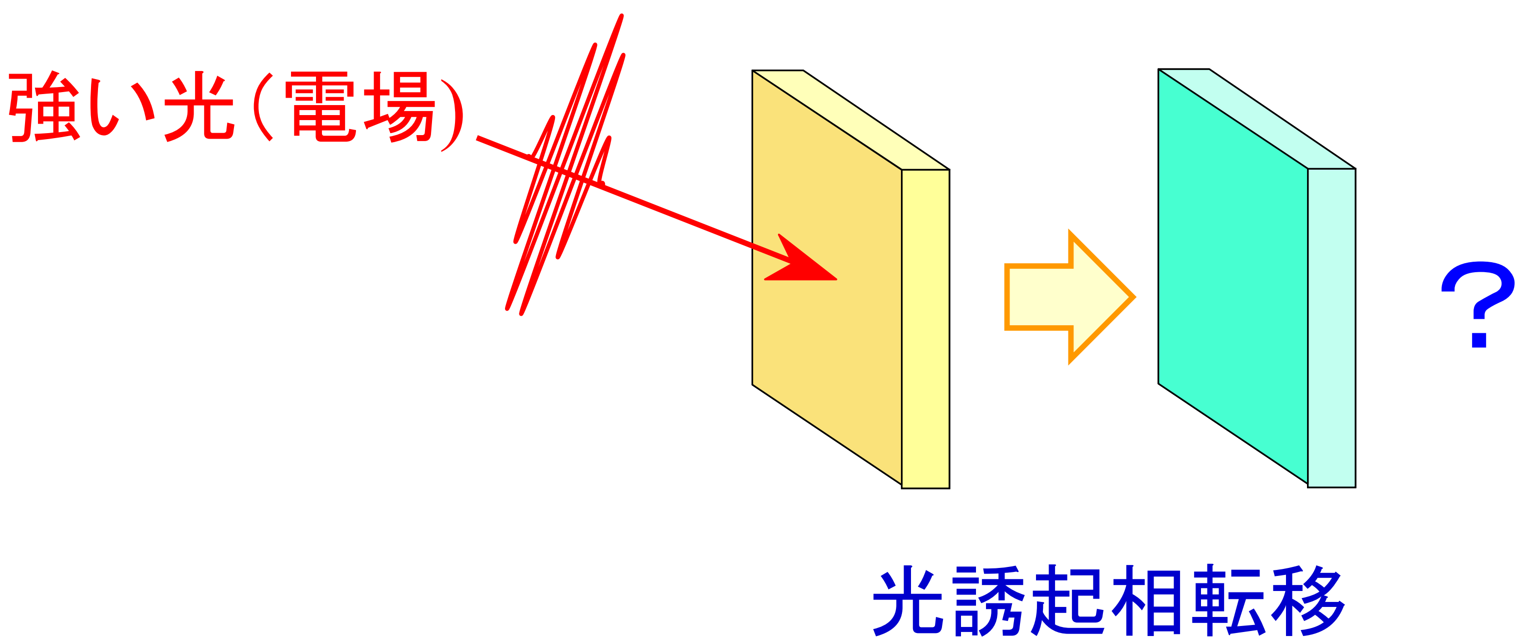
強相関系の光・電場応答と相転移

東大 新領域，産総研OIL

岡本　博



１．イントロダクション

強相関系の光誘起相転移

２．光誘起モット絶縁体－金属転移

極短光パルスによる電荷･スピンダイナミクスの検出

３．テラヘルツ(電場)誘起モット絶縁体－金属転移

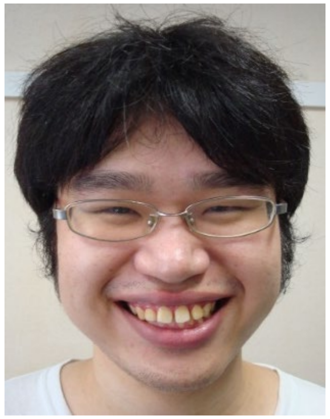
量子トンネル過程によるキャリア生成と金属化の検出

４．中赤外(強電場)パルスによる電子相制御への展開

強誘電分極制御とイオン性－ 中性転移

共同研究者

- フェムト秒レーザー分光


宮本辰也


森本剛史


山川大路


園　直樹

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北尾貴之，近藤秋洋，山川貴士，鈴木博貴，貴田徳明（東大新領域）

足立俊輔，渡部裕也（京大理）

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加藤礼三（理研），宮川和也，鹿野田一司（東大工）

- 理論

石原純夫（東北大理），遠山貴巳，新城一矢（東京理科大）
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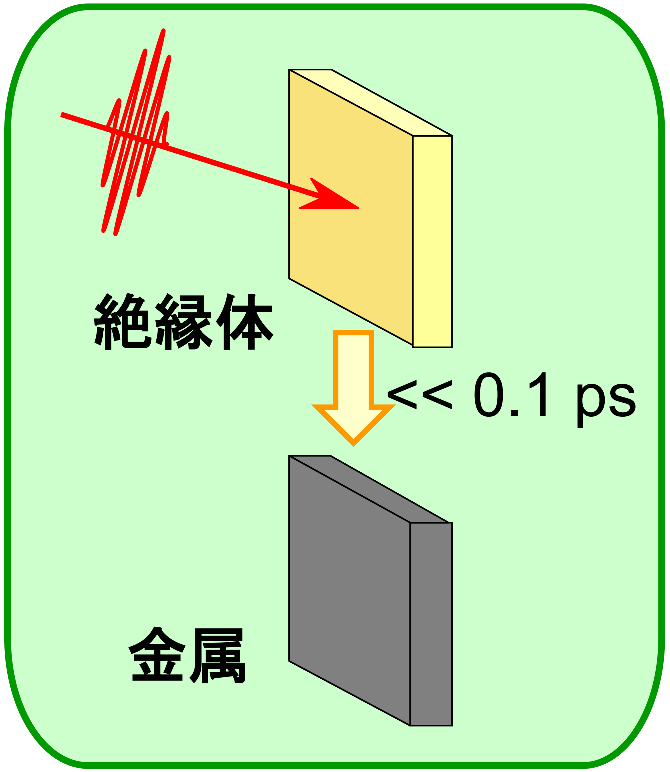
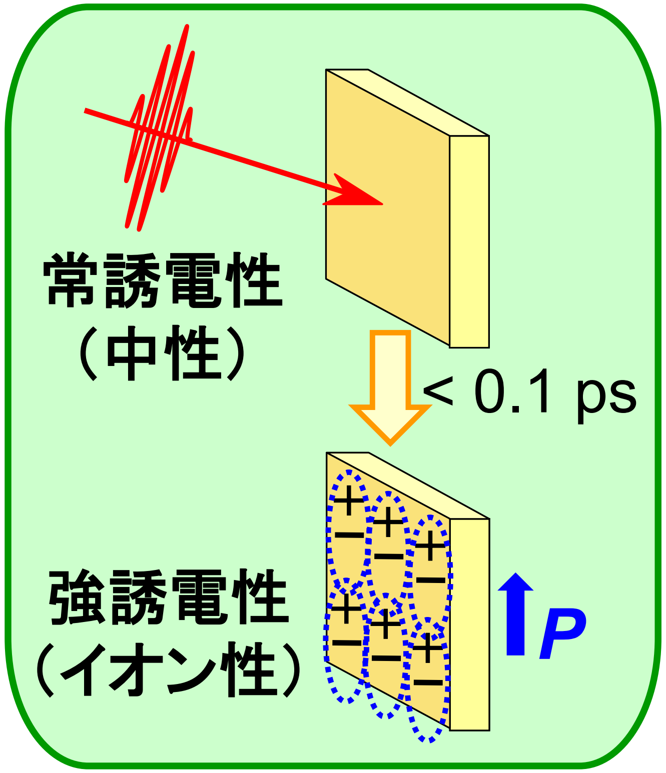
光誘起相転移

光誘起金属化



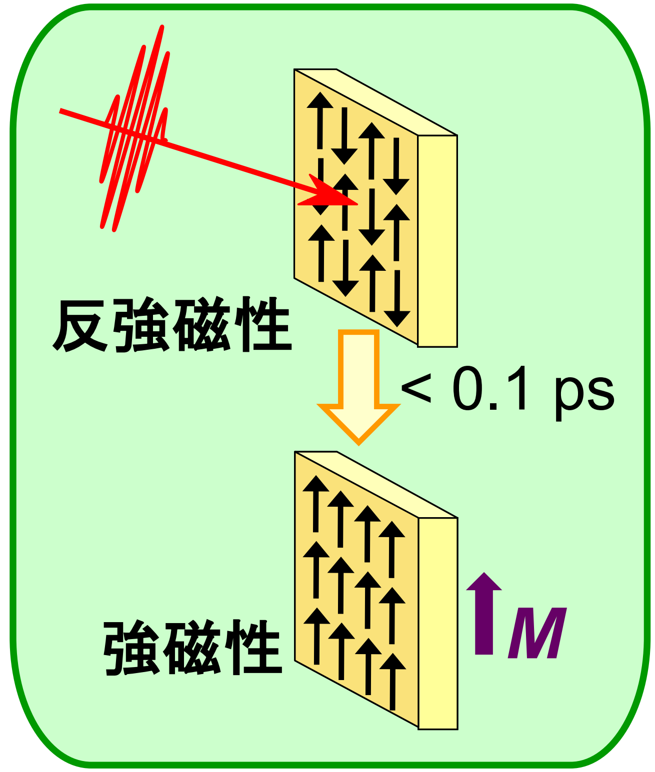
ex. 遷移金属錯体
$[Ni(chxn)_2Br]Br_2$

光誘起強誘電性



ex. 有機分子化合物
TTF-CA

光誘起強磁性

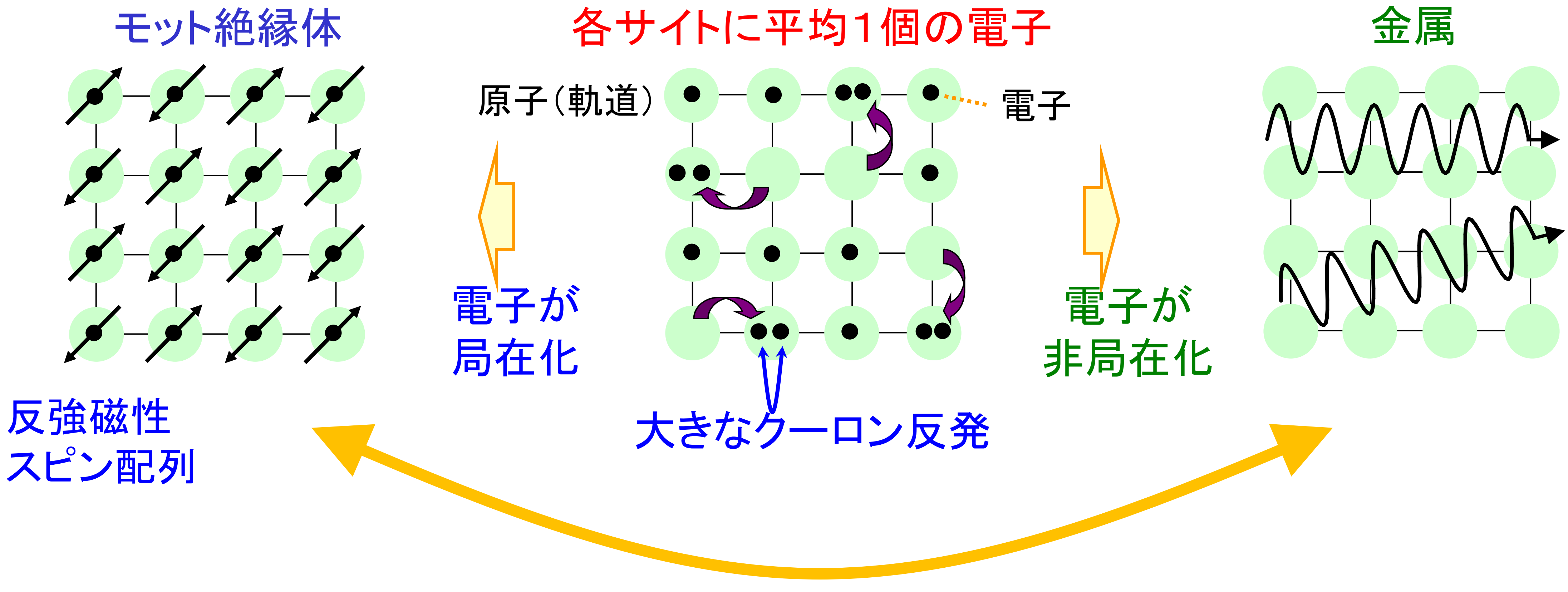


ex. マンガン酸化物
$Gd_{0.55}Sr_{0.45}MnO_3$

非平衡量子物理学 → 新しい学問分野
超高速の物性制御 → 新しい光デバイスの可能性

強相関（電子）系 ： 電子間に強いクーロン相互作用

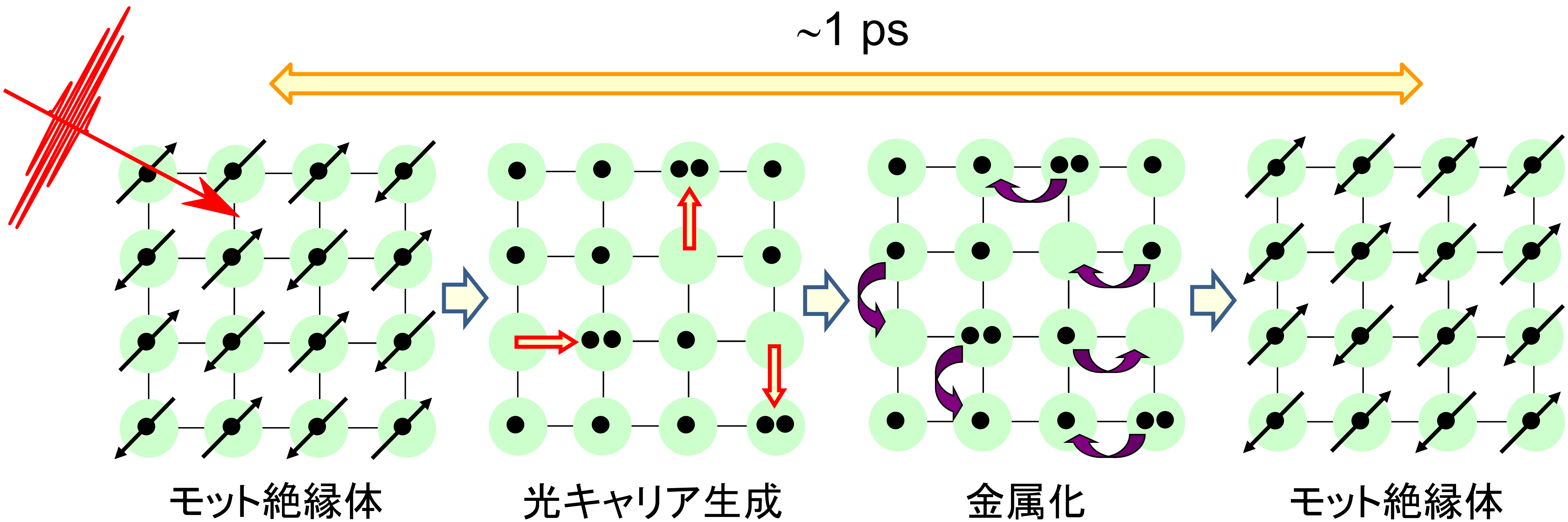
強相関電子系の特徴:　金属とモット絶縁体



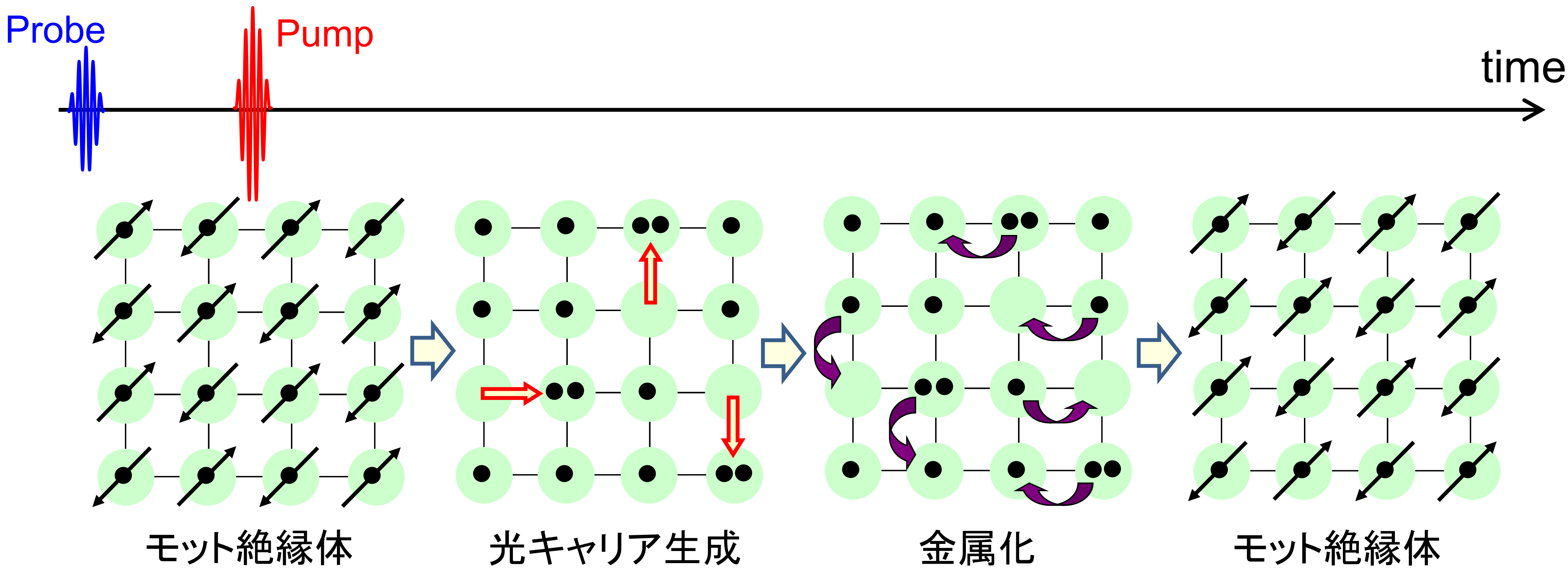
絶縁体と金属は
わずかな刺激(弱い外場)で入れ替わる
＝相転移を起こす

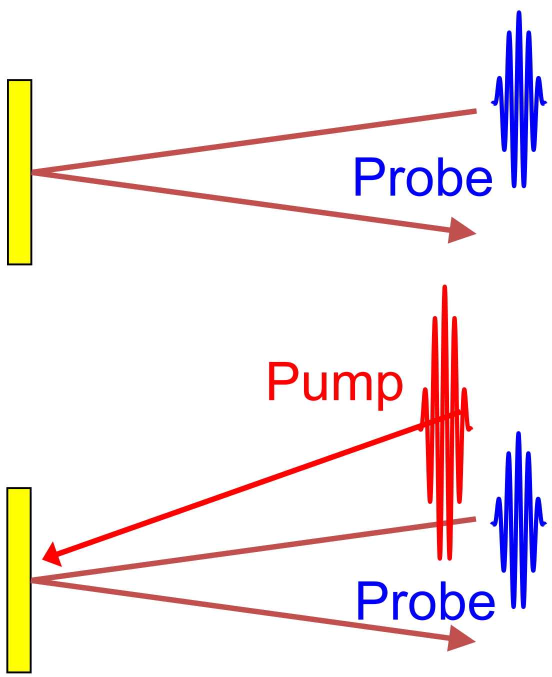
光誘起モット絶縁体→金属転移

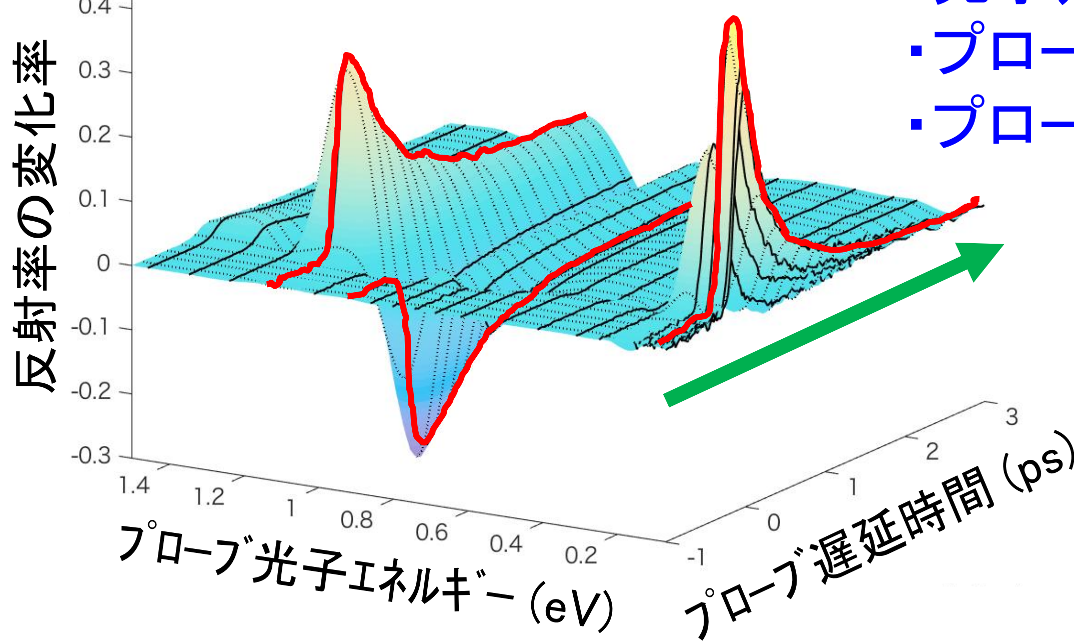
モット絶縁体の超高速光誘起金属化



フェムト秒ポンプープローブ分光



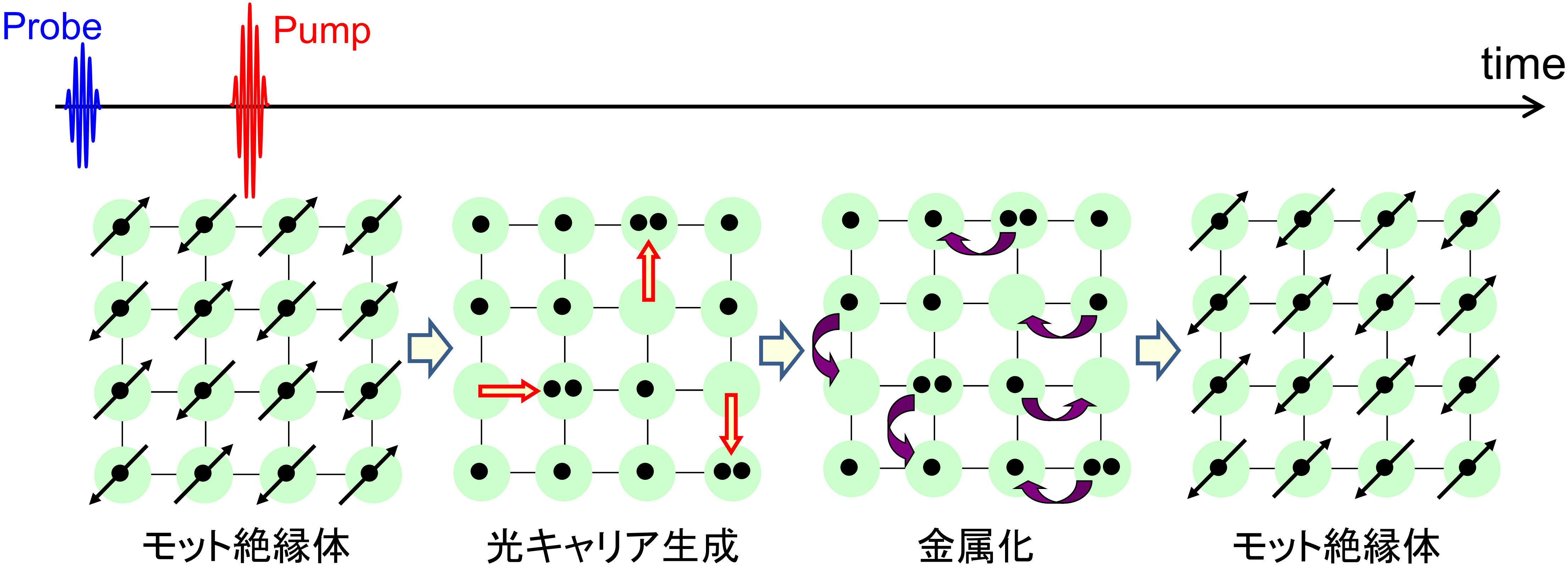


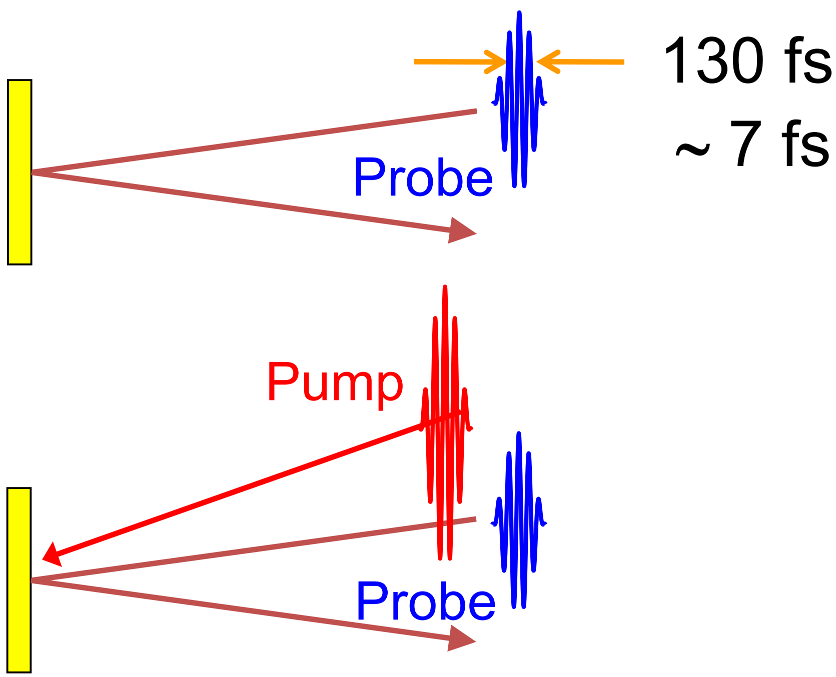


- 光学定数変化
- プローブ光子エネルギー
- プローブ遅延時間

プローブエネルギー毎に時間軸にそって光学定数の変化を検出し、過渡スペクトルに再構成

フェムト秒ポンプープローブ分光





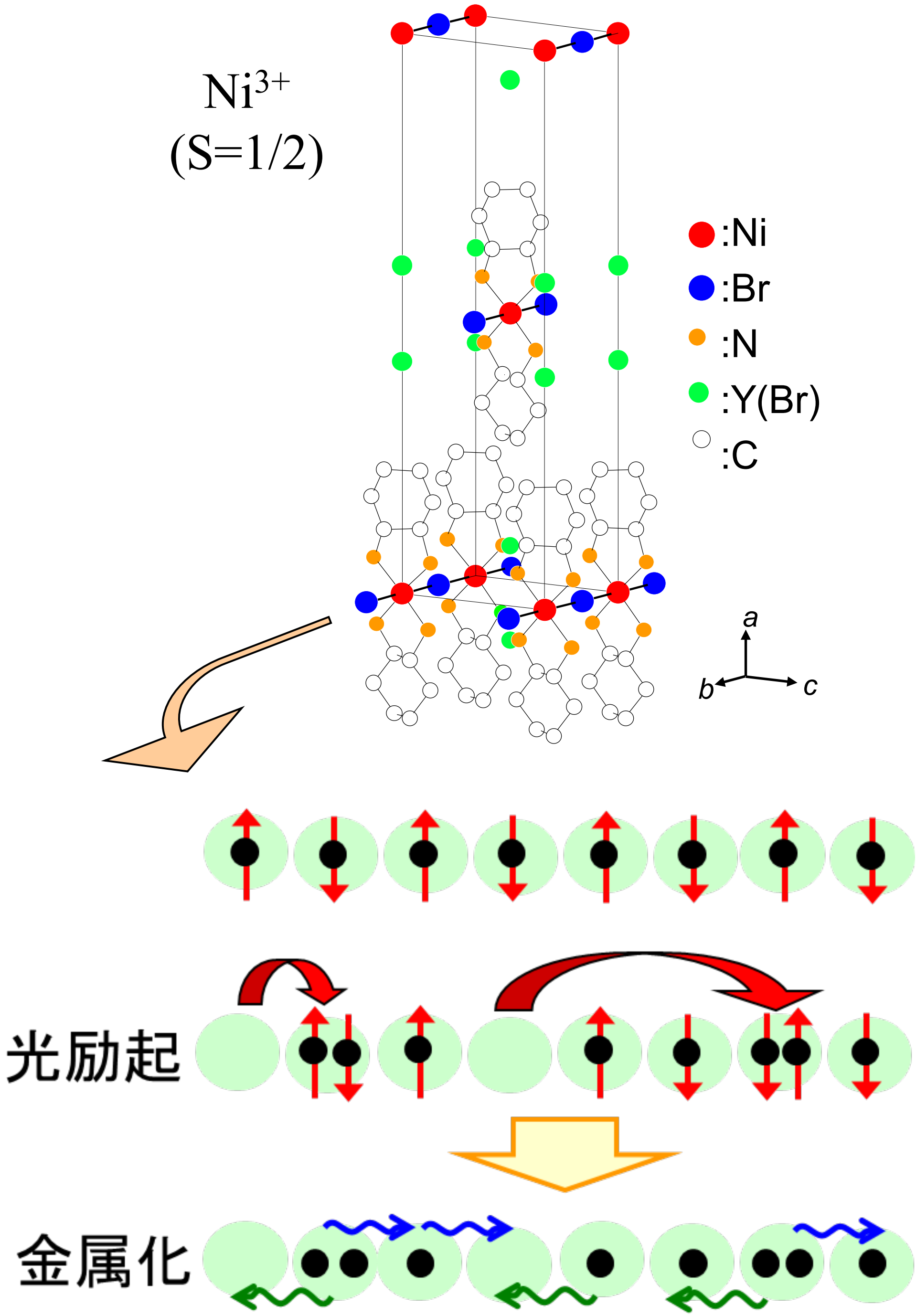
パルス幅：130 fs
時間分解能 : ~180 fs
スペクトル領域 : 4 eV - 0.06 eV

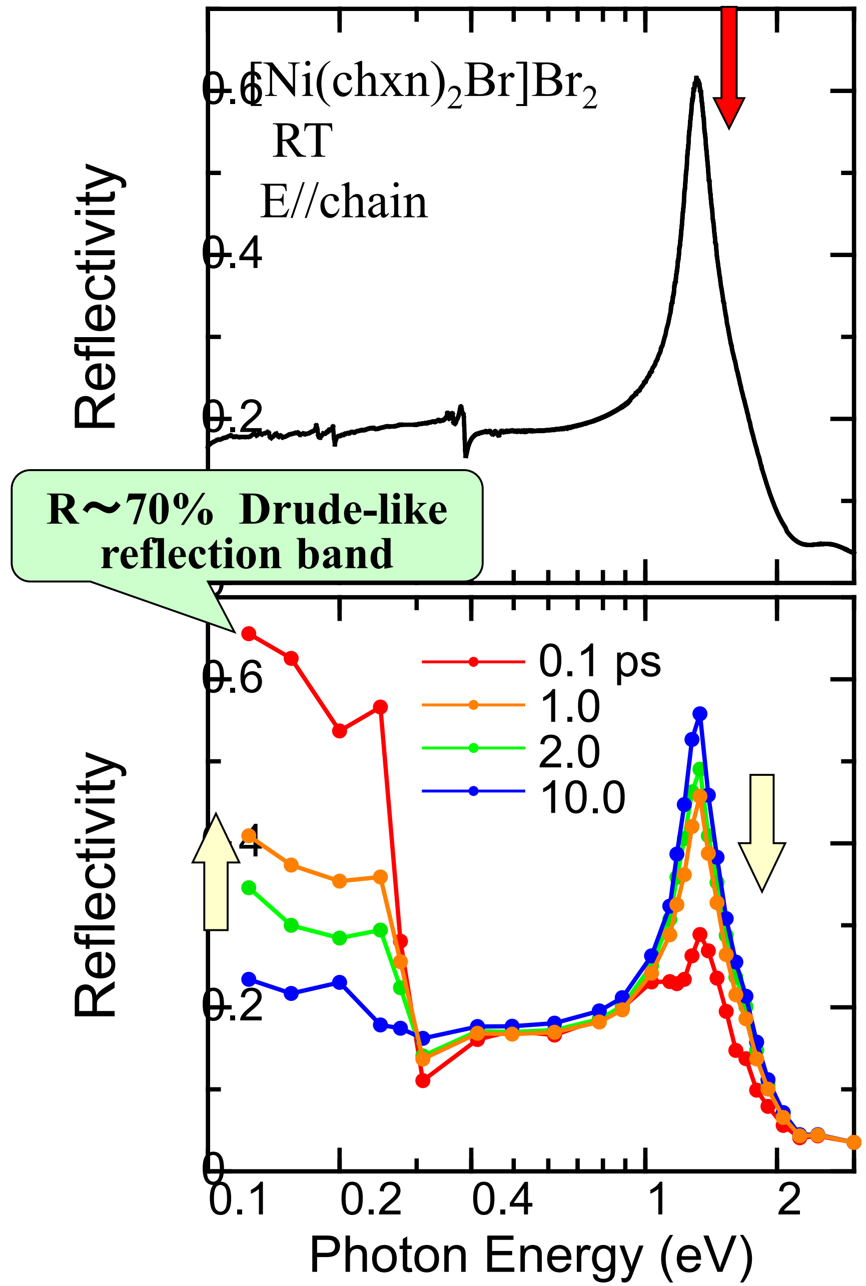
パルス幅：7 fs
時間分解能 : ~10 fs
スペクトル領域：2.2 eV – 1.7 eV

電子（スピン）ダイナミクスの実時間観測

一次元 Ni 錯体の光誘起モット絶縁体ー金属転移（180 fs time res.）

$[Ni(chxn)_2Br]Br_2$

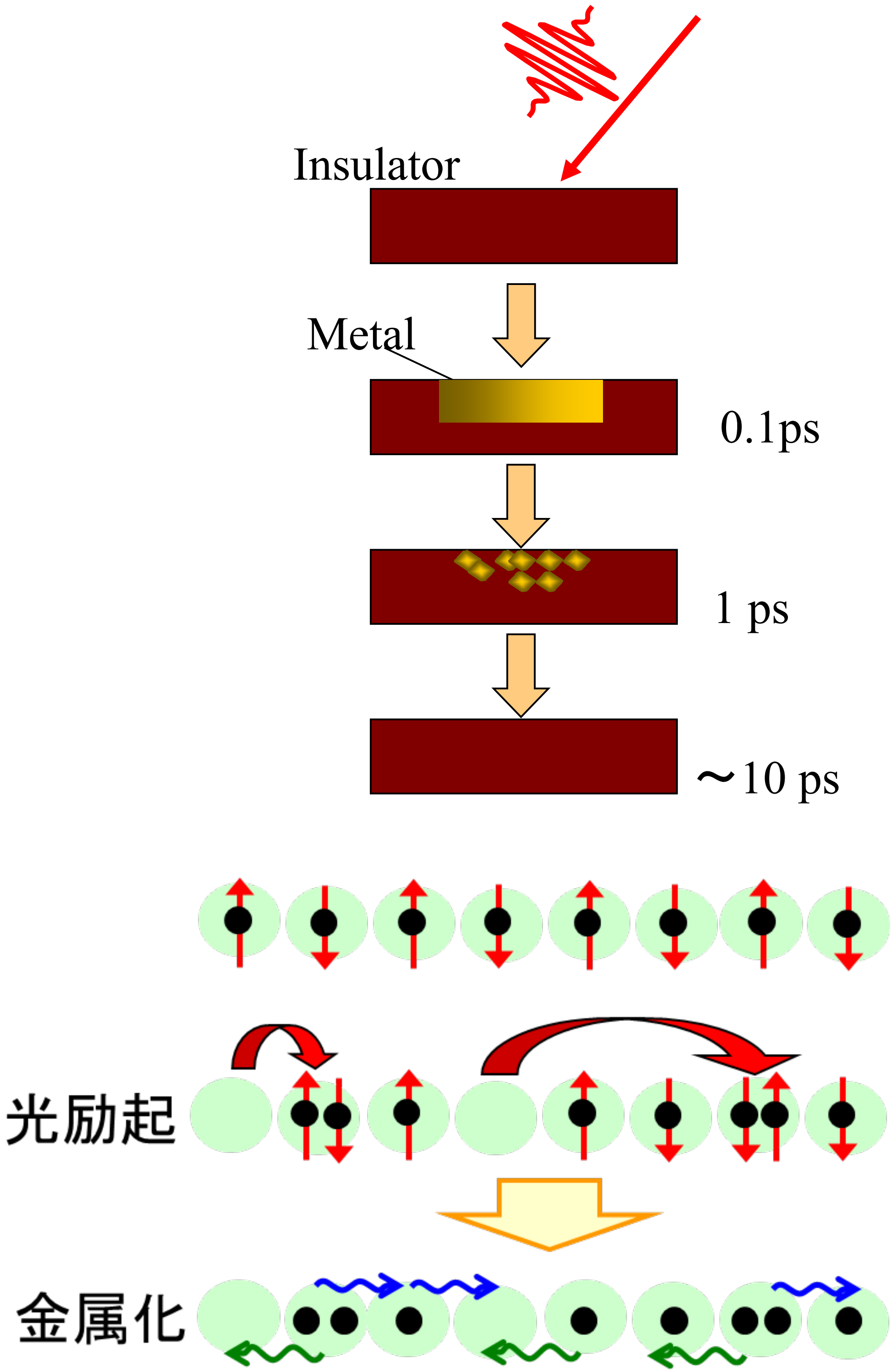


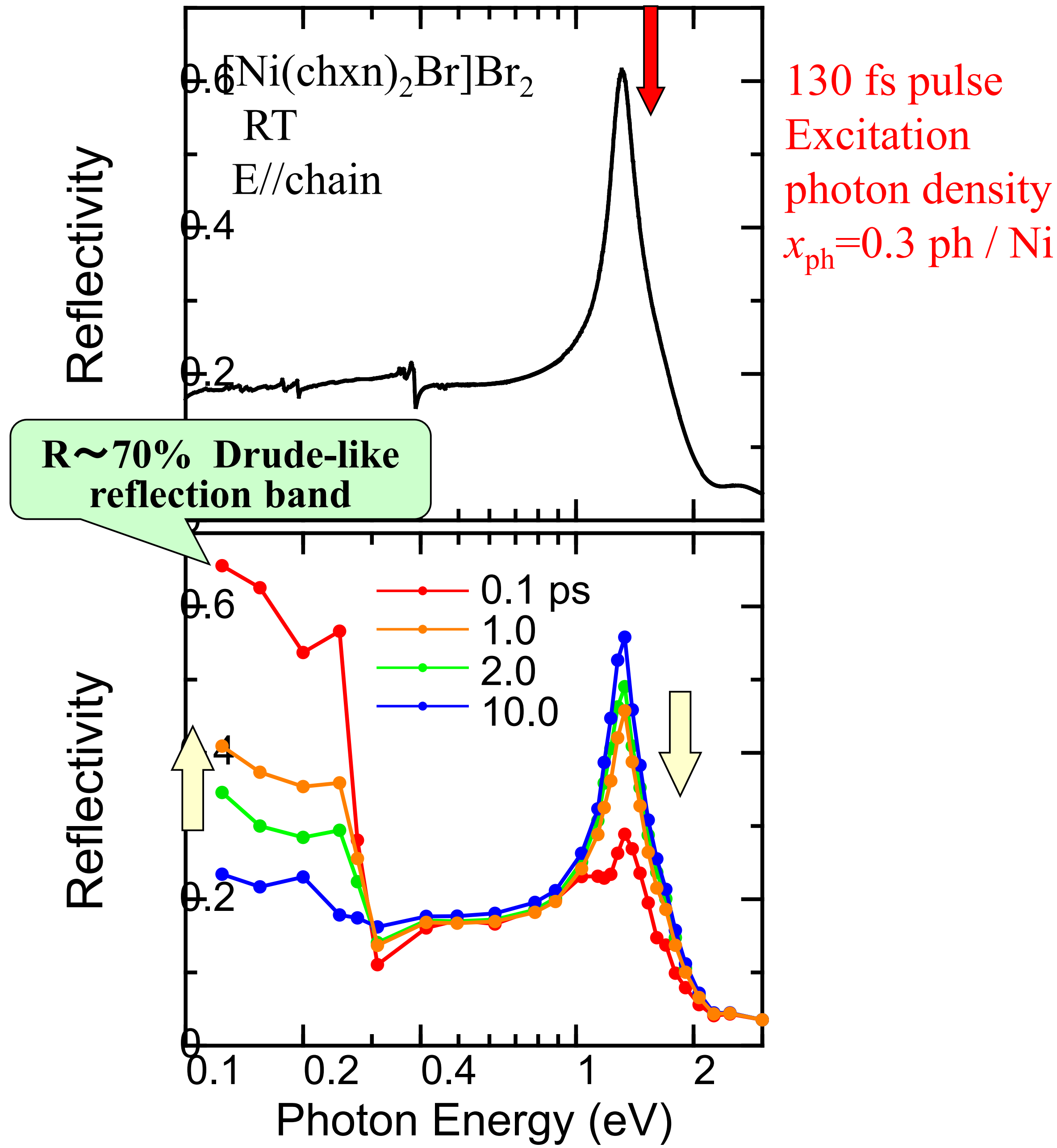


130 fs pulse
Excitation
photon density
x_{ph}=0.3 ph / Ni

PRL (2003), PRL (2007), NP (2011), PRL (2014)

一次元 Ni 錯体の光誘起モット絶縁体ー金属転移(180 fs time res.)



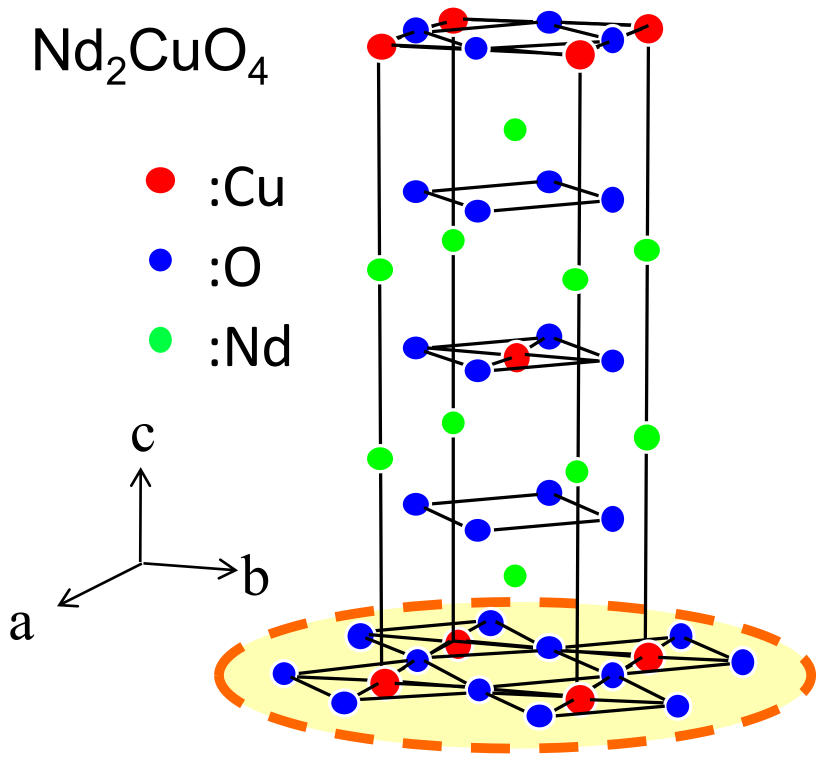


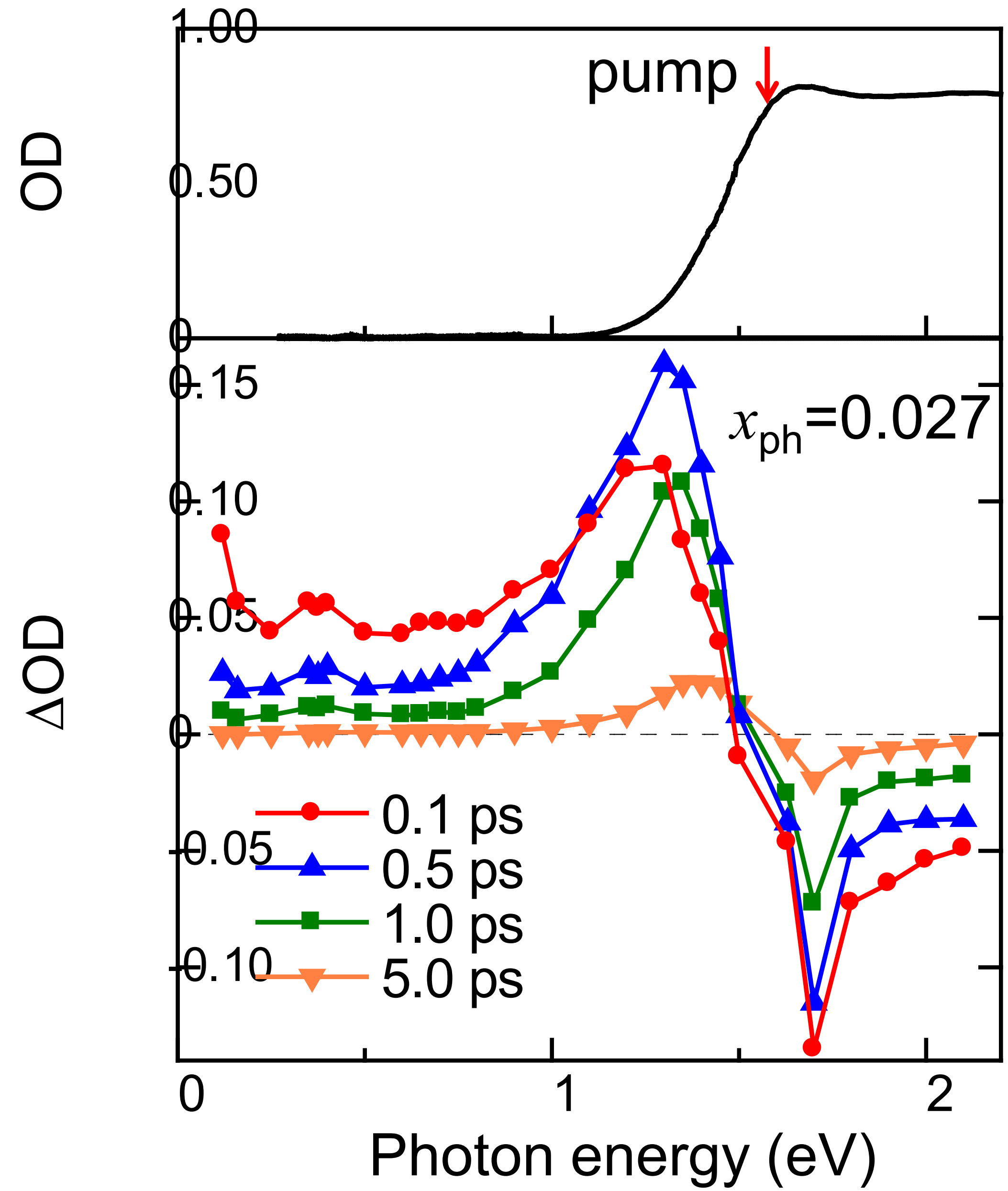
PRL (2003), PRL (2007), NP (2011), PRL (2014)

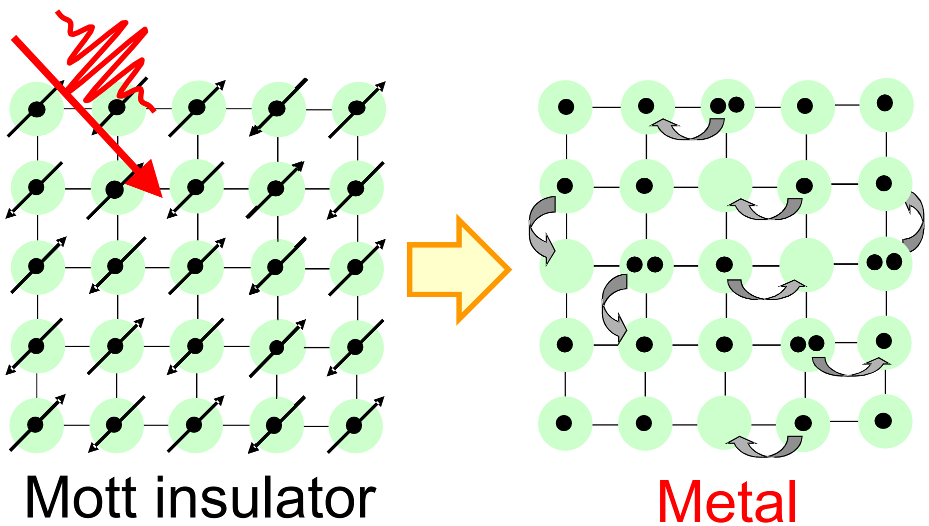
Photoinduced insulator-metal transition in Nd_2CuO_4 (180 fs res.)

*H. Okamoto et al., PRB **82**, 060513R (2010), PRB **83**, 125102 (2011).*

2D Mott insulator



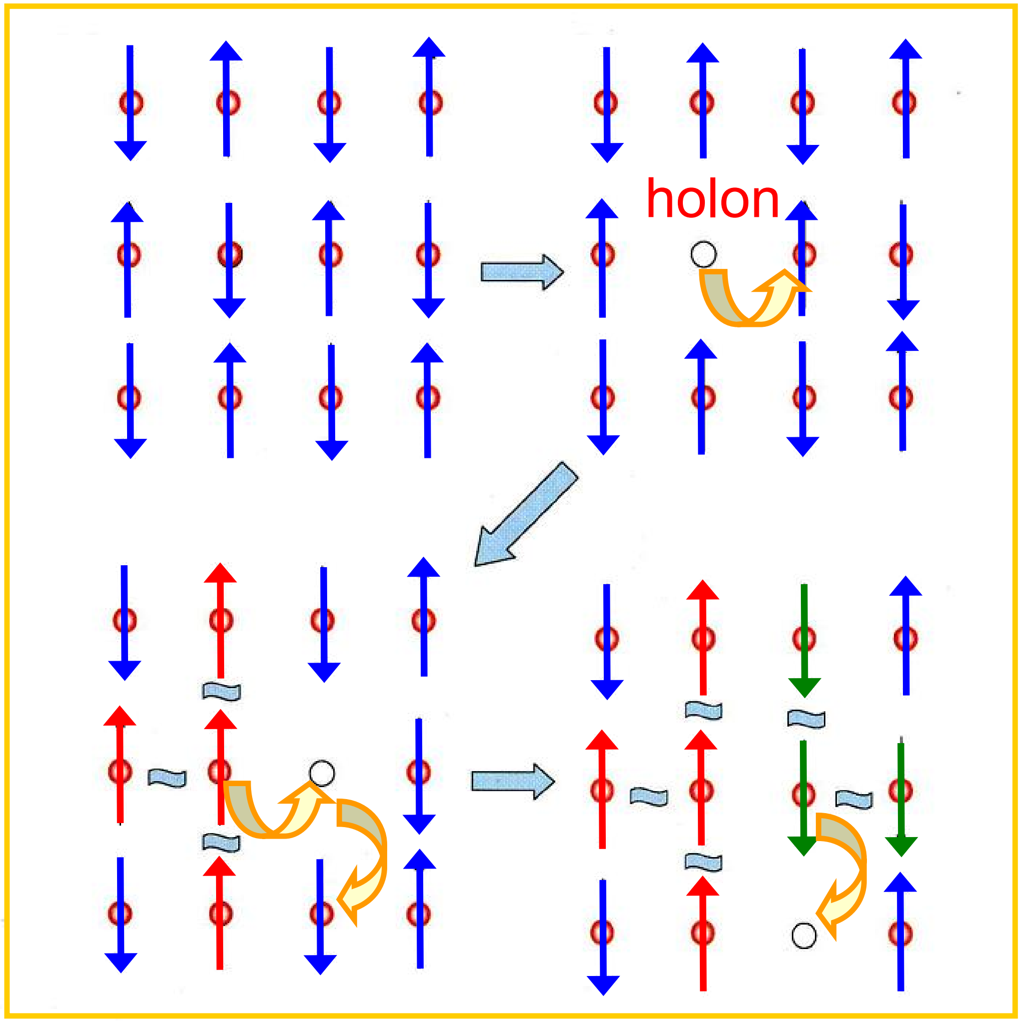




Half-filledモット絶縁体（large U）における電荷ダイナミクス

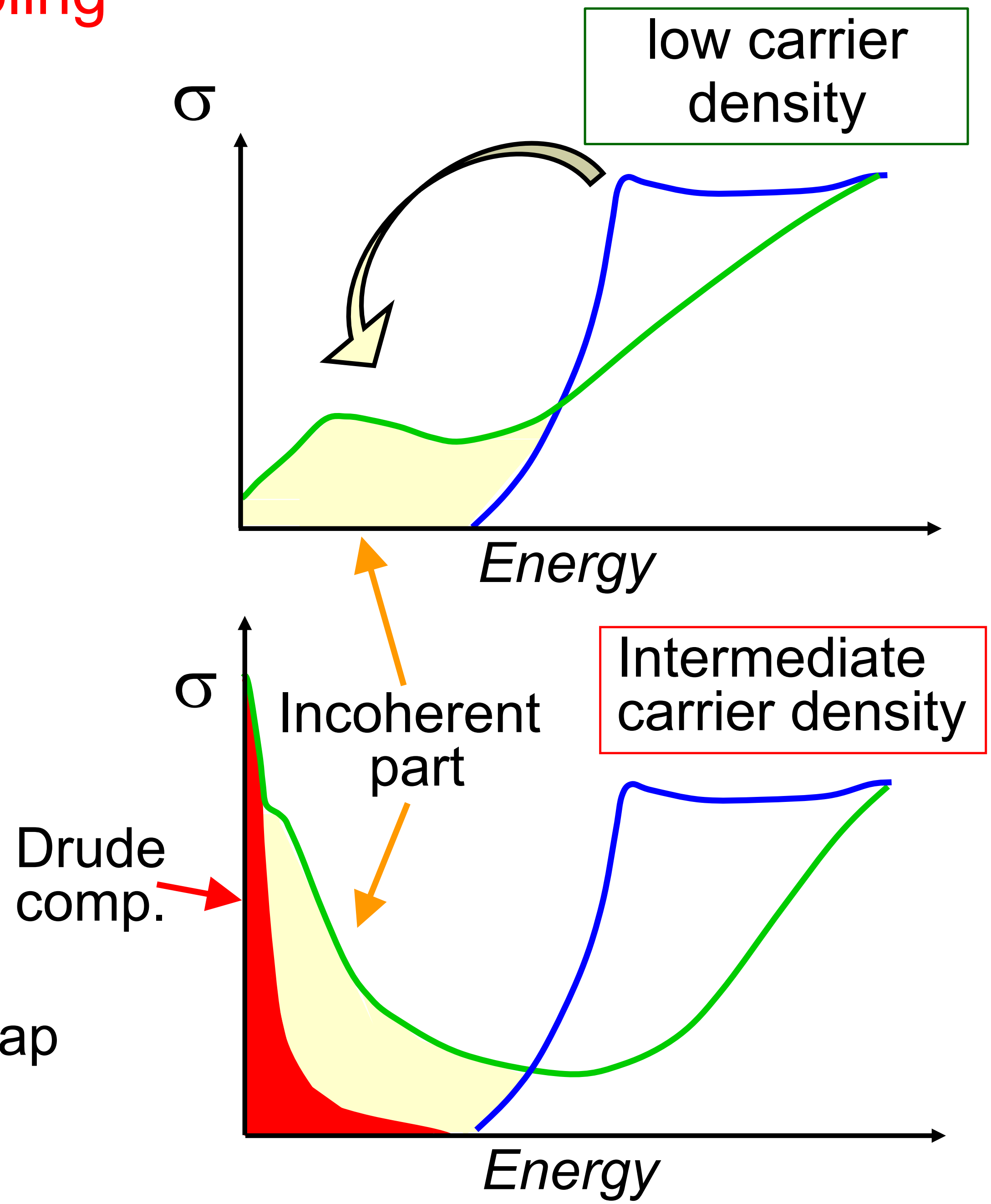
2D Mott insulator

spin-charge coupling



Introduction of carriers produces midgap absorption (incoherent part).

Drude weight is small as compared to the incoherent part.

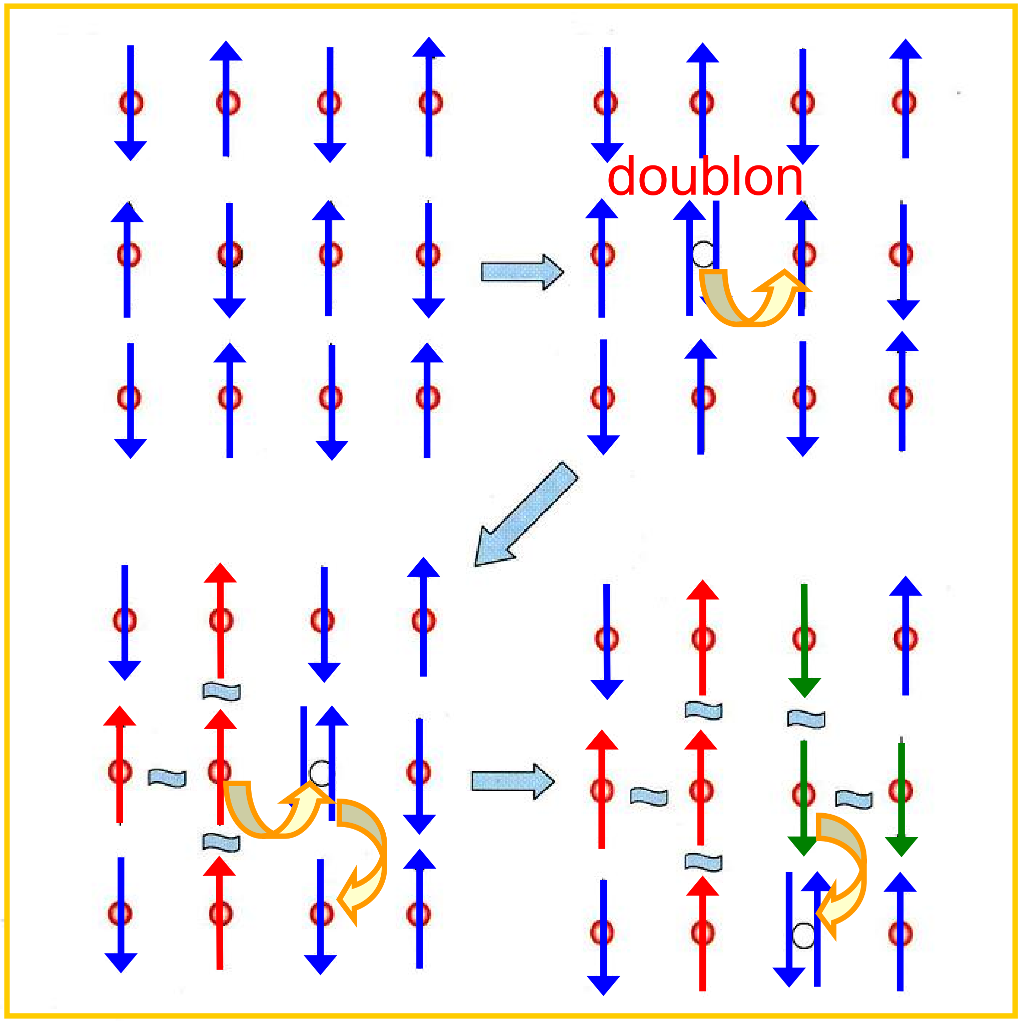


ex. LSCO, NCCO

Half-filledモット絶縁体（large U）における電荷ダイナミクス

2D Mott insulator

spin-charge coupling



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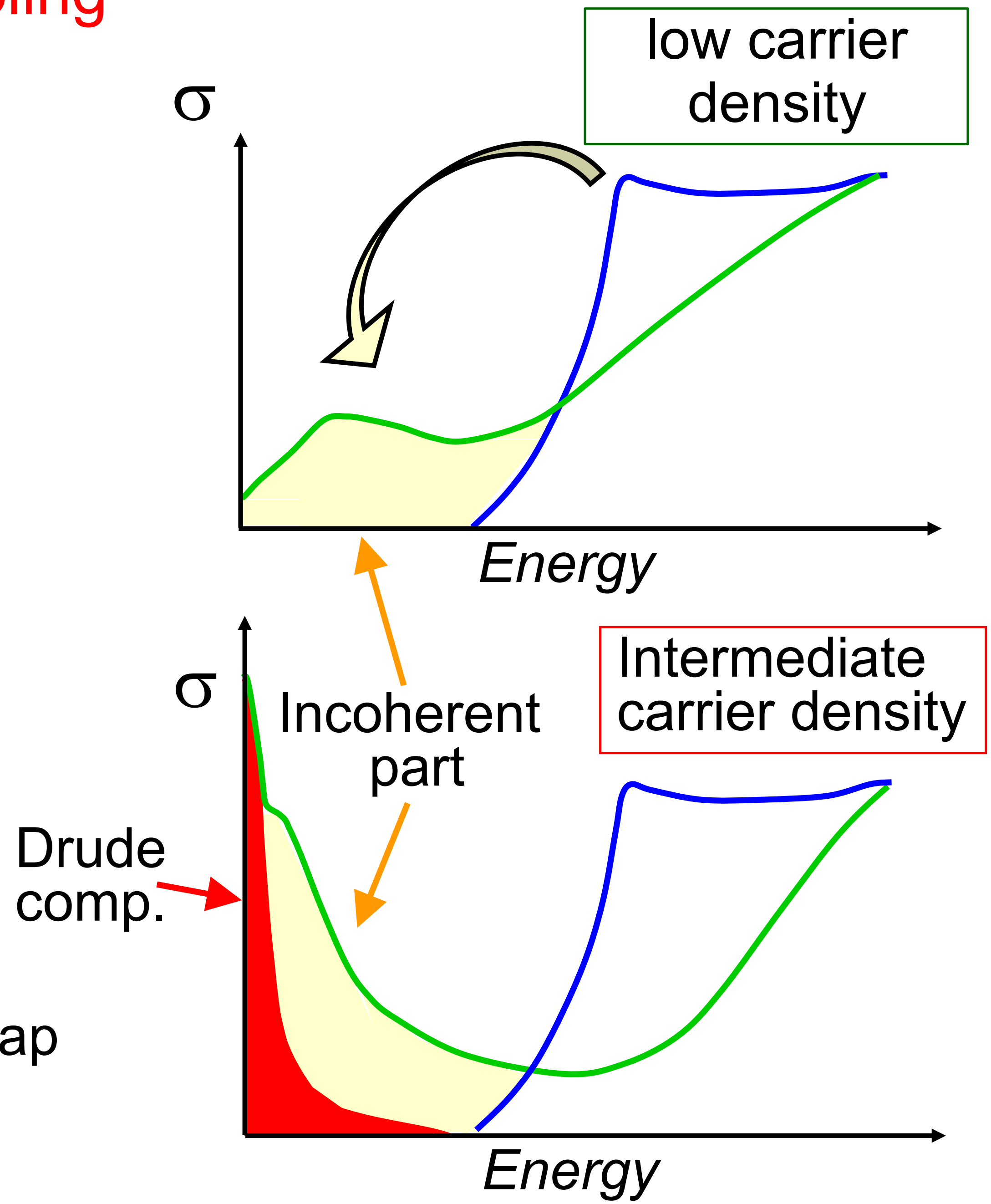
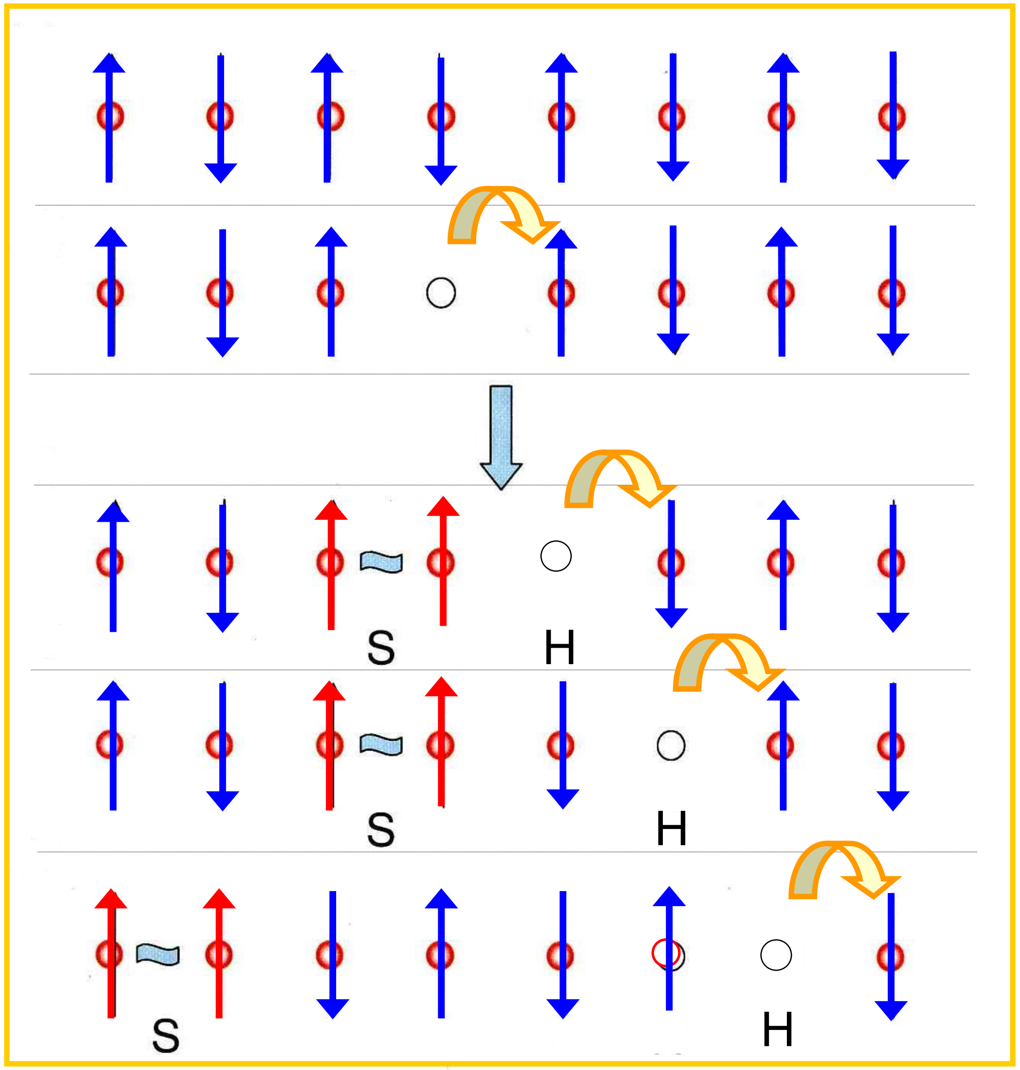
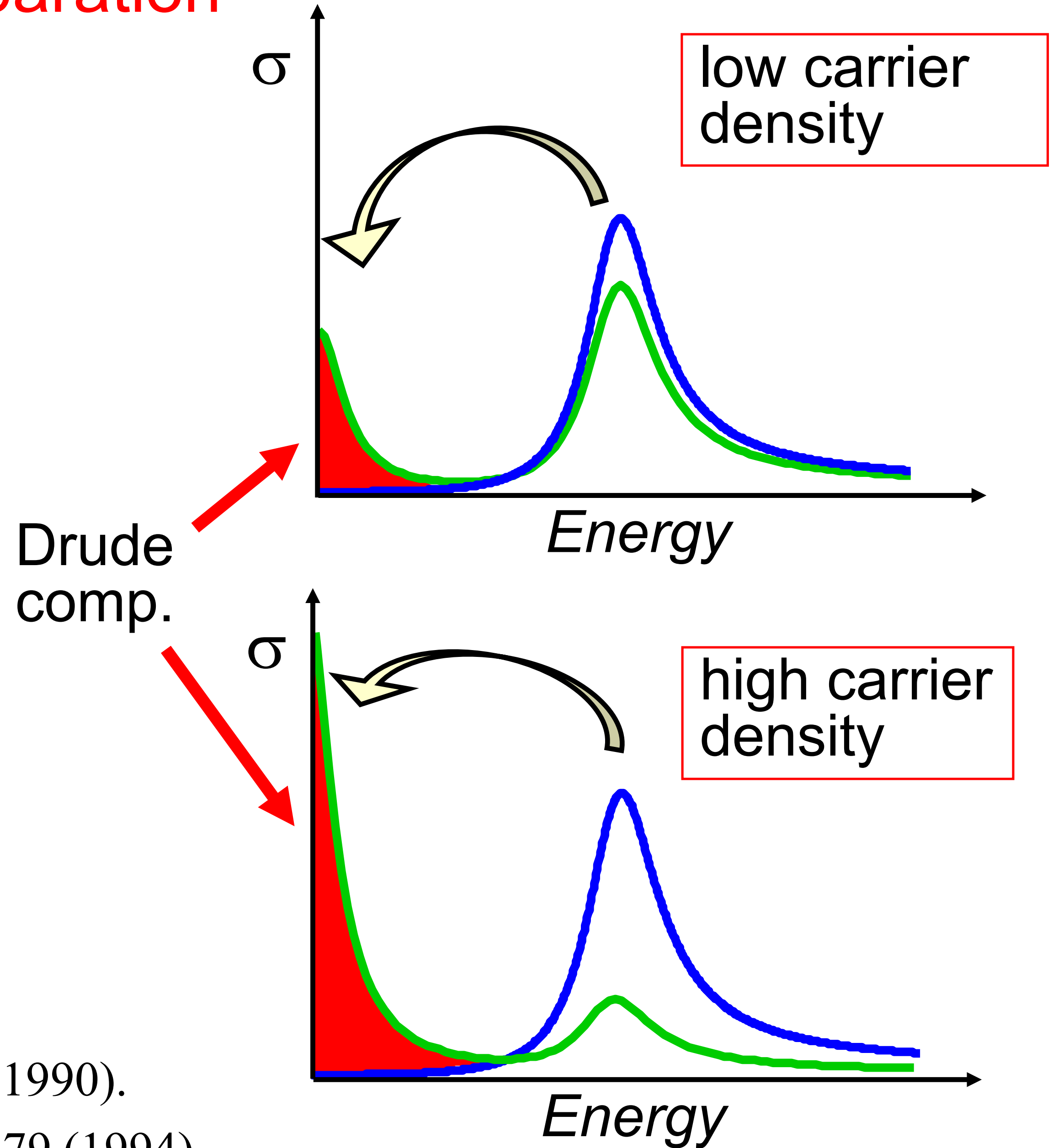


ex. LSCO, NCCO

Half-filledモット絶縁体（large U）における電荷ダイナミクス

1D Mott insulator spin-charge separation





M. Ogata and H. Shiba, Phys. Rev. B **41**, 2326 (1990).

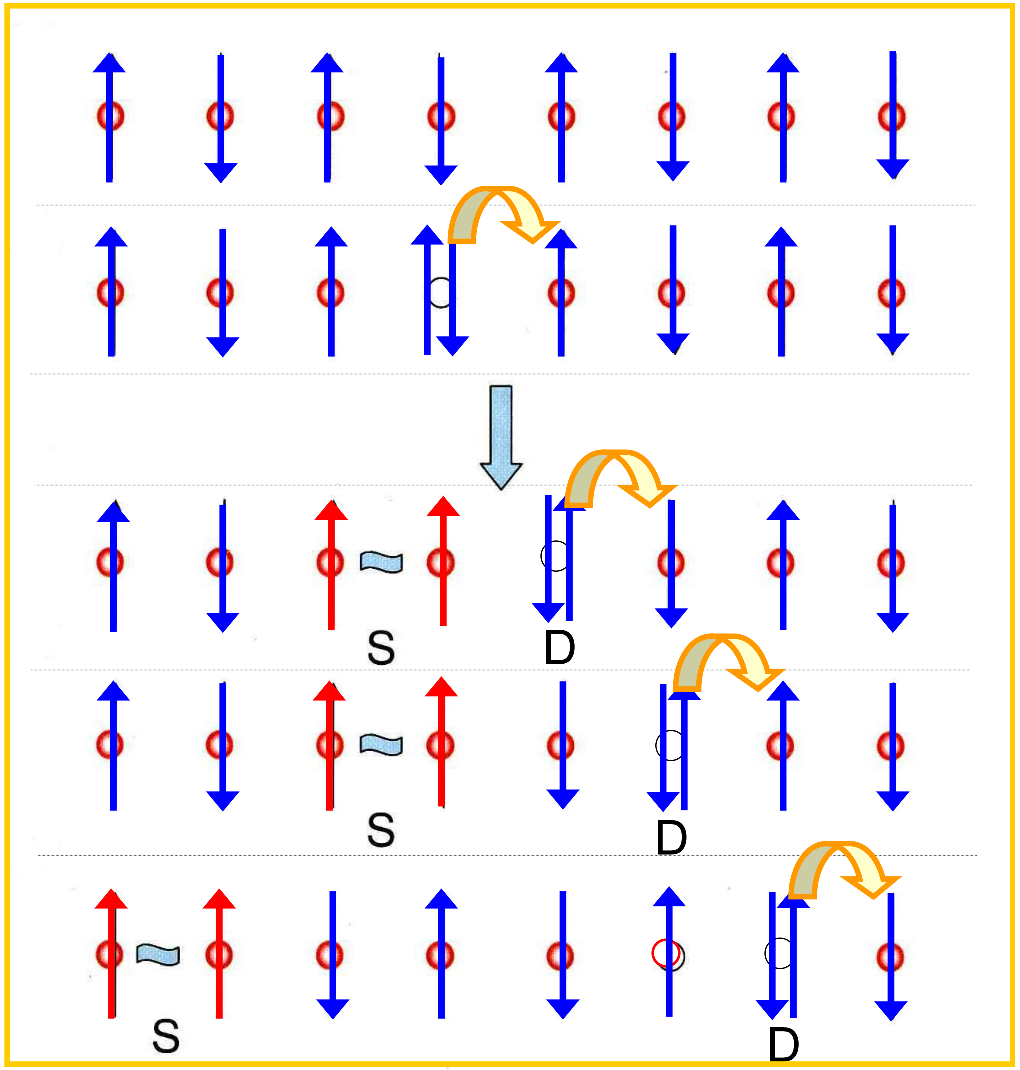
H. Eskes and A.M. Oles, Phys. Rev. Lett. **77**, 1279 (1994).

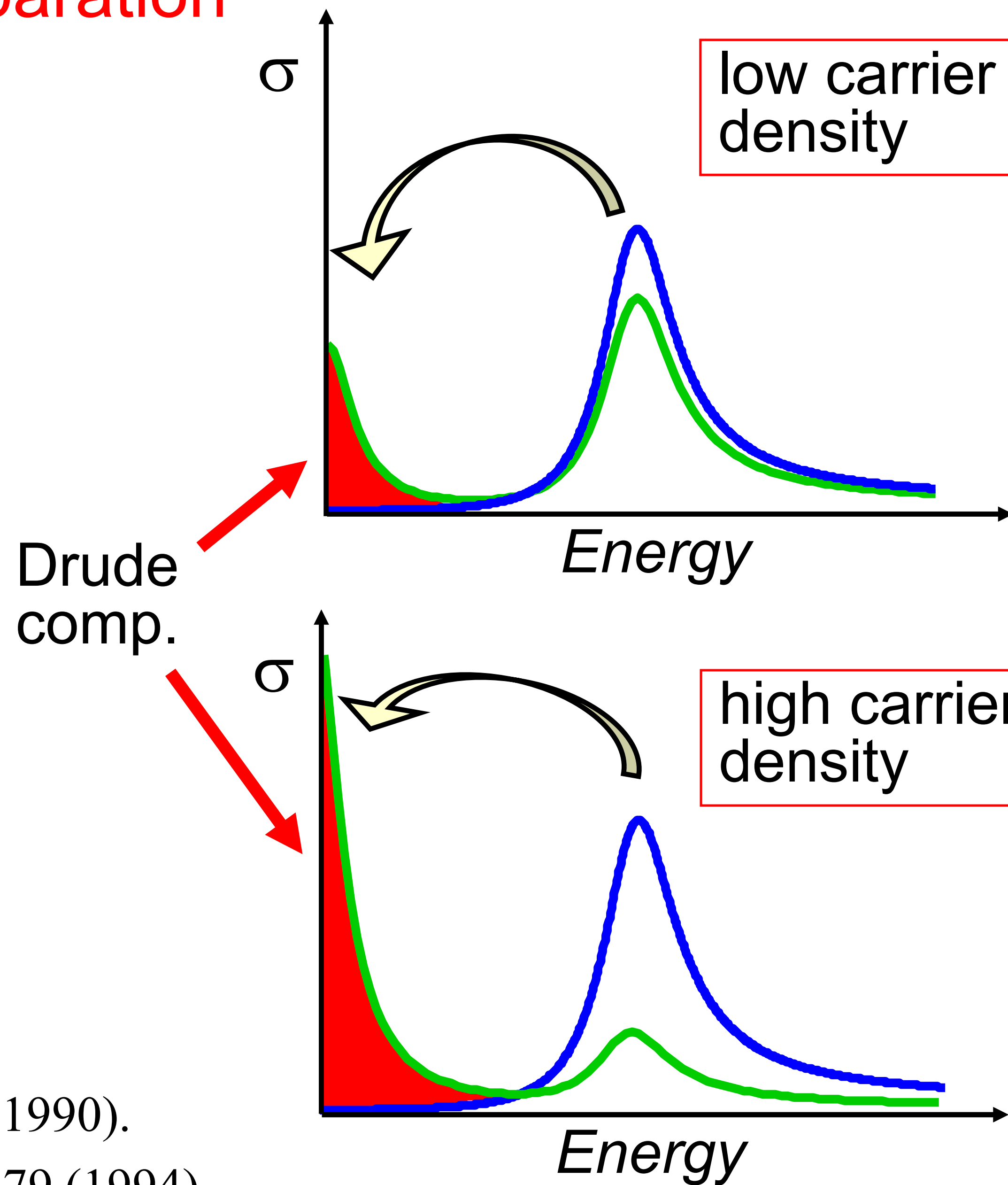
By carrier doping, spectral weight of the gap transition is transferred to the Drude component irrespective of carrier density.

ex. [Ni(chxn)$_2$Br]Br$_2$

Half-filledモット絶縁体（large U）における電荷ダイナミクス

1D Mott insulator spin-charge separation





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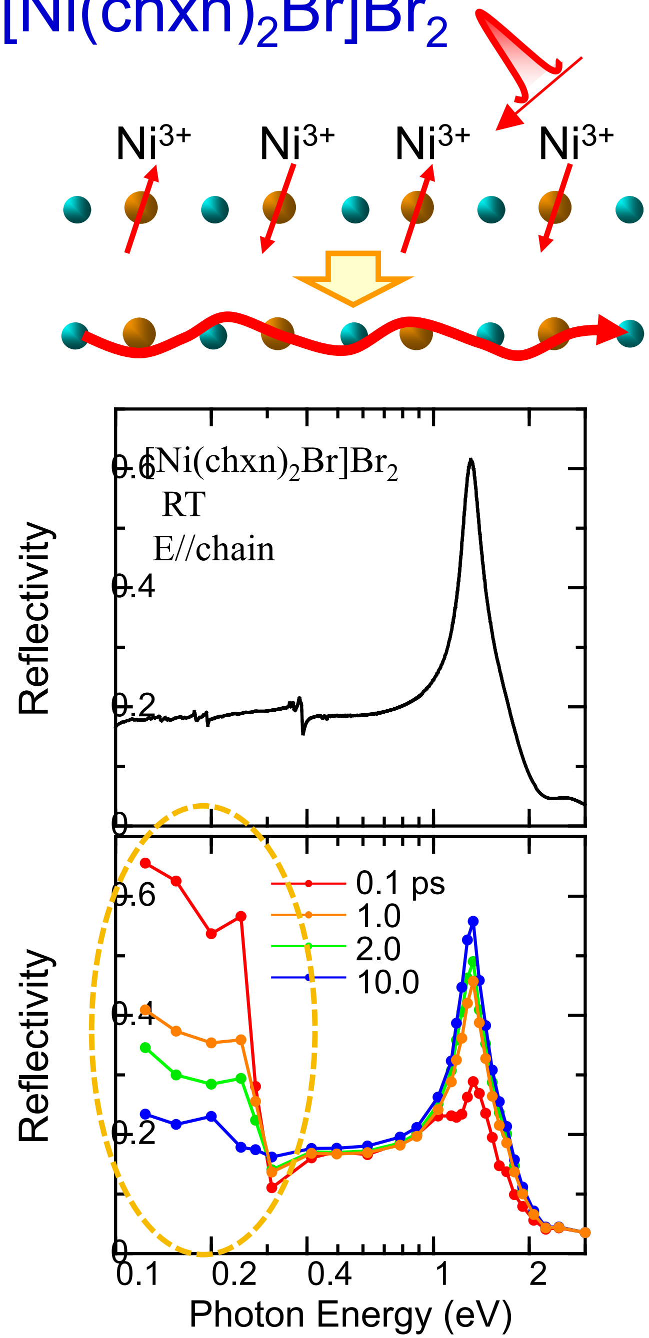
By carrier doping, spectral weight of the gap transition is transferred to the Drude component irrespective of carrier density.

ex. $[Ni(chxn)_2Br]Br_2$

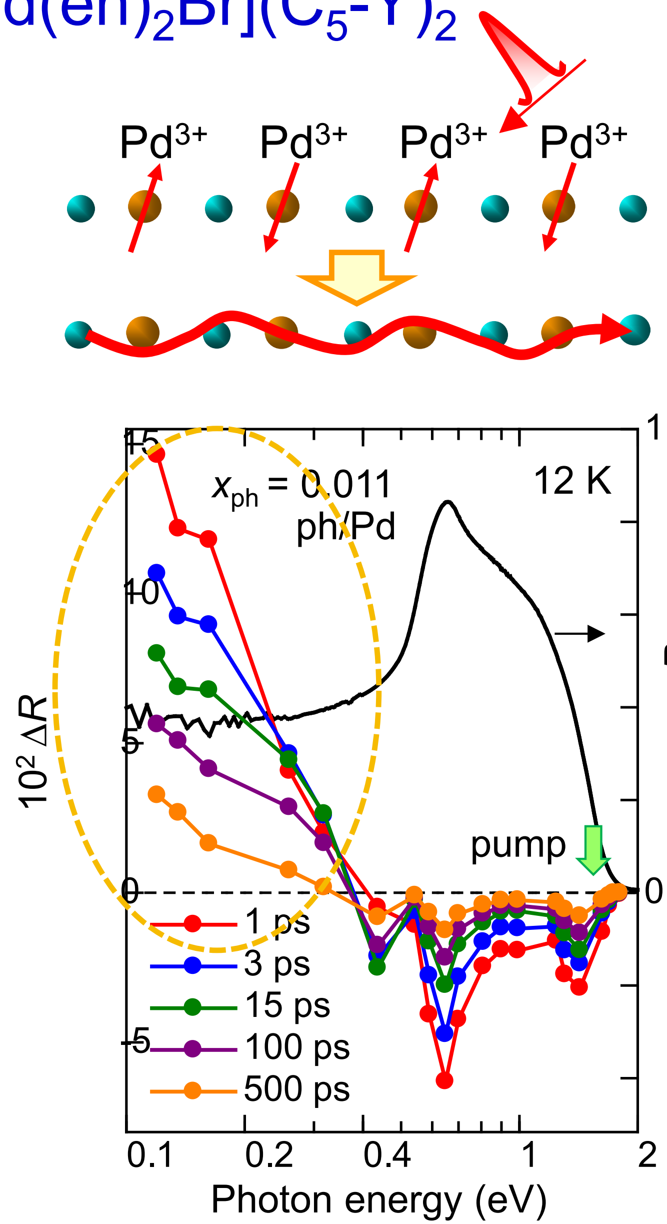
Photoinduced Mott insulator to metal transitions in 1D systems

$[Ni(chxn)_2Br]Br_2$



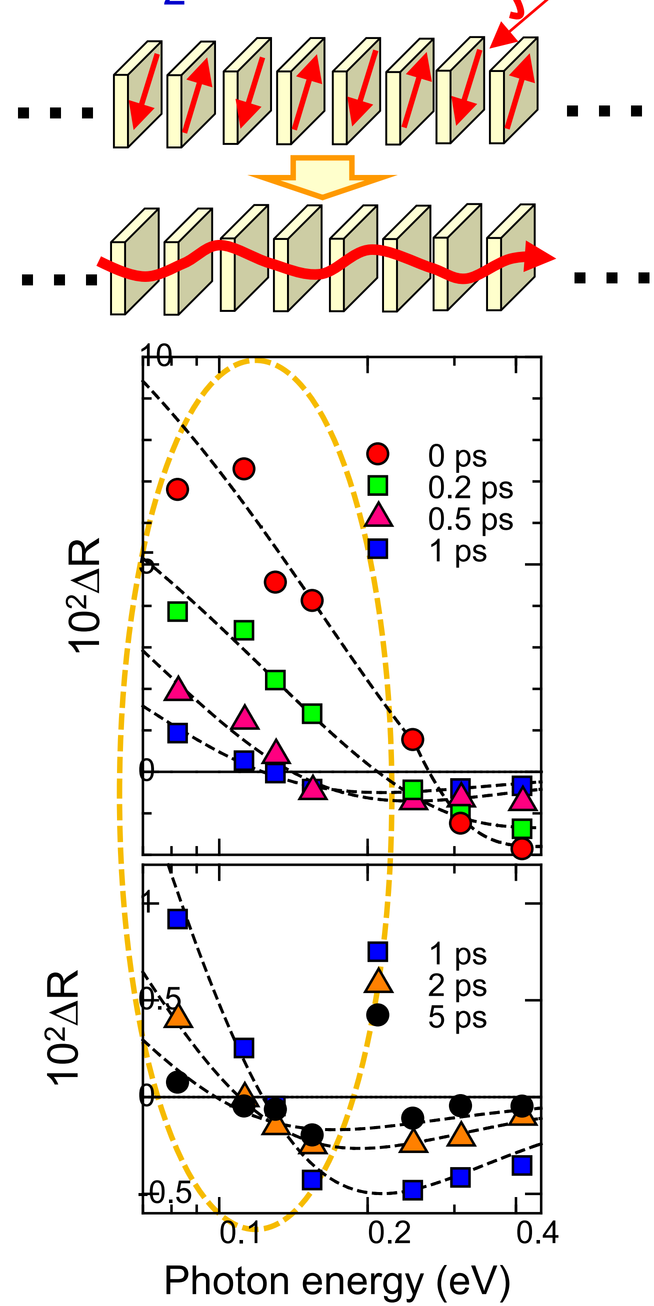
S. Iwai, et al.,
PRL ***91****, 57401 (2003).*

$[Pd(en)_2Br](C_5\text{-}Y)_2$



H. Matsuzaki et al.,
PRL ***113****, 096403 (2014).*

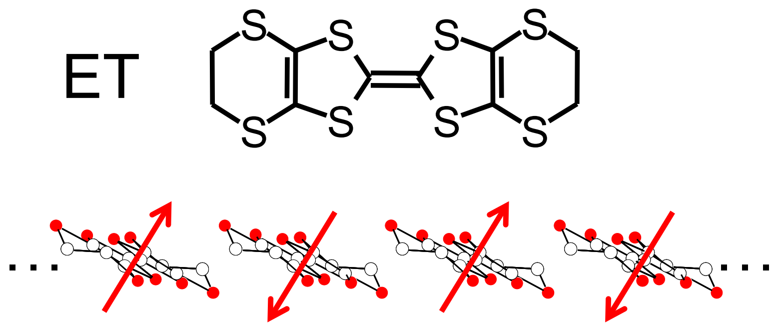
$ET\text{-}F_2TCNQ$



H. Okamoto et al.,
PRL ***98****, 037401 (2007).*

Photoinduced Mott-insulator to metal transition in a typical 1D system

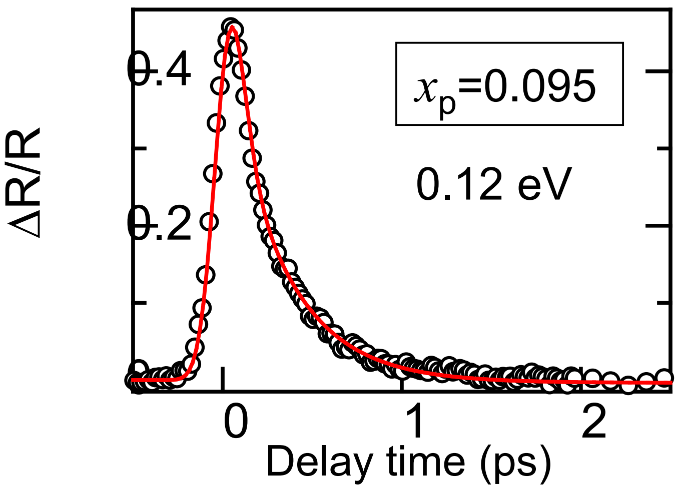
$ET\text{-}F_2TCNQ$



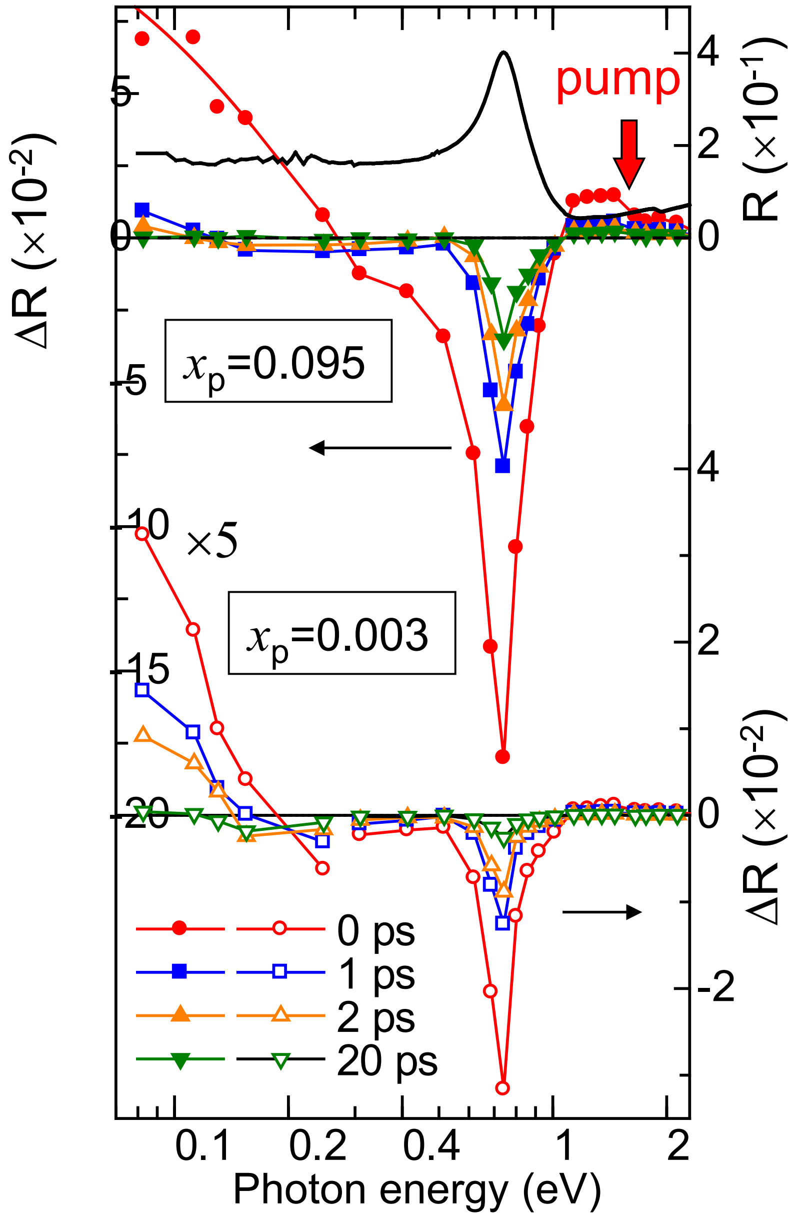
Half-filled 1D Mott insulator

Ultrafast responses

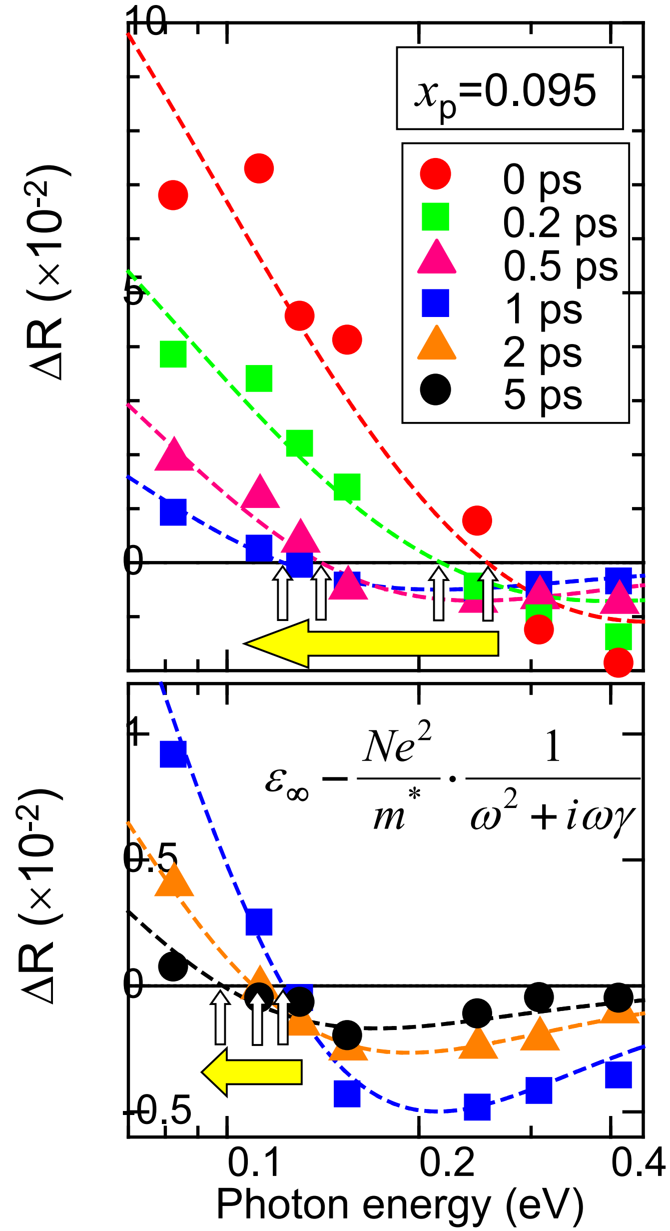
Relaxation time ~200 fs



Appearance of metallic reflection by light



Drude responses characteristic of 1D CES

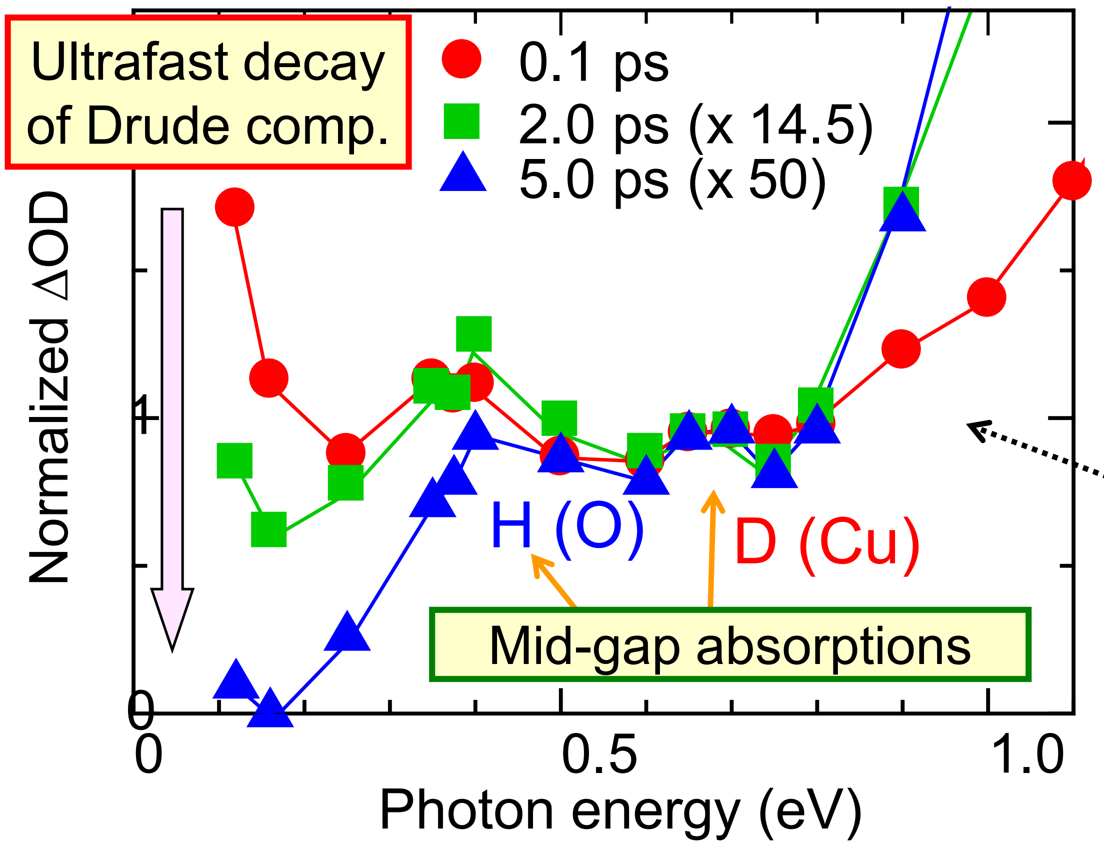


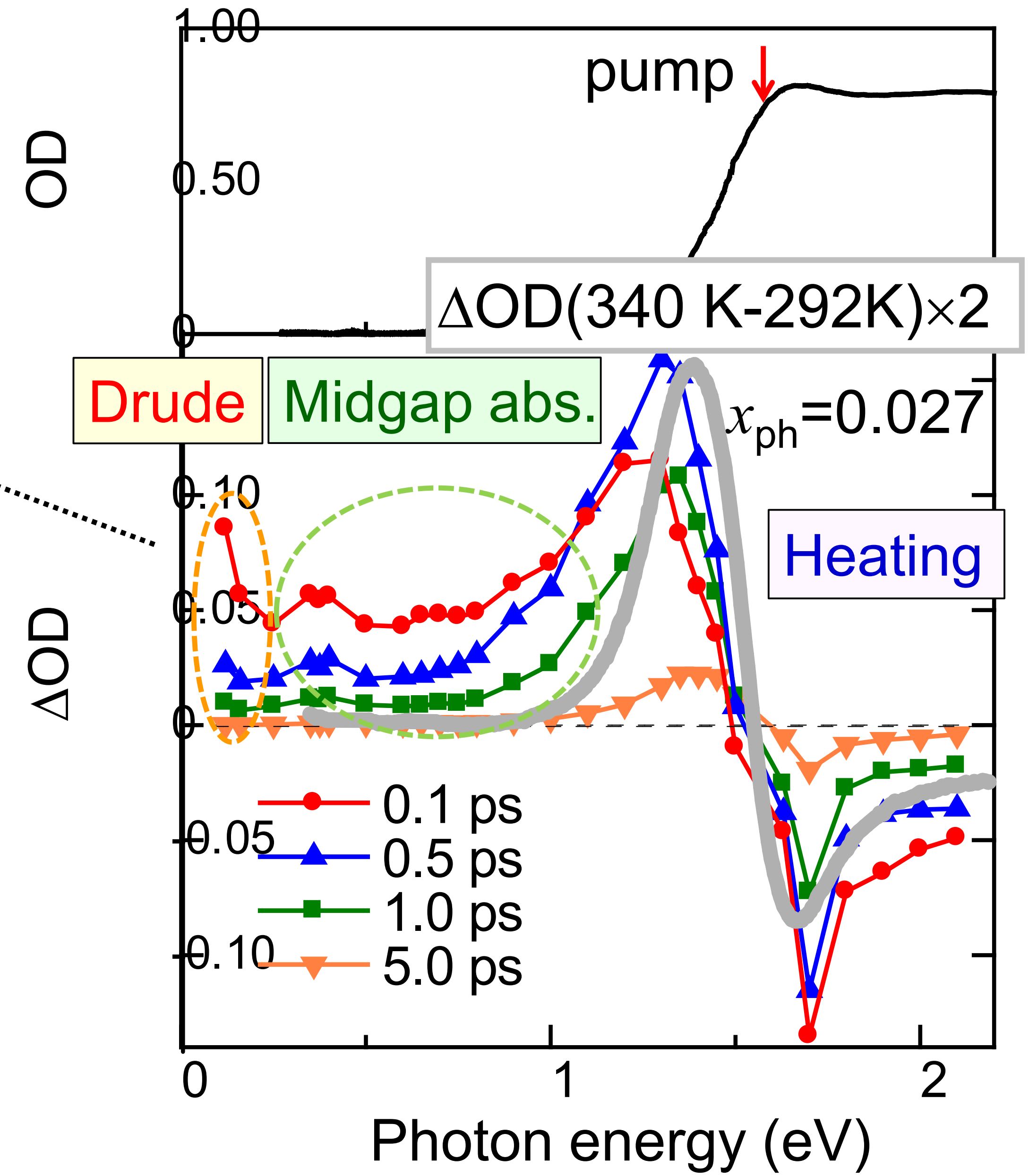
Drude responses independent of excitation photon density

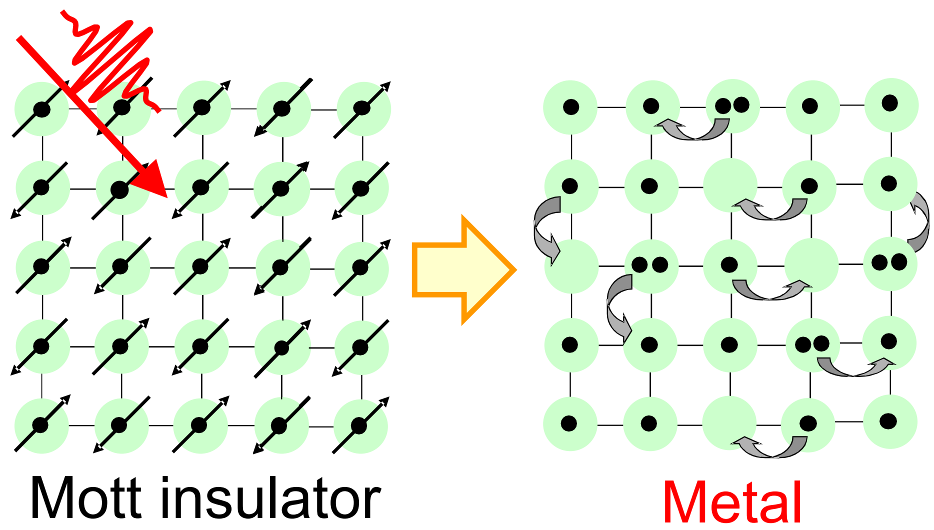
H. Okamoto *et al.*, PRL **98**, 037401 (2007), H. Uemura *et al.*, JPSJ **77**, 113714 (2008)

Photoinduced insulator-metal transition in Nd_2CuO_4 (180 fs res.)

*H. Okamoto et al., PRB **82**, 060513R (2010), PRB **83**, 125102 (2011).*







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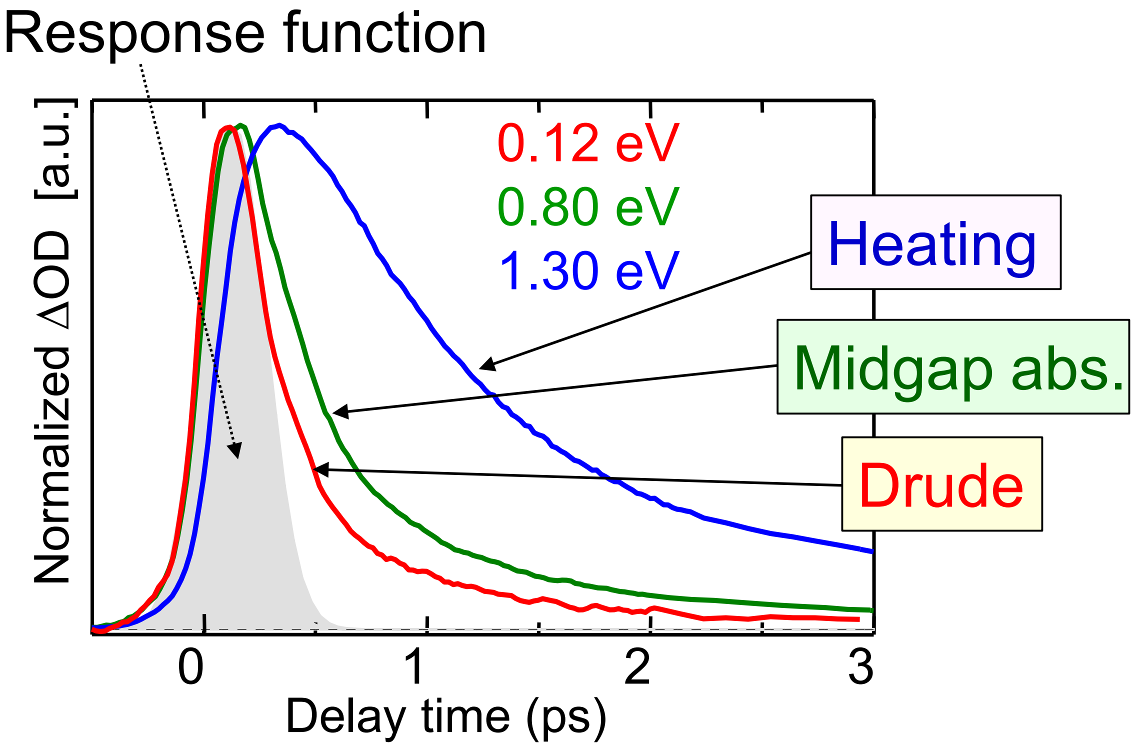
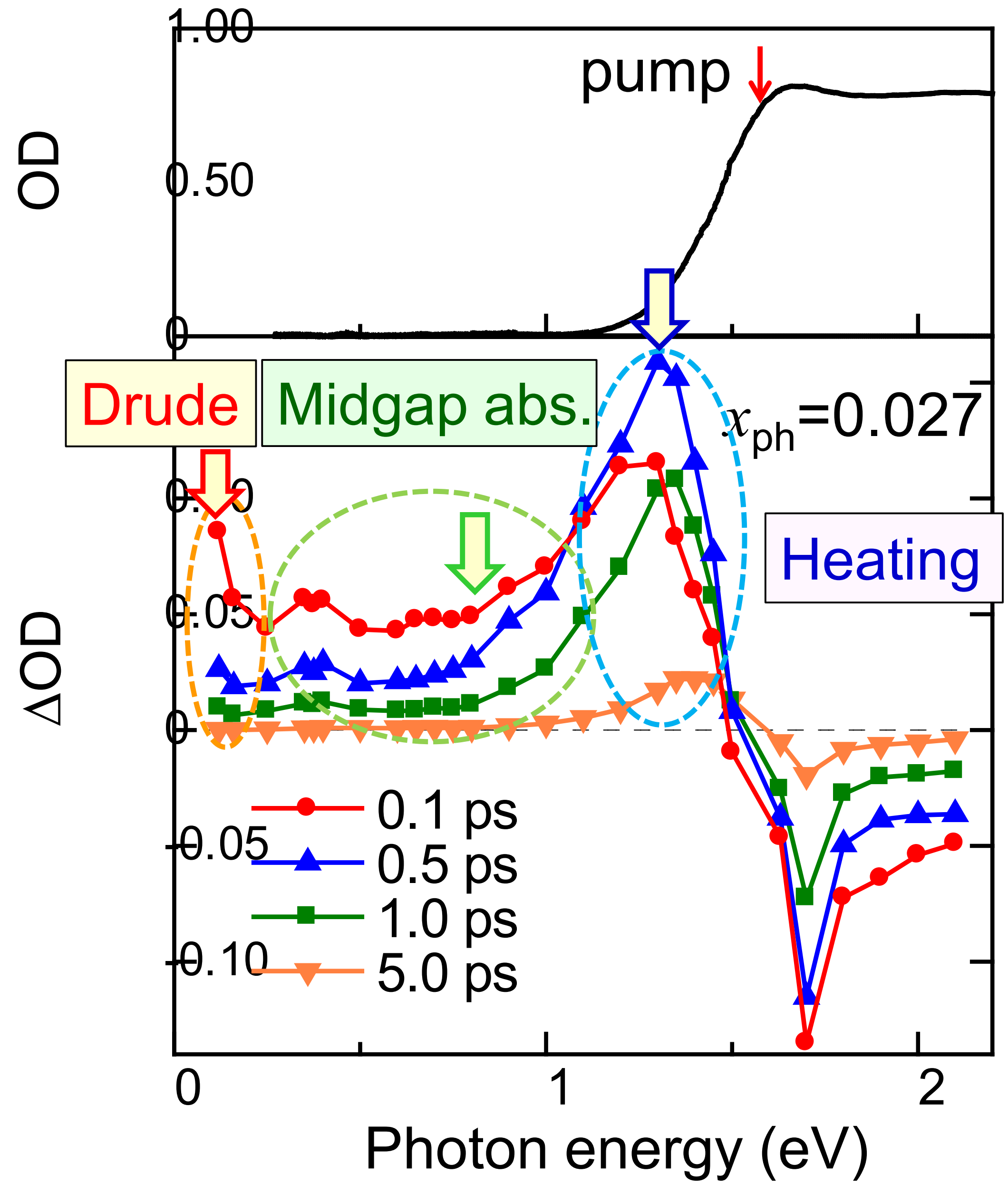
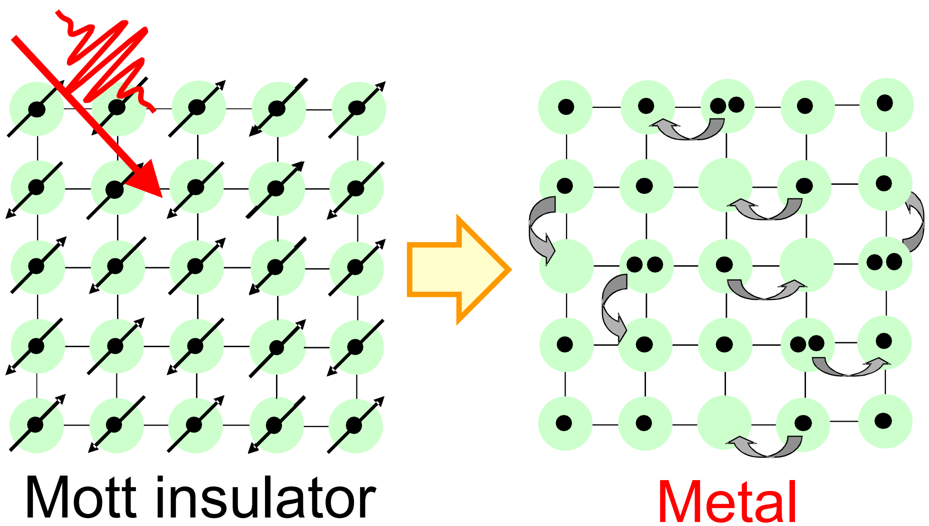
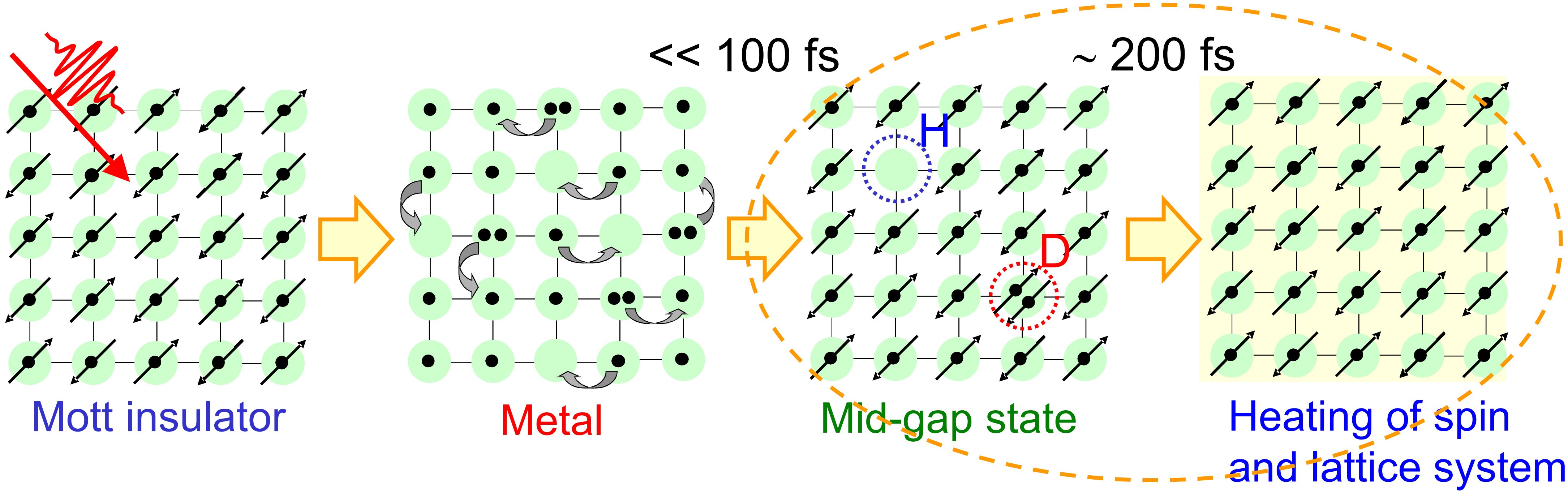






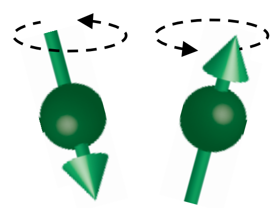
Photoinduced Mott insulator to metal transition in Nd_2CuO_4

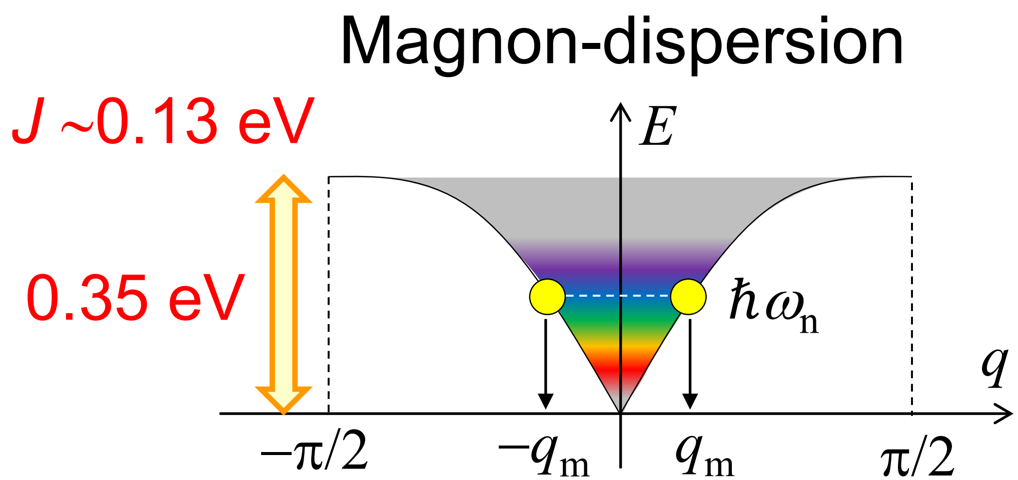


Recombination of a D-H pair ~200 fs ⇔ Rapid energy transfer to spin system?

Theory :

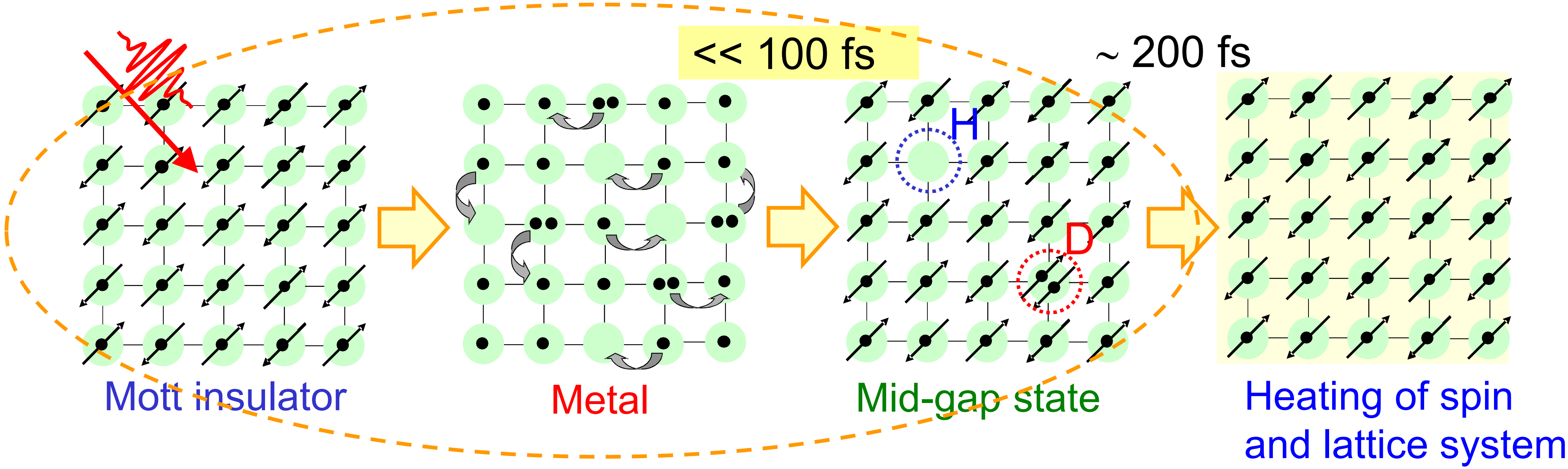
- *L. Vidmar, J. Bonca, T. Tohyama, and S. Maekawa, PRL* ***107****, 246404 (2011)*
- *Z. Lenarcic and P. Prelovsek, PRL* ***111****, 016401 (2013)*
- *E. Iyoda and S. Ishihara, PRB* ***89****, 125126 (2014)*





K. Ishii et al., Nat. Commun. ***5****, 3714 (2014)*

Photoinduced Mott insulator to metal transition in Nd_2CuO_4

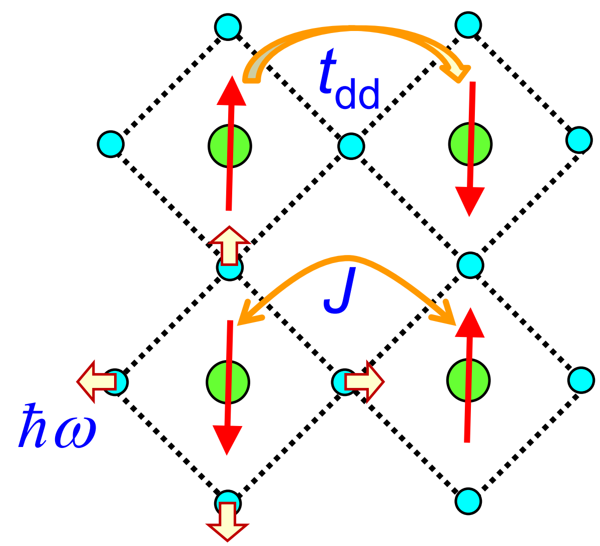


Recombination of a D-H pair ~200 fs ⇔ Rapid energy transfer to spin system?

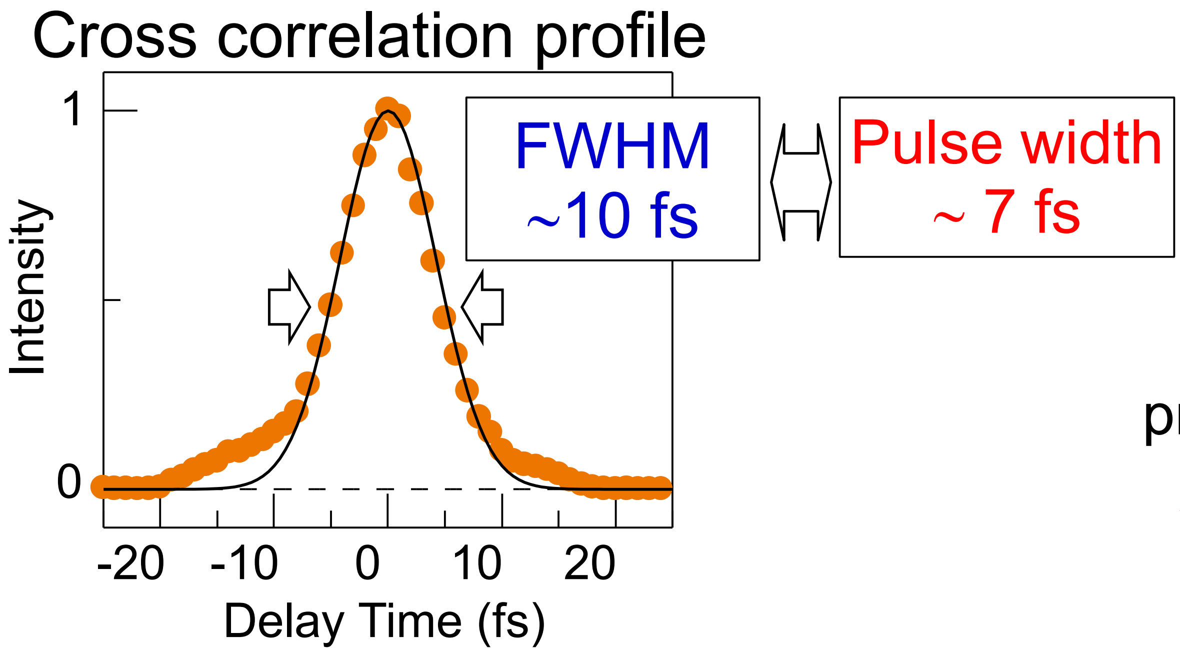
Metallization and relaxation processes << 100 fs ⇔ ????

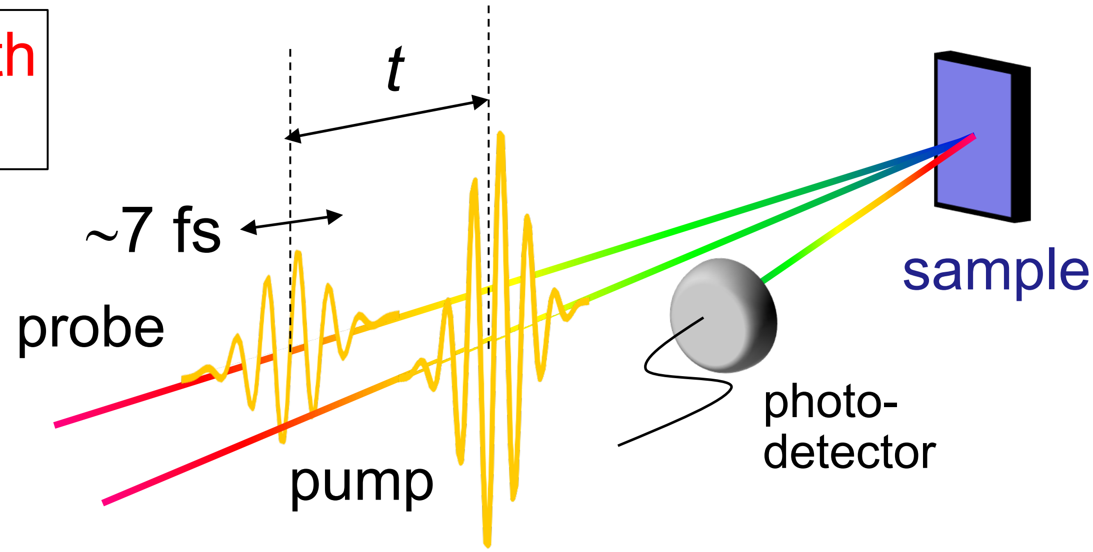
Transfer energy	t_{dd} , t_{pp} ~ 0.4 eV →	~ 10 fs
Exchange interaction	J ~ 0.13 eV →	~ 30 fs
Phonon frequency	$\hbar\omega$ ~500 cm^{-1} →	~ 60 fs

Time resolution of 10 fs is necessary.



Pump-probe experiments based on NOPA generating 7 fs pulses



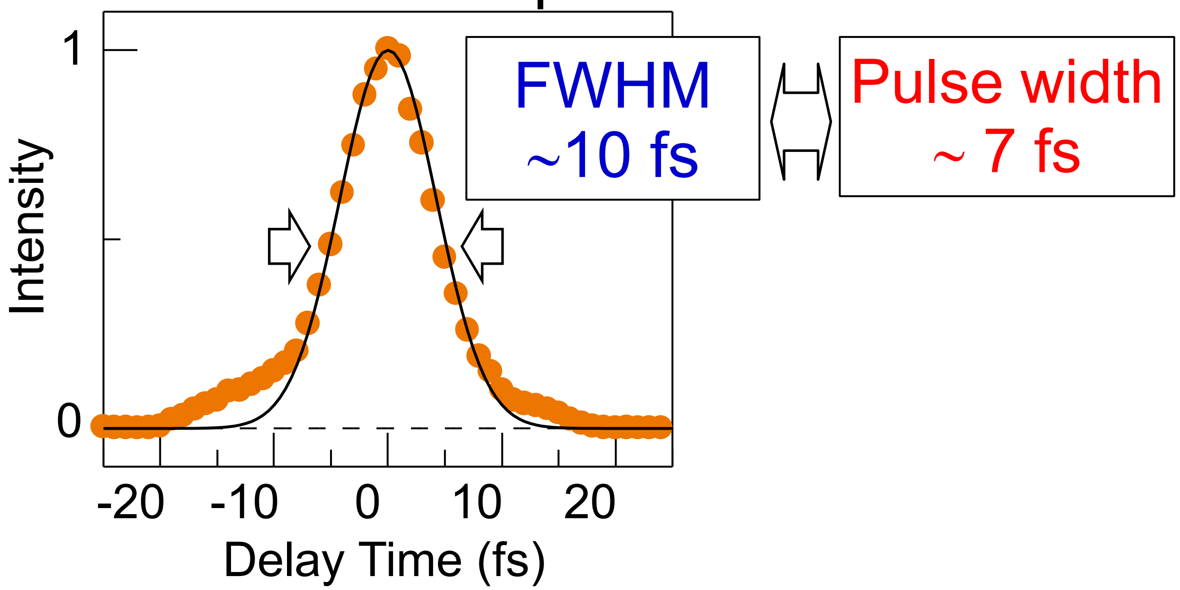


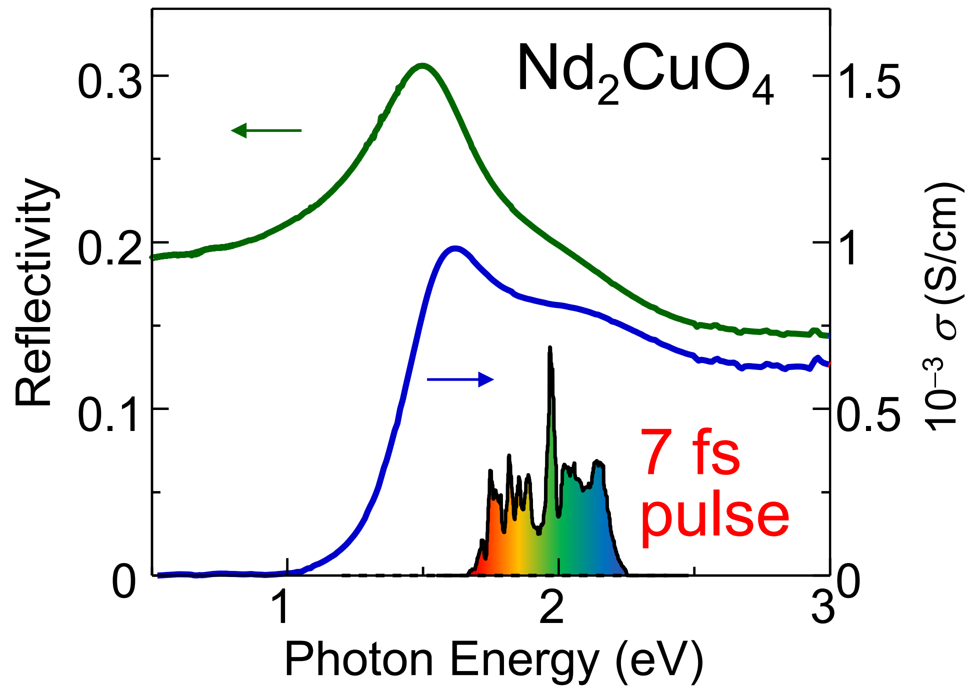
○ 7 fs pulse is divided to two beams.
→ a pump and a probe pulse

T. Miyamoto et al., Nat. Commun. ***9****, 3948 (2018)*

Pump-probe experiments based on NOPA generating 7 fs pulses

Cross correlation profile

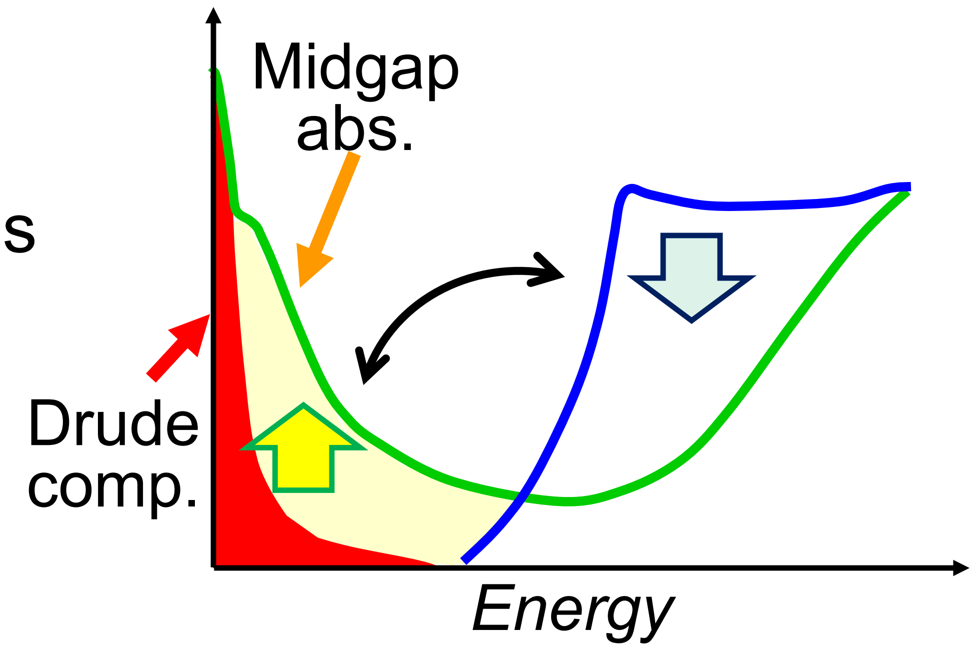




○ 7 fs pulse is divided to two beams.
→ a pump and a probe pulse

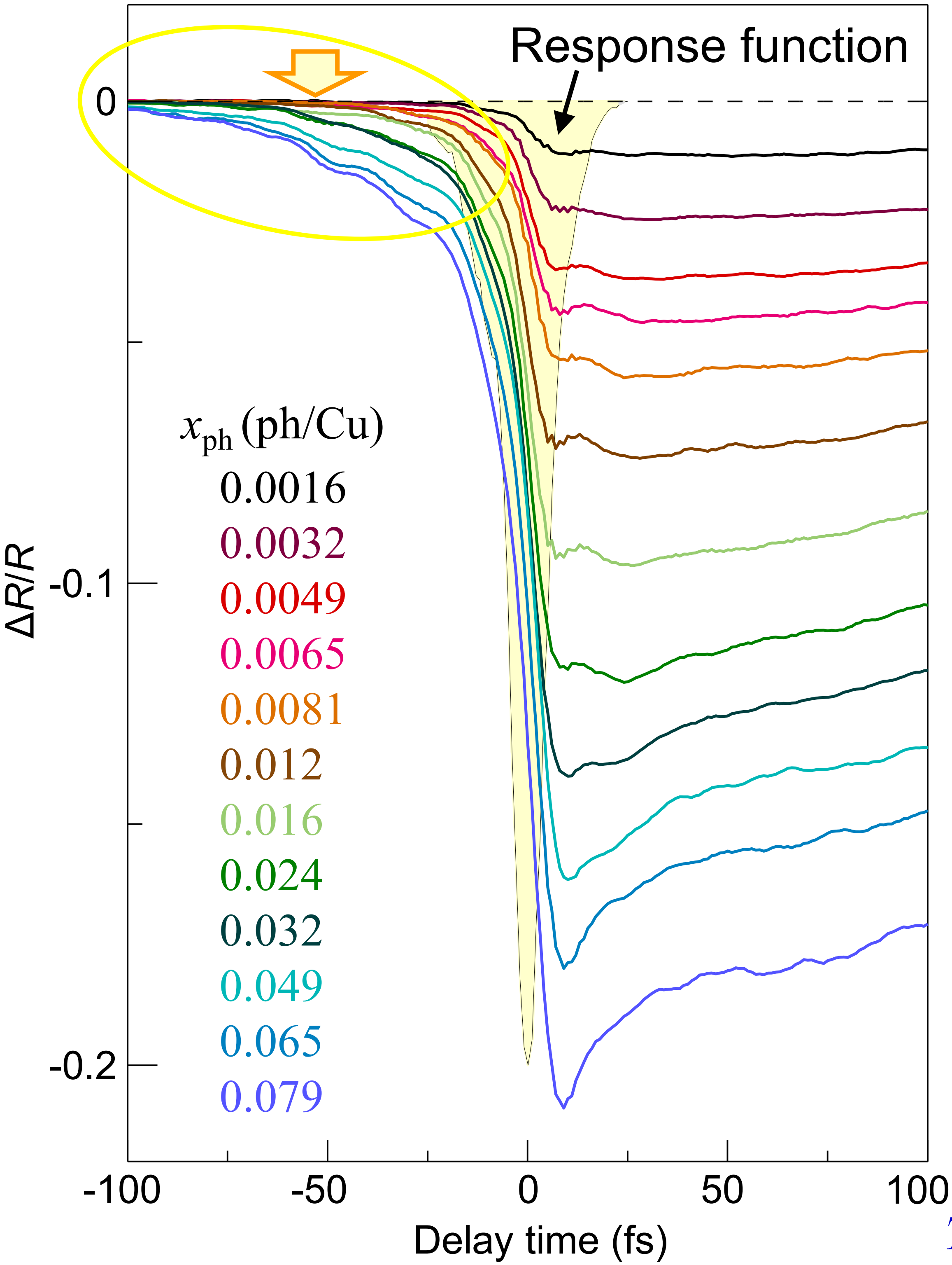
○ Time evolutions of bleaching signals
→ dynamics of metallic states
→ dynamics of mid-gap states

Transient reflectivity measurements on Nd_2CuO_4



T. Miyamoto et al., Nat. Commun. ***9****, 3948 (2018)*

Excitation photon density dependence of bleaching signals in Nd_2CuO_4



Perturbed free induction decay of probe-pulse-induced polarization

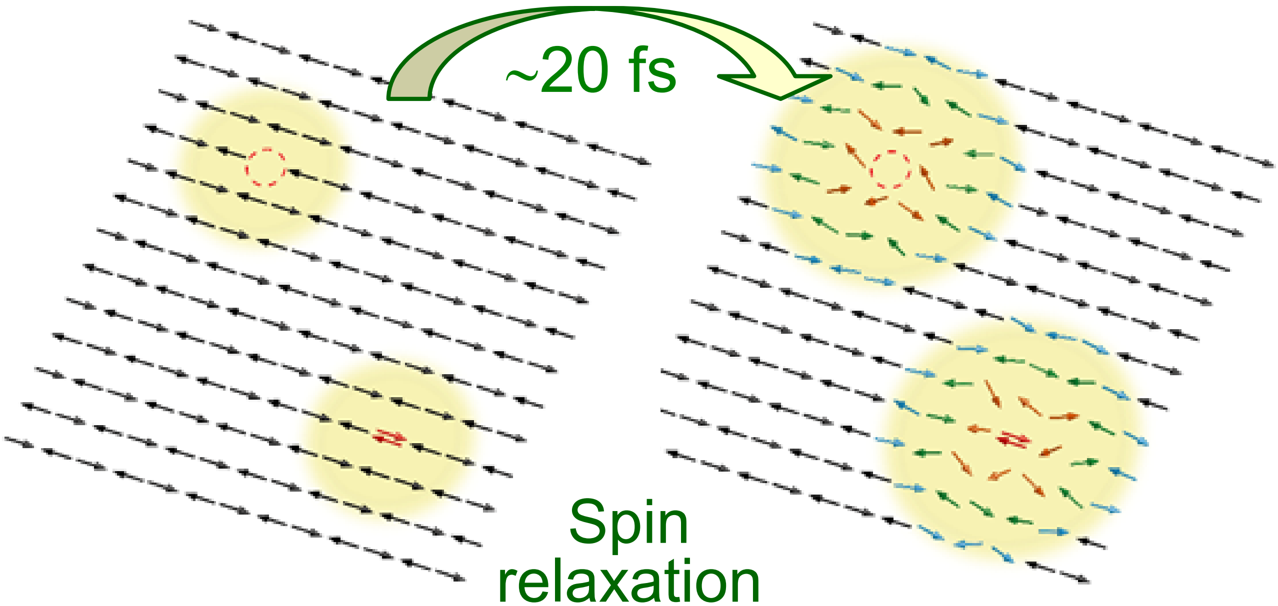
C.H. Brito Cruz et. al., IEEE J. Quantum Electron. ***24*** *261 (1988)*

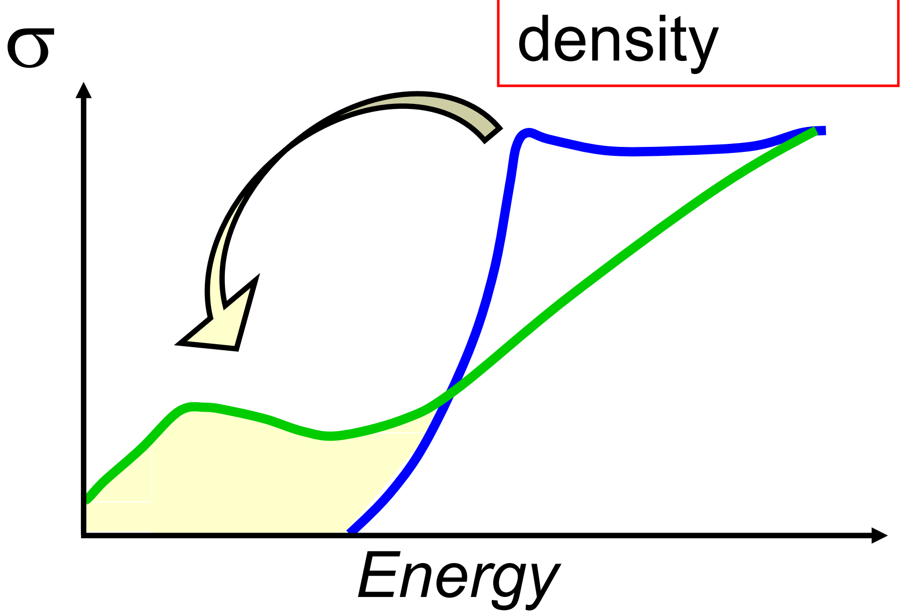
T. Miyamoto et al., Nat. Commun. ***9****, 3948 (2018)*

Excitation photon density dependence of bleaching signals in Nd_2CuO_4

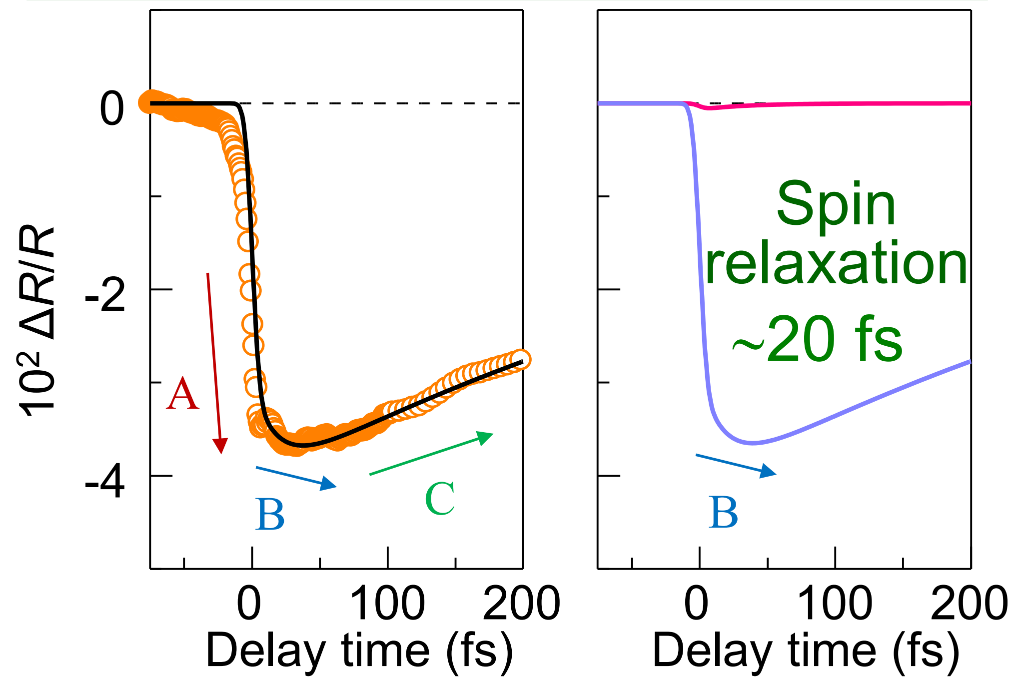
Time scale of phonon : $\hbar\omega$ ~500 cm^{-1} ⇔ 60 fs

Time scale of spin : J ~ 0.13 eV ⇔ 30 fs

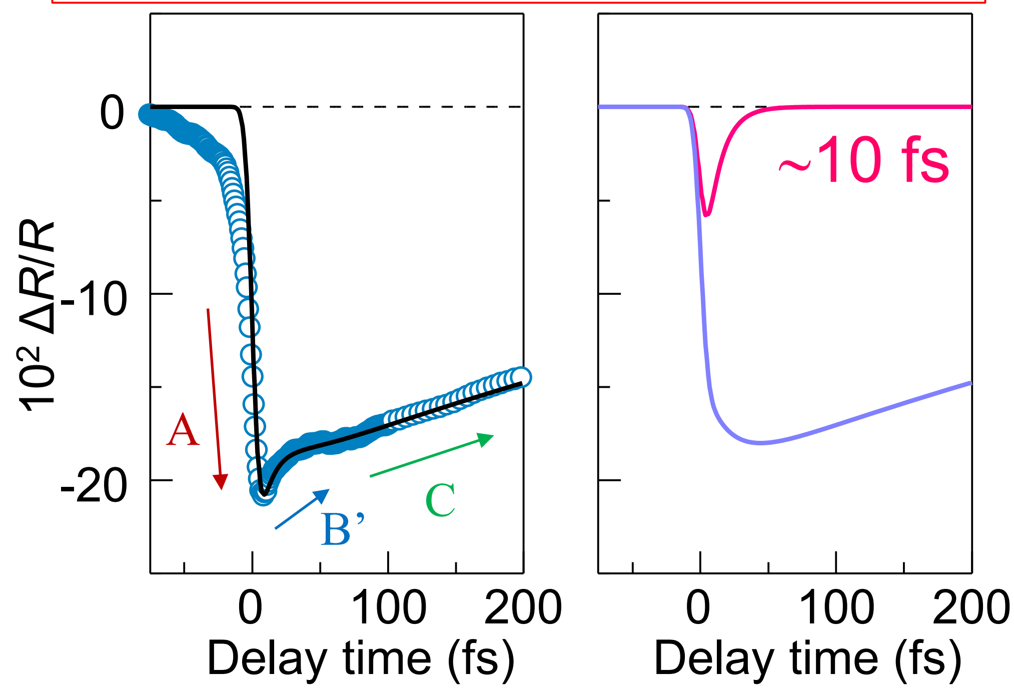




Low photon density 0.0081 ph/Cu



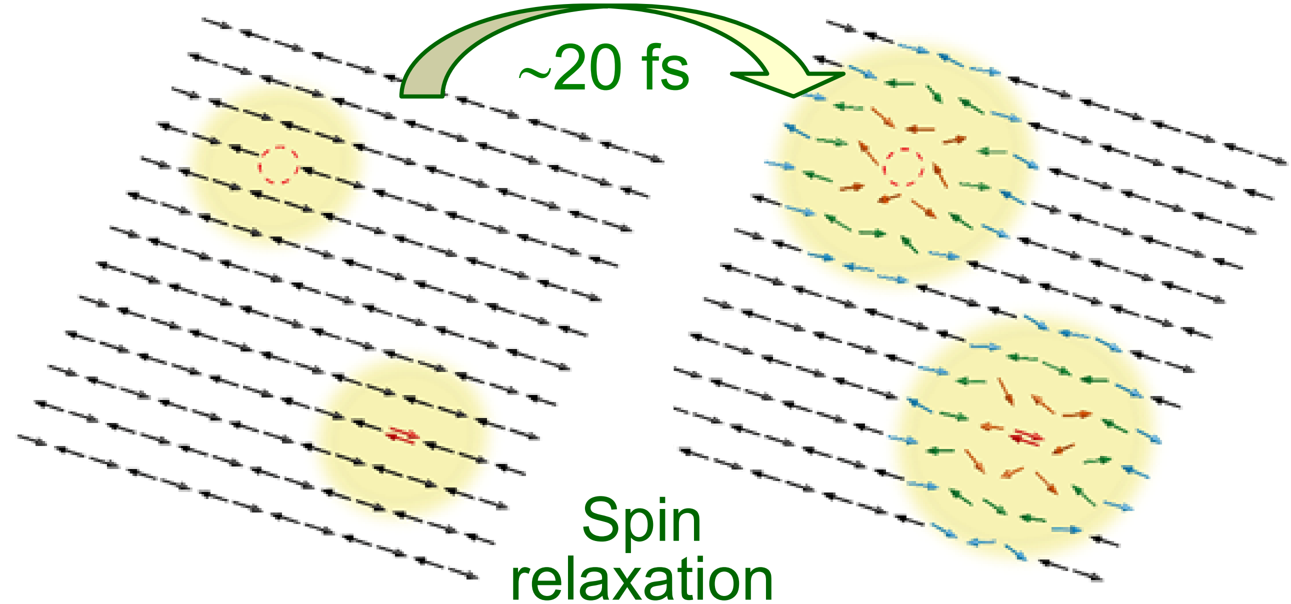
High photon density 0.079 ph/Cu



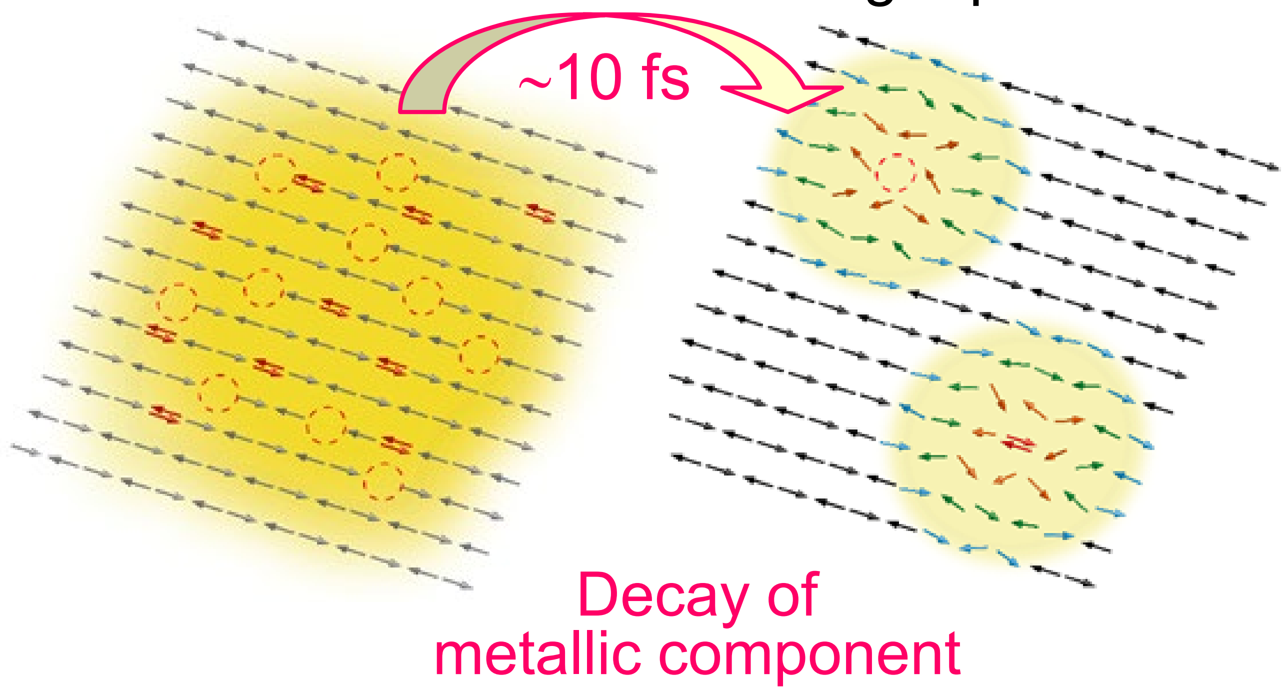
Excitation photon density dependence of bleaching signals in Nd_2CuO_4

Time scale of phonon : $\hbar\omega$ ~500 cm^{-1} ⇔ 60 fs

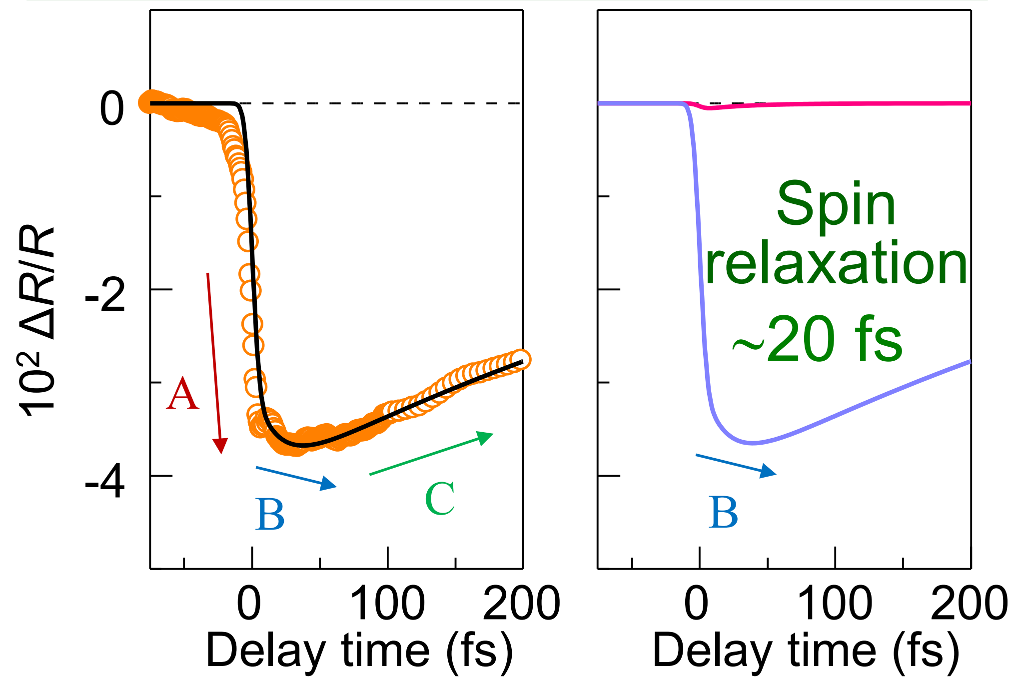
Time scale of spin : J ~ 0.13 eV ⇔ 30 fs



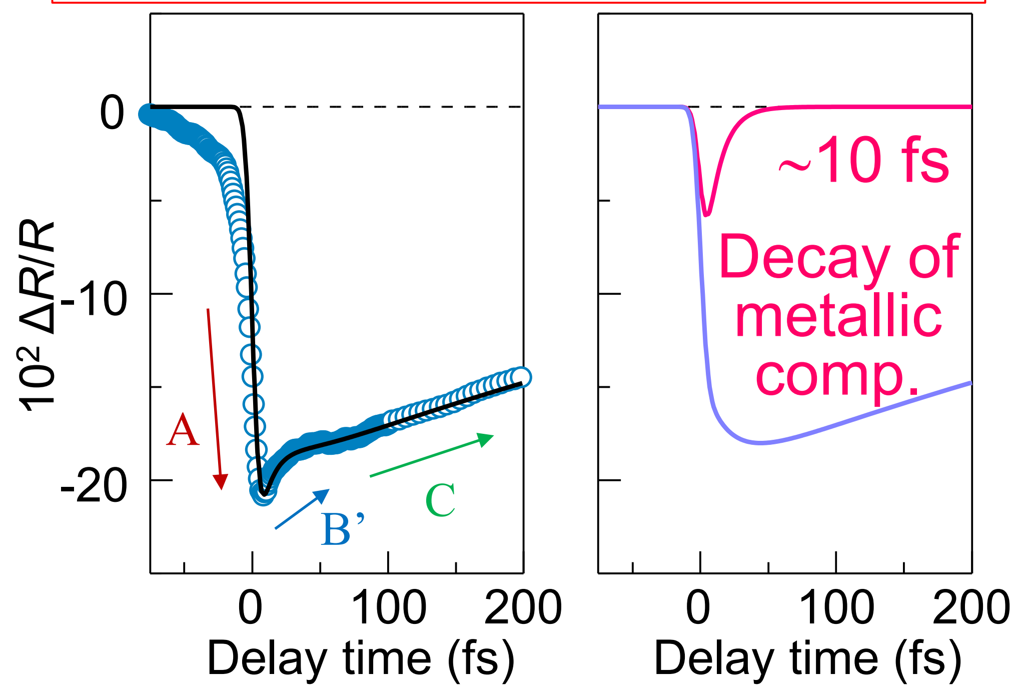
Recombination of carriers : Auger process



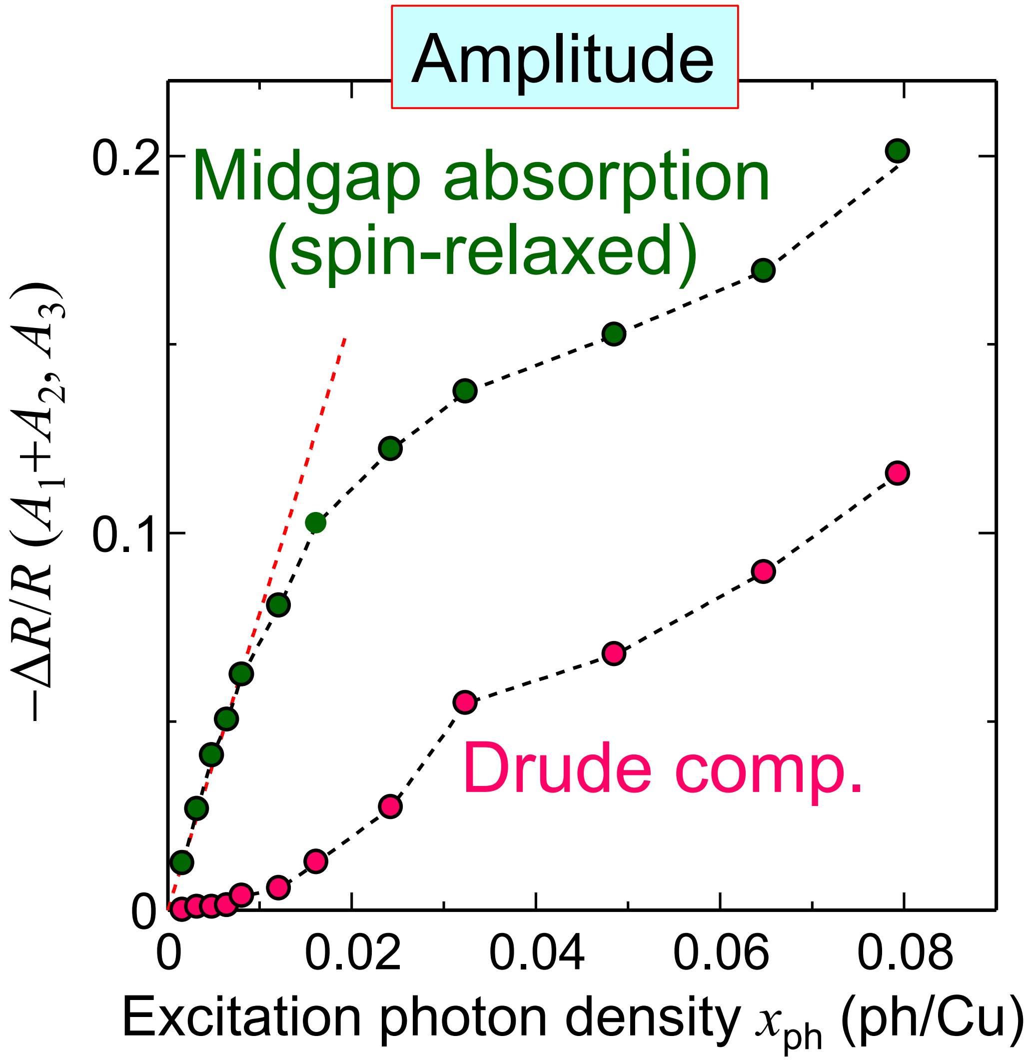
Low photon density 0.0081 ph/Cu

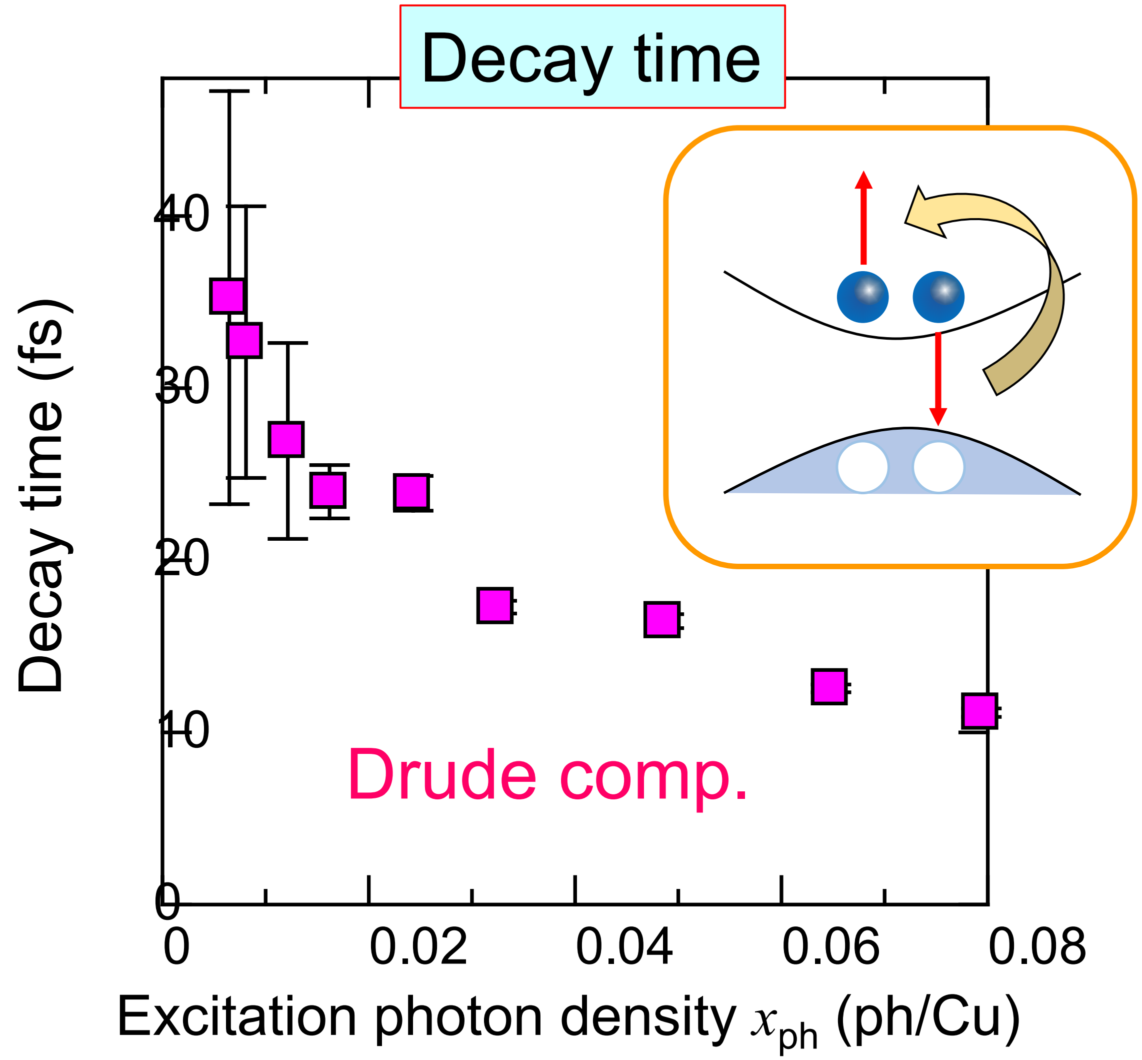


High photon density 0.079 ph/Cu



Excitation photon density dependence of the Drude component





Threshold behavior
of the Drude component

Localization of carriers

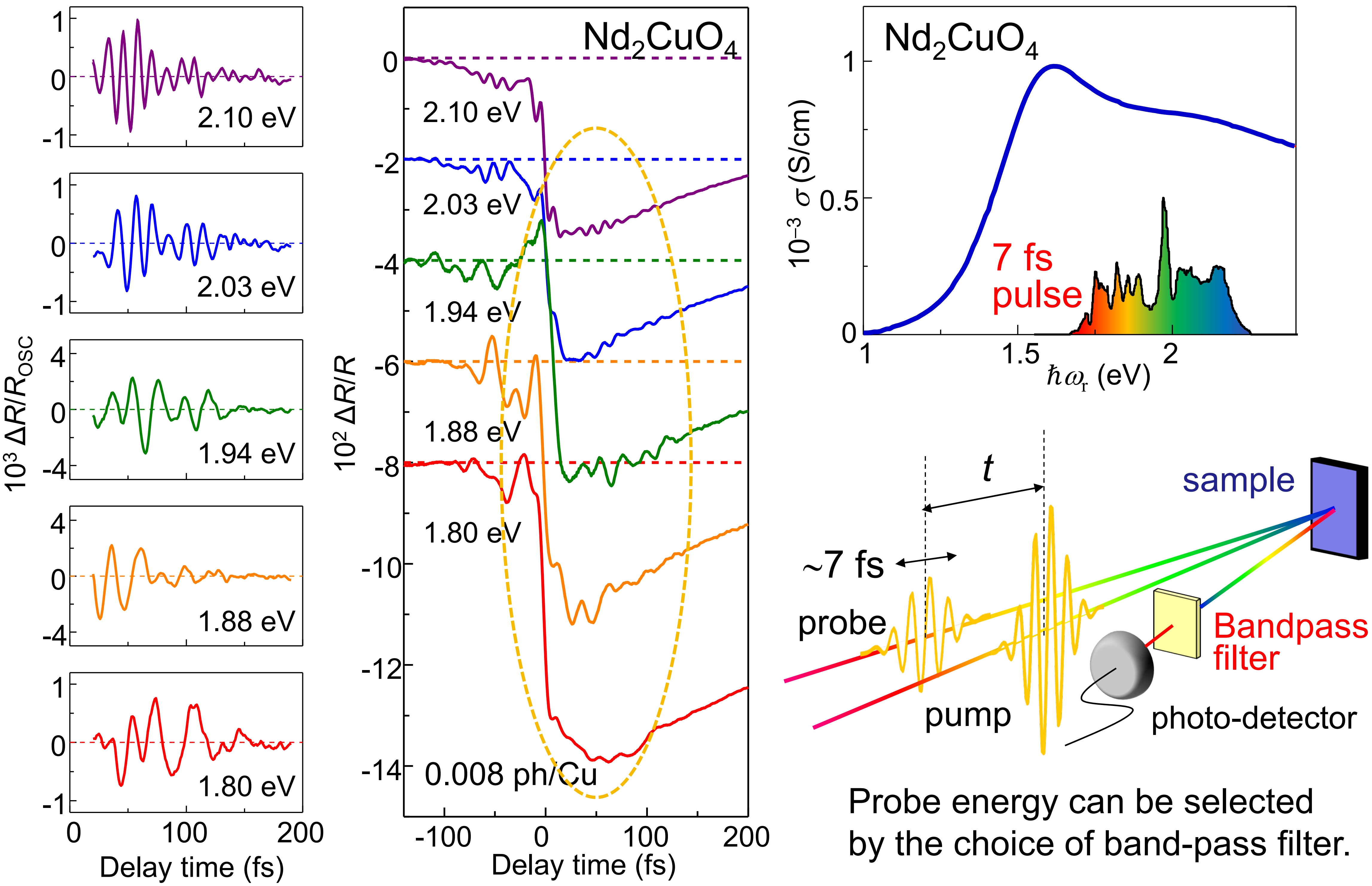
Charge-spin coupling

Decrease in the decay time
of Drude component

Auger recombination of carriers

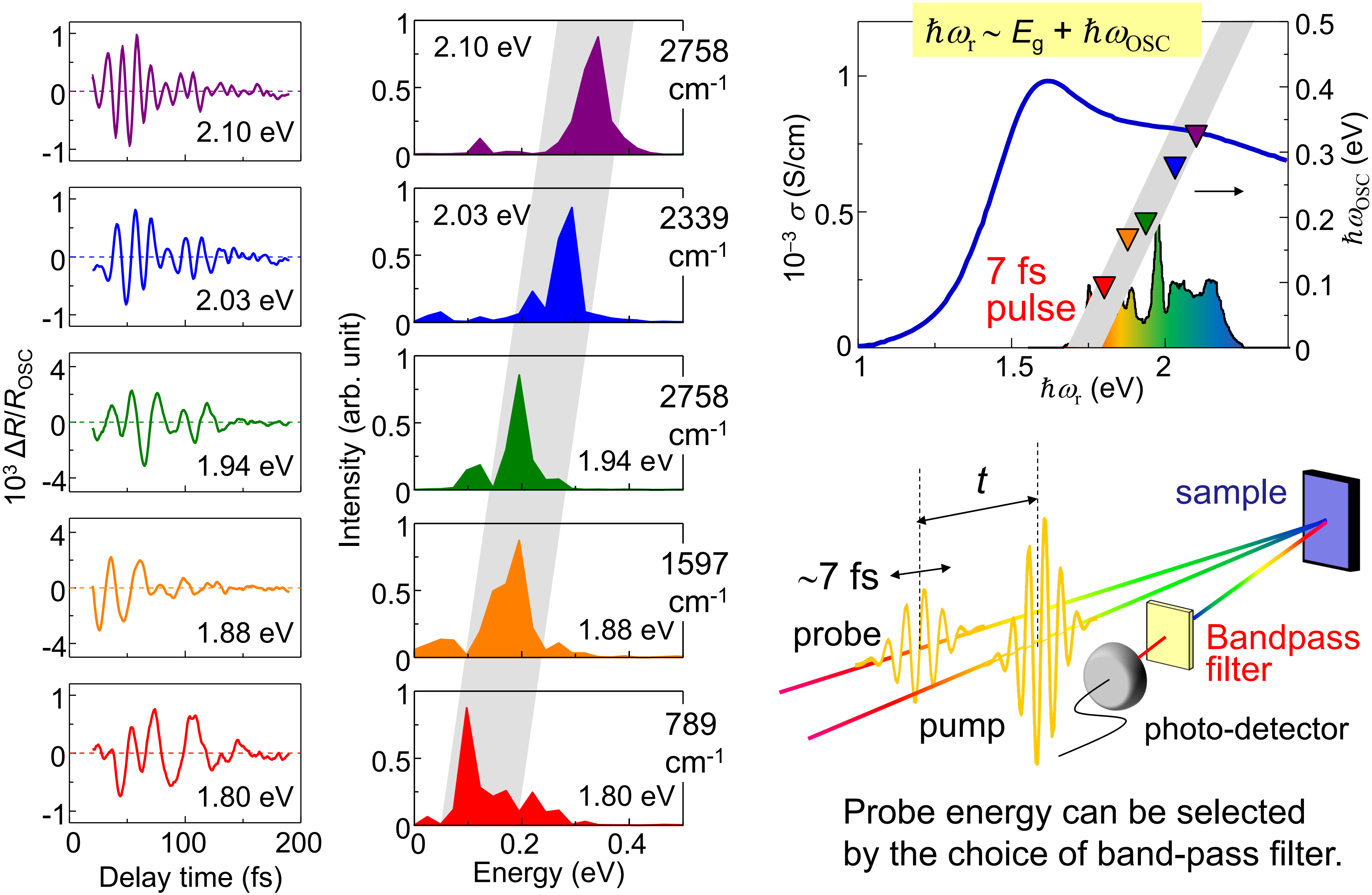
Strong e-e interaction

Pump-probe experiments based on NOPA generating 7 fs pulses



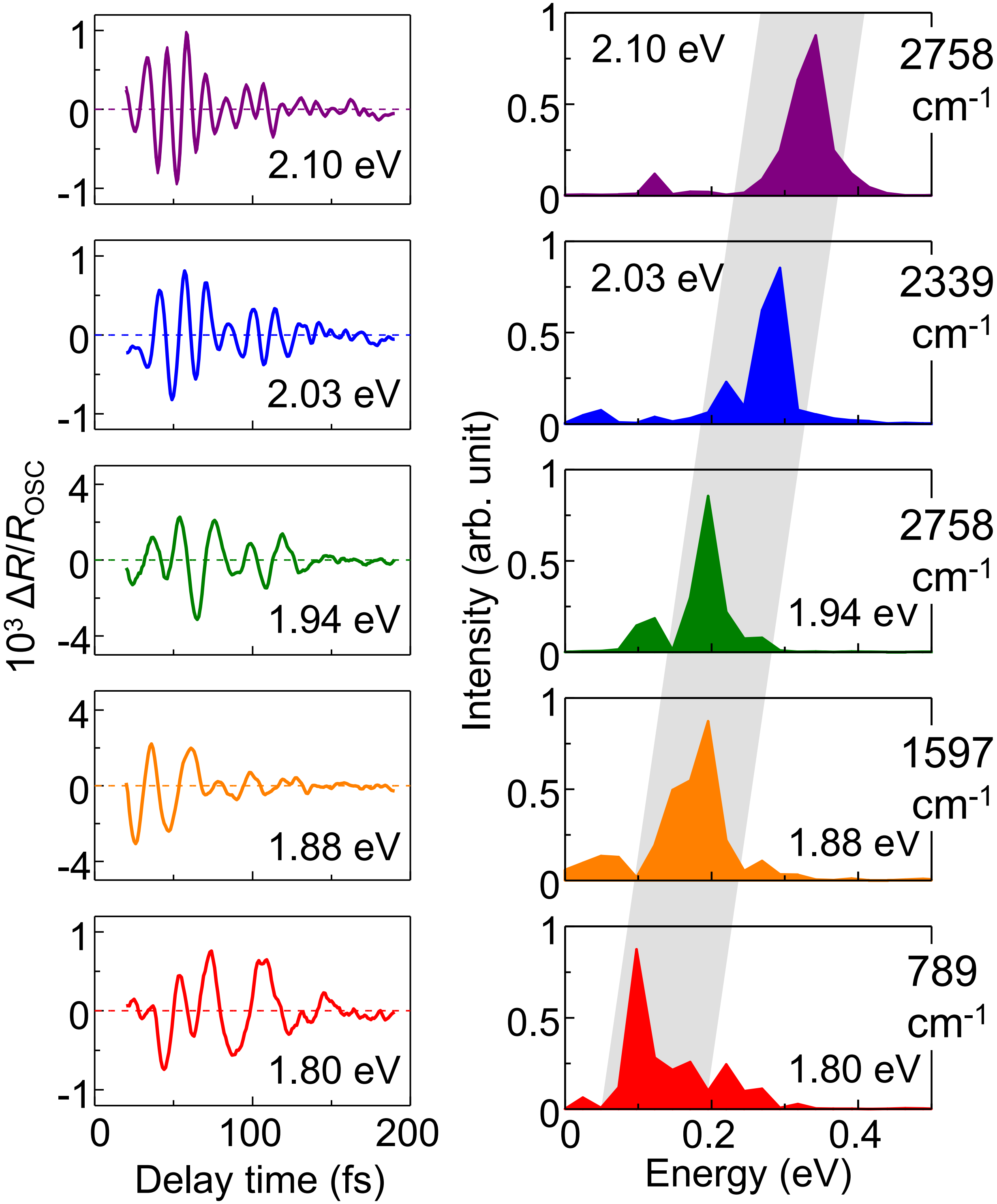
Probe energy can be selected by the choice of band-pass filter.

Pump-probe experiments based on NOPA generating 7 fs pulses



Probe energy can be selected by the choice of band-pass filter.

Pump-probe experiments based on NOPA generating 7 fs pulses



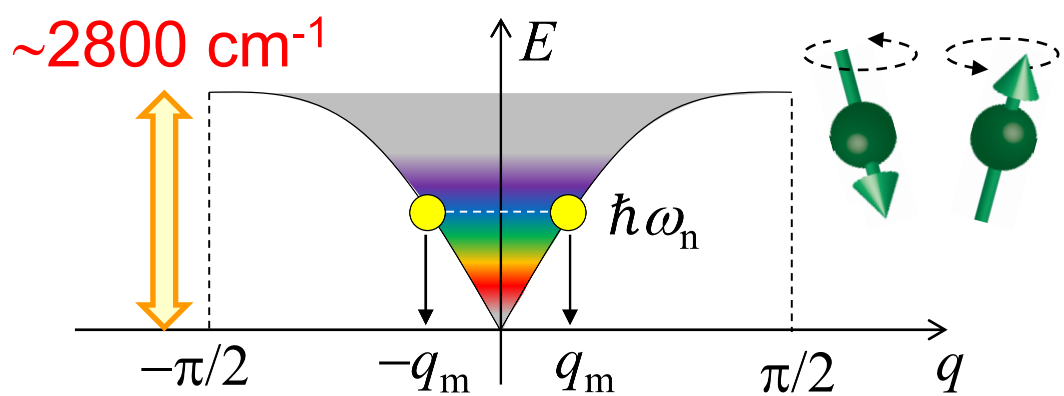
Characteristics

1. E_{OSC} depends on the probe energy (large dispersion).
2. E_{OSC} is very high.
   800–2800 cm^{-1}

✕ Optical phonon:
- no dispersion
- small energy

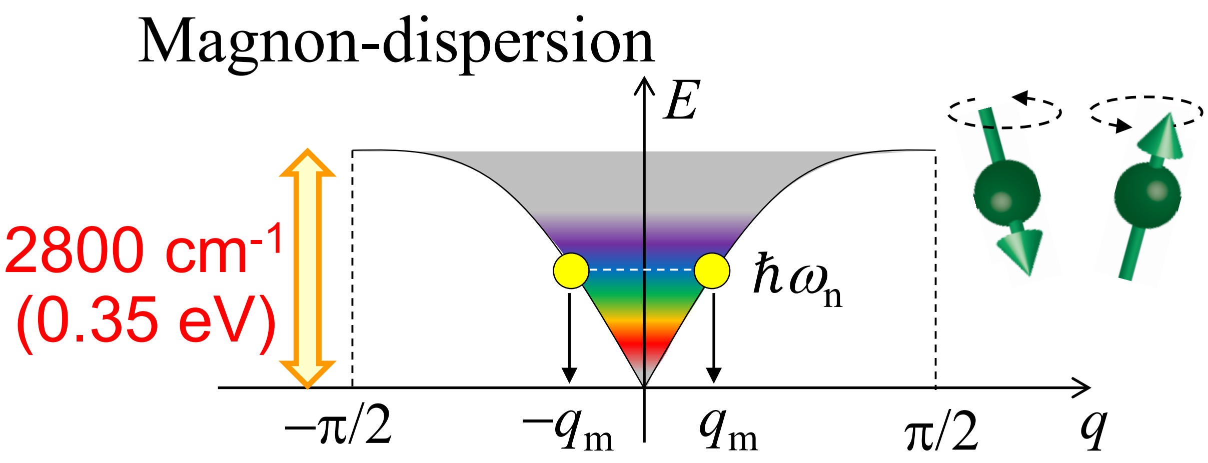
○ magnon:
- large dispersion
- large energy

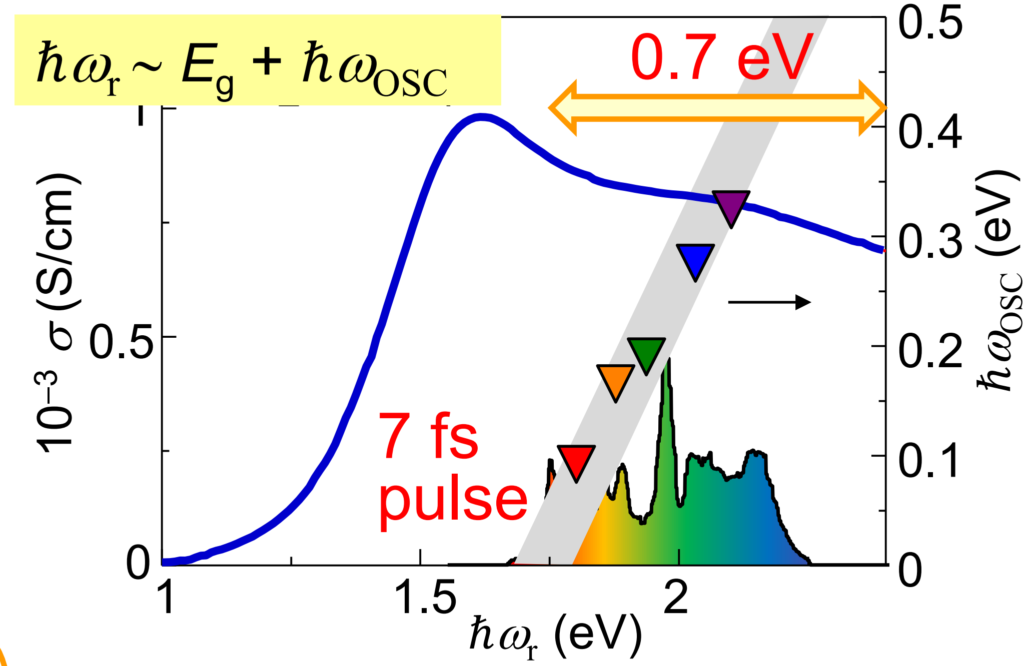
Magnon-dispersion

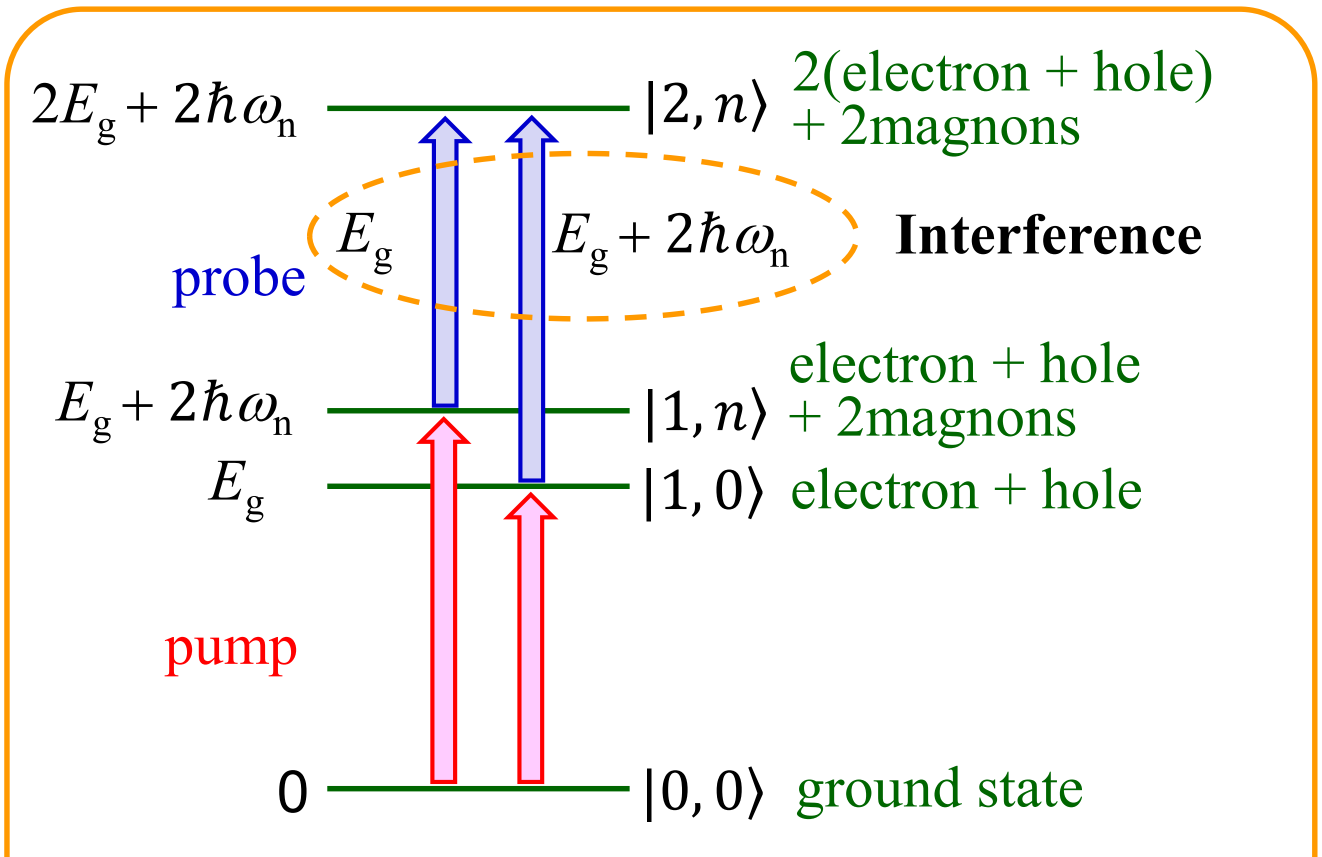


K. Ishii et al., Nat. Commun. **5***, 3714 (2014).*

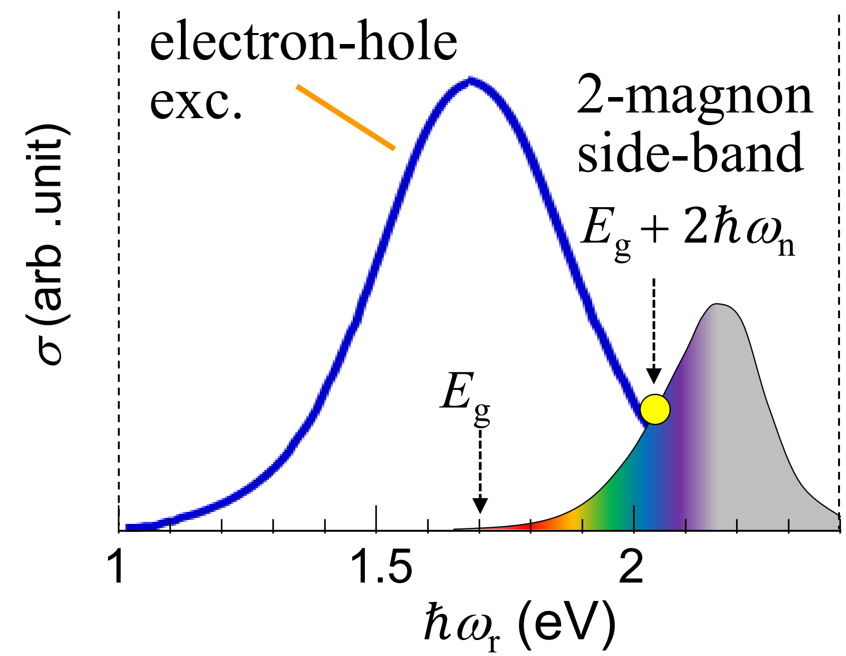
Interpretation of high-frequency oscillation (S. Ishihara)







T. Miyamoto et al., Nat. Commun. ***9****, 3948 (2018)*

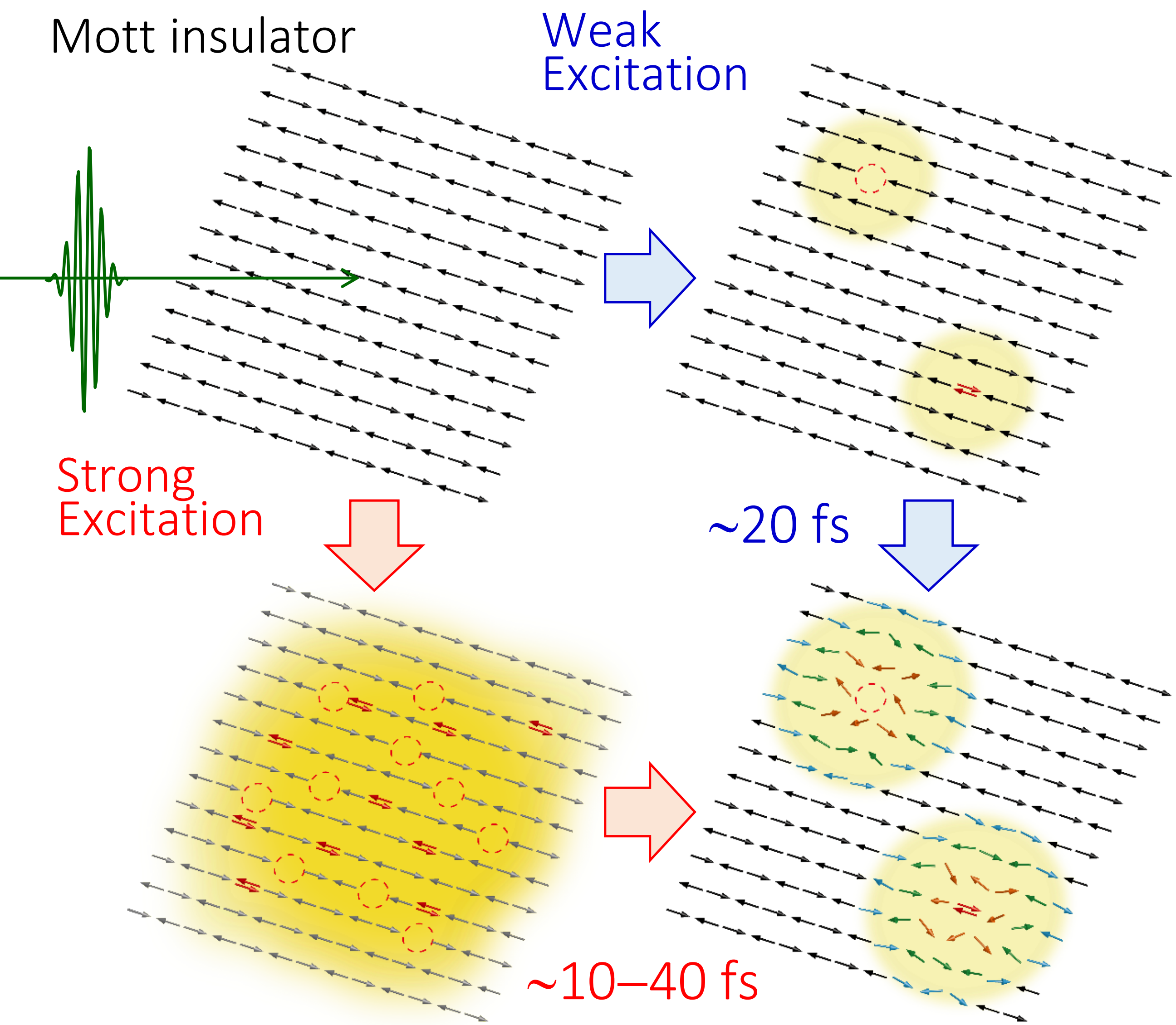


2 magnons with various frequencies

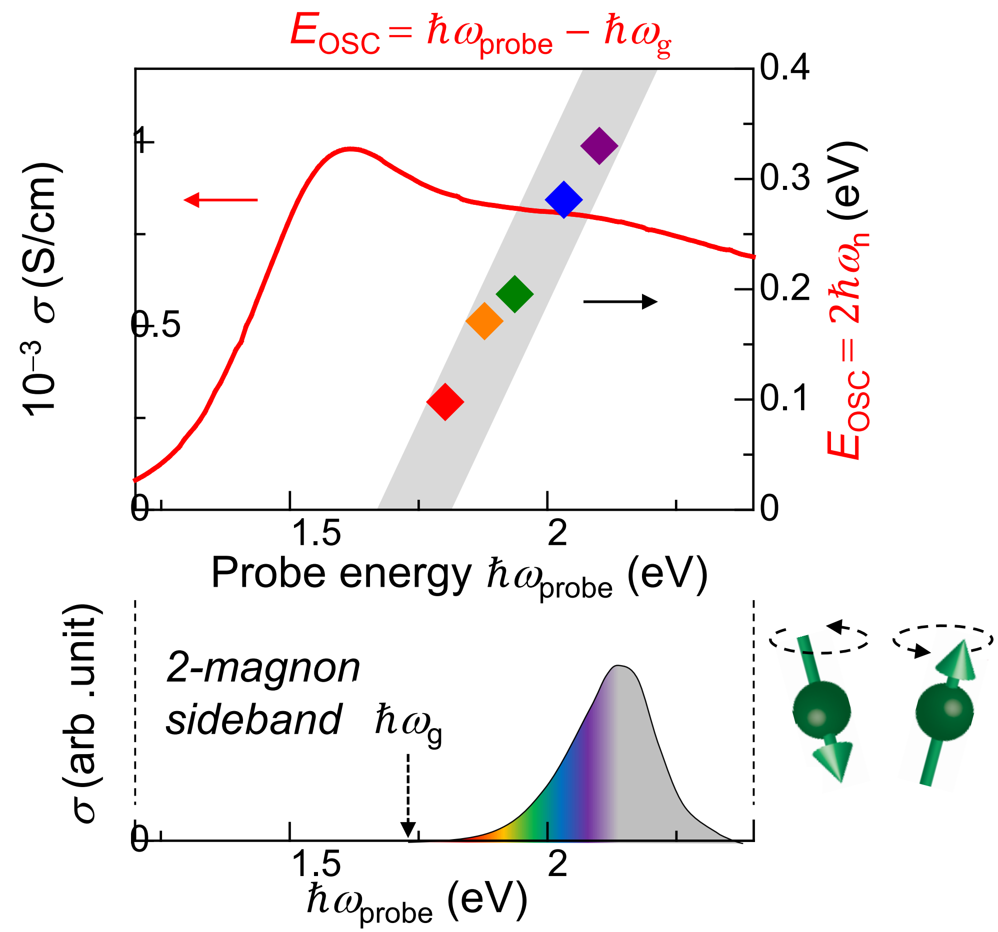
E. Hanamura, et al., PRB ***62****, 7033 (2000)*

Summary 1 : pump-probe spectroscopy with 10-fs time res. on Nd_2CuO_4

- Spin-relaxation in magnetic-polaron formations ~20 fs
- Auger carrier recombination ~10—40 fs



- Probe-energy dependent coherent oscillations
- Quantum interference of optical transitions between charge-spin coupled excited states



Strong coupling of charge and spin degrees of freedom in a 2D Mott insulator

１．イントロダクション

強相関系の光誘起相転移

２．光誘起モット絶縁体－金属転移

極短光パルスによる電荷･スピンダイナミクスの検出

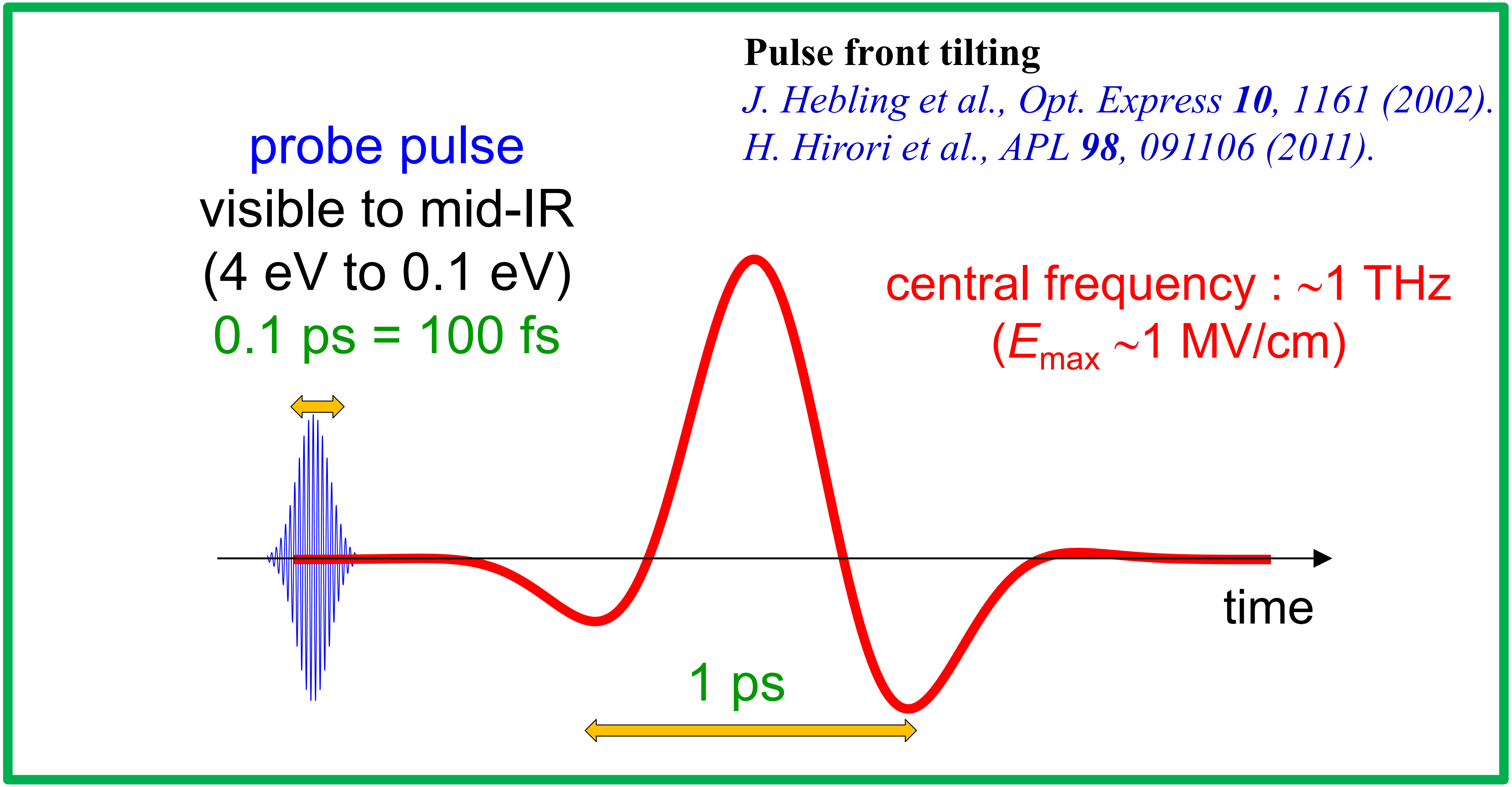
３．テラヘルツ(電場)誘起モット絶縁体－金属転移

量子トンネル過程によるキャリア生成と金属化の検出

４．中赤外(強電場)パルスによる電子相制御への展開

強誘電分極制御とイオン性－ 中性転移

Terahertz-pump optical-probe spectroscopy

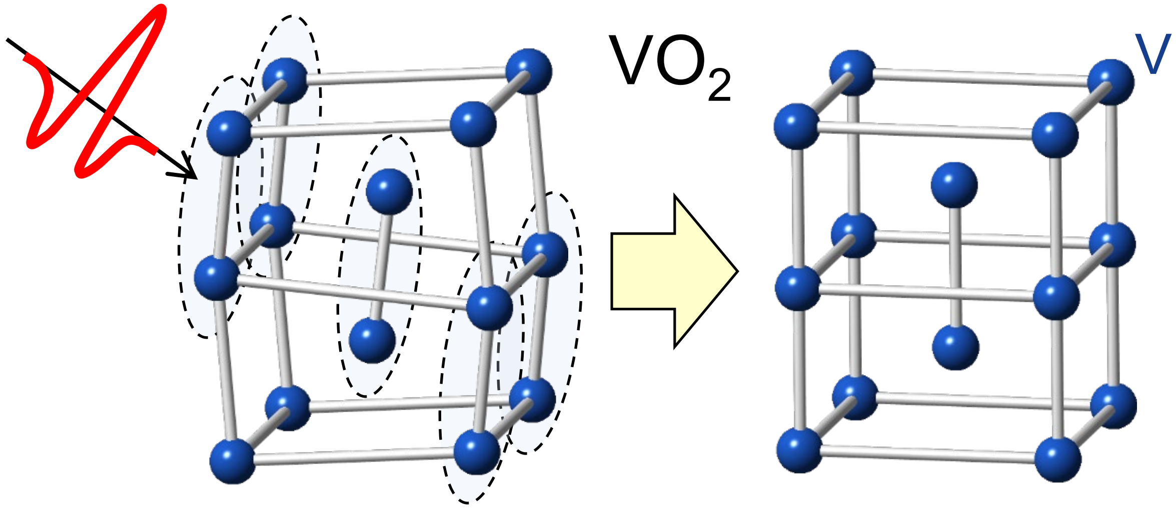


Wide-frequency range of spectral changes
along the terahertz electric fields can be obtained.

Electric-field-induced phase transitions in solids?

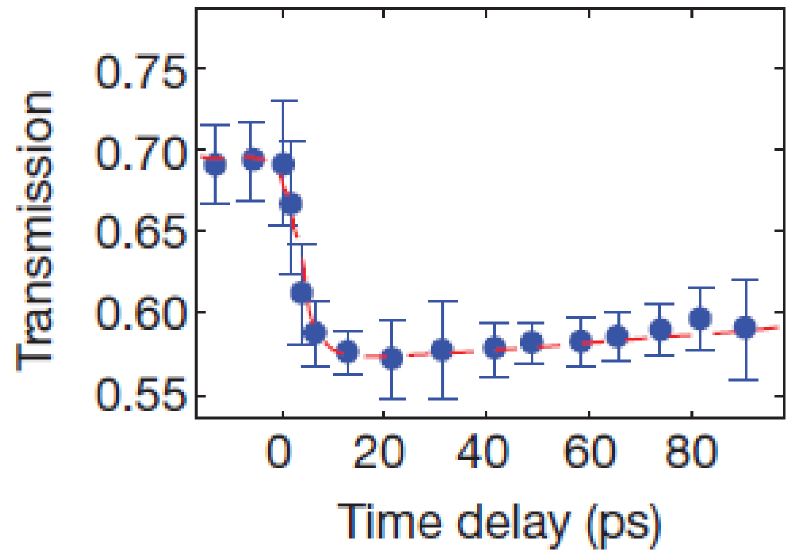
Attempts to drive a phase transition by a strong THz pulse

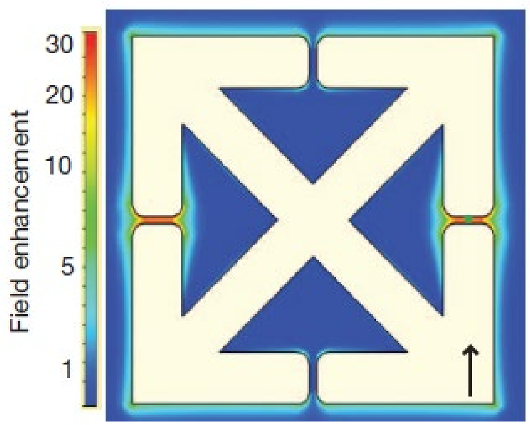
insulator-metal transition



Insulator

Metal





Split ring resonator

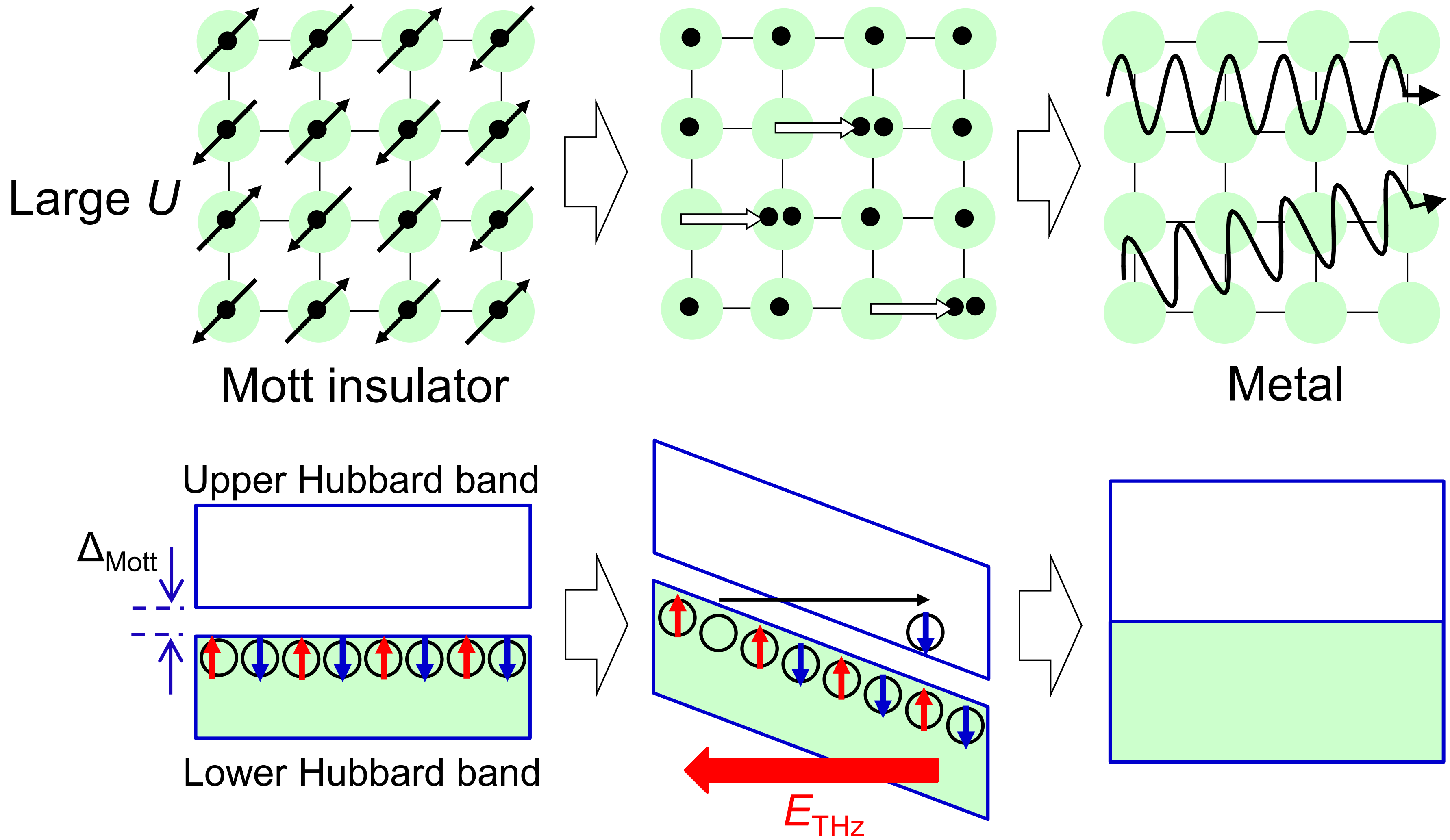
A large structural change

- A terahertz electric field enhanced in a metamaterial resonator ~2 MV/cm
- Slow dynamics >> 1 ps

M. Liu, R. Averitt, K.A. Nelson *et al*., *Nature* (2012)

To achieve an ultrafast phase control in a sub-picosecond time scale focus on electronic phase transitions without large structural changes.

THz-field-induced Mott insulator to metal transition



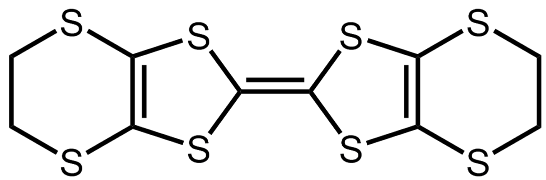
T. Oka, Phys. Rev. B ***86****, 075148 (2012)*

Theory	Δ_{Mott} (eV)	E_{th} (MV/cm)
ET-F_2TCNQ	0.7	3
Sr_2CuO_3	1.5	9

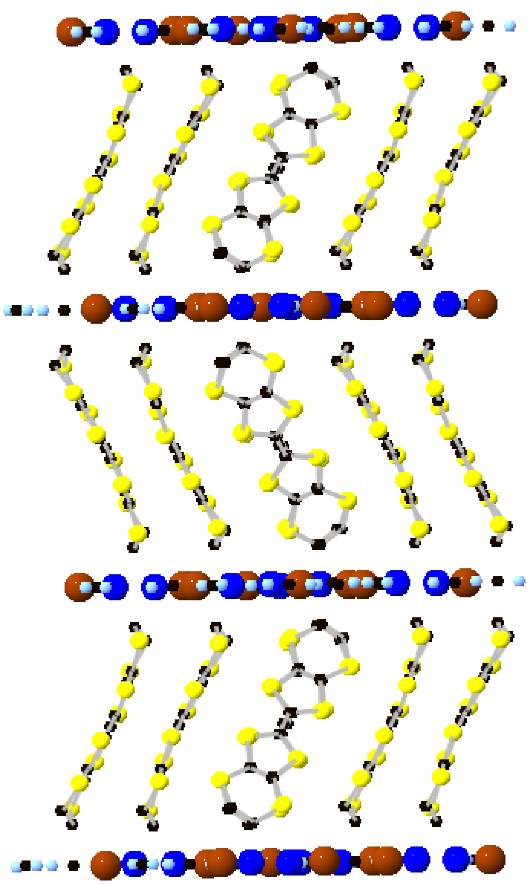
To drive a transition
by a lower electric field,
a narrow-gap Mott insulator
is advantageous.

Insulator-metal transition in κ-$(ET)_2X$

ET



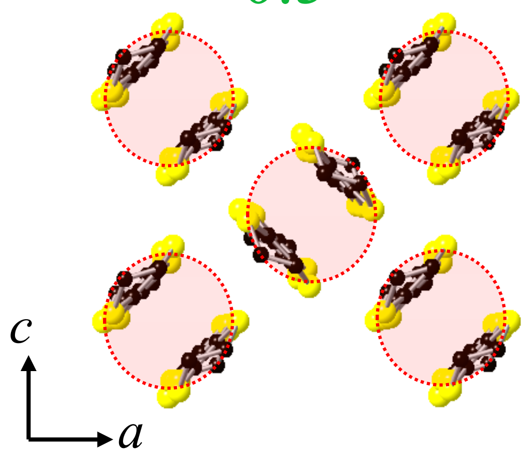
$(ET)_2^{+}X^{-}$

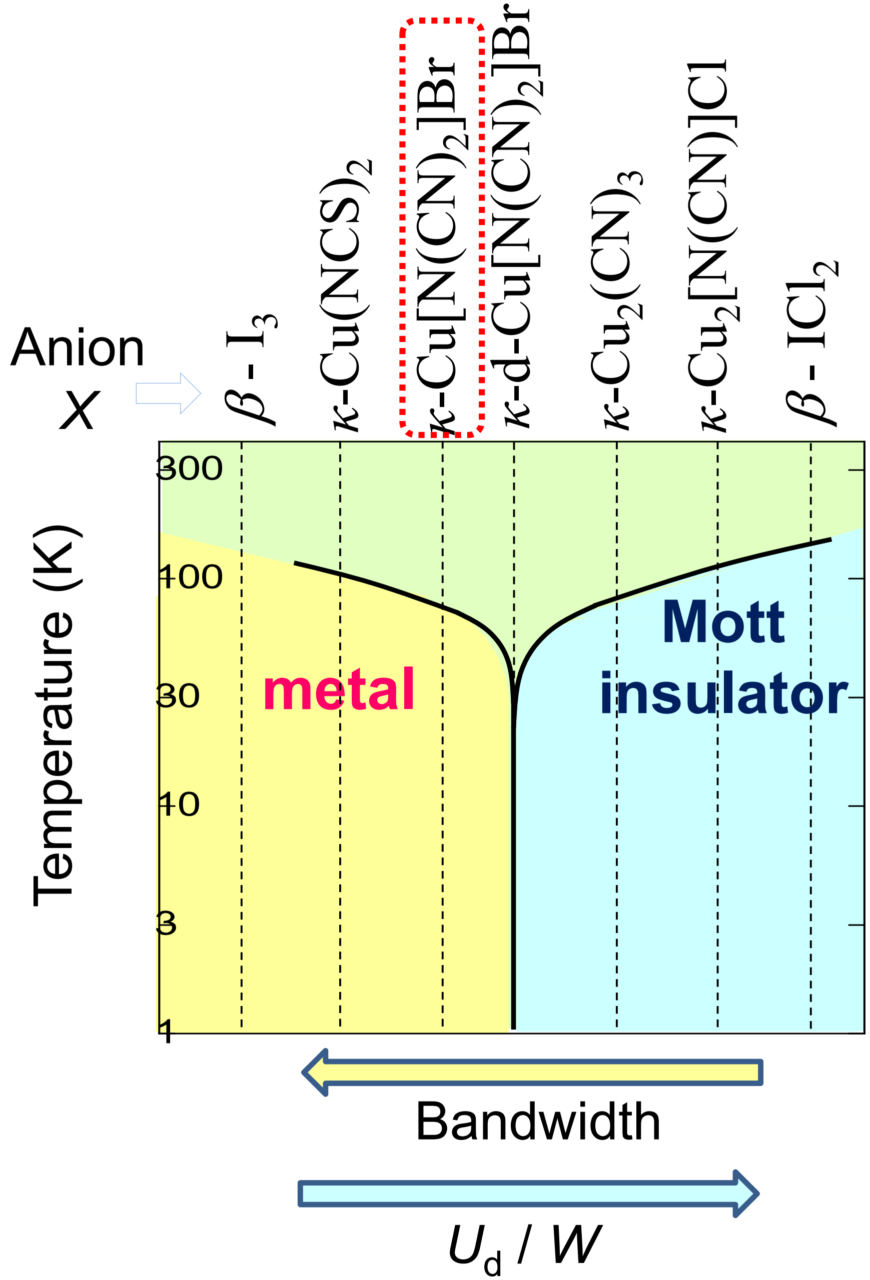


A unit is an ET dimer.
The nominal valence of each ET is +0.5.
A dimer has one hole.

2D half-filled band

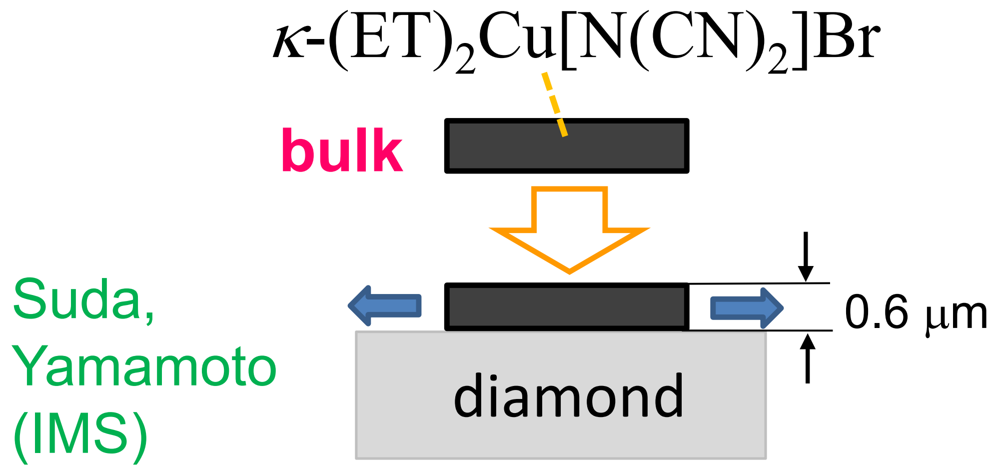
ET +0.5



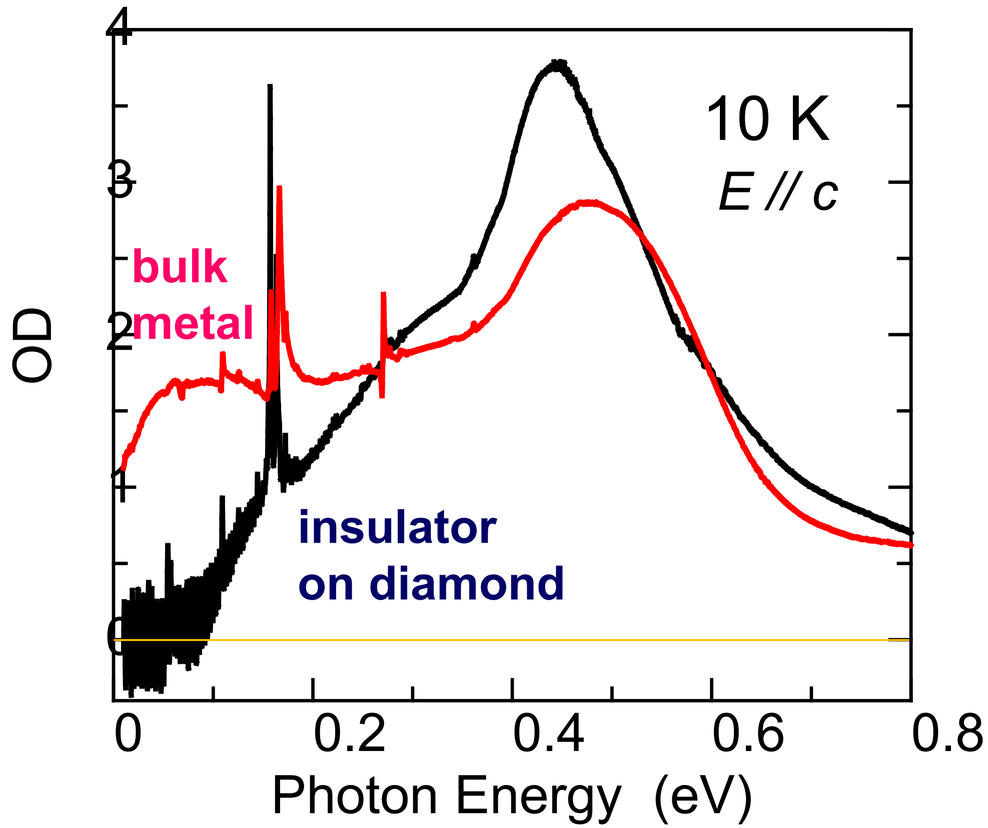


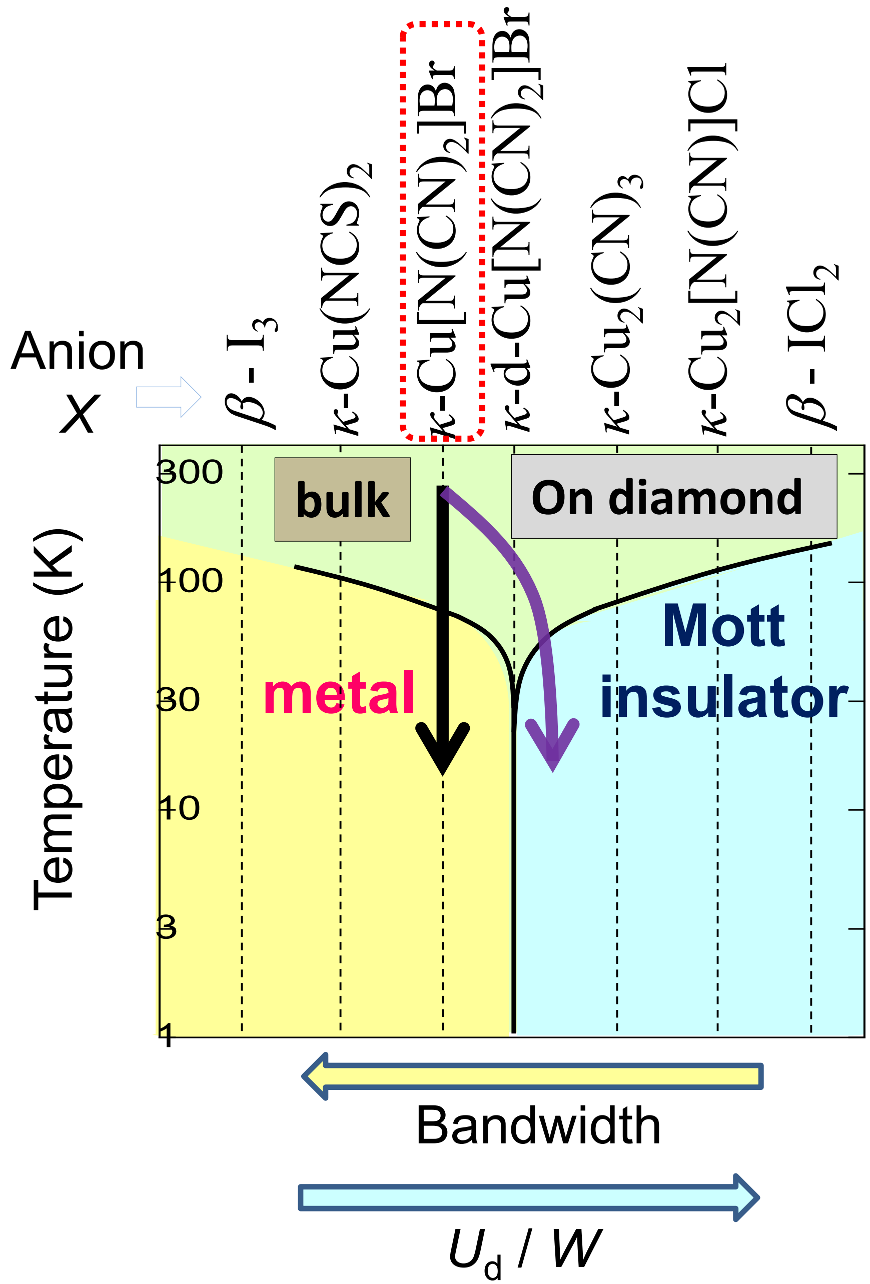
K. Kanoda, JPSJ **75**, 051007 (2006)

Insulator-metal transition in κ-$(ET)_2X$



Absorption (OD: optical density) spectra



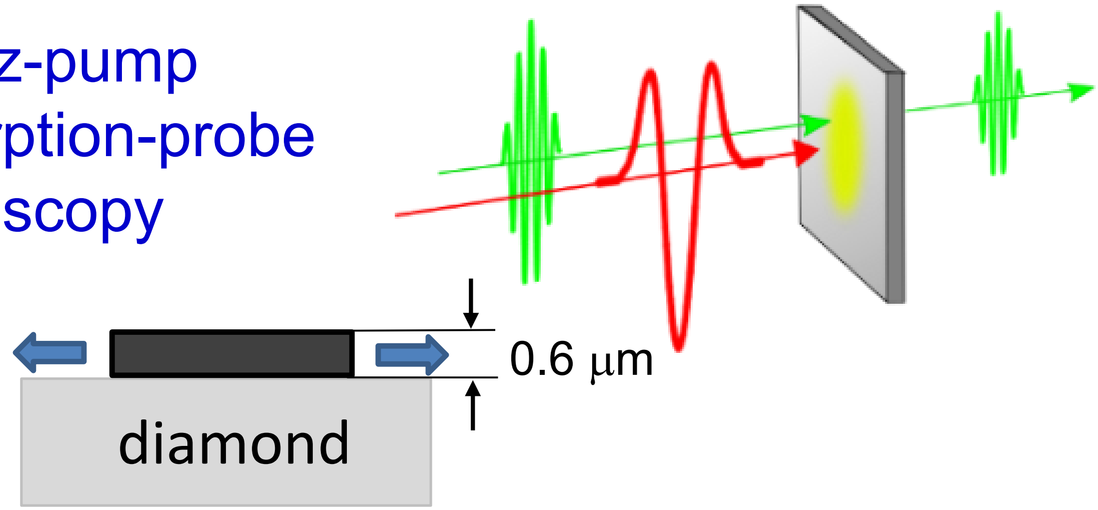


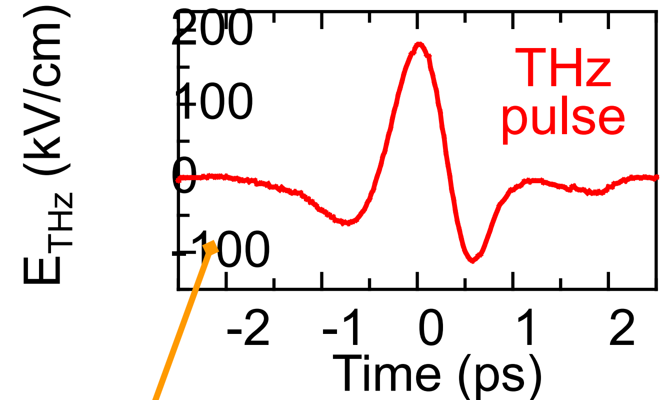
K. Kanoda, JPSJ **75**, 051007 (2006)

Insulator-metal transition in κ-$(ET)_2X$

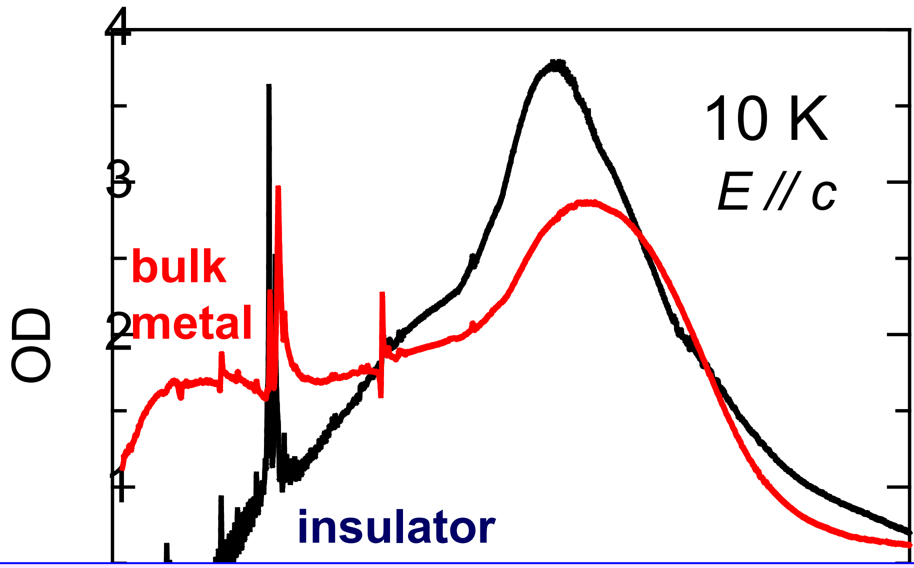
Terahertz-pump
optical-absorption-probe
spectroscopy

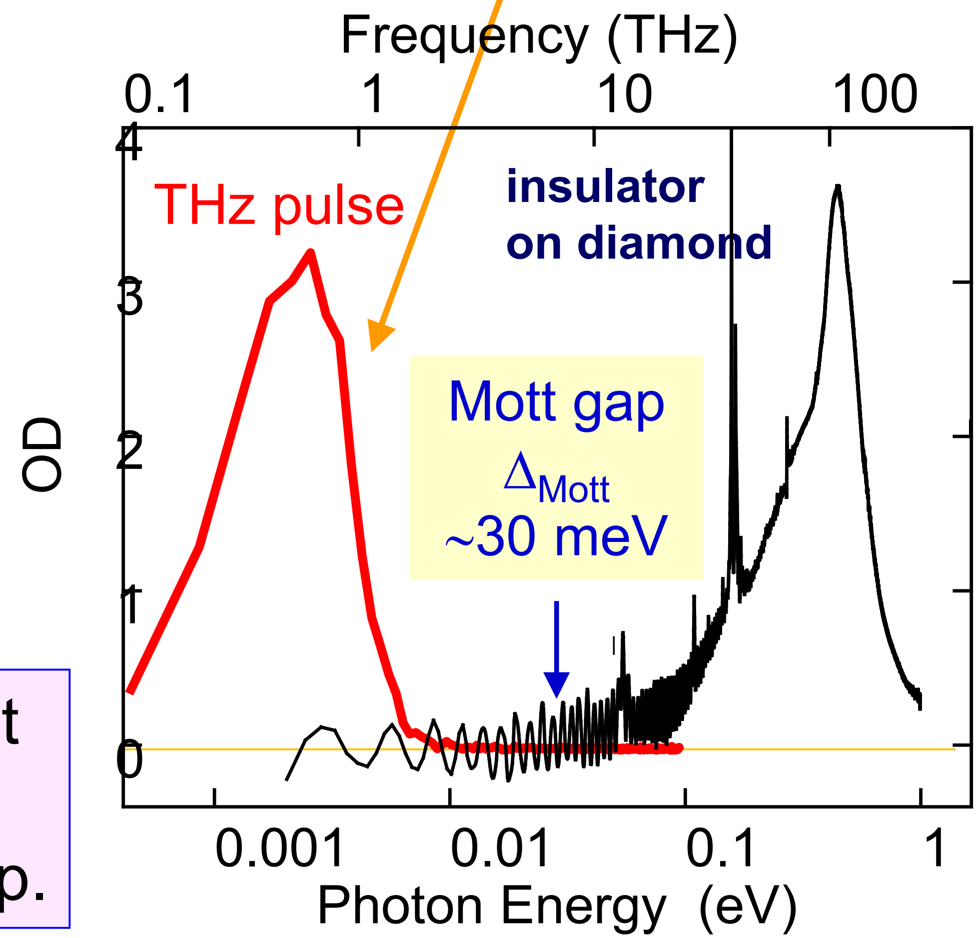
Suda,
Yamamoto
(IMS)





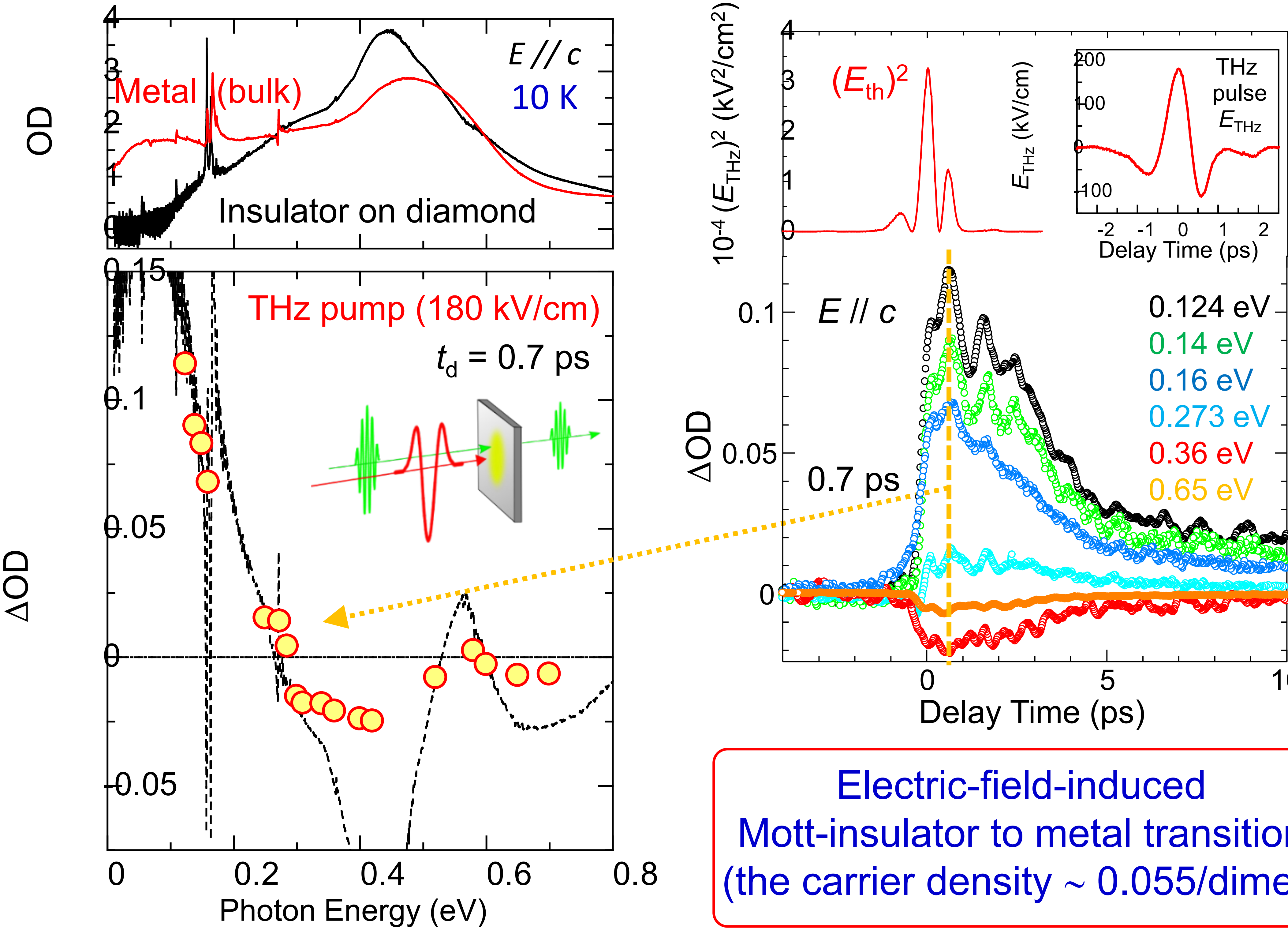
Absorption (OD: optical density) spectra





The sample is completely transparent for the THz pulse.
No carriers are excited beyond the gap.

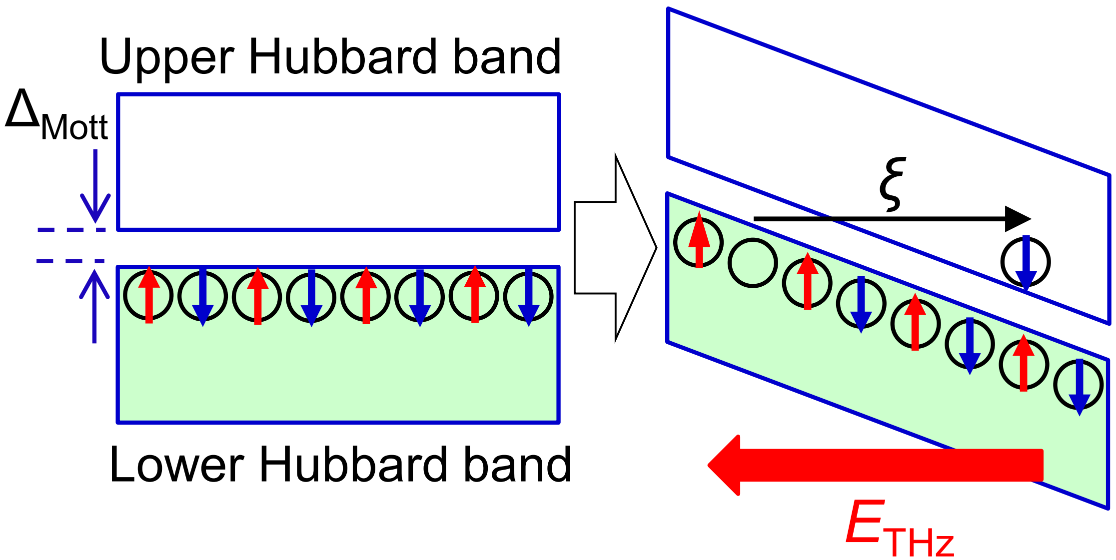
THz-pump optical-absorption-probe spectroscopy (180 kV/cm)

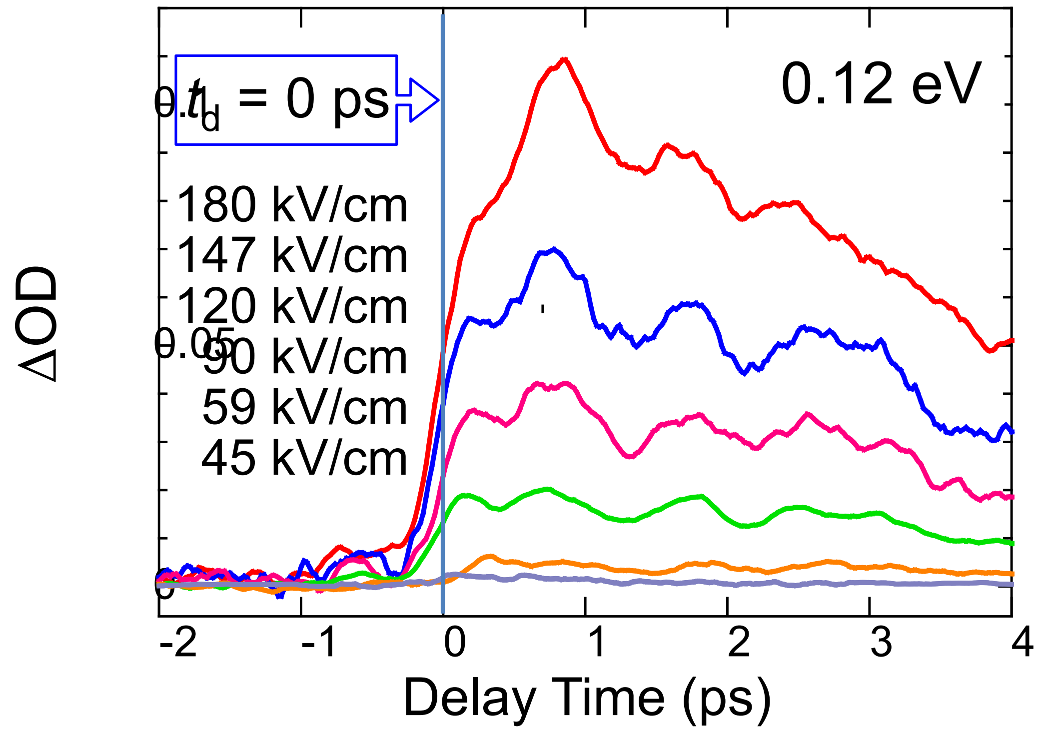


Electric-field-induced
Mott-insulator to metal transition
(the carrier density ~ 0.055/dimer)

THz electric-field dependence of absorption changes in IR region

Carrier generation via quantum tunneling



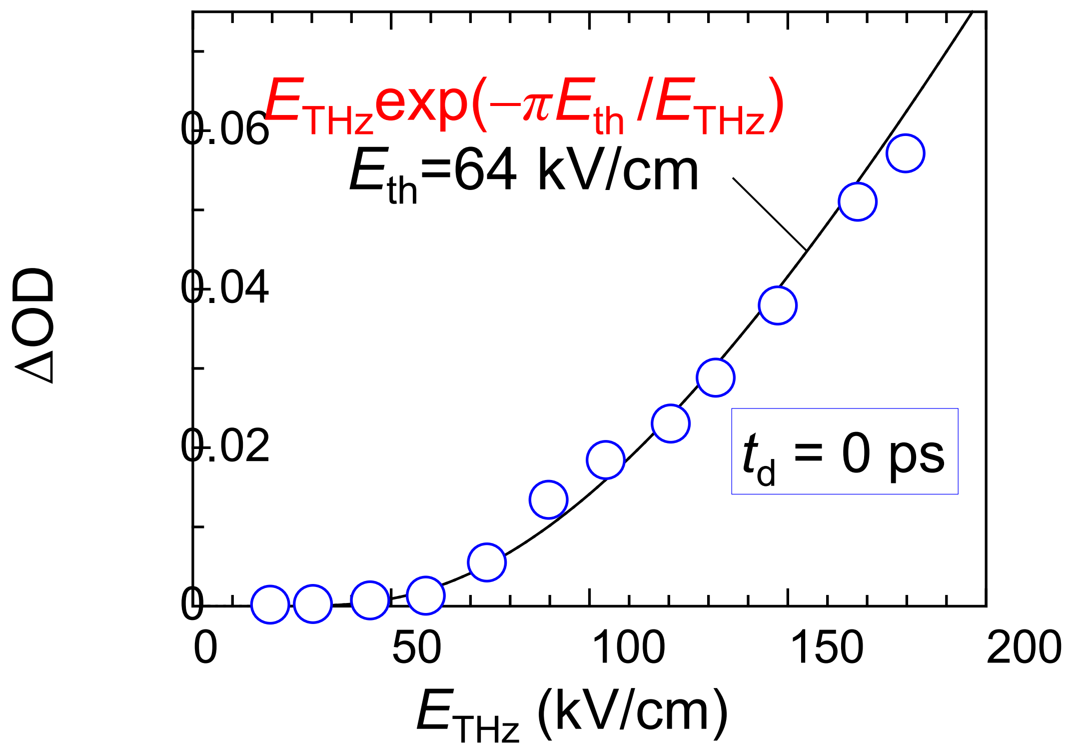


Probability of quantum tunneling

$E_{THz}\exp(-\pi E_{th}/E_{THz}) \Leftrightarrow \Delta OD$

T. Oka, Phys. Rev. B ***86****, 075148 (2012)*

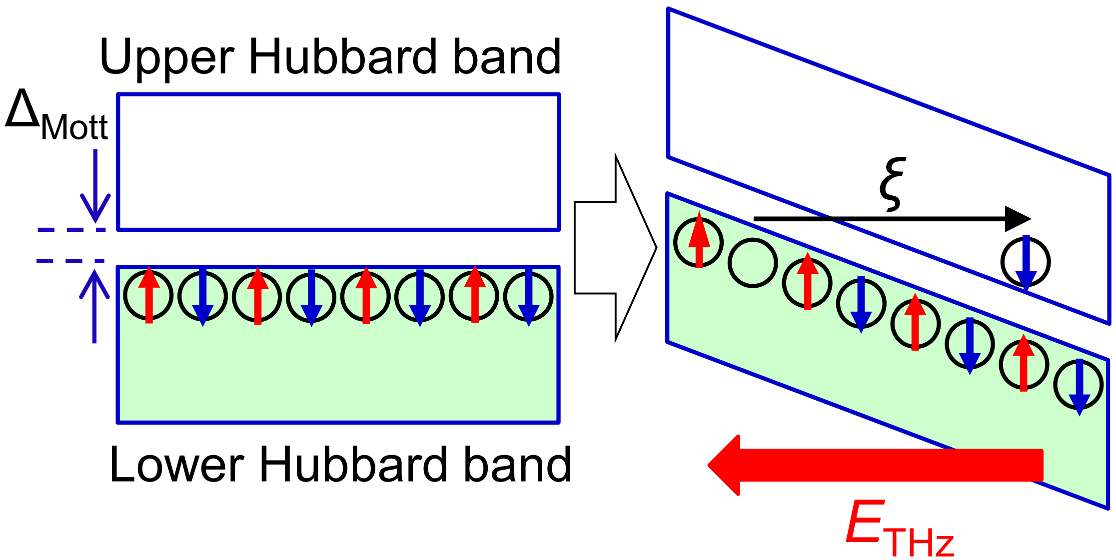
The I-M transition is driven by carrier generations via the quantum tunneling mechanism.

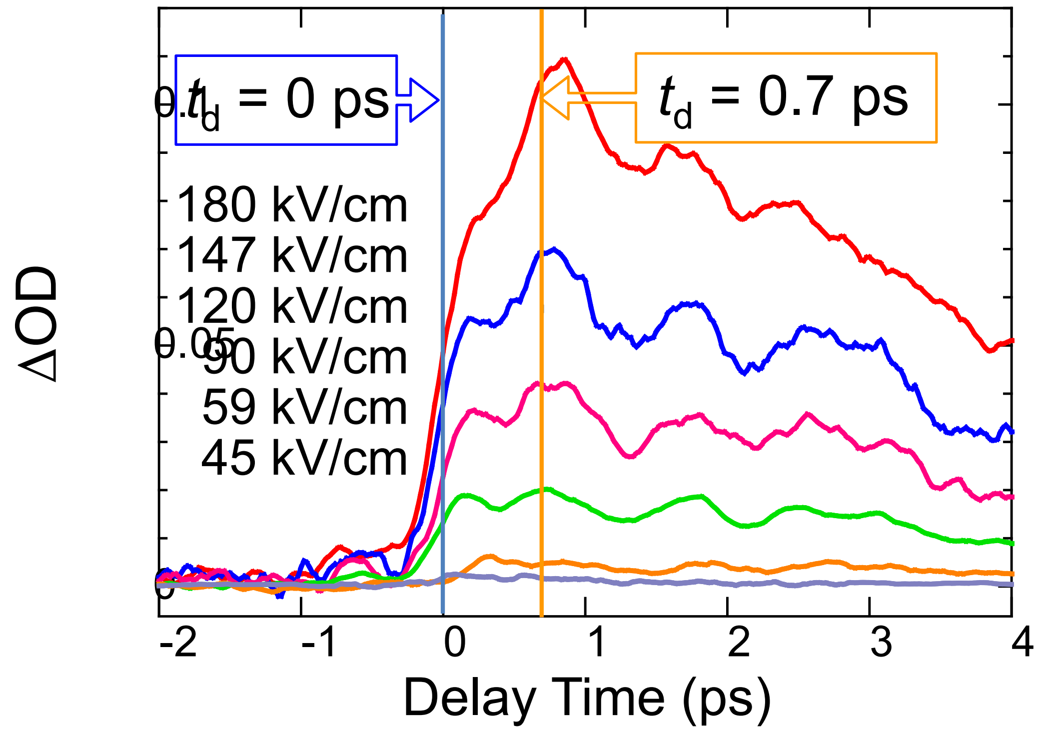


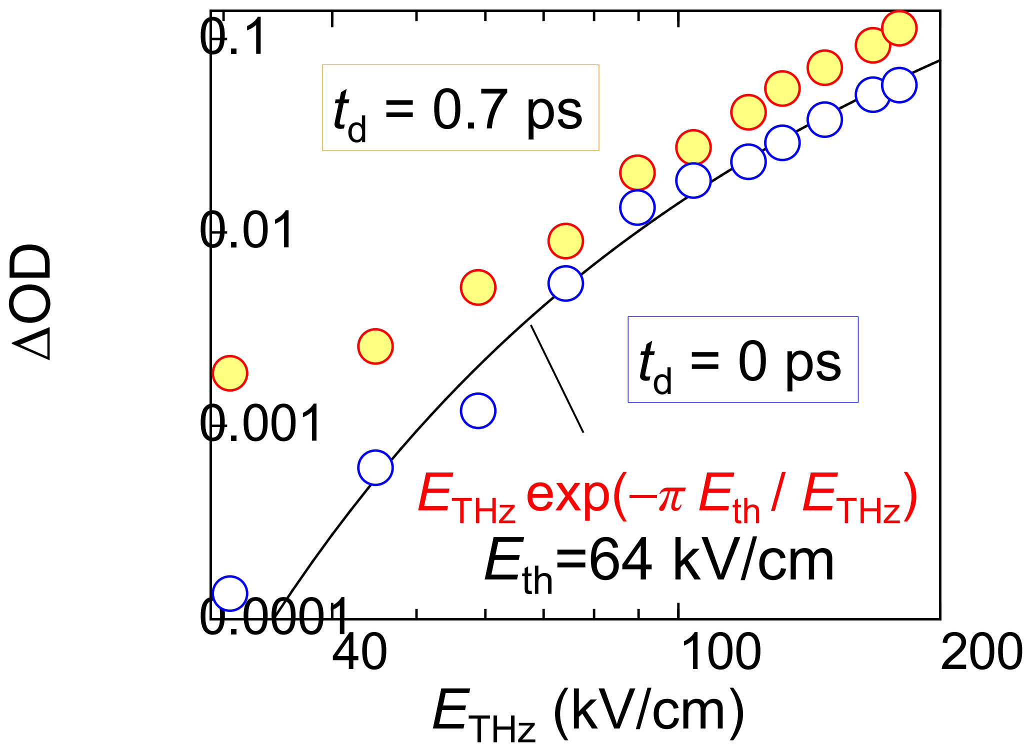
H. Yamakawa et al.,
Nature Materials ***16****, 1100 (2017)*

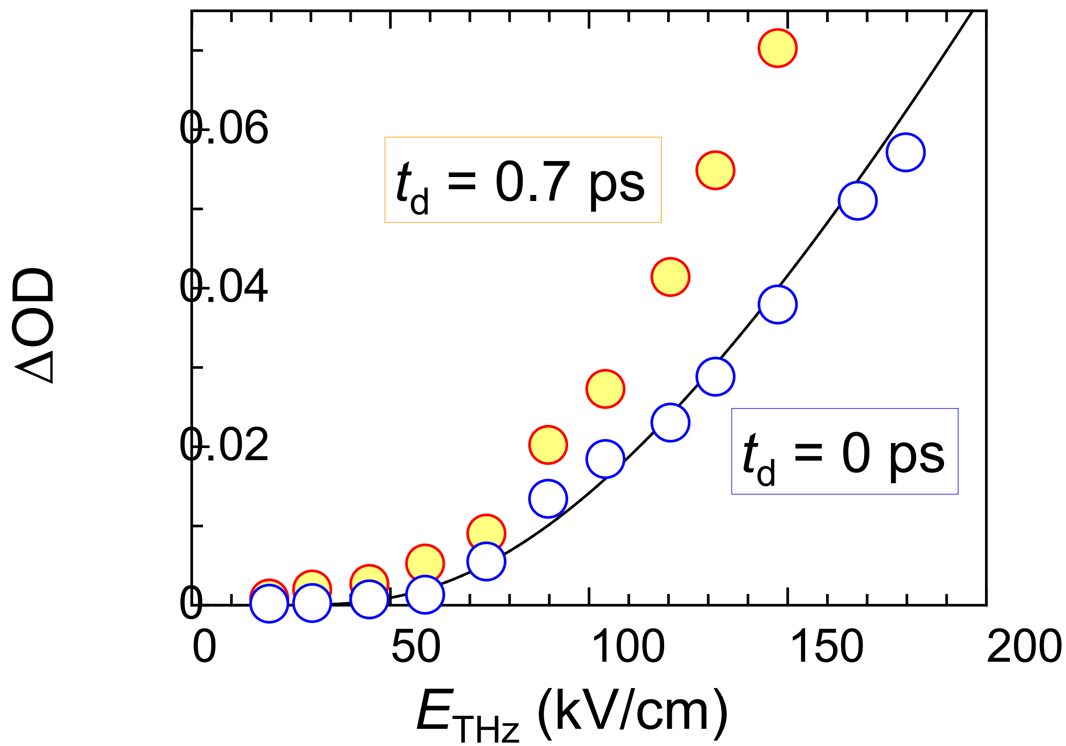
Dynamical property of THz electric-field-induced Mott transition

Carrier generation via quantum tunneling



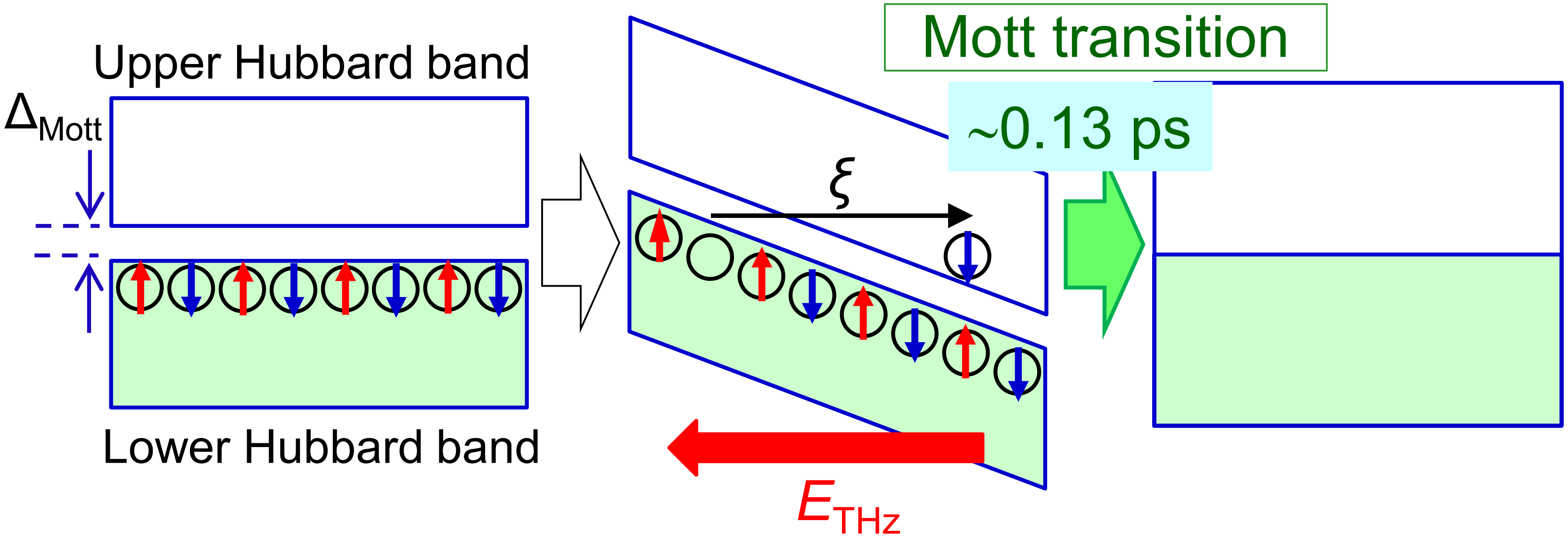




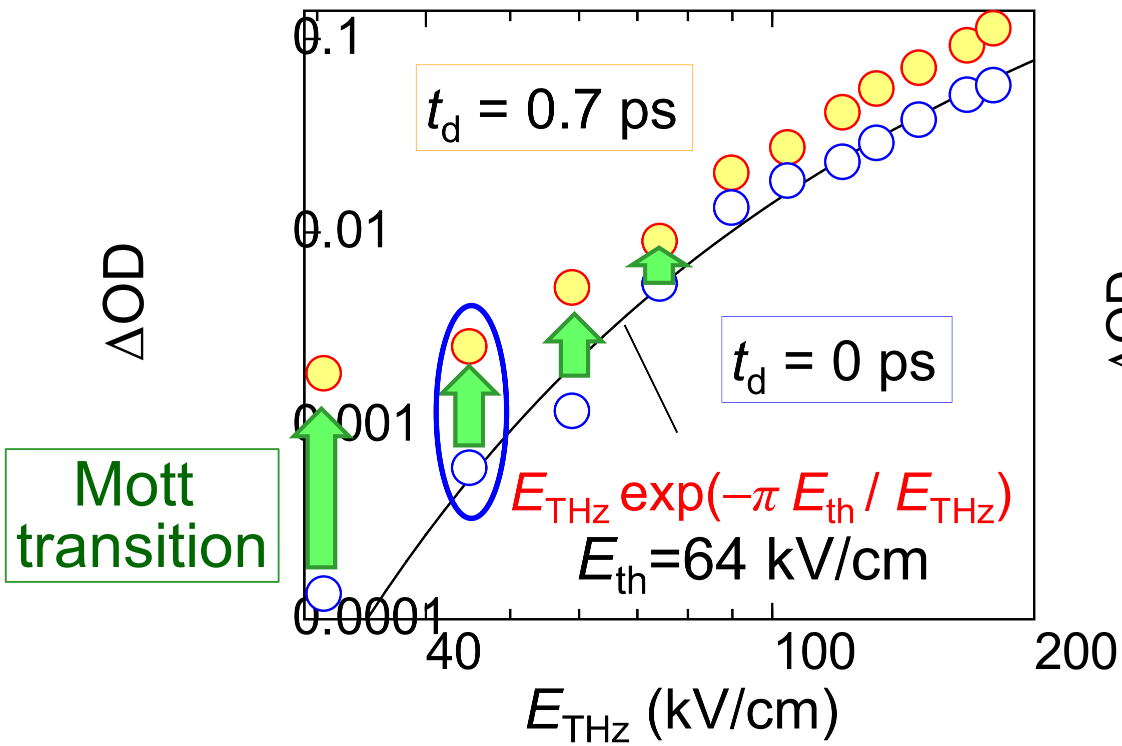


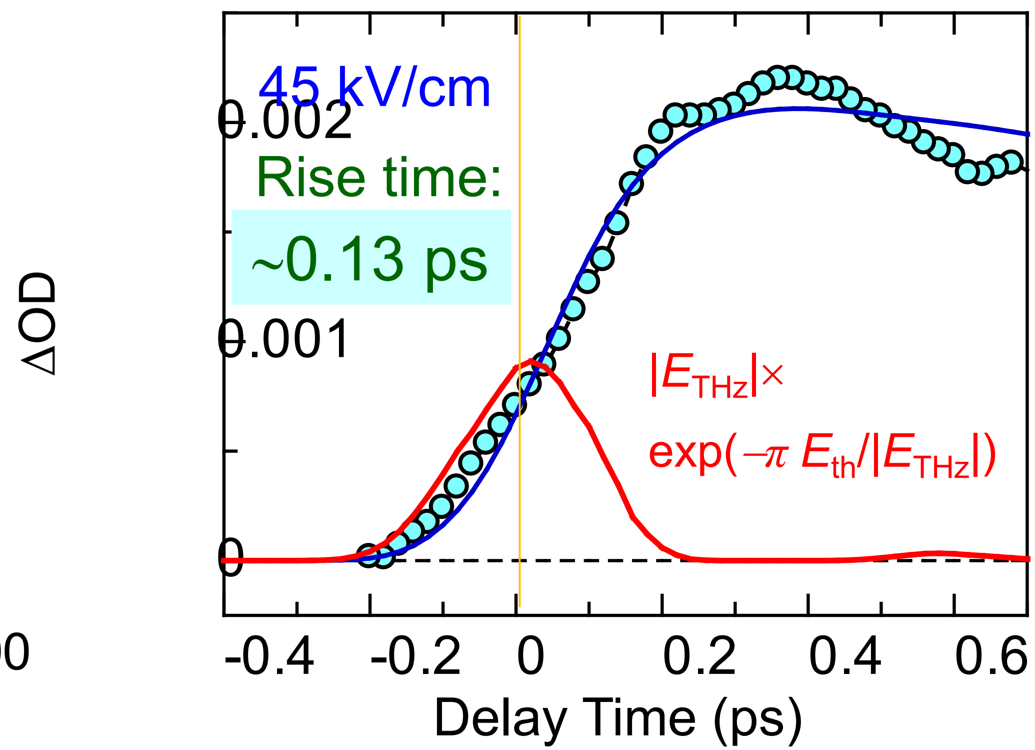
Dynamical property of THz electric-field-induced Mott transition

Carrier generation via quantum tunneling



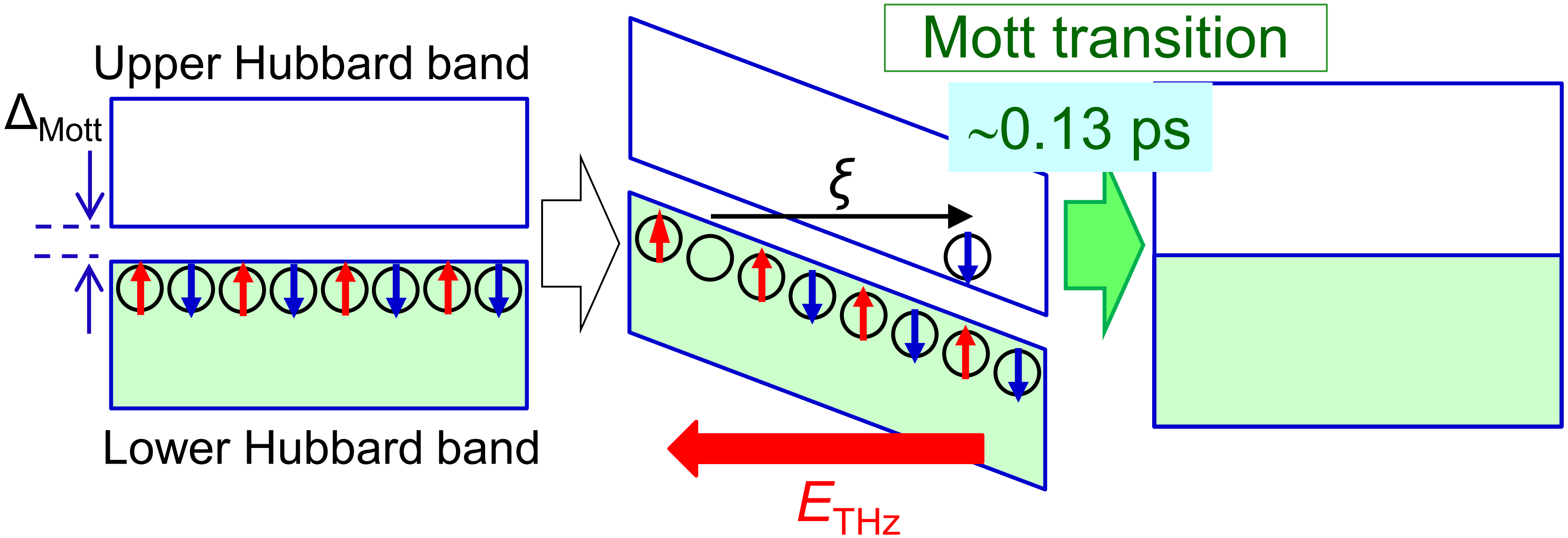
Rise time ~0.13 ps is the characteristic time of the Mott transition.



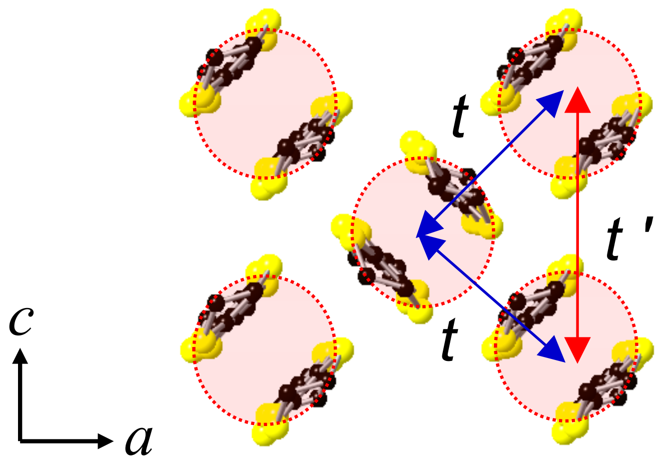


Dynamical property of THz electric-field-induced Mott transition

Carrier generation via quantum tunneling

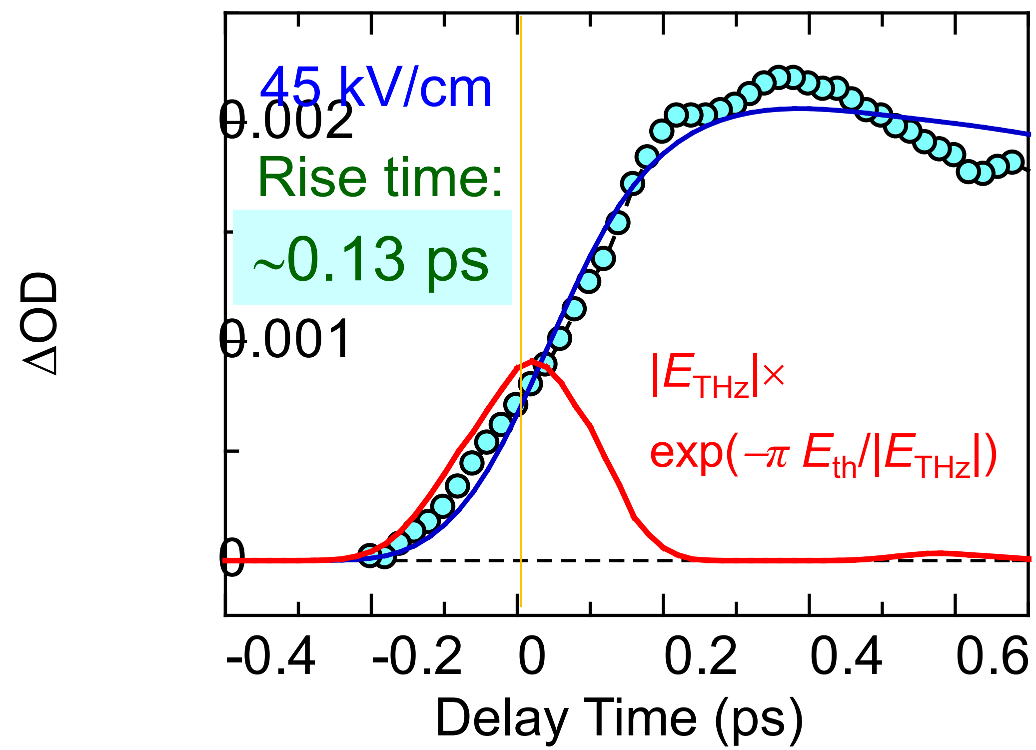


Rise time ~0.13 ps is the characteristic time of the Mott transition.



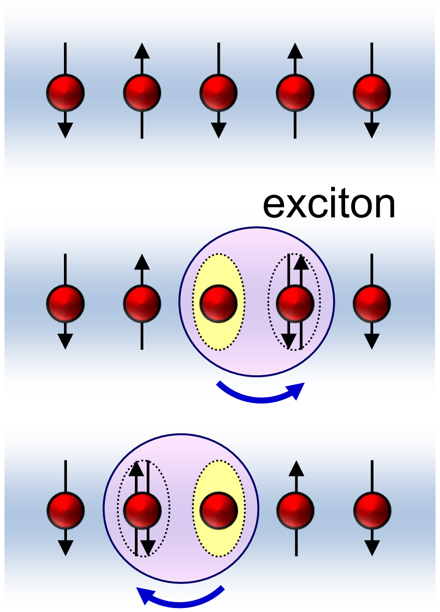
t = 78 meV ⇔ 0.051 ps

t' = 33 meV ⇔ 0.12 ps



The values of parameters
*K. Sedlmeier et al., PRB **86**, 245103 (2012).*

Large optical nonlinearity in 1D Mott insulators by THz electric fields

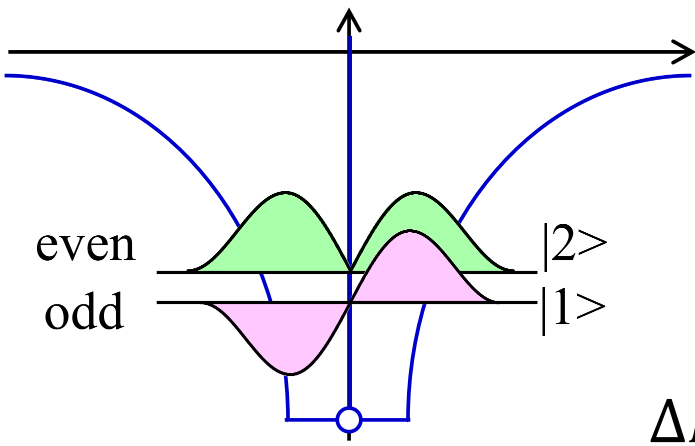


even

$$|2>=\frac{1}{\sqrt{2}}(|L>+|R>)$$

odd

$$|1>=\frac{1}{\sqrt{2}}(|L>-|R>)$$



H. Kishida et al., Nature ***405****, 929 (2000)*

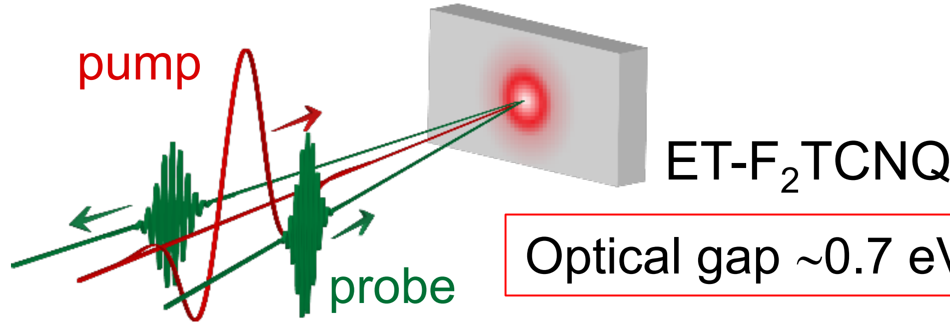
Large <1|x|2>

$$\Delta R/R \propto \chi^{(3)}(E(0))^2$$

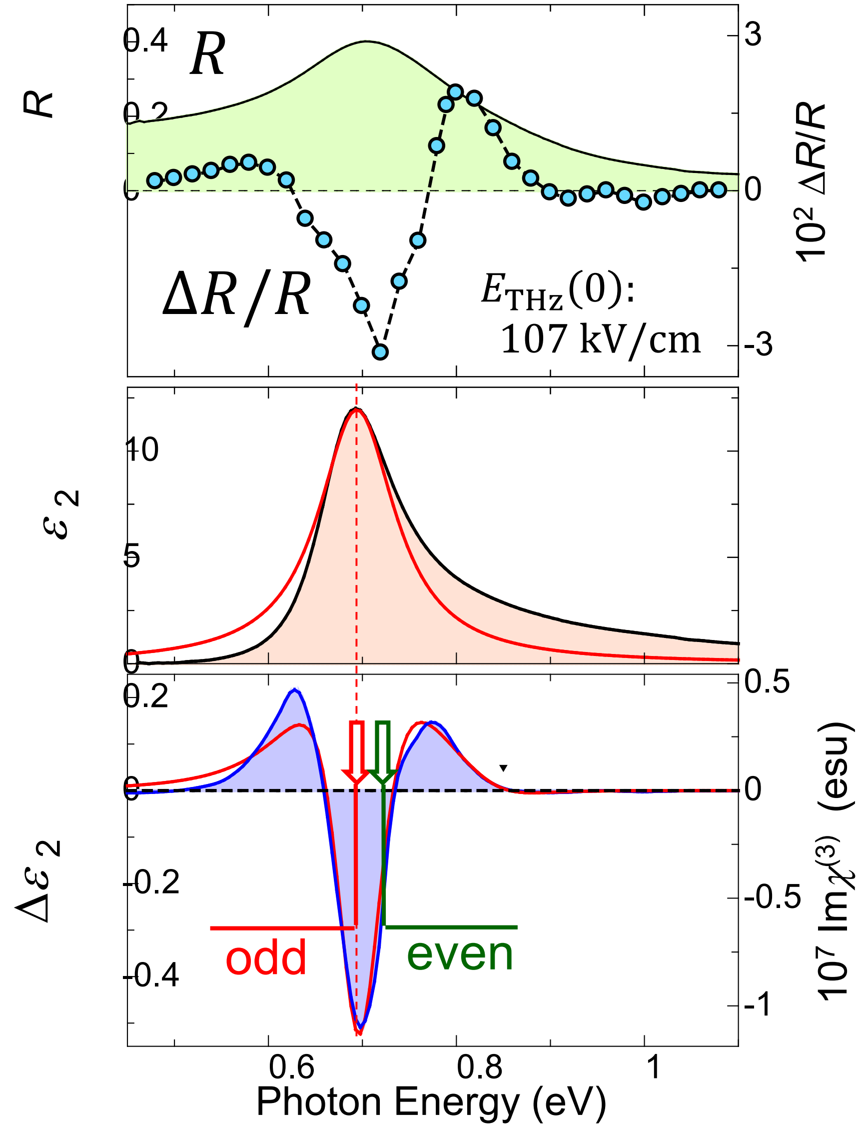
$$\chi^{(3)}(-\omega;0,0,\omega) \propto |<1|x|2>|^2 |<0|x|1>|^2$$

Large 3rd-order nonlinear optical response due to large <1|x|2> values

Theory : Mizuno et al., PRB ***62****, R4769 (2000)*

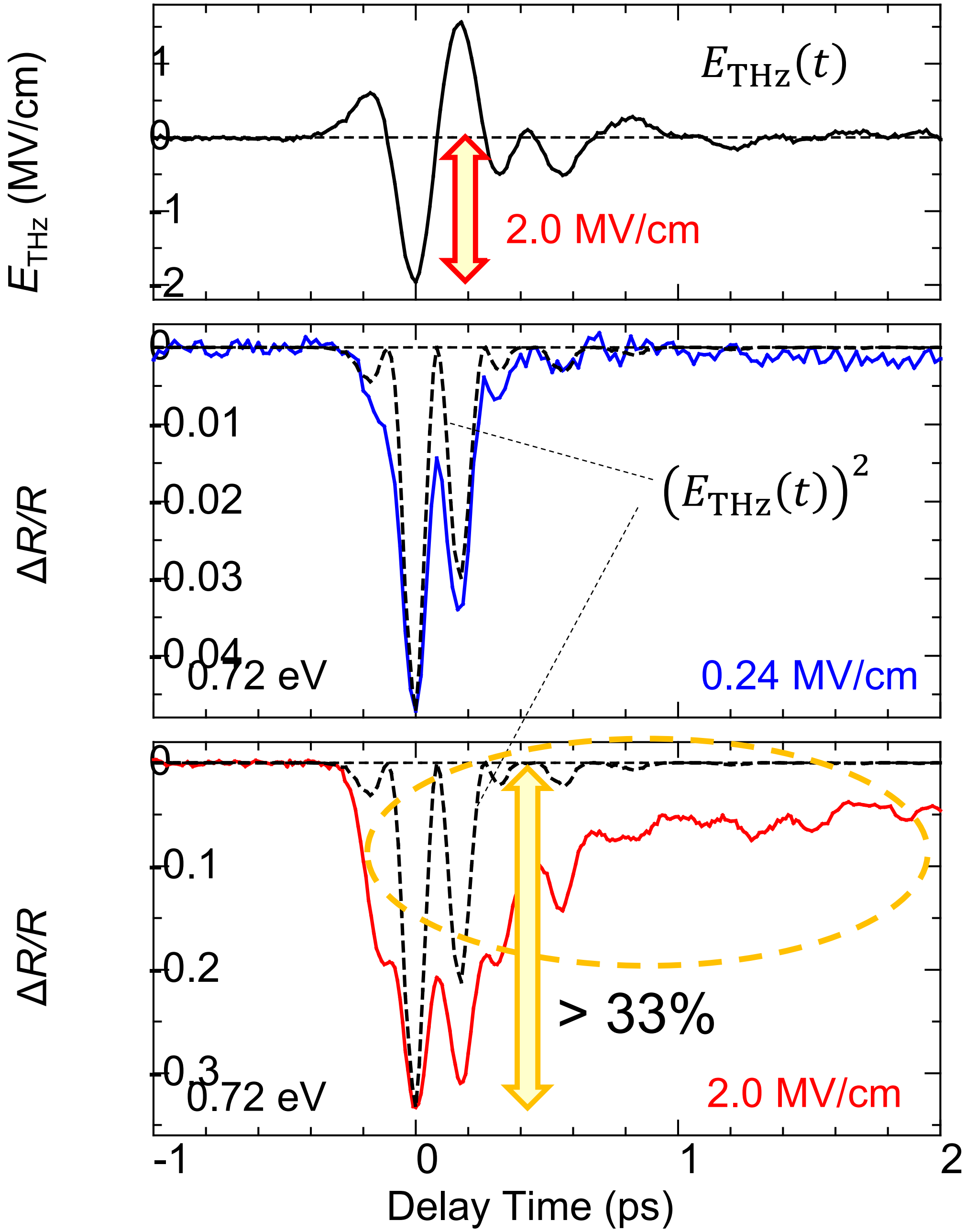


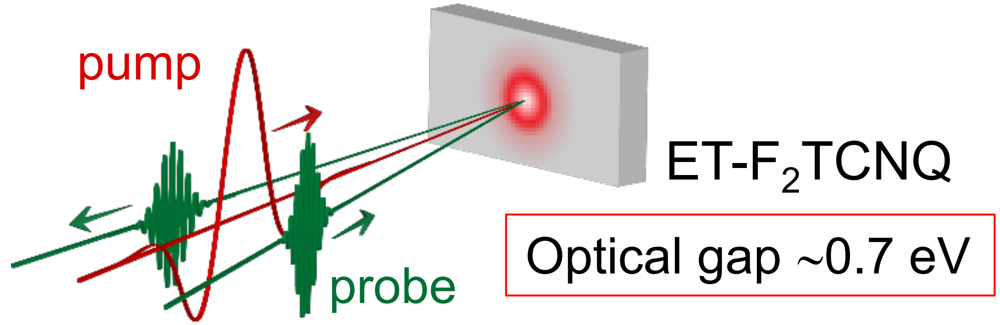
Optical gap ~0.7 eV

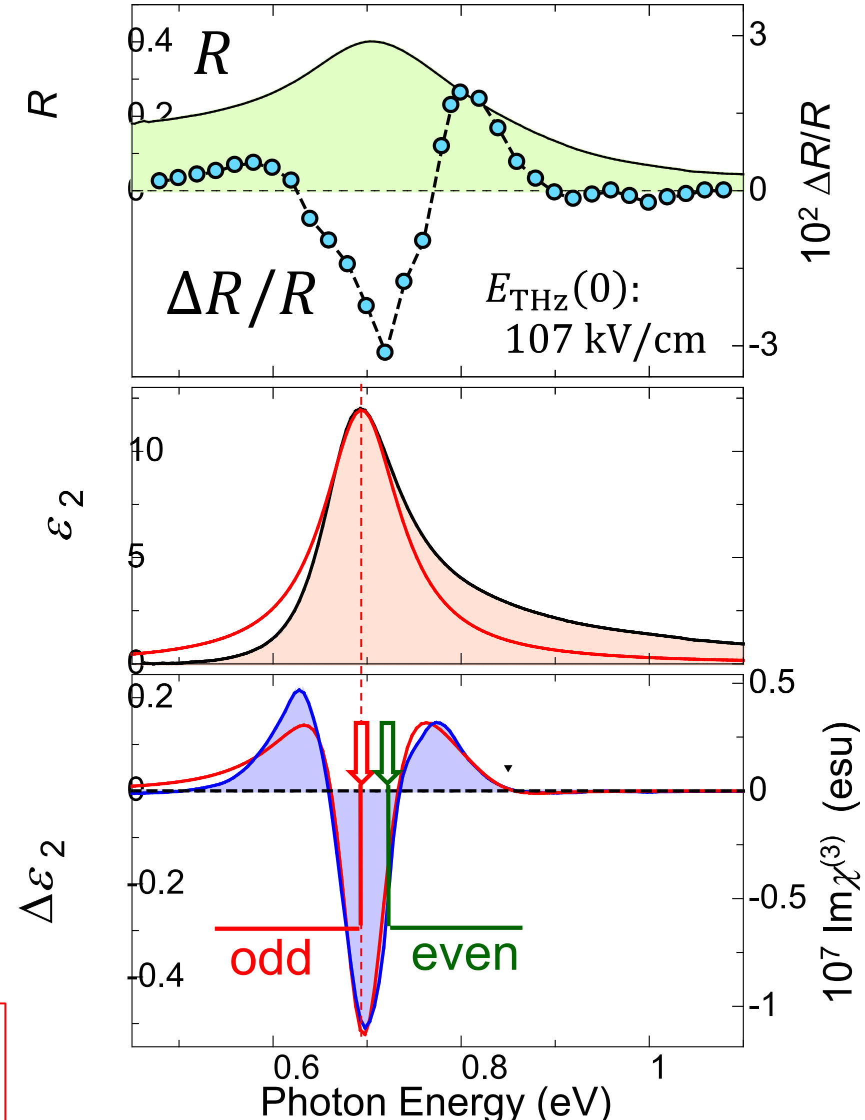


T. Miyamoto et al., to be submitted.

Large optical nonlinearity in 1D Mott insulators by THz electric fields







- ΔR/R exceeds 30%.
- Long-lived signals suggest electronic-state changes.

T. Miyamoto et al., to be submitted.

Summary 2：モット絶縁体の光/電場応答