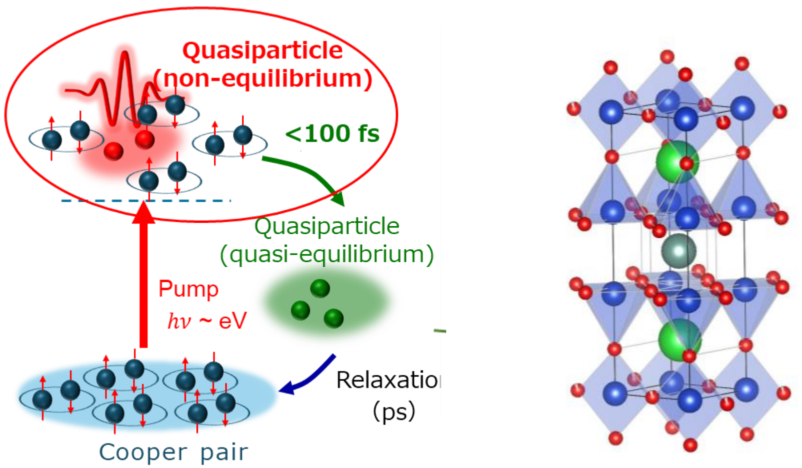
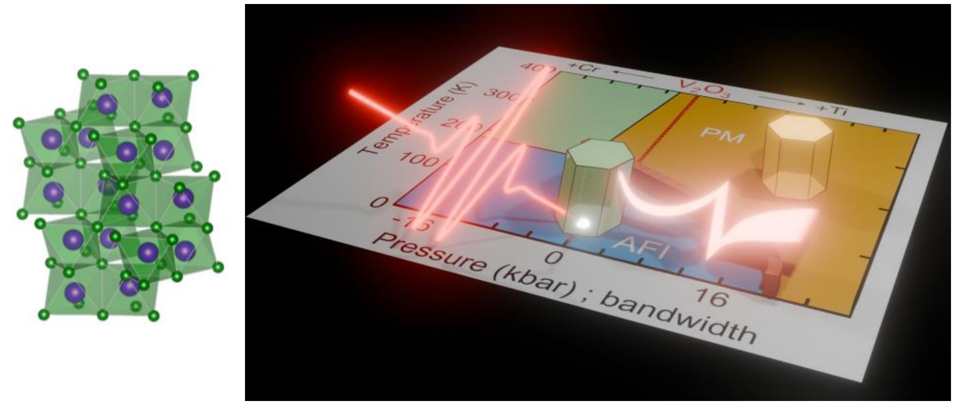
Correlated charge dynamics in transition metal oxides

Tohoku University   Shinichiro Iwai



High-T_c SC cuprate (YBCO)

Kato et al.,
Ultrafast Phenomena 2024



Ferroelastic Mott insulator V_2O_3

Amano et al.,Nat. Phys. accepted

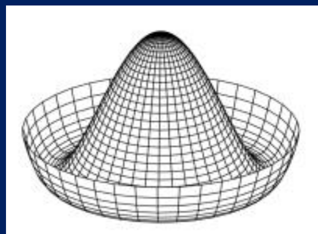
Supported by

JST-CREST **JP19198318**

Q-leap **JPMXS0118067426**

Symmetry breaking in correlated system

Gauge

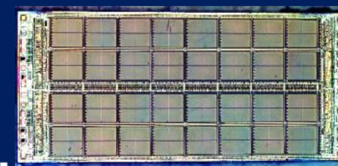


Superconductivity

K-(ET)$_2$X Nat. Photon **12, 474 (2018)**

K-(ET)$_2$X Nat. Commun. 11, 4138(2020)

YBCO Ultrafast Phenomena 2024

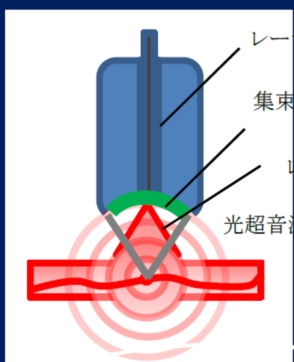


(TMTTF)$_2$X

Phys. Rev. Res. 3、L032043（2021）

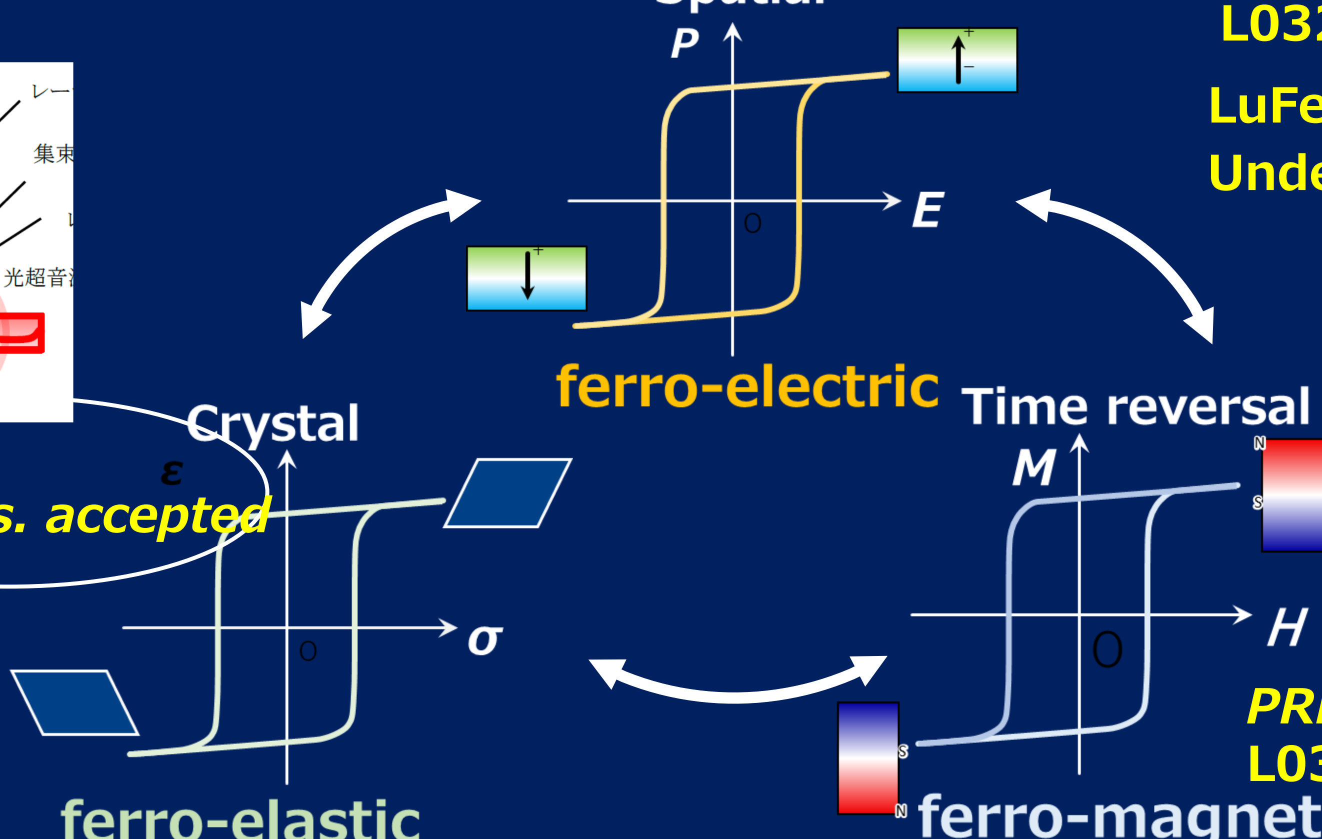
$LuFe_2O_4$

Under review



V_2O_3

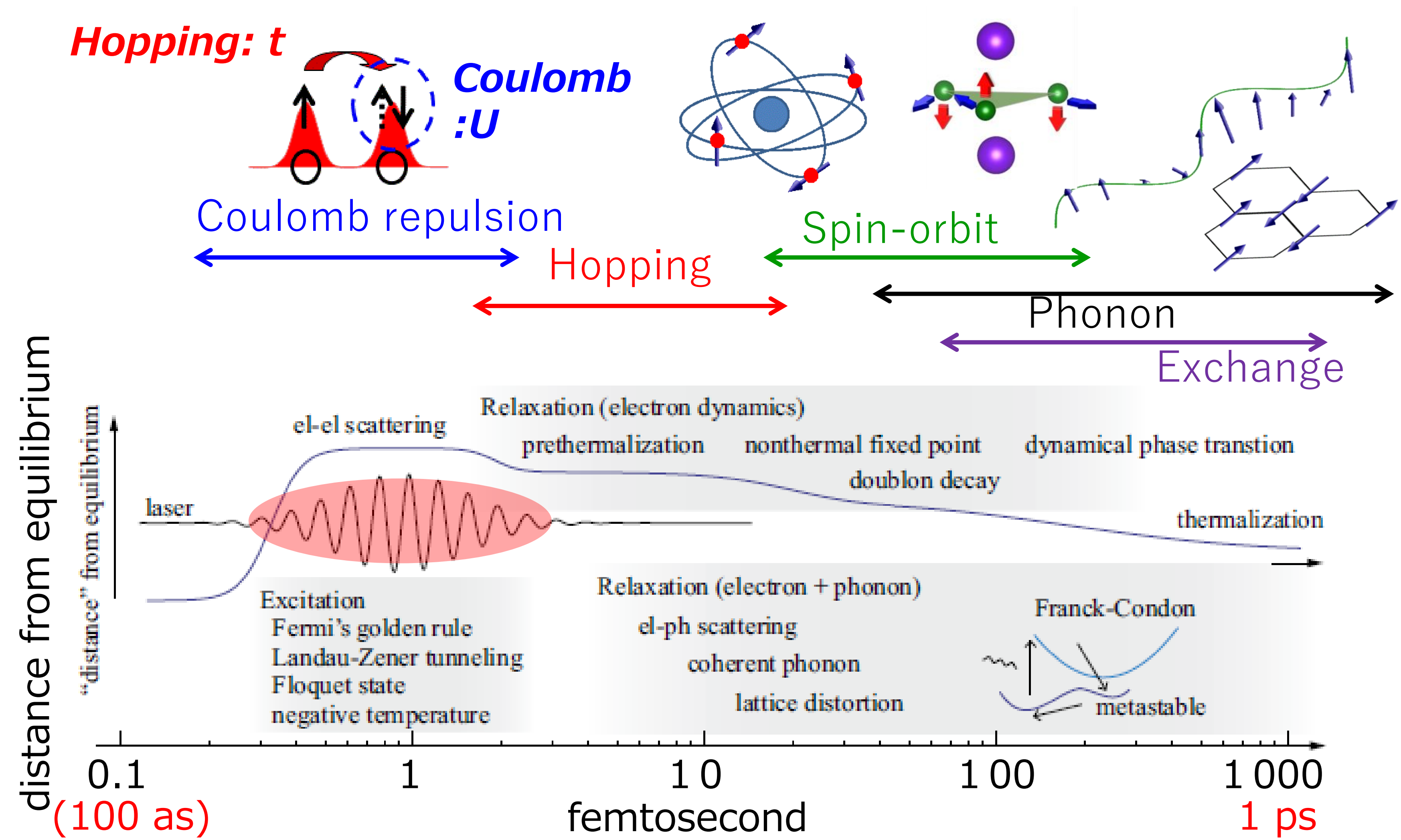
Nat Phys. accepted



α-RuCl$_3$

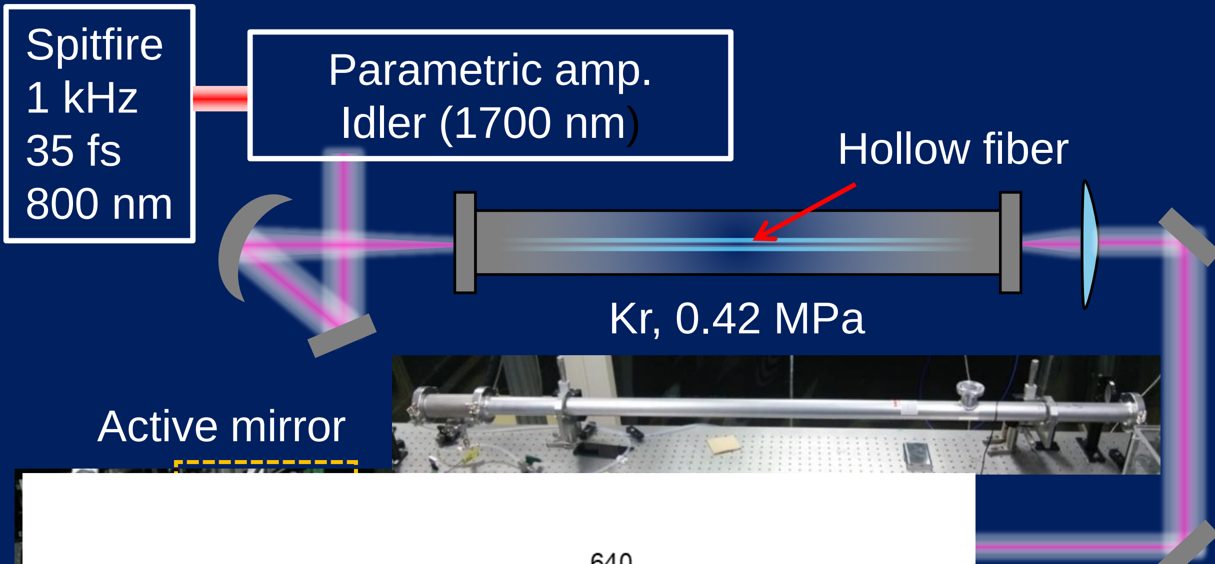
PRRes. 4, L032032(2022)

How fast can we manipulate ?



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

6 fs NIR pulse (1.3 cycle, CEP stabilized)



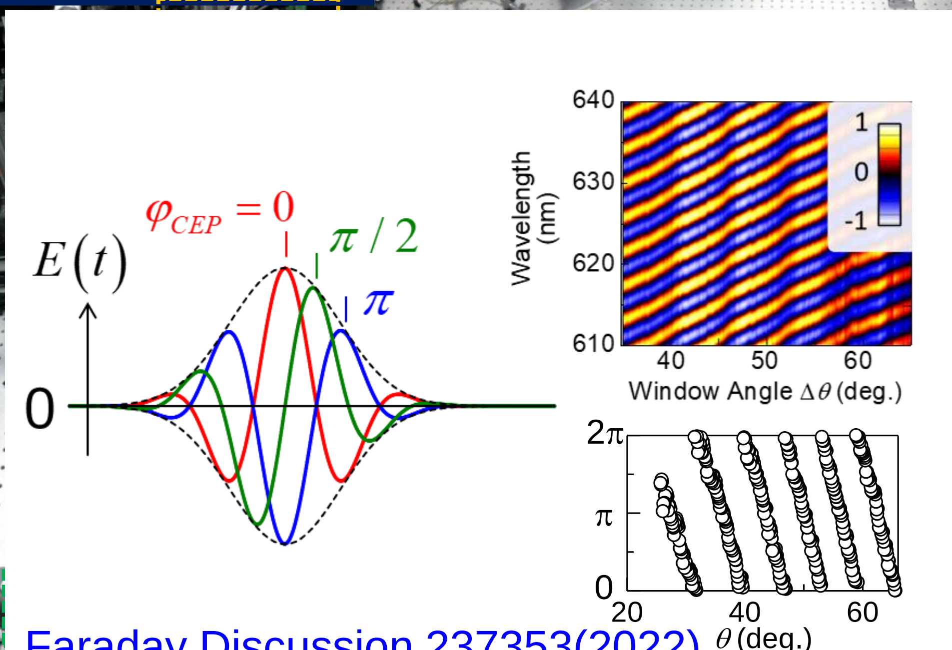
✓ **OPA Idler ($\chi^{(2)}$)**

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

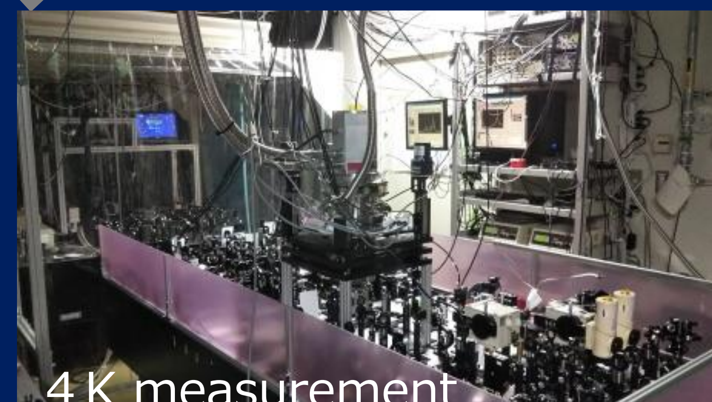
✓ **SPM($\chi^{(3)}$)**

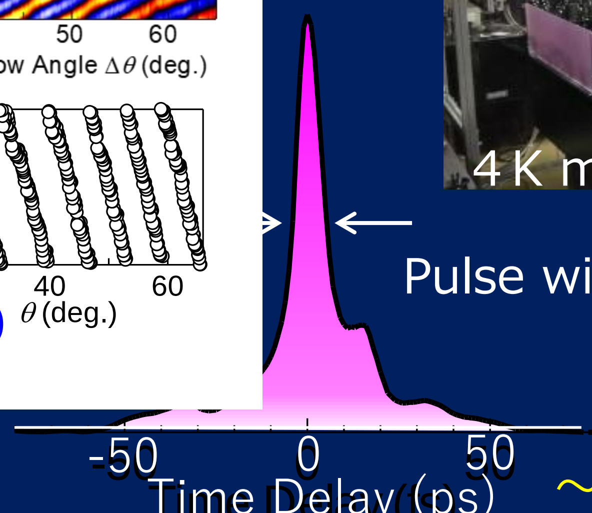
$$\varphi_{SPM} = \frac{\pi}{2} + \varphi_1 + \varphi_2 - \varphi_3$$

→ CEP stable



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





Pulse width =6 fs

30 μJ/pulse
Φ200 μm

Max >50 MV/cm !

～20 MV/cm for organics

J. Phys. B 2018

Collaborators

Ultrafast & THz spectroscopy

Tohoku Univ.

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Y. Minakami, R. Kato,
Y. Taniguchi


Kawakami


Amano


Itoh


Minakami

Kansei-hakuin Univ. H. Itoh

(YBCO)

Nagoya Univ. Y. Nakamura, H. Kishida
Kyusyu sangyo University T. Nishizaki
Tohoku University K. Ohgushi


Kato


Taniguchi

(V_2O_3) Rennes, Nantes, MaXIV, ESRF (→ lnext page)

($LuFe_2O_4$)

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Okayama Univ. K. Fujiwara, N. Ikeda

Theory

Chuo Univ. N. Arakawa, K. Yonemitsu

Thank to Christian Bernhard (*Fribourg*) , Jure Demsar (*Mainz*)

Collaborators (V_2O_3)

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H. Cailleau, M. Lorenc

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MAX IV Laboratory, Lund University,
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ESRF, The European Synchrotron
C. Mariette, M. Levantino, Mickael Kozhaev

Generation dynamics of quasi-particles in high-Tc superconducting quprate

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H. Itoh

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Y. Nakamura, H. Kishida

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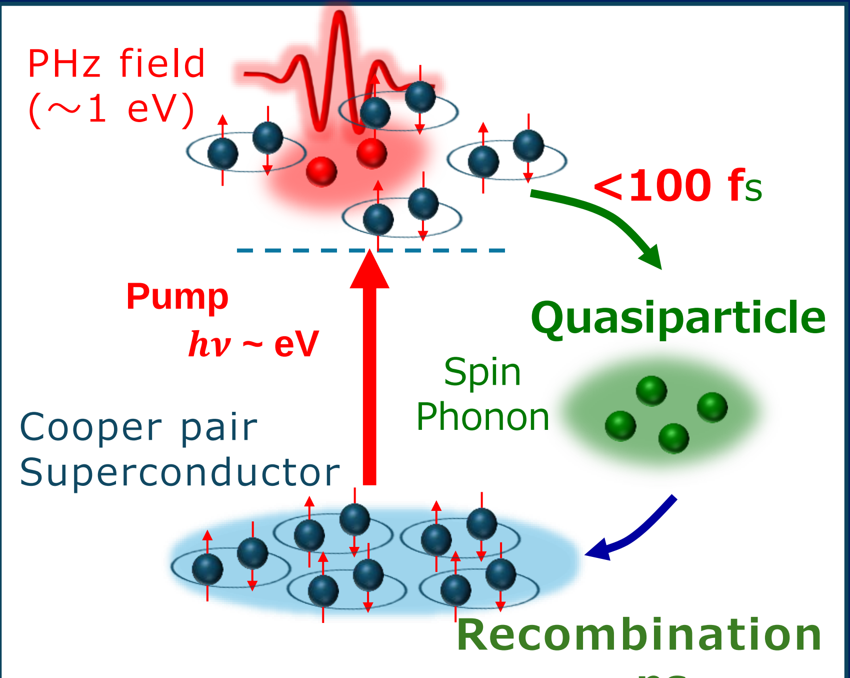
Tohoku University
K. Ohgushi

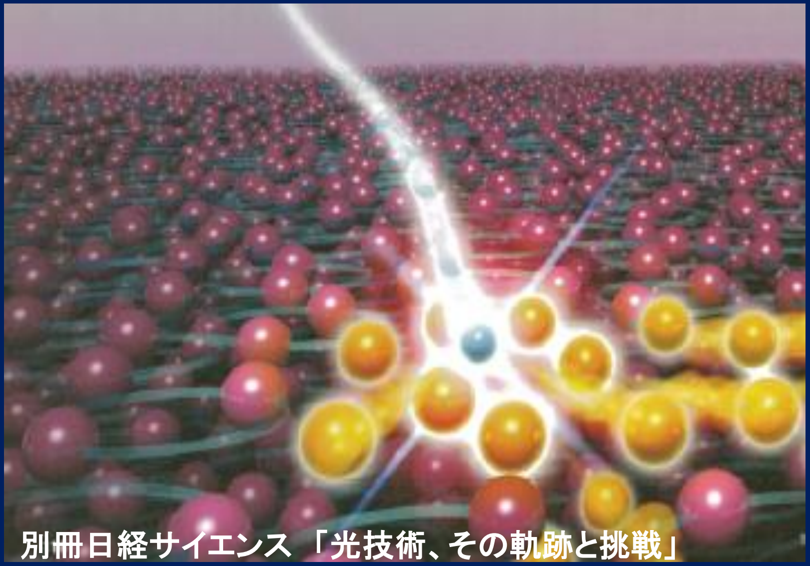
Chuo Univ.
N. Arakawa, K. Yonemitsu

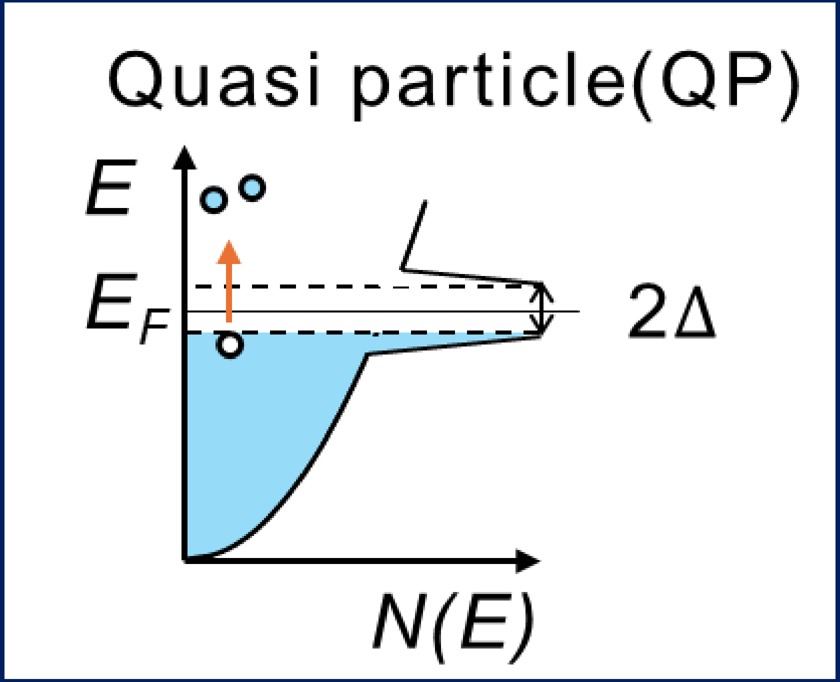
JST-CREST JP19198318
Q-leap JPMXS0118067426

QP dynamics in HT-SC cuprates

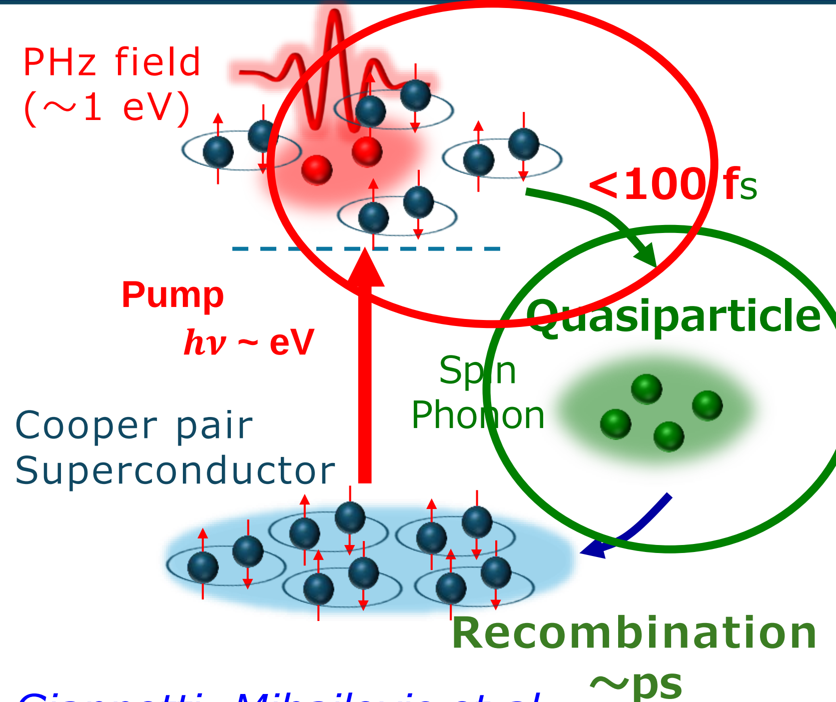


Giannetti, Mihailovic et al., Advances in Physics, **65**, 58-238(2016).





QP dynamics in HT-SC cuprates



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

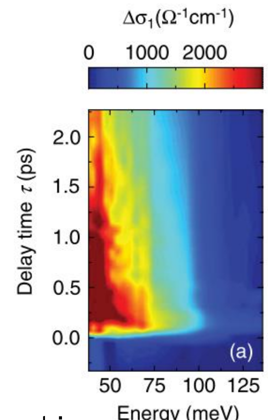
Relaxation of QP

Pashkin *et al.*,
PRL105, 067001(2010) **YBCO**

Cortés *et al.*,
PRL 107, 097002(2011). **BSCCO**

THz TDS

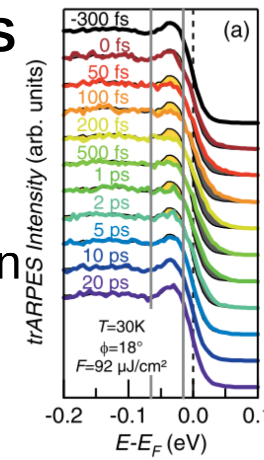
Closing of SC gap



Recombination
~3 ps

tr-ARPES

non-eq. population



→SC gap(~10 meV)

S. G. Han et al.
Phys. Rev. Lett., 65, 2708 (1990)

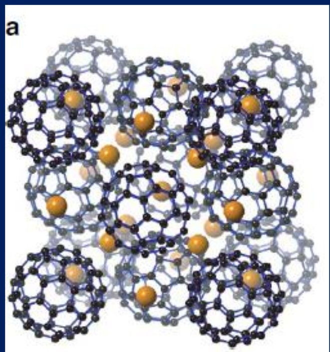
S. V. Chekalin et al.
Phys. Rev. Lett., 67, 3860 (1991)

D.H. Reitze et al.
Phys. Rev. B46 , 14309(1992)

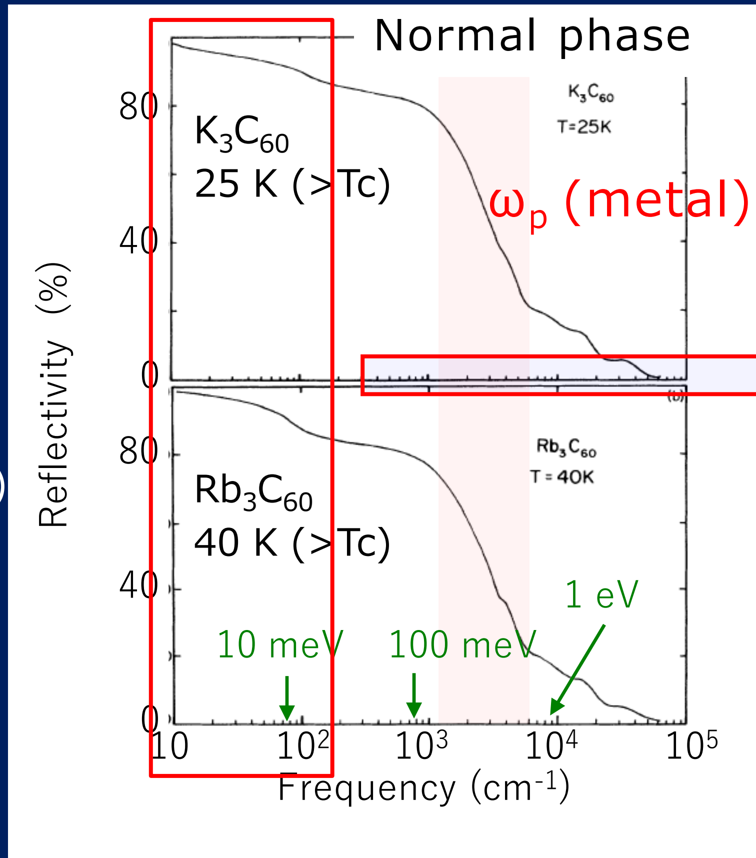
J. Demsar et al.,
Phys. Rev. Lett. 82, 4918(1999)

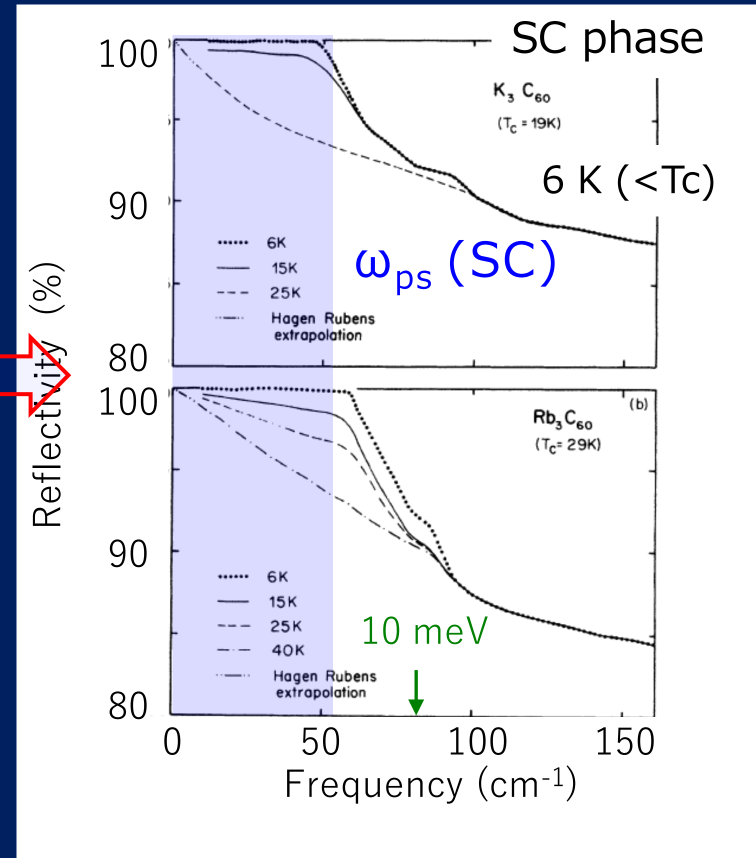
- **Spectral weight transfer (spectral range of 0.1 ～ 4 eV)**
- **Build-up of coherent phonons (3.5 & 4.5 THz)**

Conventional SC: low enery (~meV)



A_3C_{60}
(typical example)
Kasahara et al., Nat Commun. 2017





Degiorgi et al., Phys. Rev. B49, 7012(1994)

Drude model (metal)

$$\varepsilon = \varepsilon_\infty - \frac{\omega_p^{\,2}}{\omega(\omega + i\gamma)}$$

$$\omega_p \propto \sqrt{\frac{n}{m^*}}$$

n: carrier number

m^*: effective mass

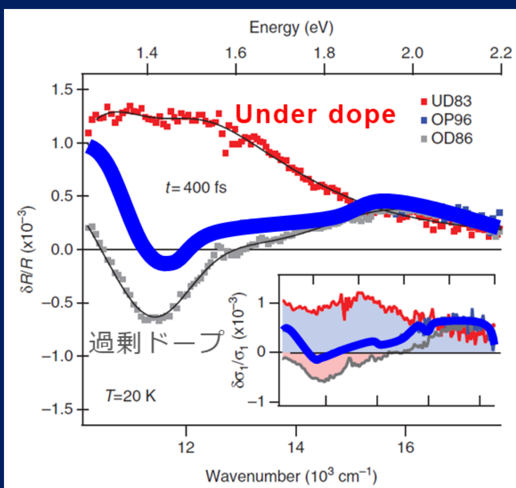
$$\varepsilon = \varepsilon_\infty - \frac{\omega_p^{\,2}}{\omega(\omega + i\gamma)}$$ for γ = 0

ω_{ps} : SC plasma freq. ~meV

Spectral weight (SW) transfer

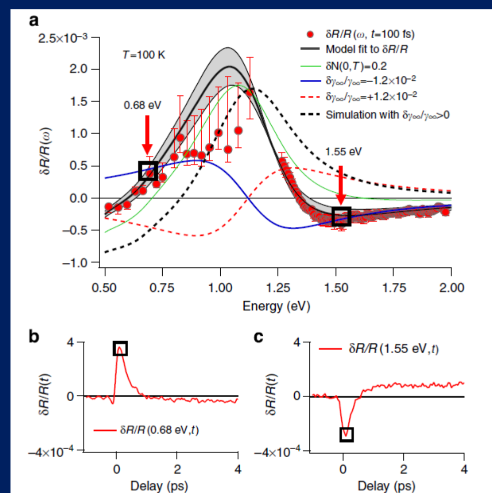
Can be characterize the dynamics of QP generation ?

$Bi_2Sr_2Ca_{0.92}Y_{0.08}Cu_2O_{8+\delta}$,
20 K
pump 1.55 eV
probe: 1.2–2.2 eV

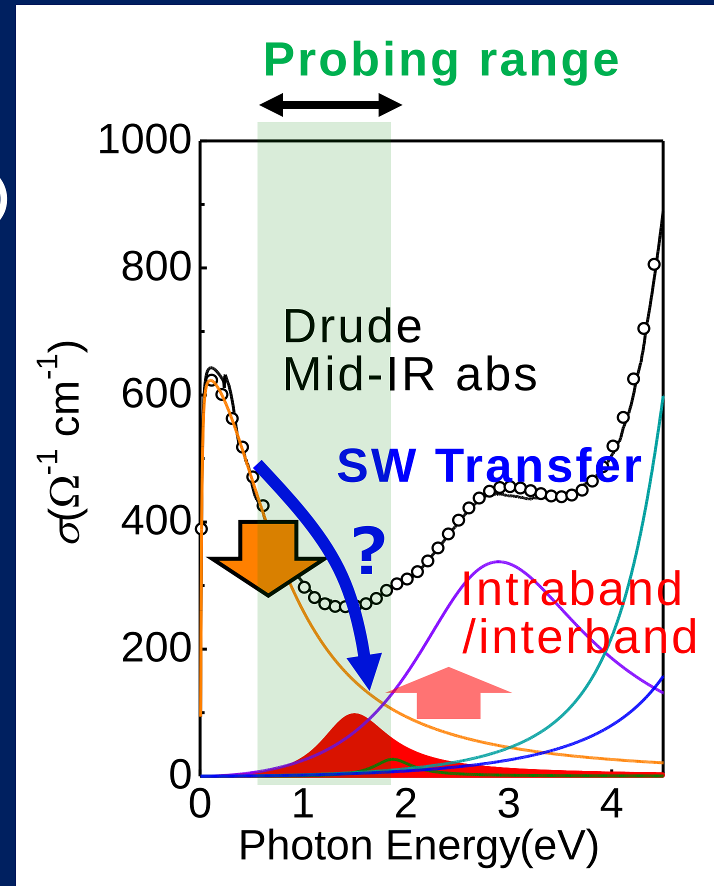


C. Giannetti *et al.*,
Nat. Commun.(2011).

$Bi_2Sr_2CaCu_2O_{8+\delta}$,
100 K
pump: 1.55 eV
probe: 0.5–2.0 eV)



F. Cilento *et al.*,
Nat. Commun.(2014).



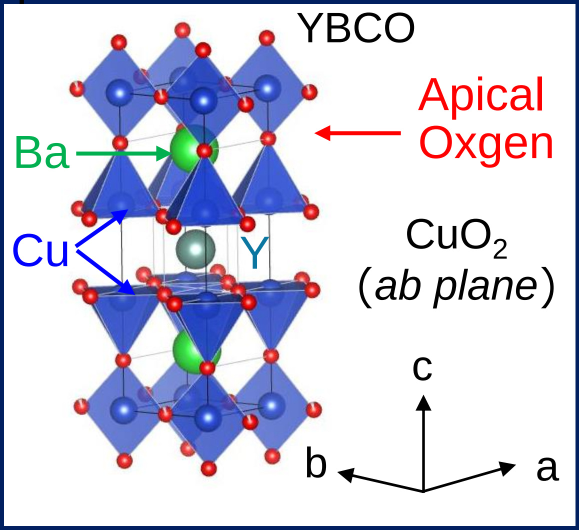
Optimally dope
Increase of SW in NIR-Vis.

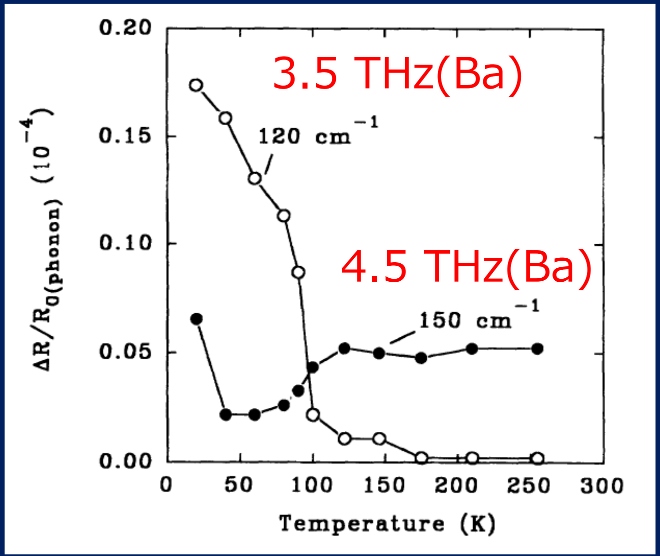
From ext. Drude (mid-IR) to Interband tr. ?

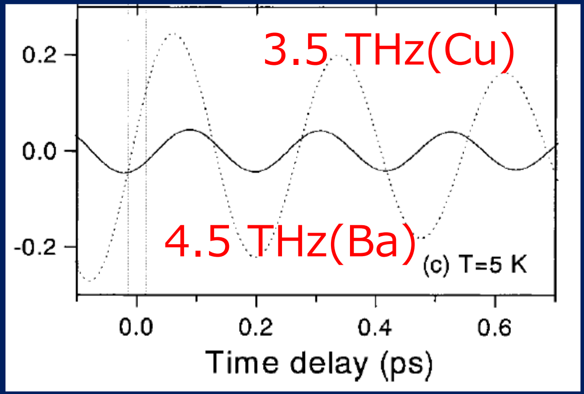
- **Expansion of spectral range is necessary**
- **SW transfer reflects the QP dynamics ?**

3.5 & 4.5 THz coherent phonons (Ba and Cu)

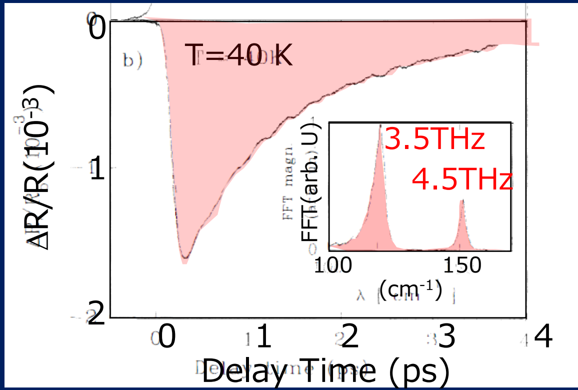
Y







O. V. Misochko et al.
PRB61, 4305 (2000)
25 fs pulse

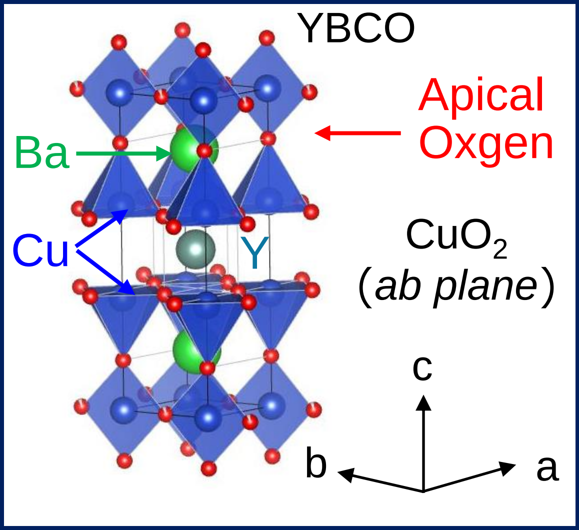


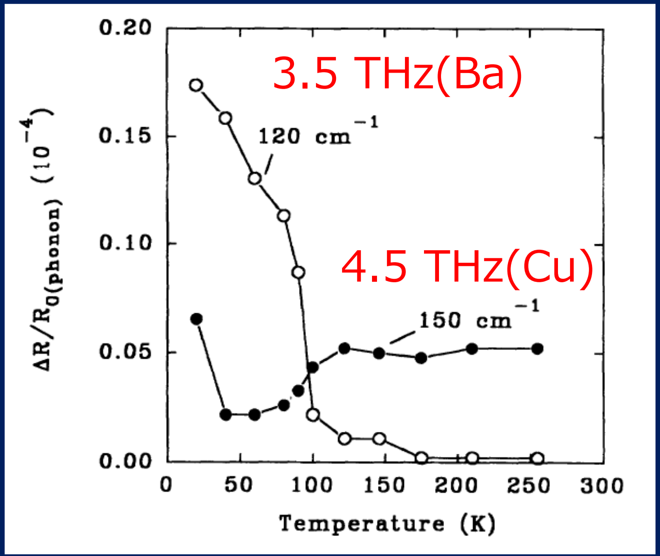
W. Albrecht et al.
PRL69, 1451 (1992)
60 fs pulse, 2 eV

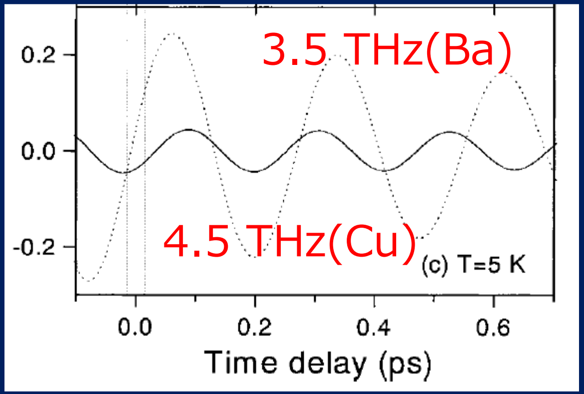
4.5 THz → Cos like (DECP ?)
3.5 THz → Sin like (ISRS ?)

✓ Sensitive to SC (3.5 THz)
backling of CuO_2 plain and e-ph interactions

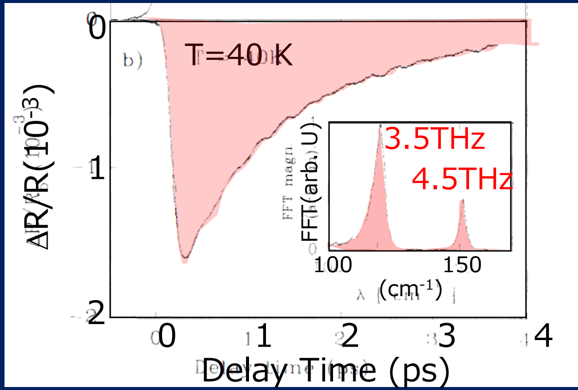
3.5 & 4.5 THz coherent phonons (Ba and Cu)







O. V. Misochko et al.
PRB61, 4305 (2000)
25 fs pulse



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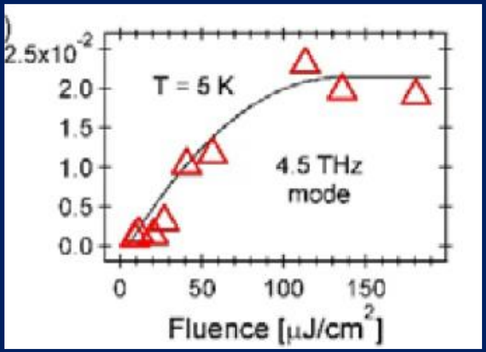
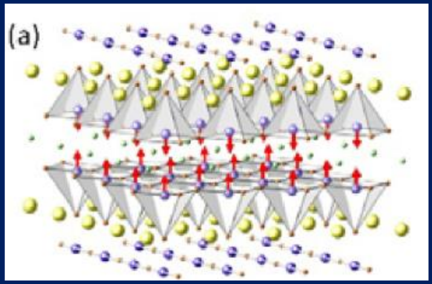
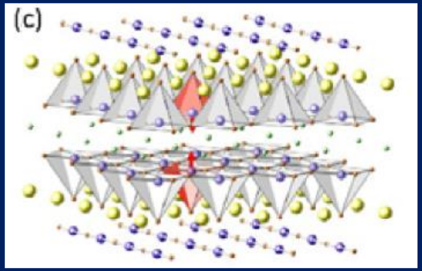


F. Novelli et al.
Phys. Rev.B95, 174524 (2017)



3.5 THz (Ba)
→ macroscopic



4.5 THz (Cu)
→ local mode

✓ Probe energy dependence of CP
✓ DMFT calculation

Objectives: Generation dynamics of QP

Target material $YBa_2Cu_3O_y$ (T_C~92 K)

1. Spectral weight transfer
(From where ?, To where ? , How fast ?)

✓ *Transient reflectivity 0.1 – 4 eV (100 fs)*

✓ *Spectrum analysis (Drude · Lorentz model)*

✓ Time scale of SW transfer
(6 fs pulse measurement)

2. Build-up mechanism of coherent phonons
(*build-up times of coherent phonon & SW ransfer)*

✓ *Modulation during 100-200 fs*

ΔR/R spectrum YBCO 0.1～4 eV (T_c=92 K)

** Using 100 fs pulse*

Previous studies on SC YBCO

1) S. G. Han *et al*., PRL65, 2708(1990)
(1～4 μm films, 2 eV, 60 fs pulse)

2) G. L. Eesley *et al*., PRL65, 3445(1990)
(1 μm film, 2 eV, 80 fs pulse)

3) S. V. Chekalin et al., PRL67, 3860(1991)
(150 film/MgO, ～2 eV, 100 fs pulse)

4) J. Demsar, PRL82, 4918(1999)
(Bulk ～1.5 eV pump, 80 fs pulse)

5) F. Novelli et al.
Phys. Rev. B95, 174524 (2017)
(80 nm film/STO, 0.95 eV pump)

✓ < 1.3 eV, > 3 eV ; the first observation

ΔR/R spectrum YBCO 0.1～4 eV (T_c=92 K)

** Using 100 fs pulse*

✓ < 1.3 eV, > 3 eV ; the first observation

✓ characteristic to SC

ΔR/R spectrum YBCO (T_c=92 K, 10 K)

** Using 100 fs pulse*

Objectives: Generation dynamics of QP

Target material $YBa_2Cu_3O_y$ (T_C~92 K)

1. Spectral weight transfer

(From where ?, To where ? , How fast ?)

- ✓ *Transient reflectivity 0.1 – 4 eV (100 fs)*
- ✓ *Spectrum analysis (Drude · Lorentz model)*
- ✓ Time scale of SW transfer (6 fs pulse measurement)

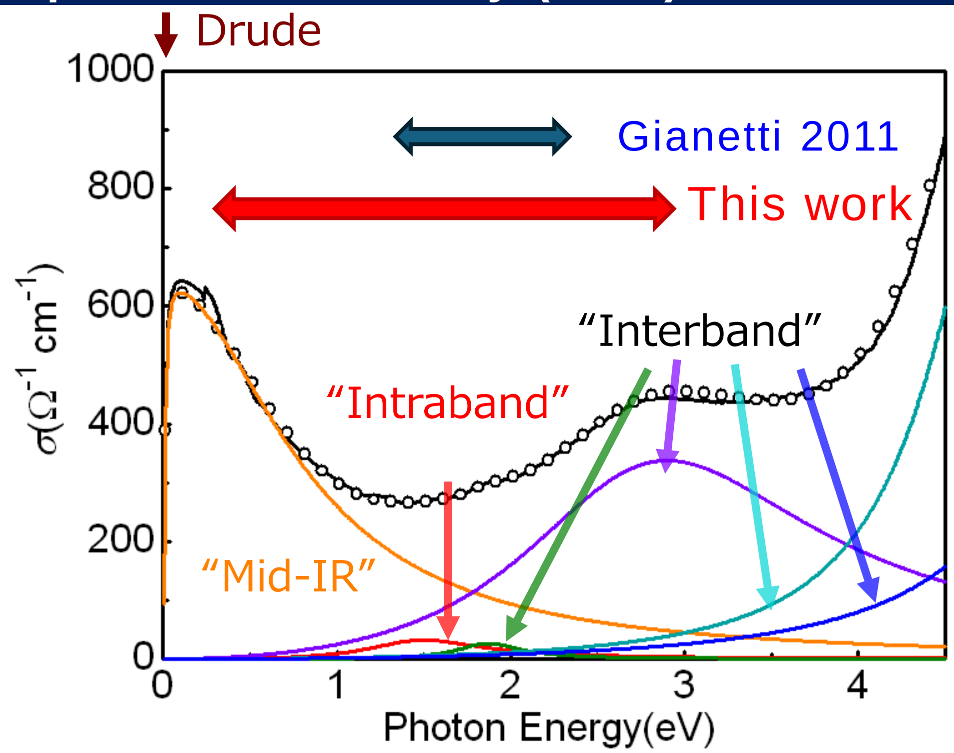
2. Build-up mechanism of coherent phonons

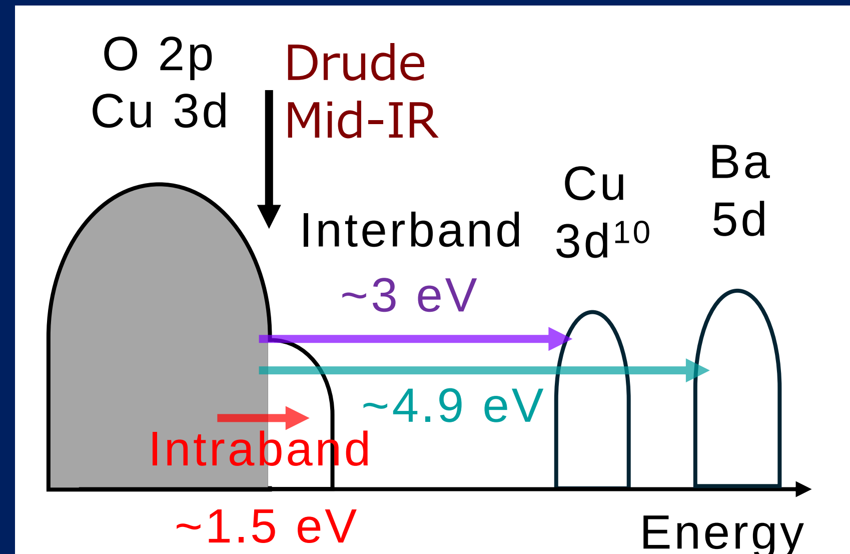
(*build-up times of coherent phonon & SW ransfer)*

- ✓ *Modulation during 100-200 fs*

Drude・Lorentz analysis (steady state)

Optical conductivity (10 K)





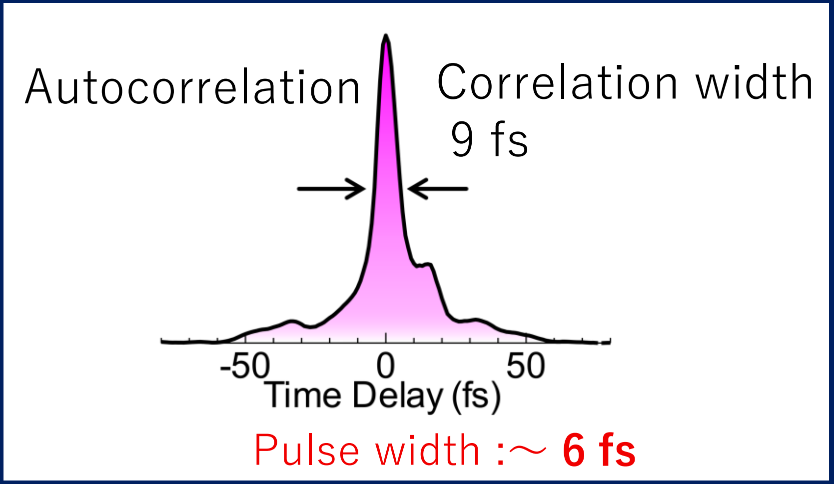
- **"Intraband": partially occupied O2p-Cu3d orbit**
- **But, in Bi2212, it is assigned to Zhang Rice singlet-Fermi level**

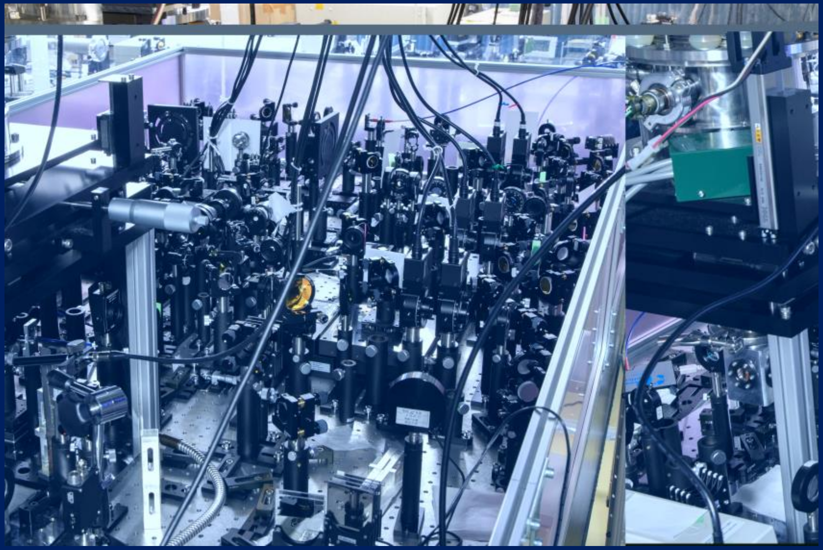
Quijada *et al* ., PRB 60, 14917 (1999).
Lee *et al* ., PRB 72, 054529 (2005).

Ishioka *et al* ., PRB 107, 184302 (2023).
Romberg *et al* ., Z. Phys. B Cond. Matter 78, 367 (1990).

Thank to
Christian Bernhard (Fribourg) , Jure Demsar (Mainz)

Dynamics of SW transfer and increase of γ





Photoinduced SW transfer ;
～100 fs ?

Dynamics during <100 fs time domain

Objectives: Generation dynamics of QP

Target material $YBa_2Cu_3O_y$ (T_C~92 K)

1. Spectral weight transfer

(From where ?, To where ? , How fast ?)

✓ *Transient reflectivity 0.1 – 4 eV (100 fs)*

✓ *Spectrum analysis (Drude · Lorentz model)*

✓ Time scale of SW transfer
(6 fs pulse measurement)

2. Build-up mechanism of coherent phonons

(*build-up times of coherent phonon & SW ransfer)*

✓ *Modulation during 100-200 fs*

$\Delta R/R$ measurement using 6 fs pulse

6 fs pulse

✓ $\Delta R/R<0$

→ "Mid-IR" scattering rate ↑

✓ **Oscillations**

- T 200～300 fs osc.
- 0–100 fs→ T ~ 30–40 fs

✓ $\Delta R/R>0$

→"Mid-IR", "Intreband" spectral weight ↑

$\Delta R/R$ measurement using 6 fs pulse

6 fs pulse

100 fs pulse

✓ $\Delta R/R<0$ (< 10 fs buildup)

→ "Mid-IR" scattering rate ↑

✓ $\Delta R/R>0$ **(~70 fs buildup)**

→"Mid-IR", "Intreband" spectral weight ↑

Time scales of SW transfer is determined as ~70 fs

Objectives: Generation dynamics of QP

Target material $YBa_2Cu_3O_y$ (T_C~92 K)

1. Spectral weight transfer
(From where ?, To where ? , How fast ?)

✓ *Transient reflectivity 0.1 – 4 eV (100 fs)*

✓ *Spectrum analysis (Drude · Lorentz model)*

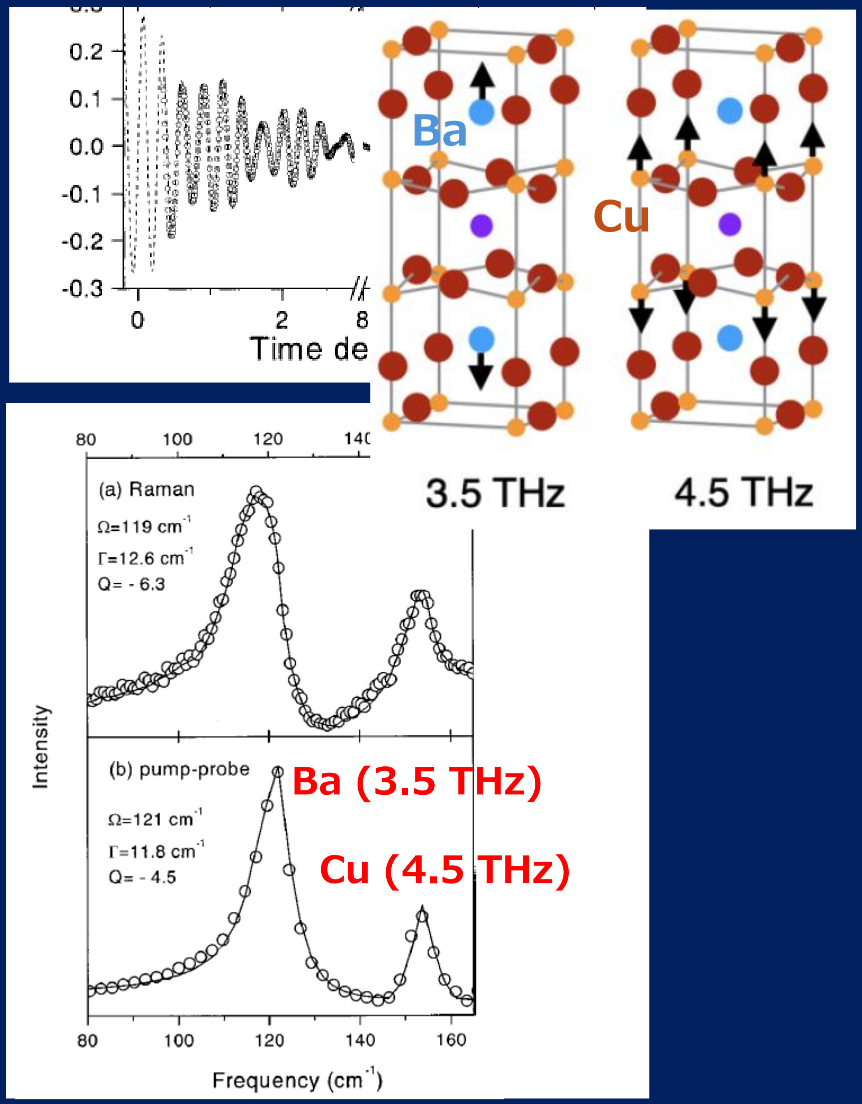
✓ Time scale of SW transfer
(6 fs pulse measurement)

2. Build-up mechanism of coherent phonons
(*build-up times of coherent phonon & SW ransfer)*

➡✓ *Modulation during 100-200 fs*

3.5 & 4.5 THz coherent phonons (Ba and Cu)

80 K



O. V. Misochko et al. PRB 61, 4305 (2000)

Summary (YBCO)

1. **Spectral weight transfer (From where ?, To where ?)**

✓ $T<T_c$:

Interband (1.9 eV, 2.9 eV)
→ Intraband(1,5 eV) and mid-IR(0.1 eV)

✓ $T>T_c$:

Interband (4.9 eV) and mid-IR(0.1 eV)
→ Interband(2.9, 1.9 eV)
and Intraband (1.5 eV)

2. **<100 fs dynamics (6 fs p-p)**

✓ **SW transfer ~70 fs**

✓ **3.5 THz (Ba) modes**
is related to SW transfer ?

✓ **Apical Oxgen CP**
is enhanced below Tc

✓ **30-40 fs oscillation**
2-phonon ? magnon ?

Ultrafast precursory dynamics of photoinduced insulator to metal transition

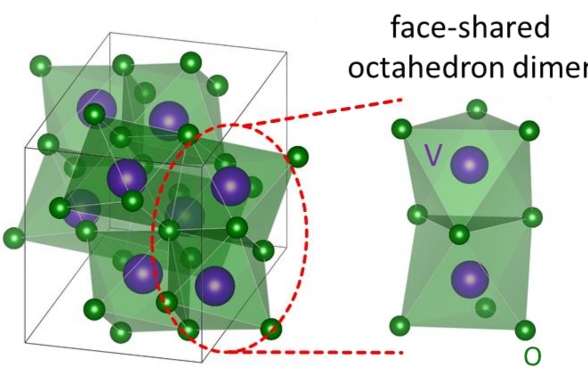
Tohoku University Shinichiro Iwai

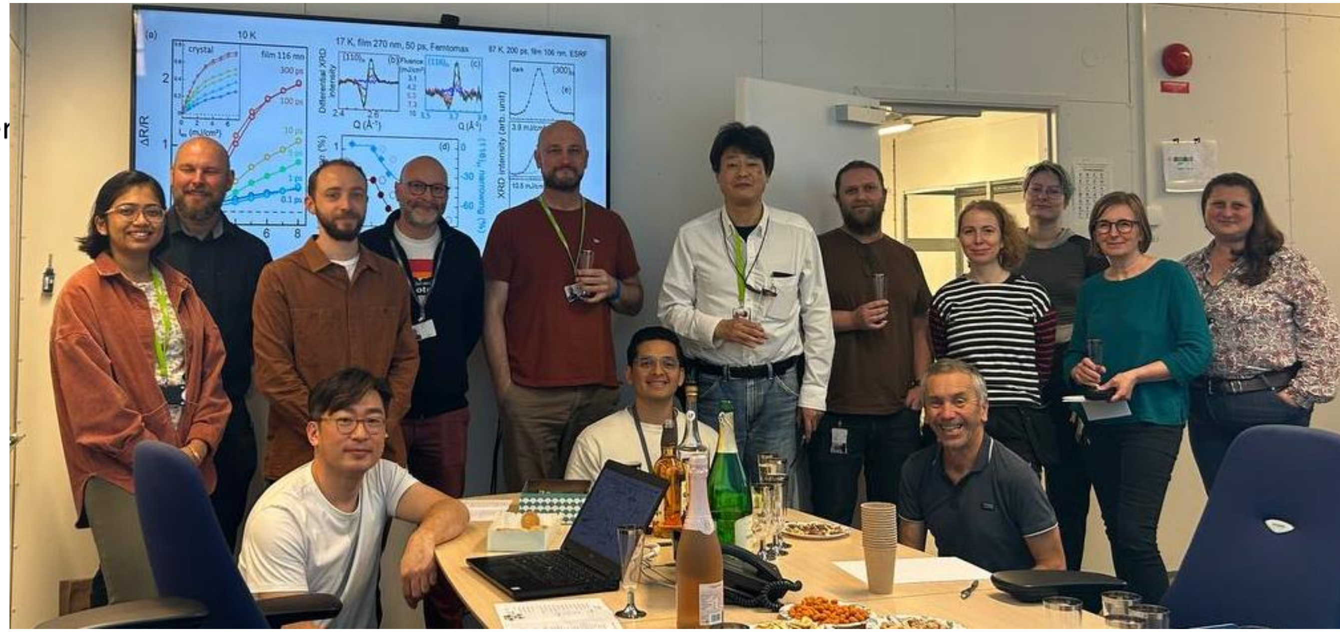
mano

M. Lorenc

E. Janod

H. Cailleau





Amano et al.,
Nat. Phys. accepted

Outline

i) Electronic and Crystal Structures in V_2O_3

ii) Thermal vs. nonthermal PIPT in V_2O_3

iii) Moivation & strategy

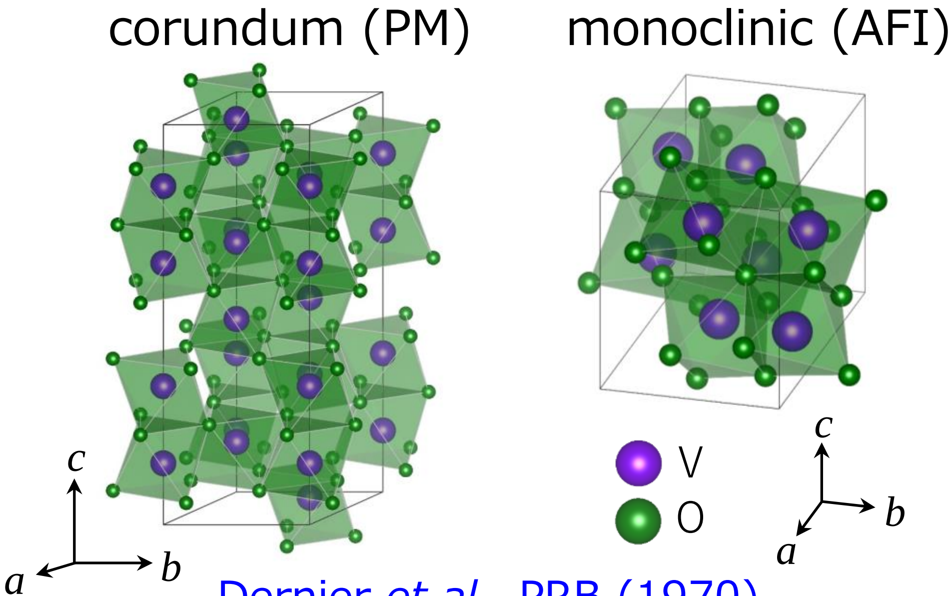
iv) Dynamics of optical gap (100 fs pulse)

- 100 % phase transition & cooperativity
- Structural dynamics (transient XRD)
- Coherent prpopagation of I-M trasition

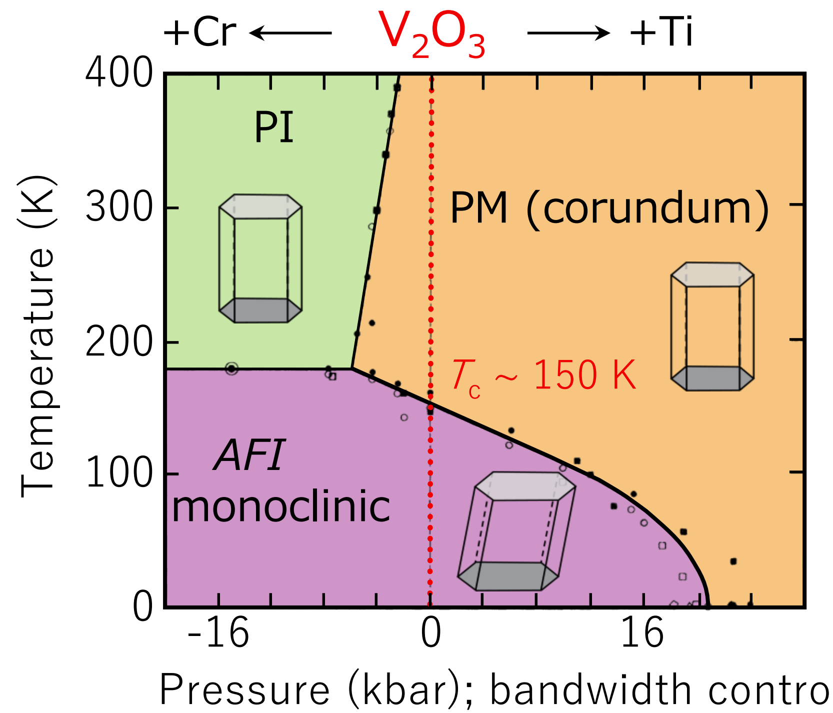
v) Precursory dynamics captured by 6 fs pulse

- I_{ex} dependence of coherent phonons (amplitude, decay, initial phase)

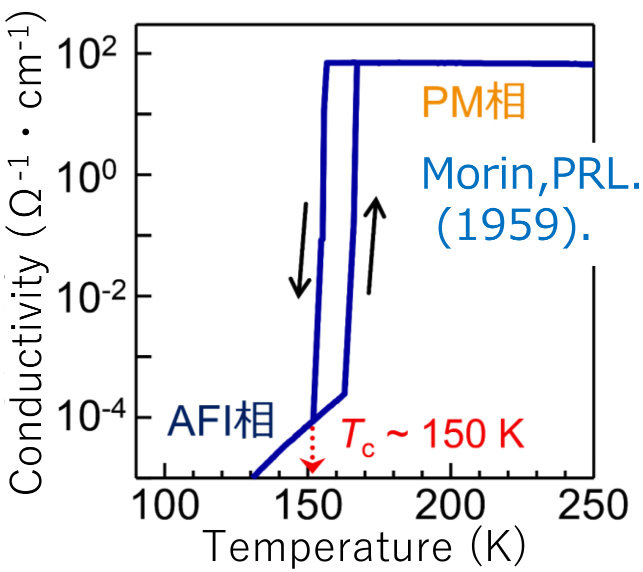
V_2O_3 ; "textbook" Mott insulator, but not so simple



Dernier *et al*., PRB (1970).
Robinson, Acta Crystallogr. B. (1975).

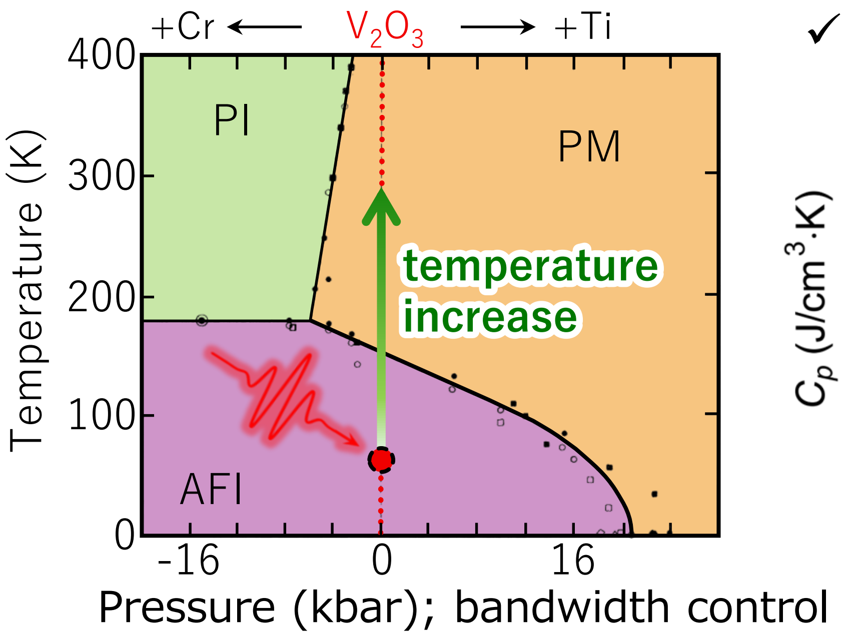


McWhan *et al*., PRL (1971).

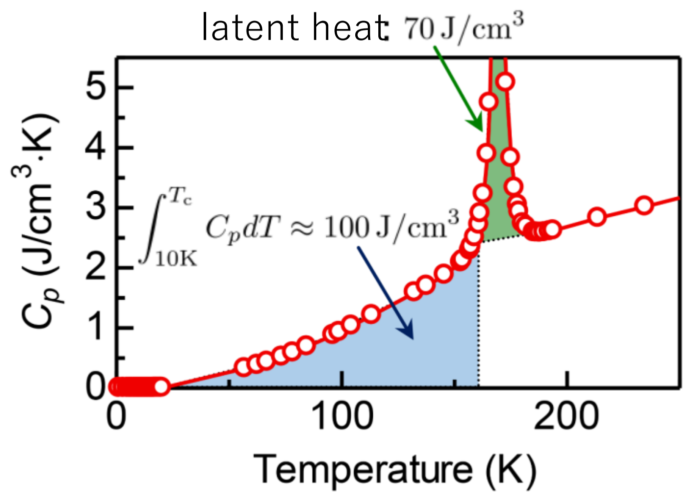


- **AFI-PM: Symmetry change & Volume change**
- **Electron correction and electron-phnon interaction cooperatively work**
- heat capacity: Anderson, J. Am. Chem. Soc. (1936).
- neutron scattering: Word *et al*., PRB (1981).
- Raman scattering: Kuroda *et al.*, PRB (1977).
- optical conductivity: Barker *et al*., Solid State Commun. (1970).

Early studies on PIPT (thermal process)



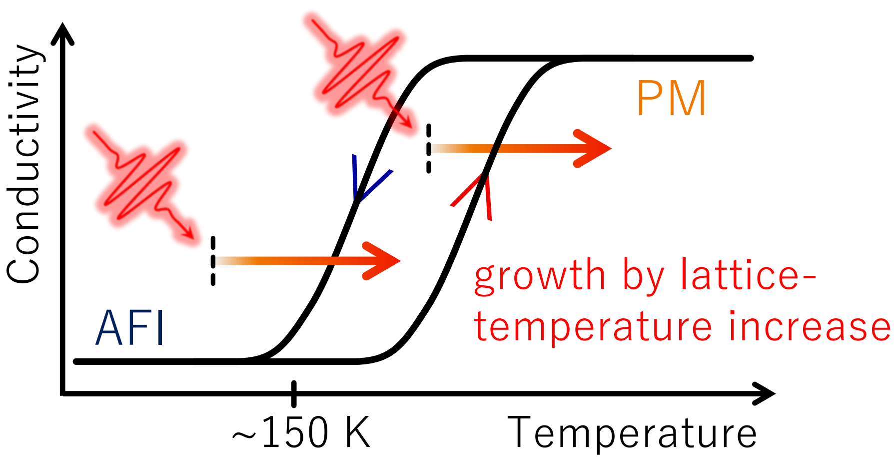
✓ **thermally-driven I-M transition**



temperature increase (10 K-) ~ 100 J/cm^3

latent heat ~ 70 J/cm^3

Total ~ 170 J/cm^3

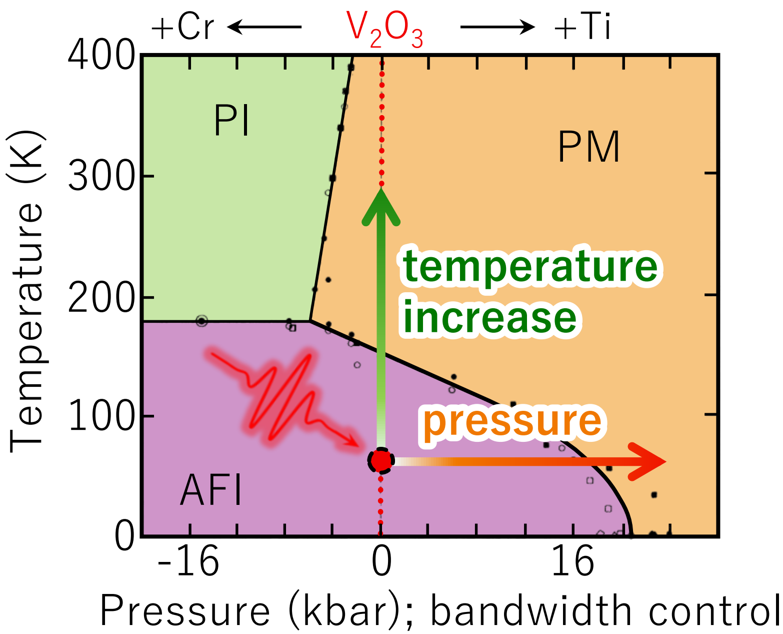


Transient (optical) abs. spectroscopy
(epitaxial film: AFI) Liu *et al*., PRL (2011).
Abreu *et al*., PRB (2015).

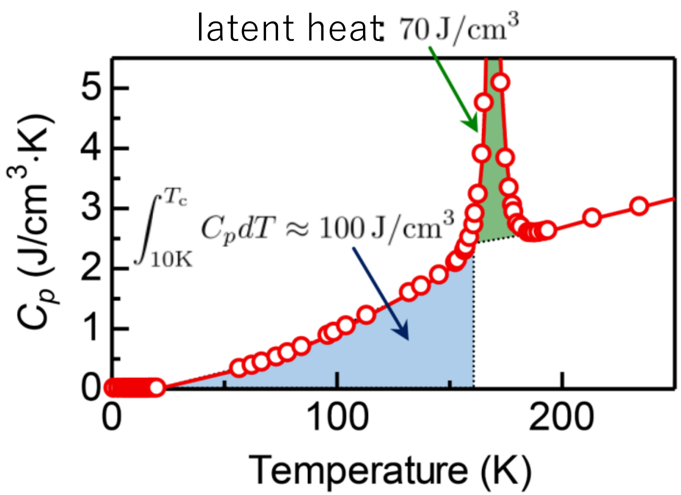
Transient X-ray scattering
(epitaxial film: AFI) Singer *et al*., PRL (2018).

- **Near the transition temp.**
- **Nucleation growth by laser heating**

Thermal vs. Pressure (non-thermal)



✓ **thermally-driven I-M transition**

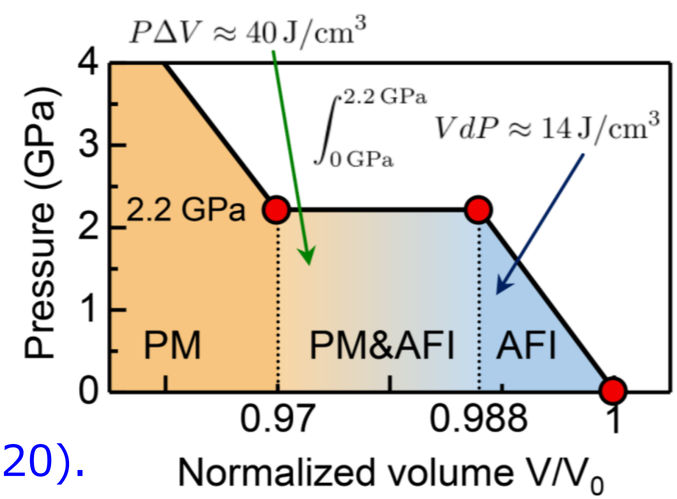


temperature increase (10 K-) ~ 100 J/cm³

latent heat ~ 70 J/cm³

Total ~ 170 J/cm³

✓ **pressure-driven I-M transition**



heat(AFI-PM) ~ 54 J/cm³

D. Babich, PhD thesis, University of Nantes (2020).

(Light induced) Pressure-driven IMT is more likely to occur ?

Moivation

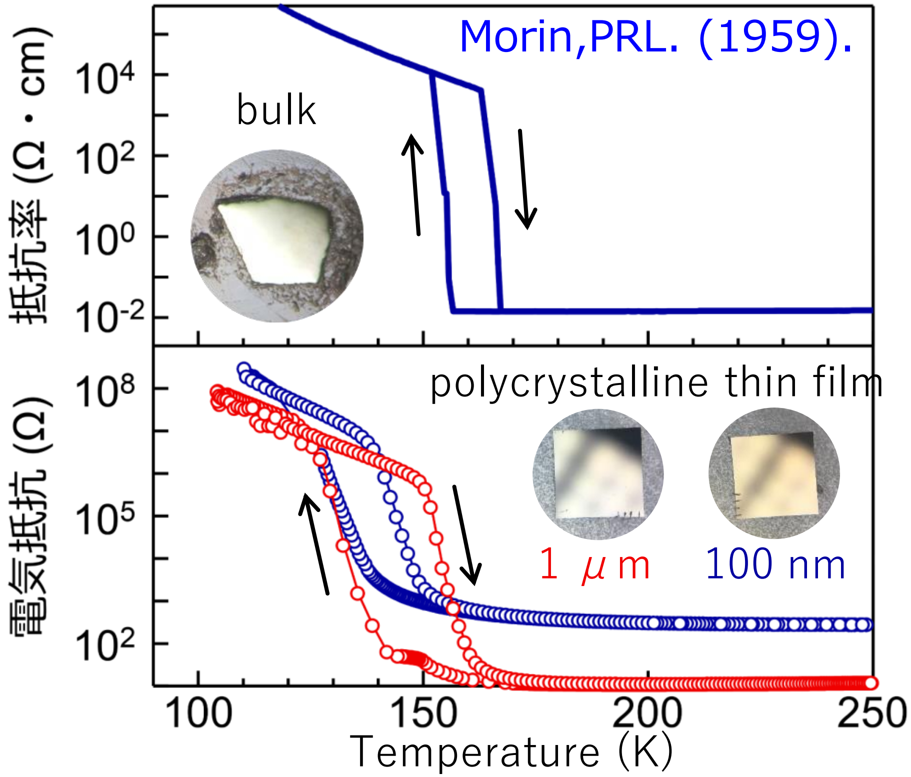
To demonstrate nonthermal IMT in V_2O_3

Strategies

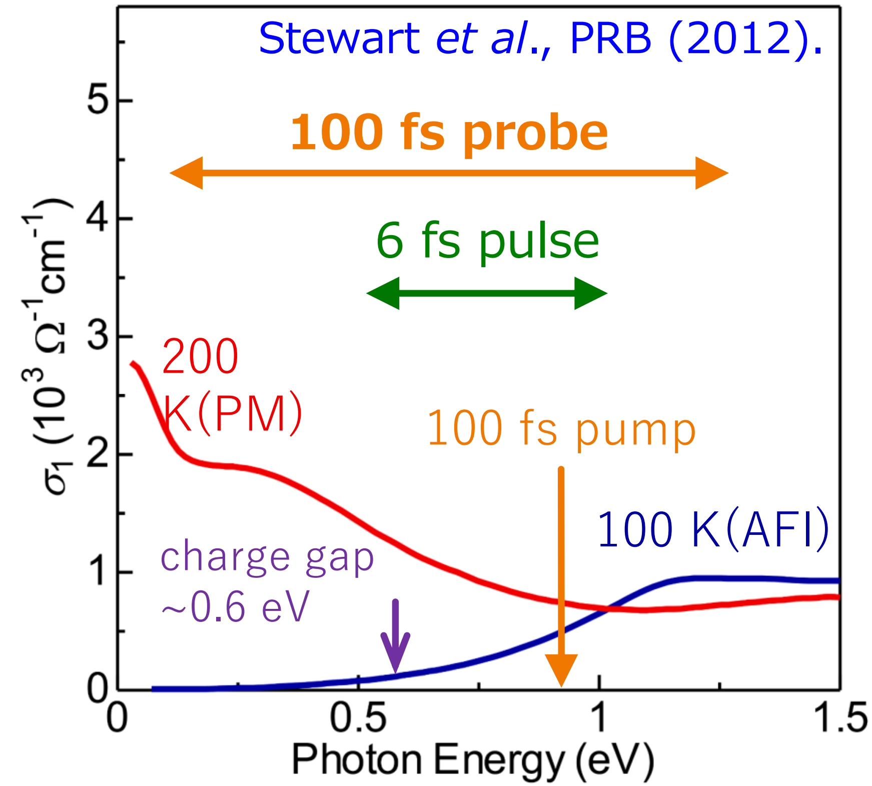
- Low Temperature (10 K, Laser heating does not reach T_c)
- Optical detection to measure the gap closing
  (mid-IR～NIR 100 fs, 6 fs pulses)
- Time resolved XRD to confirm symmetry and volume changes
  (Time resolution ～200 fs)
- Nanocrystal (domain 20 nm)

Experimental conditions

Temperature dependences of conductivity



Optical conductivity spectra



Sample

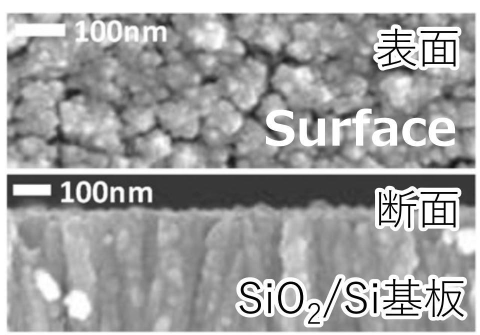
i) bulk crystal

- || c-axis, ⊥c-axis

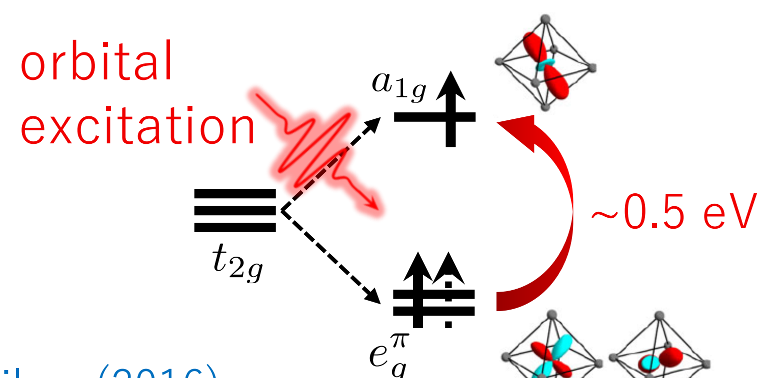
ii) polycrystalline thin film

thickness: 116, 270 nm

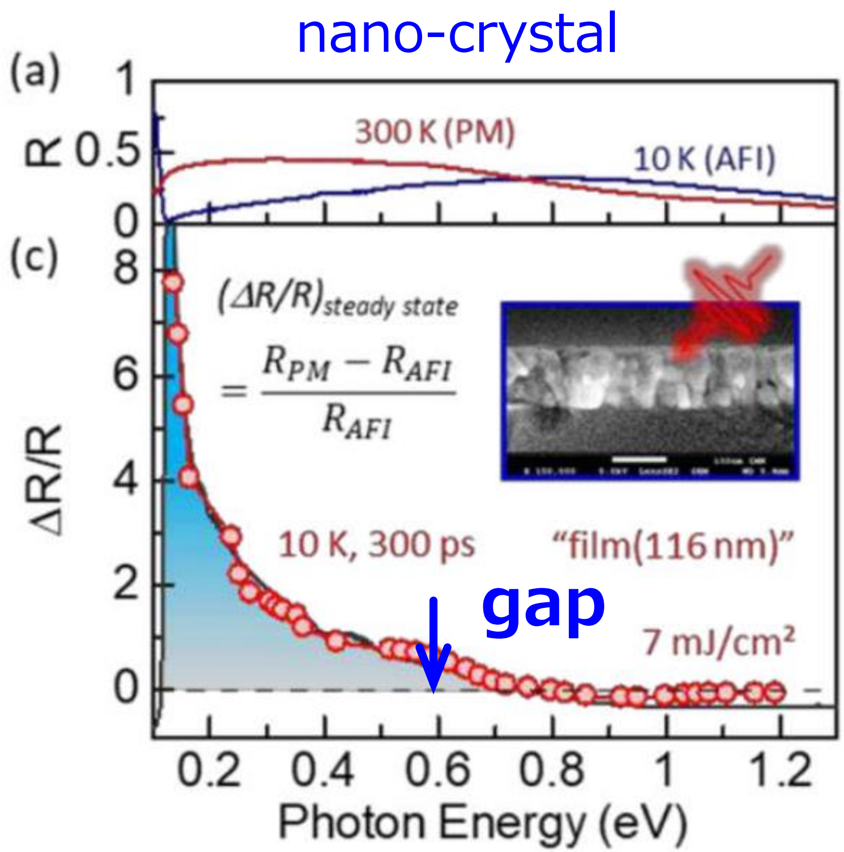
domain size: ~20 nm(AFI)

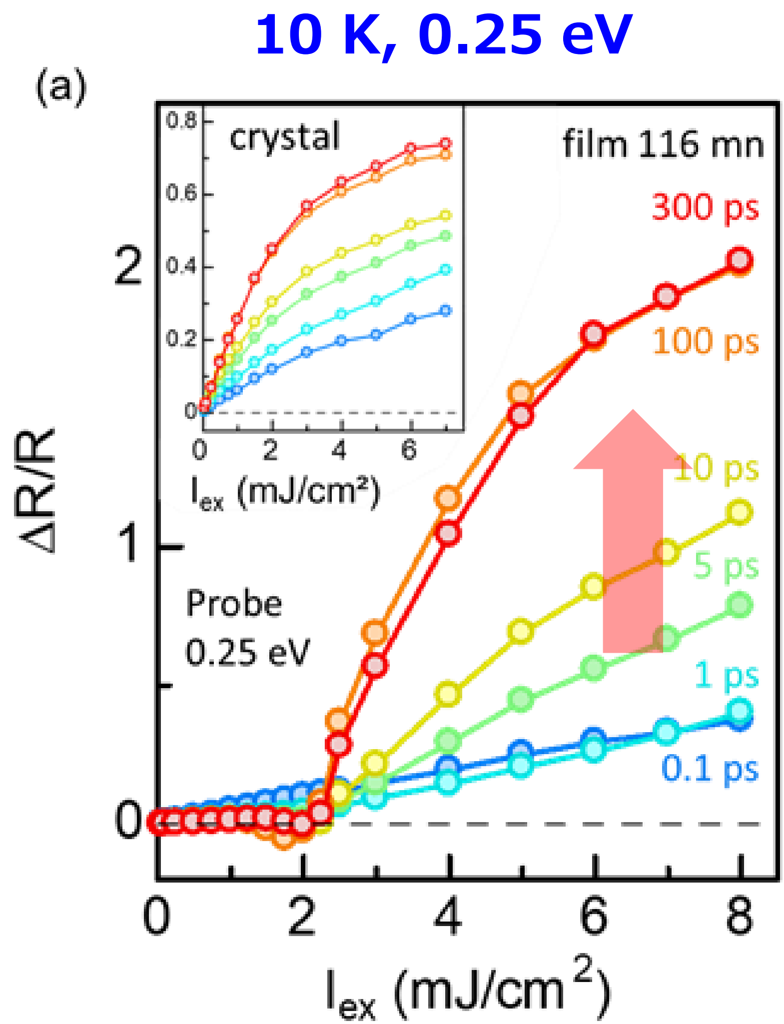


Querré *et al*., Thin Solid Films (2016).



PIPT in V_2O_3 nanocrystal vs. bulk at 10 K

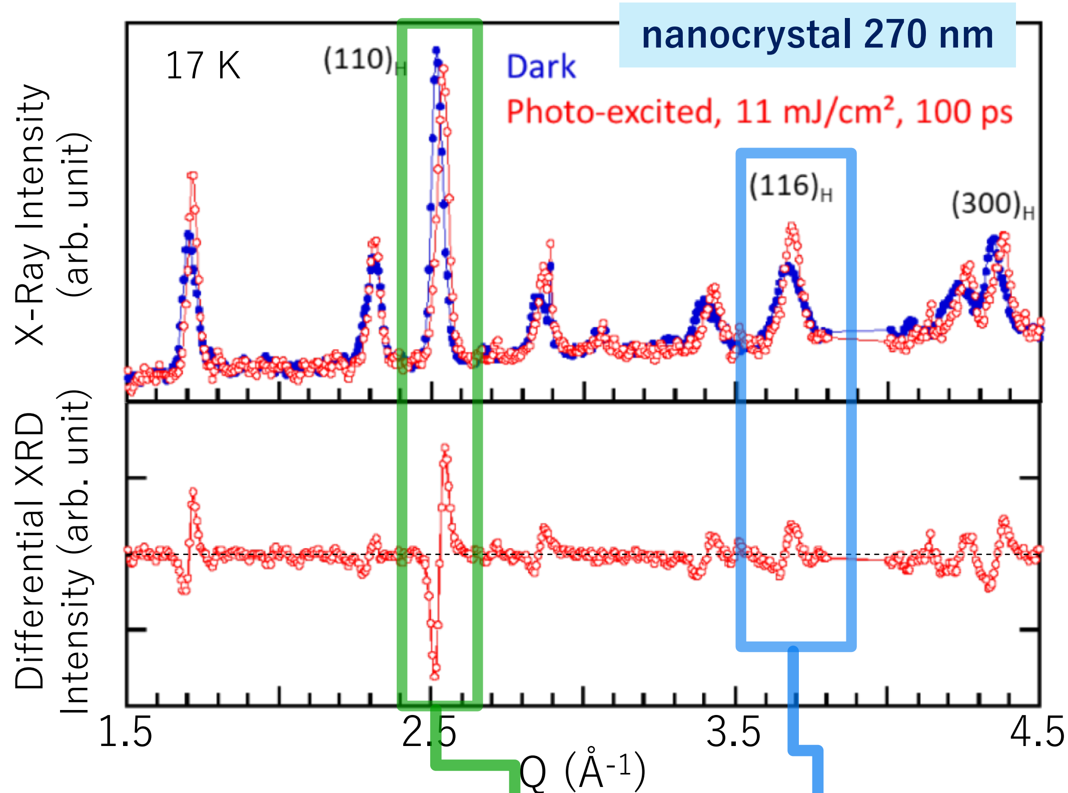




Amano et al.,
Nat. Phys. accepted

optical gap is closed in 30ps (nano-crystal)

Transient XRD （symmetry & volume）



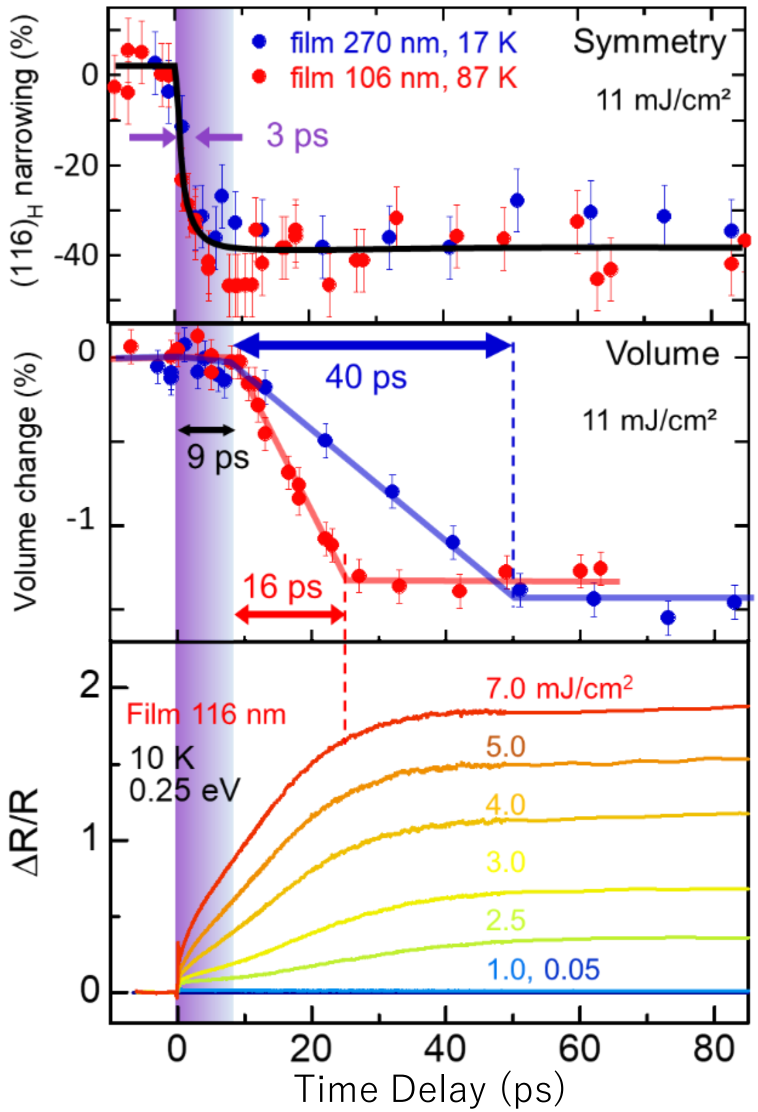
	V-V distance	Symmetry
PM	2.700 Å	Colundam
AFI	2.745 Å	monoclinic

Amano et al.,Nat. Phys. accepted

FemtoMAX

- Pump: 0.95 eV (from OPA)
- Time resolution ~210 fs

Transient XRD : Synmetry & Volume changes



Symmetry change

Monoclinmic→Corundum

・**completed in ~3 ps**

v_{sound}=4.2 nm/ps (transverse)

Domain size ～20 nm

Yelon and Kenn,

Solid State Comm. 29, 775 (1979)

Volume decrease (～1.5 % I→M)

Delayed ~9 ps (incubation time)

rise time depends on film thicjness

d = 106 nm	**~16 ps**
d = 270 nm	**~40 ps**

v_{sound}=7.4 nm/ps (Longitudunal)

independent of temperature

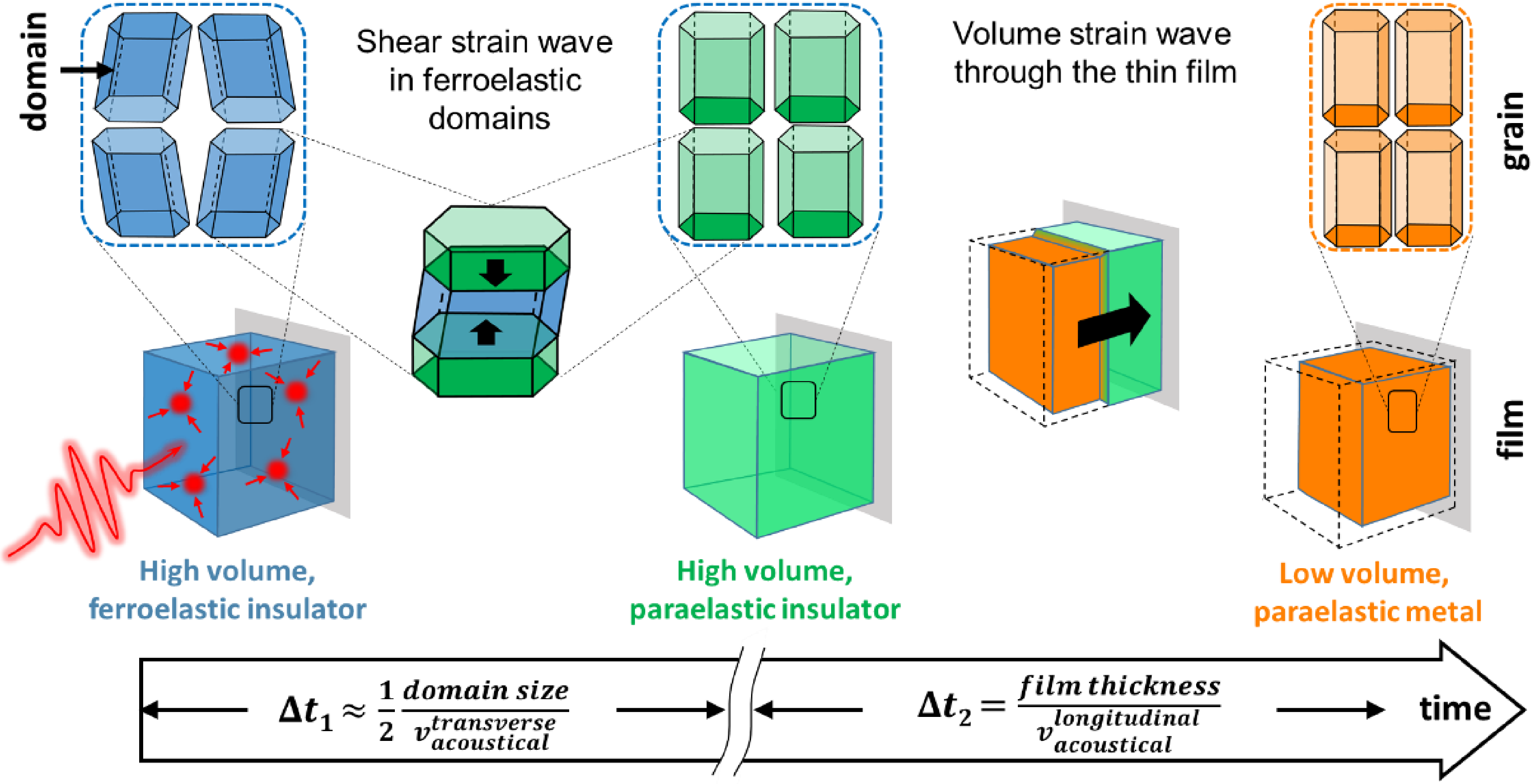
→ **nonthermal process**

Amano et al., Nat. Phys. accepted

Summary

strainwave
in domain (Transverse)
v_{sound}=4.2 nm/ps

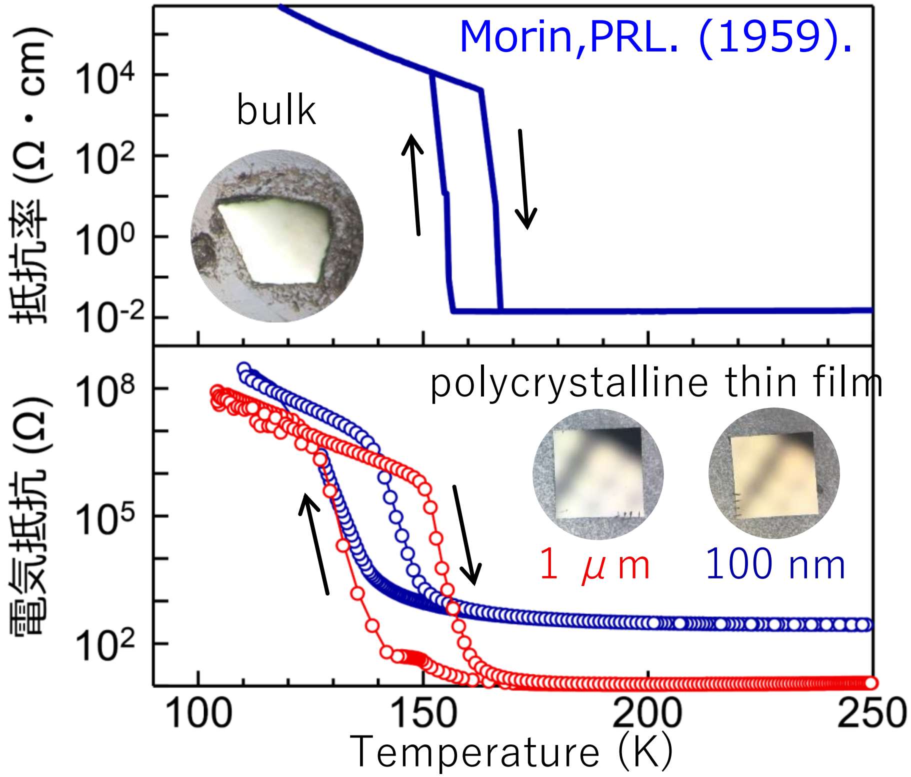
strainwave
in thin film (longitudinal)
v_{sound}=7.4 nm/ps



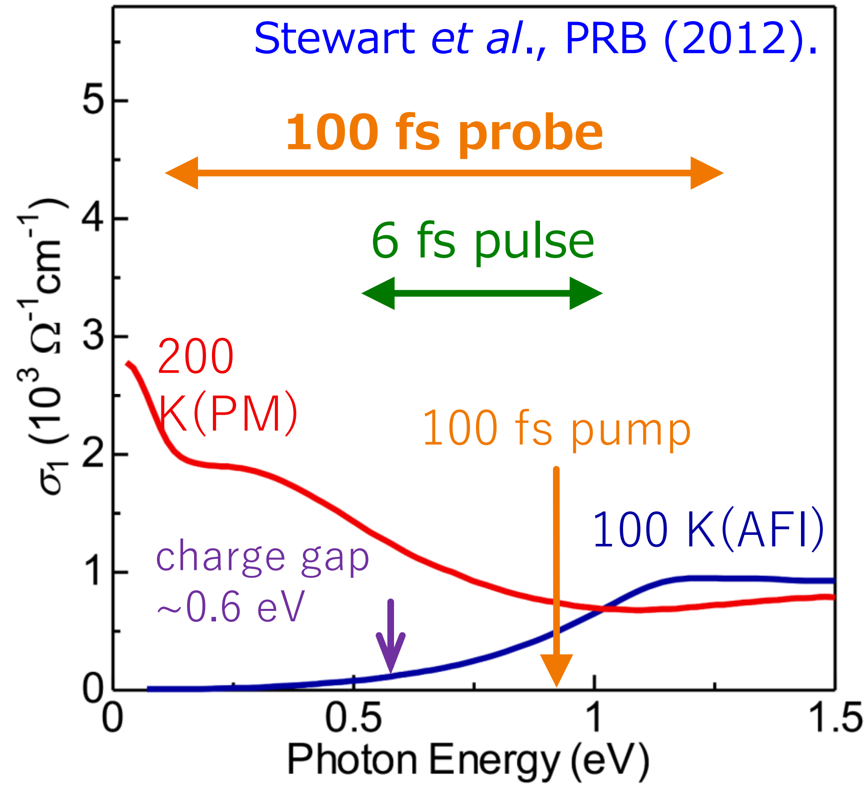
strainwave
in domain (Transverse)

Experimental conditions

Temperature dependences of conductivity



Optical conductivity spectra



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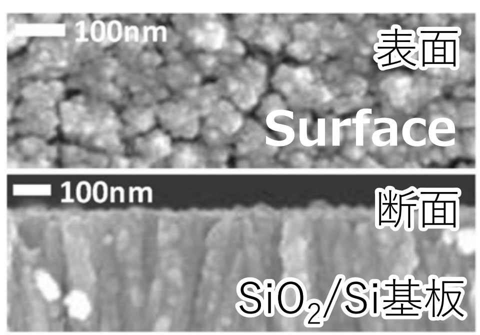
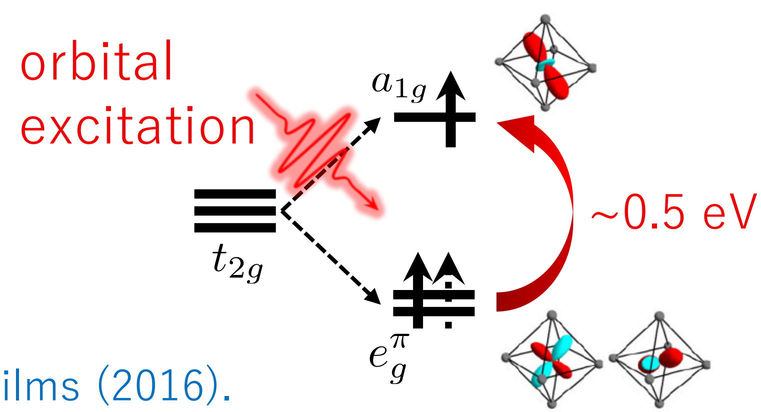
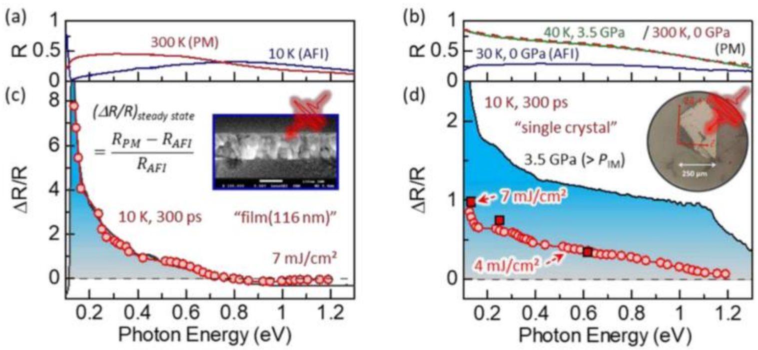


Querré *et al.*, Thin Solid Films (2016).



Summary (V_2O_3)

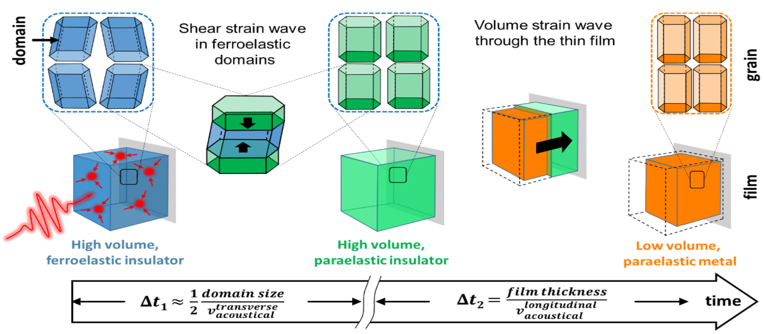


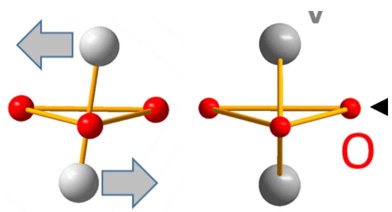
1: Mott gap dynamics

- 100 % change of ΔR/R only in the nanocrystal
- Gap closing is characterized by 30ps (100 nm)
- Temporal change of isobestic point

2: Structural dynamics

- Symmetry change < 3 ps
- Volume change 25 ps (106 nm), 50 ps(270 nm)
- Incubation time for volume change (~9 ps)



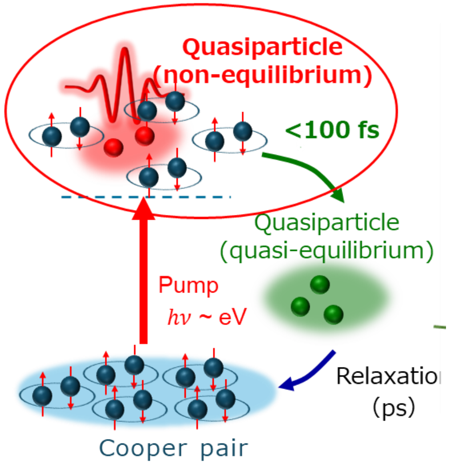


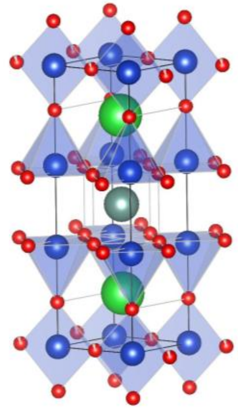
3: precursory dynamics

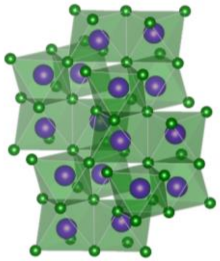
- 3 characteristic V-V modes (237, 276, 322 cm^{-1})
- Only 237 cm^{-1} (shear) mode shows nonlinearity
- 237 cm^{-1} also shows lifetime shortening (< 1/10 !)

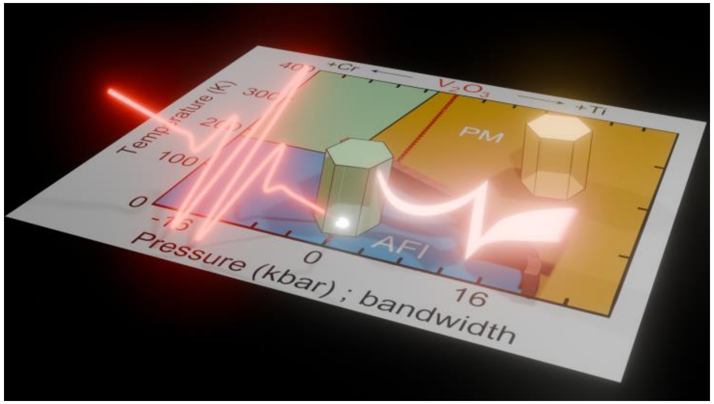
Immediate dissapearence of the af interaction ?

Summary









High-T_c SC cuprate (YBCO)

Kato et al.,
Ultrafast Phenomena 2024

Ferroelastic Mott insulator V_2O_3

Amano et al.,Nat. Phys. accepted

Ultrafast dynamics (toward < 1 fs) can clarify the microscopic mechanmism of symmetry change