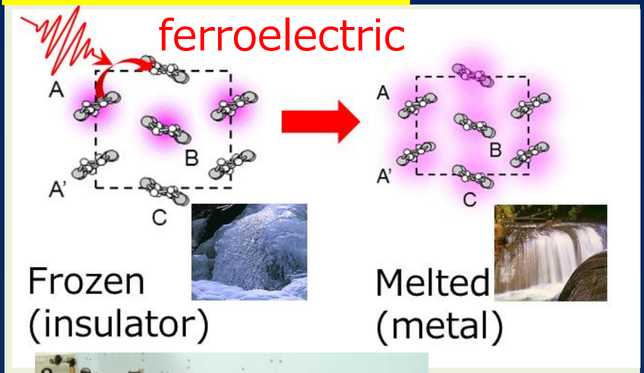
i) 有機物質　電子のコヒーレンス時間　～40 fs（h/(0.

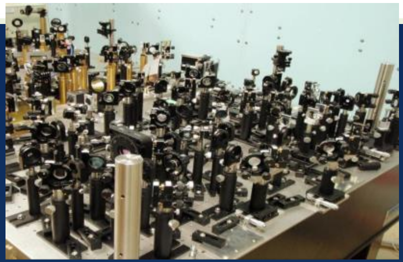
ii) < 10 fs の時間分解測定

CREST(JST)　2008-2013
先端光源を駆使した光科学・光技術の融合展開（伊藤正 総括）
「先端超短パルス光源による光誘起相転移現象の素過程の解明」

Capturing coherent charge oscillation

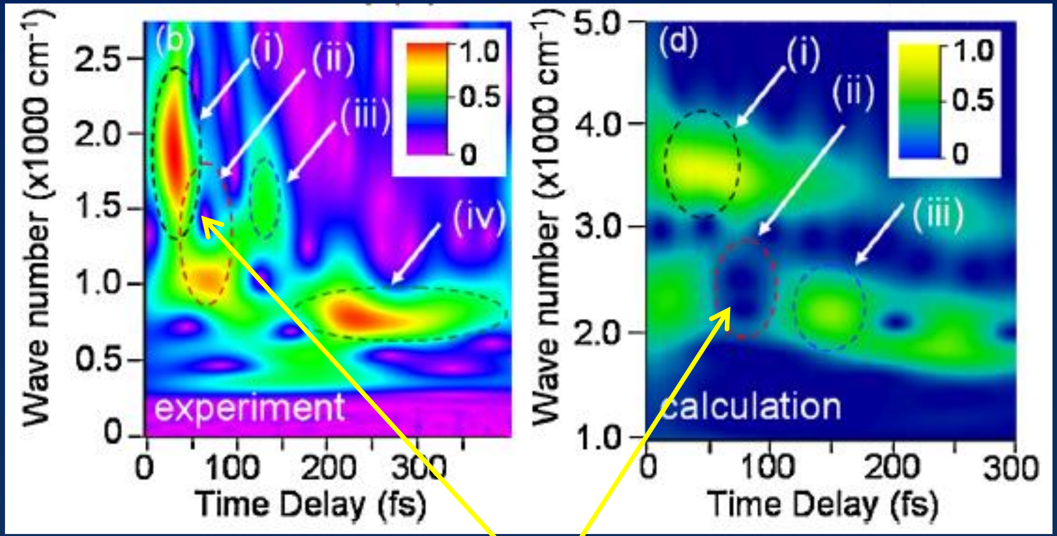
12 years ago





OPA @1.6μm

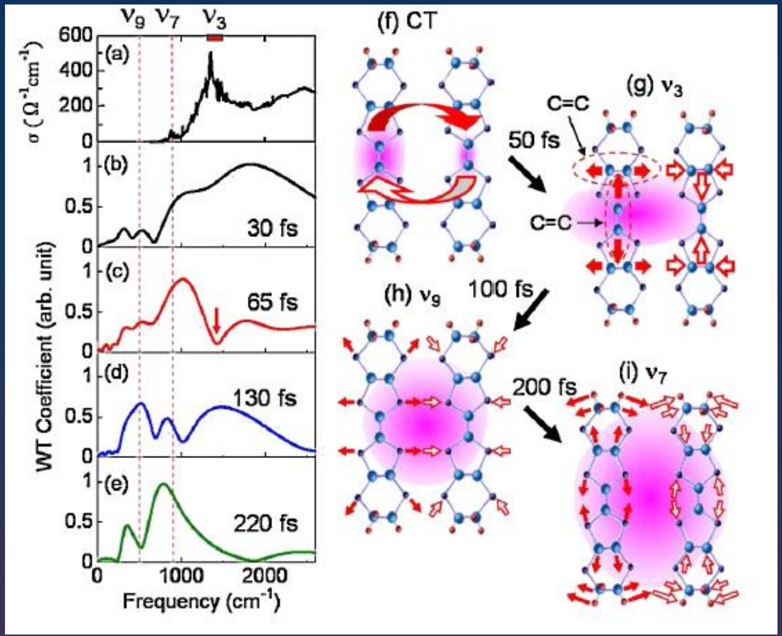
10 fs spectroscopy + quantum calculation

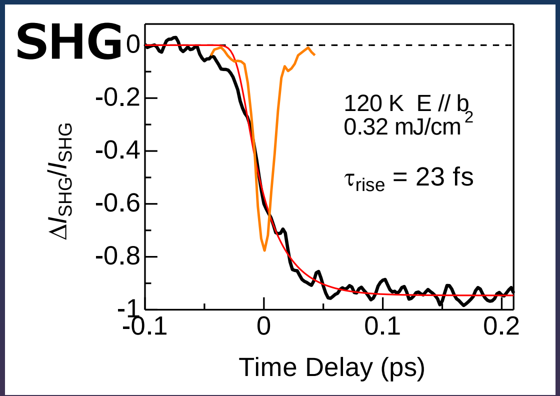


Fano interference

Coherent charge oscillation (T=18 fs)
(dephasing ～ 40 fs)

Phys. Rev. Lett. 98, 097402 (2007)
Phys. Rev. Lett. 105, 246402 (2010).

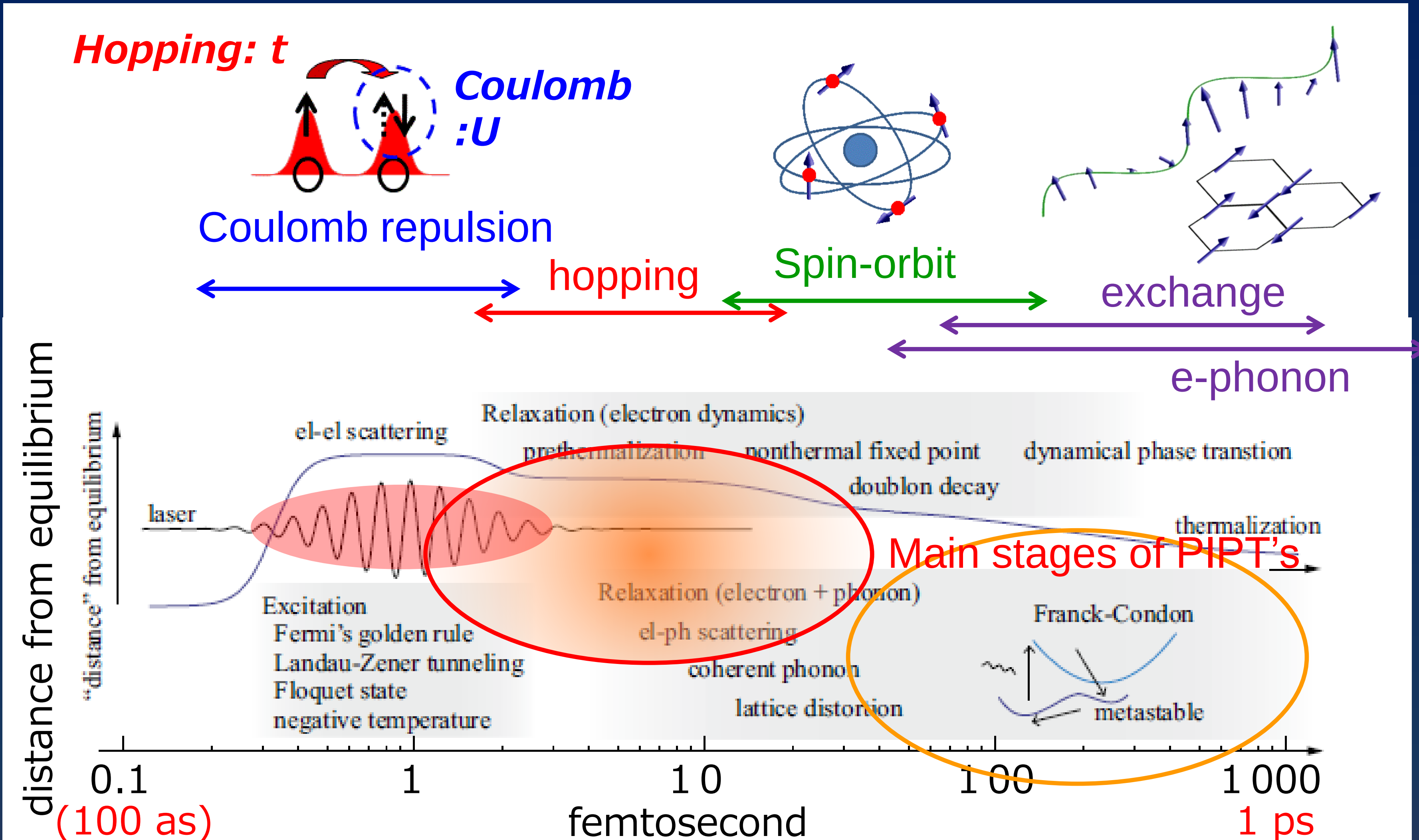




Ferroelectric nature disappears in 20 fs < hopping

(using 7 fs pulse)

Photo-excited states in correlated system

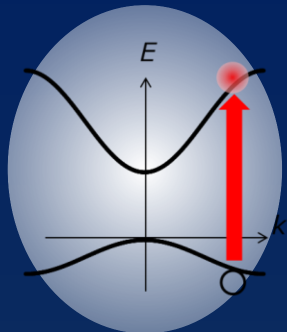


Aoki, Tsuji, Eckstein, Oka, Werner *Rev. Mod. Phys*. 86, 779 (2014).

✓ **Electronic coherence survives (depends on material)**

✓ **Thermalization is not completed**

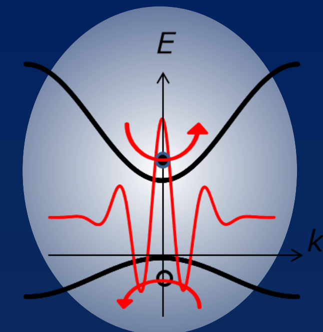
PHz charge motion deiven by PHz field



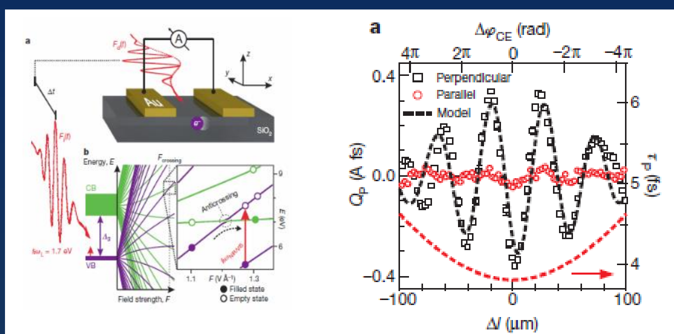
Paradigm shift toward quantum manipulation

Position, Density ➡ **Phase**

~100 attosecond $\Delta t = \Delta E / \hbar$ **~eV**

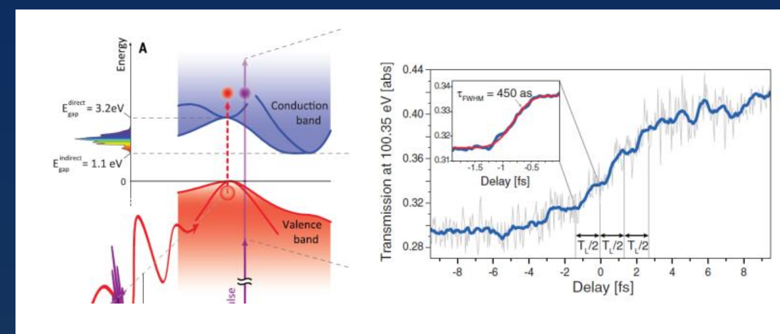


Attosecond transient current (SiO_2)

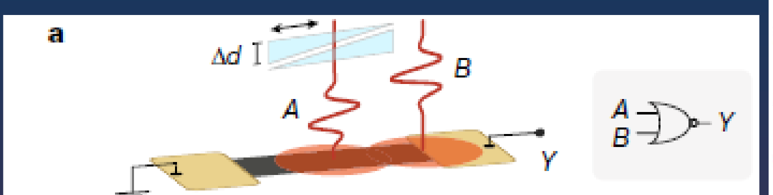


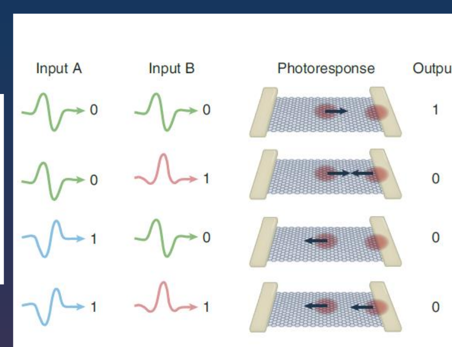
Schiffrin, Krauszet al., *Nature* 2013

Attosecond XUV spectroscopy (Si)



Schulltze et al.,*Science* 2014



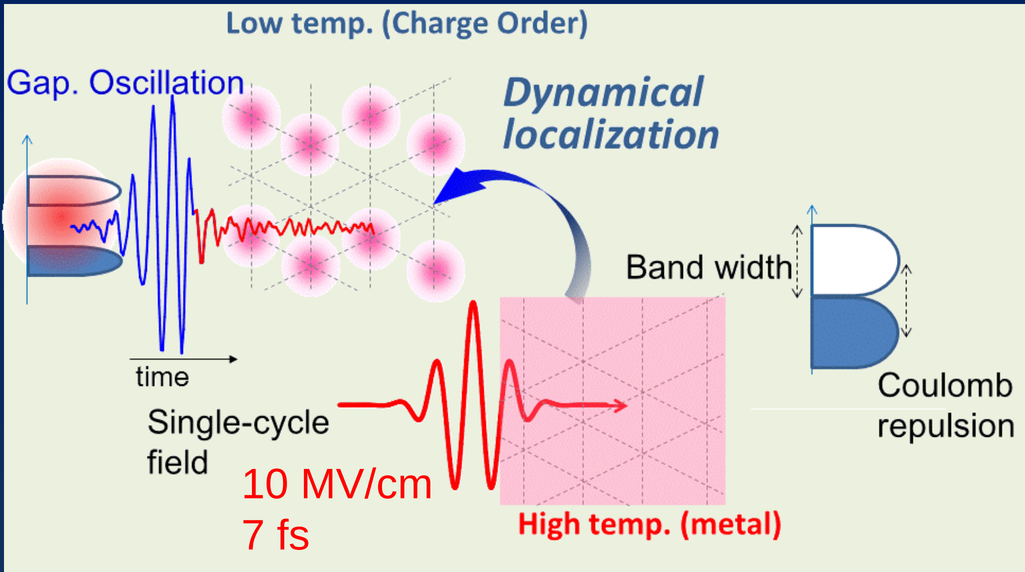


PHz logic gate
(interference: real & virtual)

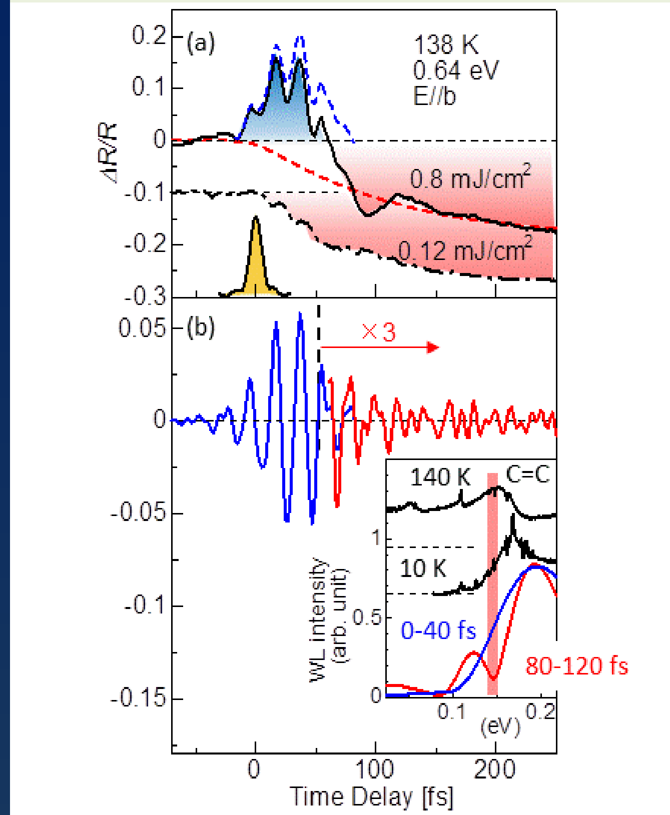
T. Boolakee et al.,
Nature 605, 251(2022)
K-J Tielrooij Nature Mat.

Semiconductors, Graphene (e-e interaction is small)

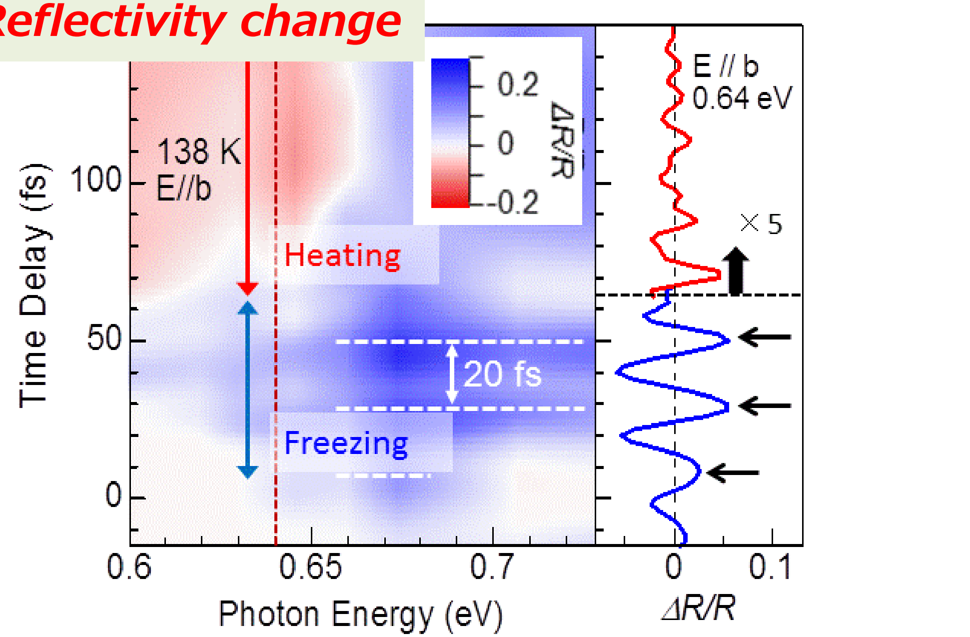
Optical freezing of charge motion



Insulating gap in time axis



Reflectivity change



Ishikawa *et al., Nature commun*. 2014

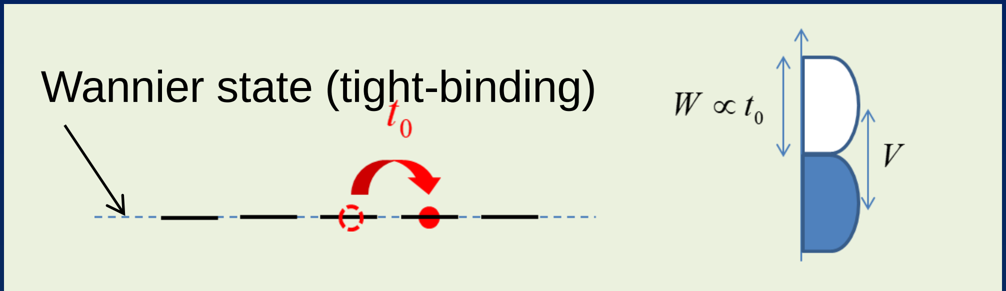
Nature commun. 5, 5528(2014)

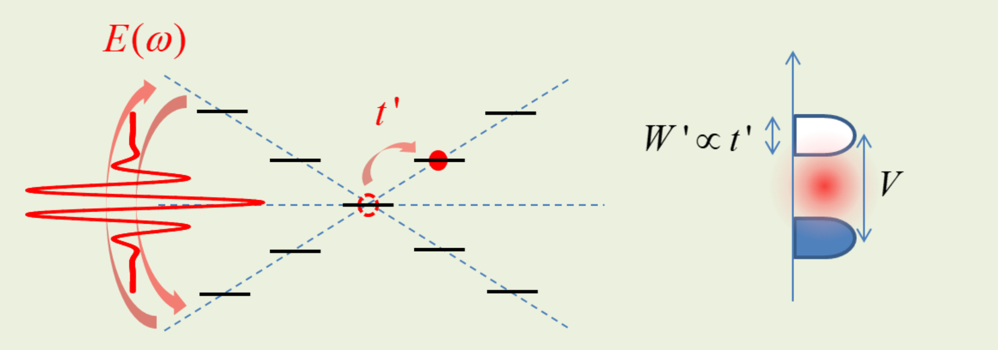
PRB 93, 165126 (2016)

PRB 95, 201106(R) (2017)

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

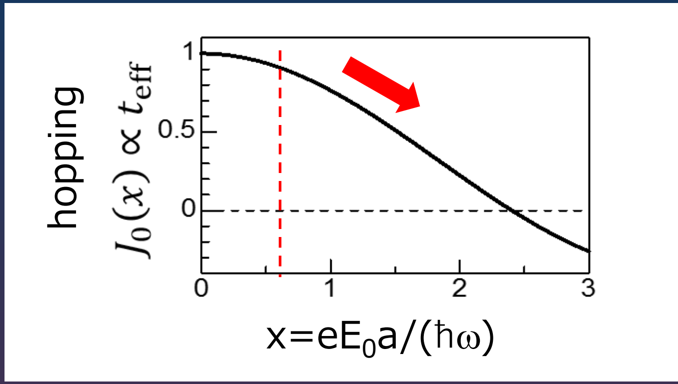
Dynamical localization

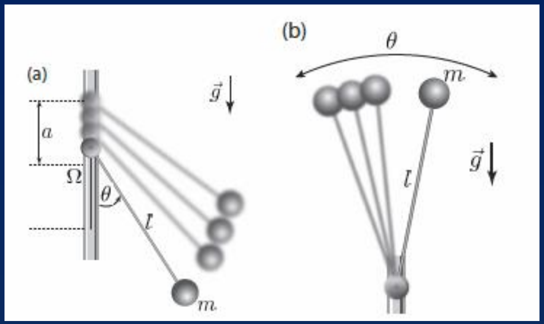




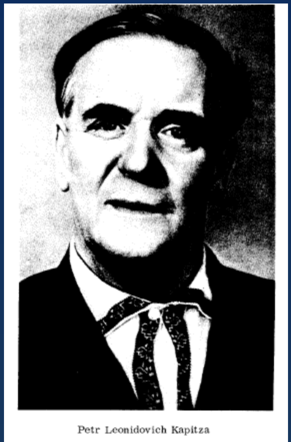
High-frequency strong field
disturbs hopping (ω=0；stark ladders)

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





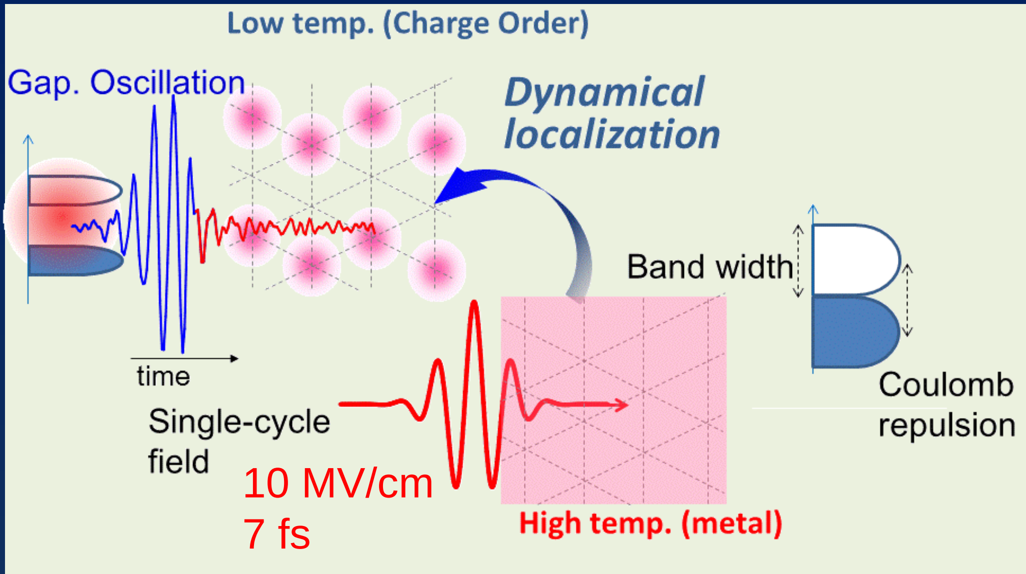
Dynamical stabilization
M. Bukovet al.,
Advances in Physics, **64**, 139(20



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

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

Optical freezing of charge motion



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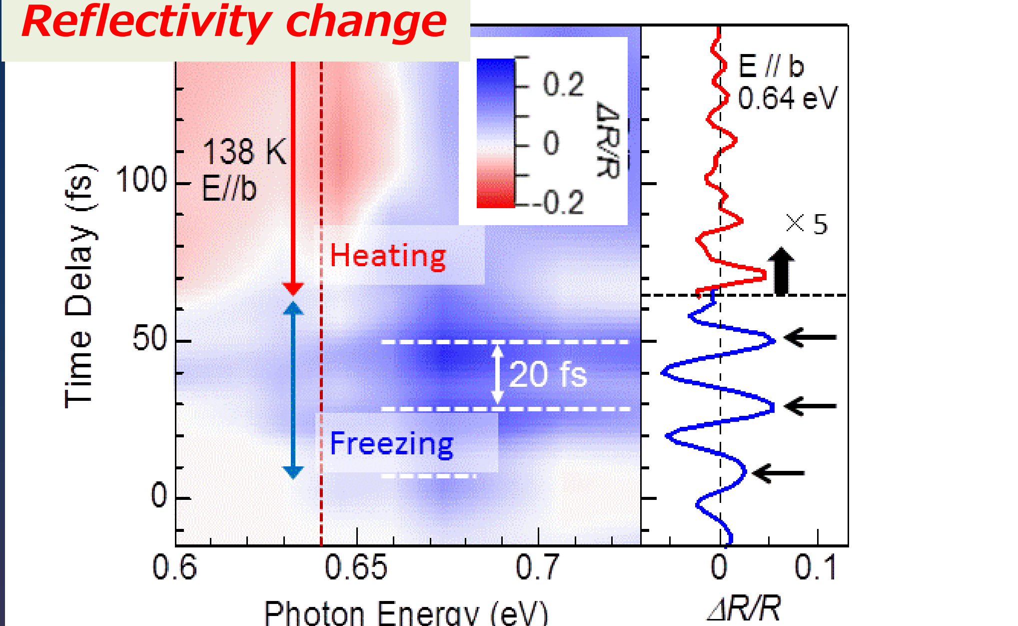

Ishikawa *et al., Nature commun*. 2014

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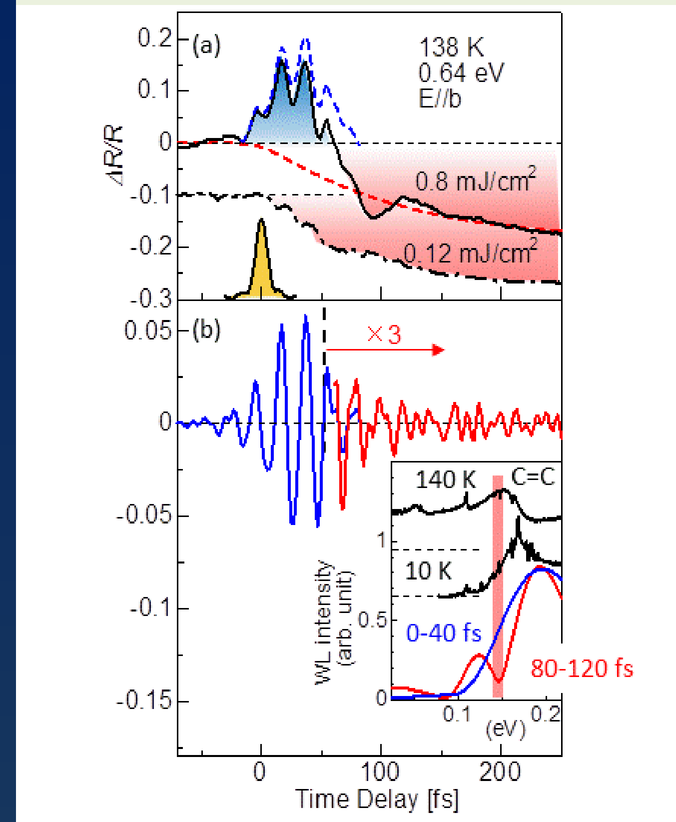

Nature commun. 5, 5528(2014)

PRB 93, 165126 (2016)

PRB 95, 201106(R) (2017)

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

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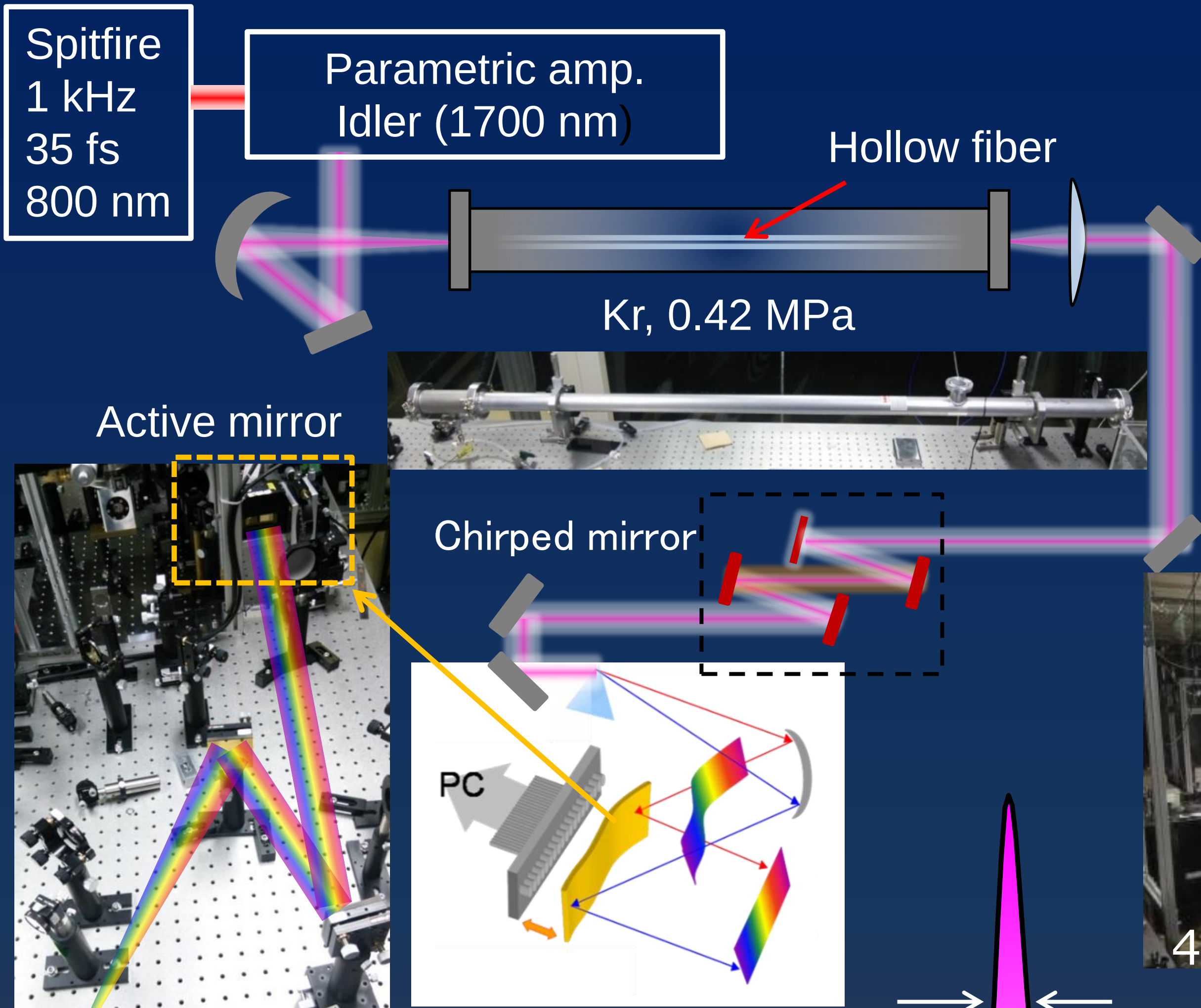
Phys. Rev. Res. (L) 4, L032032(2022)
arXiv: 2207.03877

v) Summary & inprogress

- ✓ Correlated Dirac semimetal ($SrIrO_3$)
- ✓ Electronic ferroelectricity ,,,,,,,,,,,

6 fs NIR pulse (1.3 cycle, CEP stabilized)



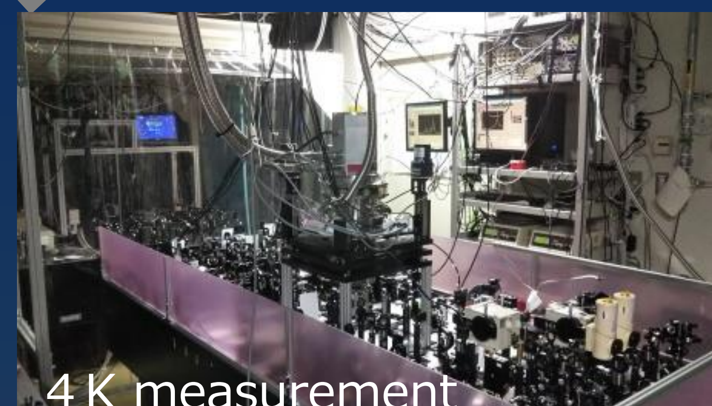
✔ **OPA Idler ($\chi^{(2)}$)**

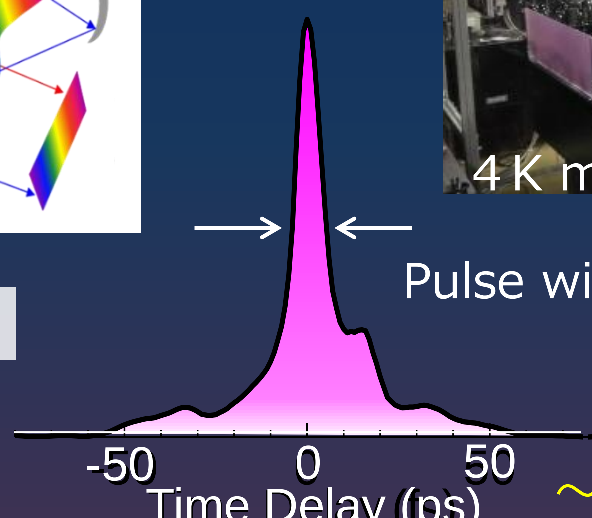
$$\varphi_{Idler} = -\frac{\pi}{2} + \varphi_{Pump} - \varphi_{Signal}$$

✔ **SPM($\chi^{(3)}$)**

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

$\rightarrow$ CEP stable





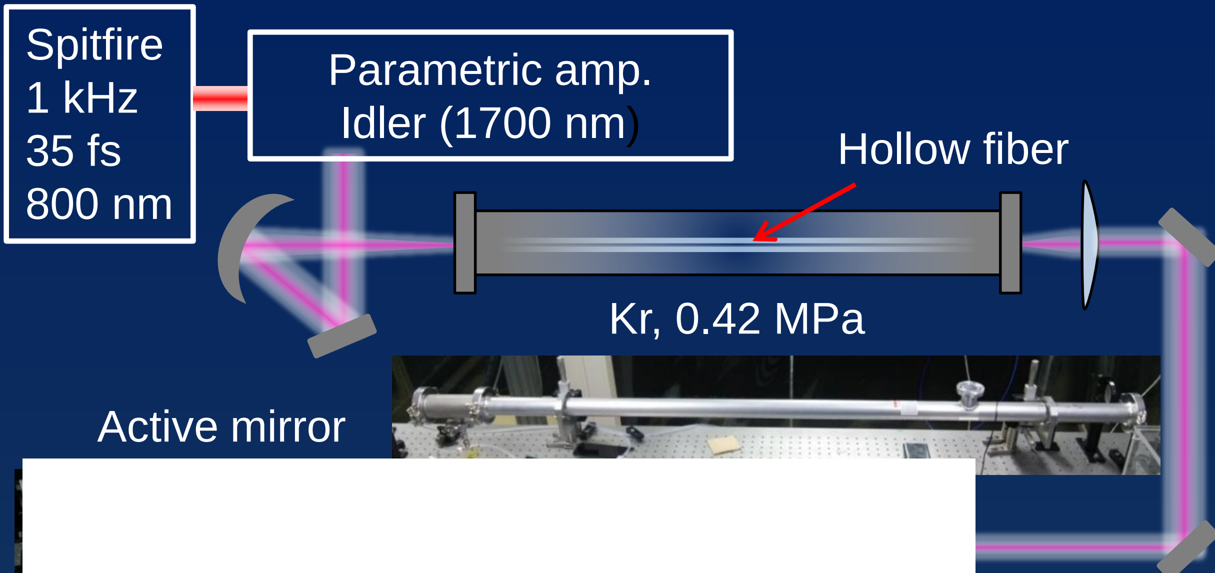
Pulse width =6 fs

30 μJ/pulse
Φ200 μm

J. Phys. B51, 174005(2018)
(invited paper)

～20 MV/cm for organics

6 fs NIR pulse (1.3 cycle, CEP stabilized)



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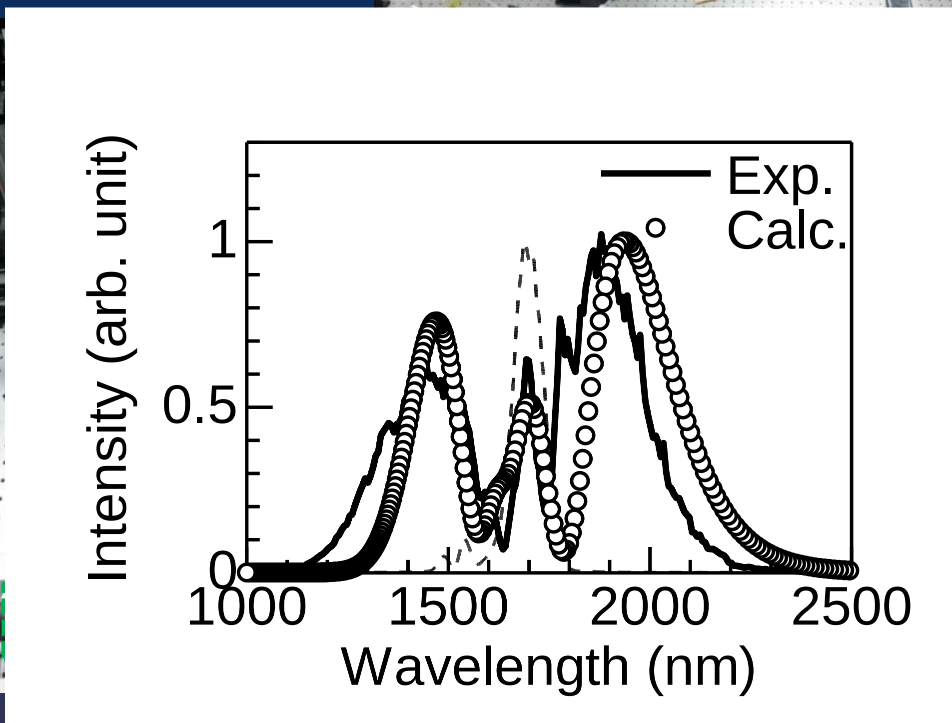
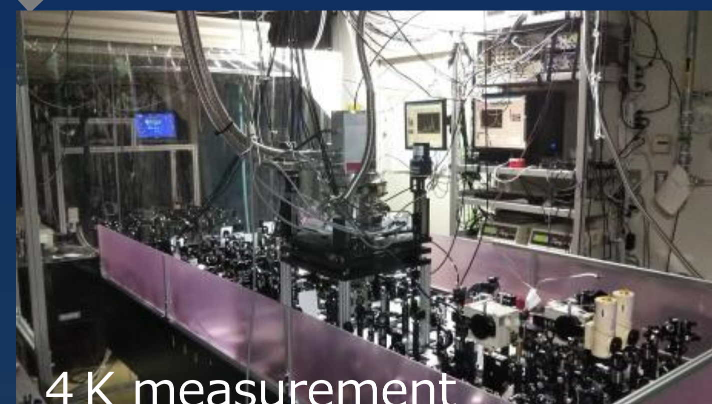
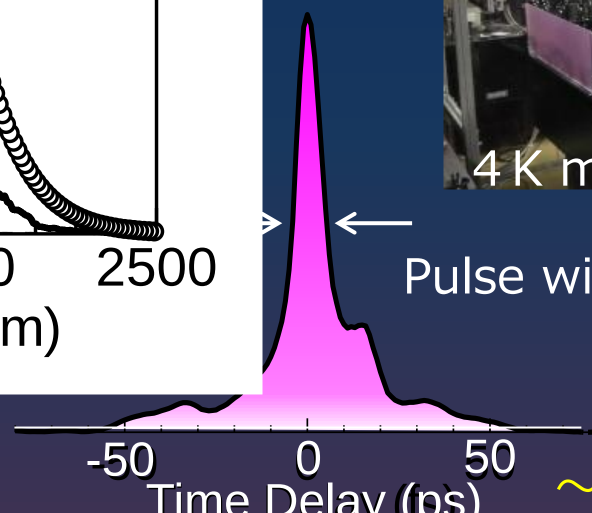






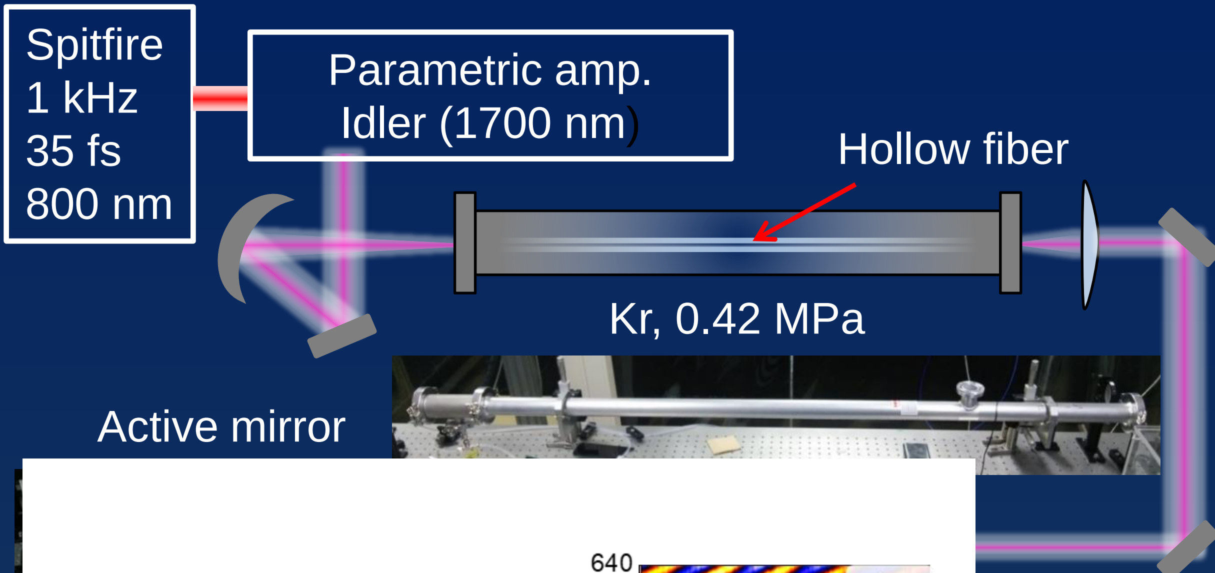
Pulse width =6 fs

30 μJ/pulse
Φ200 μm

J. Phys. B51, 174005(2018)
(invited paper)

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

6 fs NIR pulse (1.3 cycle, CEP stabilized)



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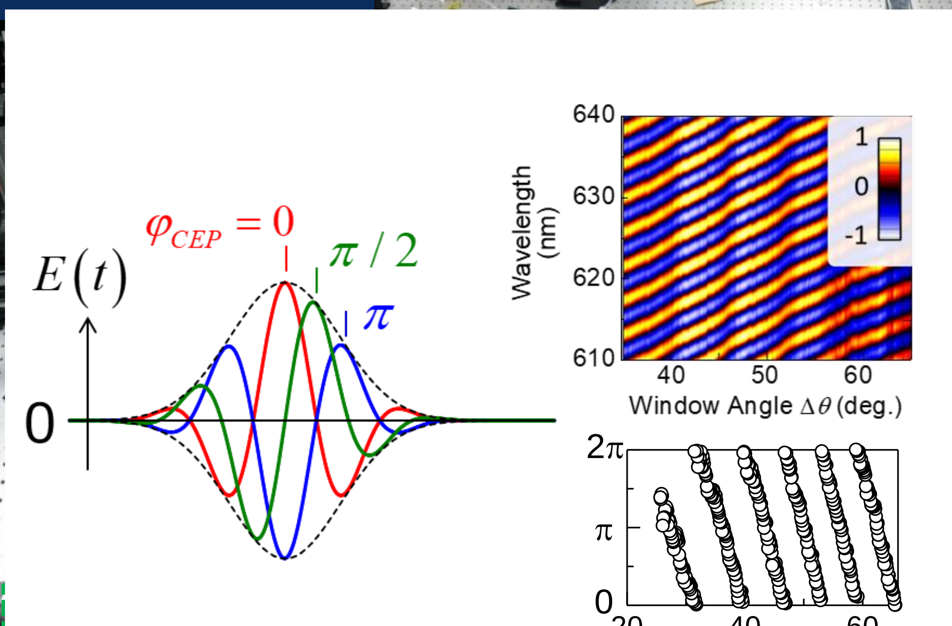
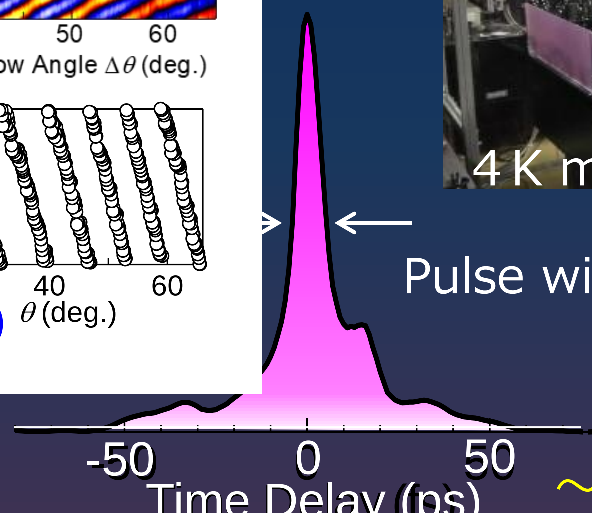


Faraday Discussion 237353(2022)
レーザー研究, 50, 313 (2022)



Pulse width =6 fs

30 μJ/pulse
Φ200 μm

J. Phys. B51, 174005(2018)
(invited paper)

～20 MV/cm for organics

Outline

i) Introduction

- ✓ Coherent charge motion in correlated system
  (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
- ✓ Temperature dependence (anomaly around T_{SC})

Nat. Photon 2018

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

Nat. Commun. 2020

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

- ✓ Ultrafast magnetization (larger for $T>T_N$)
- ✓ Coherent carrier dynamics & theory

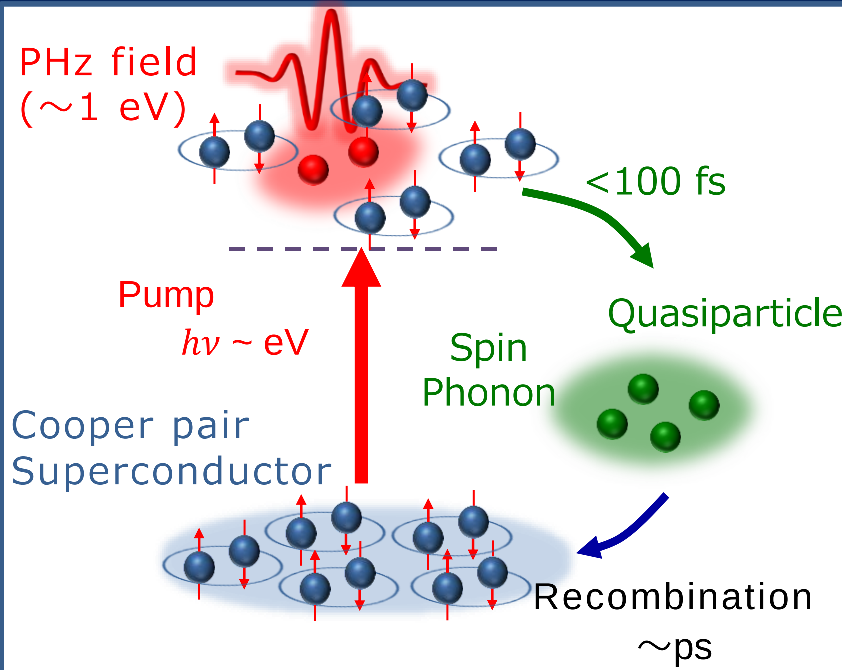
Phys. Rev. Res. (L) 4, L032032(2022)
arXiv: 2207.03877

v) Summary & inprogress

- ✓ Correlated Dirac semimetal ($SrIrO_3$)
- ✓ Electronic ferroelectricity ,,,,,,,,,,,

Excitation of superconductors

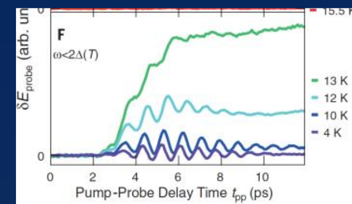
Excitation by light (NIR-Visible)
→ rise of electron temp. (scattering)

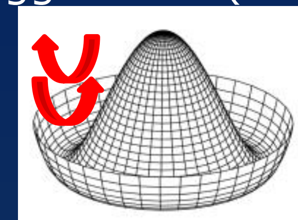


Giannetti, Mihailovic et al., Advances in Physics, **65**, 58-238(2016).

✓ Avoid increasing temp
→low energy excitations

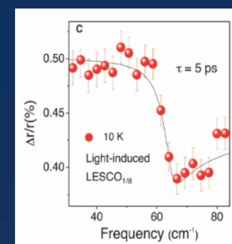
- Coherent exc. of Higgs mode (BCS)

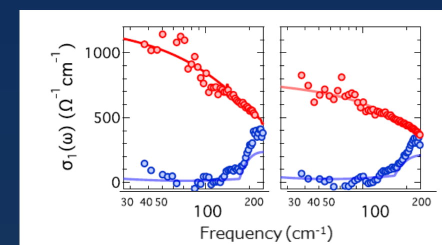




Matsunaga, Shimano et al. Science 2014.

- Photoinduced SC Cuprates, Organics





D. Faustii et al., Science 2011

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

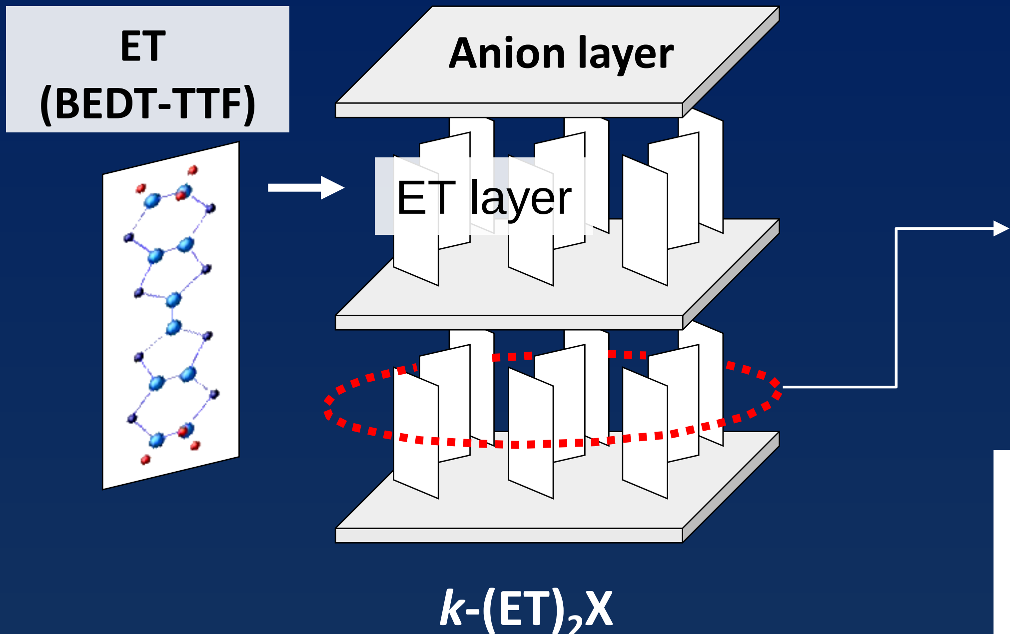
✓ Another approach

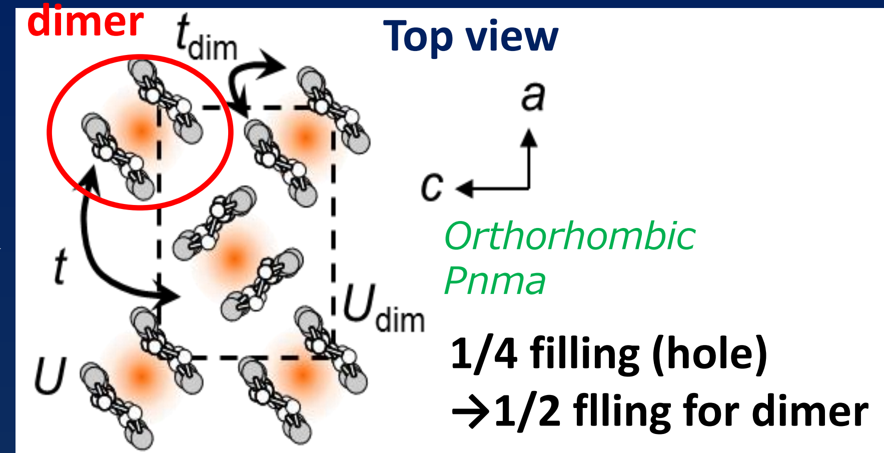
e-e scattering time

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

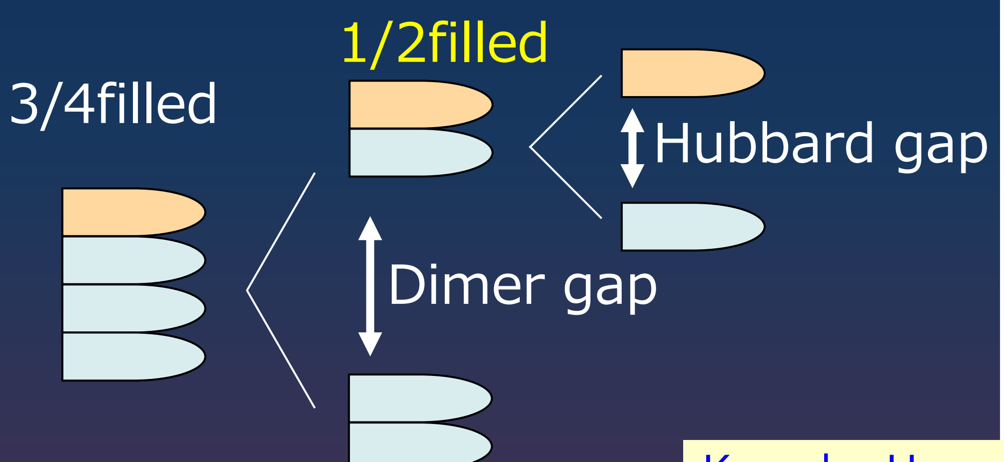
- **6 fs pulse can control electrons in no-scattering time window ?**

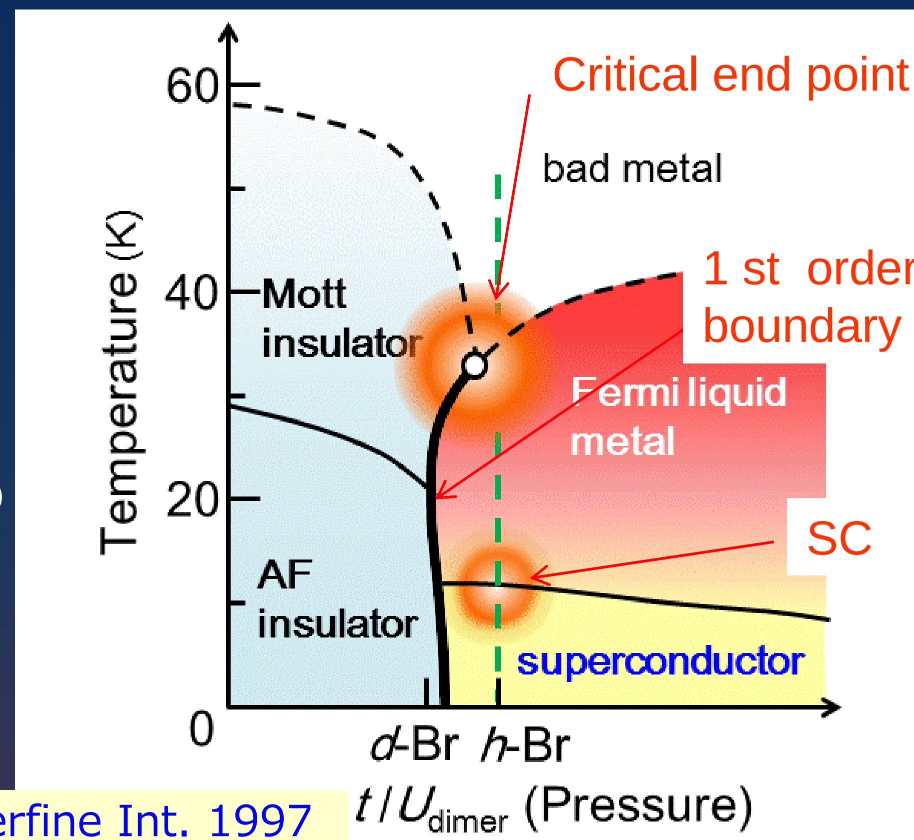
Organic superconductor κ-$(ET)_2Cu[N(CN)_2]Br$





Dimer Mott insulator



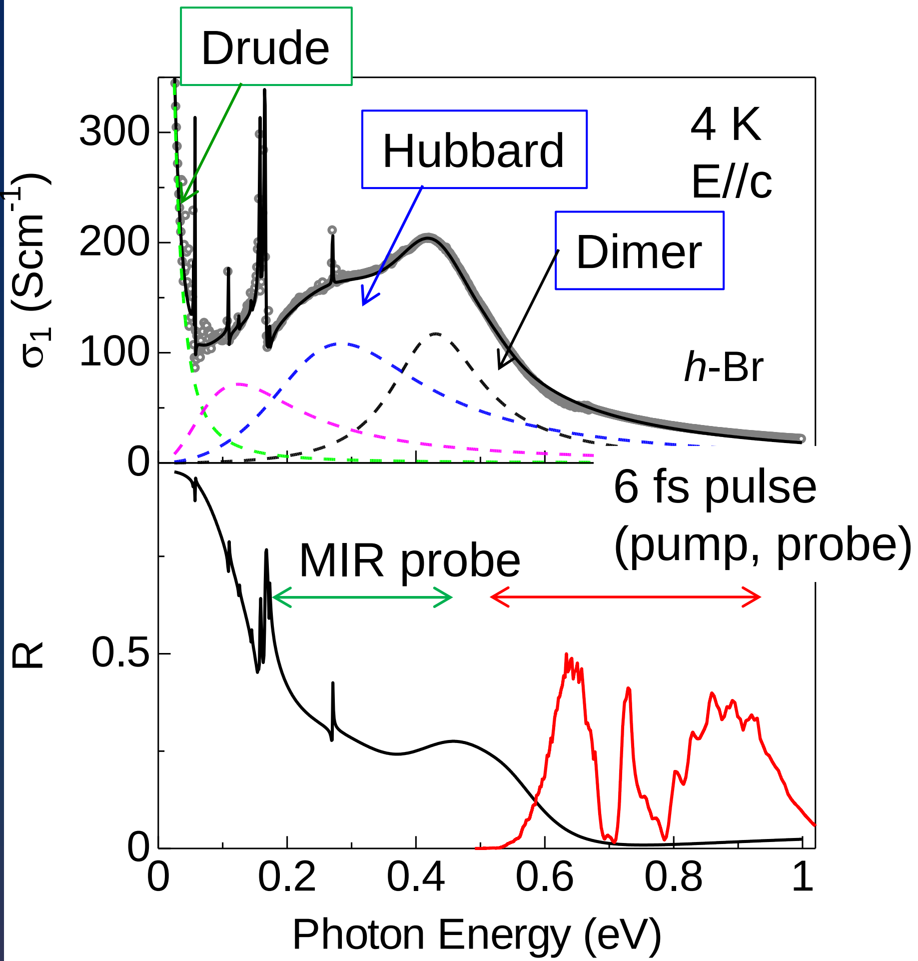


Kanoda, Hyperfine Int. 1997

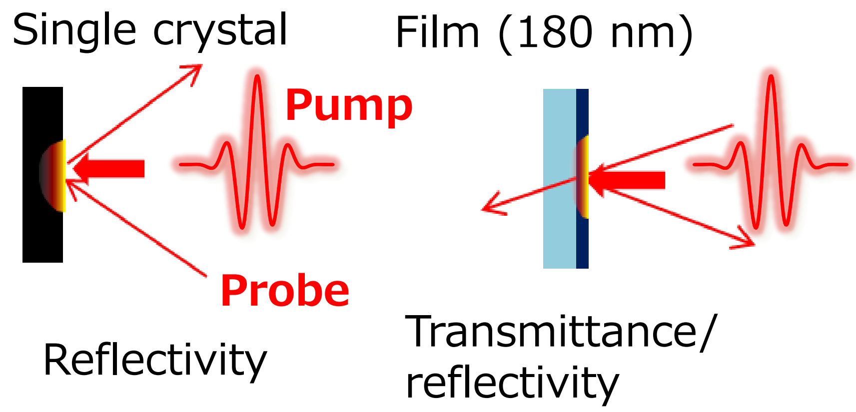
κ-(h-ET)$_2$Cu[N(CN)$_2$]Br (h-Br)

Reflectivity/Optical conductivity (h-Br)

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

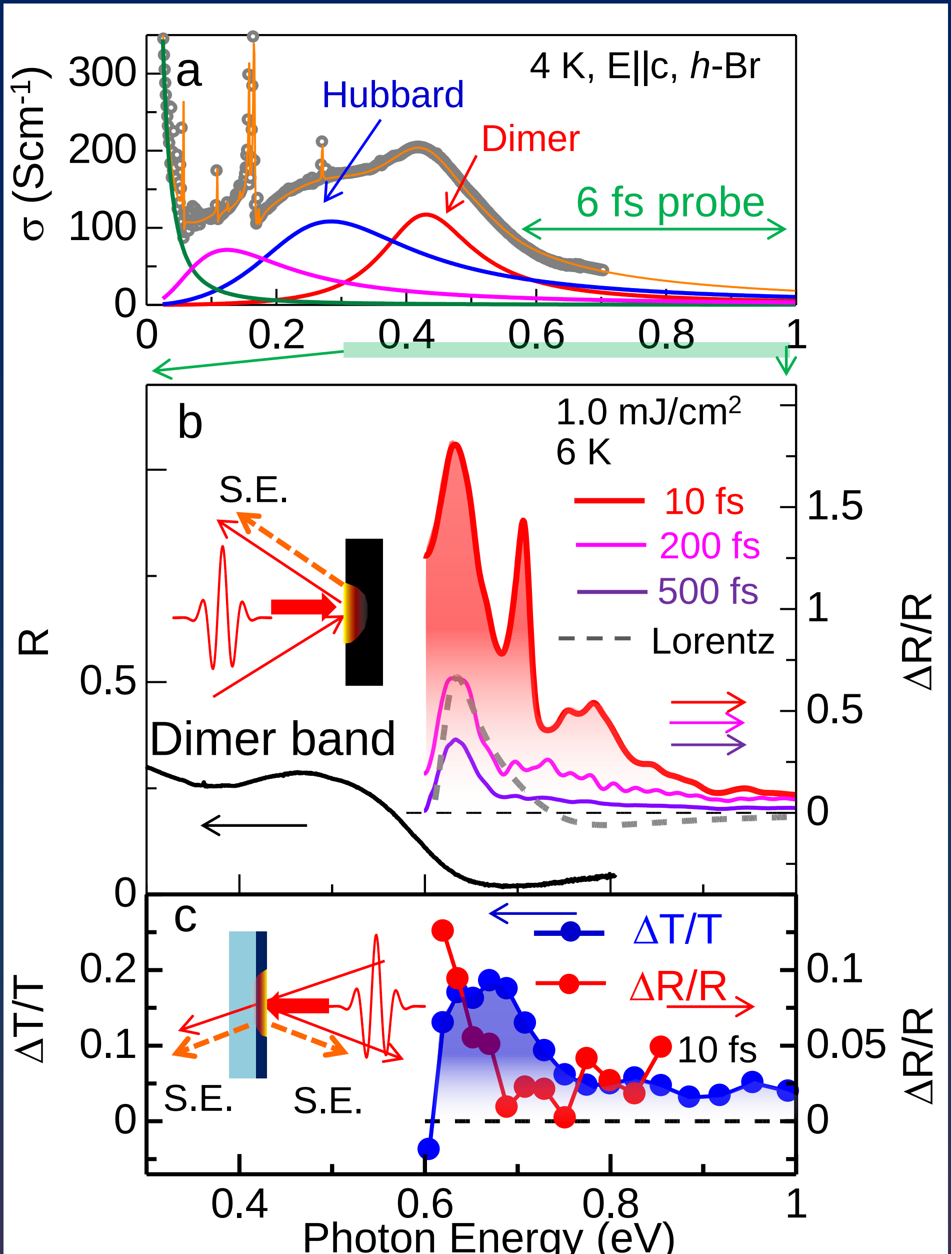


Pump-probe(2-or 3- beam)



Pump/Probe∼ 1000/1

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



Nat. Photon 12, 474 (2018)

✓ Same-color pump & probe

✓ New reflectivity peak
0.63 eV, at 10 fs (FID at 0.7 eV)
~200 % reflectivity increase !

✓ Lorentz analysis

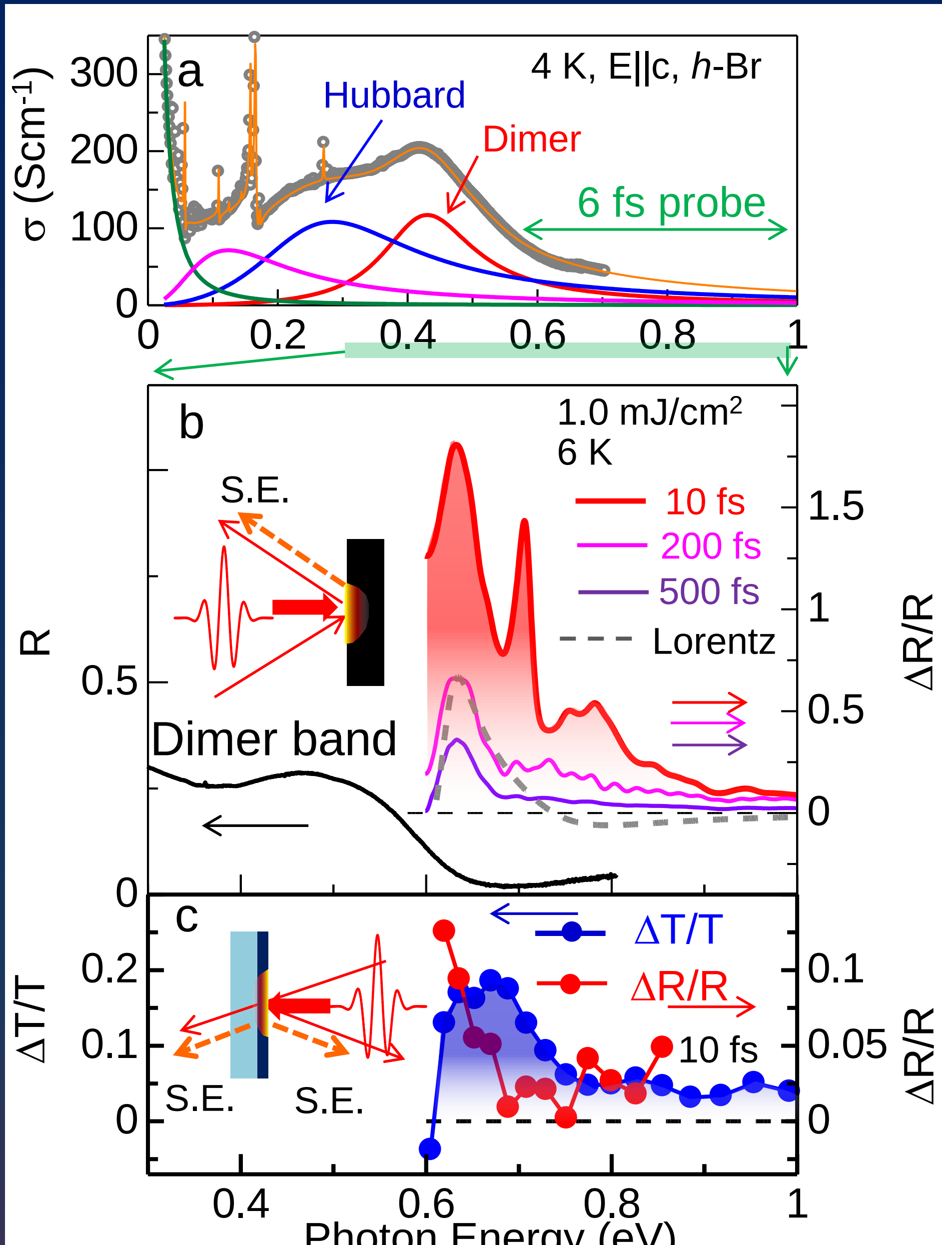
Additional oscillator
frequency: 0.61 eV
damping: 0.04 eV

✓ Thin film (180 nm on CaF_2)
H. Yamamoto et al.,
Nature commun. 2013

Increases of
transmittance and reflectivity

→ **Stimulated emission**
(Peak energy > dimer band)

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



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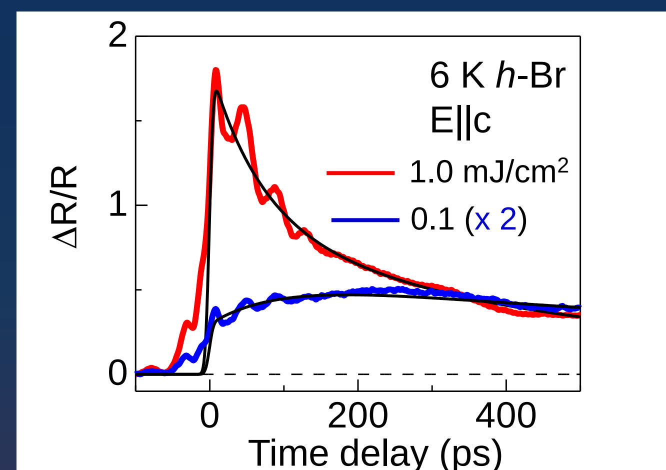
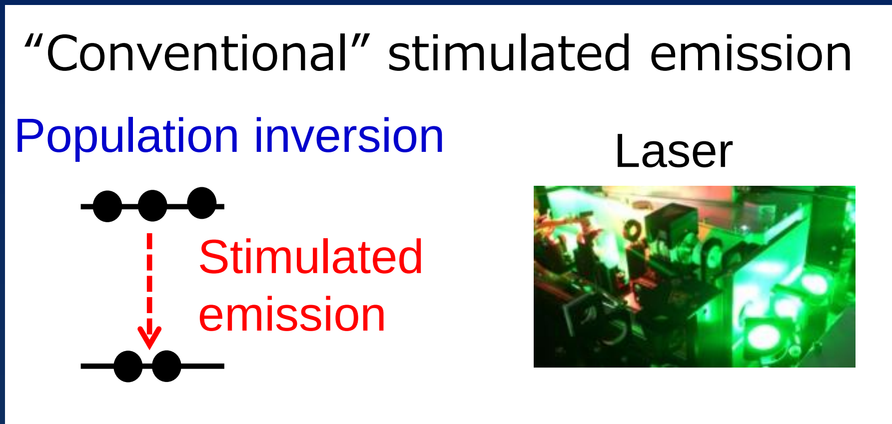
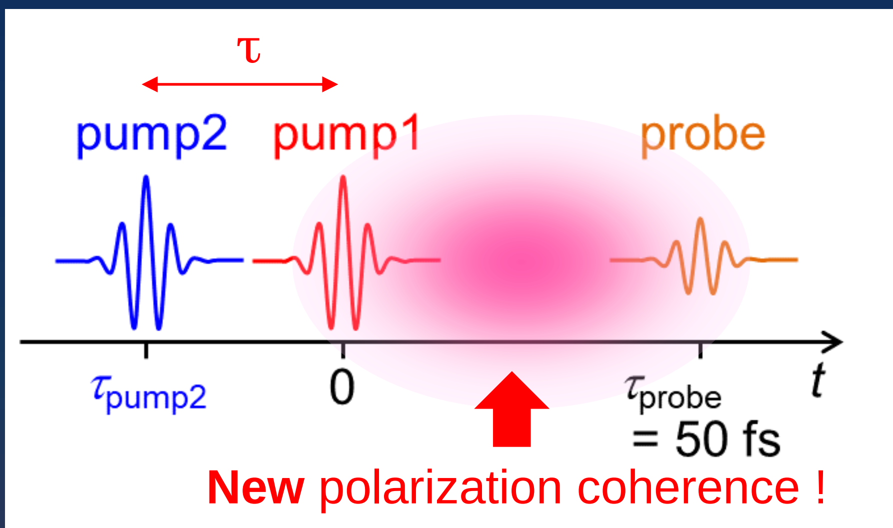


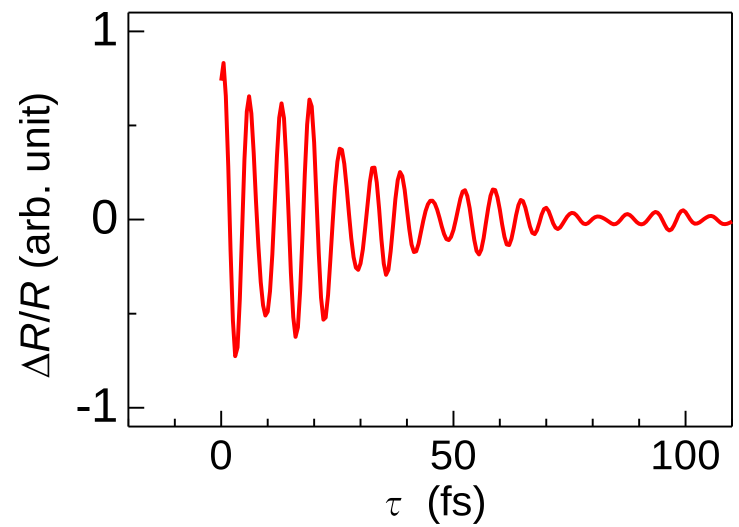
Fast (65%) rise <10 fs,
decay 70 fs

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)

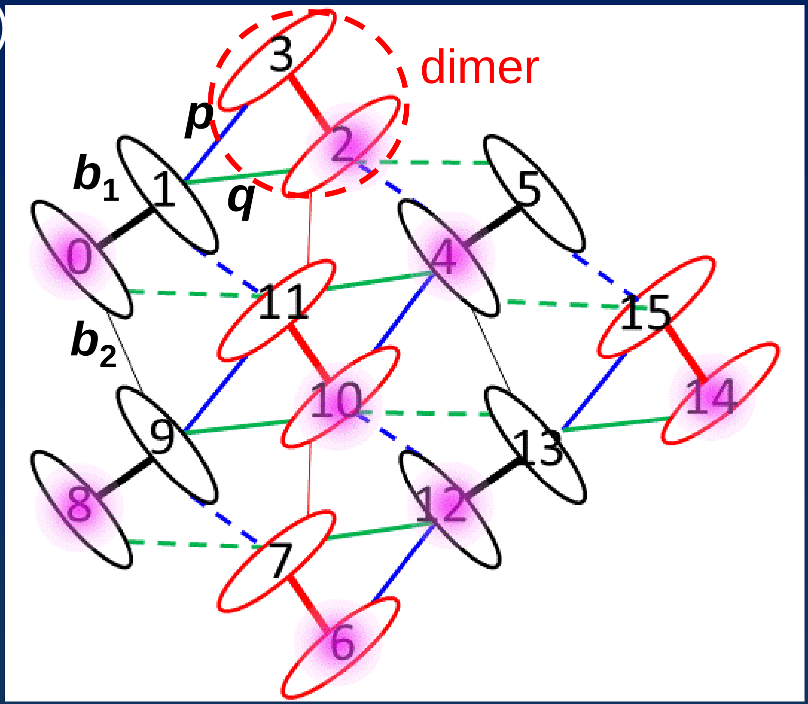
✓ Decay time of SE (70 fs)
is approximately equal to the
electronic coherence time

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)



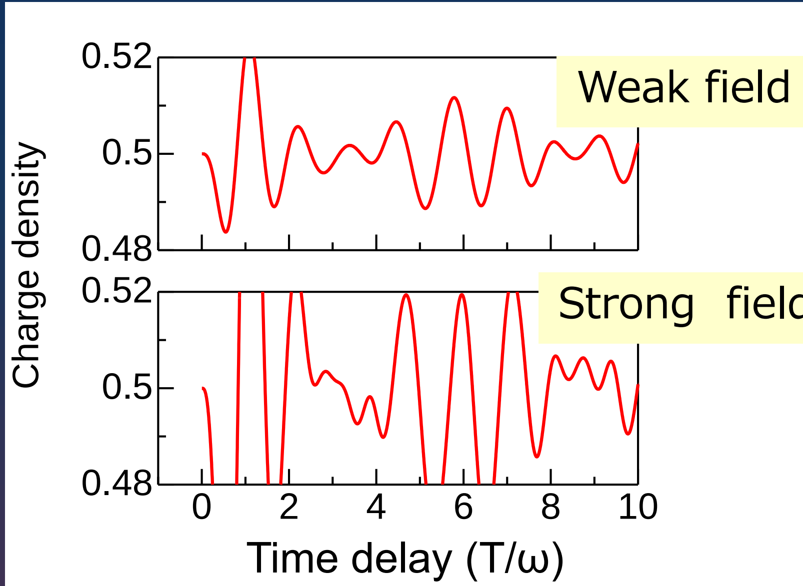
$$H_{2D} = \sum_{\langle i,j\rangle\sigma} t_{ij}\left(c^{+}_{i,\sigma}c_{j,\sigma} + c^{+}_{j,\sigma}c_{i,\sigma}\right) + U\sum_{i} n_{i\uparrow}n_{i\downarrow} + \sum_{\langle i,j\rangle} V_{ij}n_in_j$$

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

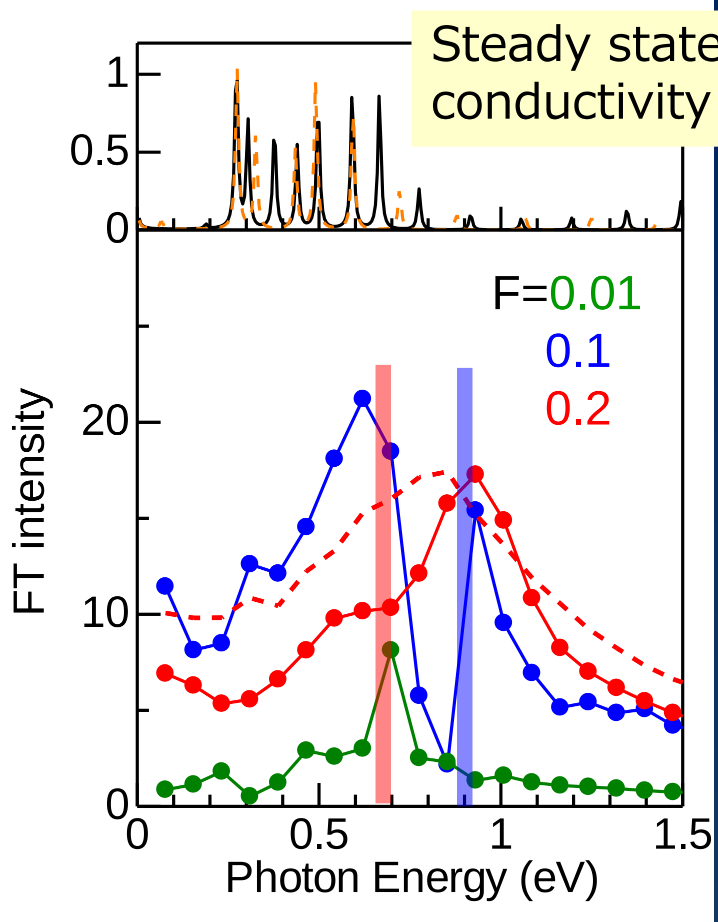
$$\boldsymbol{A}(t) = \frac{c\boldsymbol{F}}{\omega}\left[\cos(\omega t) - 1\right]\theta(t)\theta\left(\frac{2\pi}{\omega} - t\right)$$

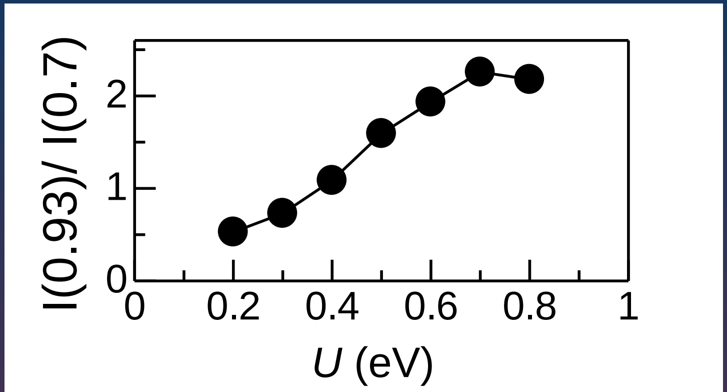


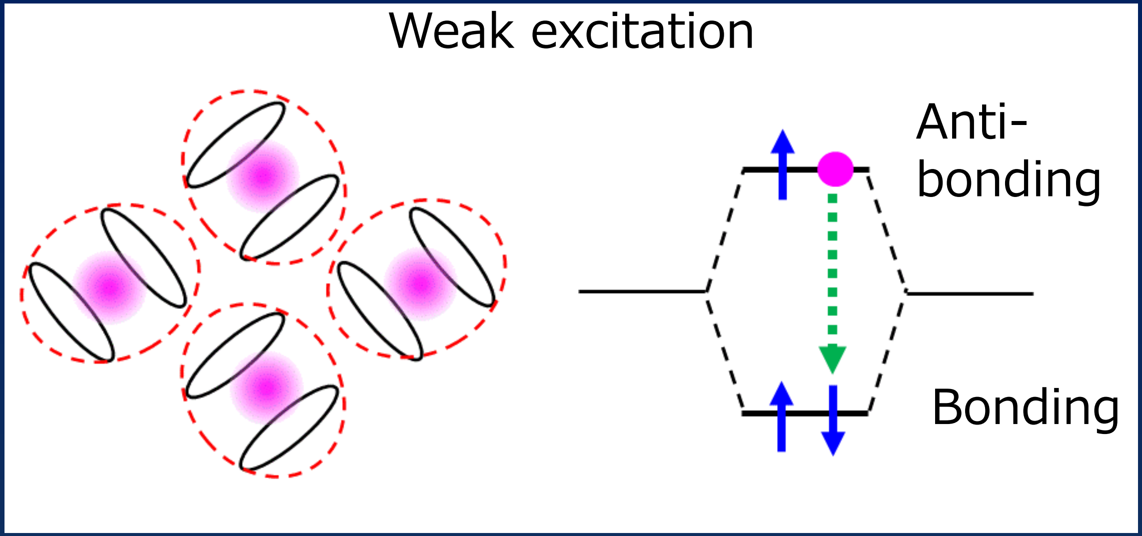
Time evolution of charge density on a molecule

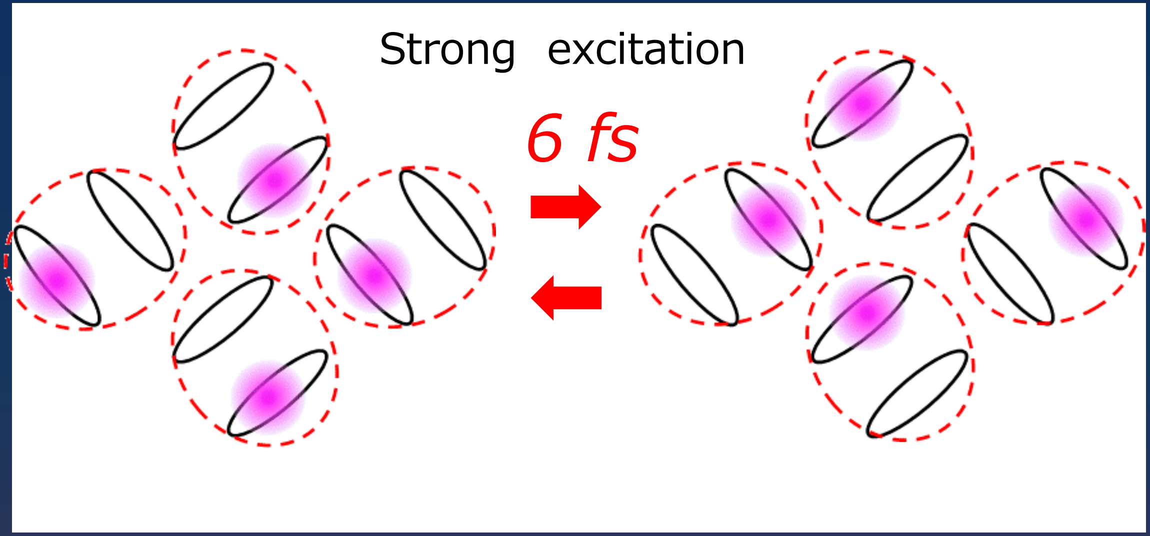
Yonemitsu, JPSJ87, 044708(2018).

U dependence of non-linear charge motion









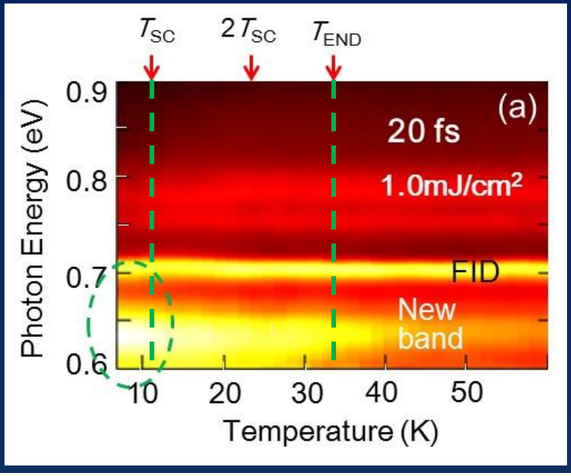
Non-linear oscillation is sensitive to *U* !

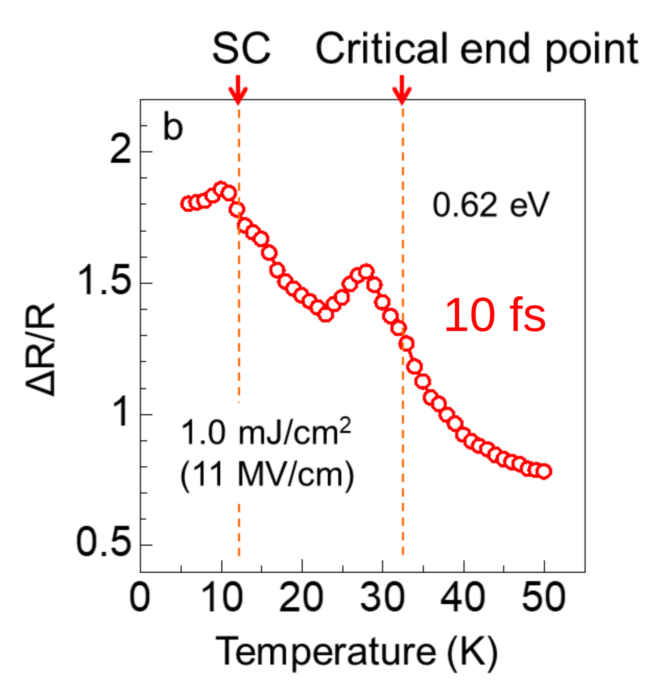
Anomaly at T_{SC}

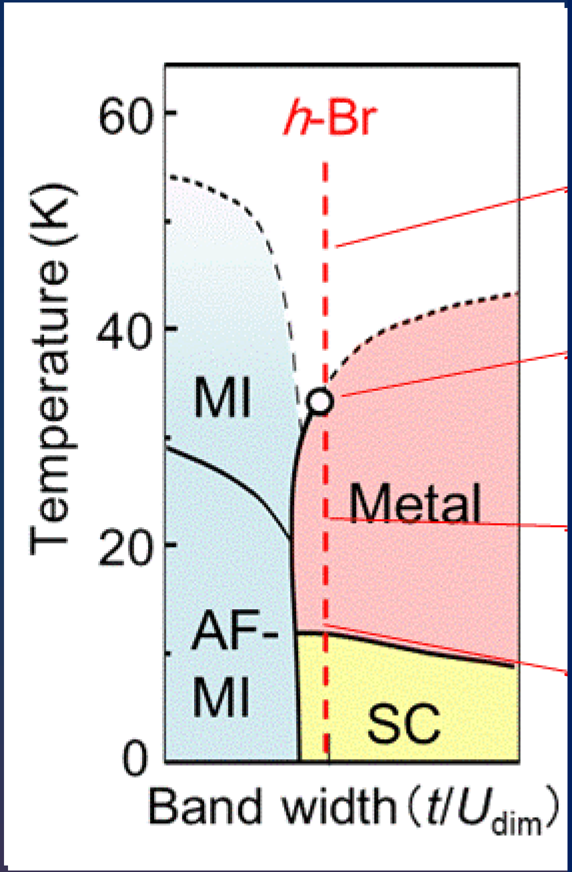
✓ Gradual increase from 20 K ~2 T_{SC}

✓ 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

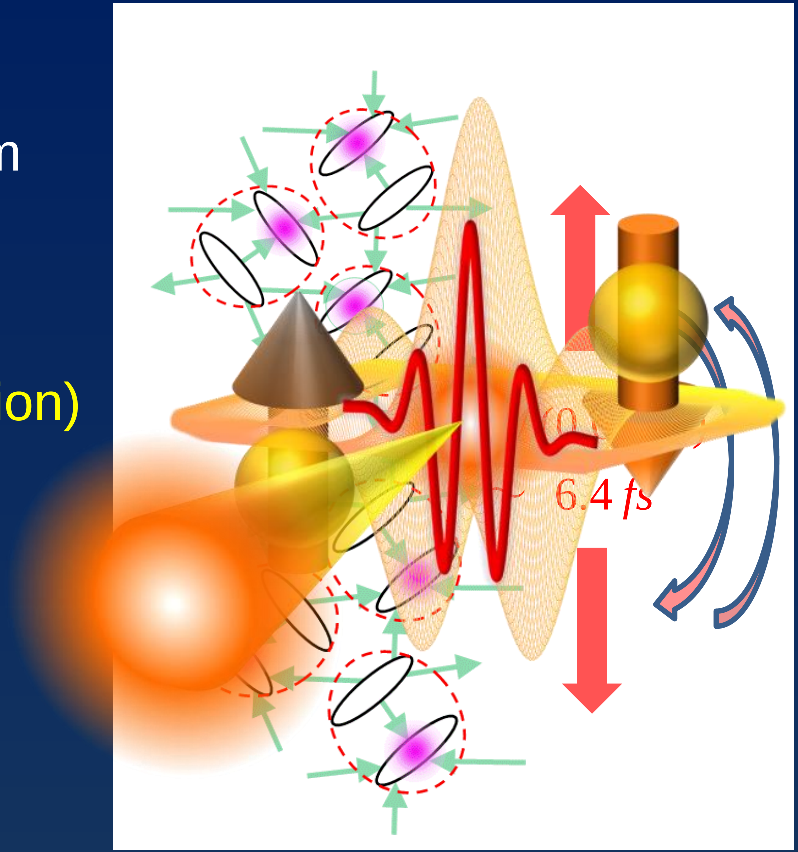
Summary 1

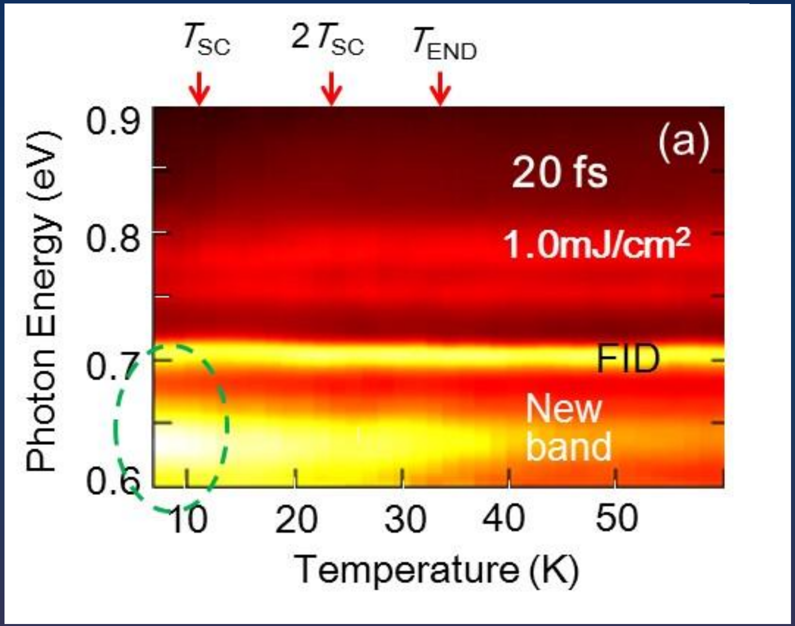
Strong light field effect on dimer Mott system
(6 fs NIR ～single-cycle pulse)

✔ Stimulated emission at 0.63 eV

✔ Nonlinear charge oscillation (synchronization)

✔ Anomalous increase at CEP, SC at 10 fs





Mott criticality and superconductivity are sensitive to 10 fs strong field effect

Kawakami, Sasaki, H. Yamamoto, Yonemitsu, Iwai et al., Nat. Photon. 12, 474 (2018)

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(Coherent chare motion, Dynamical localization)

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Nat. Commun. 2020

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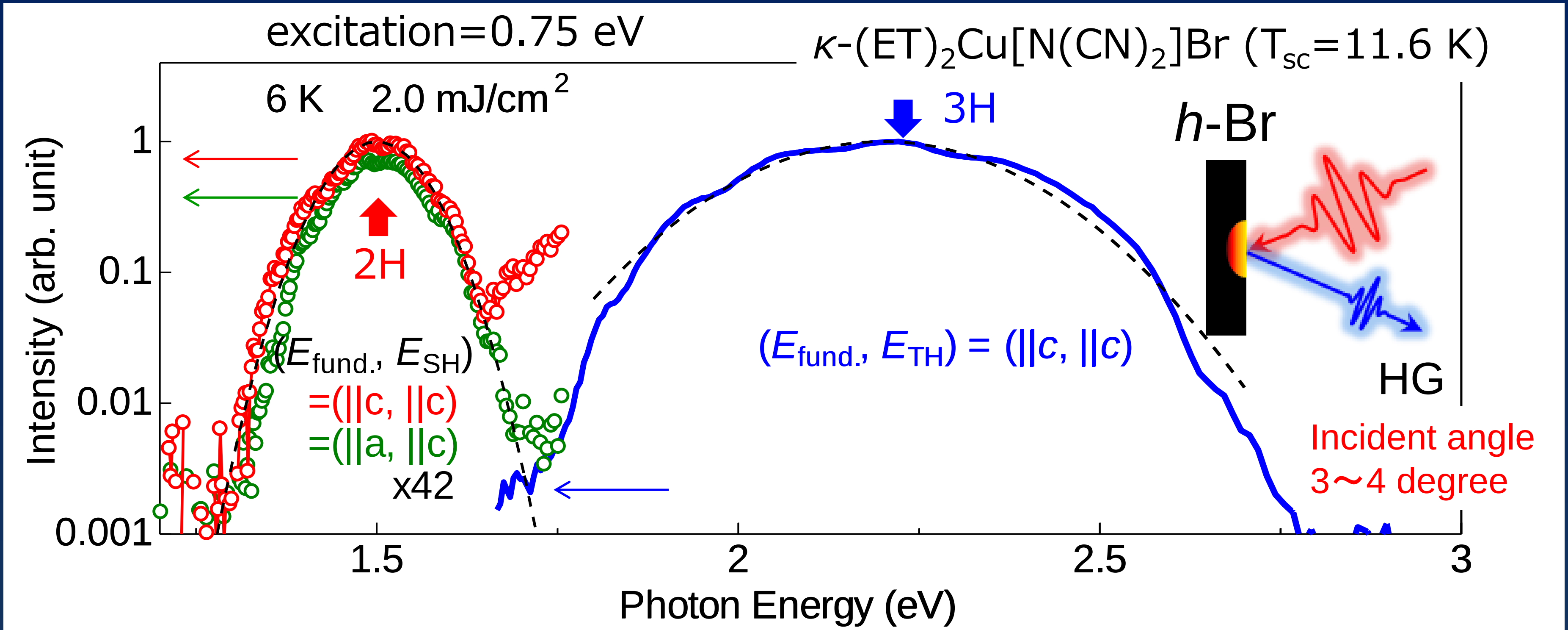
Phys. Rev. Res. (L) 4, L032032(2022)
arXiv: 2207.03877

v) Summary & inprogress

✓ Correlated Dirac semimetal ($SrIrO_3$)

✓ Electronic ferroelectricity ,,,,,,,,,,,

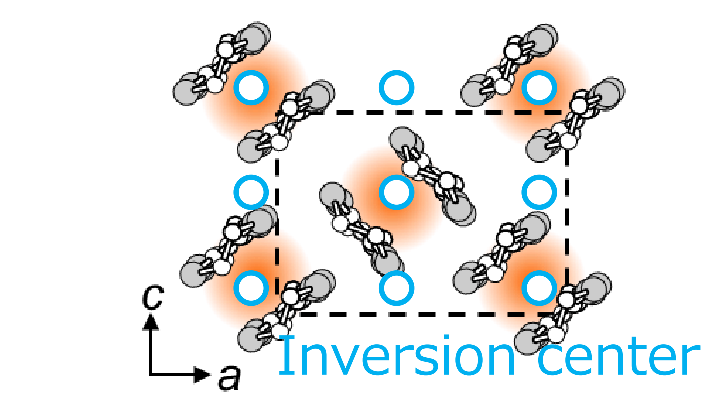
SHG &THG in organic superconductor



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

→ unconventional SHG

Nat. Commun. 10, 1038 (2020)



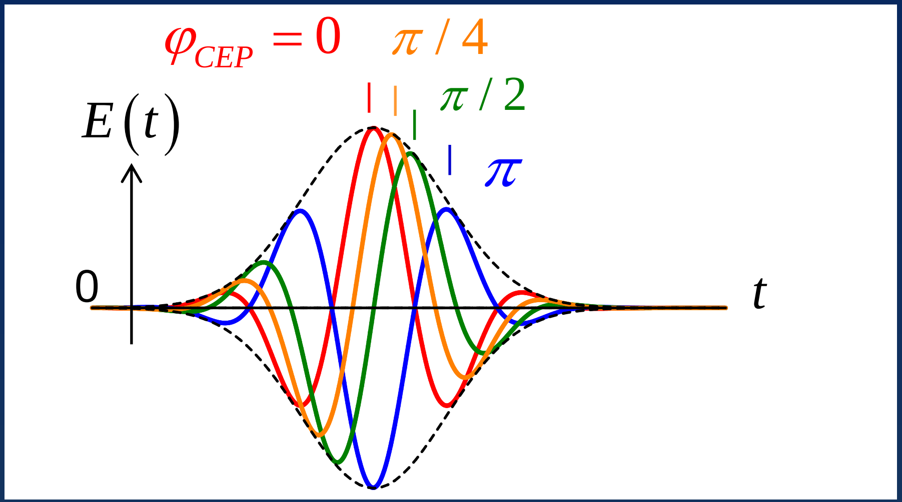
Orthorhombic

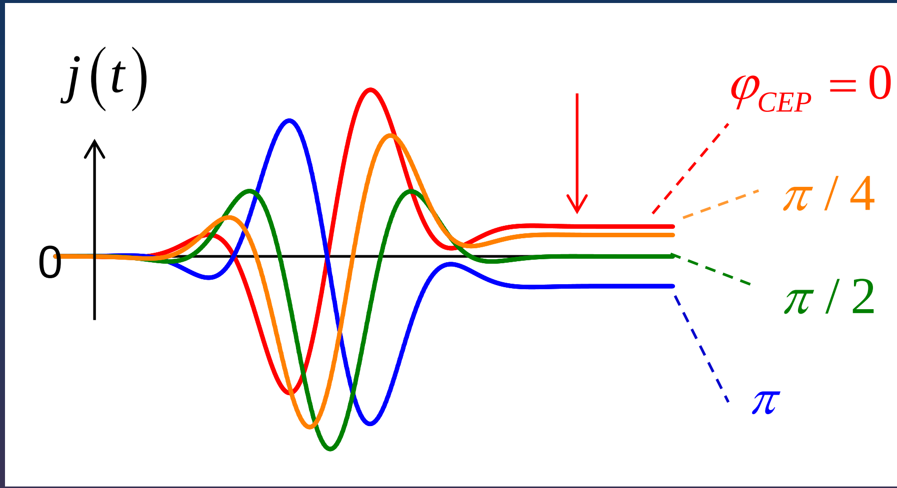
Pnma → $\chi^{(2)} = 0$

Carrier Envelope Phase (CEP)

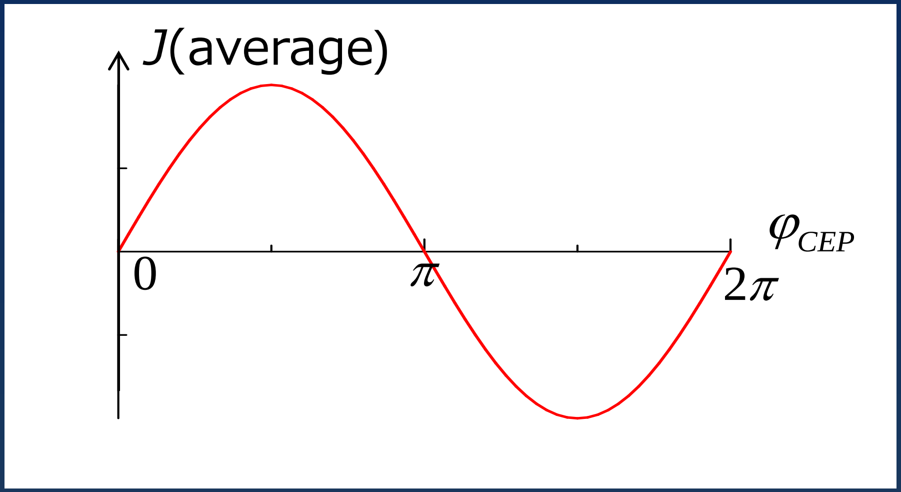
$$E(t) = E_0(t)\sin(\omega t - \varphi_{CEP}) \qquad \varphi_{CEP} : \text{CEP}$$

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





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



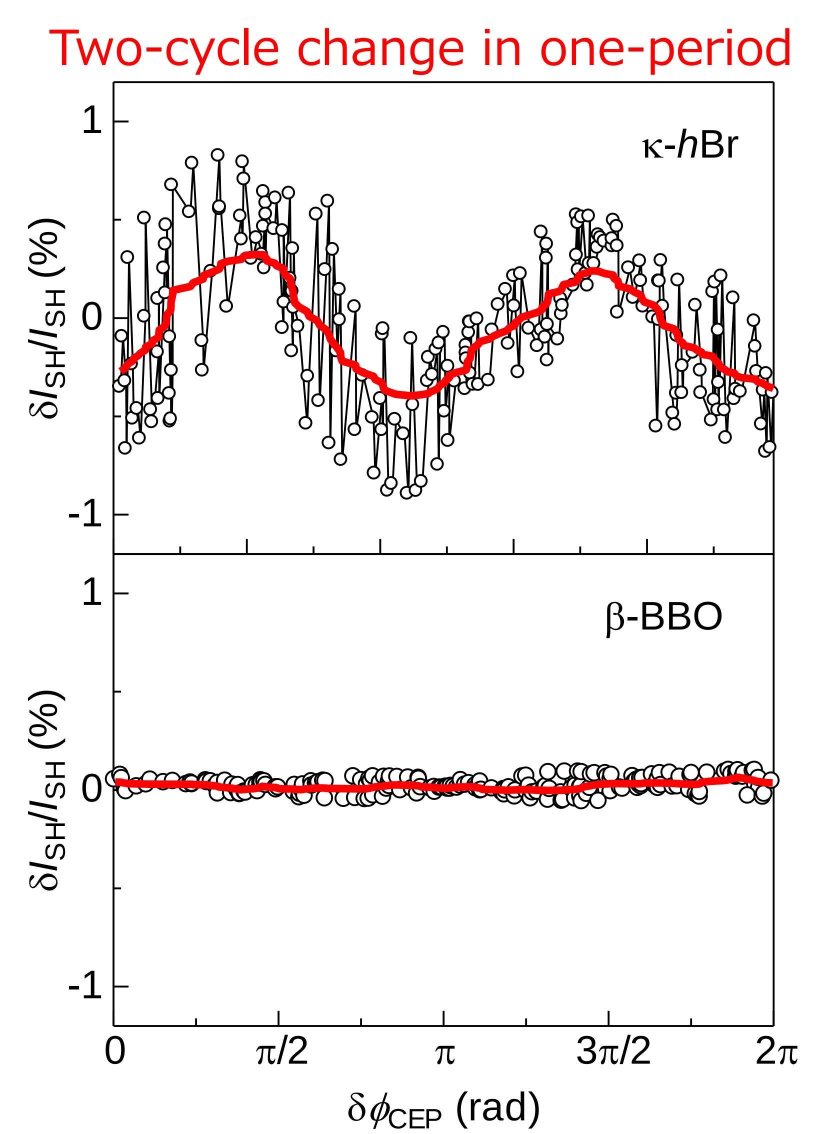
✓ Non-dissipative J is CEP sensitive

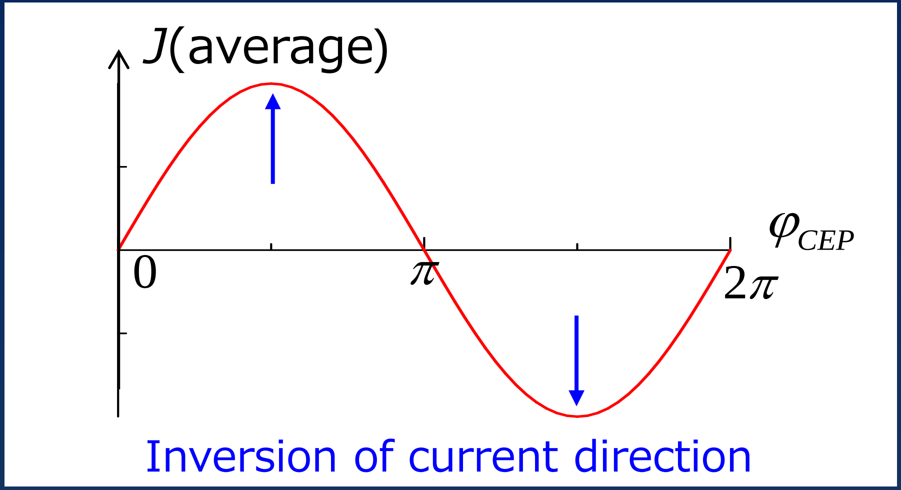
One-cycle change in one-period

Nat. Commun. 10, 1038 (2020)

CEP dependence of SHG

Nat. Commun. 10, 1038 (2020)

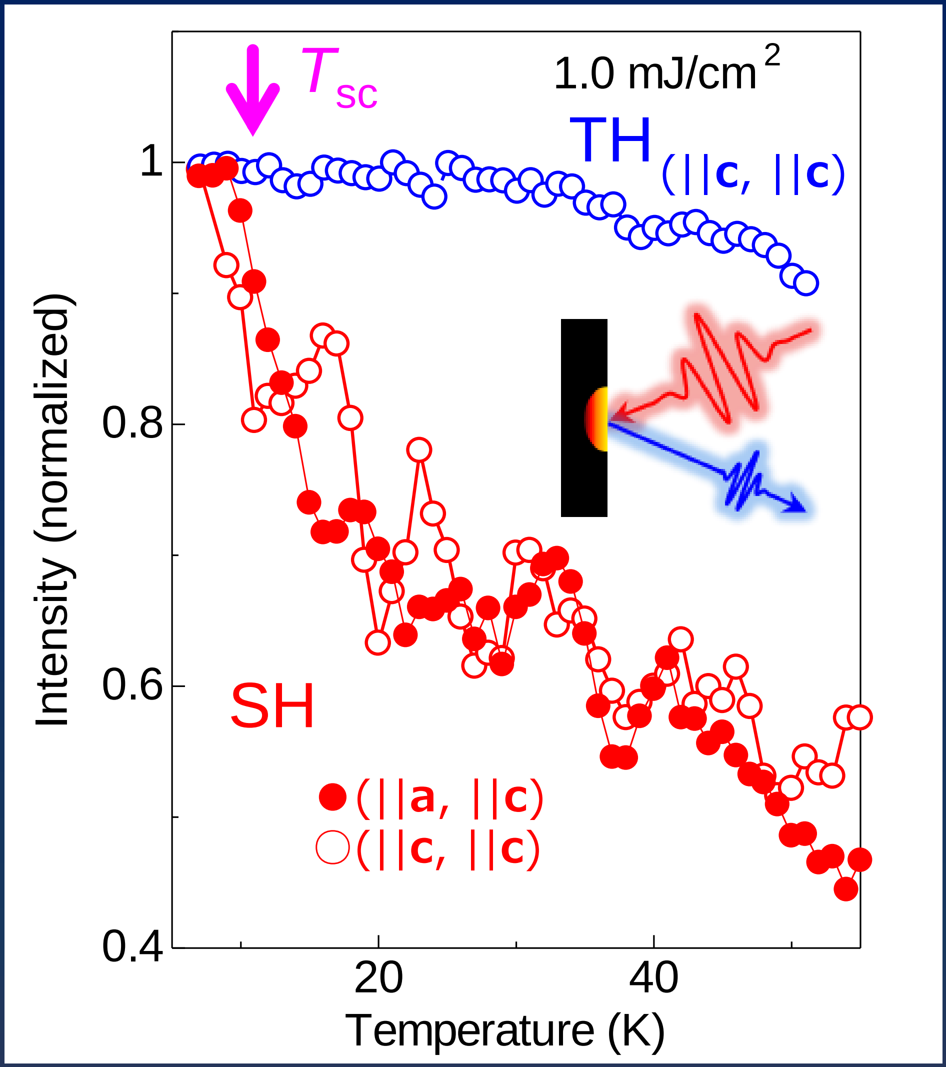


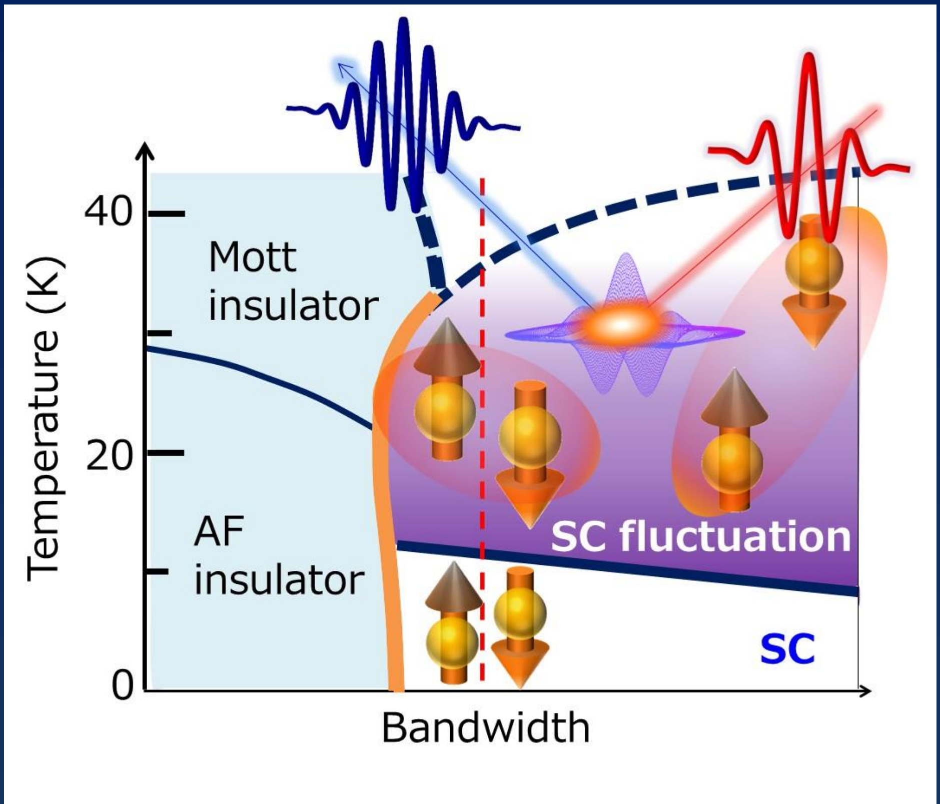


Current is modulated by *f* .
But, SHG can't distinguish direction of current
(SHG is modulated by 2 *f*)

SHG is described by non-scattering current

Temperature dependences of SHG





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

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

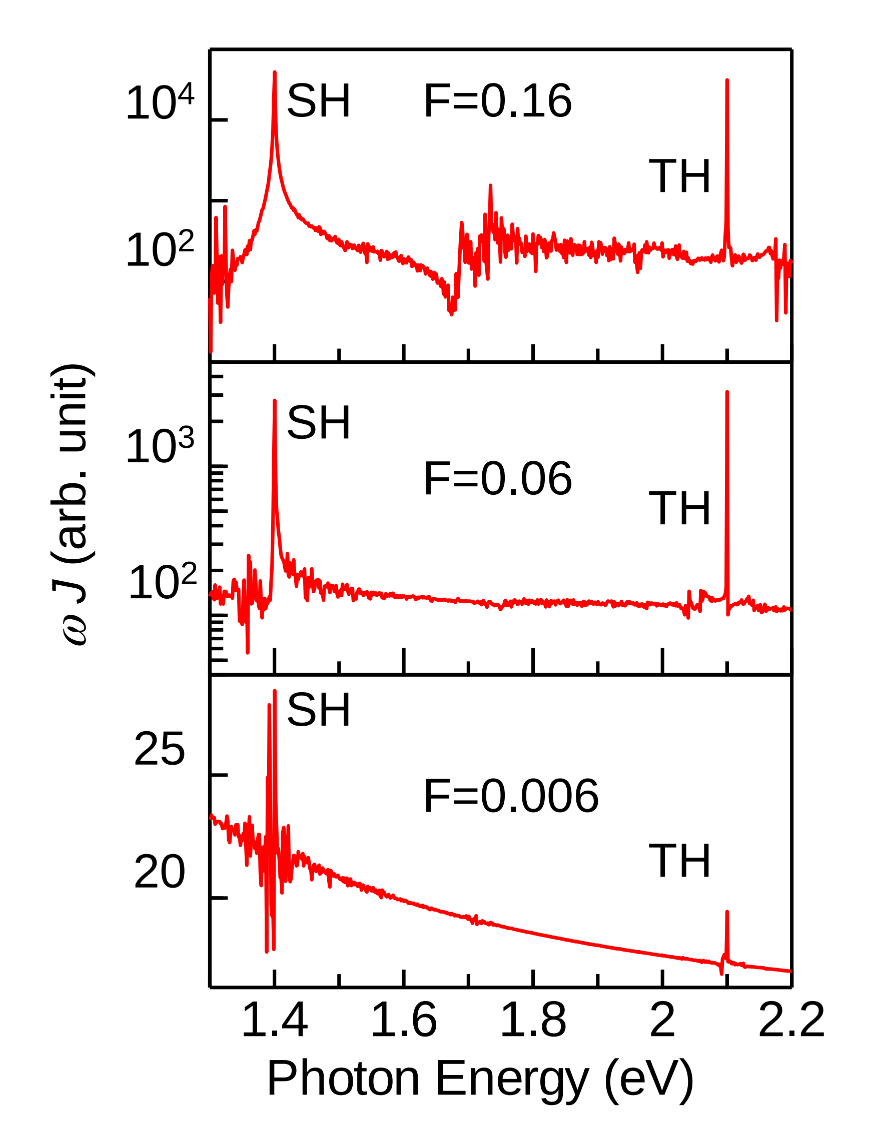
✓ SHG increases toward T_{SC}

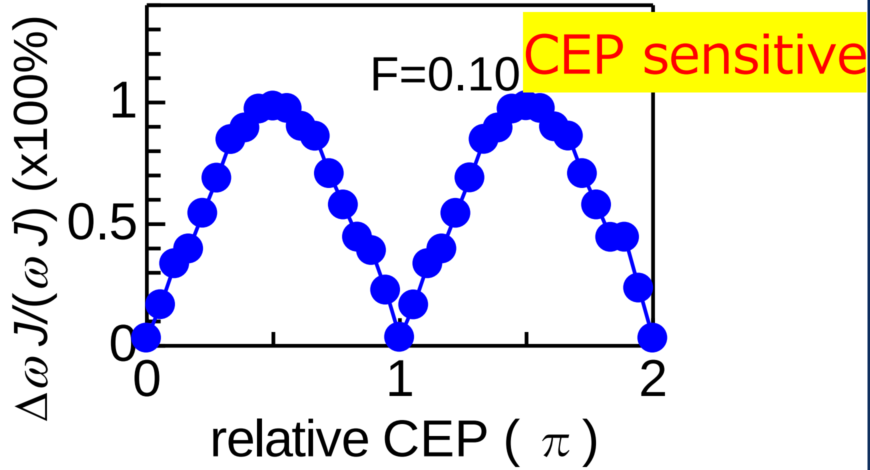
✓ SHG is sensitive to SC fluctuation (reflecting the small working distance of non-scattering current?)

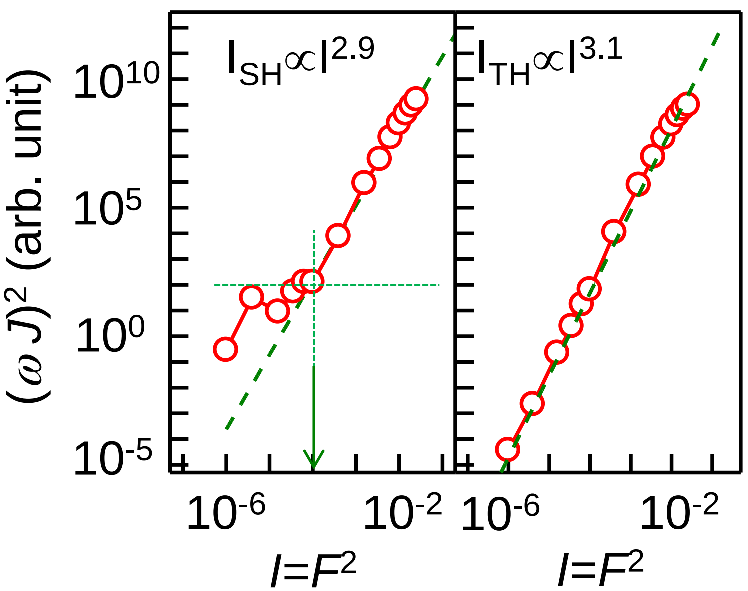
Mean field theory（Prof. Yonemitsu ）

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

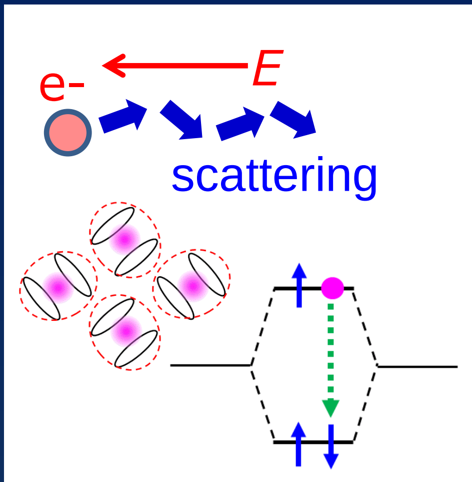
Nat. Commun. 10, 1038 (2020)







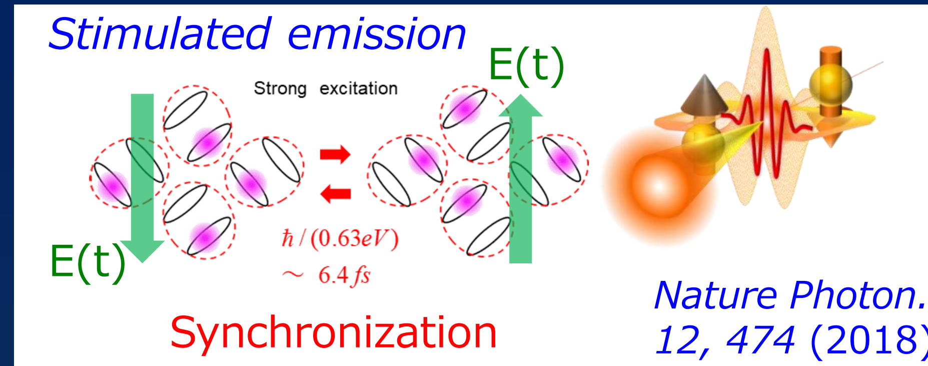
Summary　Organic superconductor *h*-Br

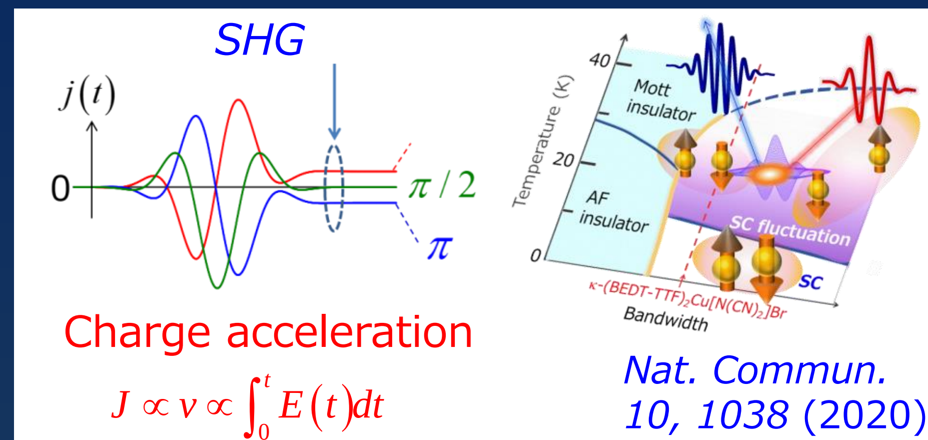


Linear response

Correlated charge motion

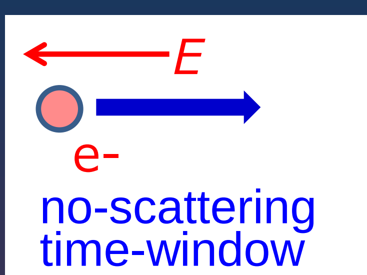
Current induced SHG





Faraday Discussion 237,353(2022)
日本物理学会誌　77, 304 (2022)
レーザー研究 50, 313(2022)

No-scattering current is sensitive to SC fluctuation ?



$$J \propto v \propto \int_0^t E(t)dt \quad \overset{?}{\Longleftrightarrow} \quad j = \frac{n_s e^2}{m} A$$

(London eq.)

(One-electron approx.)　　(Many electron system)

Open problem between ultrafast and SC ?

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PRL2010
Nat. commun. 2014
PRB 2016
PRB 2017(R)
J. Phys. B 2018
(review)

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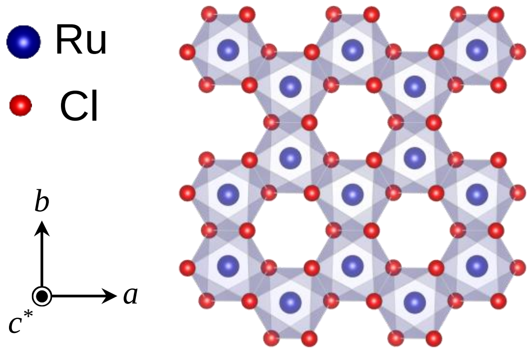
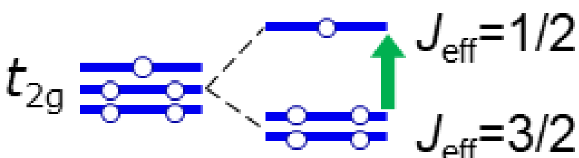
α-$RuCl_3$: Spin-orbit assisted Mott insulator

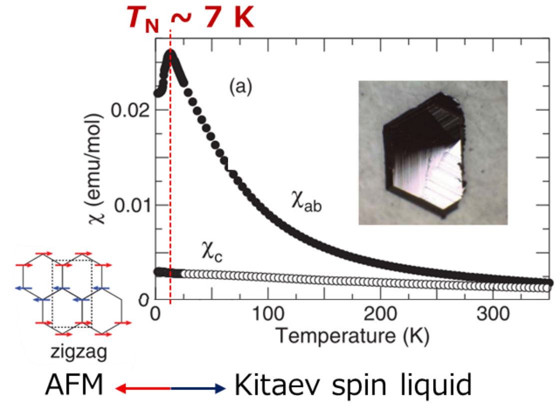


Honeycomb structure

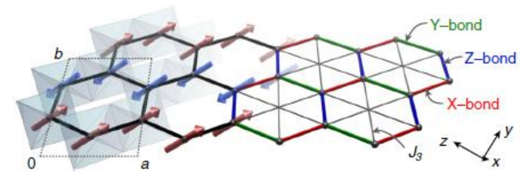


λ=0.15 eV
~t

Magnetic susceptibility



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

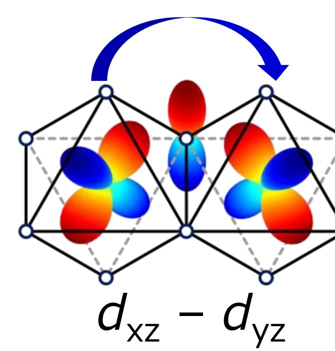
✓ Spin-orbit assisted Mott Insulator

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

Jackeli, Khaliullin PRL 2009

✓ Inter-site hopping t ~0.1 eV
(between different t_{2g} orbitals (such as $d_{xz} - d_{yz}$)

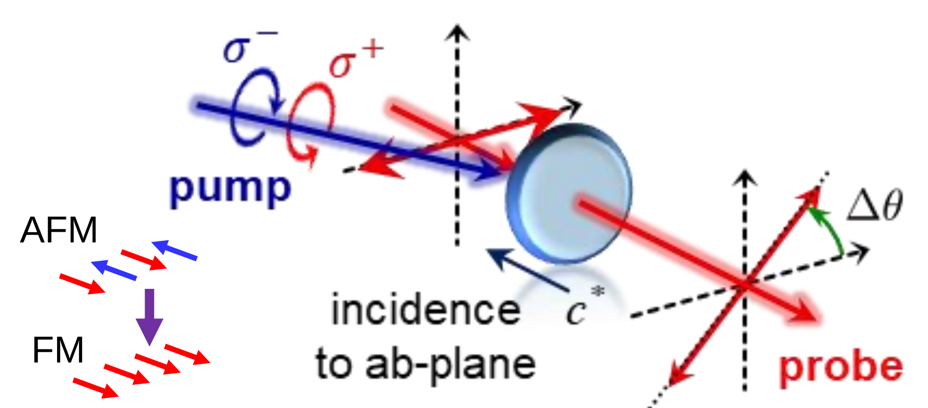
Winter al. Phys. Rev. B93, 214431(2016)



t: intersite hopping

Orbital moment?

Experimental



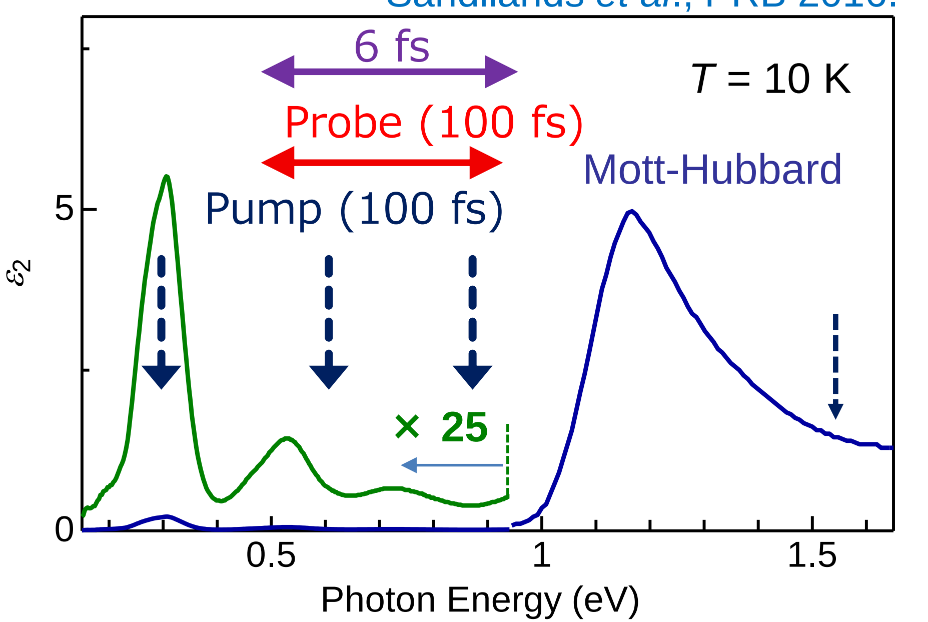
Helicity dependent polarization rotation

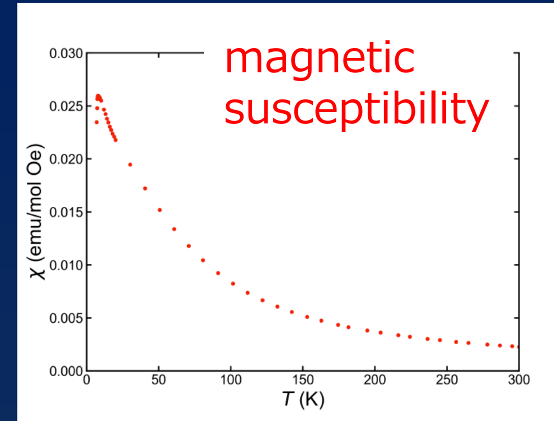
Inverse Faraday effect

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)

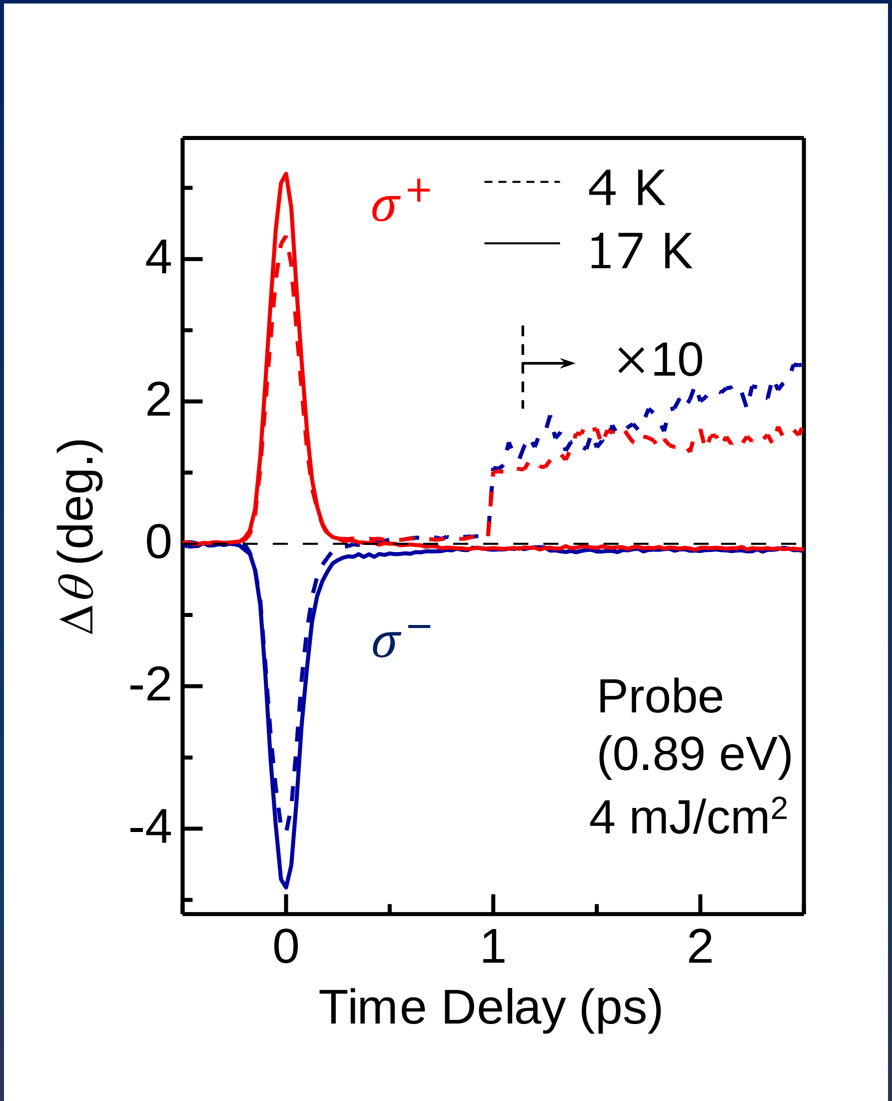
✓ Circular (σ^+, σ^-) polarization

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



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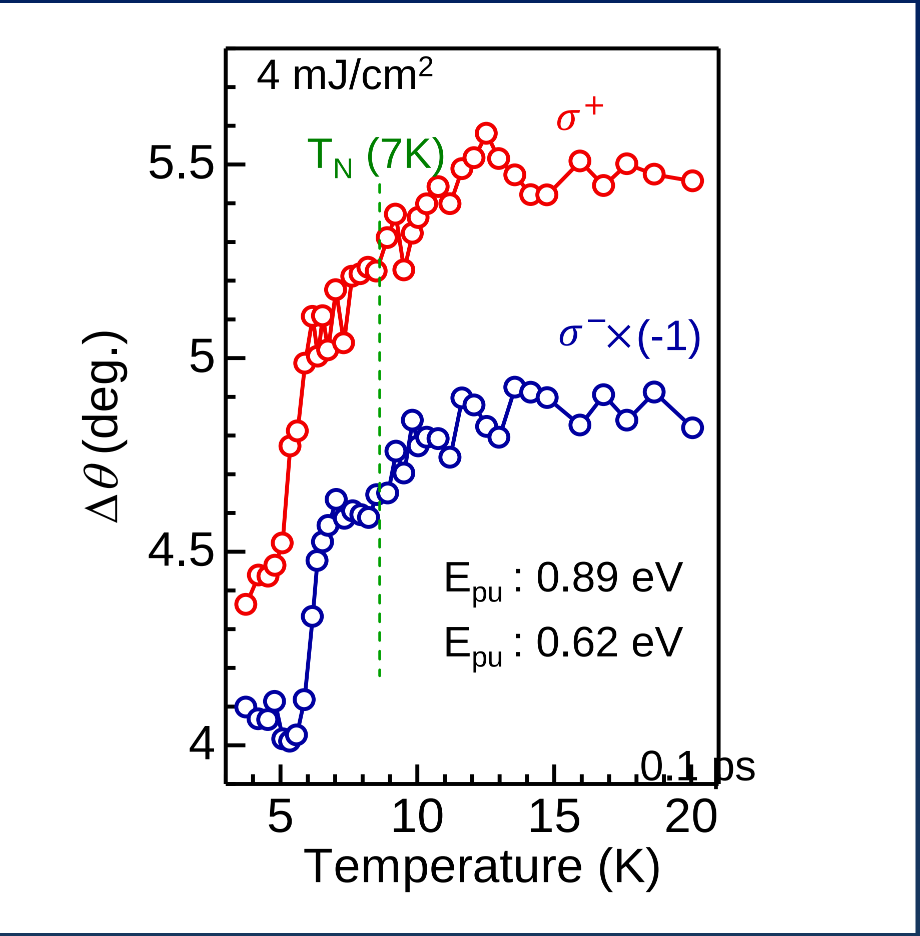
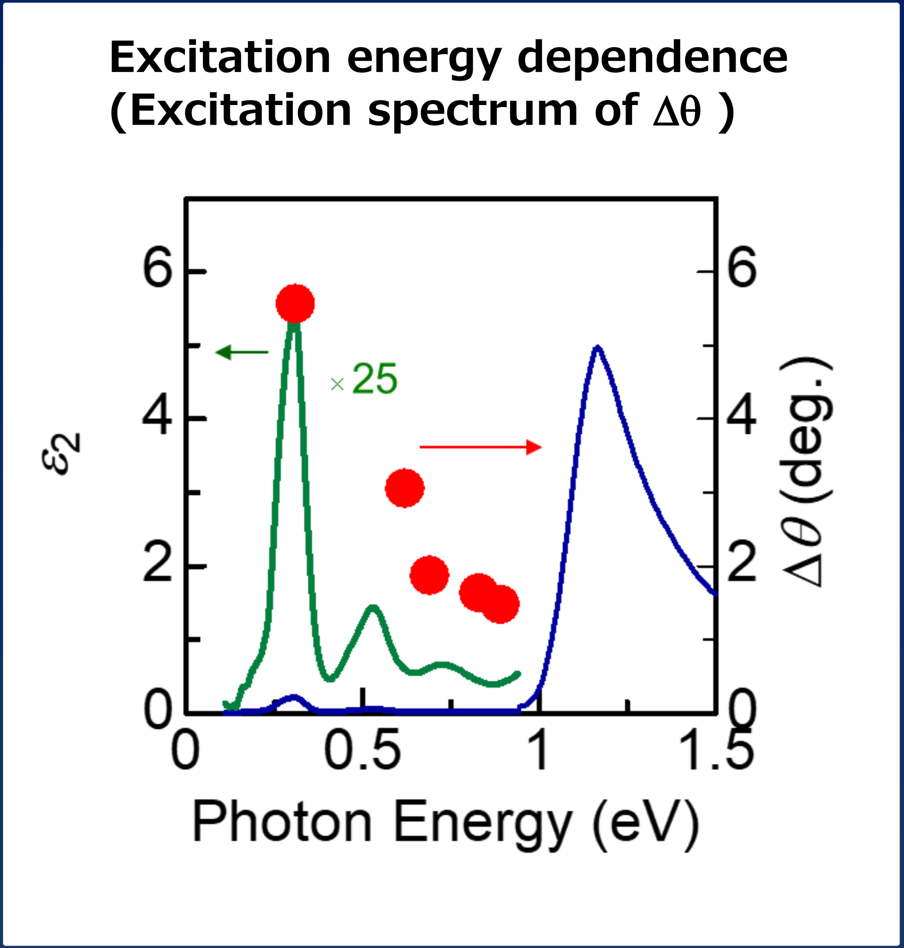
Light induced ultrafast magnetization (⊥ plane)

Unconventional behaviors of Δθ





✓ Reduction at T< T_N ?

Opposit to the expected tendency in typical IFE (increase in Δθ below T_N)

✓ Resonance to spin-orbit excitons

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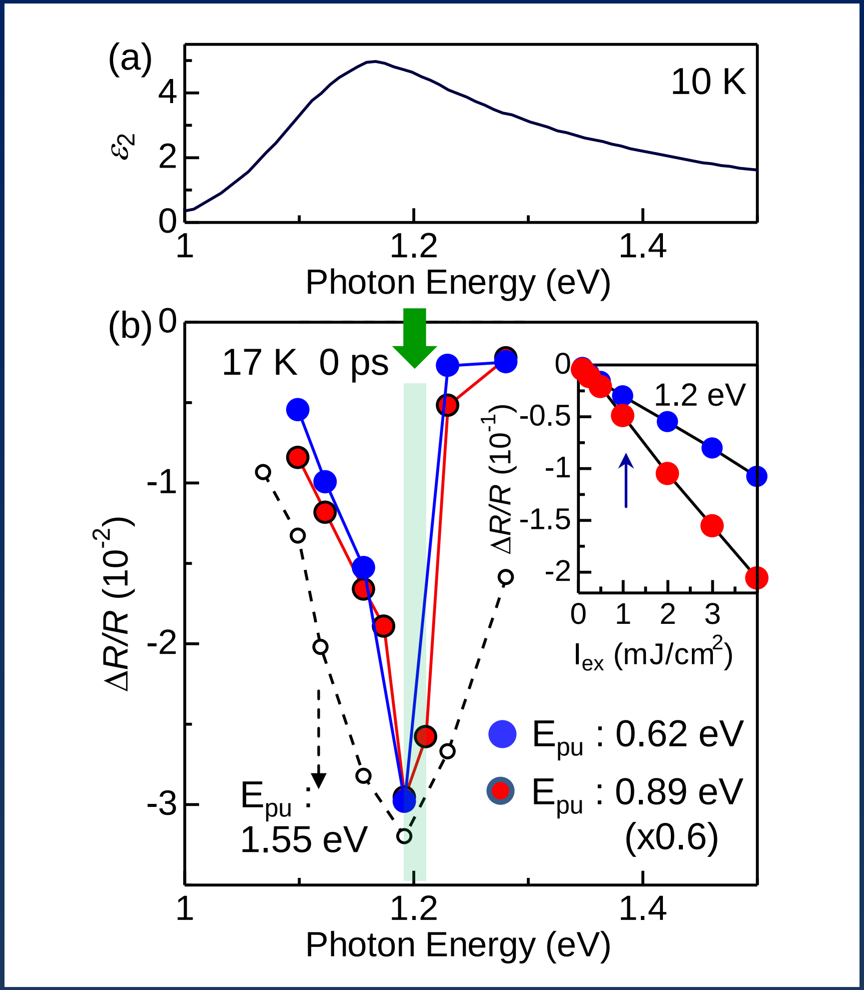
New mechanism of light-induced magnetization ?

Time profile of ΔR/R



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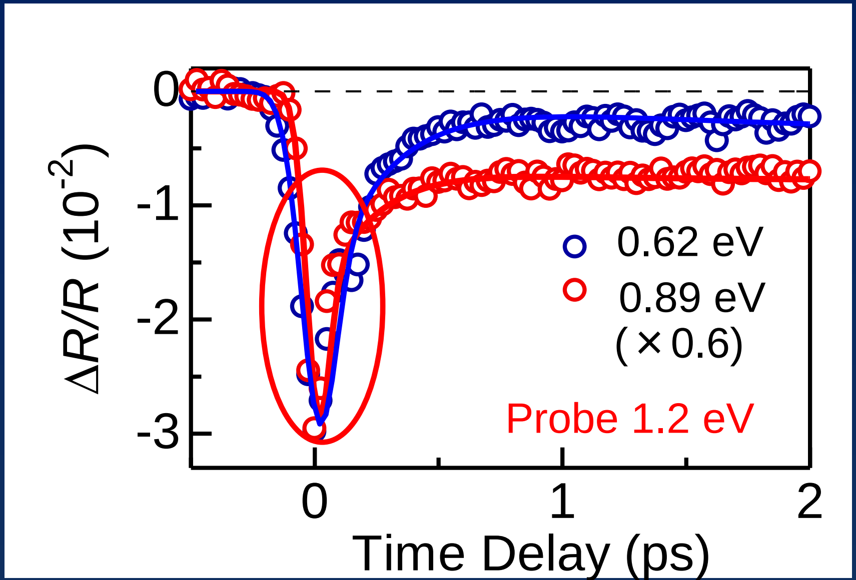

✓ **Relaxation dynamics**

i) < 100 fs

→ Ultrafast magnetization ?

ii) ~ 250 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)

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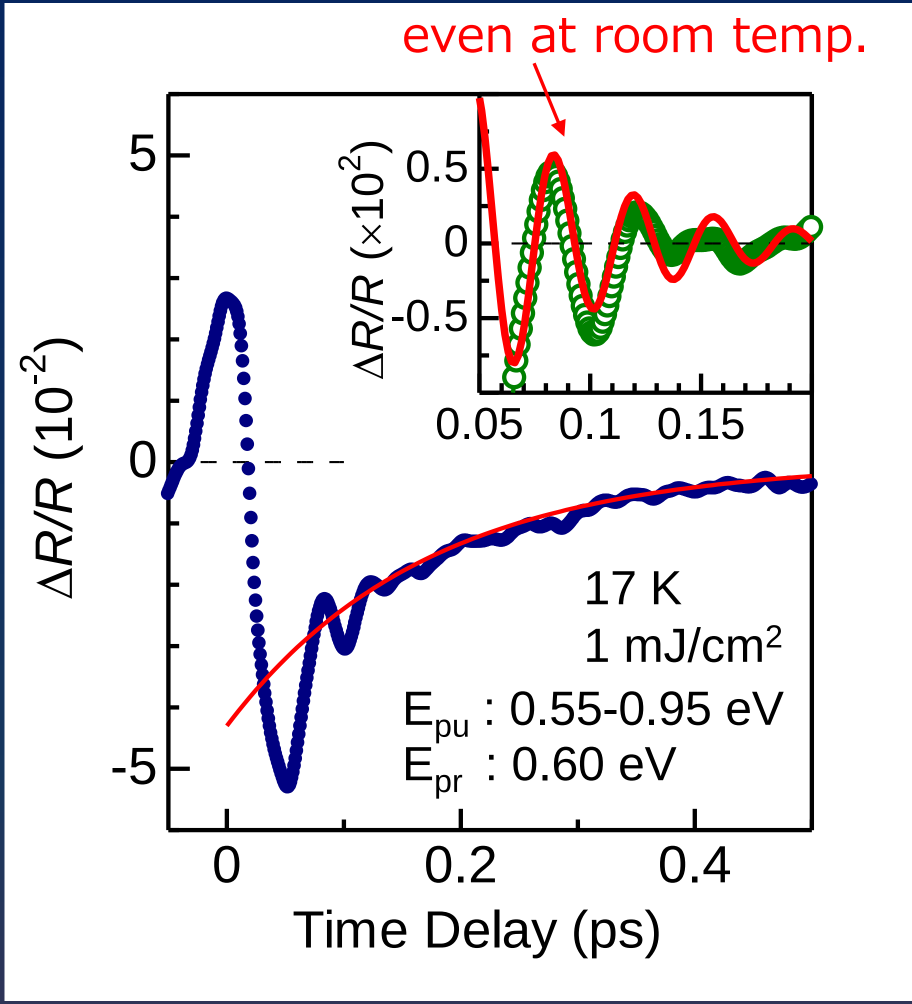
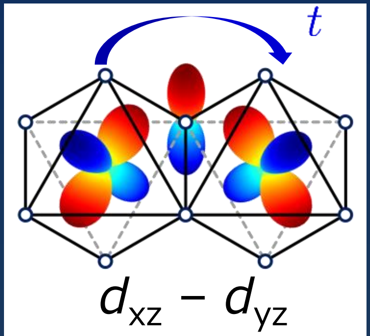


✓ oscillation period ~ 40 fs (<shortest phonon period 100 fs)

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

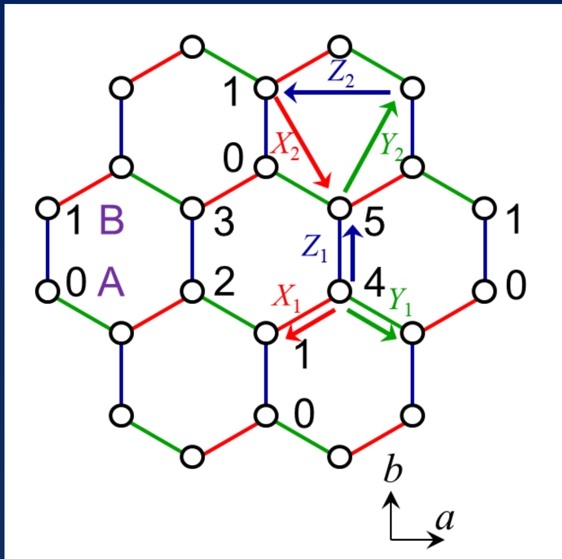
→Coherent charge hopping between different t_{2g} orbitals?



dephasing time 60 fs ~ lifetime of magnetization

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

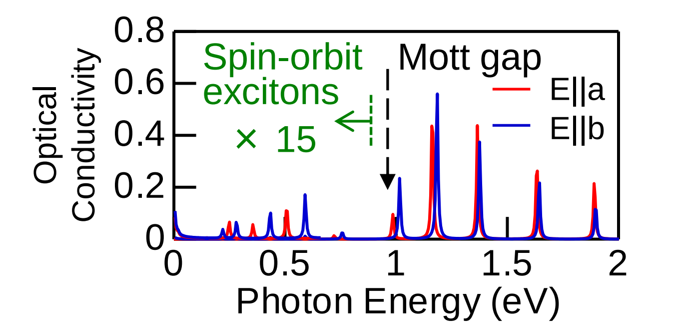
Opto-magneto effects in α-$RuCl_3$ (theory)

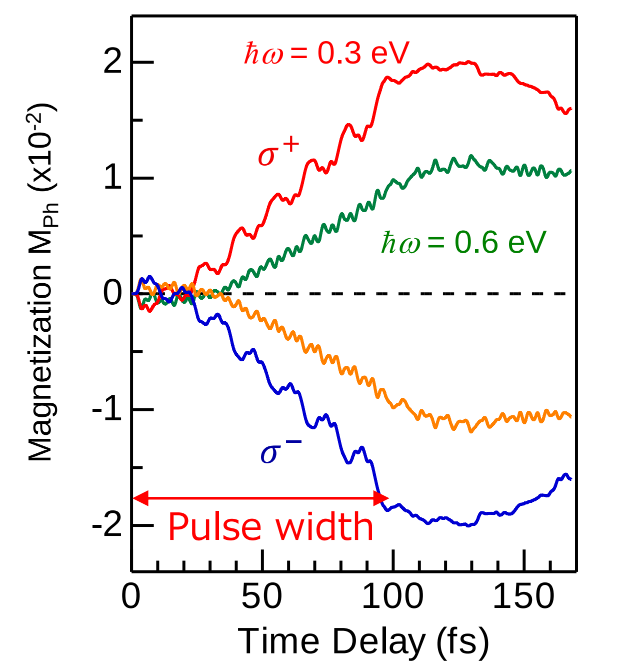


Quantum mechanical analysis (steady state)

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

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

✓Peierls substitution

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

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J. Phys. Soc. Jpn. 91,104702(2022)

High-frequency expansion in Floquet theory

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

the second-lowest order of the high-frequency expansion

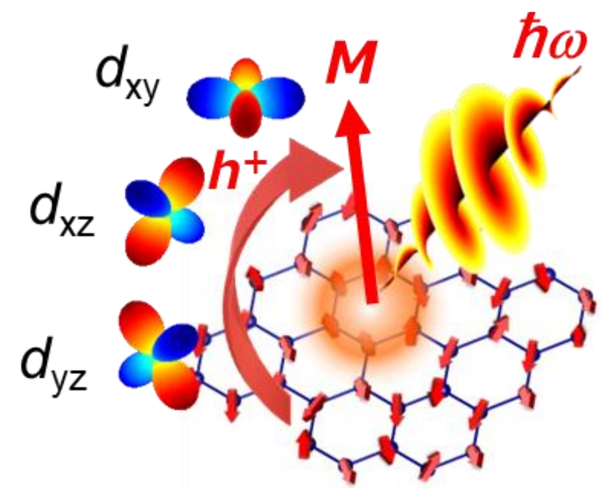
orbital moment

有効軌道角運動量

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

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

Spiral current drives magnetic moment



異なるt_{2g}軌道間の電荷移動

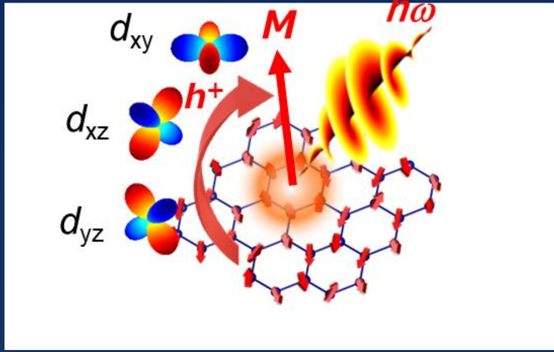
Phys. Rev. Res. 4, L032032(2022)
J. Phys. Soc. Jpn. 91,104702(2022)

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* (L) 4, L032032(2022)**
K. Yonemitsu
***J. Phys. Soc. Jpn*. 91,104702(2022)**

Summary

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

i) Introduction

✓ Coherent charge motion in correlated syatem
(Coherent chare motion, Dynamical localization)

✓ *6-fs NIR pulse, CEP control/detection*

ii) Stimulated emission in organic SC κ-ET salt

✓ Ultrafast stimulated emission (SE) driven by strong field

✓ Temperature dependence (anomaly around T_{SC})

Nat. *Photon* 2018

iii) Unconventional SHG in κ-ET salt

✓ SHG induced by Petahertz no-scattering current (CEP sensitive)

✓ Enhancement of SHG near SC fluctuation

Nat. Commun. 2020

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

✓ Ultrafast magnetization (larger for $T>T_N$)

✓ Coherent carrier dynamics & theory

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

v) Summary & inprogress

✓ Correlated Dirac semimetal ($SrIrO_3$)

✓ Electronic ferroelectricity ,,,,,,,,,,,