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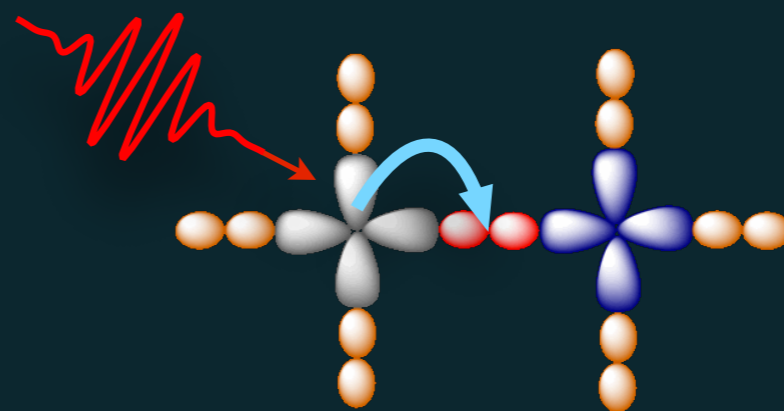
# Ultrafast optical spectroscopy of strongly correlated materials and high-temperature superconductors: a non-equilibrium approach

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ILAMP

*Interdisciplinary laboratories for advanced materials physics*  
[centridiricerca.unicatt.it/ilamp](http://centridiricerca.unicatt.it/ilamp)



# outline

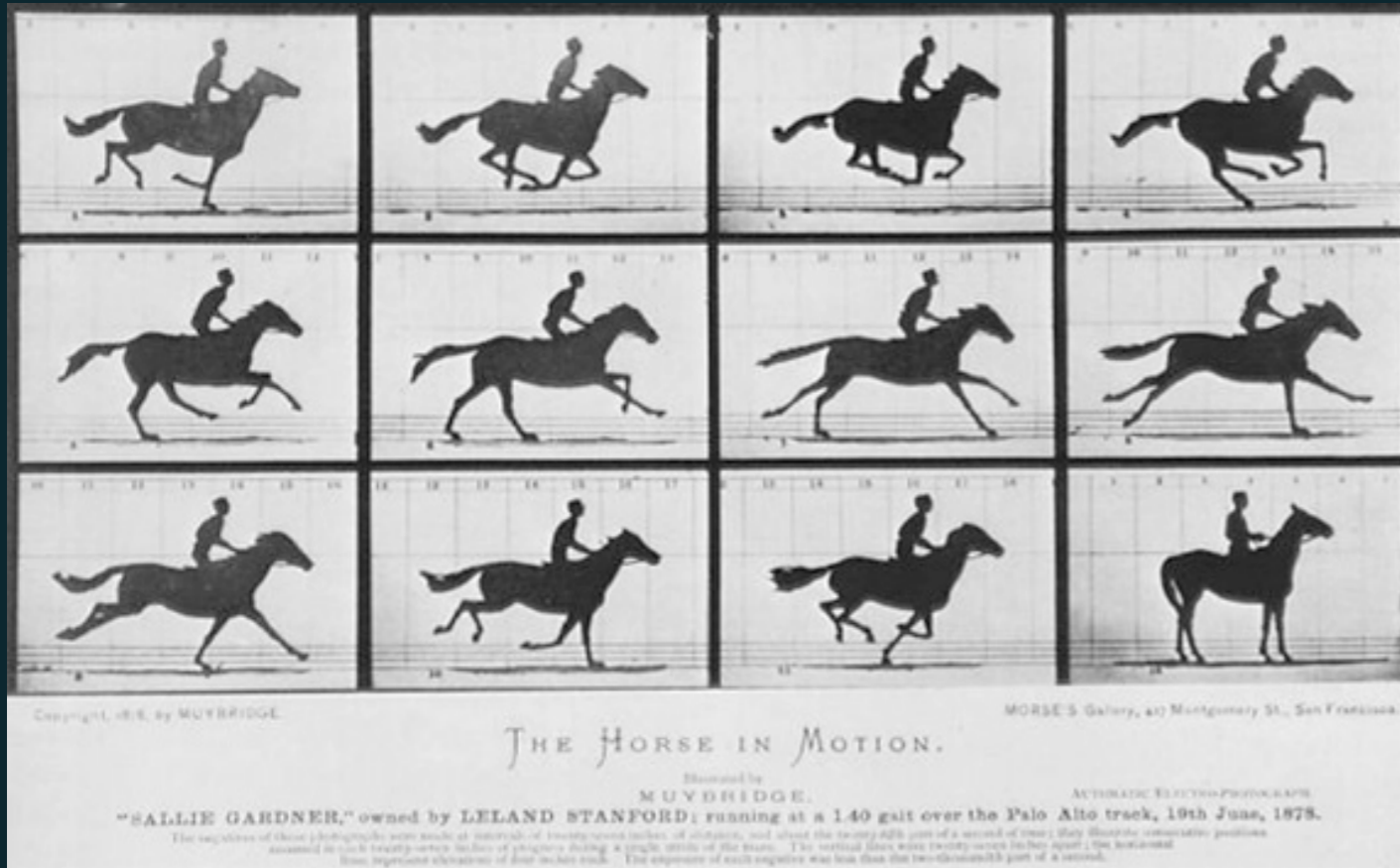
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- Ultrafast spectroscopies to investigate the electron dynamics in materials  
C. Giannetti et al. *Advances in Physics* **65**:2, 58-238 (2016)
- light pulses to UNDERSTAND equilibrium properties  
⇒ pairing glue in high- $T_c$  superconductors
- light pulses to MANIPULATE materials properties  
⇒ hidden states, transient photo-enhanced superconductivity
- light pulses to EXCITE specific degrees of freedom  
⇒ coherent lattice oscillations and Higgs modes



# time-resolved optics

High-speed photography by E. Muybridge (1872, San Francisco)



ultrafast shutter?



Nobel Lecture by A. Zewail (1999)  
(femtochemistry)

# stroboscopic pictures

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stroboscopic light



⇒ flashes of light shorter than  
the timescale of the dynamics



# stroboscopic pictures

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how fast?

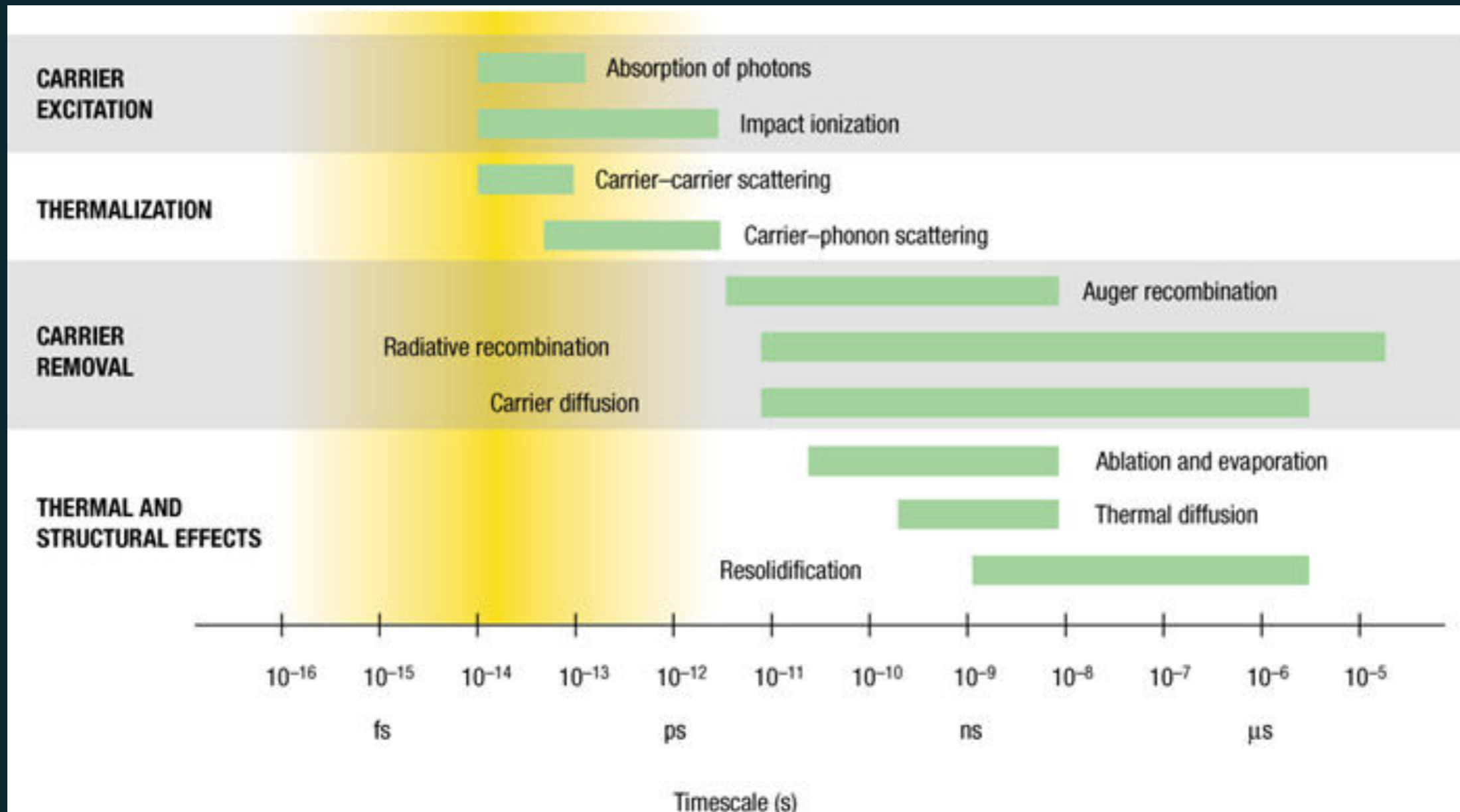


1  $\mu$ s shutter

# ultrafast dynamics in solids

Is it possible to snap the electron dynamics in real materials?

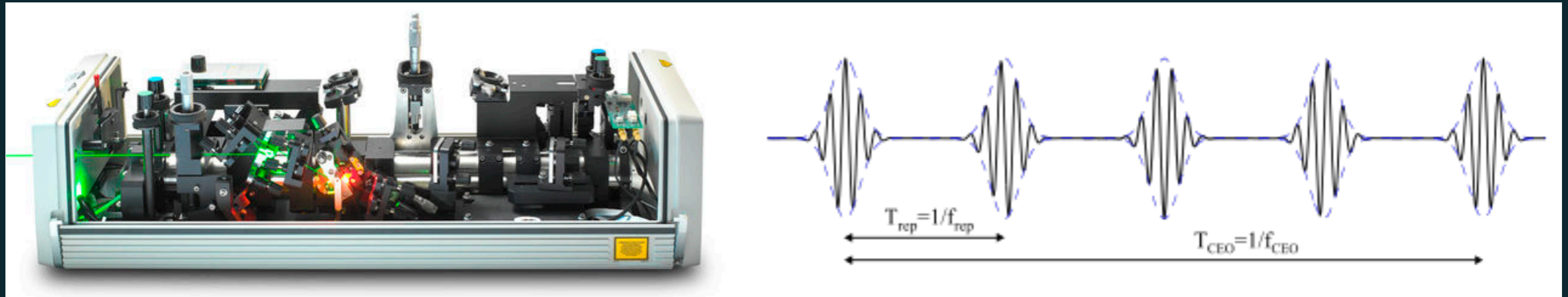
$$\hbar \approx 650 \text{ meV fs}$$



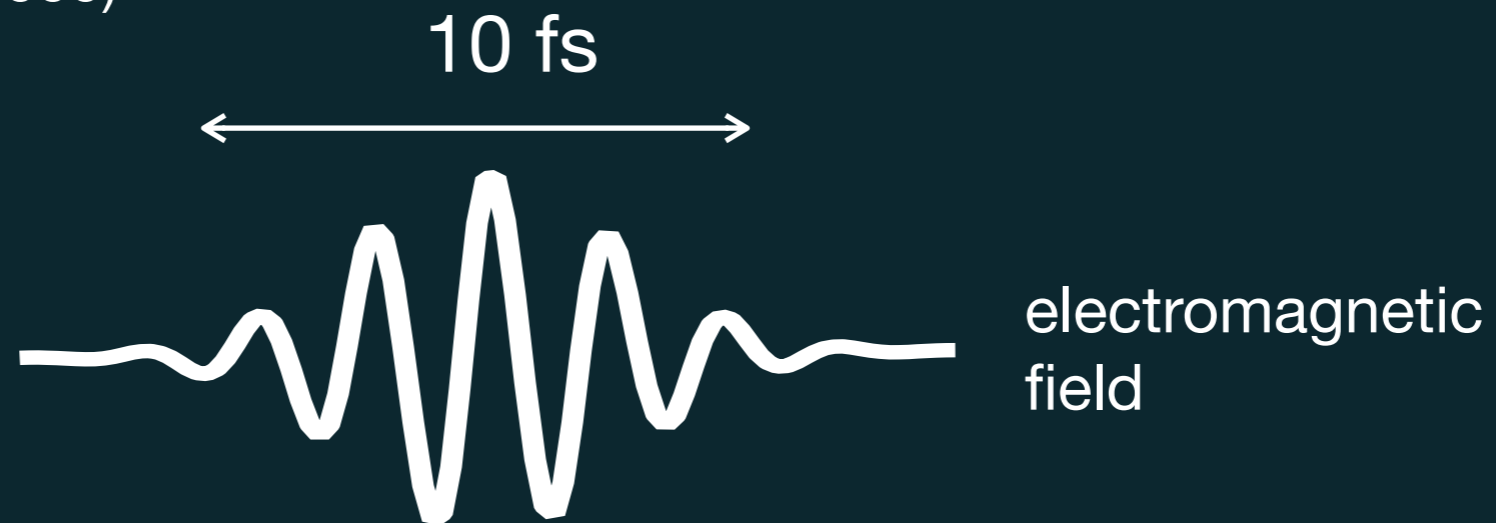


# ultrafast lasers

⇒ femtosecond flashes of lights



Nobel prize to A. Zewail (1999)  
(femtochemistry)



1 femtosecond =  $10^{-15}$  s = 0.0000000000000001 s  
optical cycle: 2 fs @ 600 nm

# femto-photography

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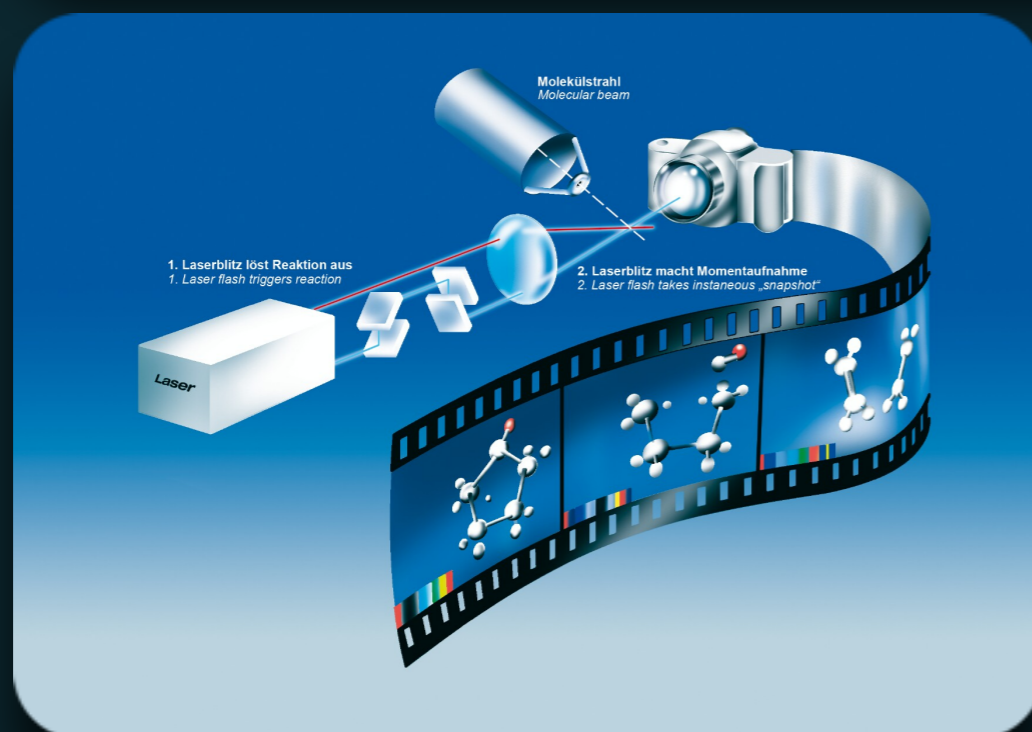
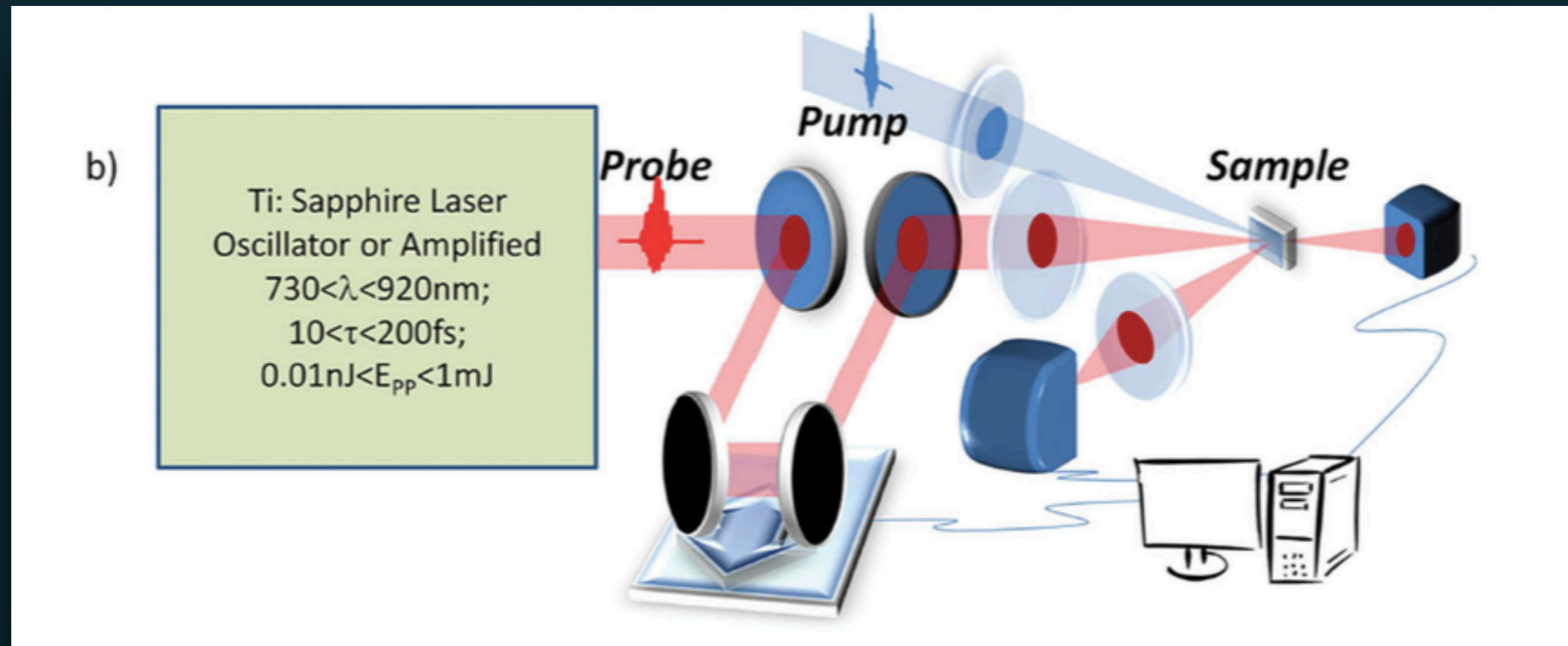


10 cm  $\Rightarrow$  100 picoseconds @ speed velocity

[femtocamera.info](http://femtocamera.info) (MIT)

# pump-probe spectroscopy in solids

C. Giannetti et al. *Advances in Physics* 65:2, 58-238 (2016)



movies of chemical reactions and  
electronic interactions in materials

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electromagnetic  
properties

bandstructure

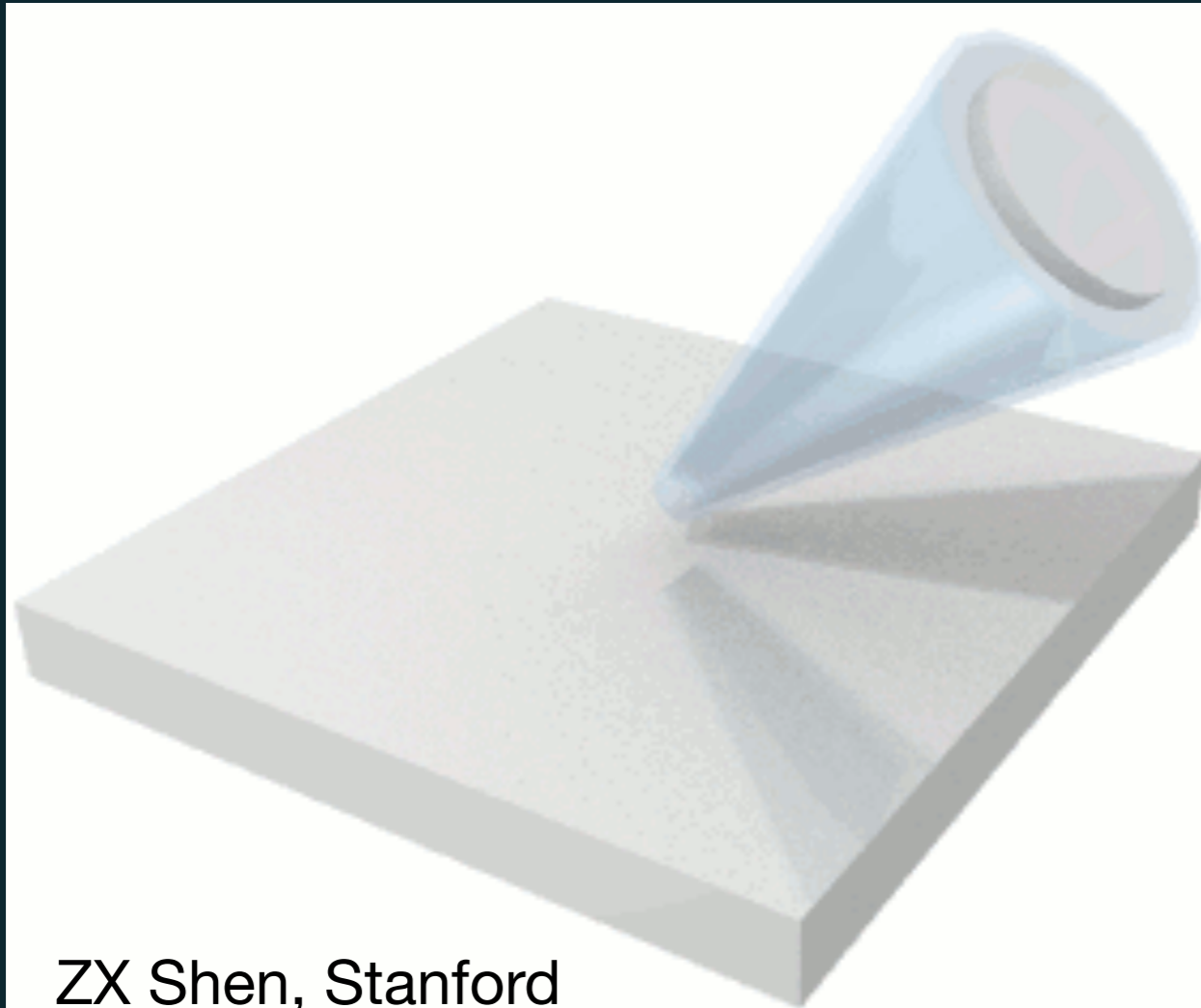
how many degrees of freedom it  
is possible to probe?

Lattice

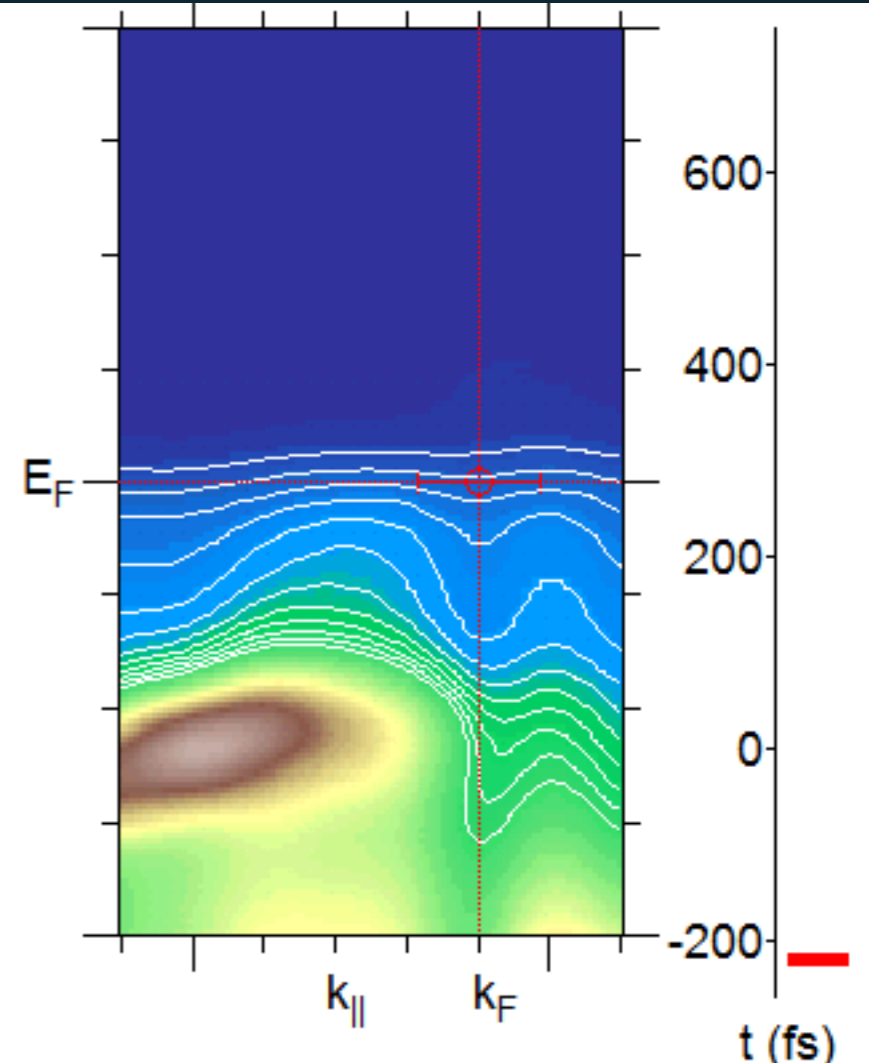
long-range  
orders



# bandstructure



ZX Shen, Stanford



Angle-Resolved-Photoemission-Electron-Spectroscopy

energy-momentum conservation

$$E_{kin} = \hbar\omega - \Phi - |E_B|$$

$$\hbar\mathbf{k}_{||} = \sqrt{2mE_{kin}} \cdot \sin\theta$$



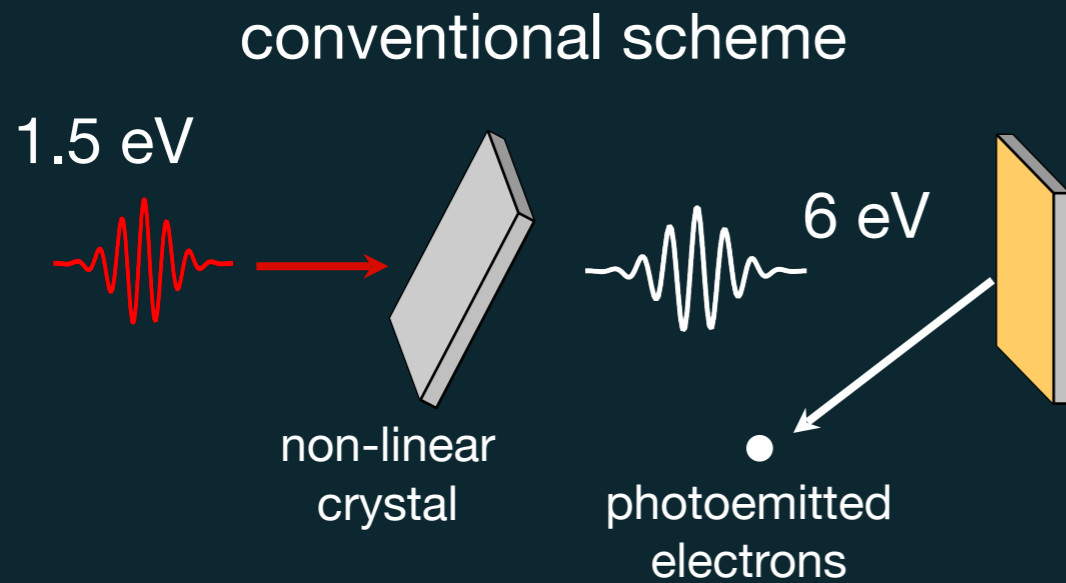
mapping band

dispersion:

$$E_B(\mathbf{k}_{||})$$

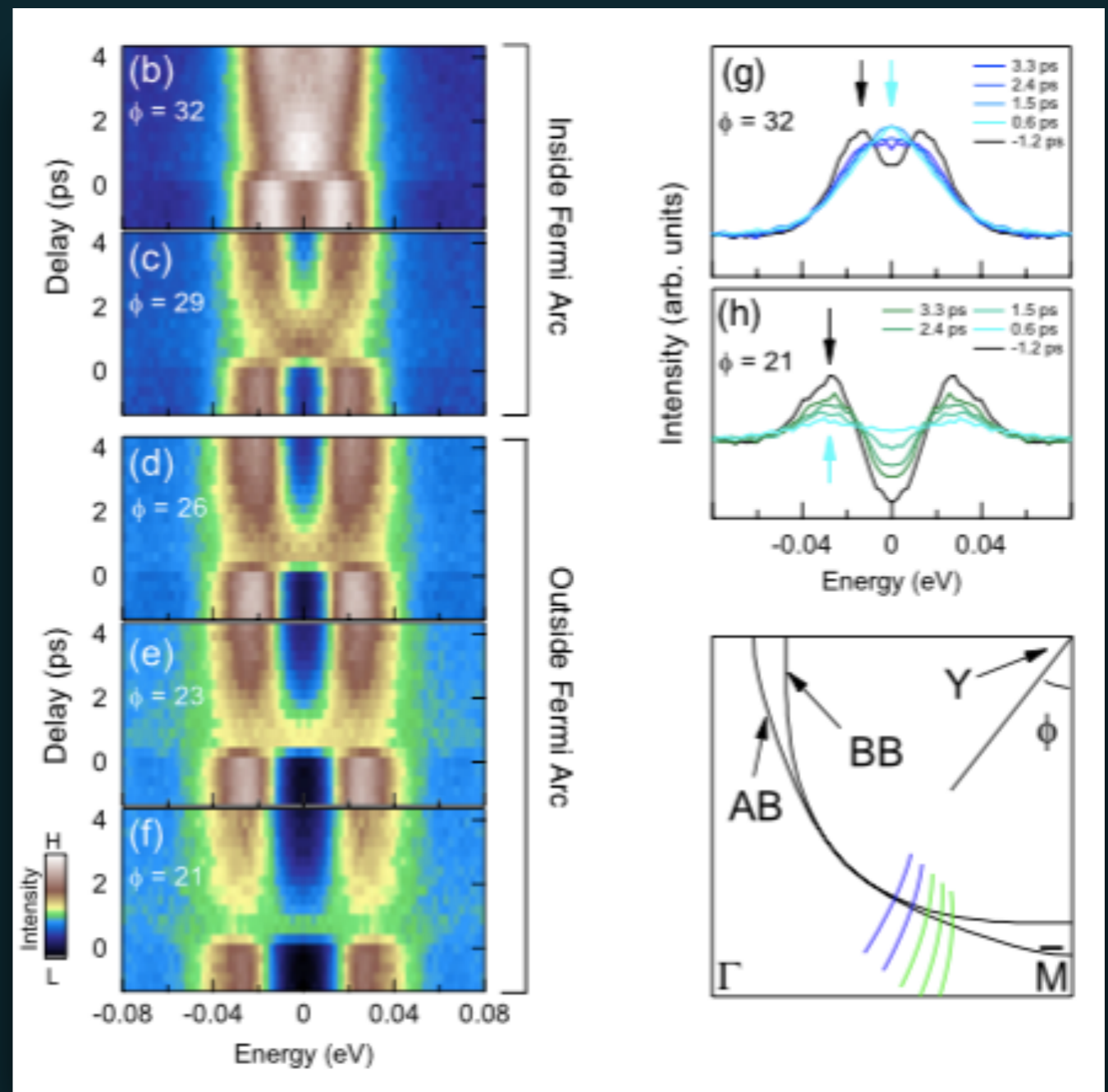
# time-resolved ARPES

## Nodal-Gap dynamics in superconducting copper oxides

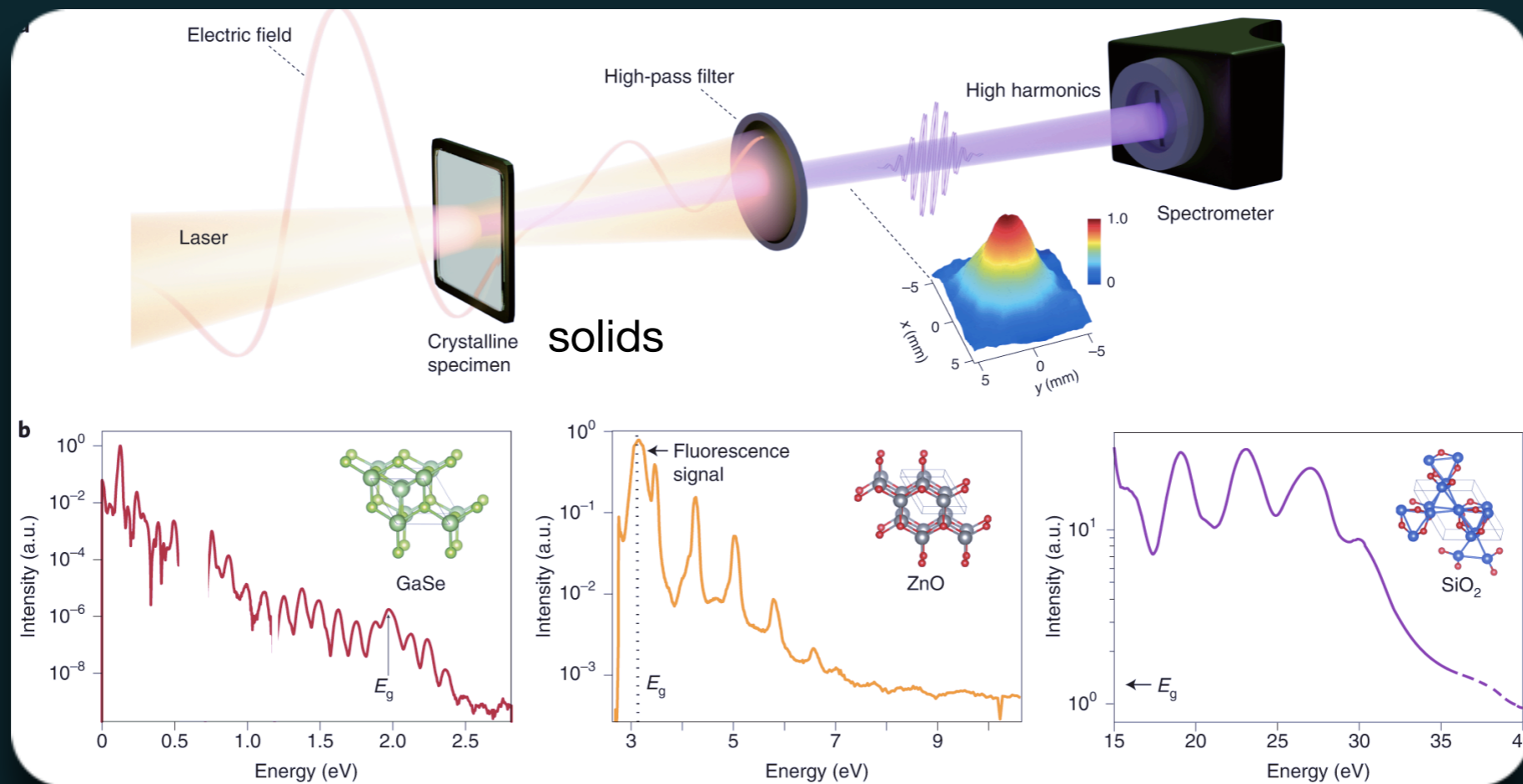
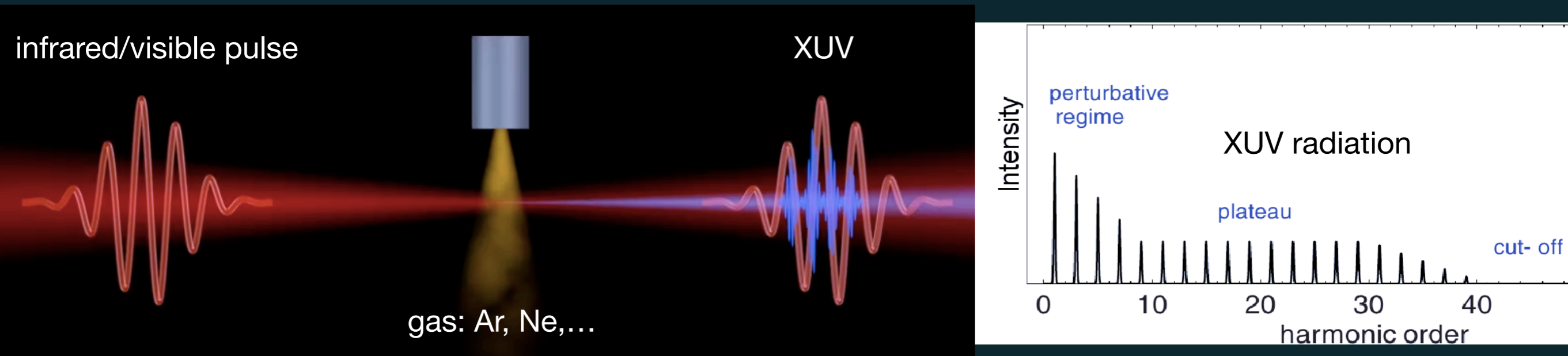


### LIMITS

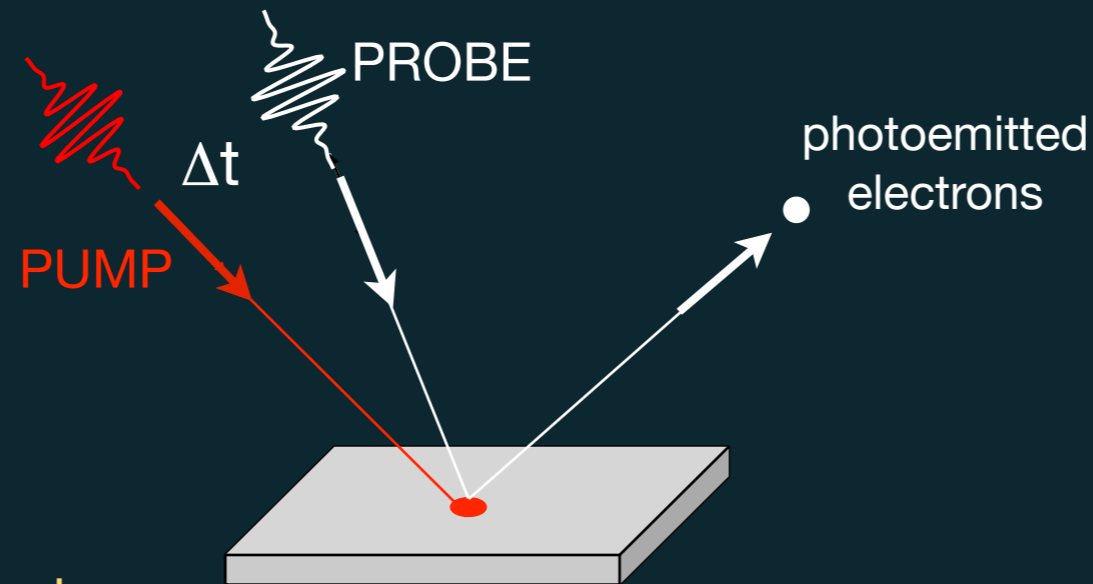
- **momentum**: no access to the antinodal region
- **energy**: no access to binding energies  $>1$  eV



# high-harmonics generation

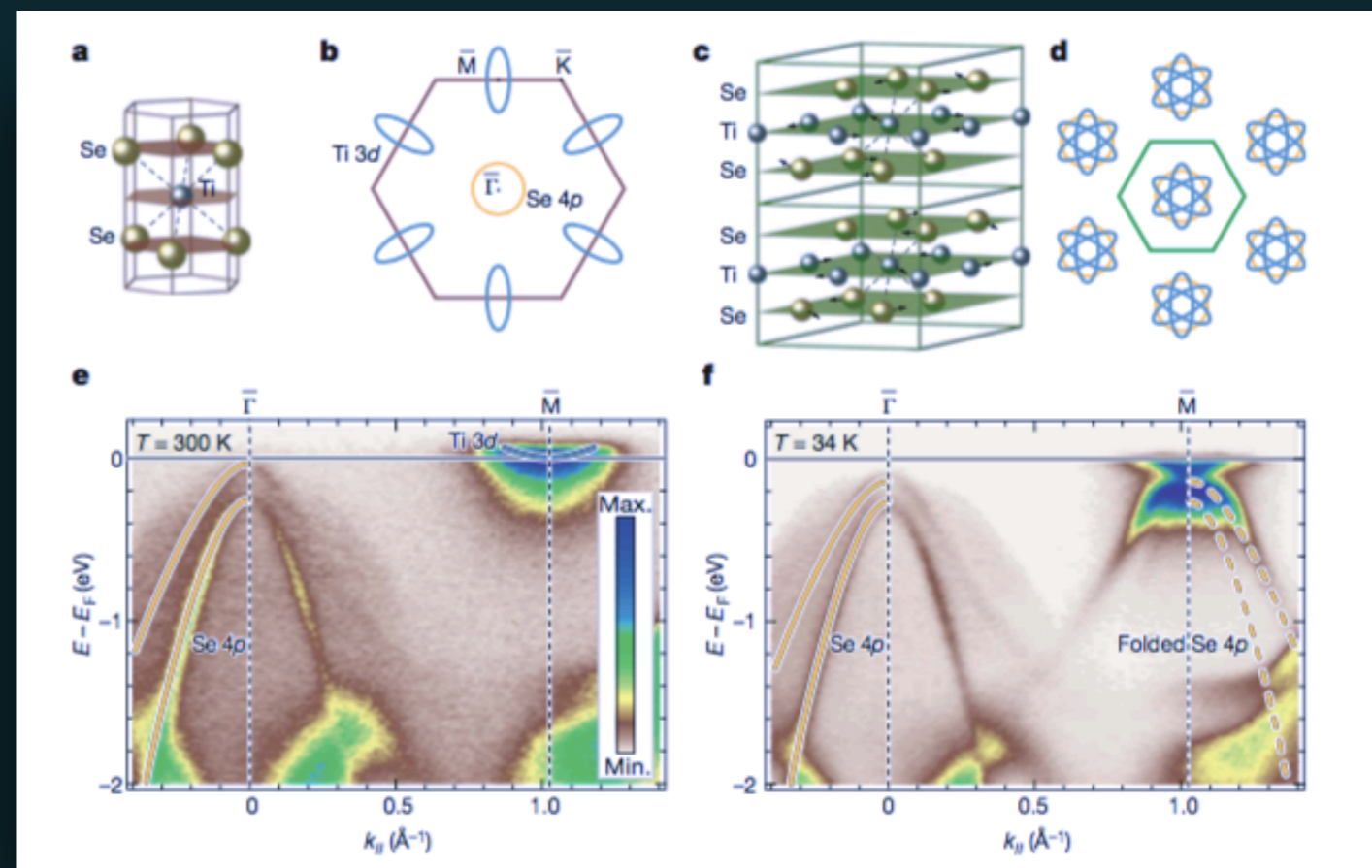
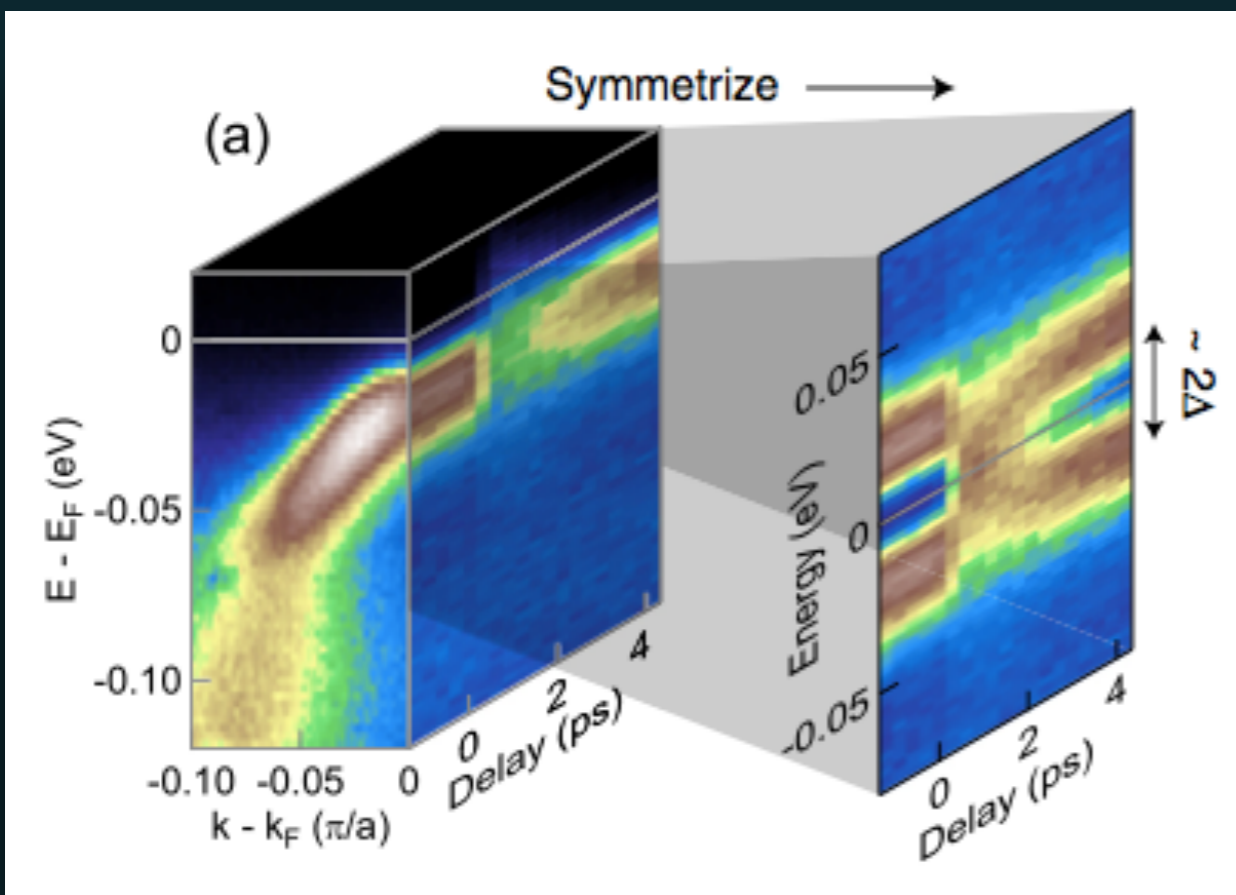


# time-resolved ARPES



Nodal-Gap dynamics in superconducting copper oxides  
UV probe

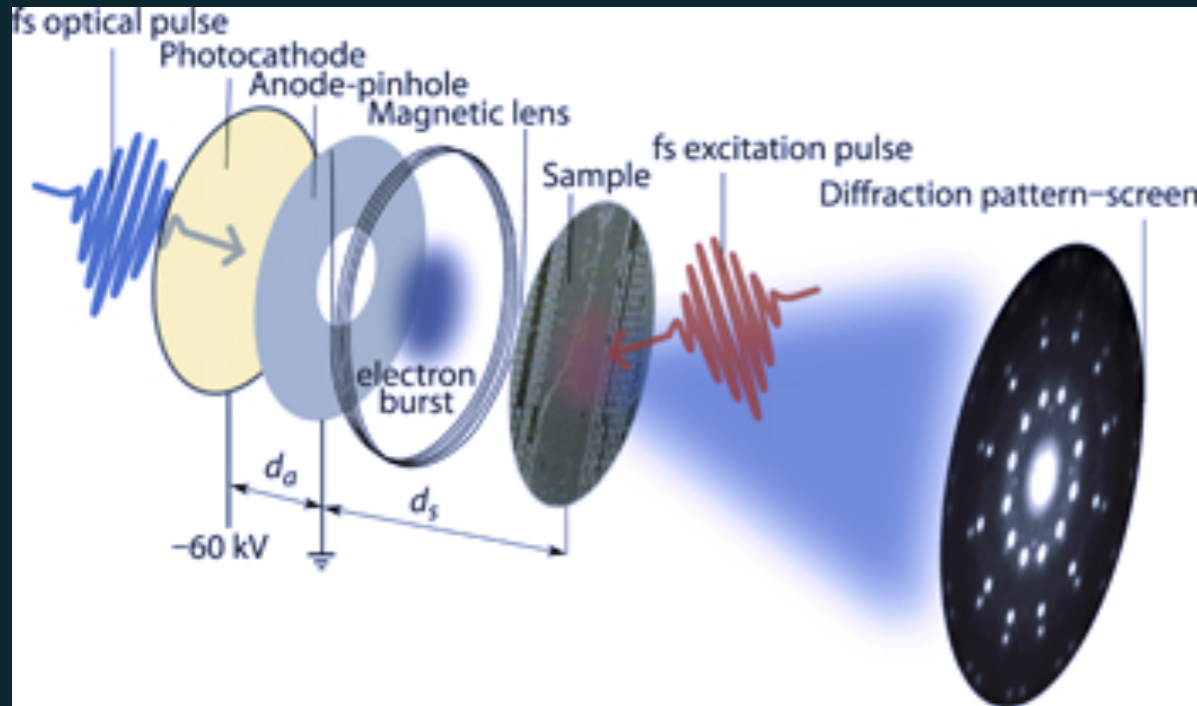
Collapse of long-range charge order in 1T-TiSe<sub>2</sub>  
X-ray probe





# time-resolved electron diffraction

## Lattice



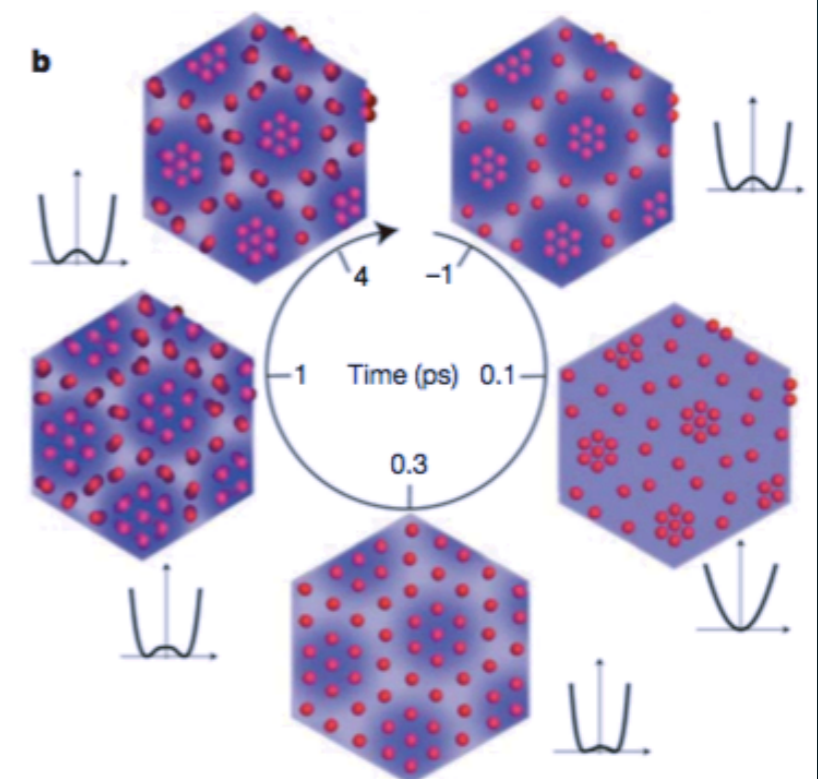
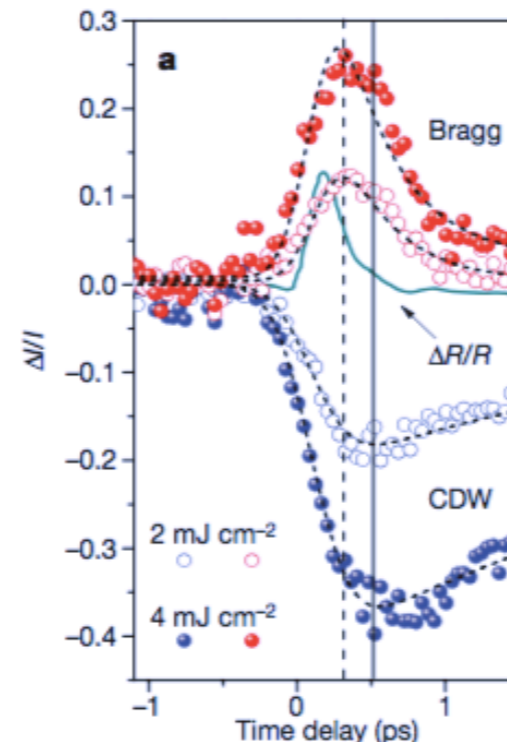
100 fs bunch of electrons

dynamics of the optical melting of  
a charge density wave in TaS<sub>2</sub>

A.J. McCulloch et al., *J. Phys. B: At. Mol. Opt. Phys.* **49**, 164004 (2016)

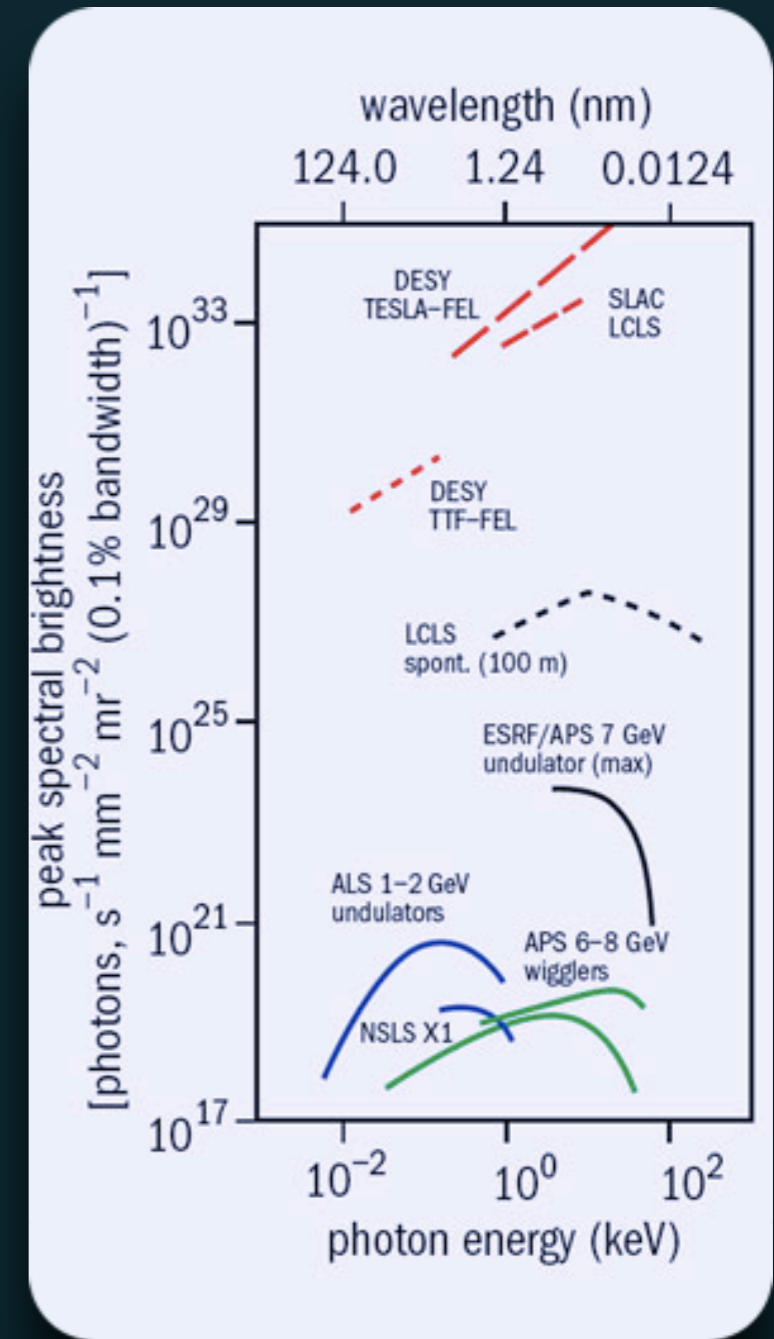
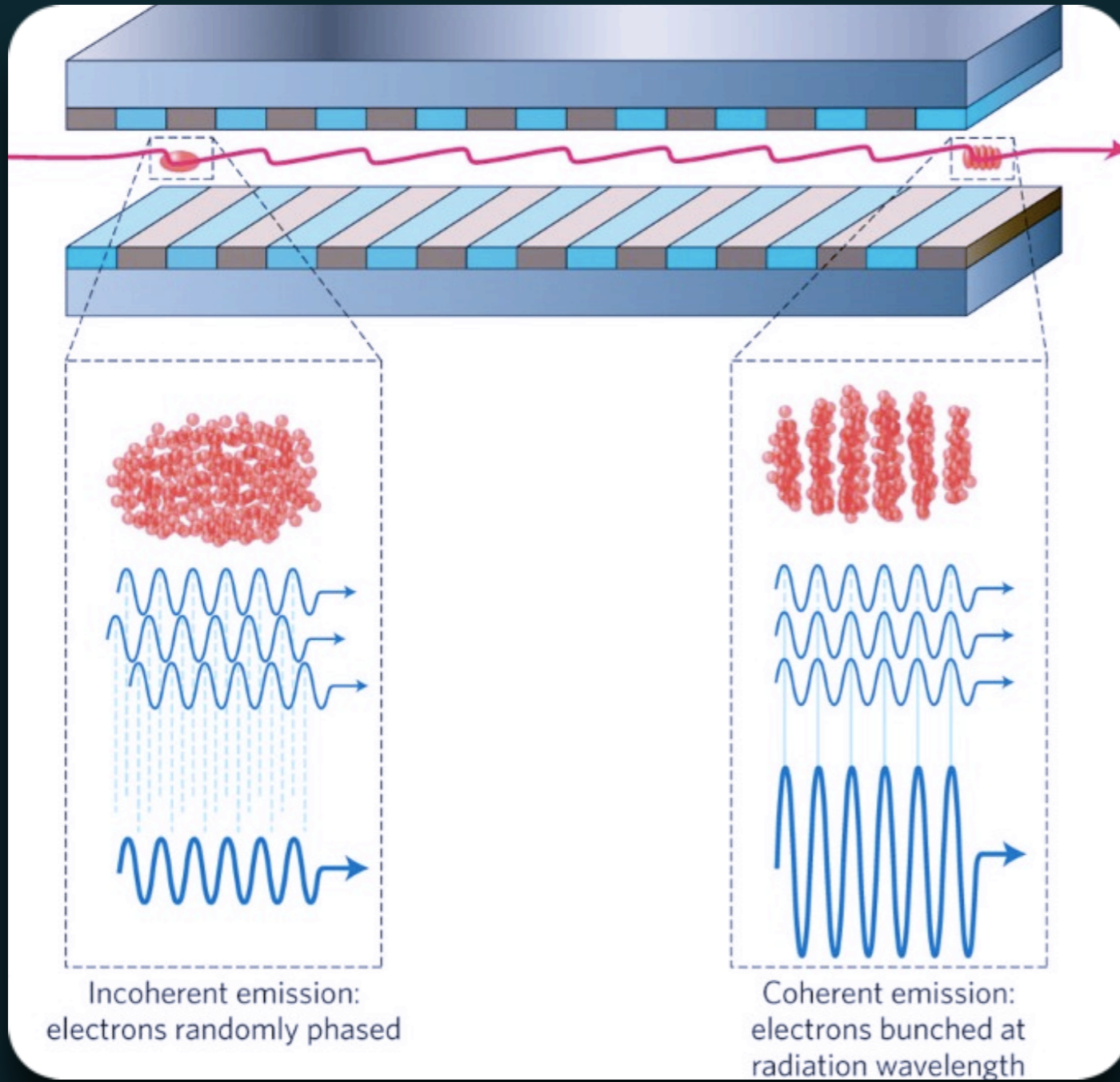
### PHYSICS:

- photo-induced structural phase transitions
- dynamics of the lattice



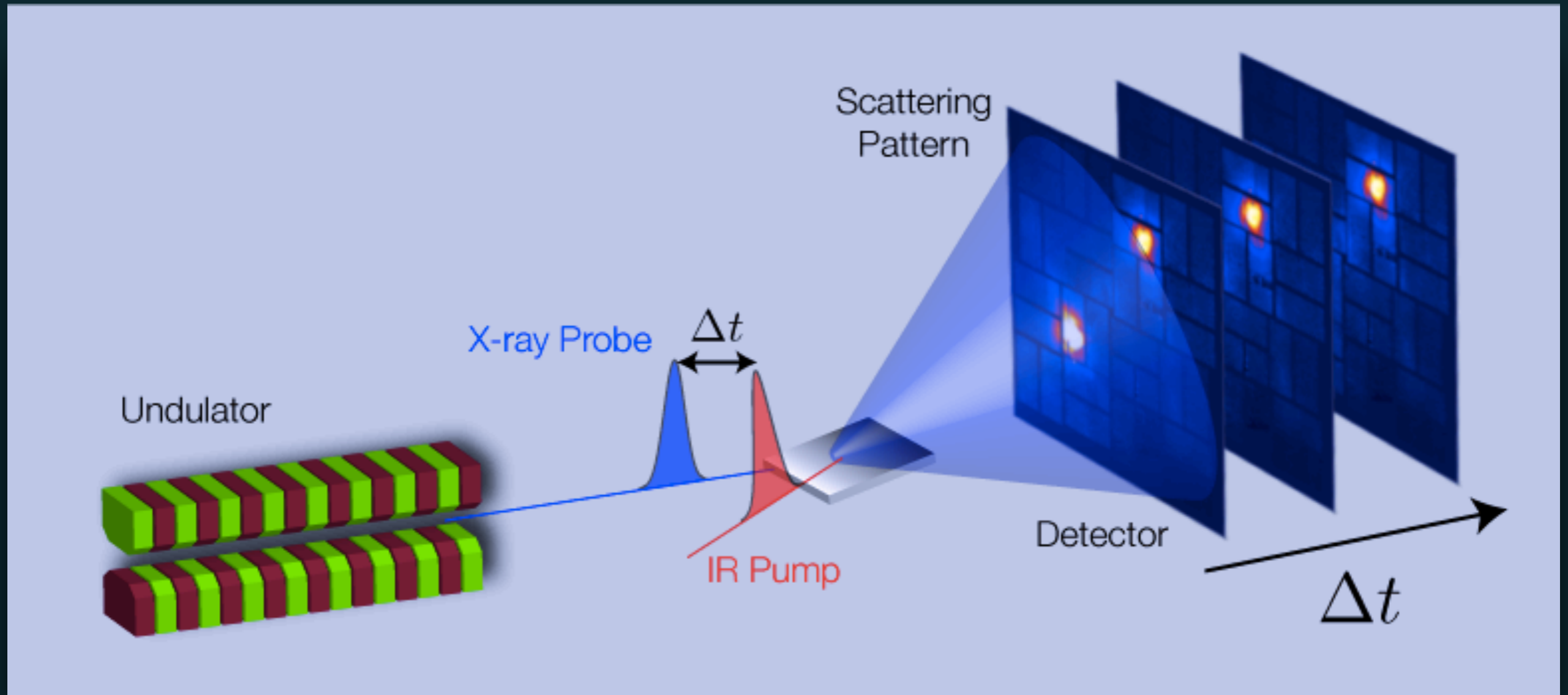
M. Eichberger et al.,  
*Nature* **468**, 799 (2010)

# Free Electron Lasers



time-resolved X-ray diffraction

# Lattice Long-range orders

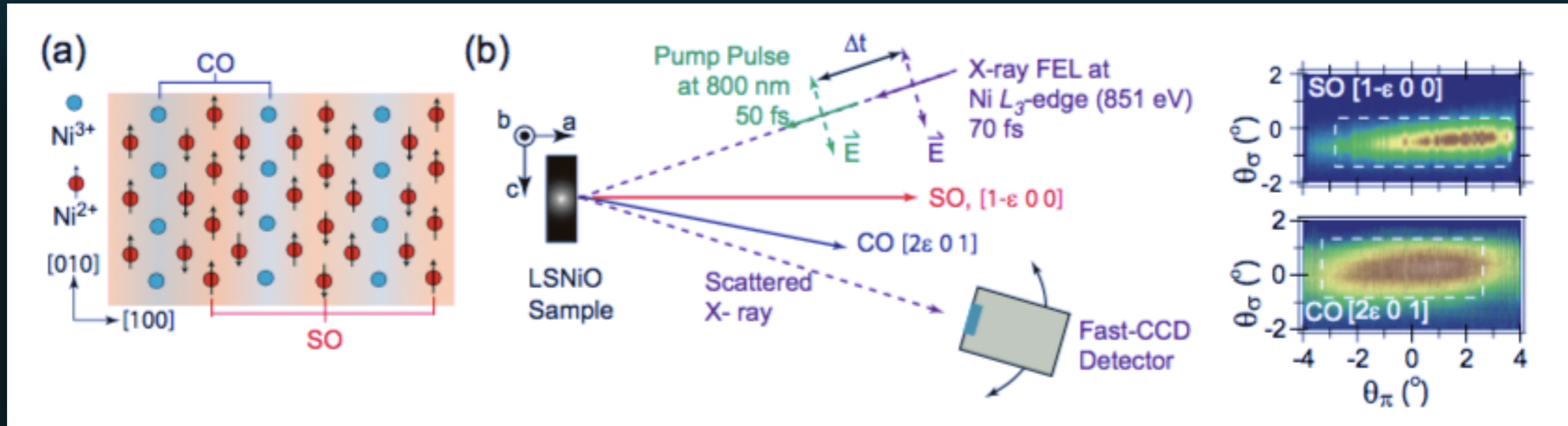


X-ray FEL pulses

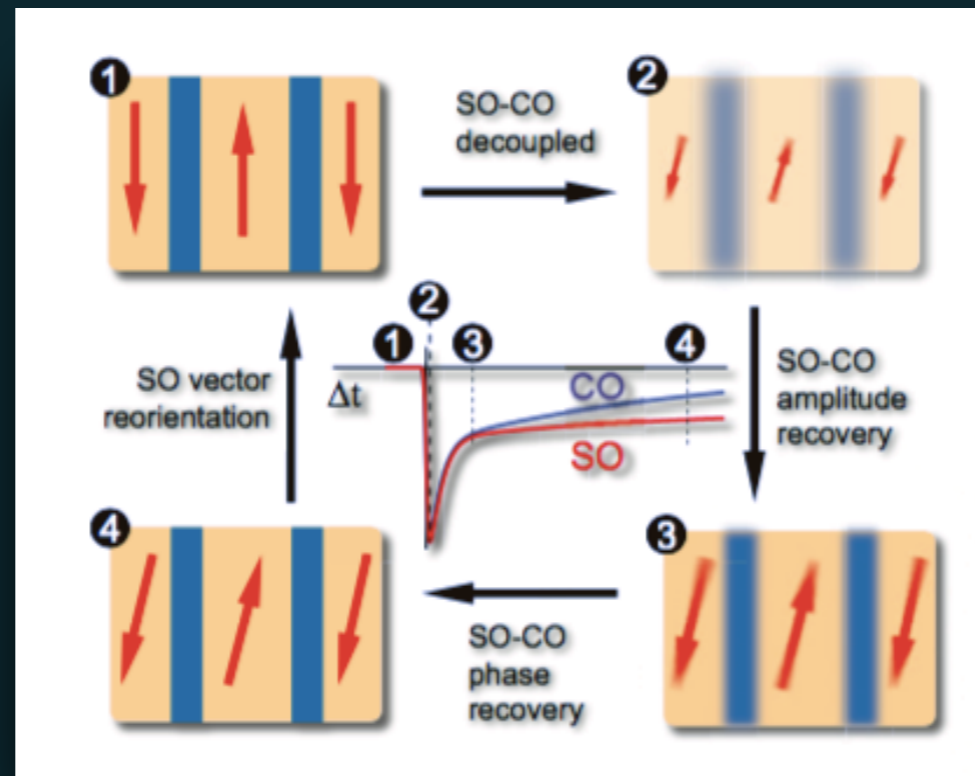


time-resolved X-ray diffraction

# Long-range orders



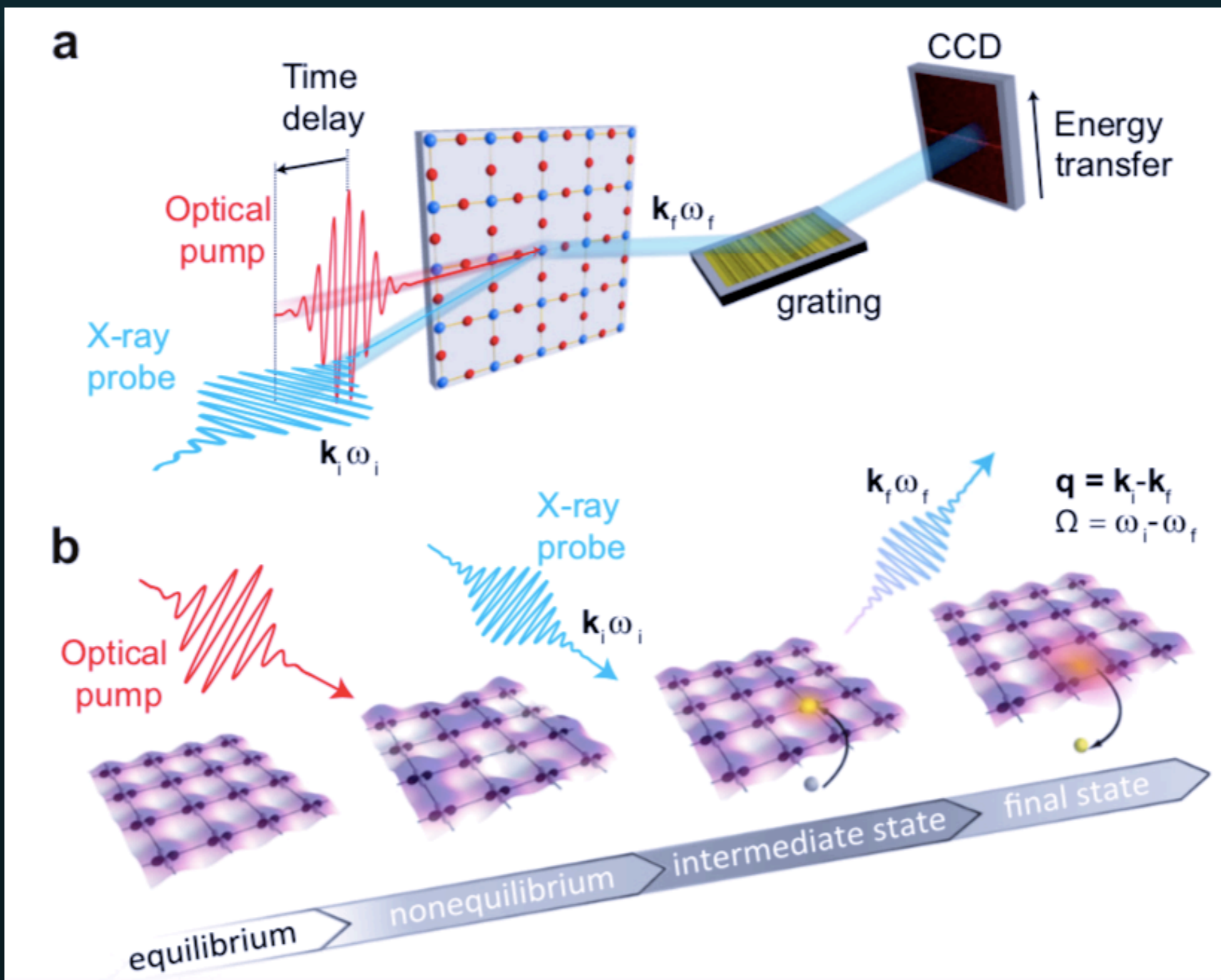
dynamics of the optical melting of intertwined charge and spin orders in a striped nickelate



Y.D. Chuang et al., *Phys. Rev. Lett.* (2012)

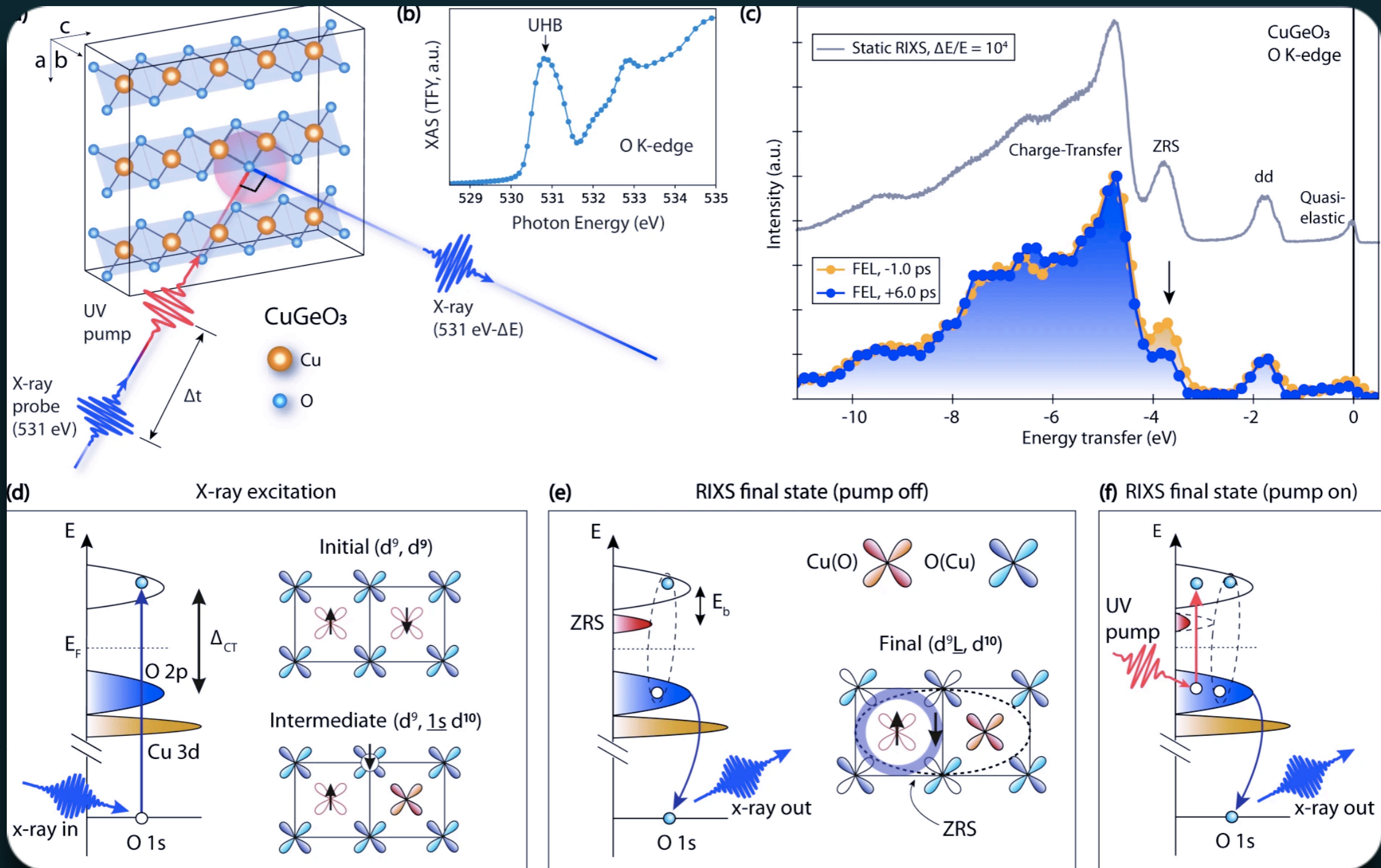


# time-resolved Resonant Inelastic X-ray Scattering



# time-resolved Resonant Inelastic X-ray Scattering

probing short-range magnetic correlations:  
dynamics of the Zhang-Rice singlet in  $\text{CuGeO}_3$



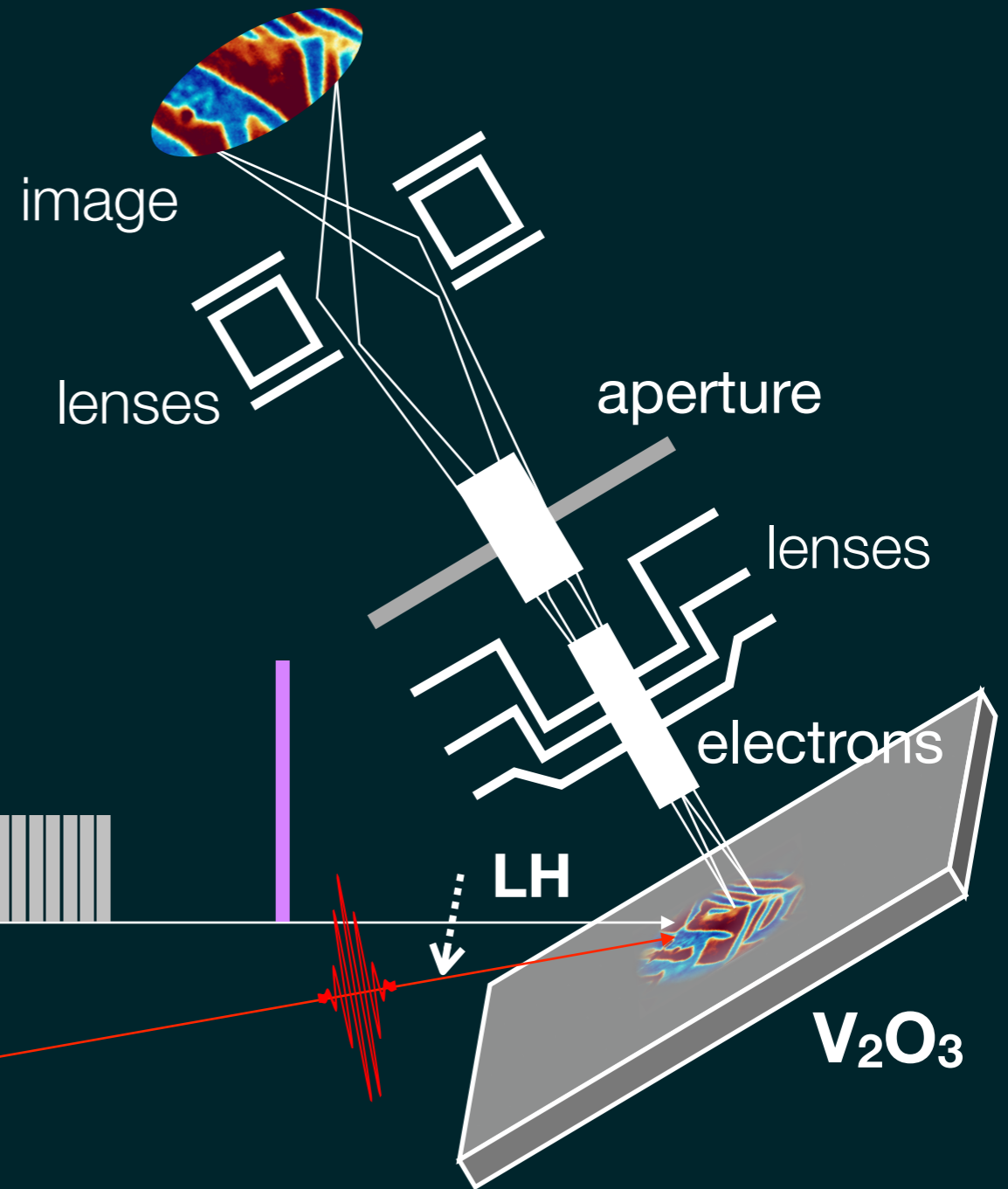
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towards time-resolved  
microscopy...

# time-resolved X-ray PEEM microscopy

hybrid injection mode

I06 beamline @ Diamond Light Source (UK)



X-rays (520 eV)

1.87  $\mu$ s

bunch

single pulse

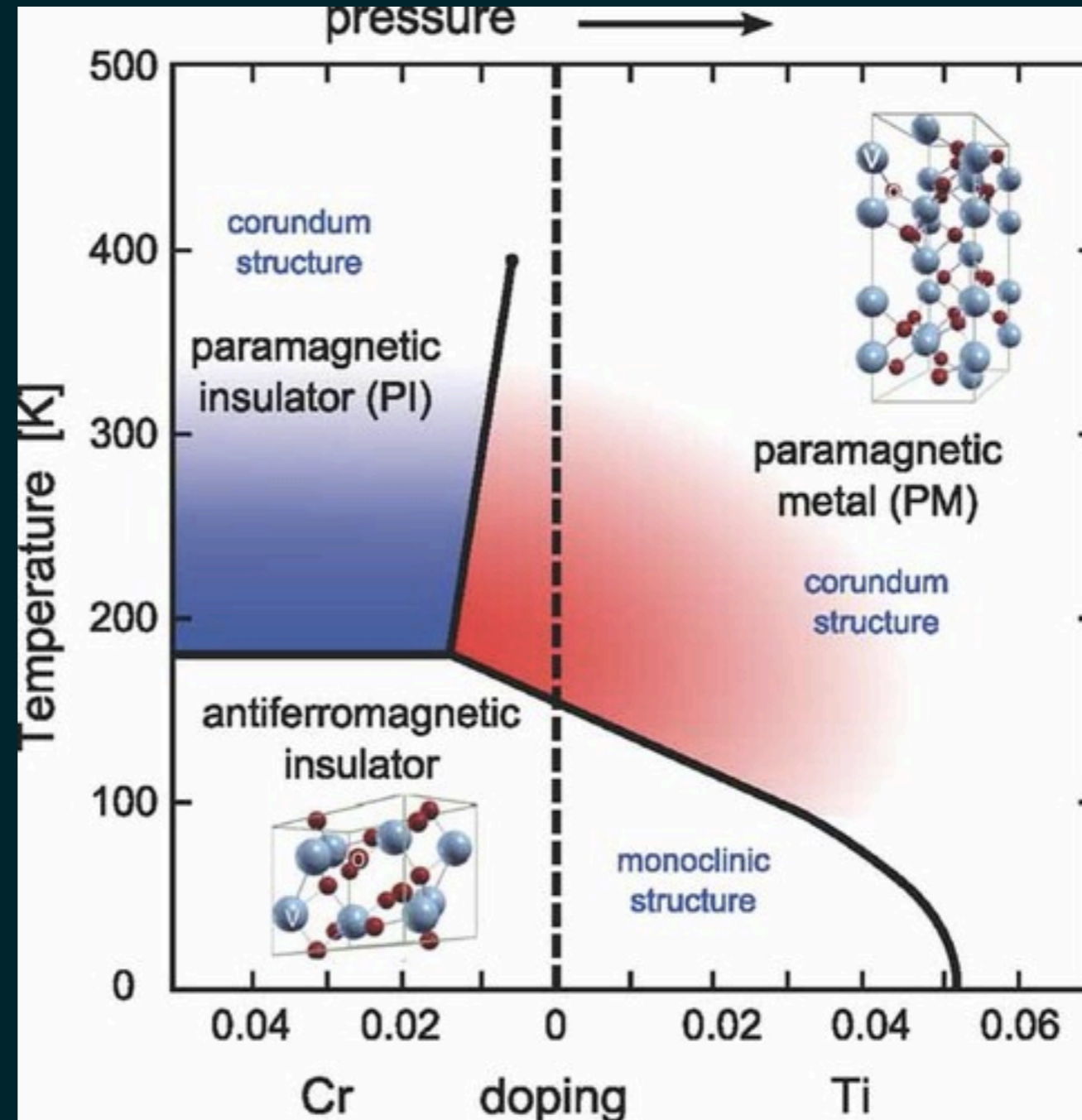
synchronized pump (1.5 eV)

time-resolved microscopy with 30 nm resolution



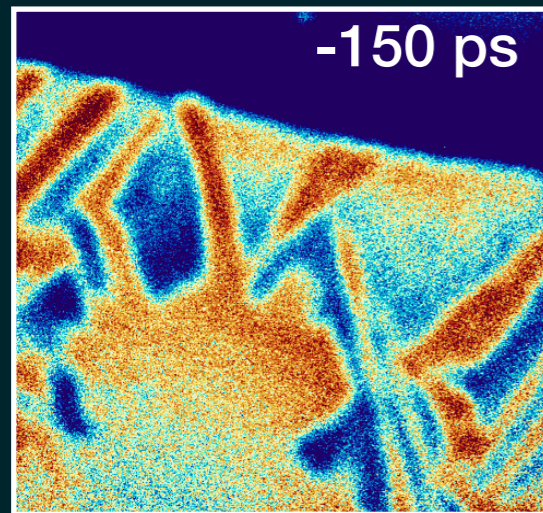
# time-resolved PEEM

## Mott transition in $V_2O_3$

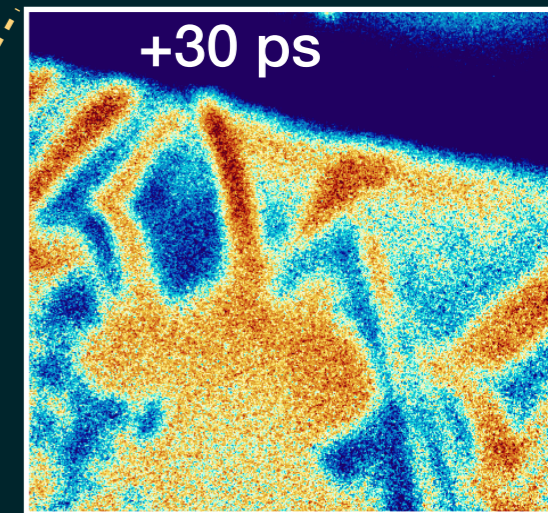
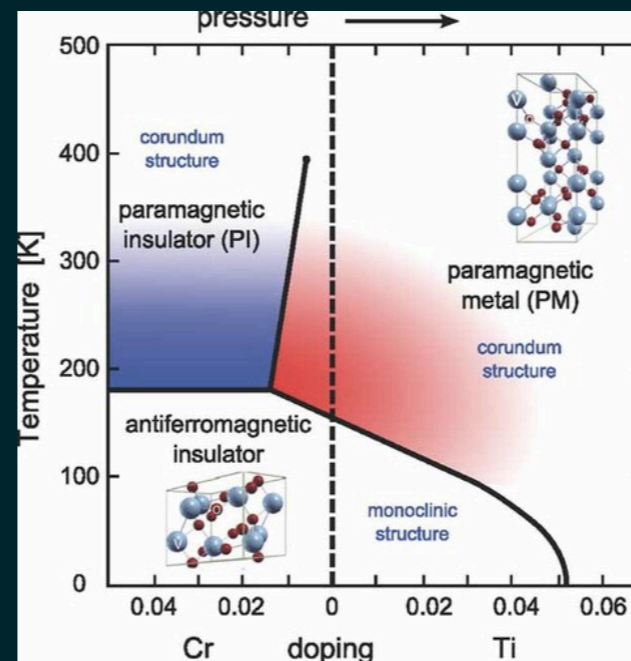


# time-resolved PEEM

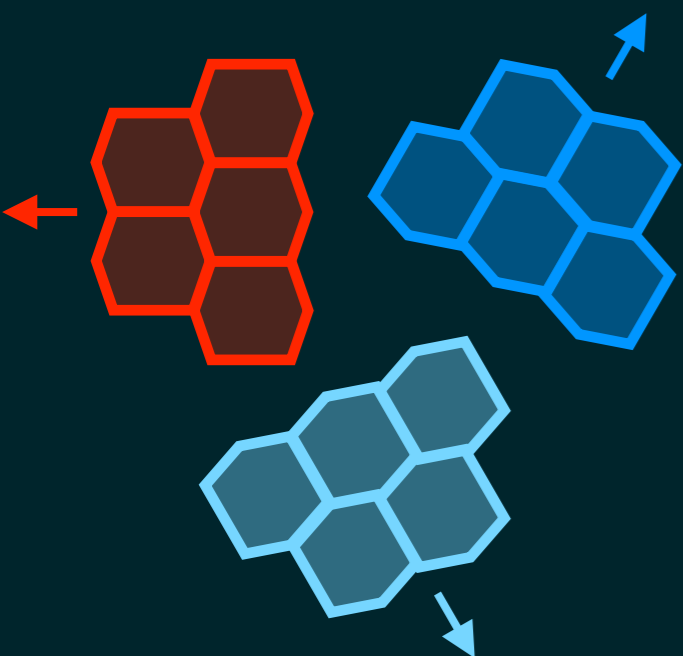
spontaneous formation of striped polydomains oriented along the crystallographic hexagonal axes



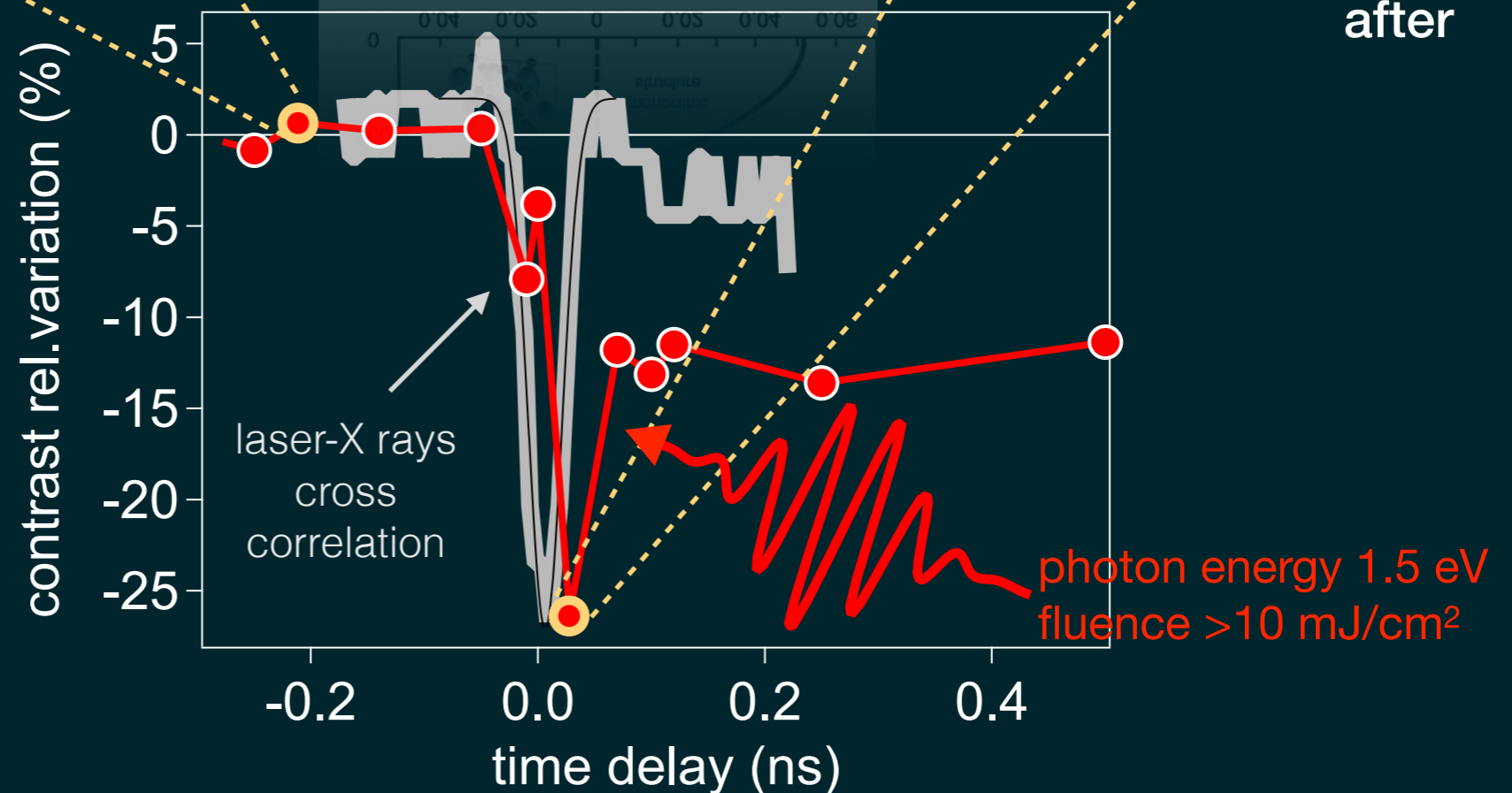
## V<sub>2</sub>O<sub>3</sub> phase diagram



before



after

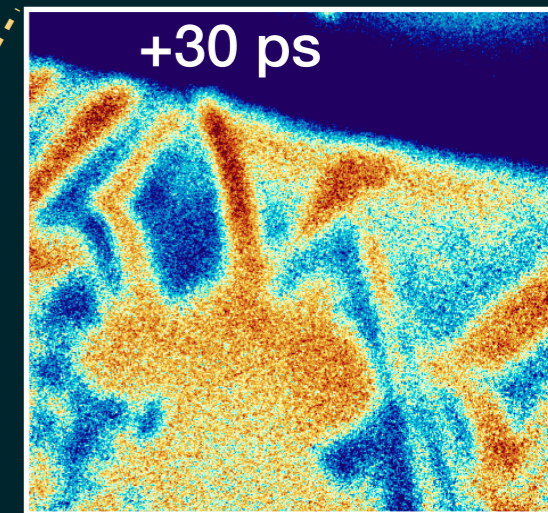
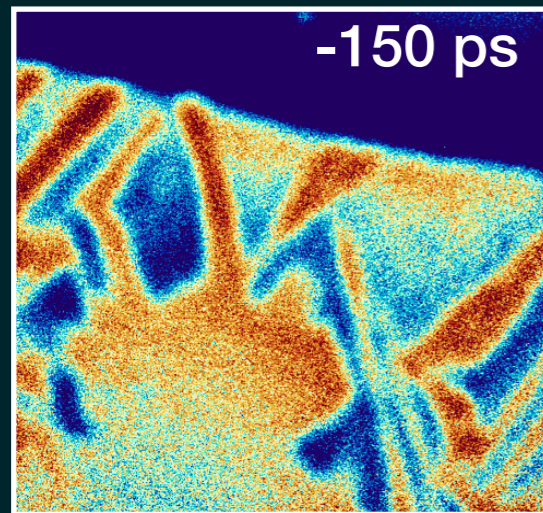
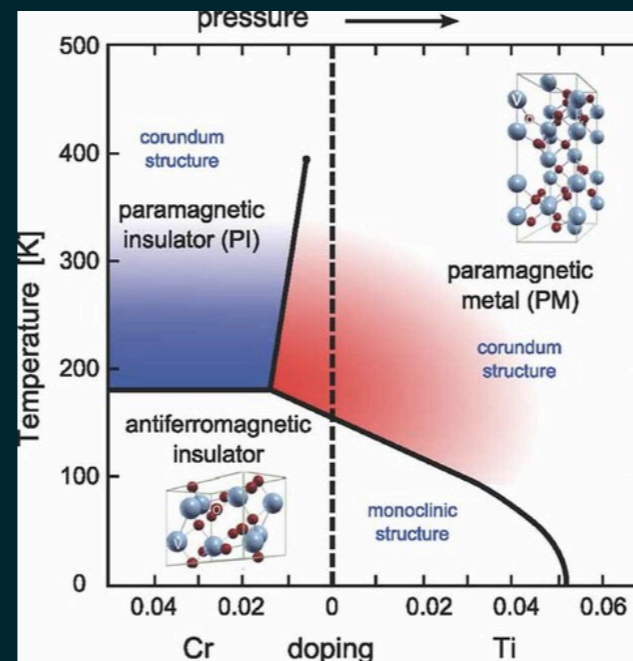




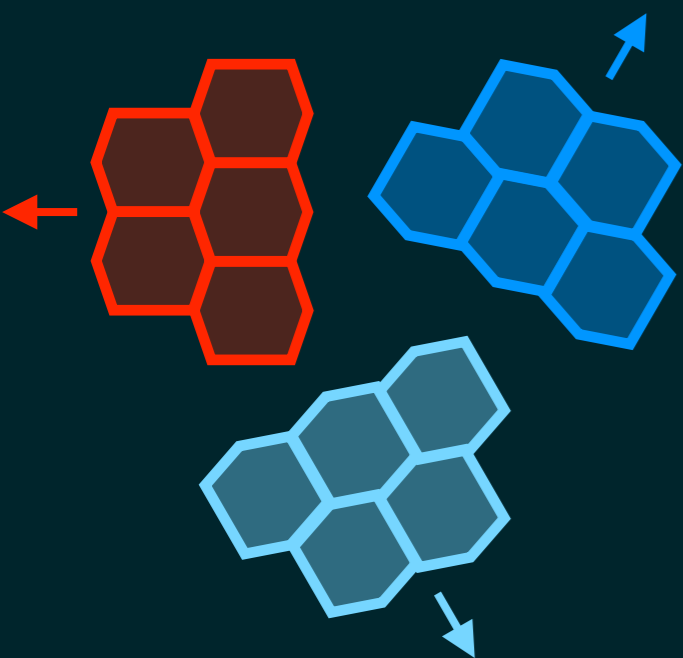
# time-resolved PEEM

spontaneous formation of striped polydomains oriented along the crystallographic hexagonal axes

$V_2O_3$  phase diagram

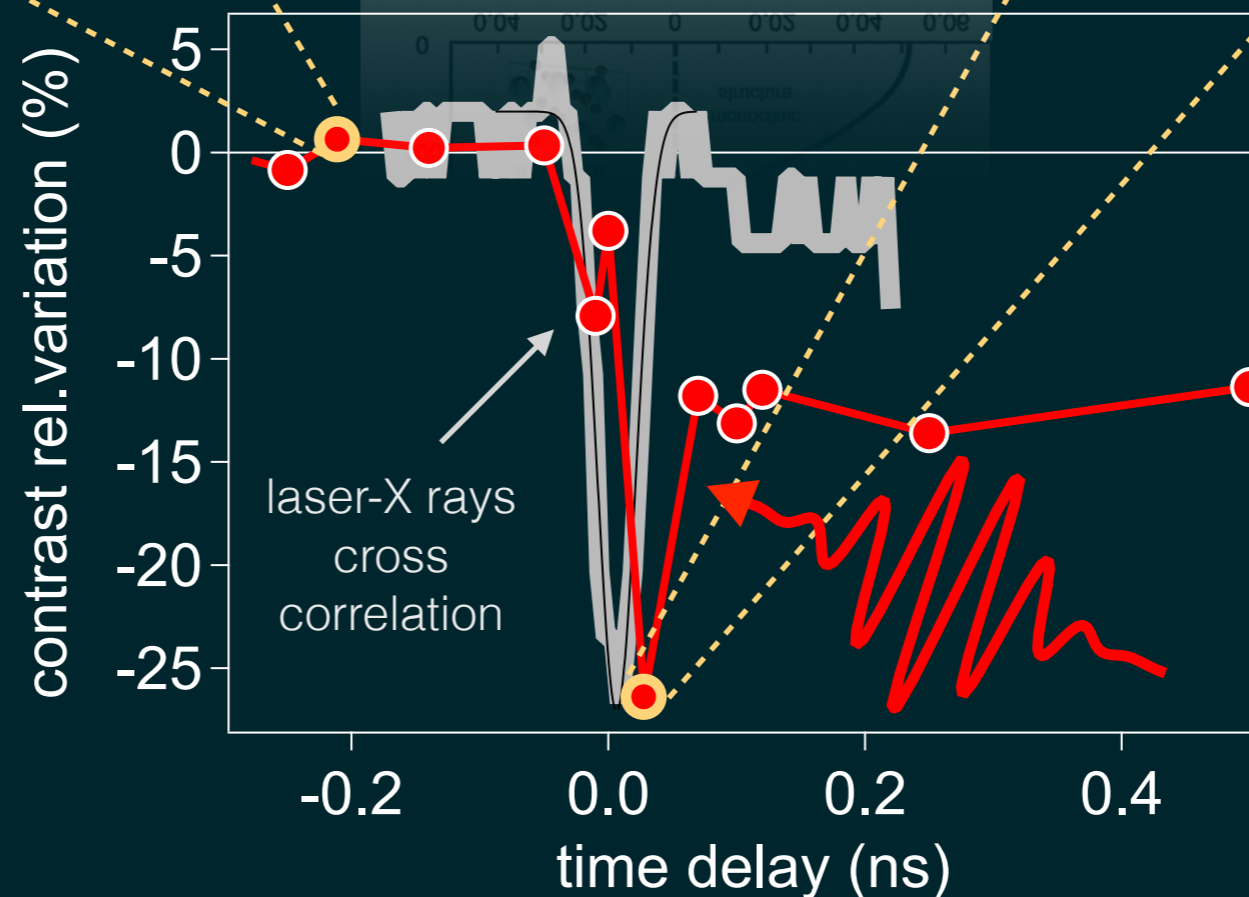
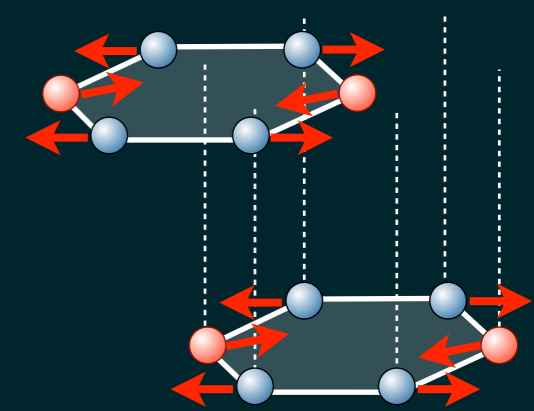


before

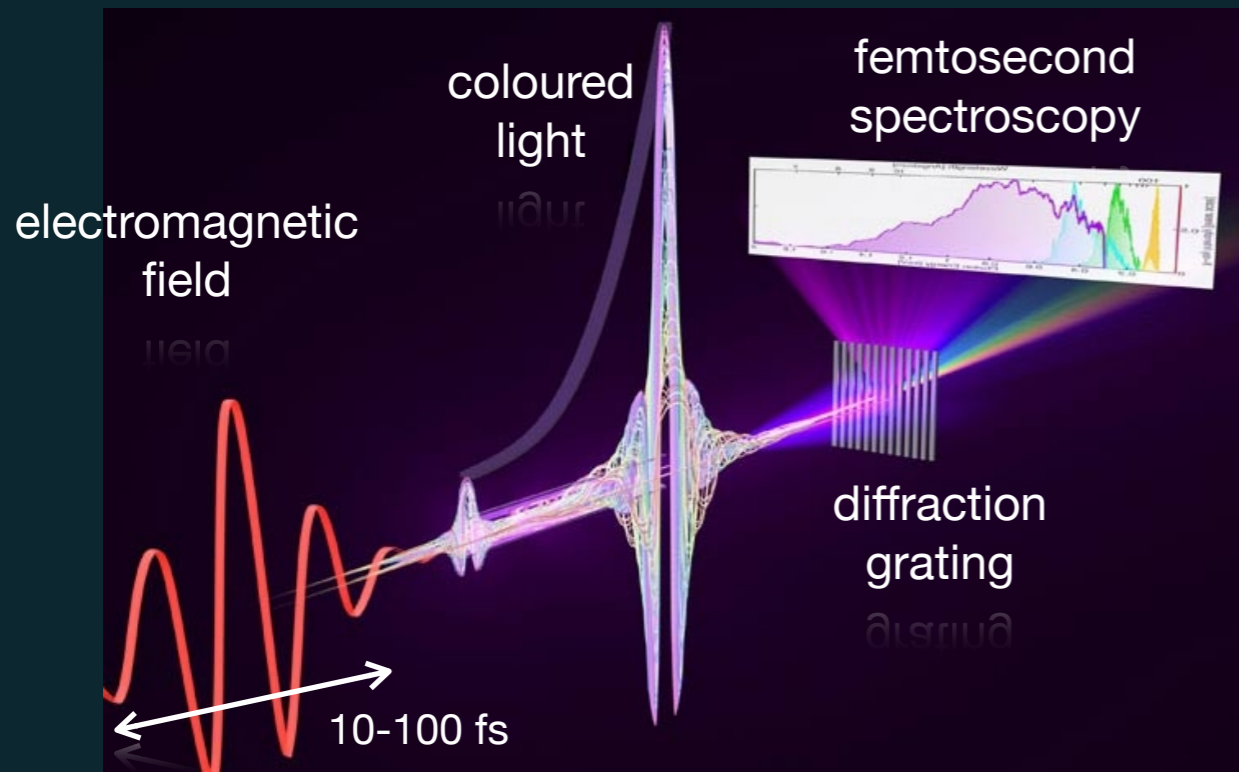


after

monoclinic metal



# electromagnetic properties

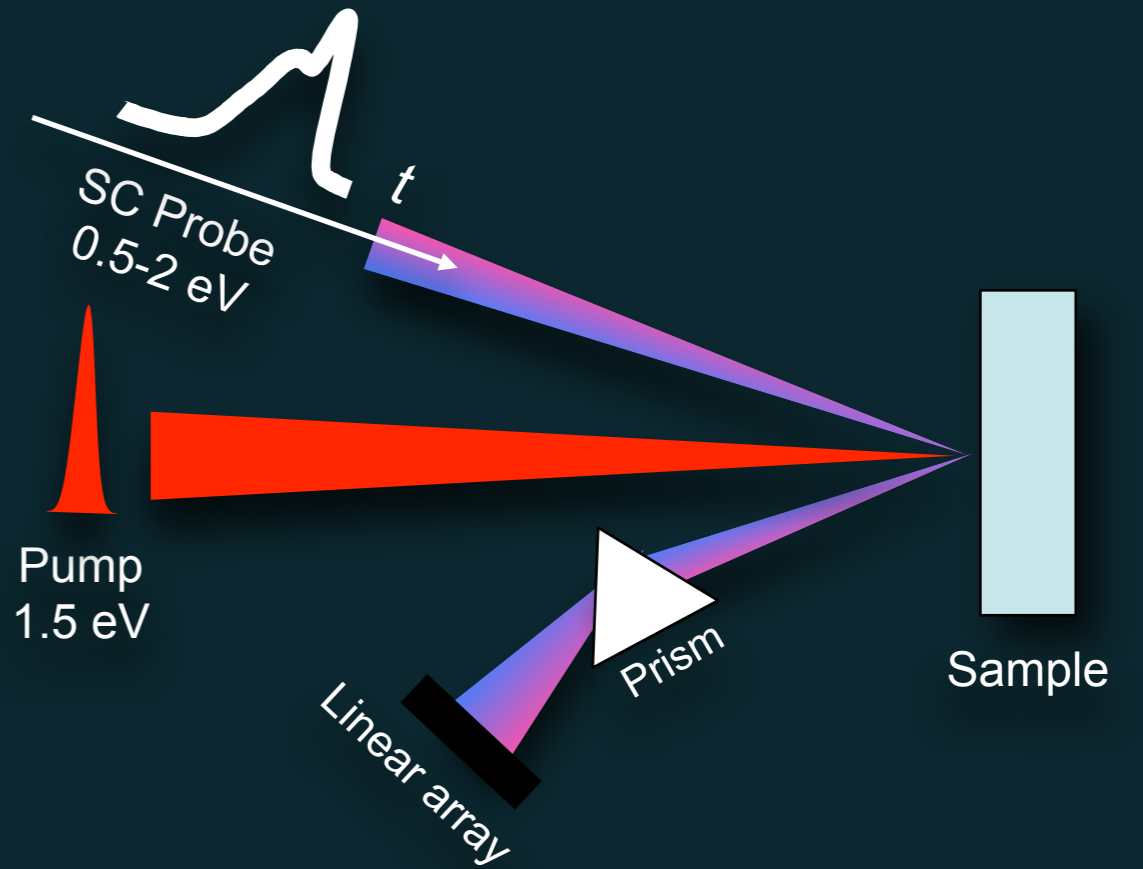
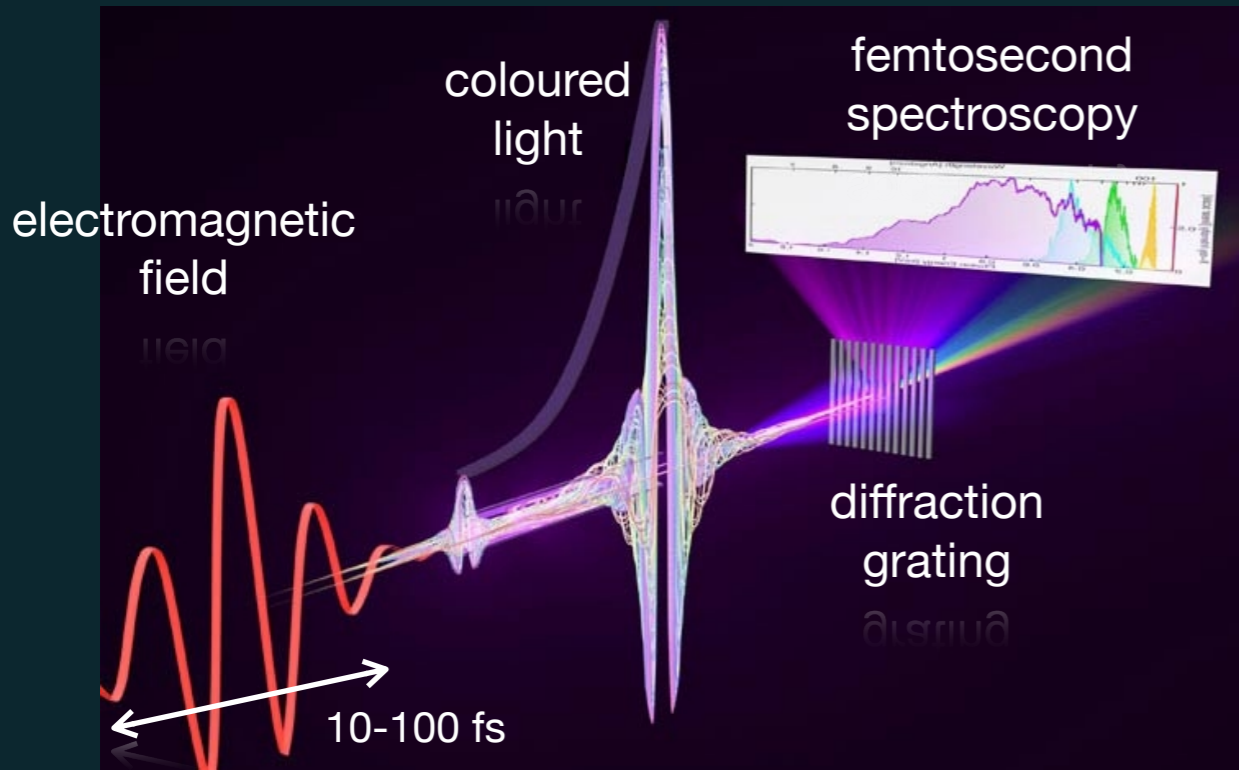




# non-equilibrium spectroscopy

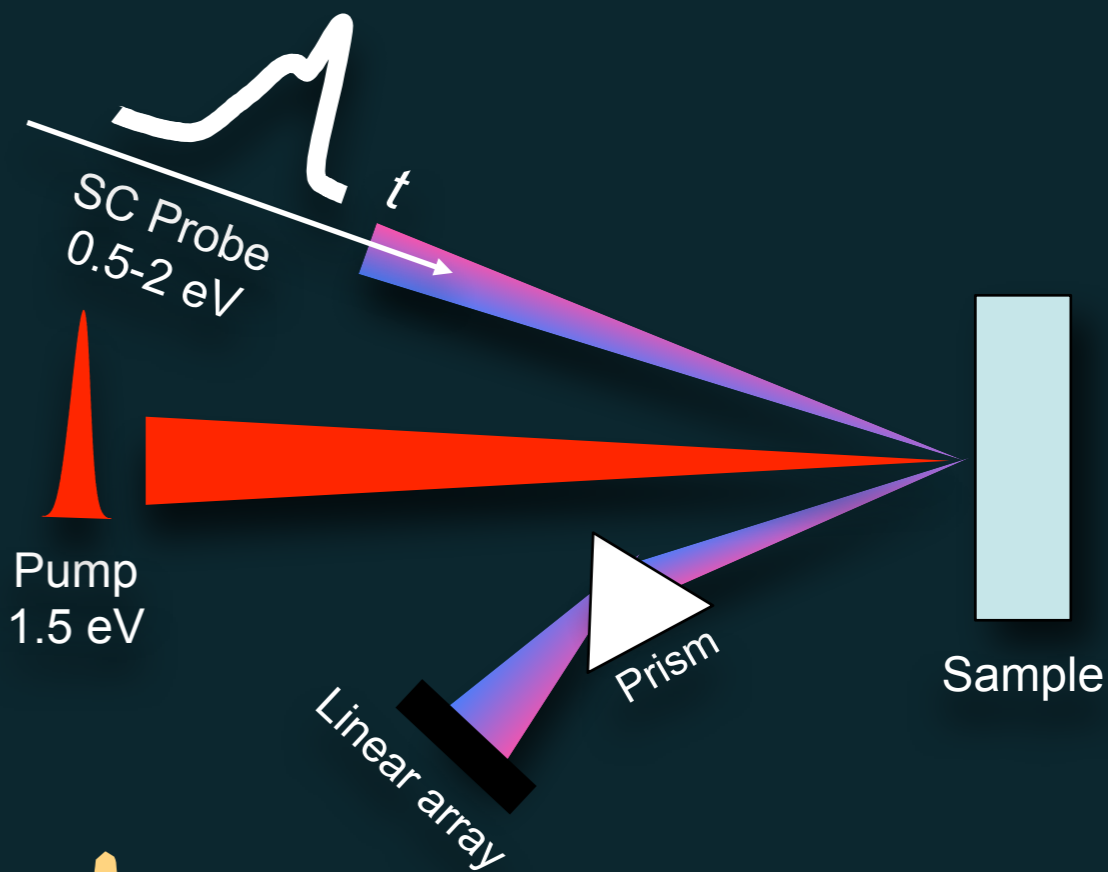
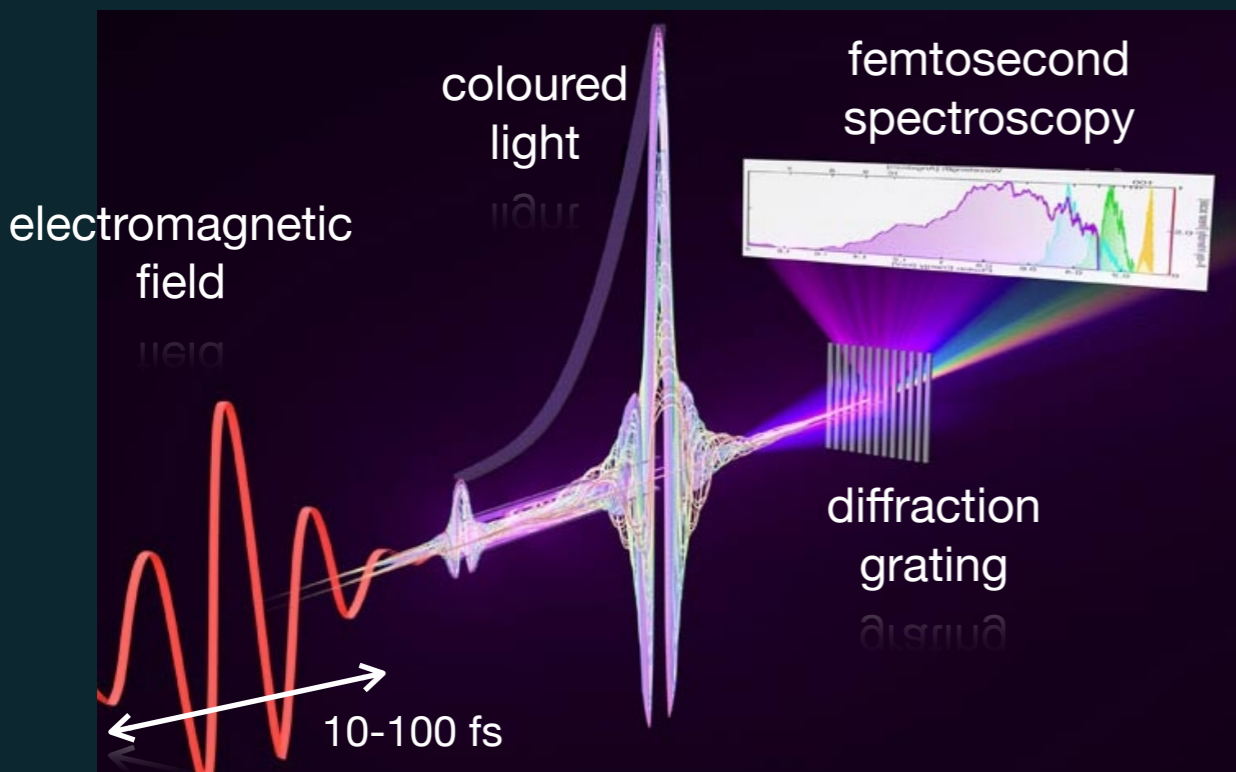
## from pump-probe to femtosecond spectroscopy

[www.nature.com/nature](http://www.nature.com/nature)



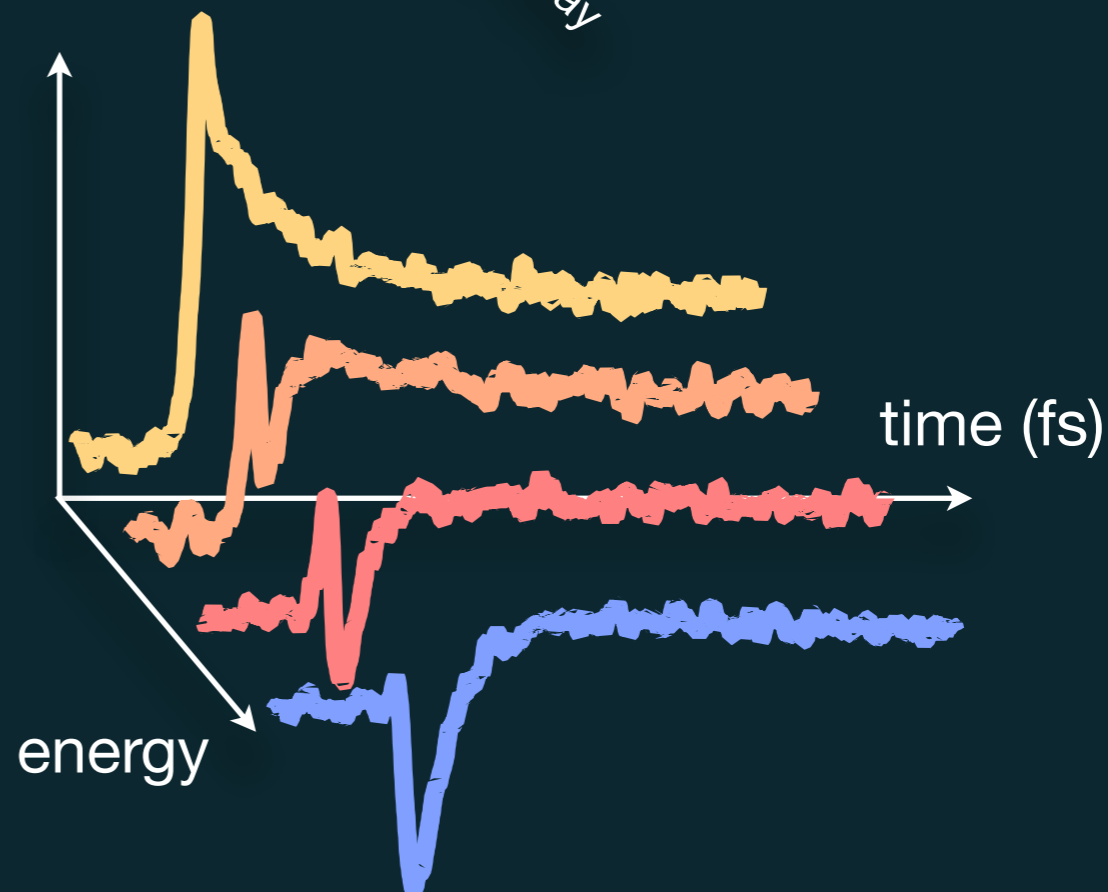
# non-equilibrium spectroscopy

## from pump-probe to femtosecond spectroscopy



$$\frac{\delta R}{R}(\omega, t) = \frac{R_{exc}(\omega, t) - R_{eq}(\omega)}{R_{eq}(\omega)}$$

time+frequency information

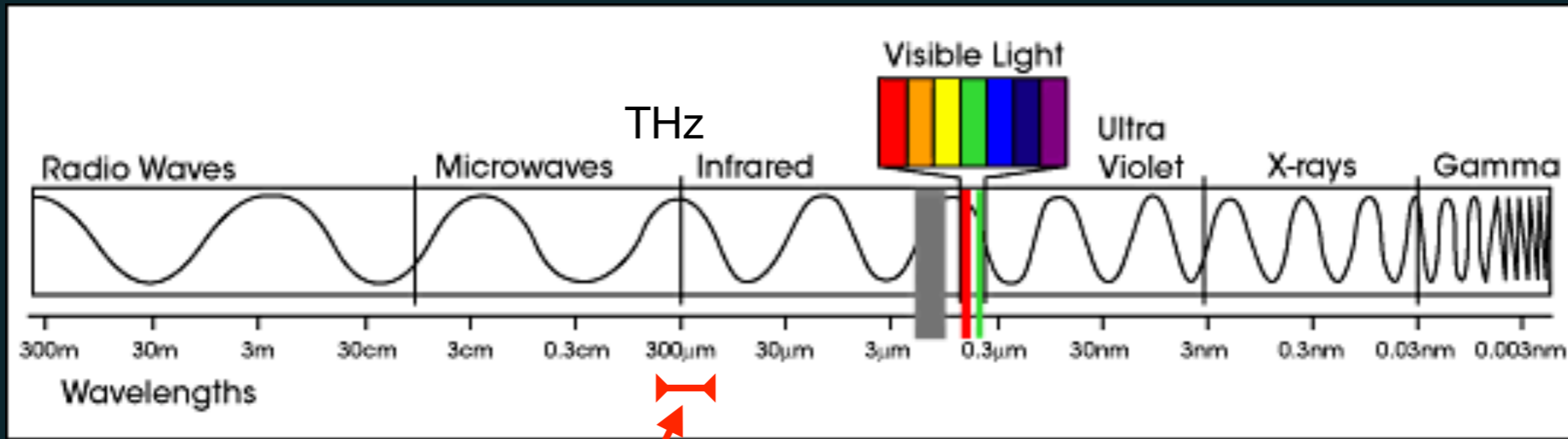


# the electromagnetic spectrum

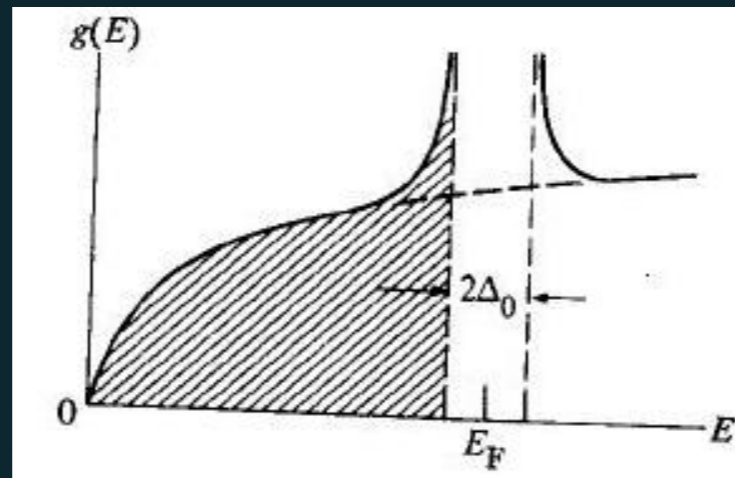
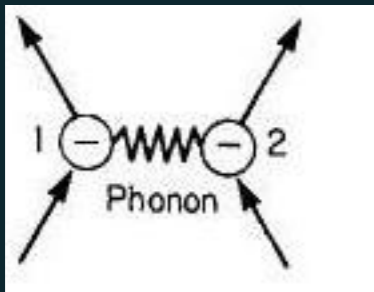
$$\omega = 2\pi c / \lambda$$

$$1 \text{ eV} = 1.24 \mu\text{m}$$

$$4 \text{ meV} = 1 \text{ THz} = 200 \mu\text{m}$$



~1-4 meV superconducting gap in BCS

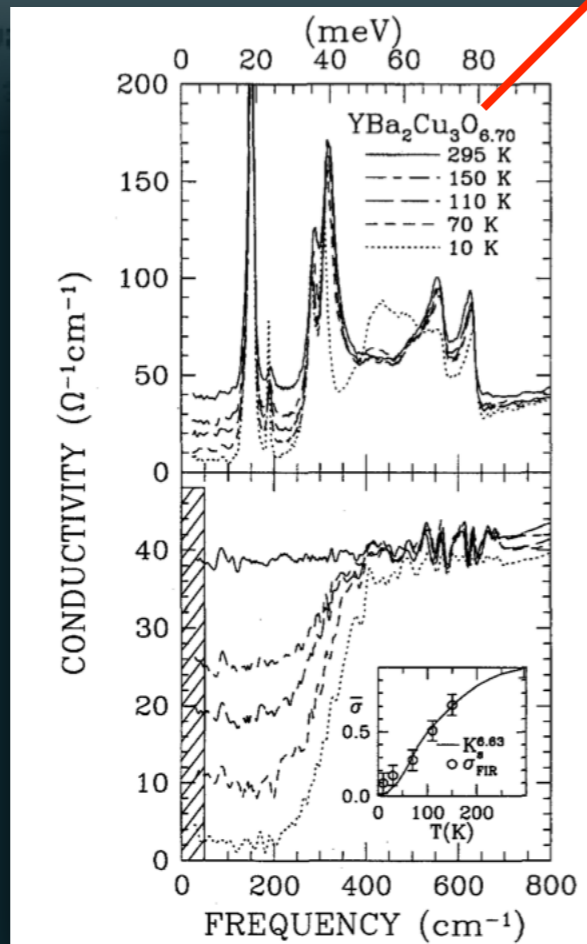
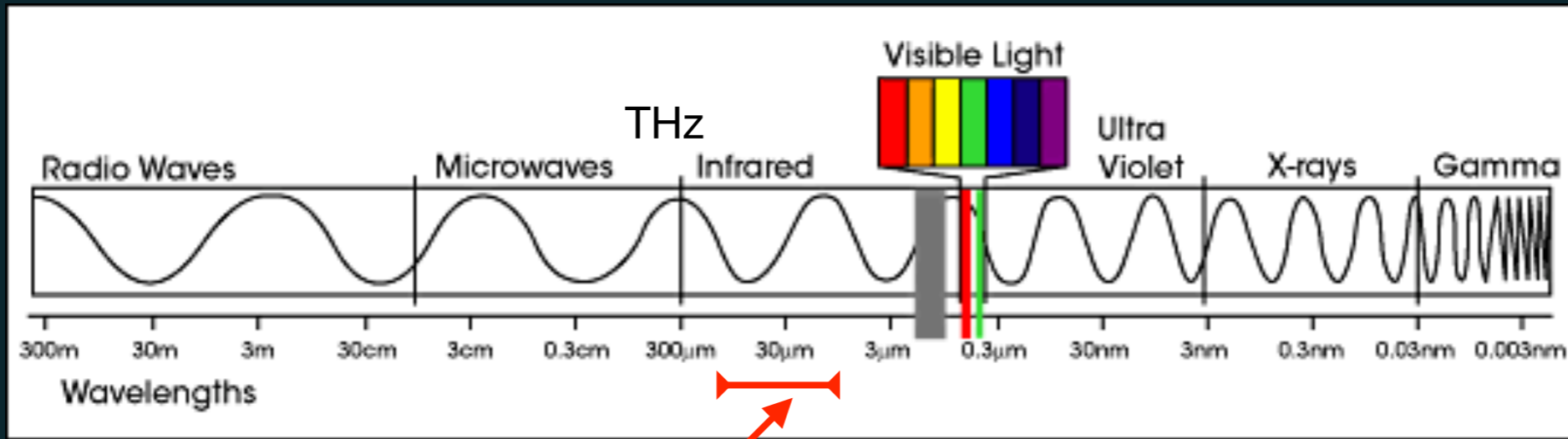


# the electromagnetic spectrum

$$\omega = 2\pi c / \lambda$$

$$1 \text{ eV} = 1.24 \mu\text{m}$$

$$4 \text{ meV} = 1 \text{ THz} = 200 \mu\text{m}$$



~30-90 meV infrared-active phonon modes

~40 meV superconducting gap and pseudogap in high- $T_c$

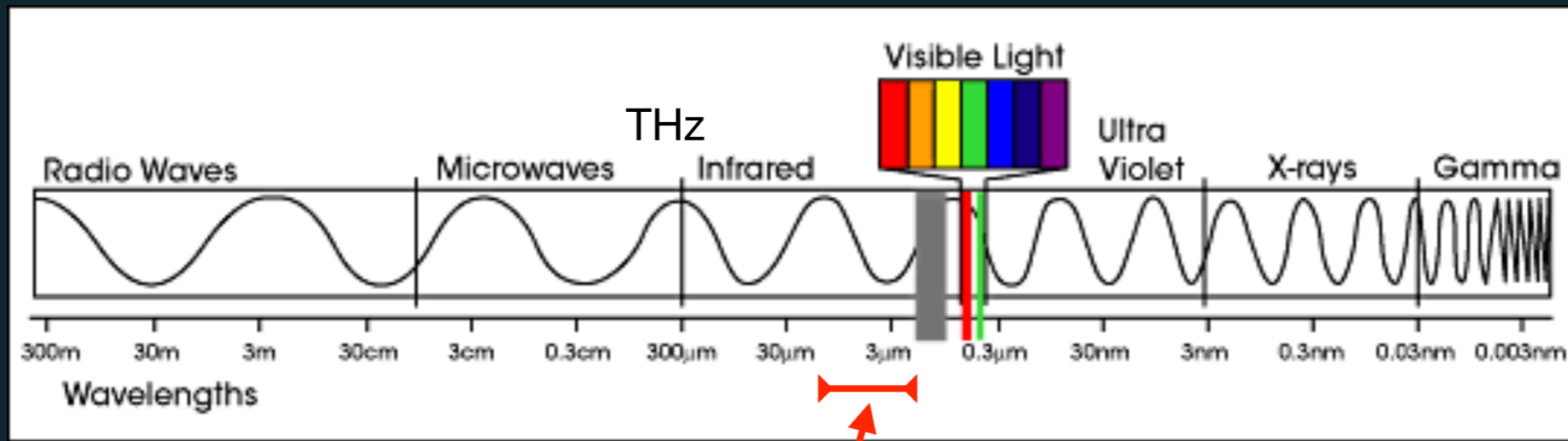
C. Homes et al, *Physica C: Superconductivity* 254, 265 (1995)

# the electromagnetic spectrum

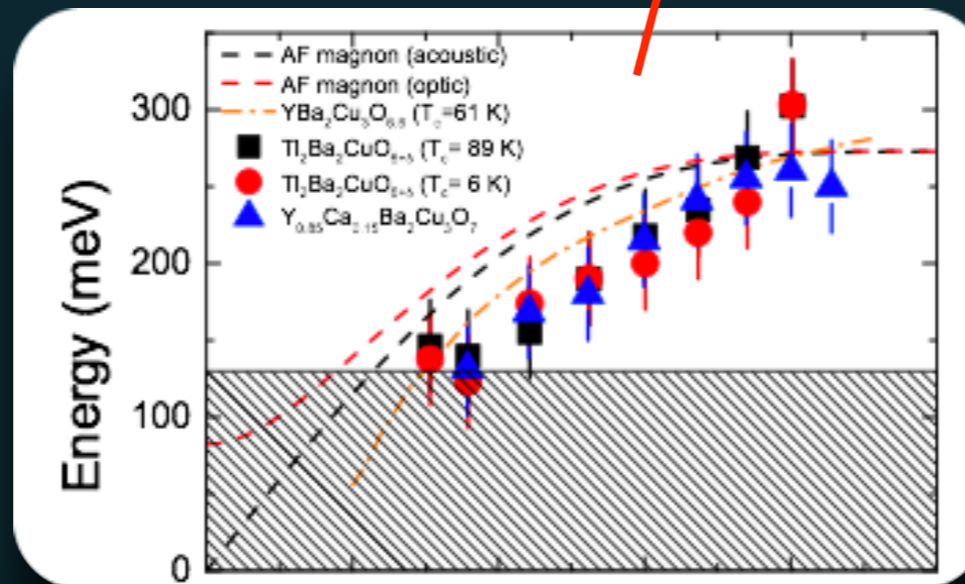
$$\omega = 2\pi c / \lambda$$

$$1 \text{ eV} = 1.24 \mu\text{m}$$

$$4 \text{ meV} = 1 \text{ THz} = 200 \mu\text{m}$$



$\sim 300 \text{ meV}$  spin fluctuations



RIXS: Dispersion of paramagnons in cuprates at all dopings

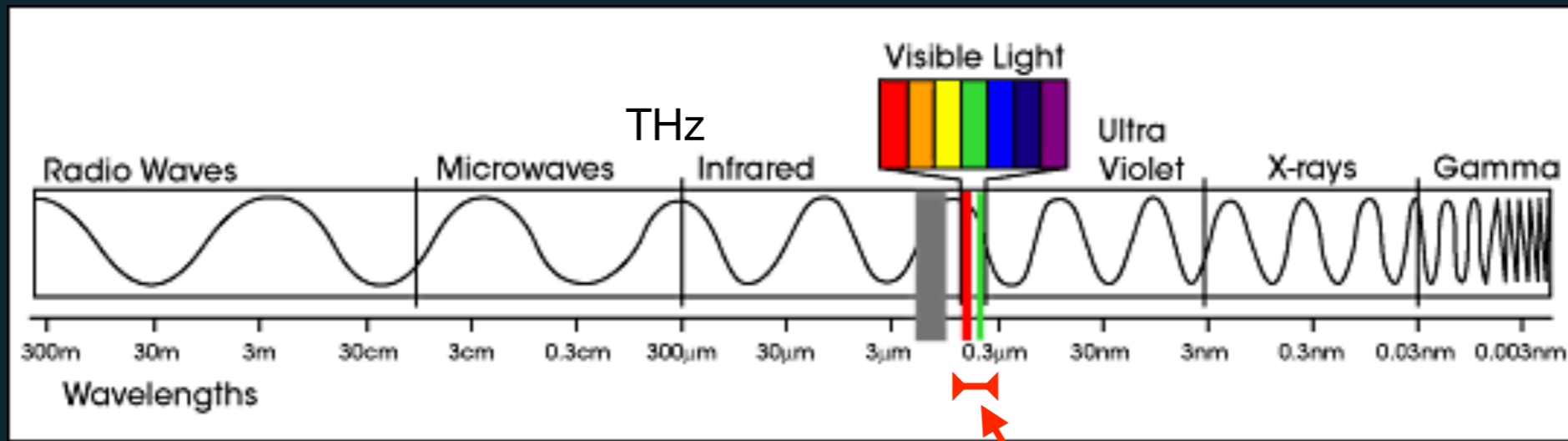


# the electromagnetic spectrum

$$\omega = 2\pi c / \lambda$$

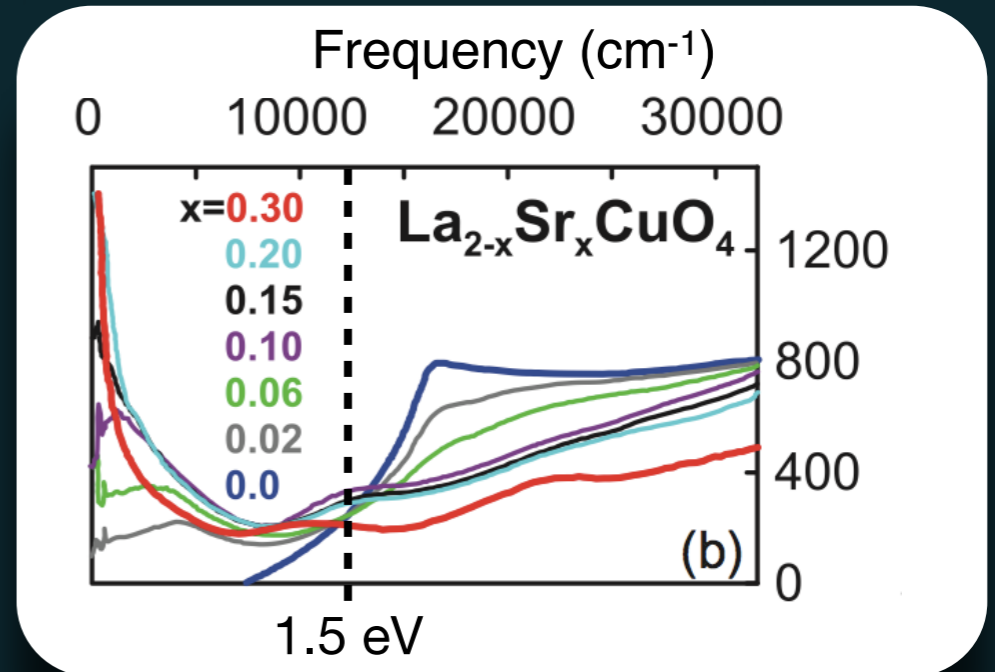
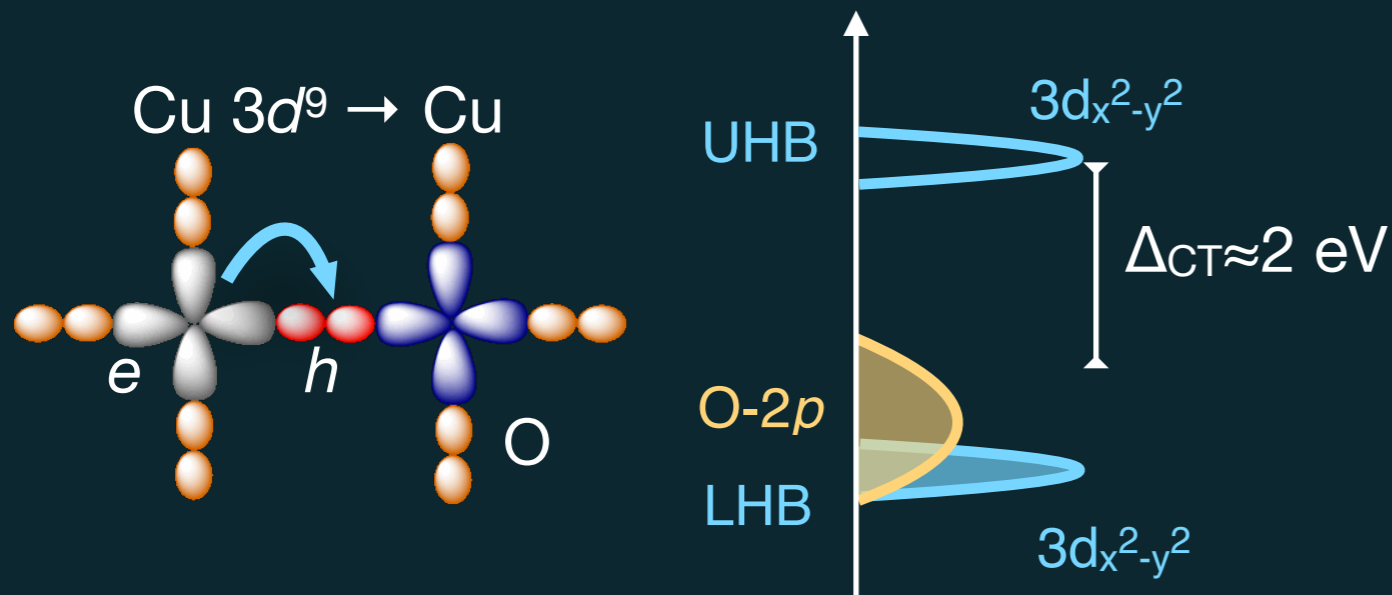
$$1 \text{ eV} = 1.24 \mu\text{m}$$

$$4 \text{ meV} = 1 \text{ THz} = 200 \mu\text{m}$$



>1.5 eV charge transfer and interband transitions

$$8064 \text{ cm}^{-1} = 1 \text{ eV}$$



S. Uchida et al, *Phys. Rev. B* 43, 7942 (1991)

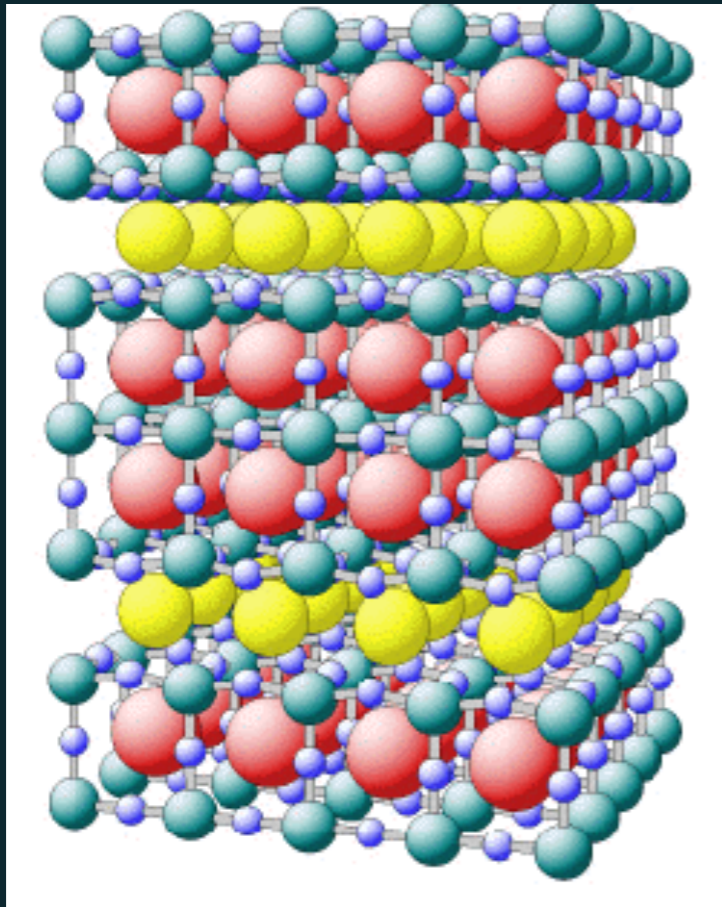
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light pulses to **UNDERSTAND**  
equilibrium properties

⇒ electron-boson coupling in  
superconductors

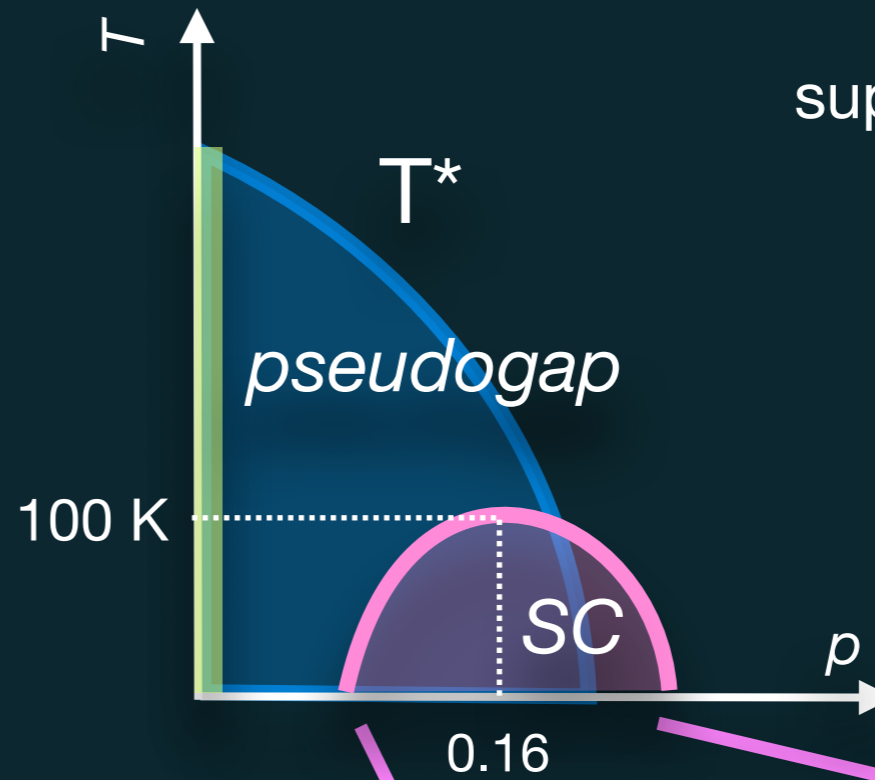
# copper oxides

bi-dimensional copper oxides



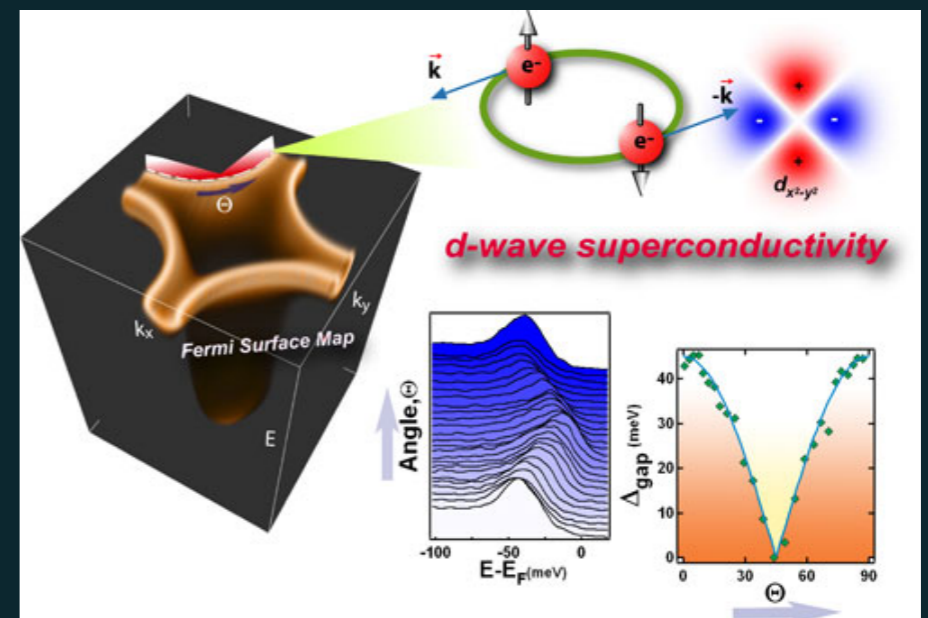
Nobel Prize 1986:  
J.G. Bednorz, K.A. Müller

which glue in high- $T_c$   
superconductors?



superconductivity on  
top of a very  
unconventional  
ground state!

$d$ -wave  
superconducting  
gap



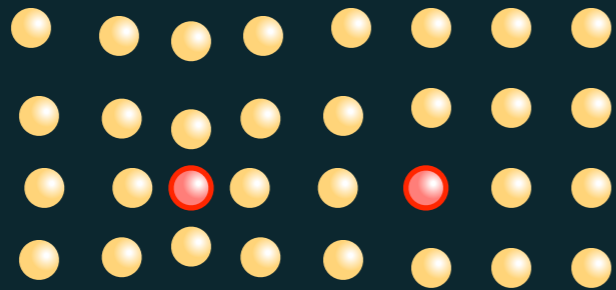
ZX Shen's group



# retarded interaction and the “glue” problem

BCS

glue: lattice distortion



space

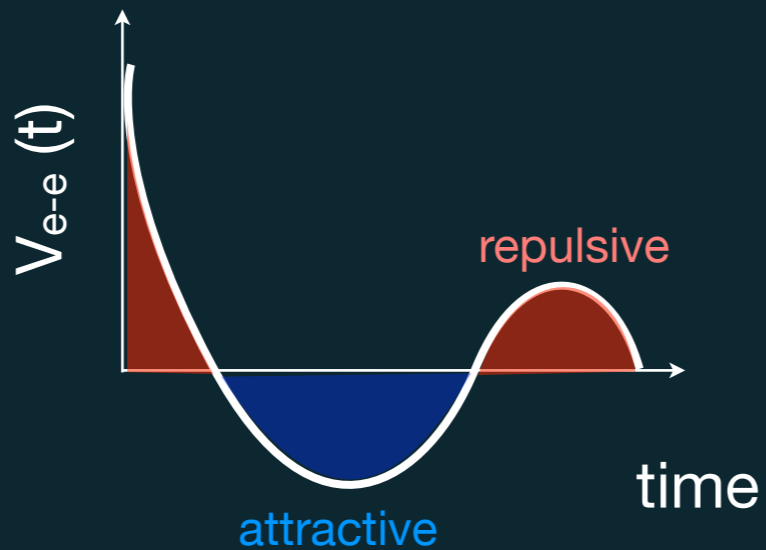
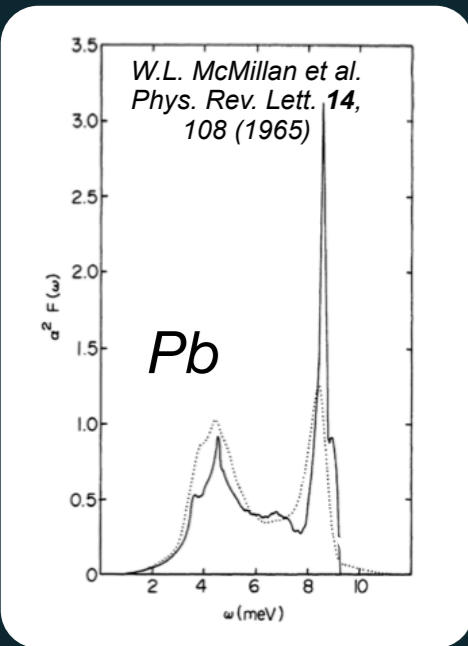
$$\lambda = 2\pi/|Q|$$



energy scale

$$\alpha^2 F(\Omega)$$

timescale

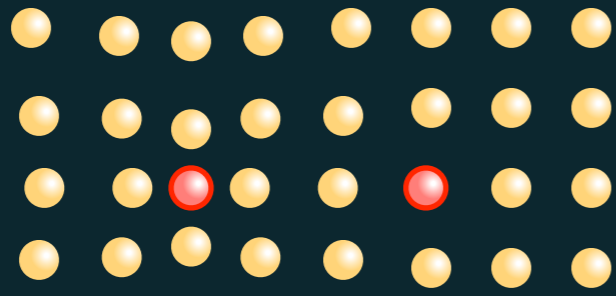


$$T = 2\pi/\Omega_Q \approx 100 \text{ fs}$$

# retarded interaction and the "glue" problem

## BCS

glue: lattice distortion



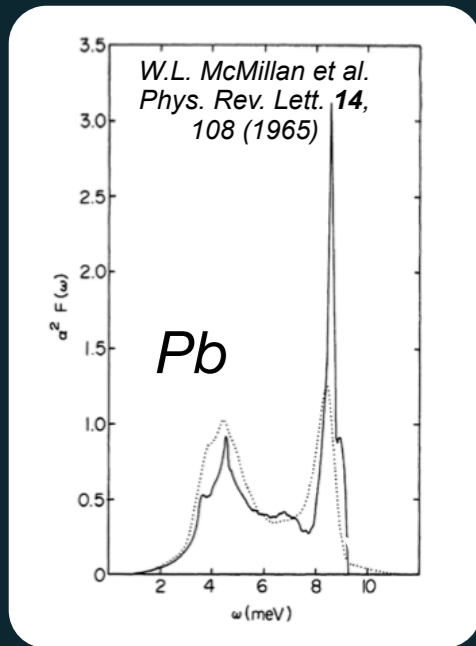
space

$$\lambda = 2\pi/|Q|$$

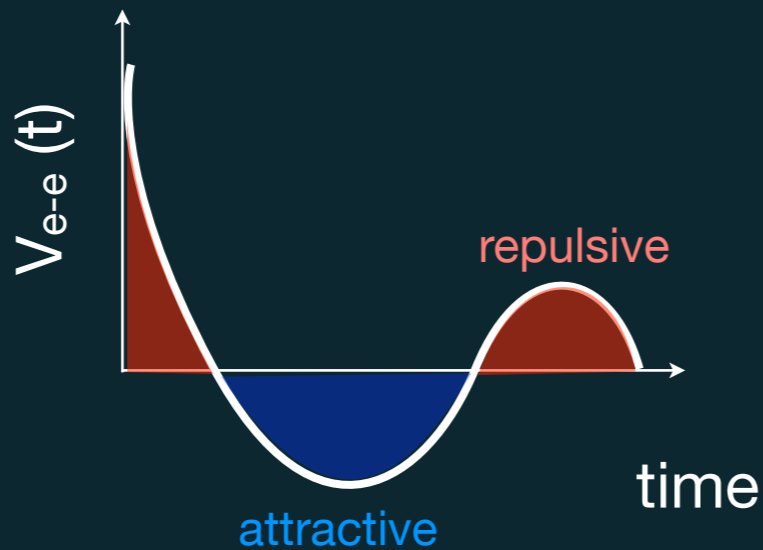


energy scale

$$\alpha^2 F(\Omega)$$



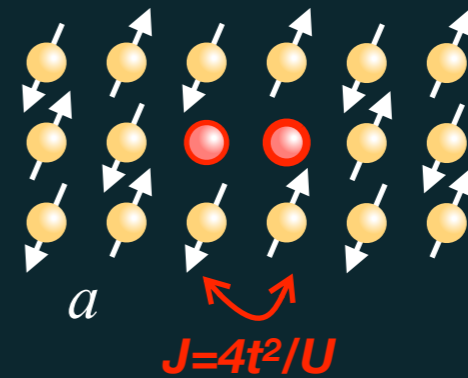
timescale



$$T = 2\pi/\Omega_Q \approx 100 \text{ fs}$$

## unconventional

d-wave: other glue?

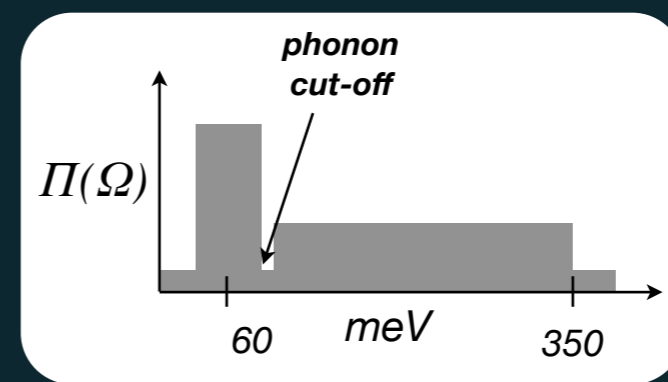


space

$$\lambda = a (\approx 4 \text{ \AA})$$

energy scale

$$\Pi(\Omega) = \alpha^2 F(\Omega) + I^2 \chi(\Omega)$$



$$\hbar/J (\approx 6 \text{ fs})$$

J. Carbotte et al. *Rep. Prog. Phys.* 74, 066501 (2011)

$$T = 2\pi/J < 10 \text{ fs}$$

# electron-boson coupling

electron-boson coupling affects the electron dynamics  $\Rightarrow$

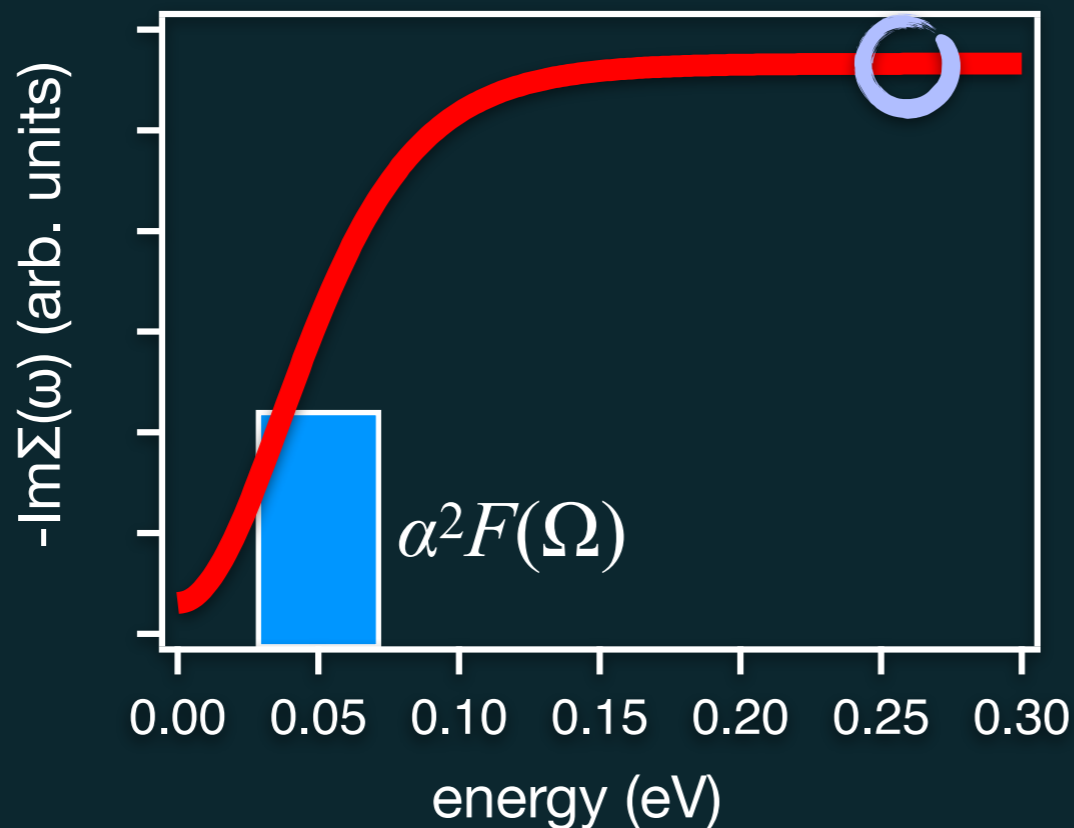
frequency-dependent electron self-energy:  $\Sigma(\omega)$

for Fermi liquids: 
$$\Sigma(\mathbf{k}, \omega, T) = \int_0^\infty \alpha^2 F(\mathbf{k}, \Omega) L(\omega, \Omega, T) d\Omega$$

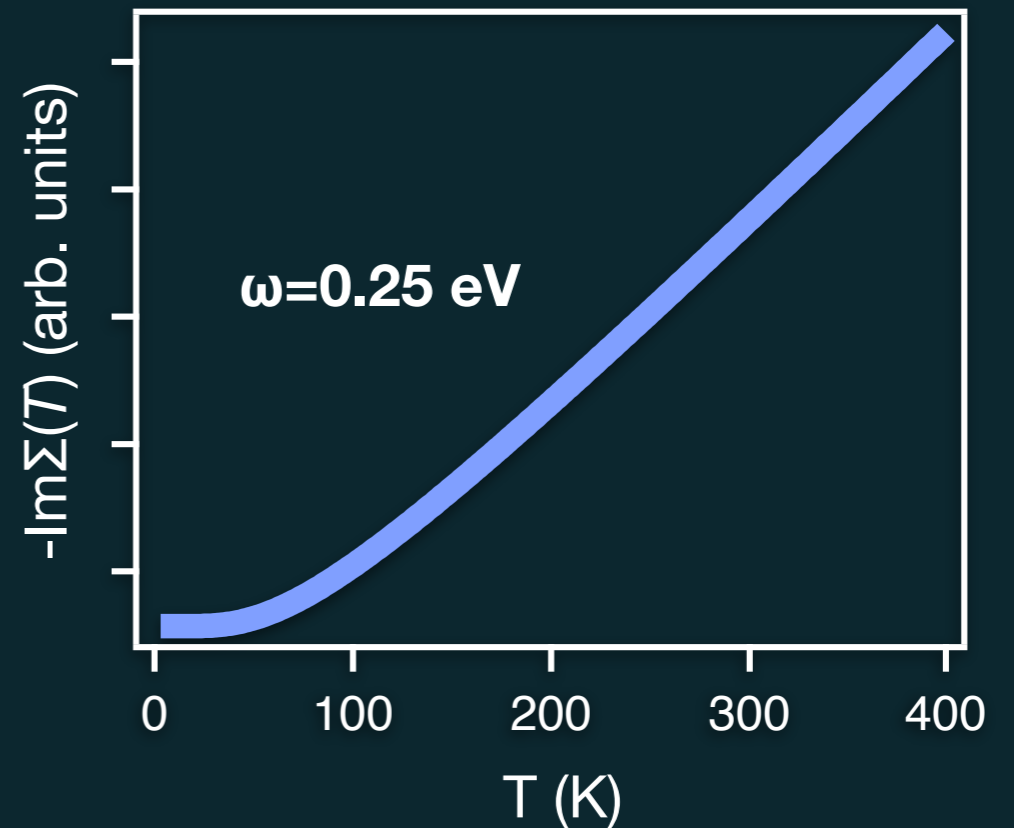
*bosonic function: the coupling*

*kernel function: the population*

frequency dependence

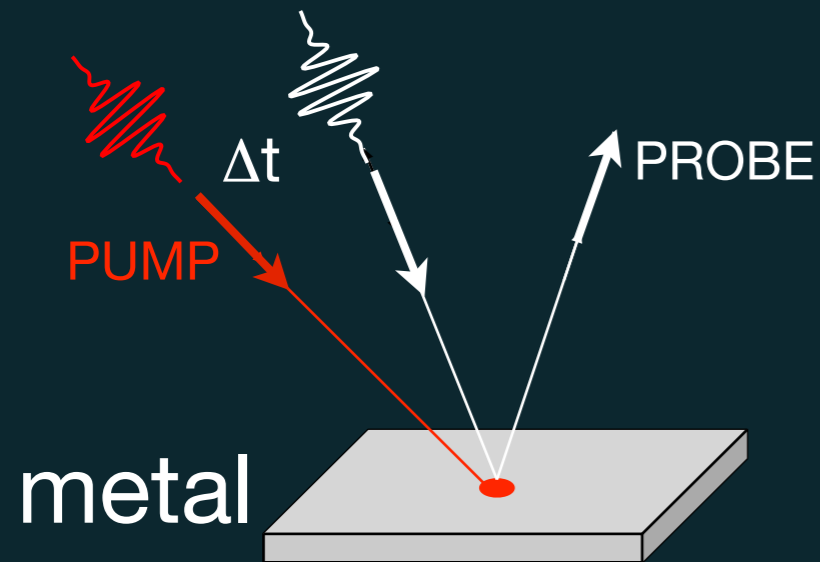


temperature dependence



# energy relaxation pathways

pump probe on metals



effective-temperature model

$$\frac{\partial T_e}{\partial t} = \frac{G(\Pi_{lat}, T_{lat}, T_e)}{\gamma_e T_e} + \frac{p}{\gamma_e T_e}$$
$$\frac{\partial T_{lat}}{\partial t} = -\frac{G(\Pi_{lat}, T_{lat}, T_e)}{C_{lat}}$$

$G/C$   
determines the dynamics in the time domain

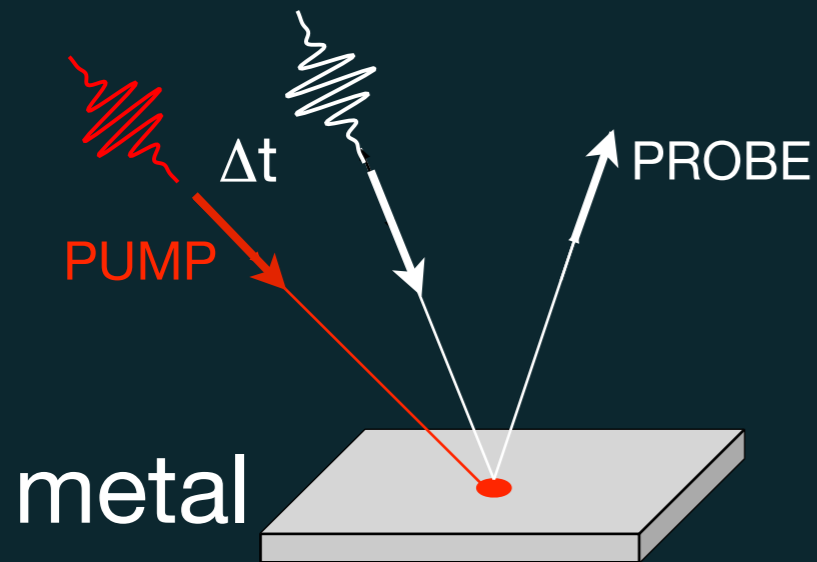
*Note: In the equations above, the term  $\frac{p}{\gamma_e T_e}$  in the first equation is annotated with a red arrow labeled "PUMP".*

P.B. Allen, *Phys. Rev Lett.* **59** 1460 (1987)



# energy relaxation pathways

pump probe on metals



effective-temperature model

$$\frac{\partial T_e}{\partial t} = \frac{G(\Pi_{lat}, T_{lat}, T_e)}{\gamma_e T_e} + \frac{p}{\gamma_e T_e}$$

$$\frac{\partial T_{lat}}{\partial t} = -\frac{G(\Pi_{lat}, T_{lat}, T_e)}{C_{lat}}$$

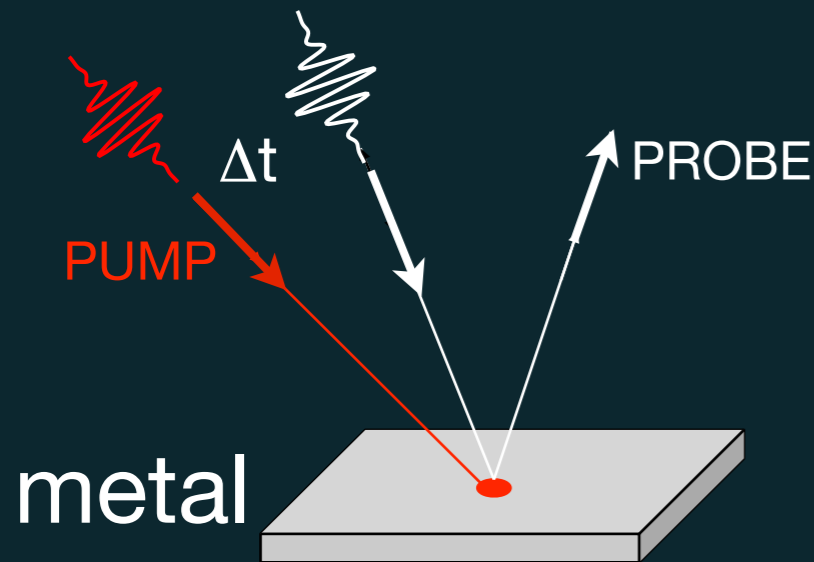
$G/C$

determines the dynamics in the time domain

$$G = \frac{3\gamma_e}{\pi \hbar k_B^2} \int_0^\infty d\Omega \Pi_{lat}(\Omega) [N(\Omega, T_{lat}) - N(\Omega, T_e)]$$

# energy relaxation pathways

pump probe on metals



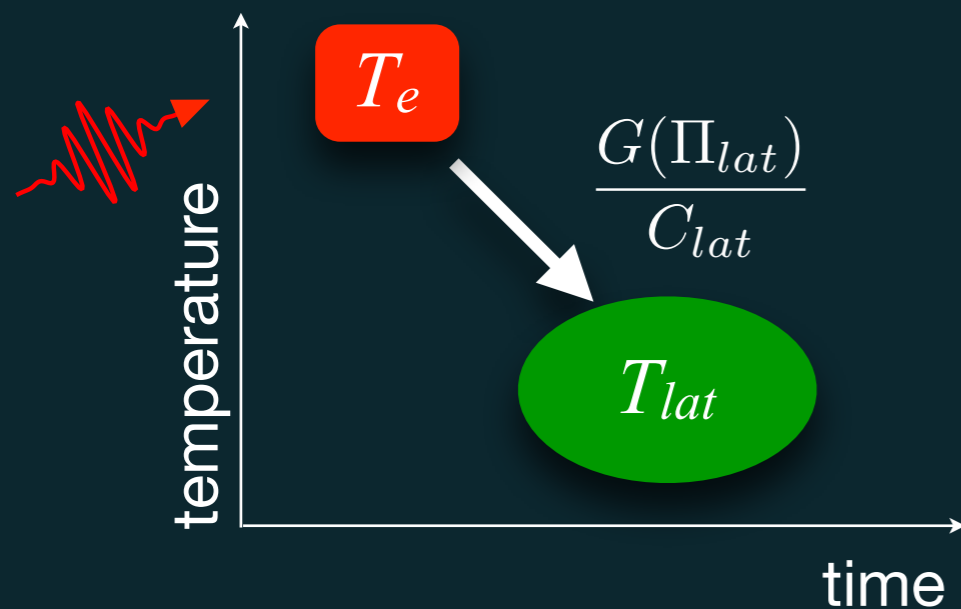
effective-temperature model

$$\frac{\partial T_e}{\partial t} = \frac{G(\Pi_{lat}, T_{lat}, T_e)}{\gamma_e T_e} + \frac{p}{\gamma_e T_e} \quad G/C$$

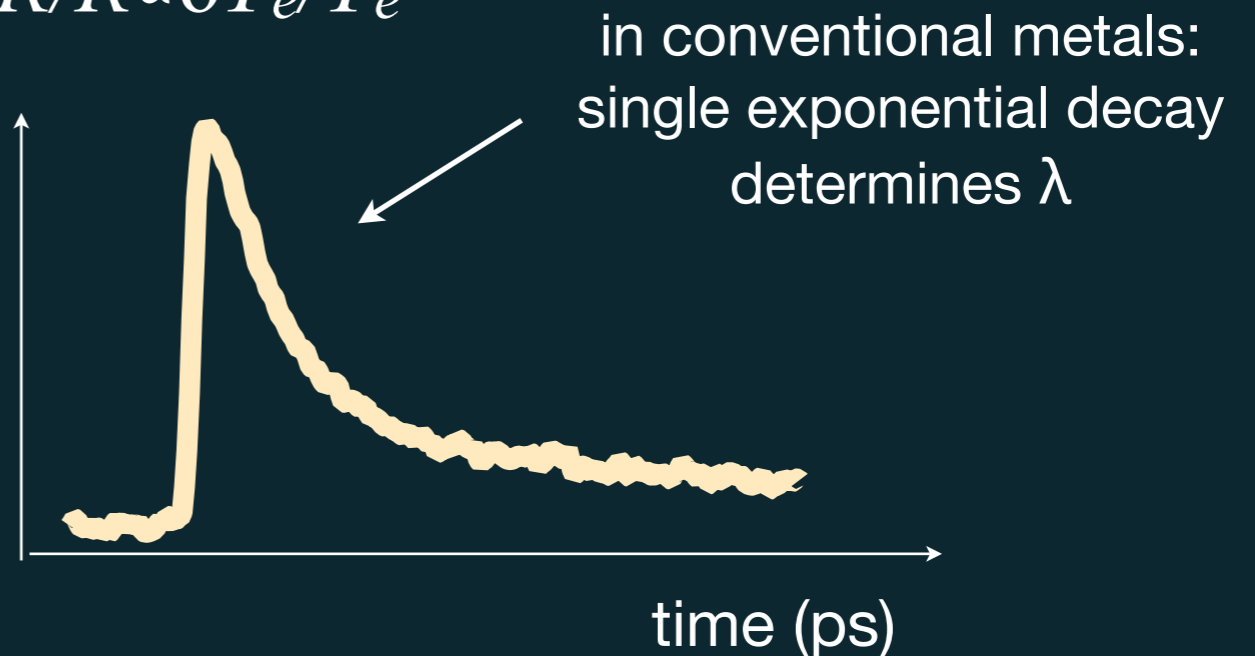
$$\frac{\partial T_{lat}}{\partial t} = -\frac{G(\Pi_{lat}, T_{lat}, T_e)}{C_{lat}} \longrightarrow \text{determines the dynamics in the time domain}$$

$$G = \frac{3\gamma_e}{\pi \hbar k_B^2} \int_0^\infty d\Omega \Pi_{lat}(\Omega) [N(\Omega, T_{lat}) - N(\Omega, T_e)]$$

hierarchy in the dynamics

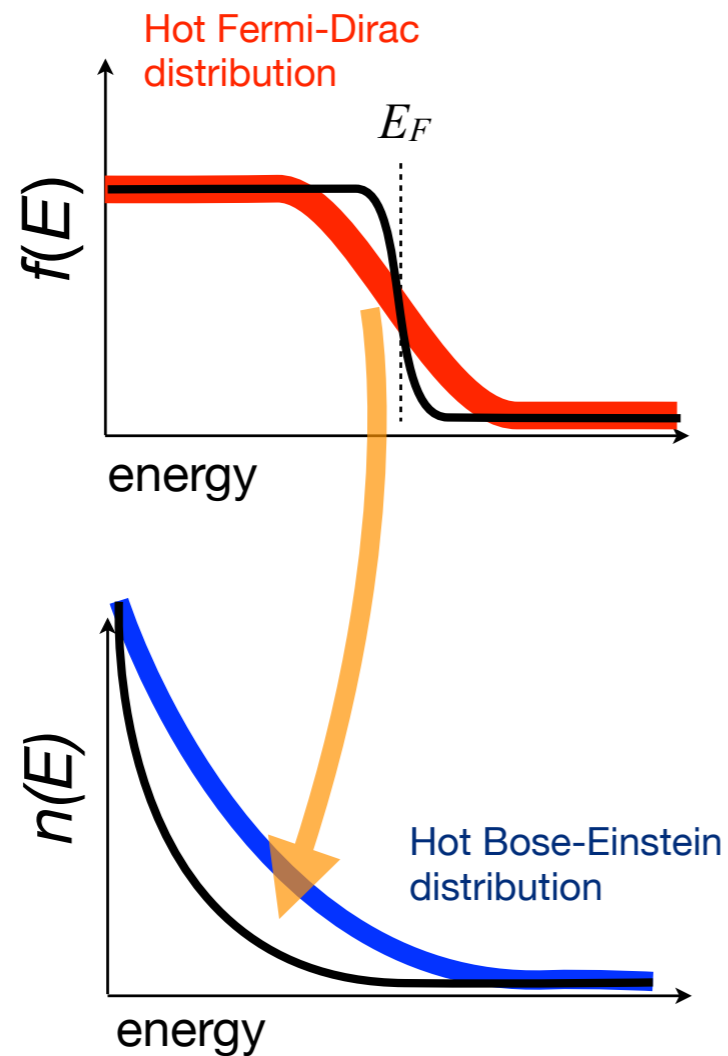


$$\delta R/R \propto \delta T_e/T_e$$

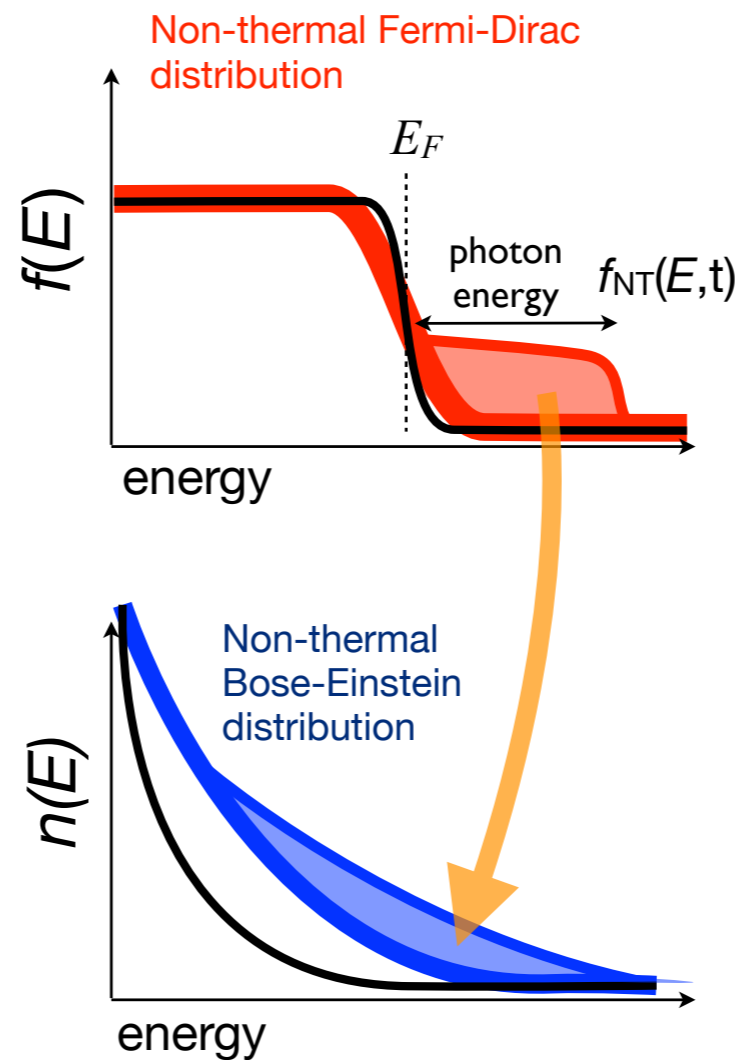


# be careful with the 2TM!

## effective temperature model



## extended effective temperature model

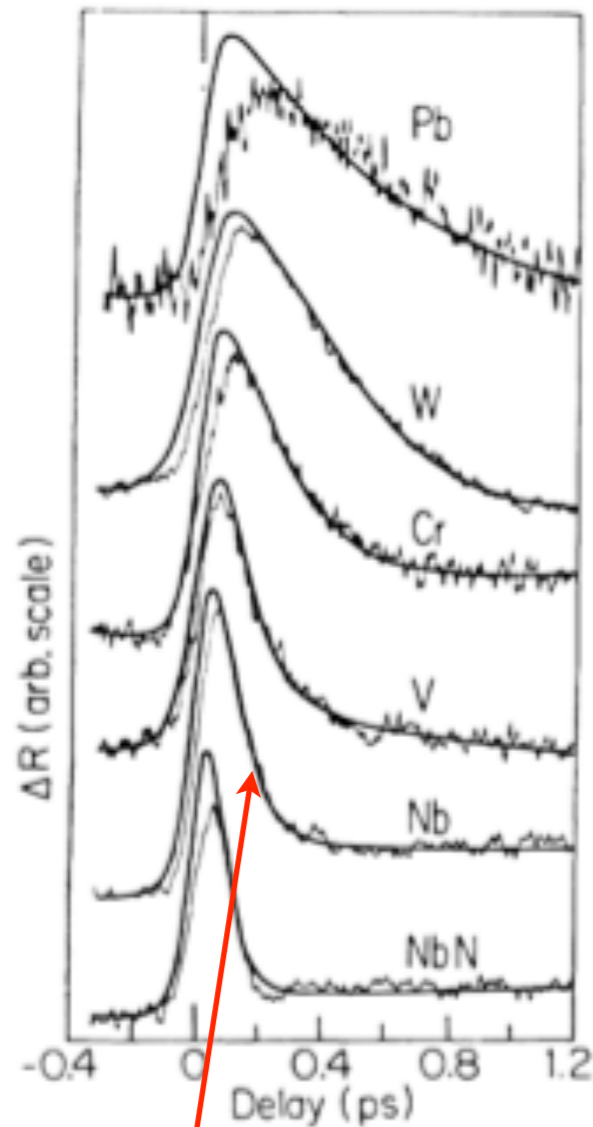


Fermi-liquid theory:  
the QP lifetime  
diverges at  $E_F$ !!

$$1/\tau_e \propto (E - E_F)^2$$

Boltzmann equations to account for  $f_{NT}(E,t)$

# non-equilibrium spectroscopy on metals



single exponential decay

	$T_e(0)$ (K) <sup>a</sup>	$\lambda_{\text{exp}}\langle\omega^2\rangle$ (meV <sup>2</sup> )	$\langle\omega^2\rangle$ (meV <sup>2</sup> )	$\lambda_{\text{exp}}$	$\lambda_{\text{lit}}$
Cu	590	$29 \pm 4$	$377^b$	$0.08 \pm 0.01$	$0.10^b$
Au	650	$23 \pm 4$	$178^c$	$0.13 \pm 0.02$	$0.15^c$
Cr	716	$128 \pm 15$	$987^d$	$0.13 \pm 0.02$	...
W	1200	$112 \pm 15$	$425^e$	$0.26 \pm 0.04$	$0.26^e$
V	700	$280 \pm 20$	$352^f$	$0.80 \pm 0.06$	$0.82^f$
Nb	790	$320 \pm 30$	$275^g$	$1.16 \pm 0.11$	$1.04^g$
Ti	820	$350 \pm 30$	$601^g$	$0.58 \pm 0.05$	$0.54^g$
Pb	570	$45 \pm 5$	$31^i$	$1.45 \pm 0.16$	$1.55^i$
NbN	1070	$640 \pm 40$	$673^j$	$0.95 \pm 0.06$	$1.46^j$
V <sub>3</sub> Ga	1110	$370 \pm 60$	$448^k$	$0.83 \pm 0.13$	$1.12^k$

S.D. Brorson et al. *Phys. Rev. Lett.* **64**, 2172 (1990)

$$\tau_{e-lat} = \frac{\pi k_B^2 T_e}{3\hbar\lambda\langle\Omega^2\rangle}$$

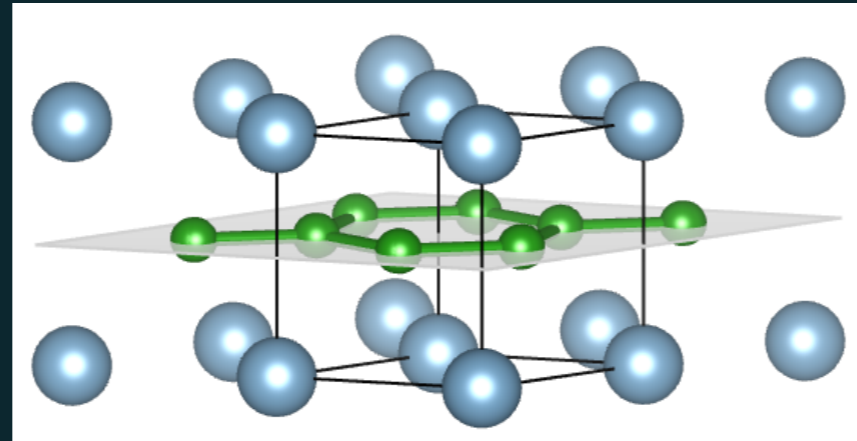
Eliashberg coupling  $\lambda = 2 \int_0^\infty \alpha^2 F(\Omega) / \Omega d\Omega$



# electron-phonon coupling in superconductors

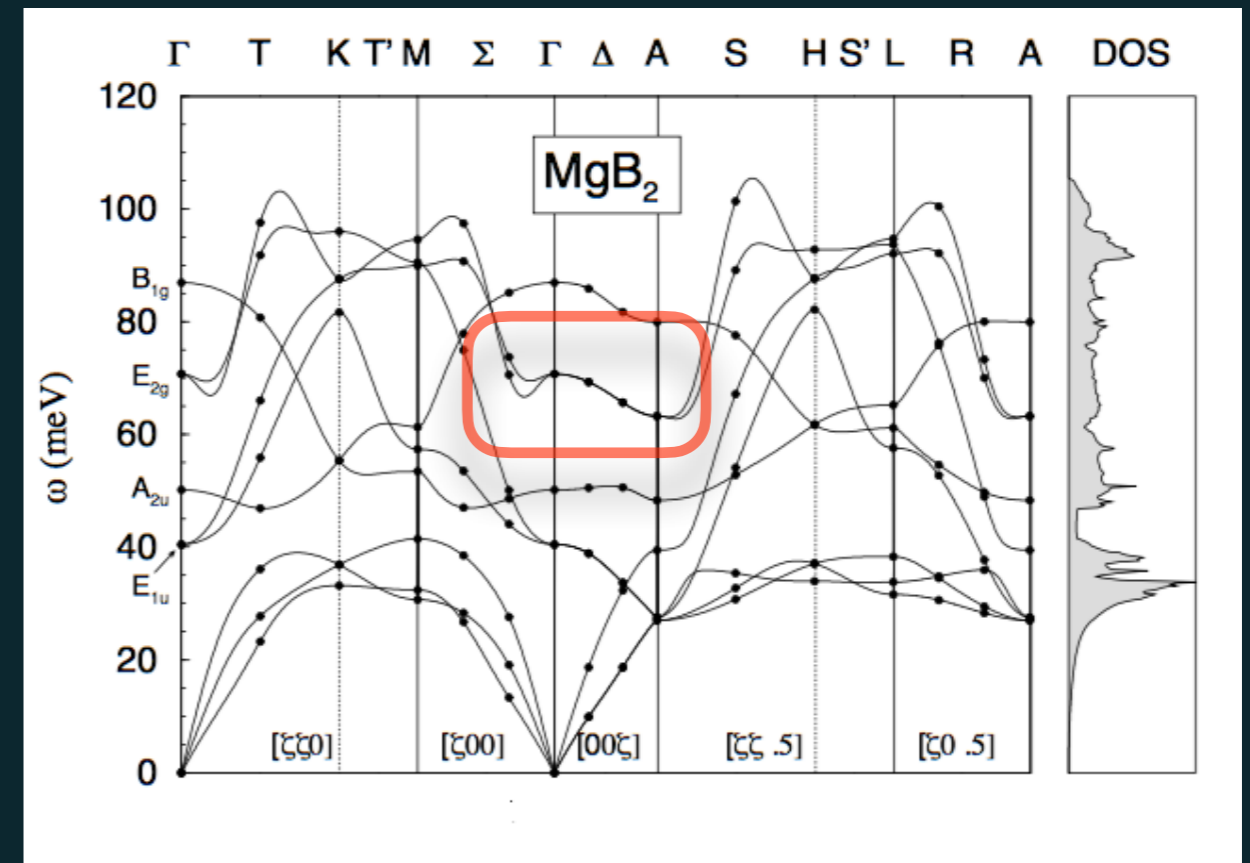
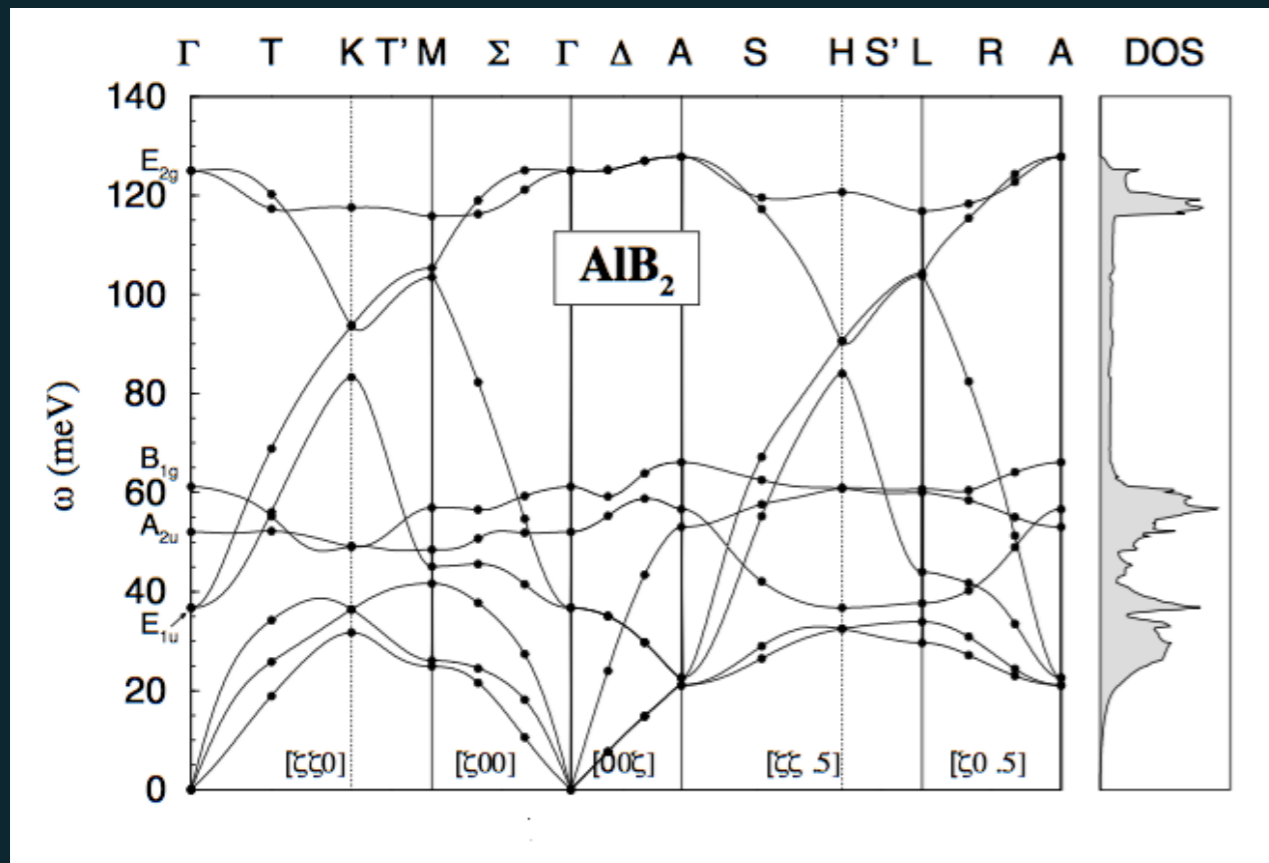
the paradigmatic example of the isostructural  $\text{MgB}_2$  and  $\text{AlB}_2$

Mg/Al



boron planes

very similar phonon dispersion

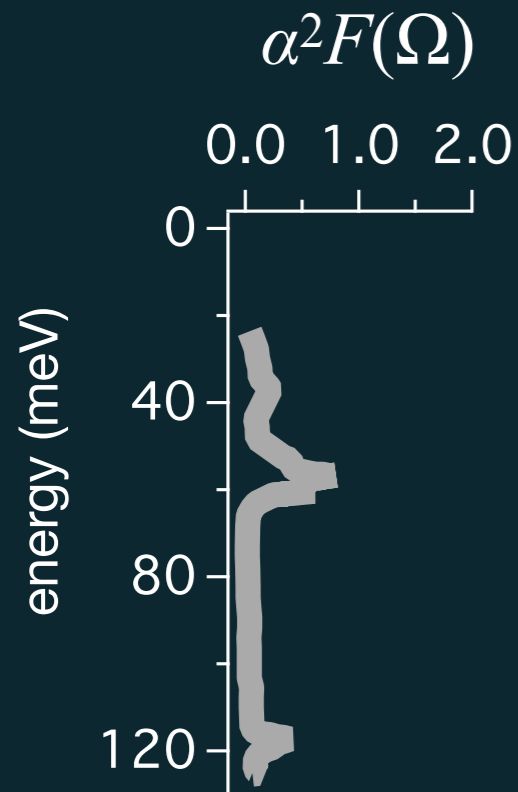


K.-P. Bohnen et al. Phys. Rev. Lett. **86**, 5771 (2001)

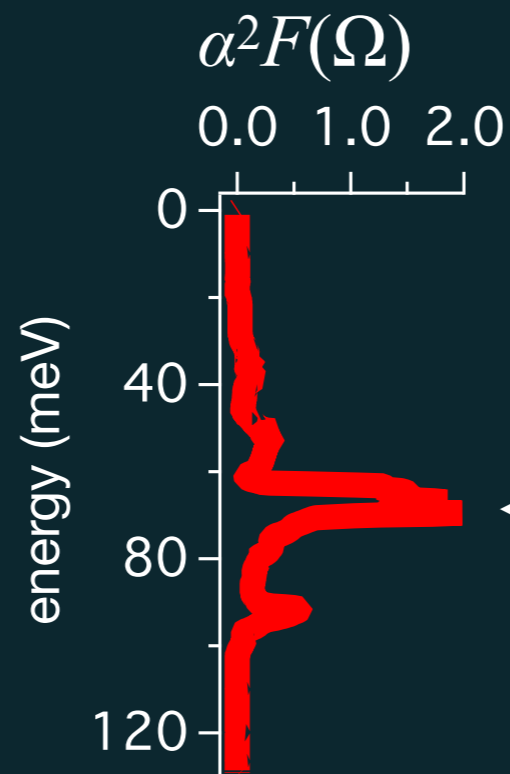
Y. Kong et al. Phys. Rev. B **64**, 020501R (2001)

# electron-phonon coupling in superconductors

$\text{AlB}_2$  (non superconducting)



$\text{MgB}_2$  ( $T_c=39$  K)



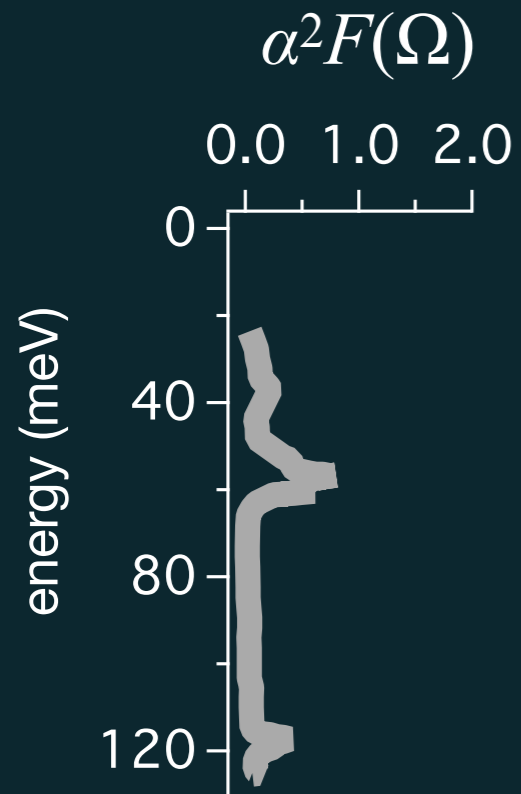
$$E_{\text{ph}}=73 \text{ meV} \Rightarrow \Delta t \approx 10 \text{ fs}$$

strong coupling with the optical  
bond-stretching  $E_{2g}$  mode

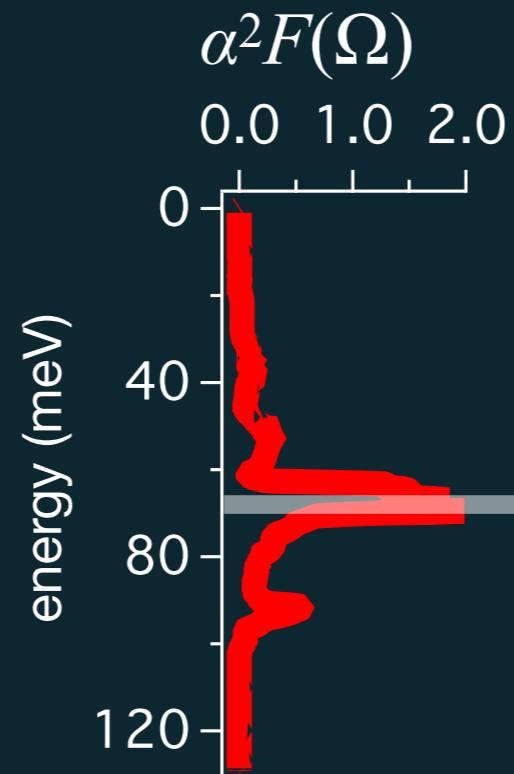
K.-P. Bohnen et al. Phys. Rev. Lett. **86**, 5771 (2001)

# electron-phonon coupling in superconductors

$\text{AlB}_2$  (non superconducting)

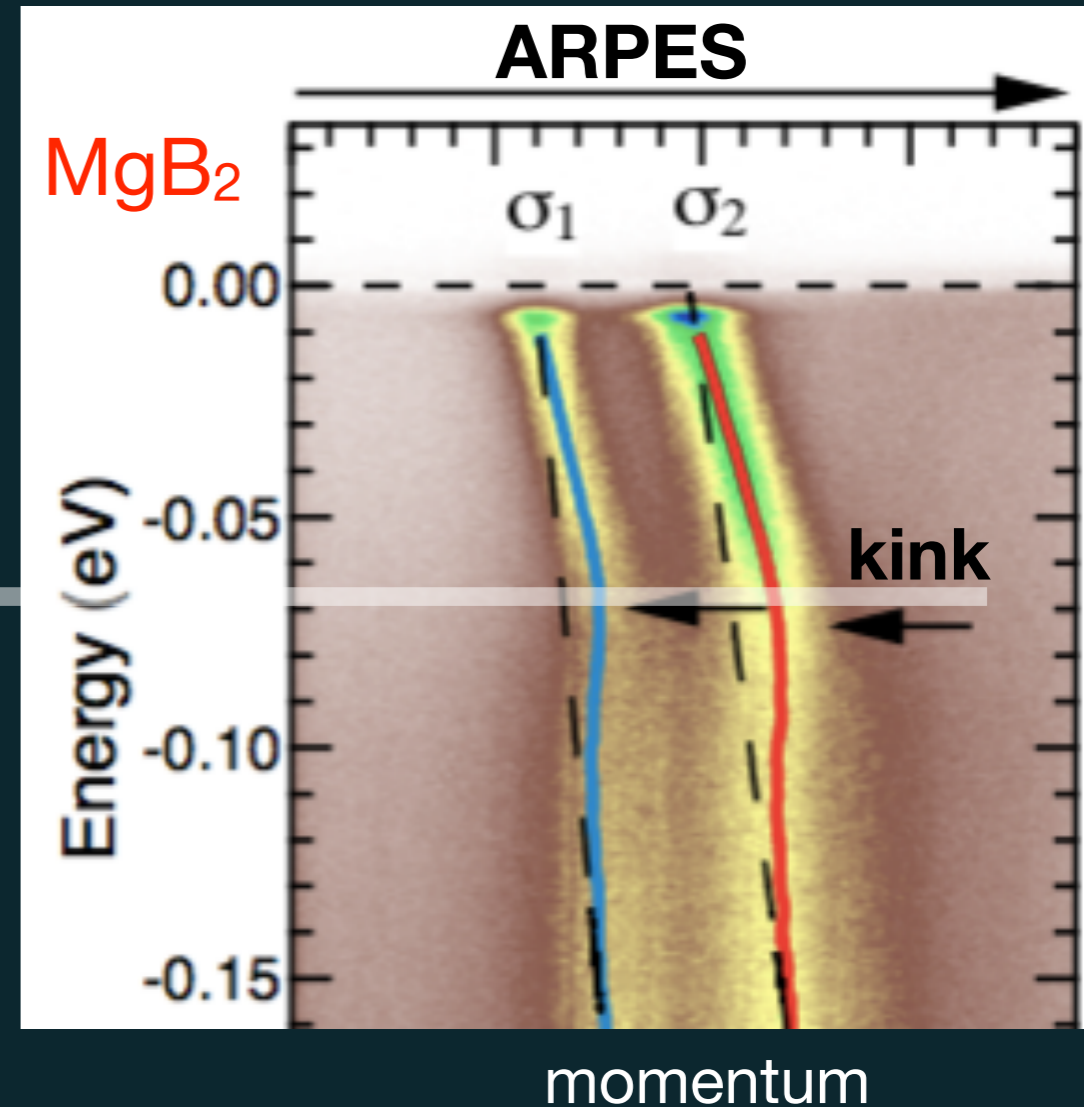


$\text{MgB}_2$  ( $T_c=39$  K)



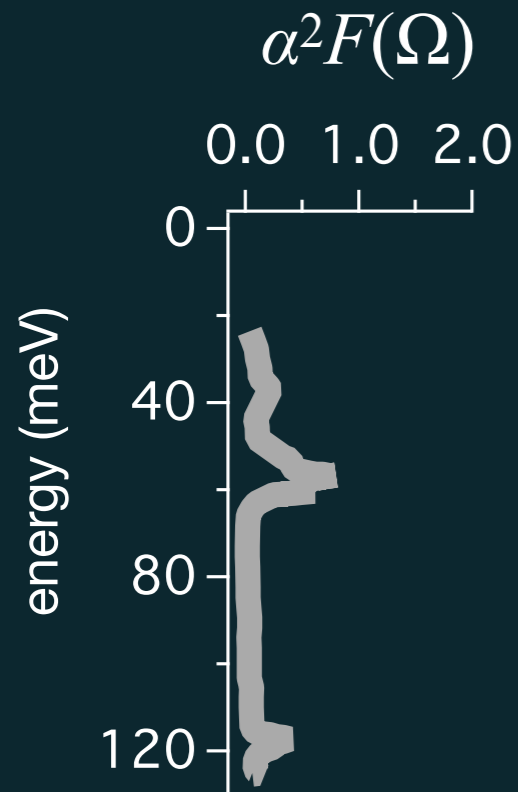
$\lambda \sim 0.62 \Rightarrow T_c \sim 40$  K

D. Mou et al. Phys. Rev. Lett. (2015)

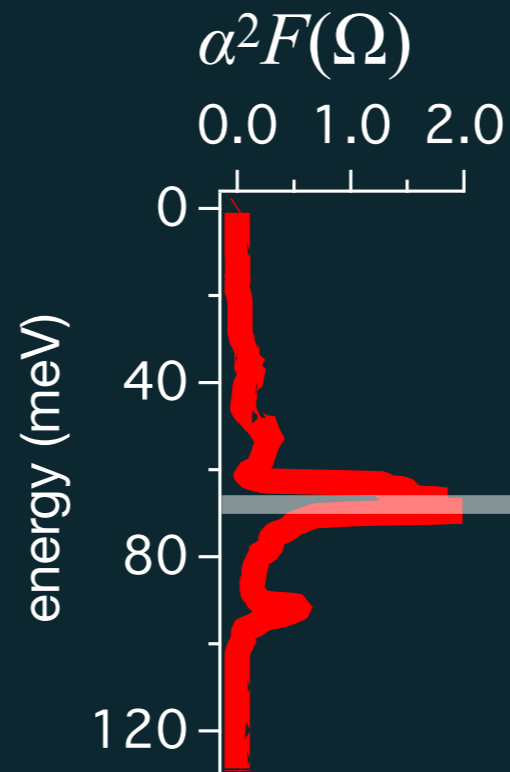


# electron-phonon coupling in superconductors

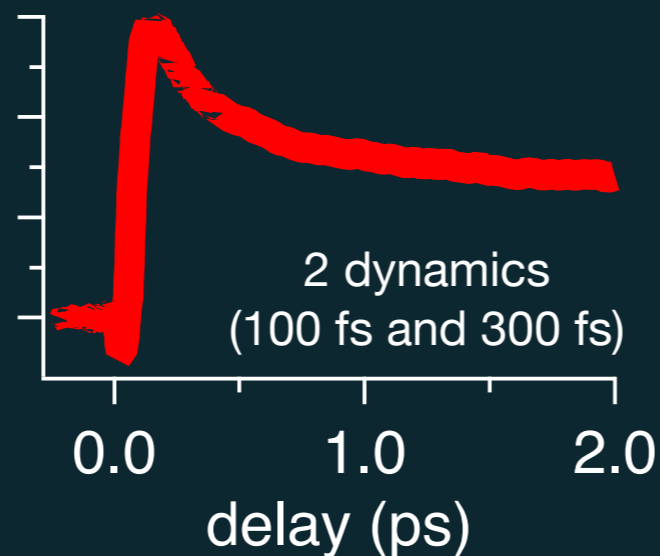
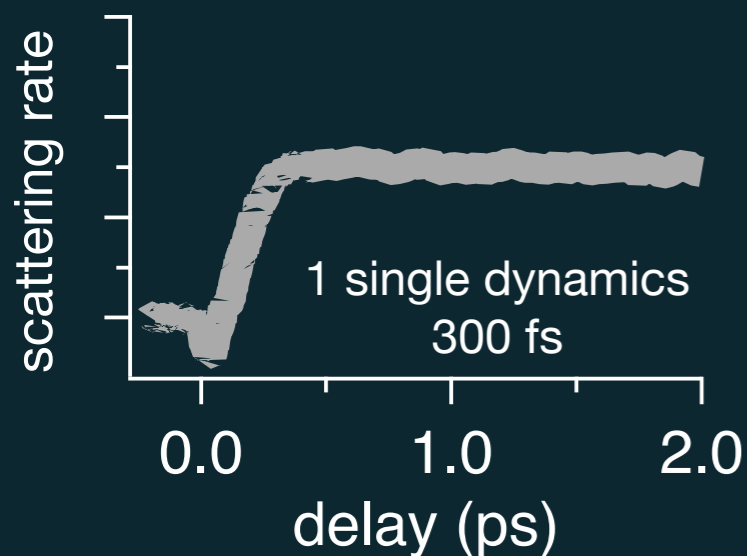
**AlB<sub>2</sub>** (non superconducting)



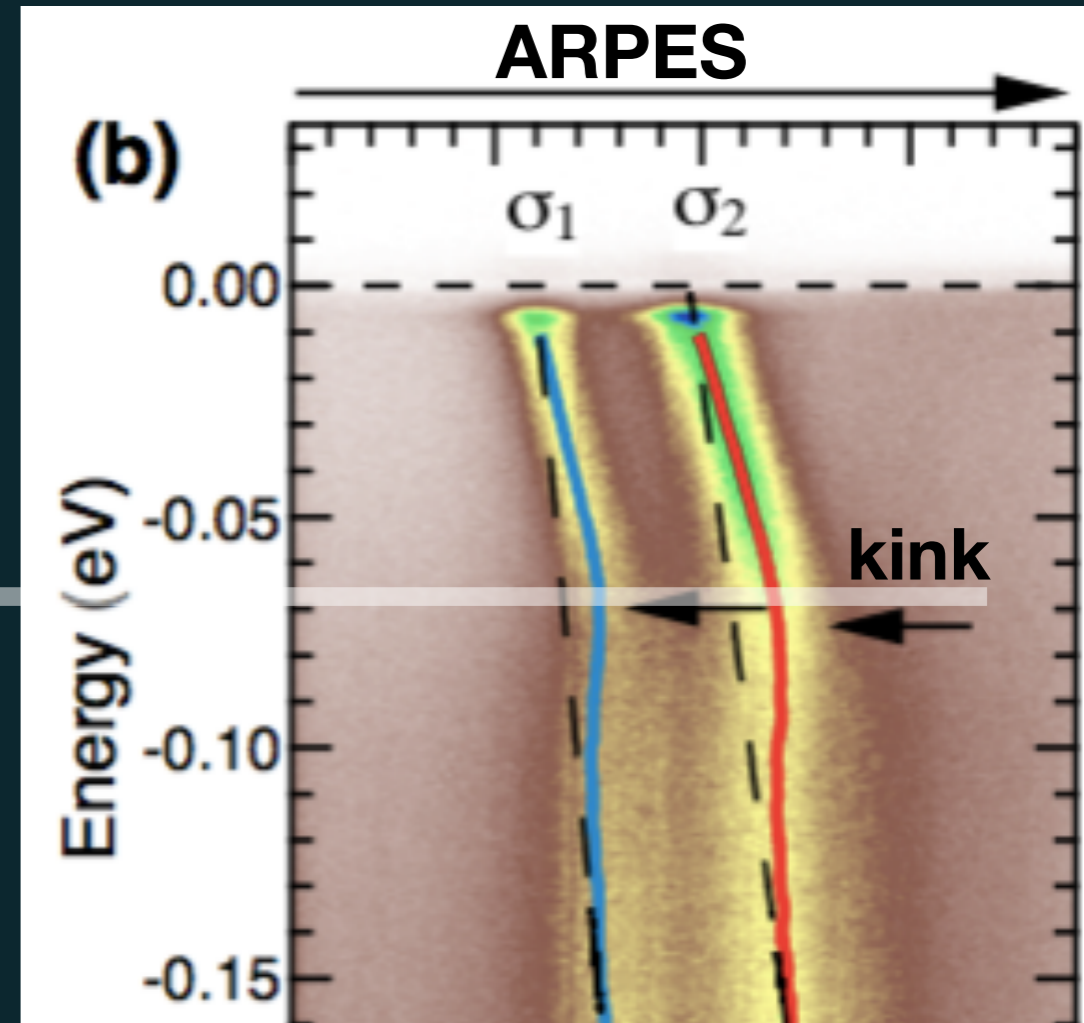
**MgB<sub>2</sub>** ( $T_c=39$  K)



in the time domain:



D. Mou et al. Phys. Rev. Lett. (2015)



the measured dynamics tracks  
the change of the boson  
temperature (density):

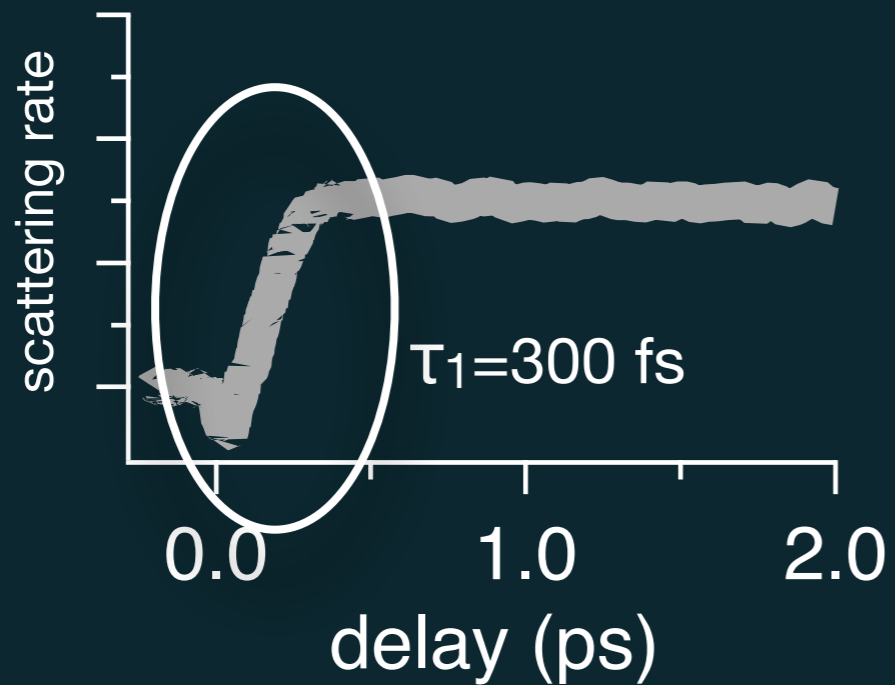
$$\delta R/R \propto \delta T_{\text{bos}}/T_{\text{bos}}$$

S. Dal Conte et al. *in preparation*

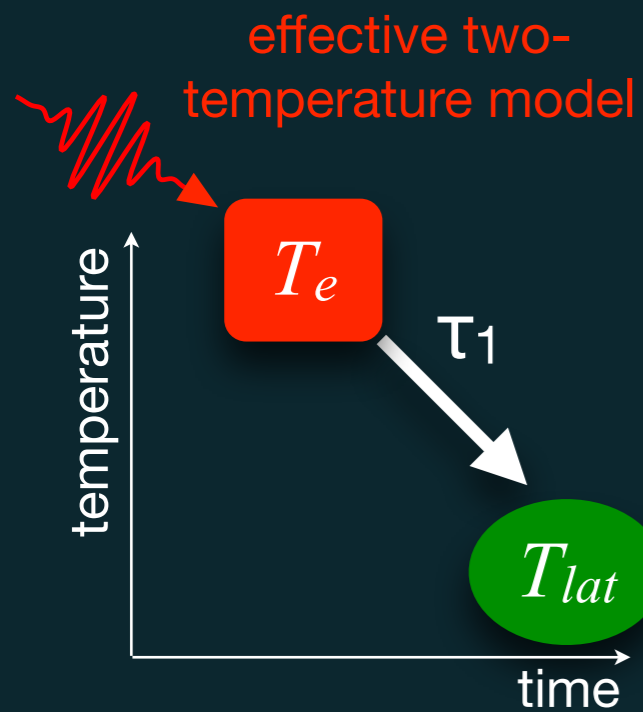
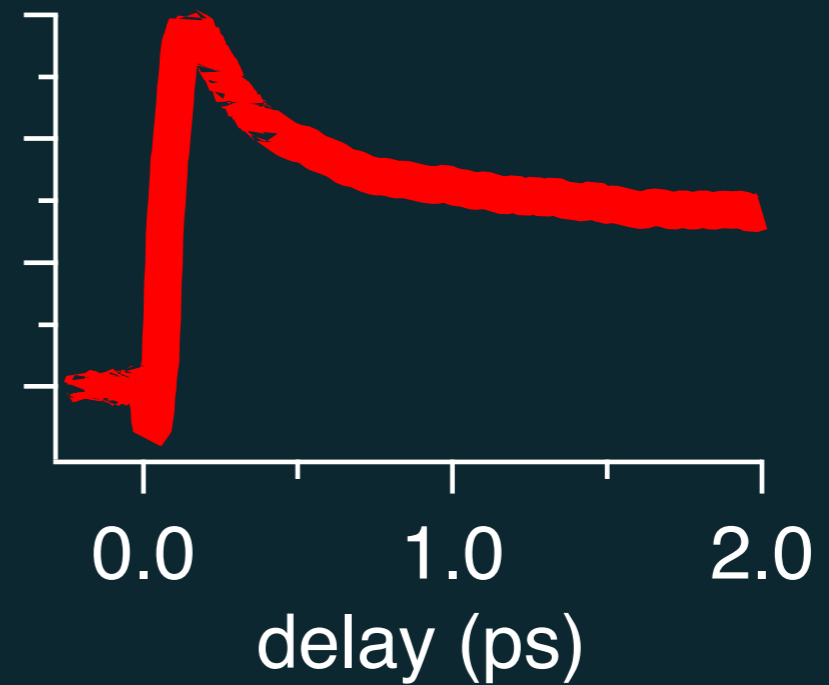


# electron-phonon coupling in superconductors

$\text{AlB}_2$  (non superconducting)

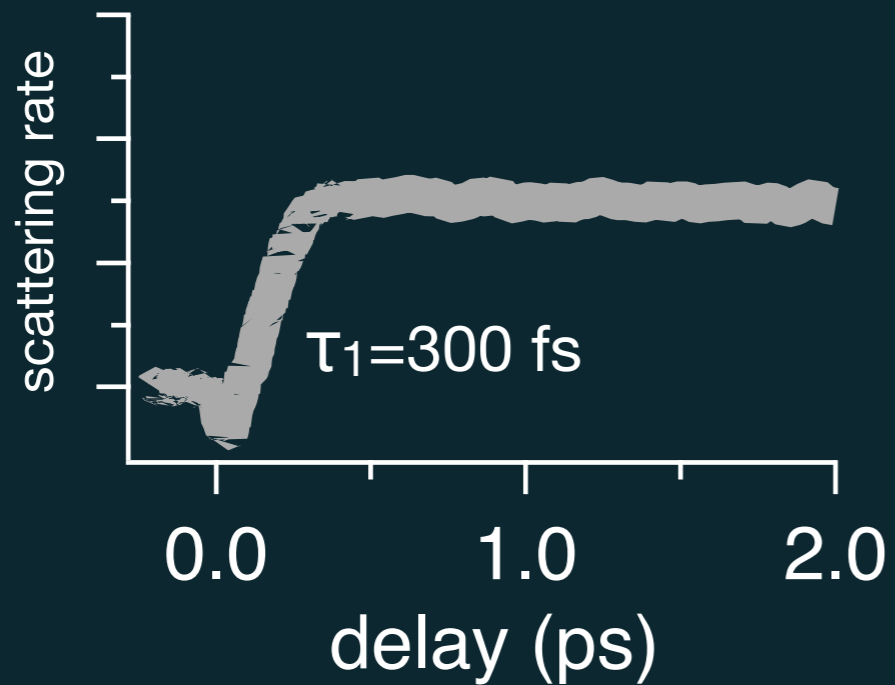


$\text{MgB}_2$  ( $T_c = 39$  K)

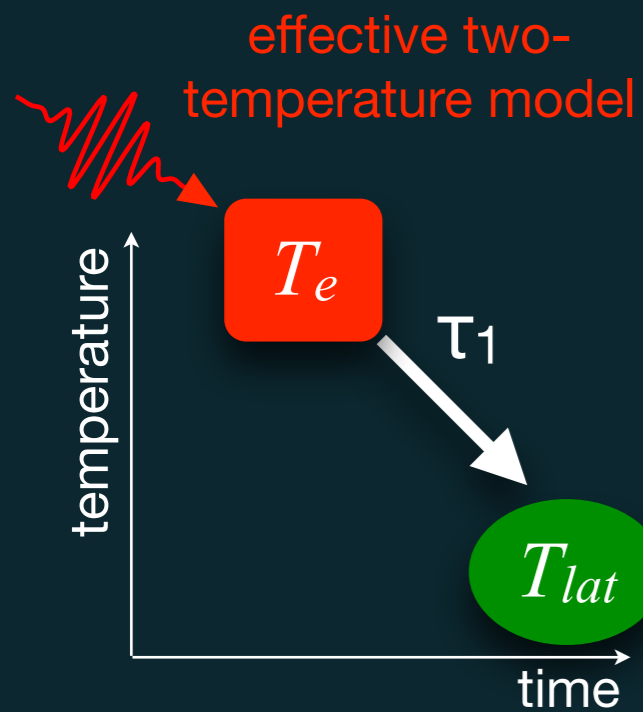
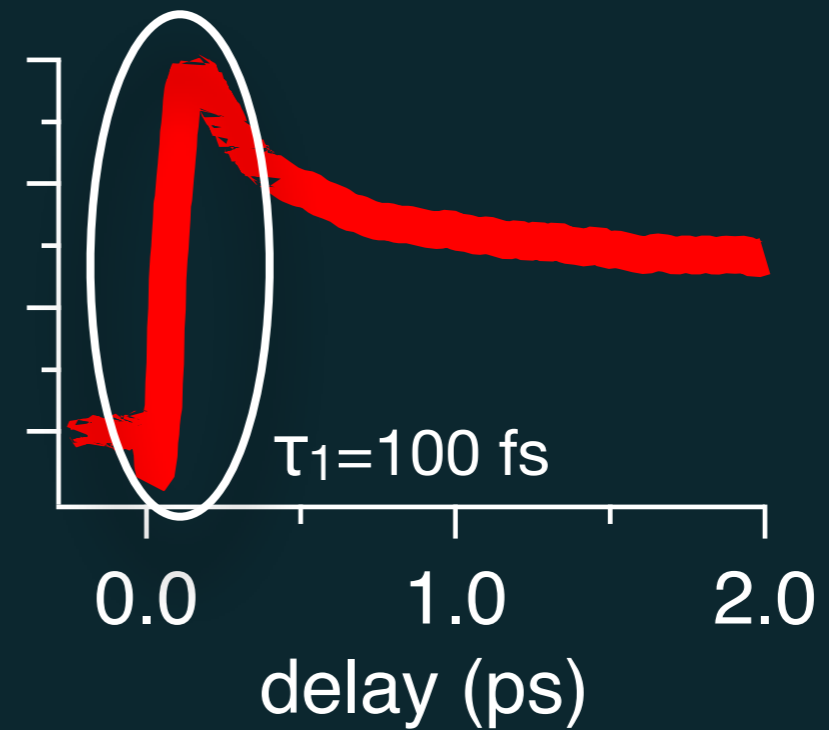


# electron-phonon coupling in superconductors

$\text{AlB}_2$  (non superconducting)

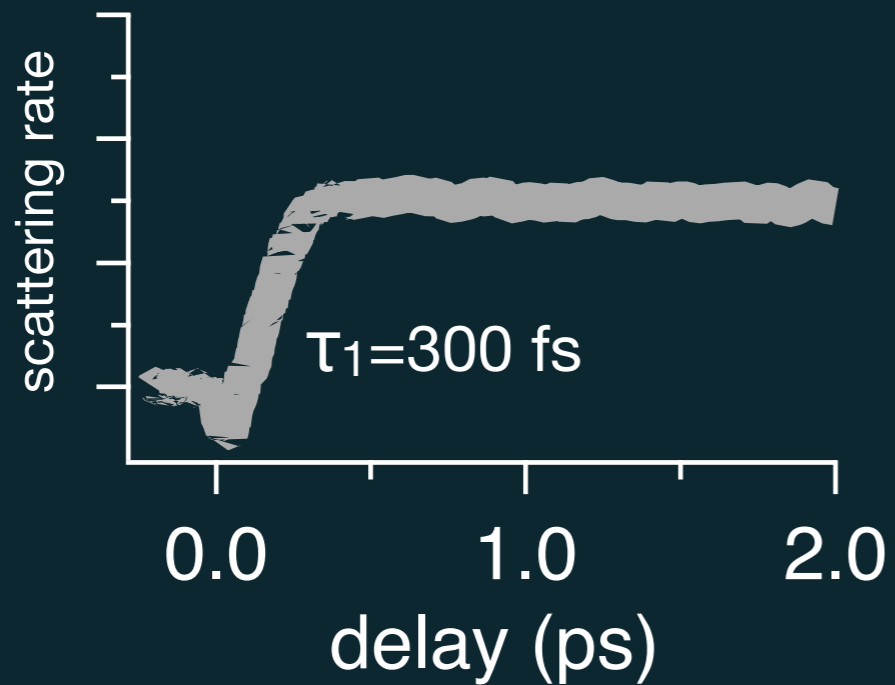


$\text{MgB}_2$  ( $T_c = 39$  K)

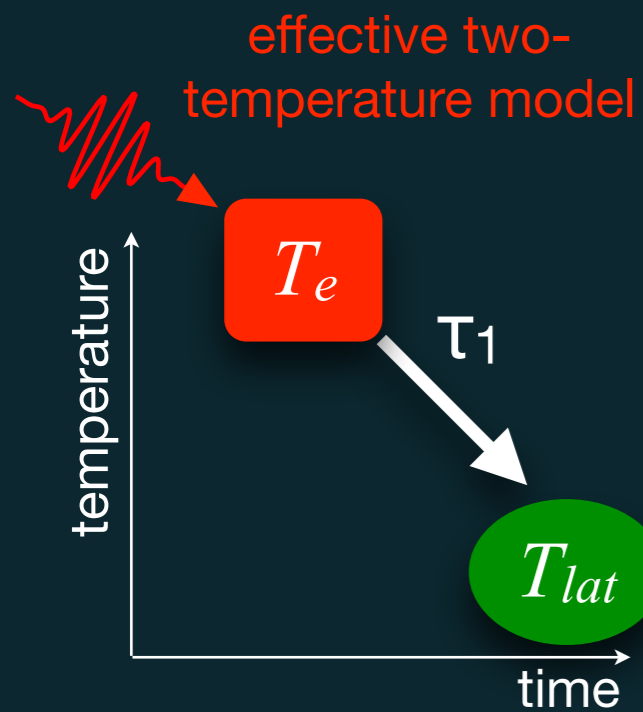
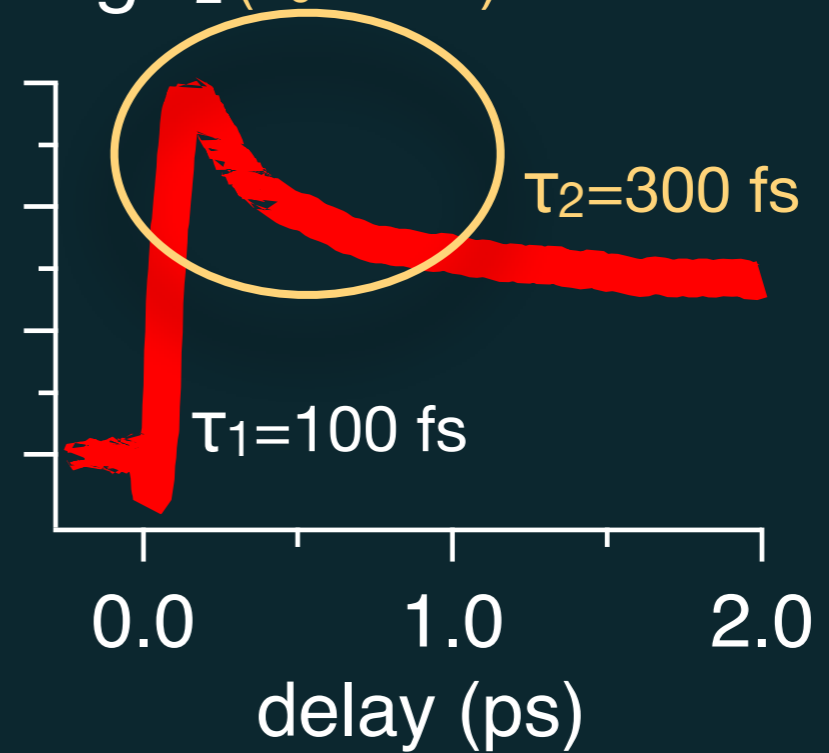


# electron-phonon coupling in superconductors

$\text{AlB}_2$  (non superconducting)

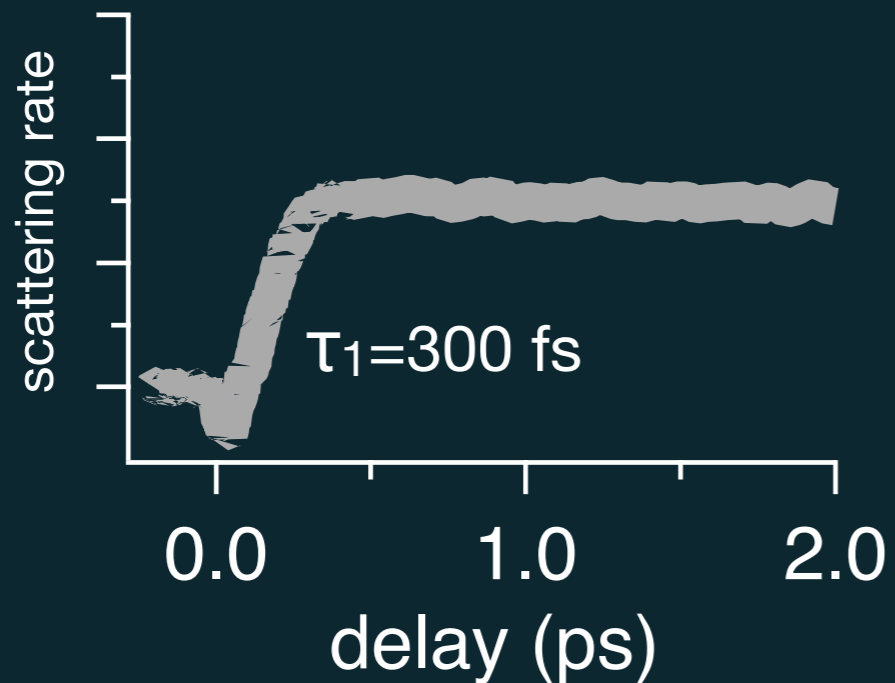


$\text{MgB}_2$  ( $T_c = 39$  K)

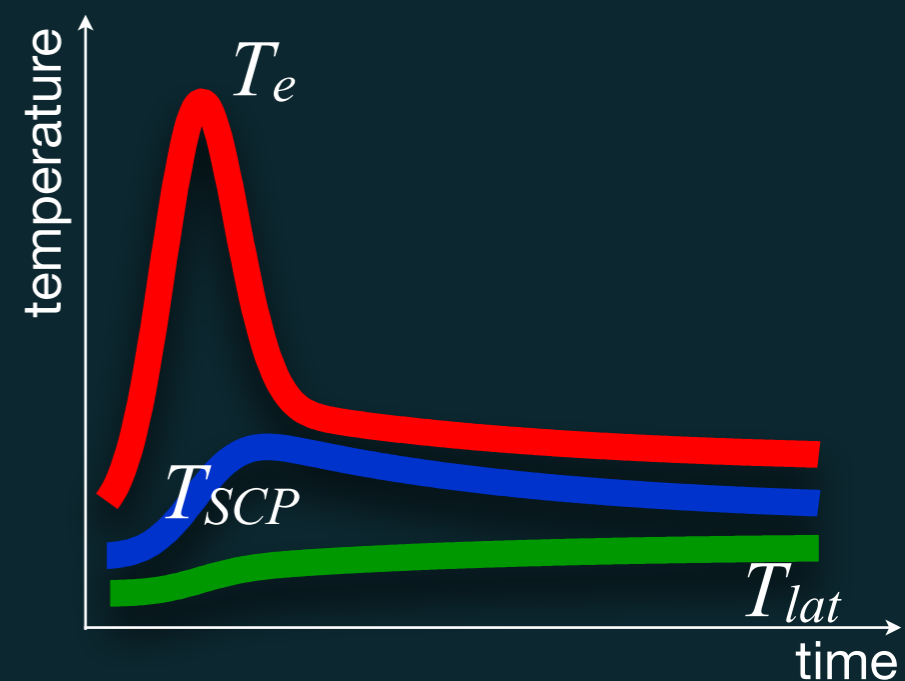
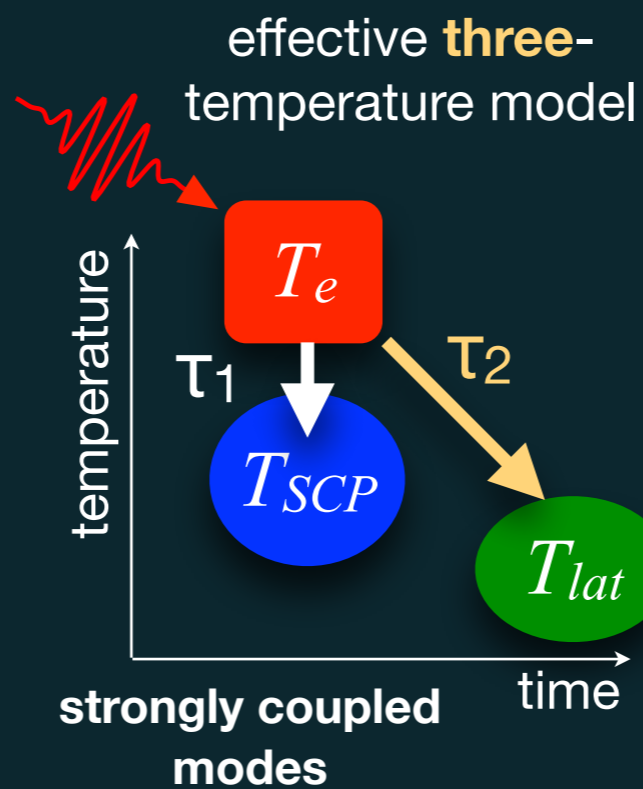
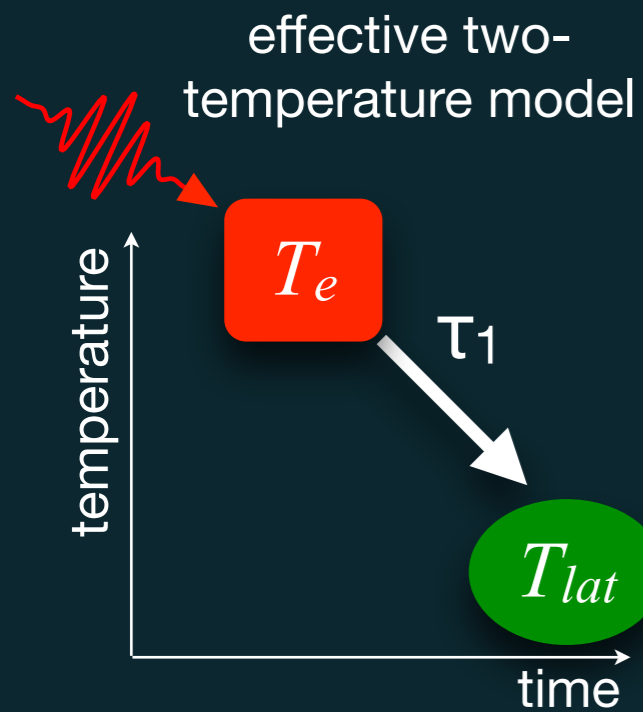
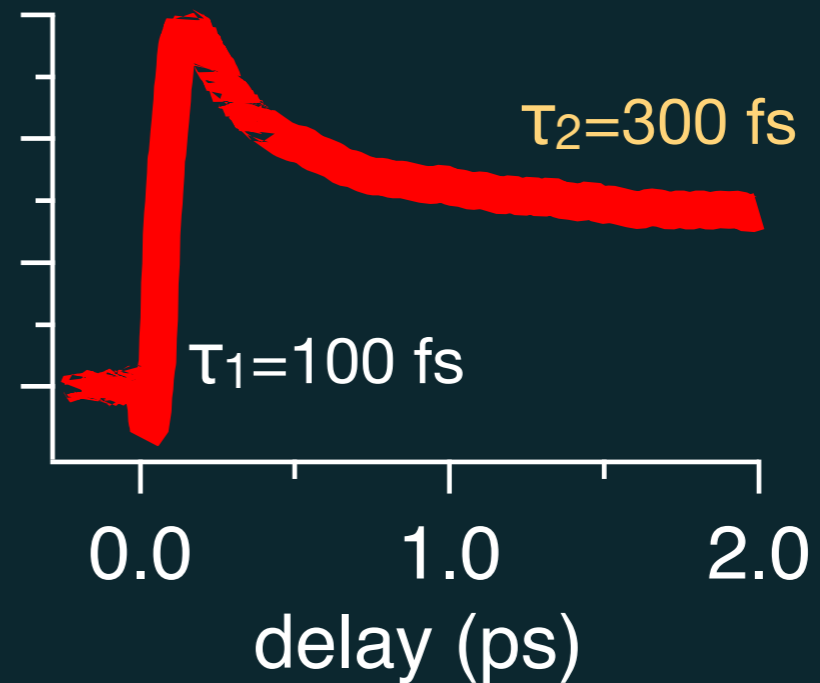


# electron-phonon coupling in superconductors

$\text{AlB}_2$  (non superconducting)



$\text{MgB}_2$  ( $T_c = 39$  K)

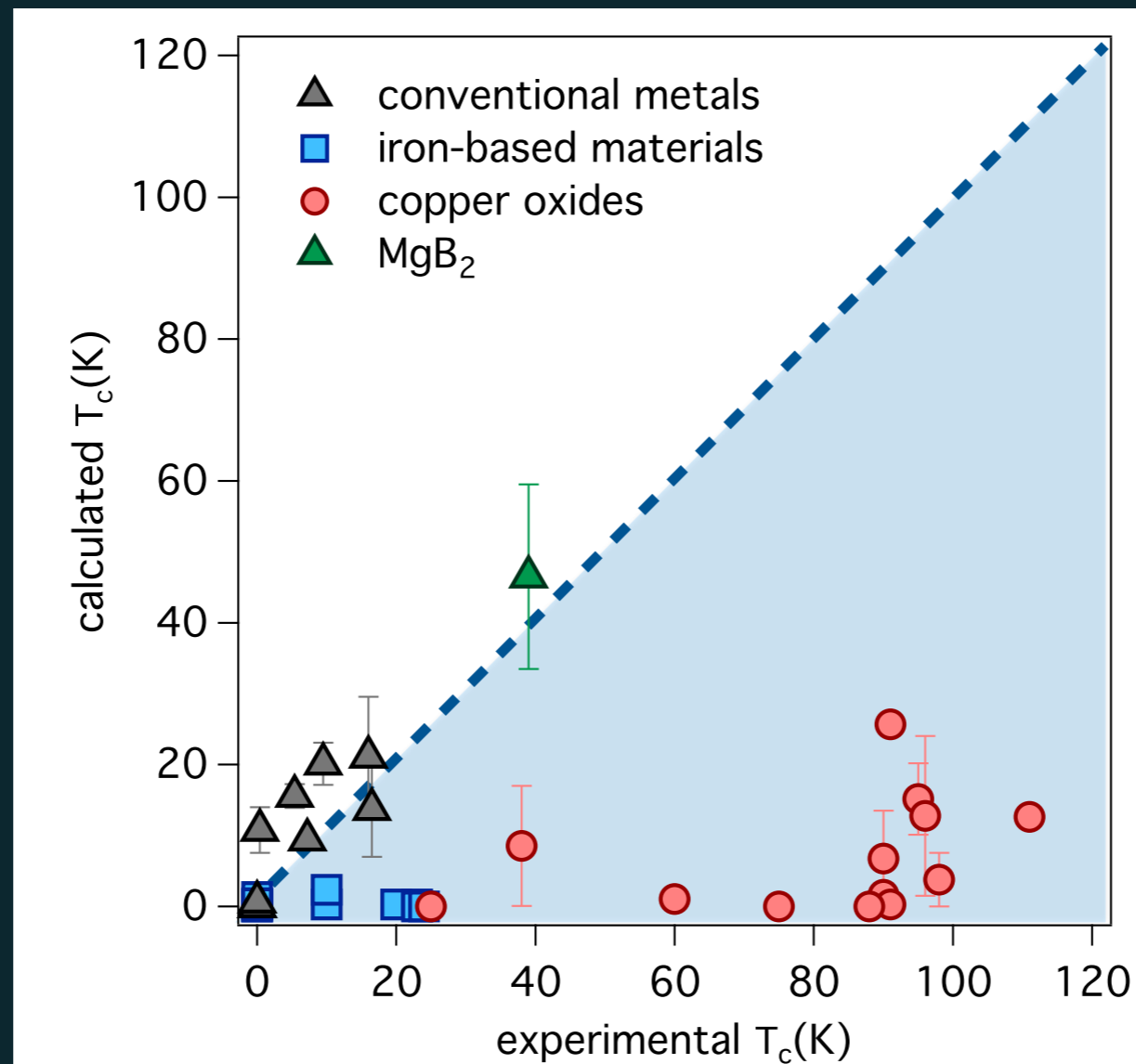




# electron-phonon coupling in unconventional superconductors

electron-phonon coupling  
 $\lambda$  obtained from time-  
 resolved techniques

C. Giannetti et al. Advances in  
 Physics **65**:2, 58-238 (2016)



**CUPRATES:**  
*some glue is missing!!!*

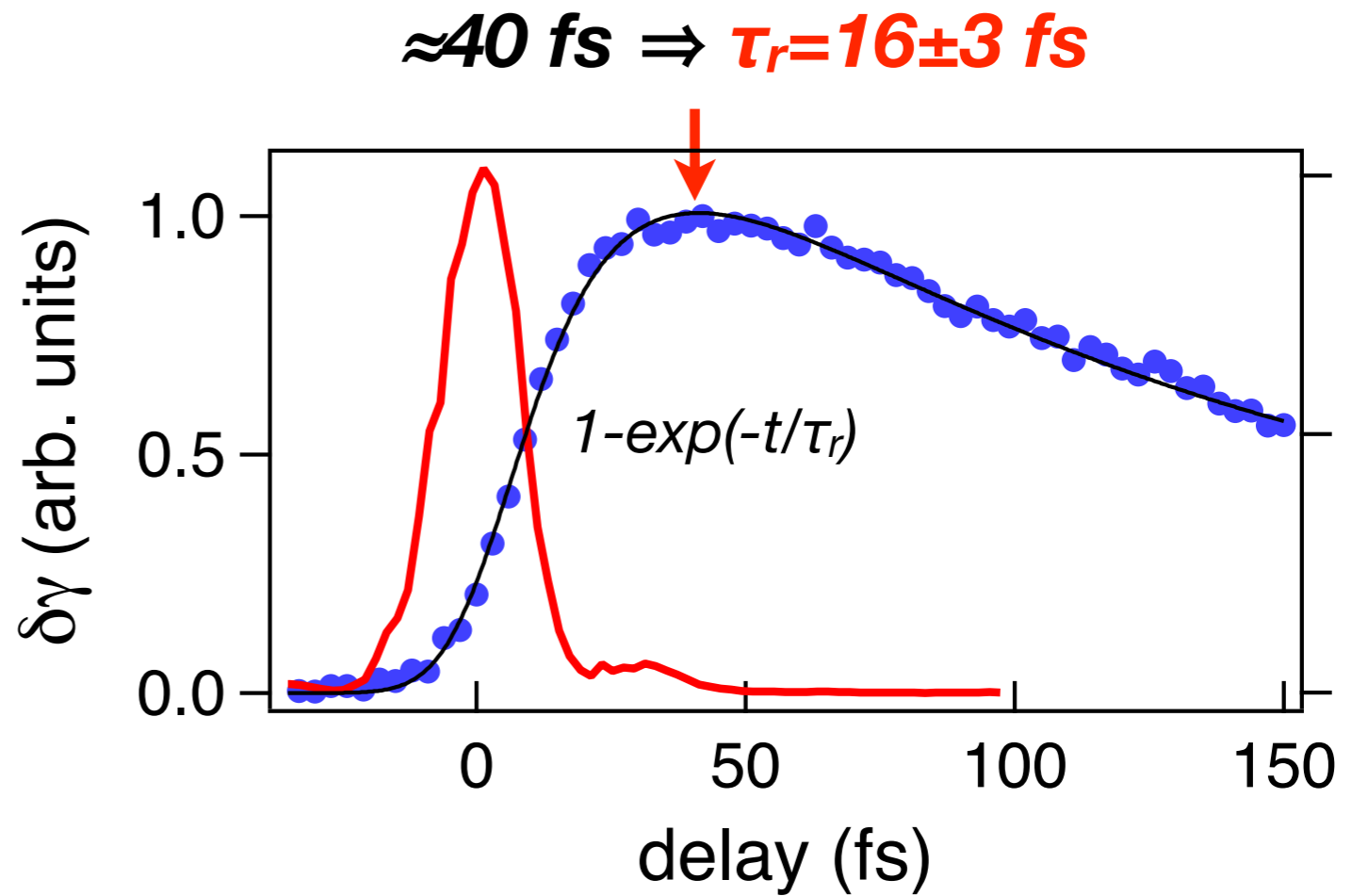
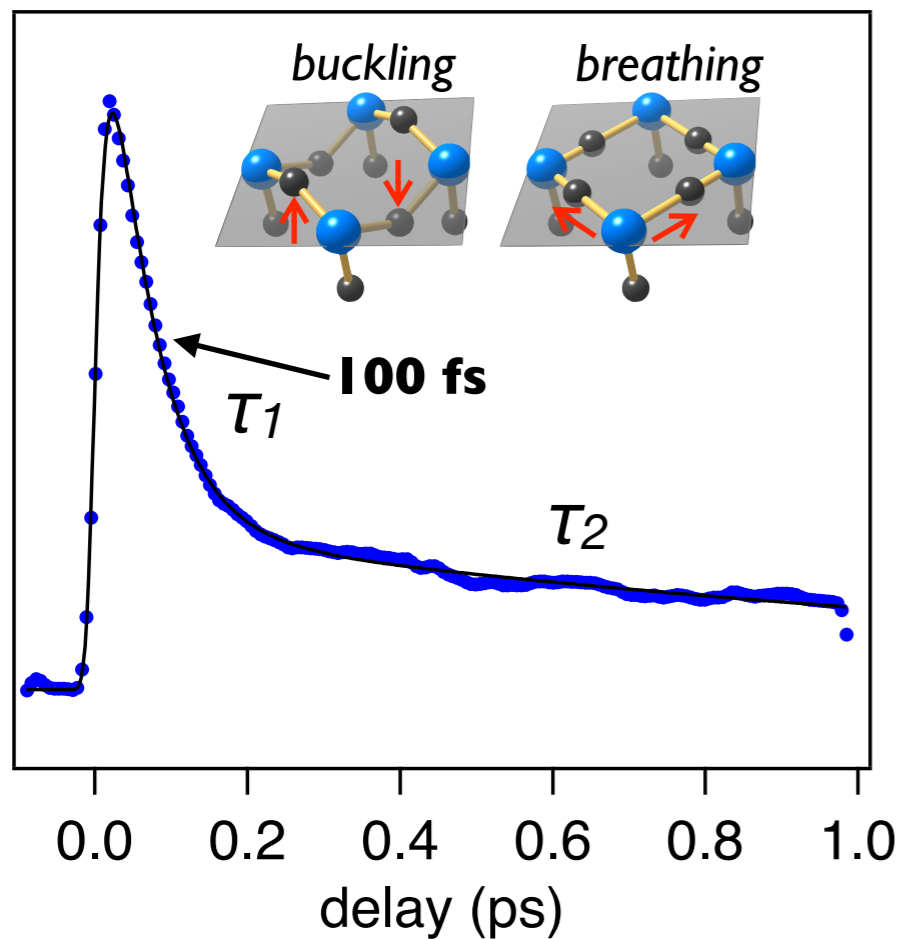
$\tilde{\Omega}$ : frequency log-average

$\mu^*$ : effective Coulomb repulsion  
 $g < 1$  for d-wave superconductors

$$T_c = 0.83\tilde{\Omega}e^{-\frac{1.04(1+\lambda)}{g(\lambda-\mu^*(1+0.62\lambda))}}$$

# non-equilibrium spectroscopy on cuprates

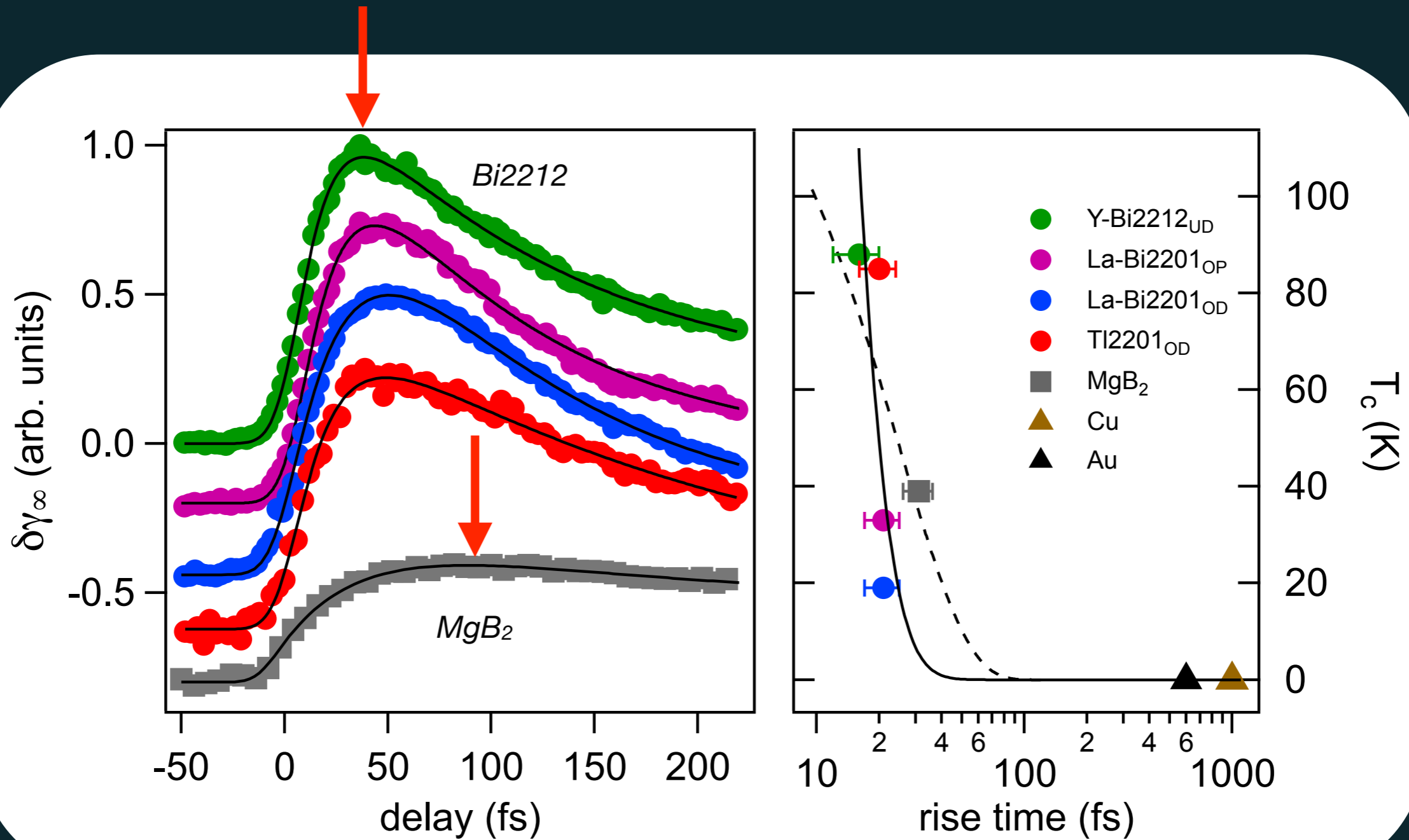
## superconducting copper oxides



$$\delta R(\omega) \propto \delta T_b$$

3 different electronic timescales!

# comparing to conventional superconductors ( $\text{MgB}_2$ )

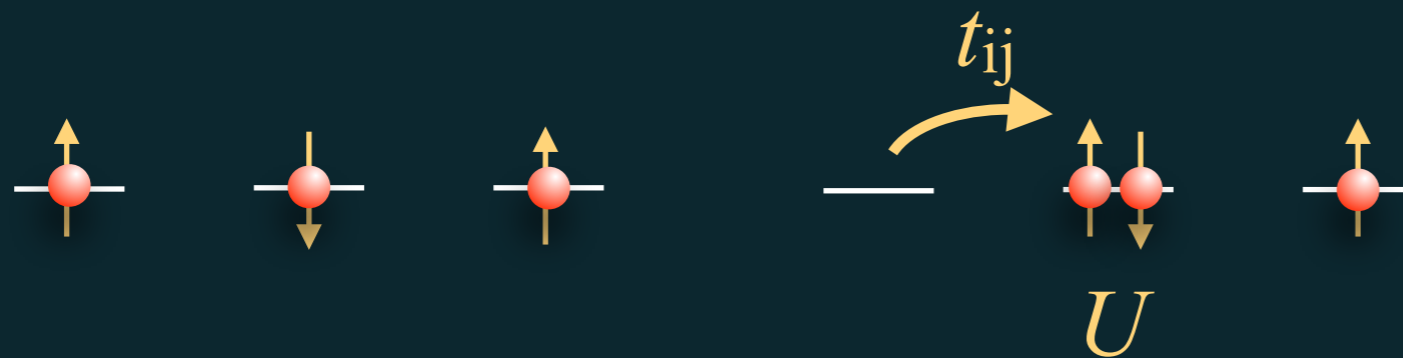


$$T_c = 0.83\tilde{\Omega}e^{-\frac{1.04(1+\lambda)}{g(\lambda-\mu^*(1+0.62\lambda))}}$$

# Hubbard model

single-band Hubbard model

$$\hat{H} = - \sum_{i,j,\sigma} (t_{ij} \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + c.c.) + U \sum_i \hat{n}_{i,\uparrow} \hat{n}_{i,\downarrow} - \mu \sum_i \hat{n}_i$$



2<sup>nd</sup> order  $J=4t^2/U$   
energy gain for  
antiferromagnetic ground  
state

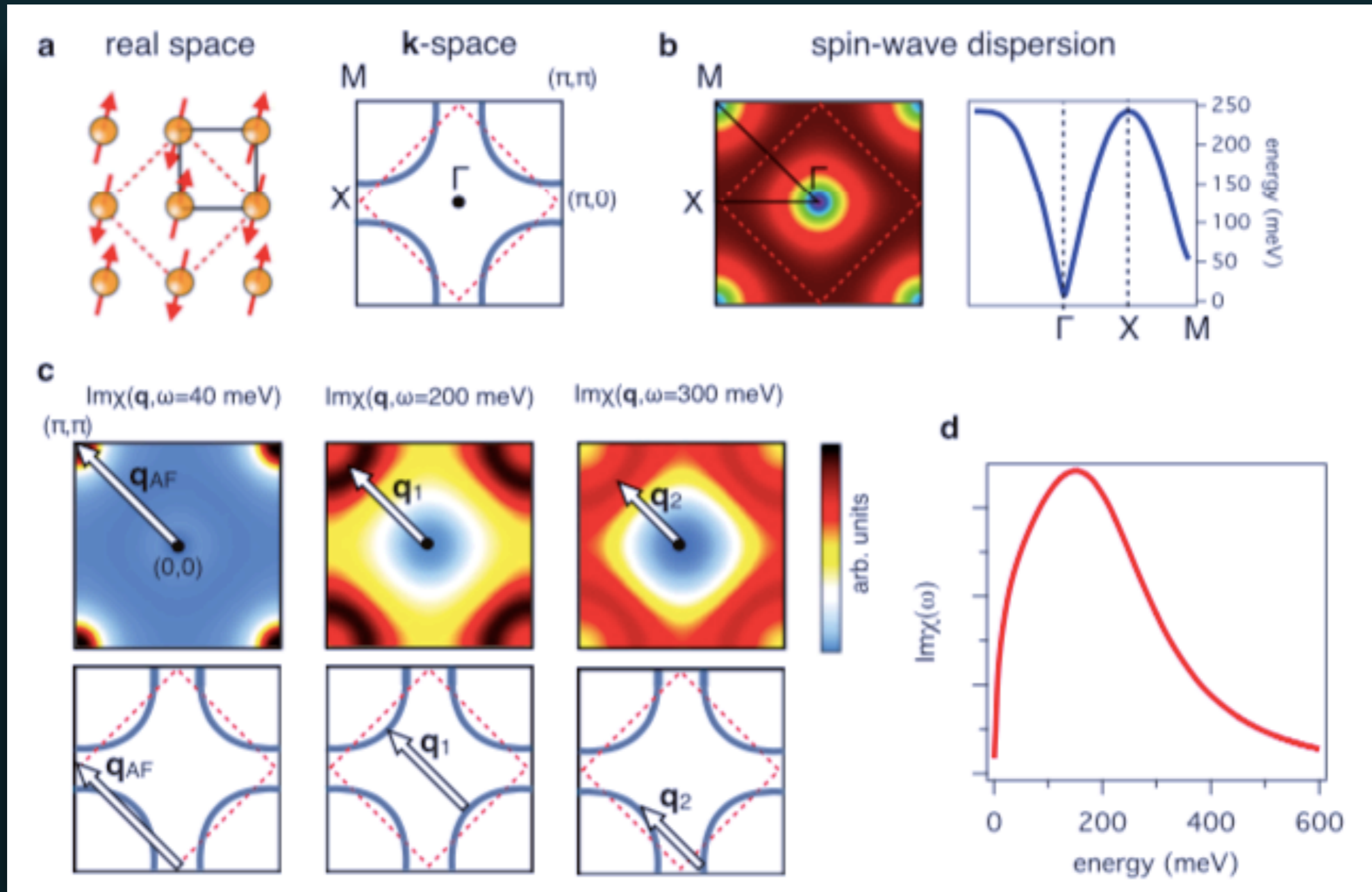
$t$ - $J$  model in the  $U \rightarrow \infty$  limit

$$H = -t_h \sum_{\langle \mathbf{l}\mathbf{j} \rangle, \sigma} (c_{\mathbf{l},\sigma}^\dagger \tilde{c}_{\mathbf{j},\sigma} + \text{h.c.}) + J \sum_{\langle \mathbf{l}\mathbf{j} \rangle} \mathbf{S}_{\mathbf{l}} \cdot \mathbf{S}_{\mathbf{j}}$$

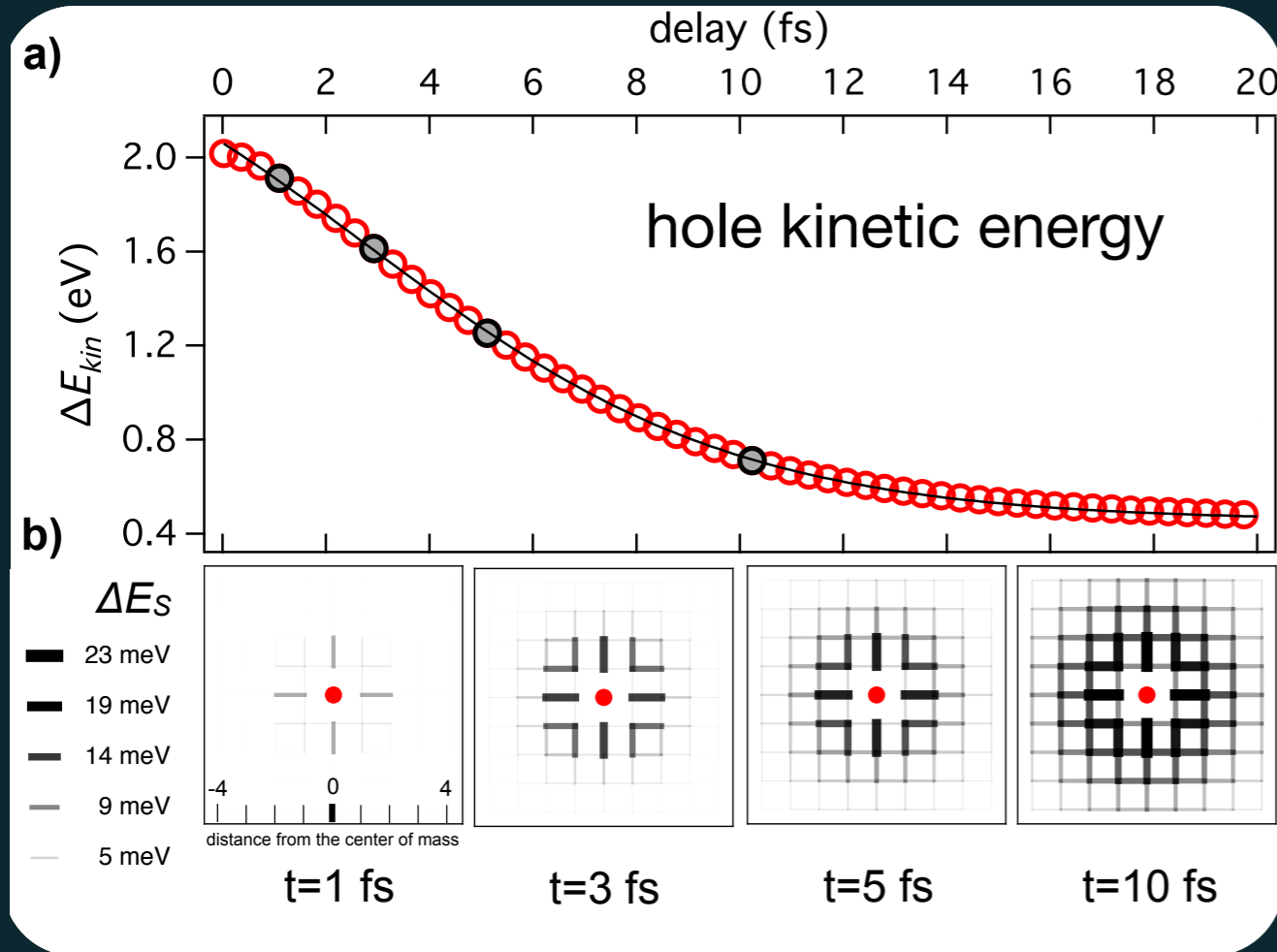
$$t=360 \text{ meV} \quad J=120 \text{ meV}$$



# Magnetic excitations in the copper sublattice

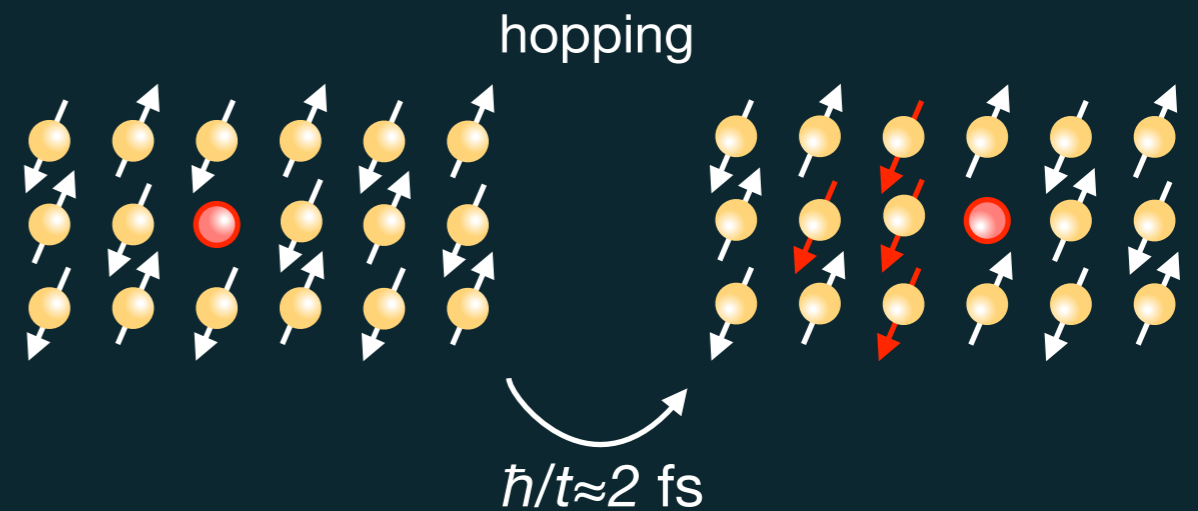


# non-equilibrium t-J model



Ultrafast coupling to high-energy short-range AF spin fluctuations:

- 10 fs coupling to short-range AF spin background



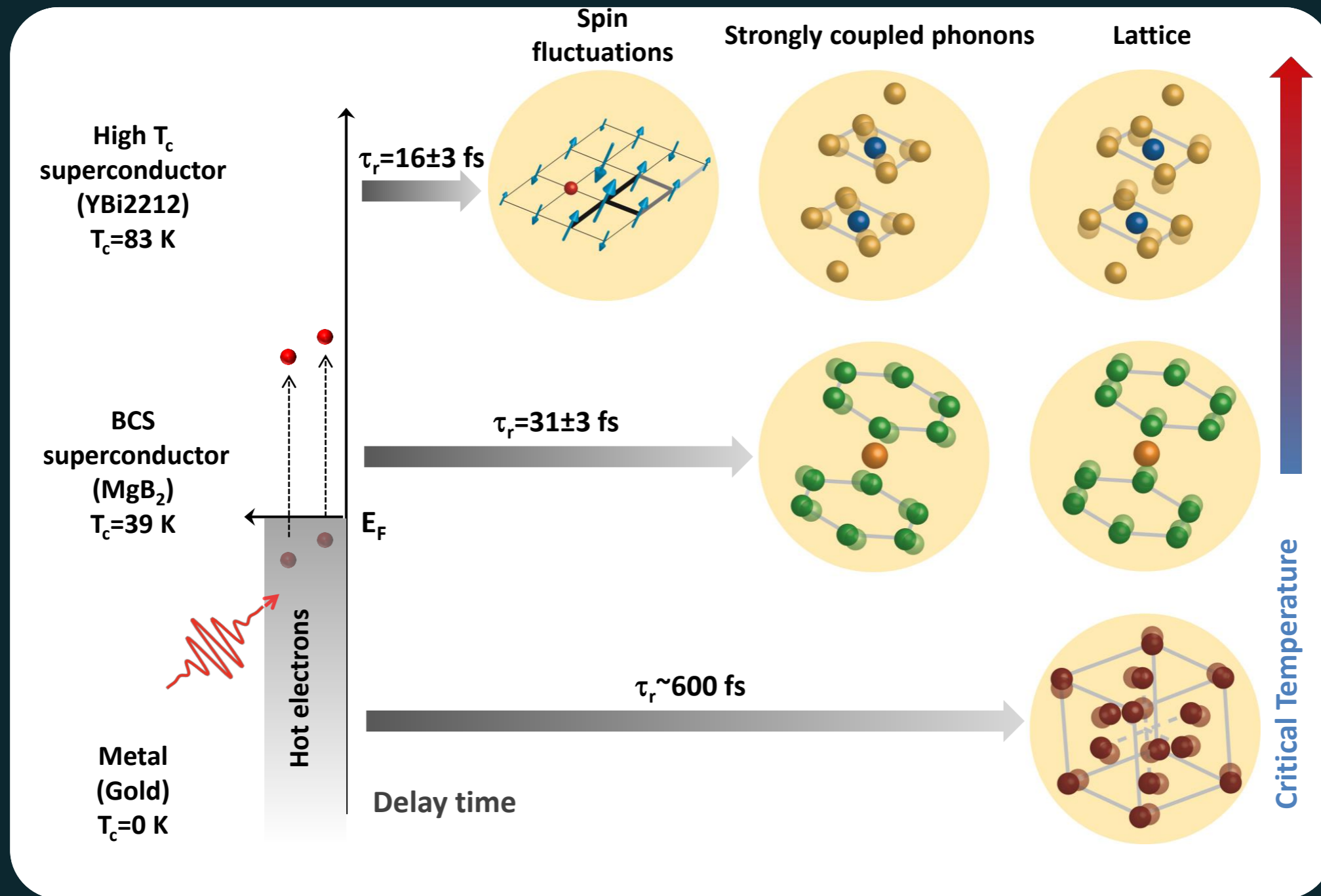
energy transfer to AF background  
 → time-dep. Schr. equation

$$H = -t_h \sum_{\langle \mathbf{l} \mathbf{j} \rangle, \sigma} (c_{\mathbf{l}, \sigma}^\dagger \tilde{c}_{\mathbf{j}, \sigma} + \text{h.c.}) + J \sum_{\langle \mathbf{l} \mathbf{j} \rangle} \mathbf{S}_{\mathbf{l}} \cdot \mathbf{S}_{\mathbf{j}}$$

$t=360$  meV    $J=120$  meV

In 16 fs photoexcited carriers can exchange energy with bosons ✓

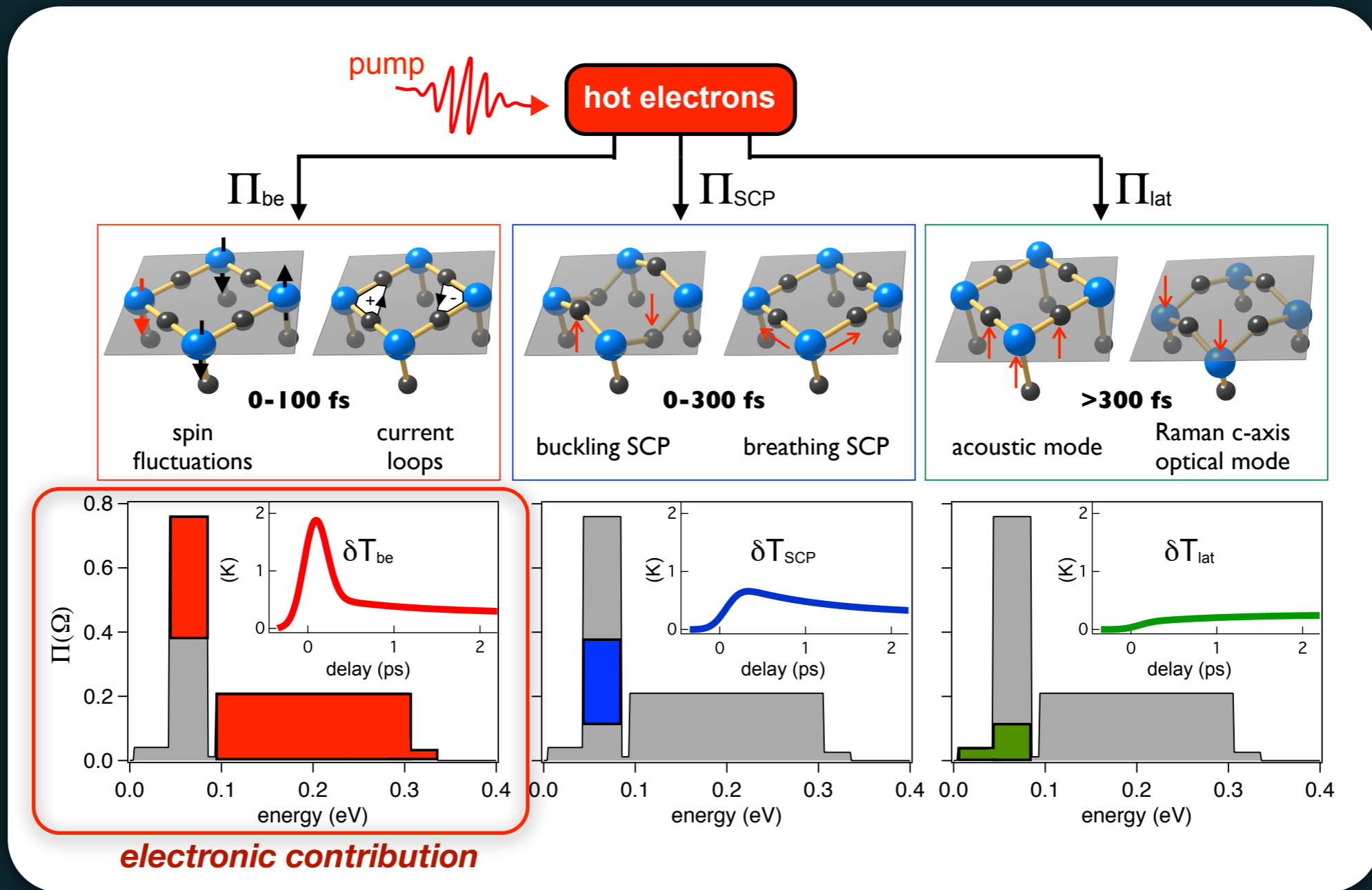
# retarded e-boson interaction



Direct observation of coupling with spin fluctuations (<20 fs) in copper oxides

# retarded e-boson interaction

S. Dal Conte et al., *Science* **335**, 1600 (2012)



retarded interaction (glue) of charge carriers with ultrafast fluctuations in cuprates

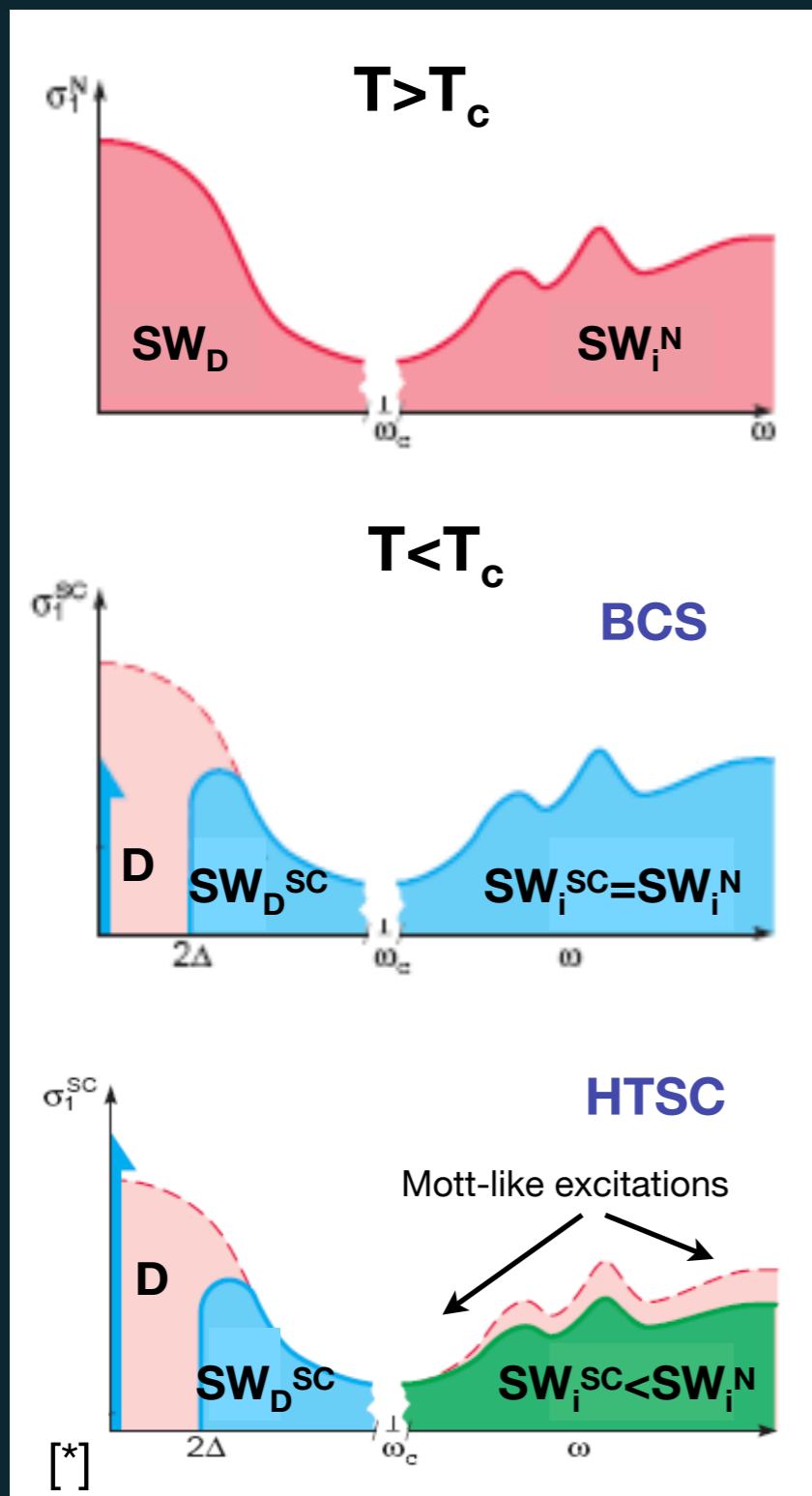
---

light pulses to **UNDERSTAND**  
equilibrium properties

⇒ optical spectral-weight  
transfer in superconductors



# Interplay between the low- and high-energy scale in HTSC?



spectral weight  $SW = \int_0^\infty \sigma_1(\omega) d\omega = \frac{\pi n e^2}{m}$

optical conductivity superconductor

$$\sigma(\omega) = \frac{ne^2}{m} \frac{1}{\tau^{-1} - i\omega} \xrightarrow{\tau \rightarrow \infty} \frac{ne^2}{m} \left[ \pi\delta(\omega) - \frac{1}{i\omega} \right]$$

Ferrel-Glover-Tinkham sum rule

$$SW_i^N - SW_i^{SC} = D - SW_D^N + SW_D^{SC}$$

Kinetic energy in a single conduction band within the nearest-neighbour tight-binding model

$$\langle K \rangle = \frac{4\hbar^2 V_{Cu}}{\pi^2 a_\delta^2 e^2} [SW_i^N - SW_i^{SC}]$$

# Interplay between the low- and high-energy scale in HTSC?

## Fundamental questions:

-Is high- $T_c$  a kinetic-energy driven phenomenon?

$$\langle E \rangle_{SC} - \langle E \rangle_N = \left[ \underbrace{\frac{\Delta^2}{V} - \frac{1}{2}N(0)\Delta^2}_{KE} \right] - \underbrace{\frac{\Delta^2}{V}}_{PE}$$

M. Tinkham, *Introduction to Superconductivity*

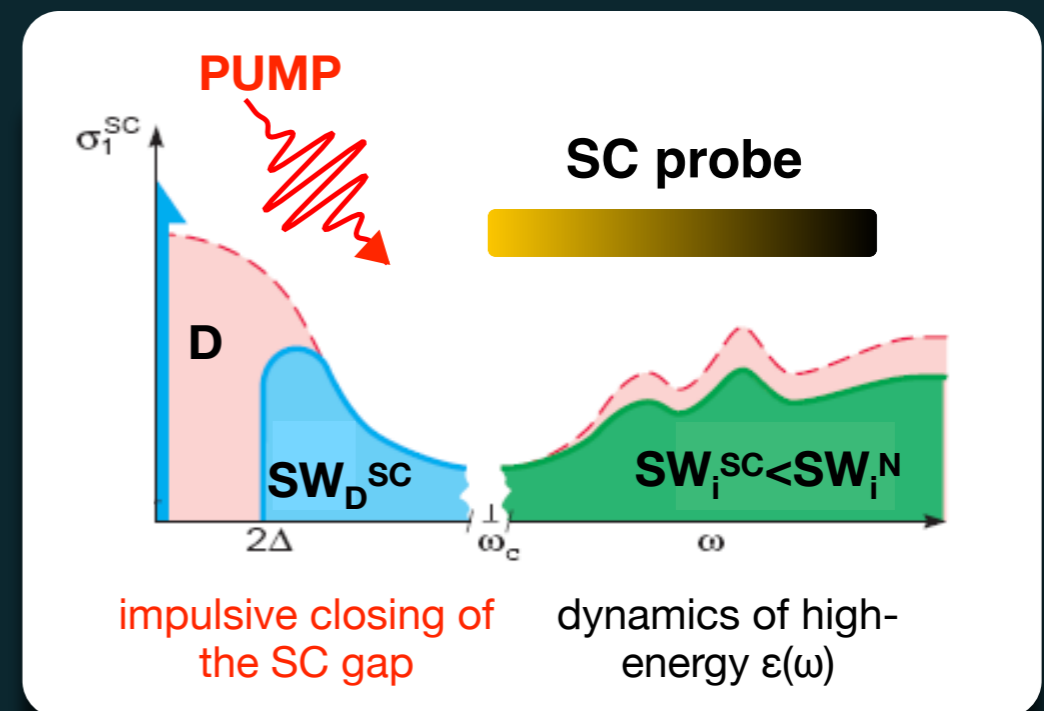
-Is the electronic structure modified at high-energy scales by the condensate?

time+spectral resolution

## Problems of equilibrium optical spectroscopies:

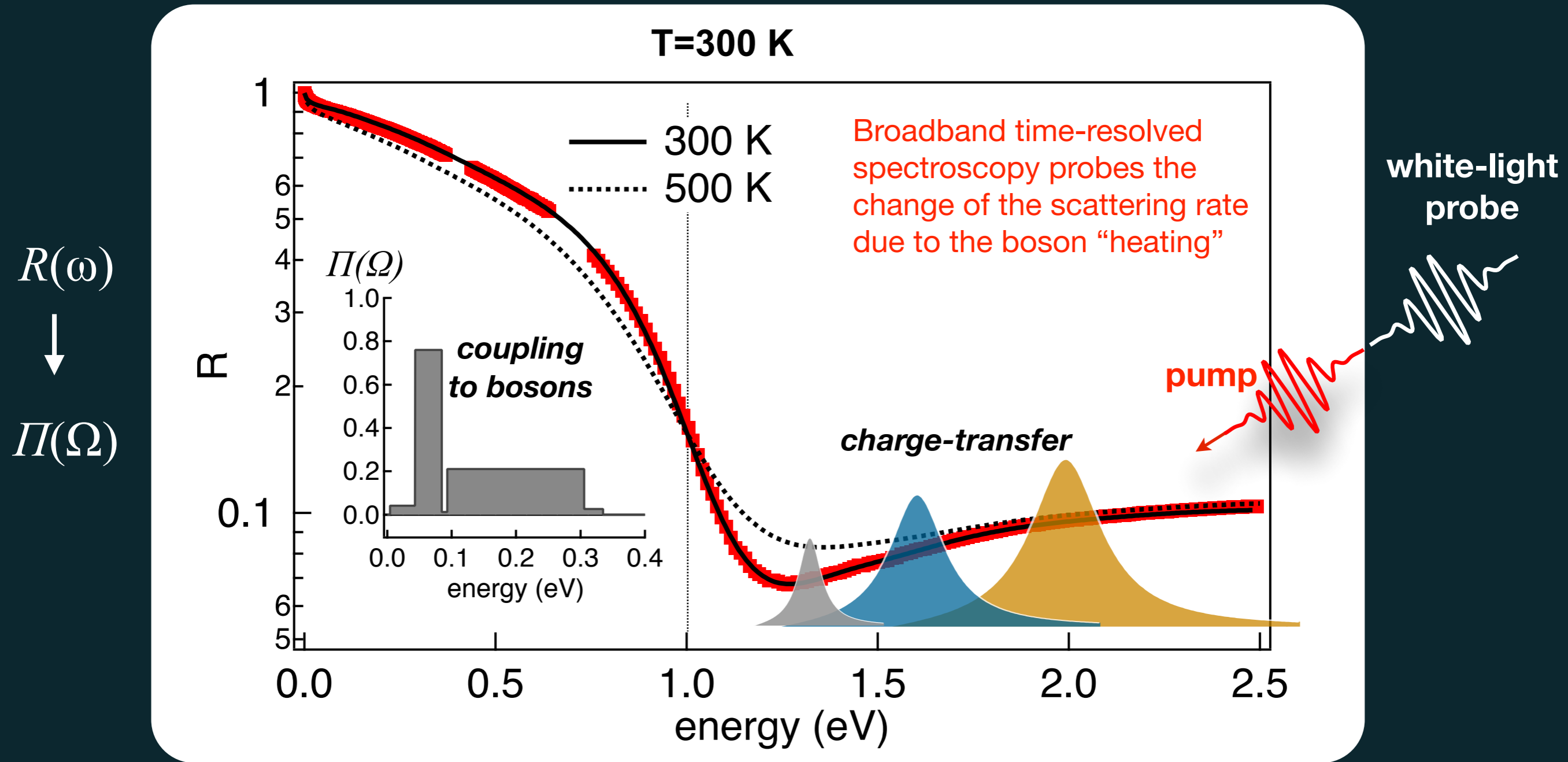
- Finite cut-off for calculating SW
- Temperature dependent Drude broadening

Holcomb, PRL (1994)  
 Rübhausen, PRL (2001)  
 Molegraaf, Science (2002)  
 Boris, Science (2004)  
 Santander-Syro, PRB (2004)



# reflectivity of cuprates

optimally doped  $\text{Bi}_2\text{Sr}_2\text{Y}_{0.08}\text{Ca}_{0.92}\text{Cu}_2\text{O}_{8+\delta}$  (YBi2212)  
 $T_c=96$  K





# Time-resolved optical spectroscopy on Y-Bi2212



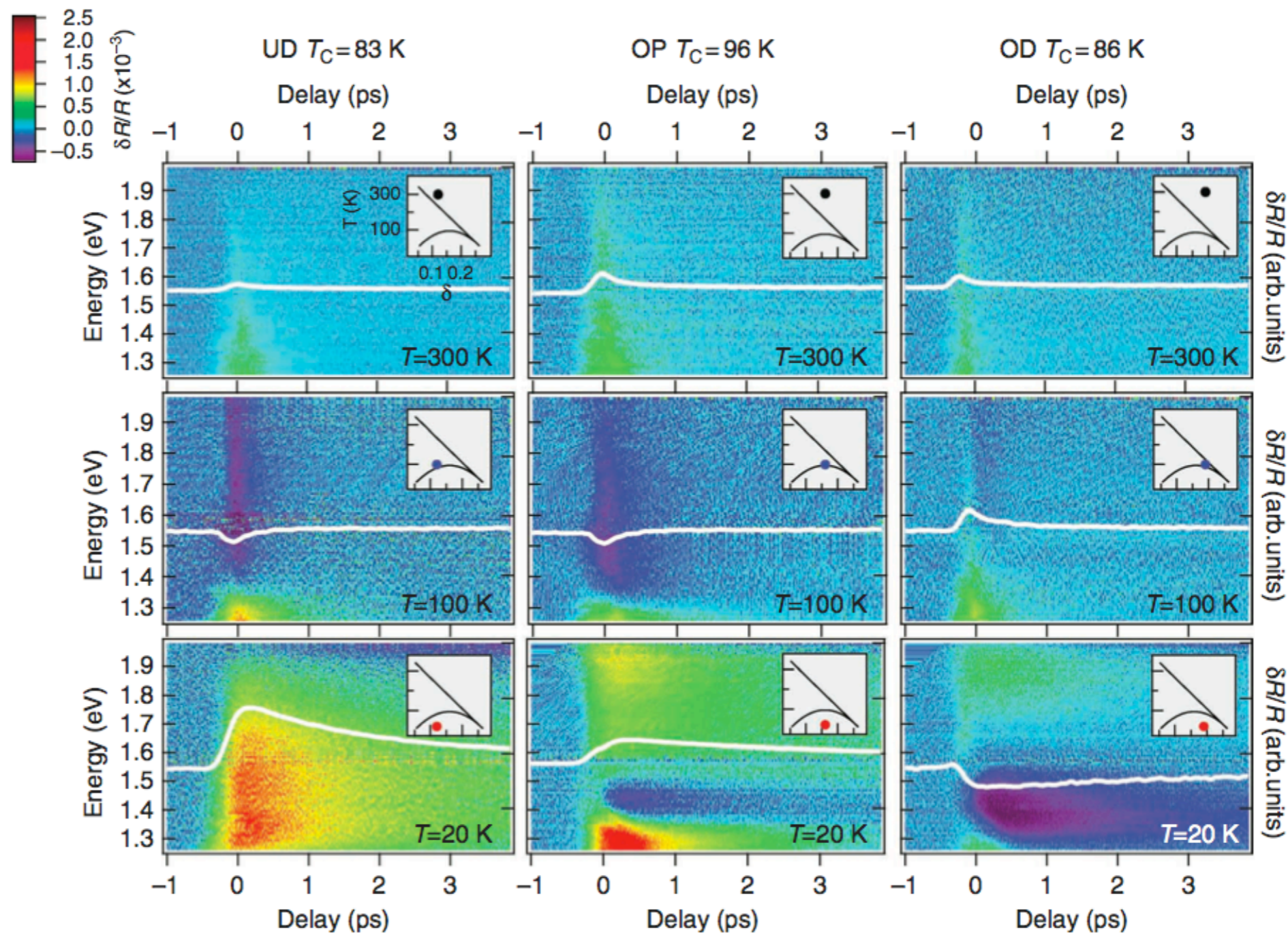
$$\frac{\delta R}{R}(\omega, t)$$

$< 10 \mu\text{J}/\text{cm}^2$

Normal  
state

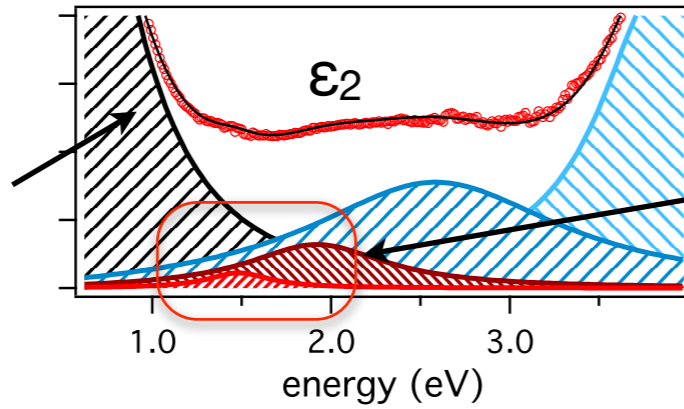
Pseudogap  
state

Superconducting  
state

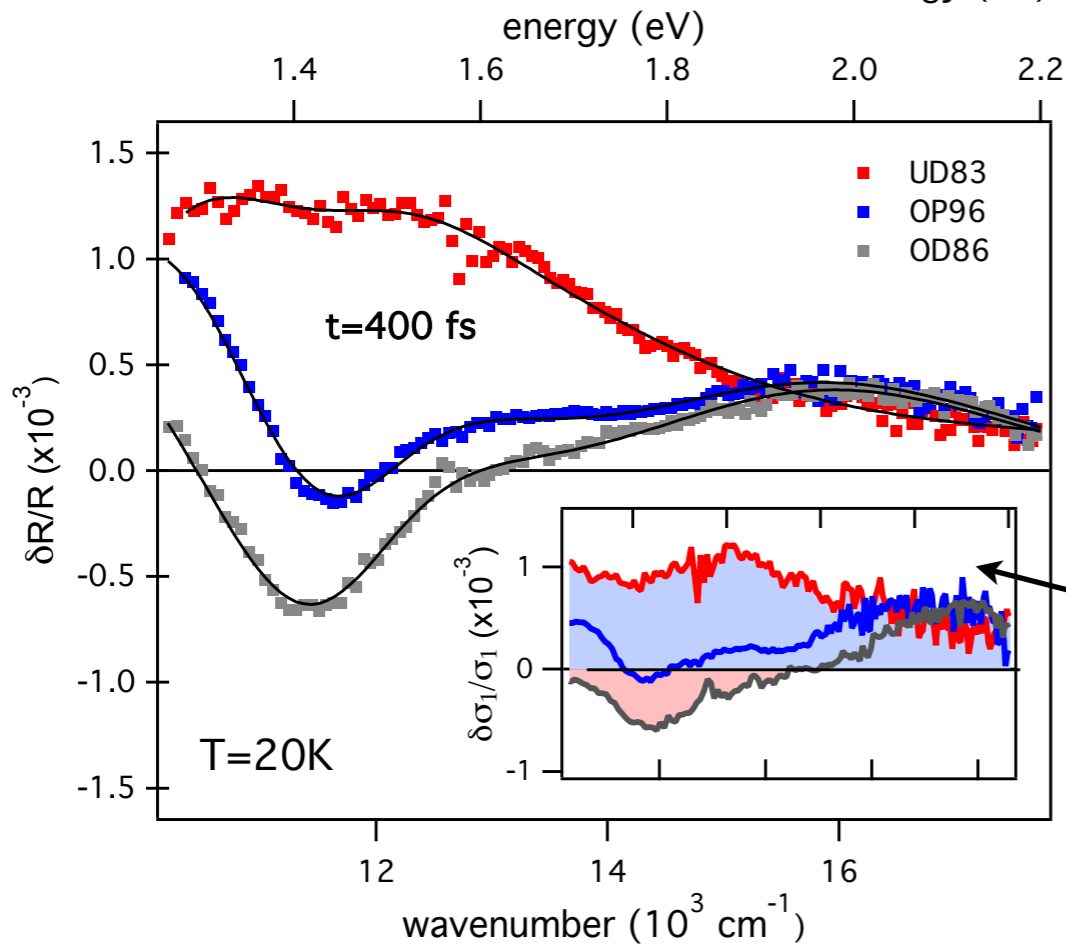


# High-energy excitations and superconductivity

Extended Drude model



interband transitions



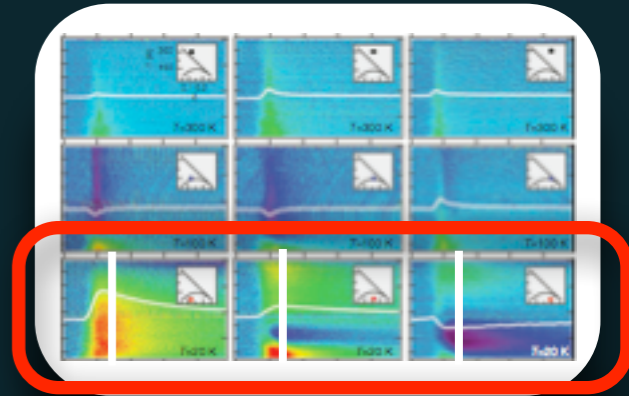
$$\delta R(\omega, t)/R$$

Kramers-Kronig constrained fit

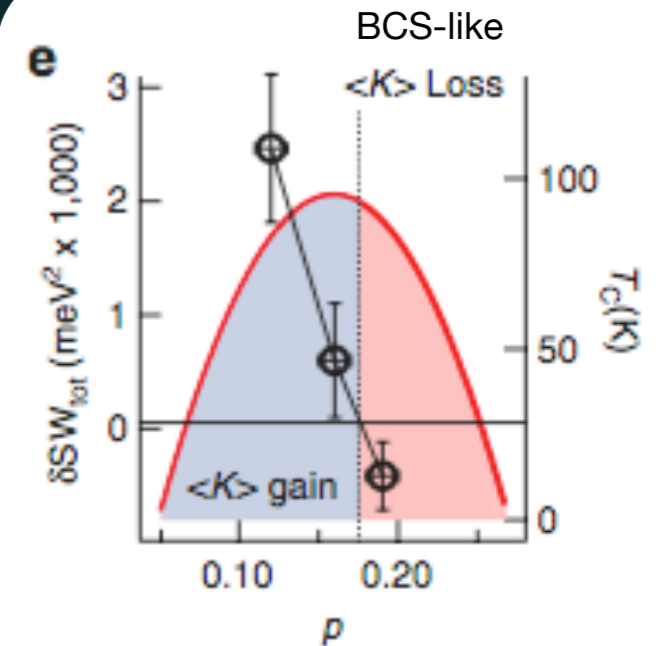
$$\delta \sigma_1(\omega, t)$$

decrease of  $\delta \sigma_1(\omega, t)/\sigma_1$  as the doping increases

20 K



$\delta=0.16$





---

light pulses to **UNDERSTAND**  
equilibrium properties

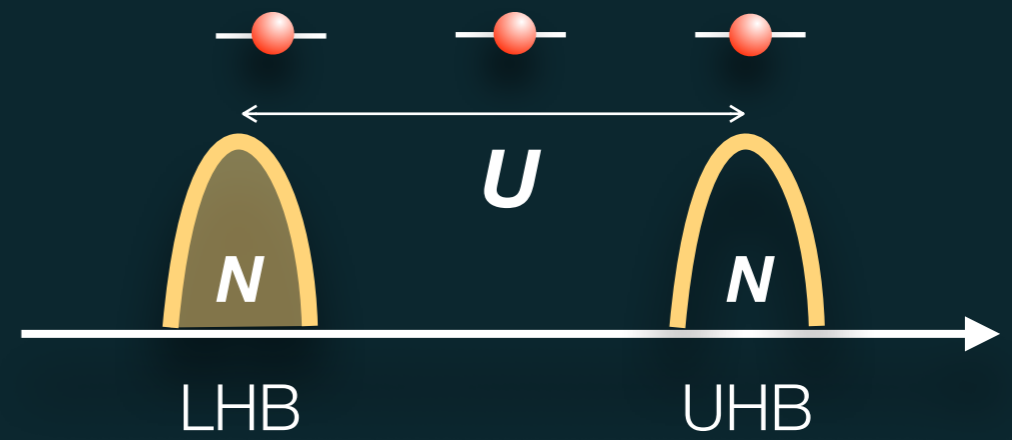
⇒ anti-nodal Mottness

# Mottness

band insulator

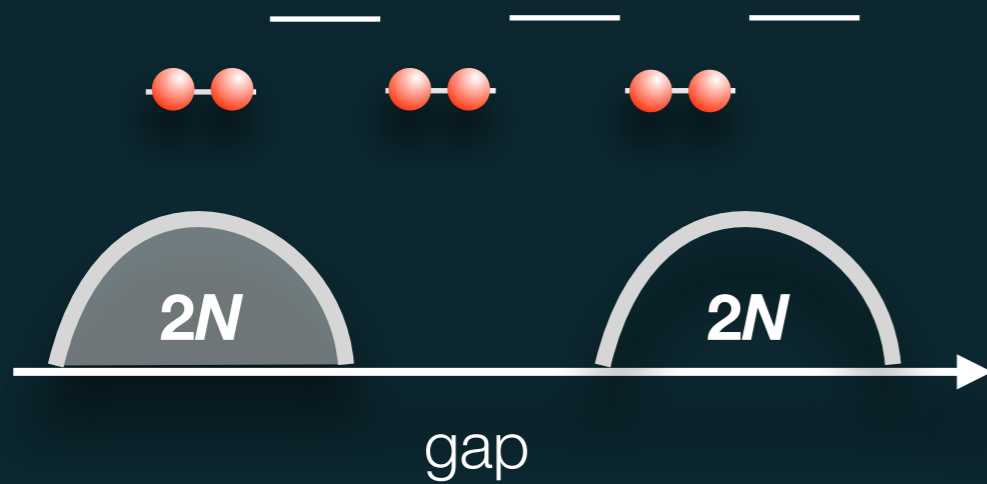


Mott insulator

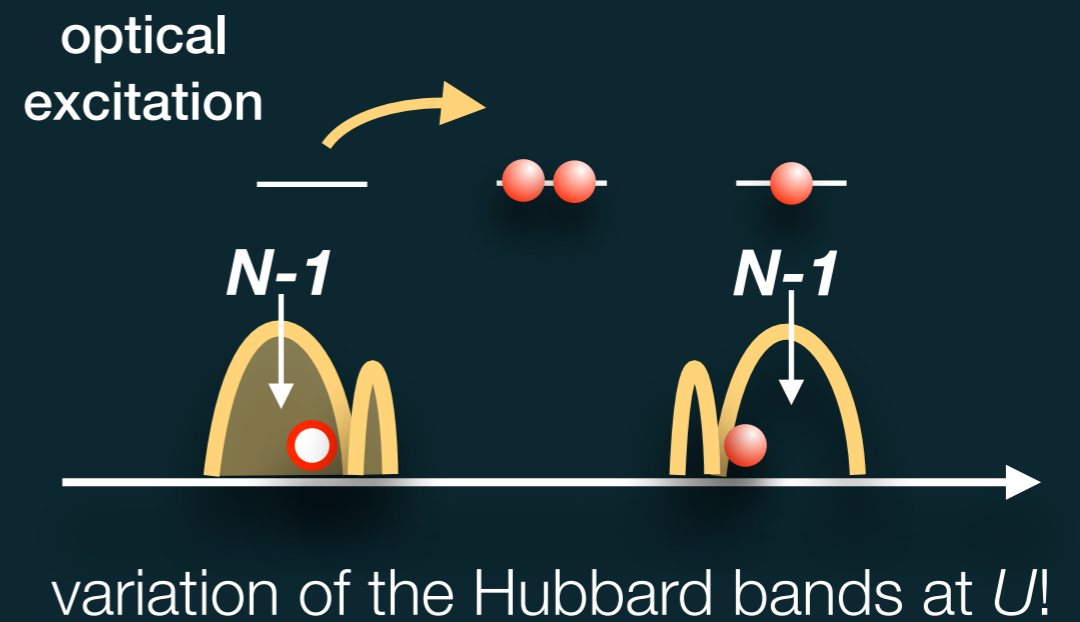
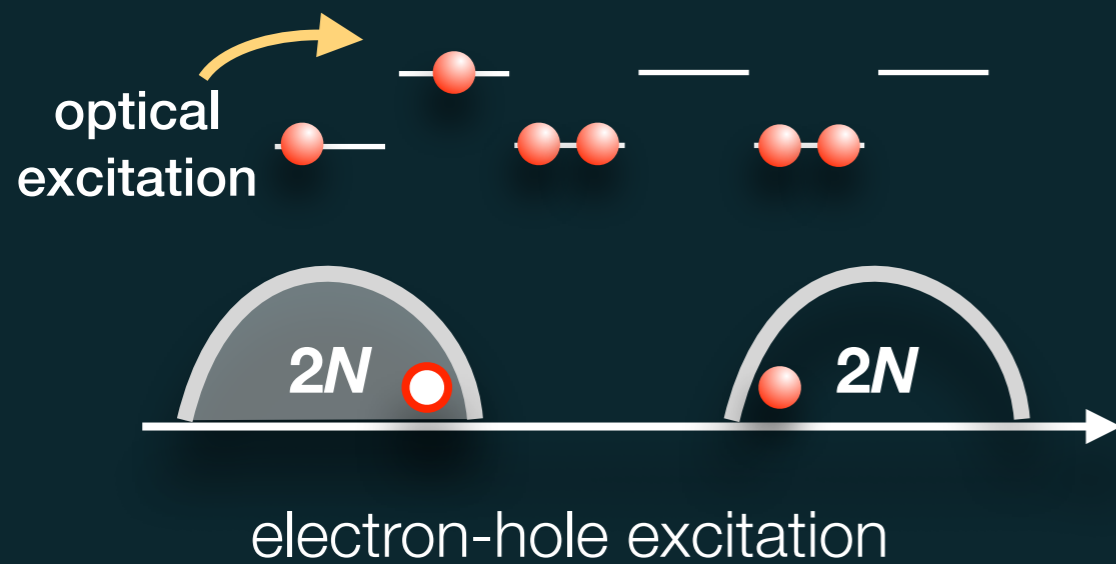
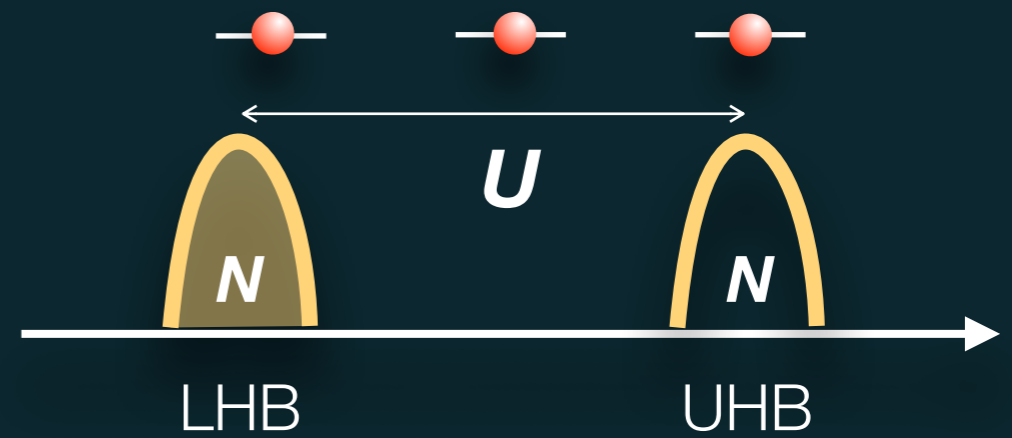


# Mottness

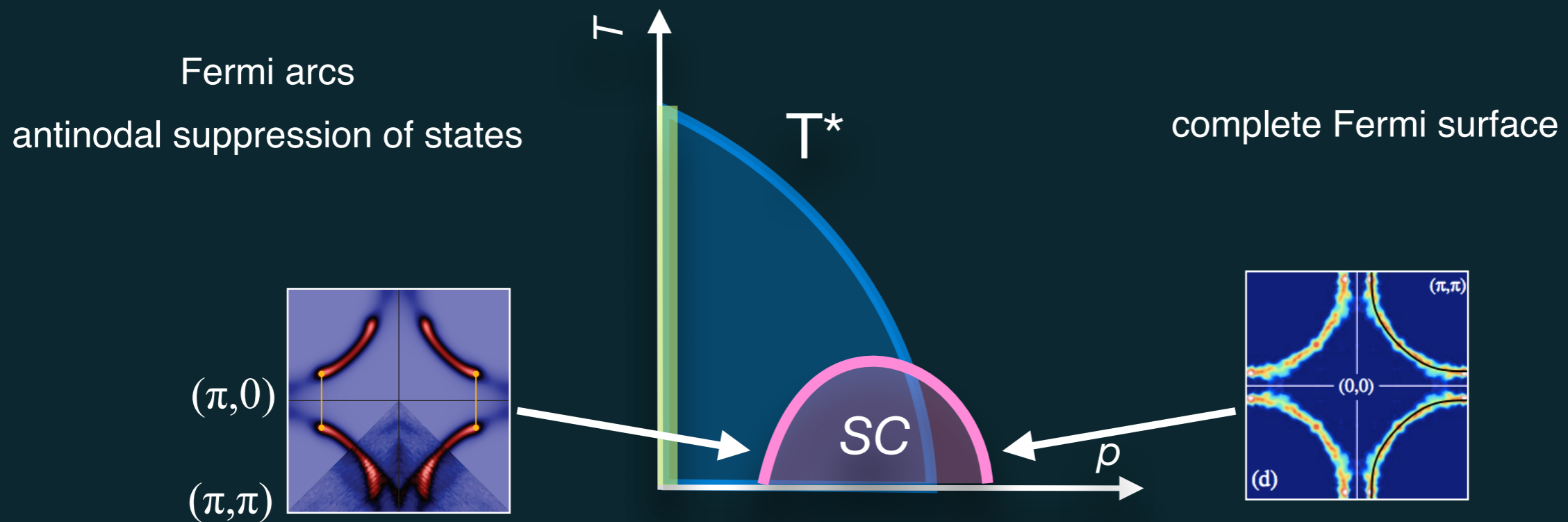
band insulator



Mott insulator



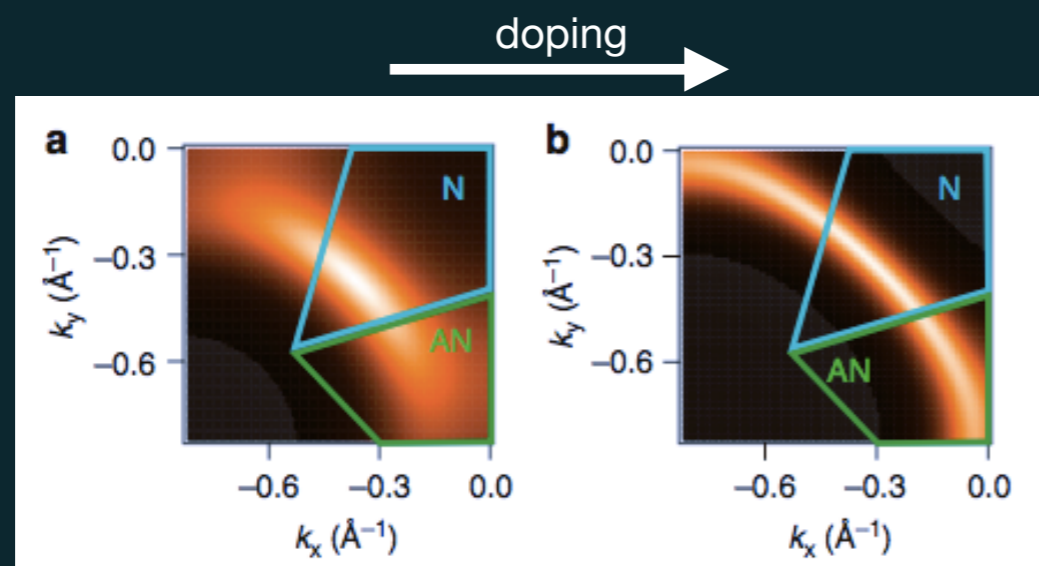
# the puzzle of antinodal gap in cuprates



R. Comin et al. *Science* **343**, 390 (2014)

M. Platè et al. *Phys. Rev. Lett.* **95**, 077001 (2005)

Is the universal antinodal suppression of states a consequence of the correlations?



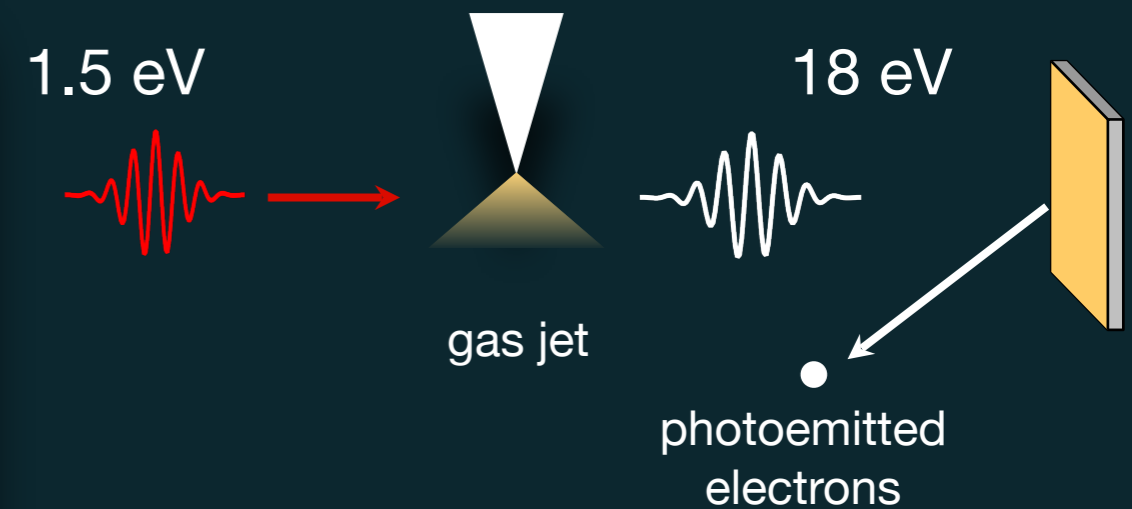
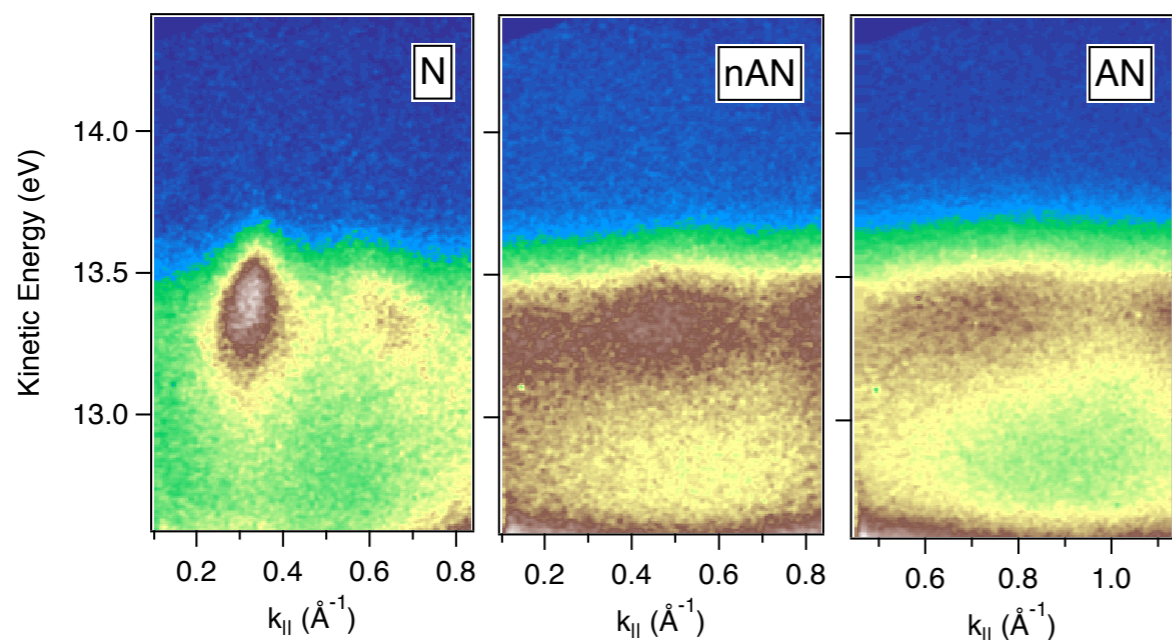
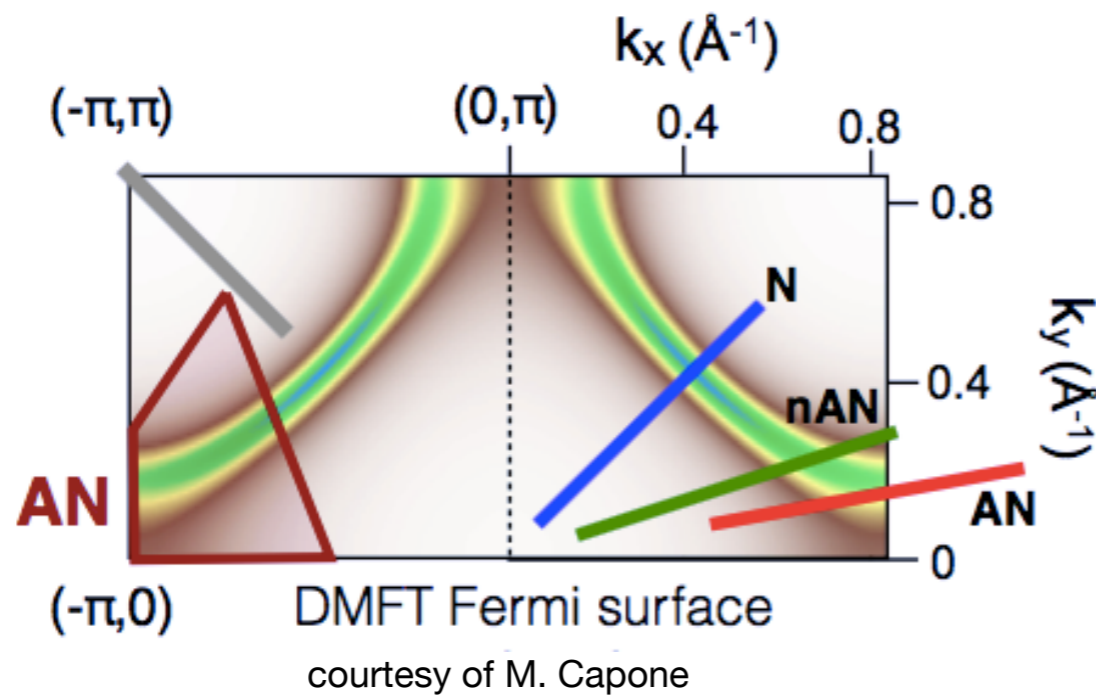
F. Cilento et al. *Nat. Commun.* **5**:4353 (2014)

It is already within the Hubbard model!

***k*-selective Mottness**

E. Gull et al. *Phys. Rev. B* **82**, 155101 (2010)

# antinodal dynamics

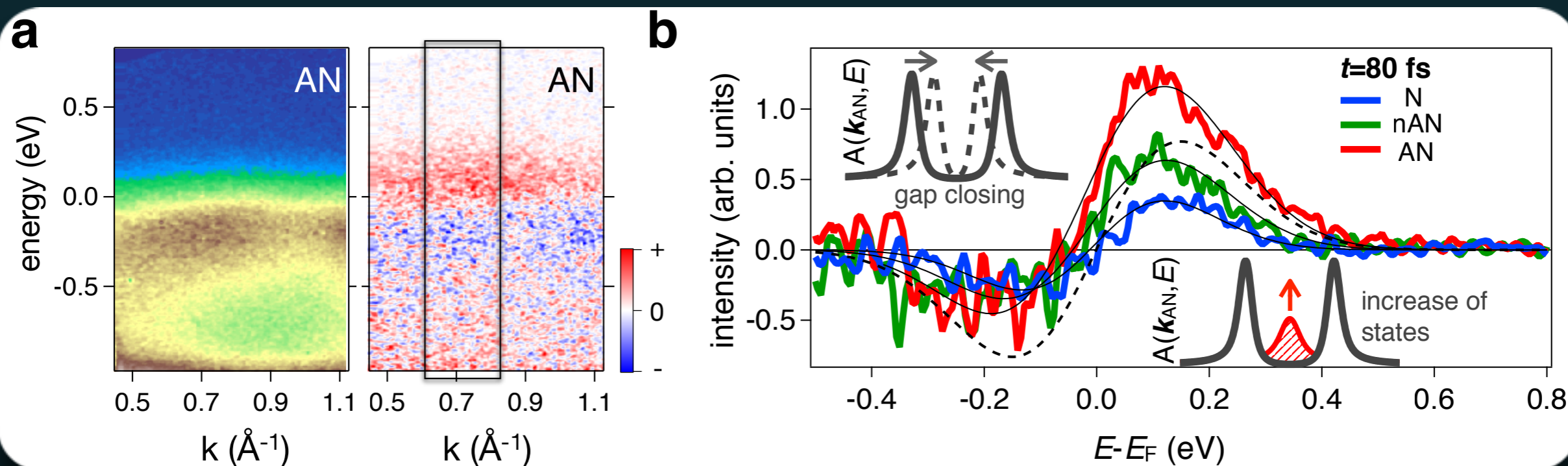


complete mapping of the  
dynamics in the full  
Brillouin zone in optimally-  
doped ( $T_c=96\text{K}$ )  
 $\text{Bi}_2\text{Sr}_2\text{Ca}_{0.92}\text{Y}_{0.08}\text{Cu}_2\text{O}_{8+\delta}$

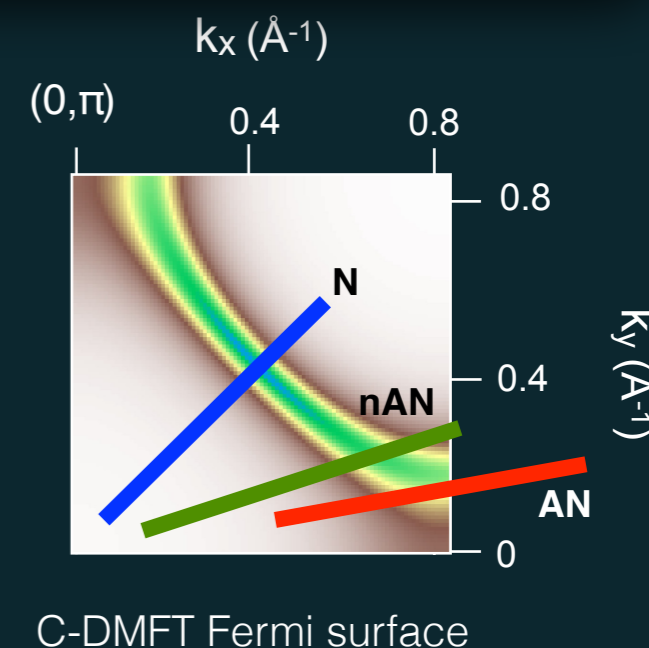


# AN dynamics

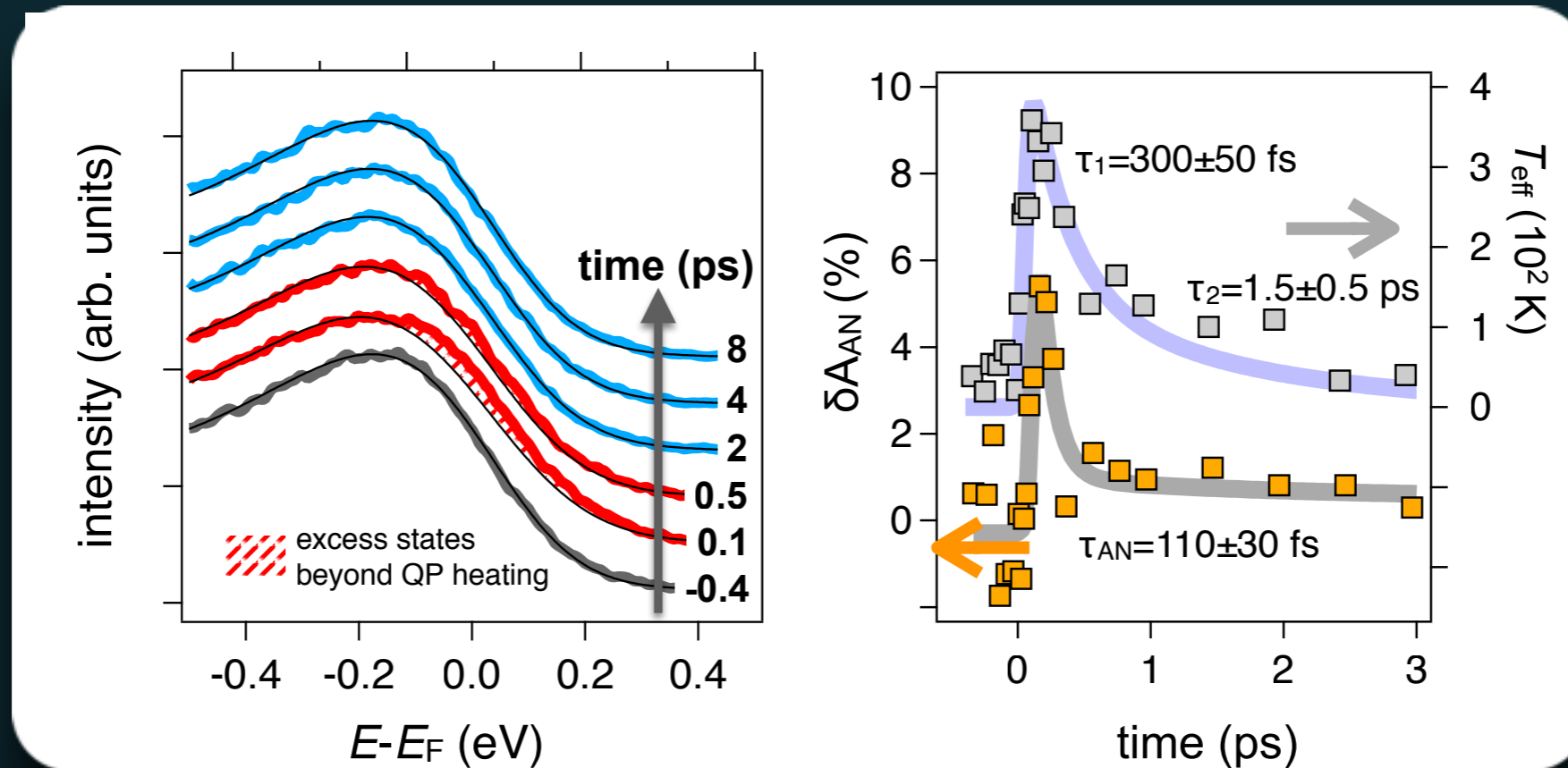
$t=80$  fs



- the antinodal asymmetric signal can be reproduced by assuming a **transient increase of states at the Fermi level**



# AN dynamics



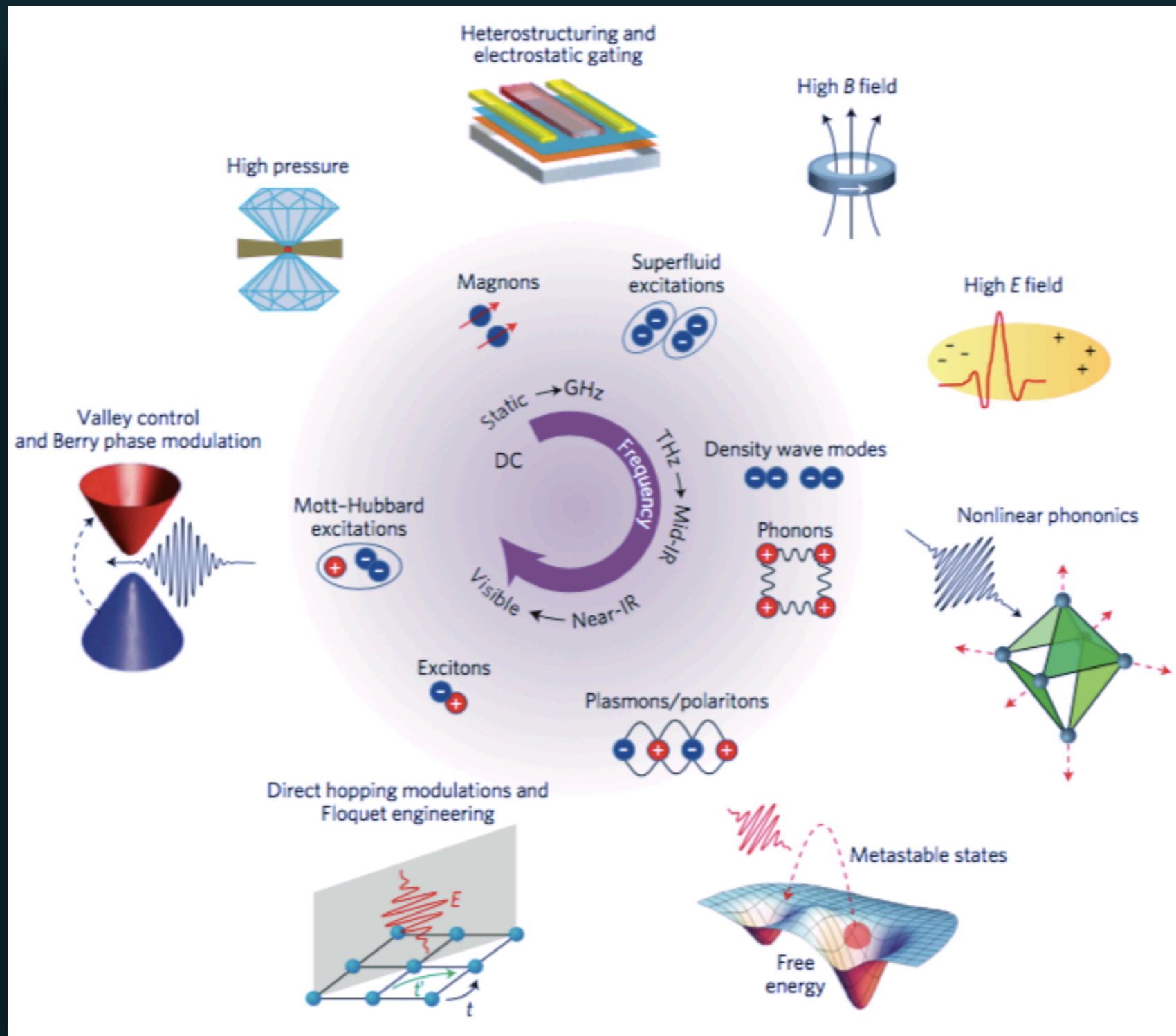
- the AN increase of states ( $\approx 100$  fs) is decoupled from the effective temperature
- the excitation at 1.5 eV drives the transient increase of antinodal states, which become more metallic

---

light pulses to **MANIPULATE**  
materials properties

⇒ hidden states, transient photo-  
enhanced superconductivity,  
competition of different orders

# properties on demand in quantum materials



**new functionalities:** methods for controlling quantum phases on demand

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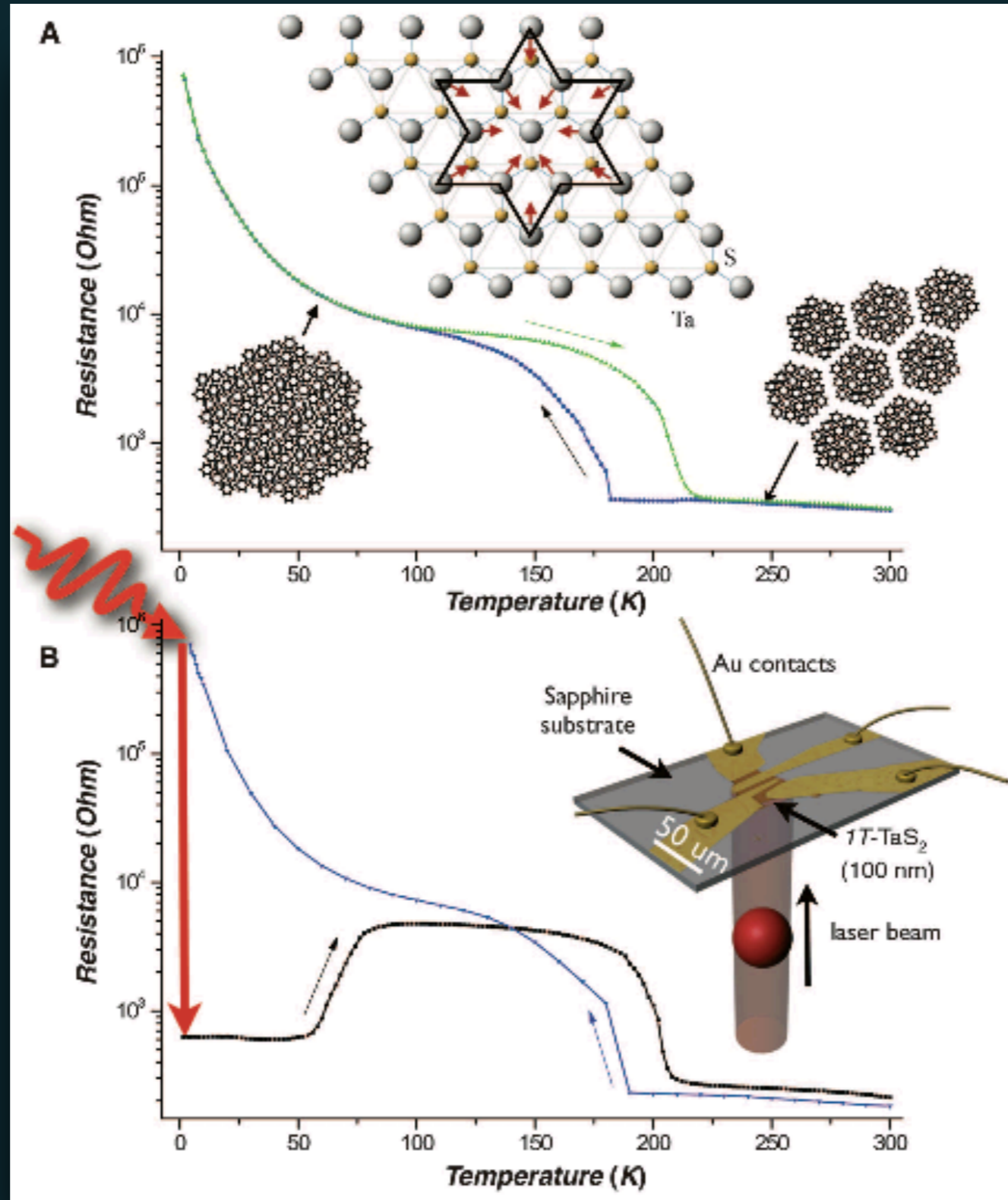
light pulses to **MANIPULATE**  
materials properties

⇒ hidden states



# photo-induced hidden states

## Ultrafast switching to a stable hidden state in $1T\text{-TaS}_2$



L. Stojchevska et al.,  
*Science* **344**, 177 (2014)

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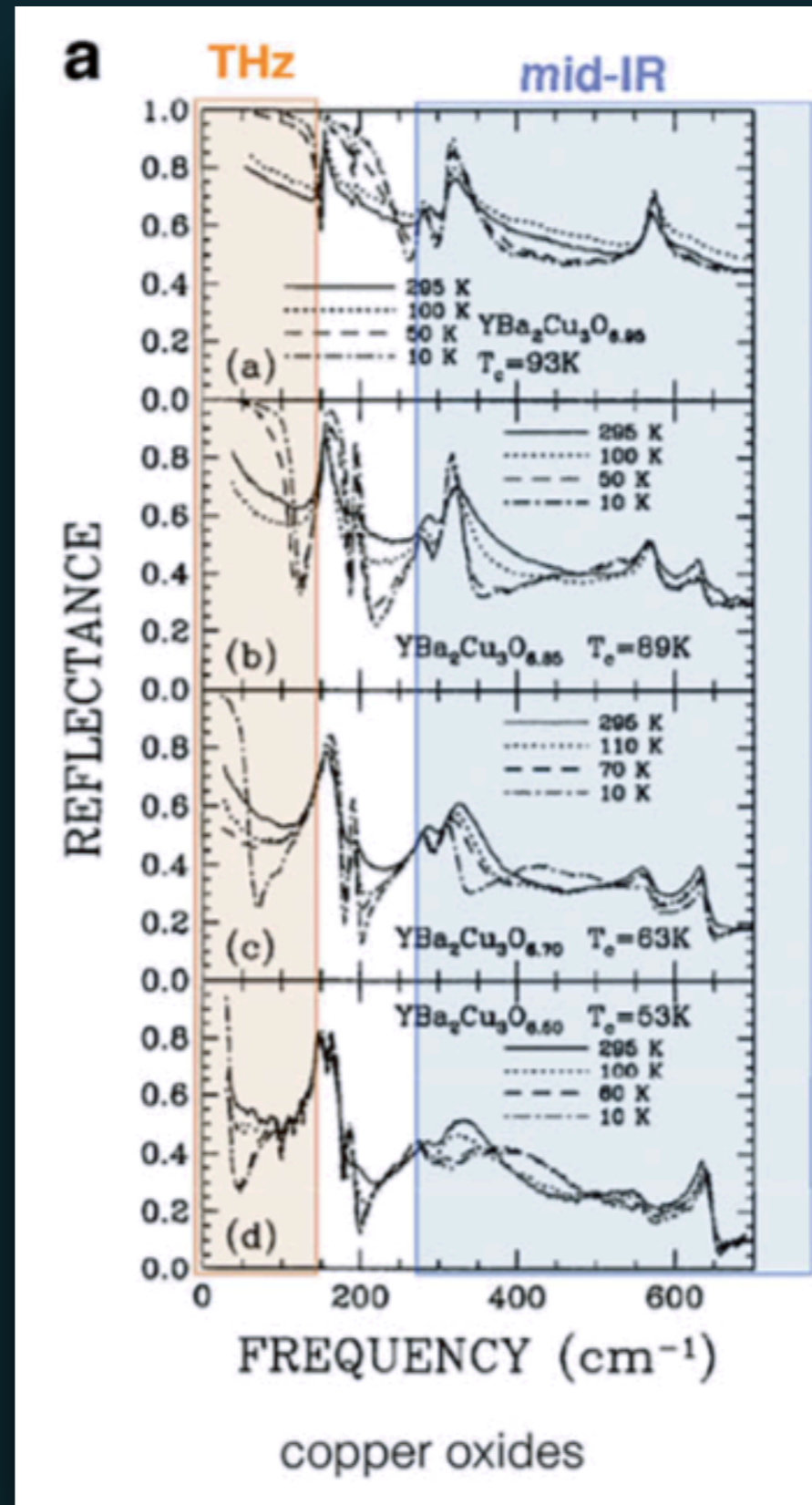
light pulses to **MANIPULATE**  
materials properties

⇒ transient photo-enhanced  
superconductivity

# phonon pumping

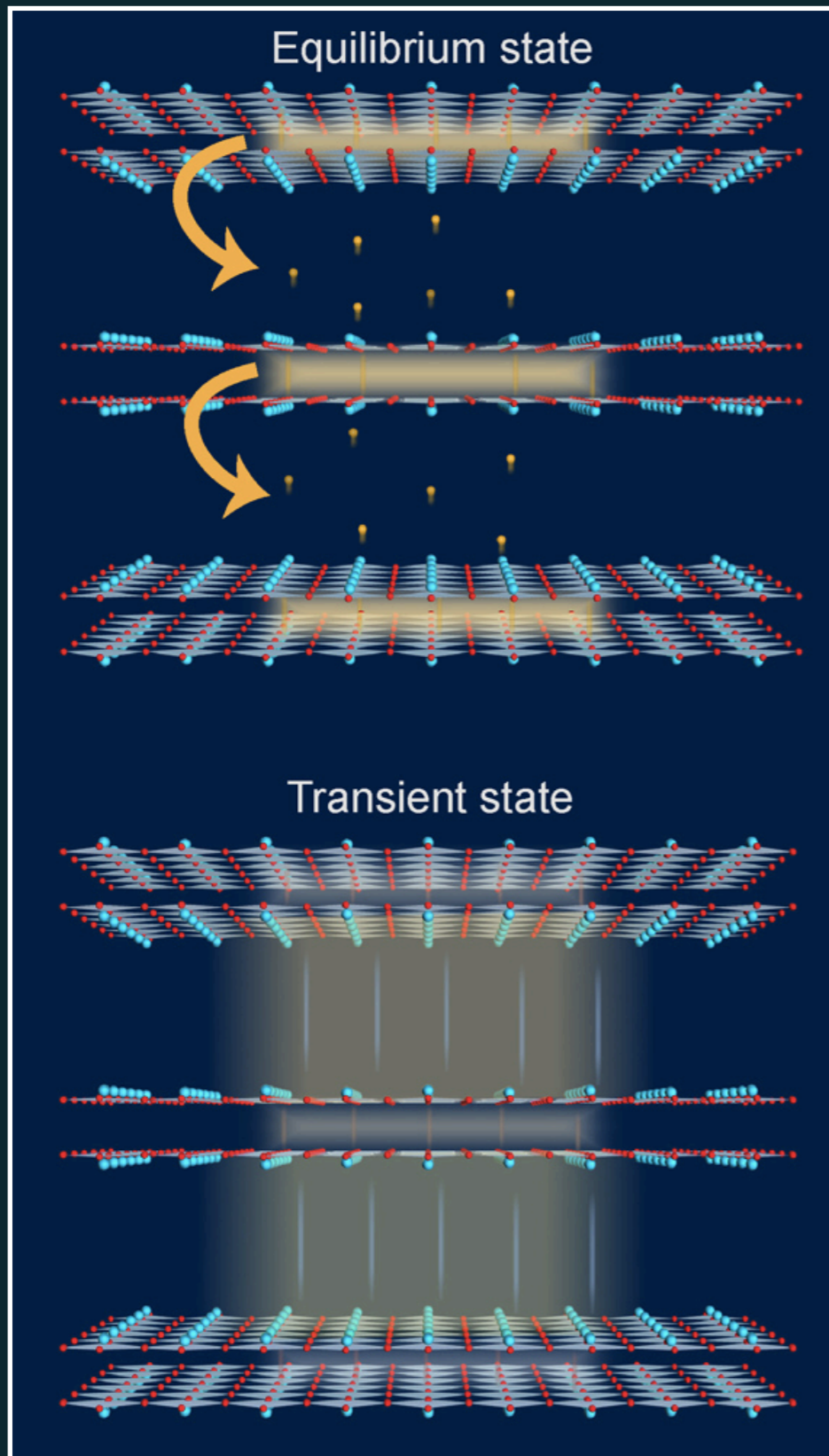
mid-IR enhancement of superconductivity in high- $T_c$ ?

role of the lattice?

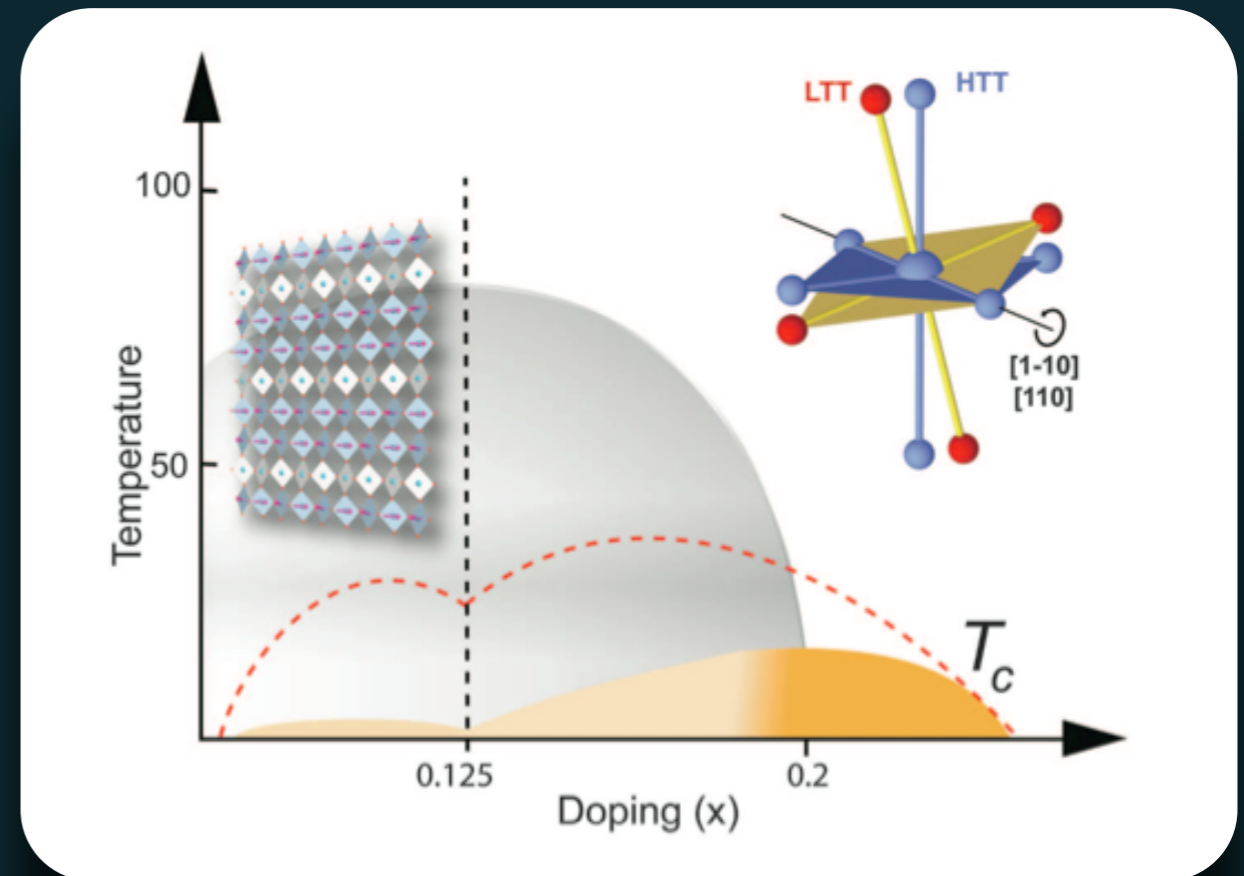


C.C. Homes et al., *Phys. C: Supercond.* **254**, 265 (1995)

# possible light-induced transient superconductivity



Transient superconductivity by removing a competing order

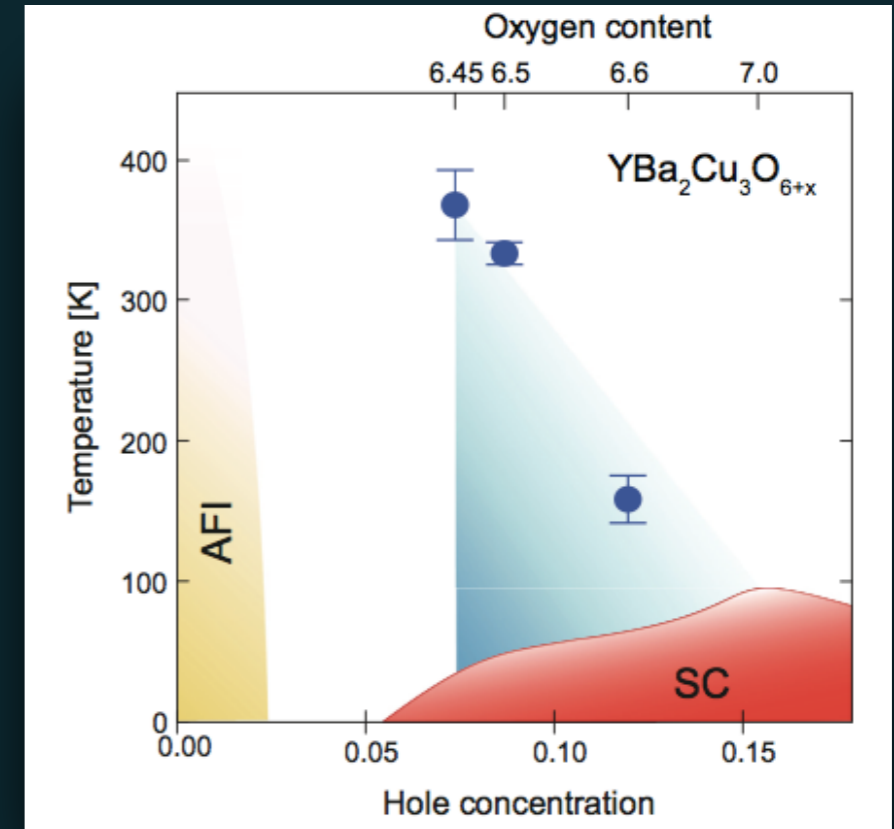
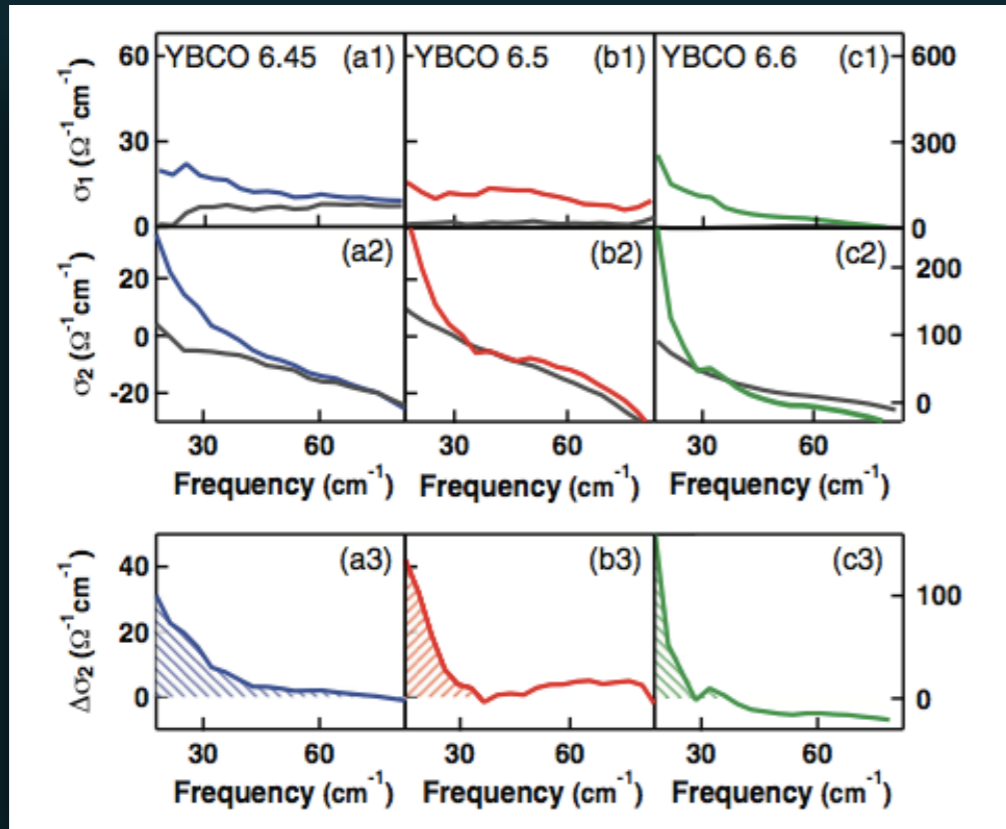


D. Fausti et al., *Science* **331**, 189 (2011)



# optical-control of phase transitions

## transient enhancement of superconductivity in copper oxides



S. Kaiser et al., *Physical Review B* **89**, 184516 (2014)  
 W. Hu et al., *Nature Materials* **13**, 705 (2014)

mid-IR pump  
 THz probe (c-axis)

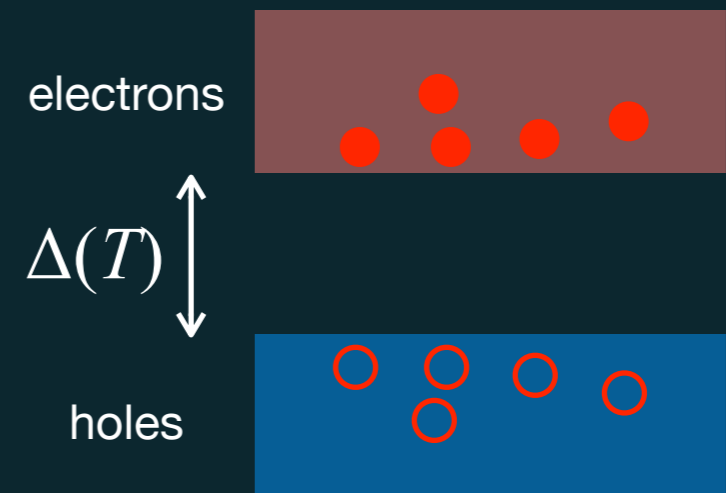
Drude opt. cond.  $\sigma(\omega) = \frac{ne^2}{m} \frac{1}{\tau^{-1} - i\omega} \xrightarrow{\tau \rightarrow \infty} \frac{ne^2}{m} \left[ \pi\delta(\omega) - \frac{1}{i\omega} \right]$



# gap enhancement via sub-gap excitation

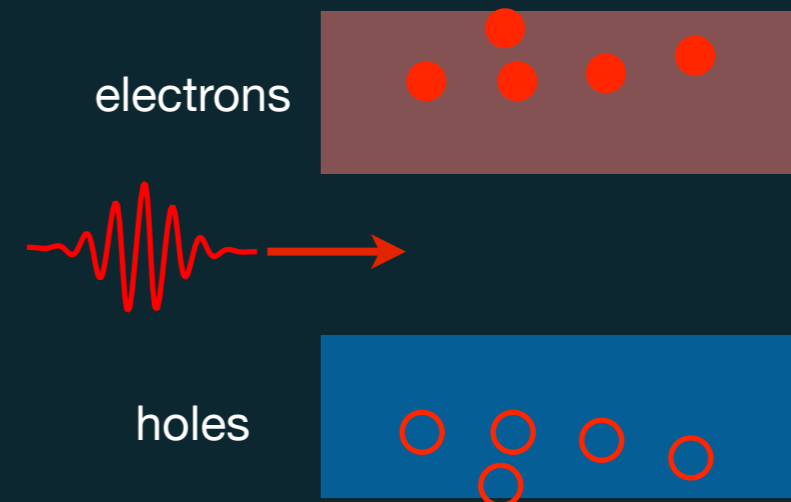
BCS gap equation at finite temperature

$$1 = VN(0) \int_{-\infty}^{\infty} \frac{\overbrace{f(T)}^{\text{thermal distribution of excitations}}}{\sqrt{\epsilon^2 + \Delta^2(T)}} d\epsilon$$



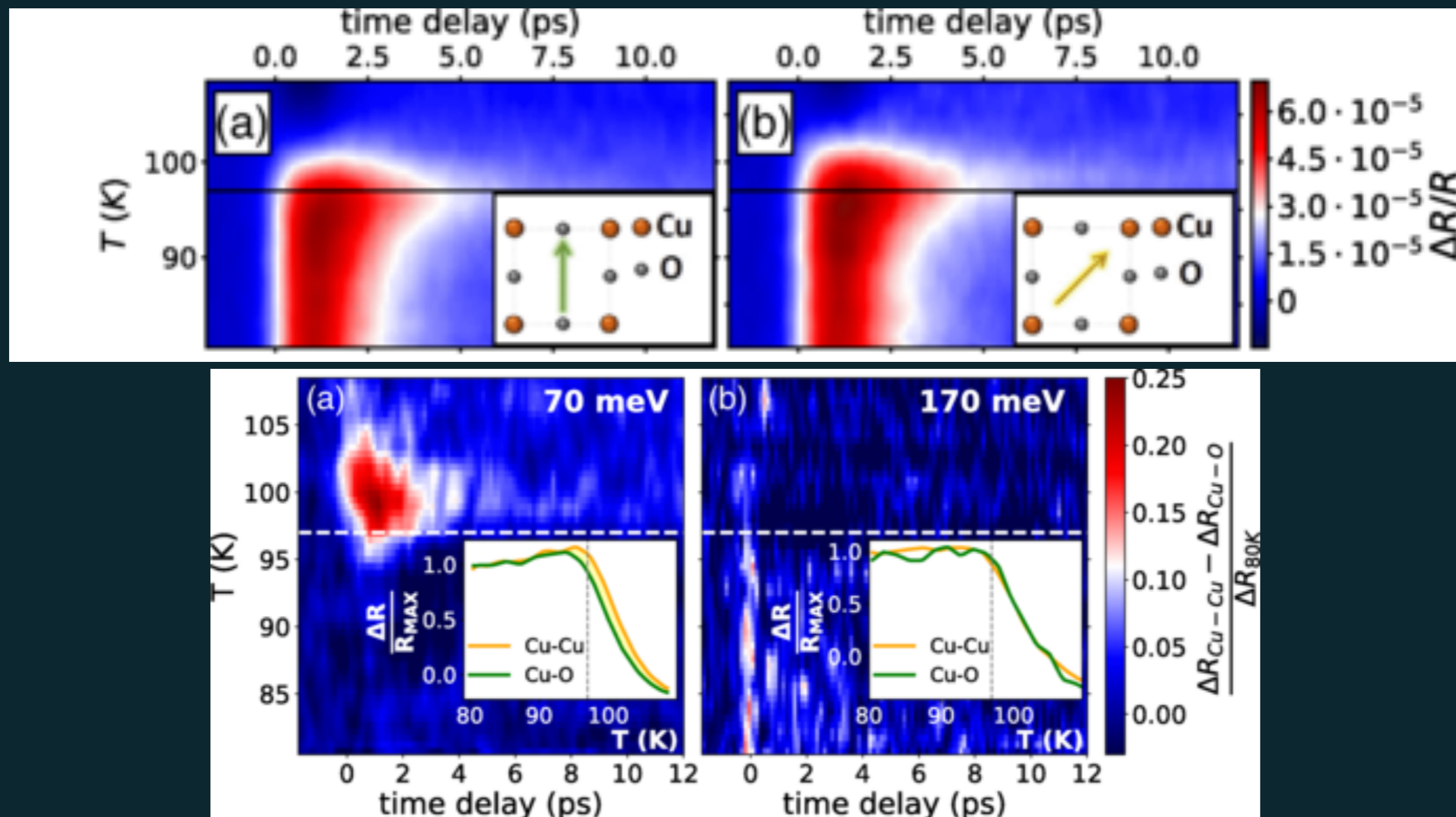
changing the distribution without injecting new excitations

$$\frac{\delta T_{eff}}{T} = \int_{-\infty}^{\infty} \frac{\delta f}{\sqrt{\epsilon^2 + \Delta^2(T)}} d\epsilon$$



# gap enhancement via sub-gap excitation

- microwave gap enhancement in Al  
T. Kommers et al., *Phys. Rev. Lett.* **38**, 1091 (1977)
- THz gap enhancement in NbN  
M. Beck et al., *Phys. Rev. Lett.* **110**, 267003 (2013)
- mid-IR gap enhancement in  $\text{Bi}_2\text{Sr}_2\text{Y}_{0.08}\text{Ca}_{0.92}\text{Cu}_2\text{O}_{8+\delta}$   
F. Giusti et al., *Phys. Rev. Lett.* **122**, 067002 (2019)



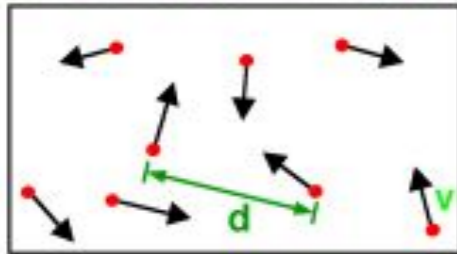
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light pulses to **MANIPULATE**  
materials properties

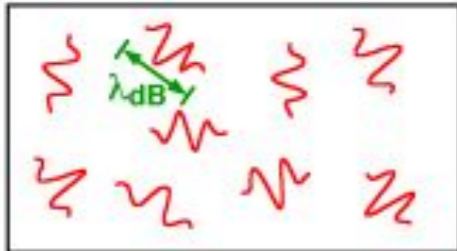
⇒ quenching the phase coherence

# destroying phase coherence with light

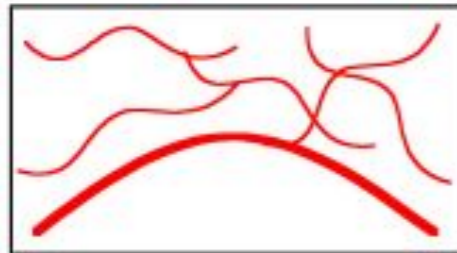
What is Bose-Einstein condensation (BEC)?



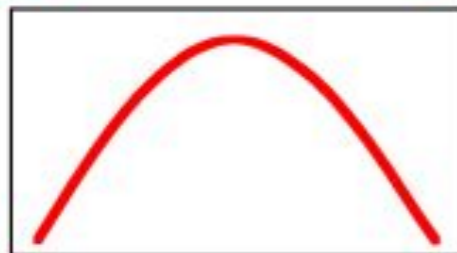
High Temperature T:  
thermal velocity  $v$   
density  $d^{-3}$   
"Billiard balls"



Low Temperature T:  
De Broglie wavelength  
 $\lambda_{dB} = h/mv \propto T^{-1/2}$   
"Wave packets"



$T = T_{crit}$ :  
Bose-Einstein Condensation  
 $\lambda_{dB} = d$   
"Matter wave overlap"



$T = 0$ :  
Pure Bose condensate  
"Giant matter wave"

kinetic energy  $\frac{1}{2}mv^2 = \frac{3}{2}K_B T$  thermal energy

De Broglie wavelength

Planck const.

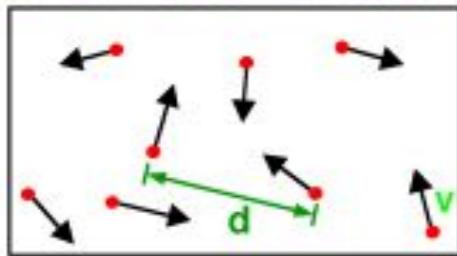
$$\lambda_{DB} = \frac{h}{mv} \propto \frac{1}{\sqrt{T}}$$

light can be used to destroy the phase coherence of the condensate without heating it up

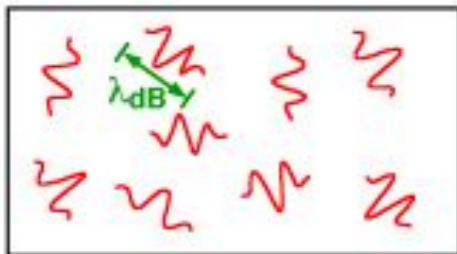


# destroying phase coherence with light

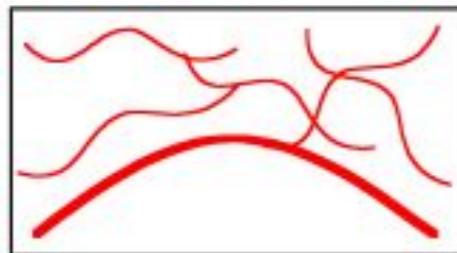
What is Bose-Einstein condensation (BEC)?



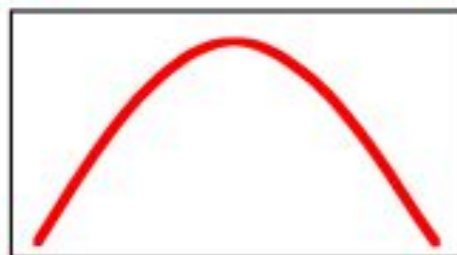
High Temperature T:  
thermal velocity  $v$   
density  $d^{-3}$   
"Billiard balls"



Low Temperature T:  
De Broglie wavelength  
 $\lambda_{dB} = h/mv \propto T^{-1/2}$   
"Wave packets"

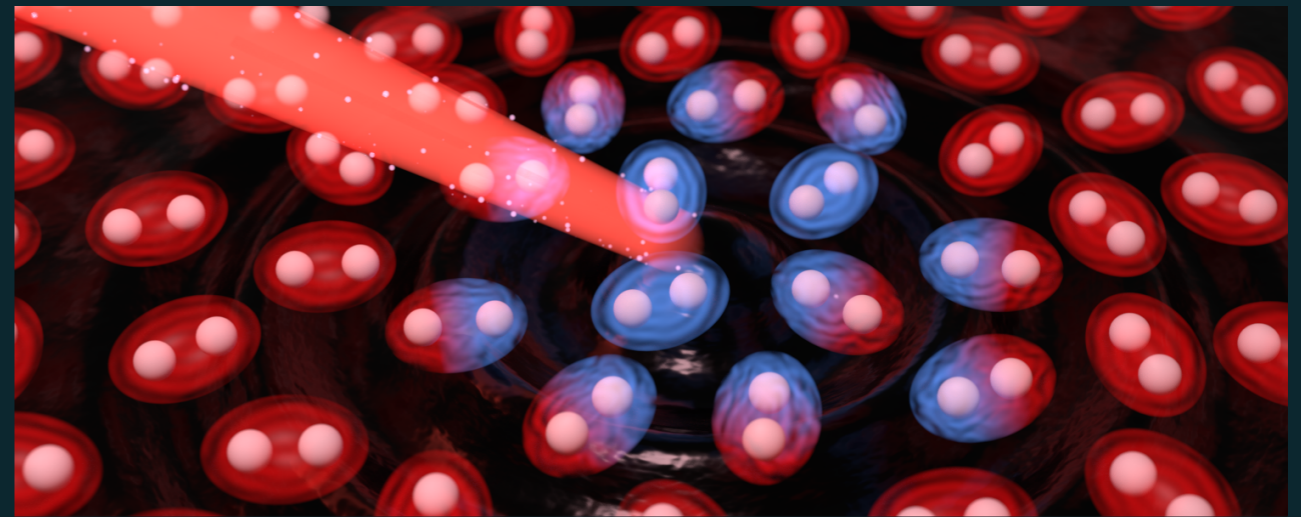


$T = T_{crit}$ :  
Bose-Einstein  
Condensation  
 $\lambda_{dB} = d$   
"Matter wave overlap"



$T = 0$ :  
Pure Bose  
condensate  
"Giant matter wave"

cartoon: light excitation of the  
Cooper pair condensate



courtesy of F. Boschini (UBC, Vancouver)

light can be used to destroy the phase coherence of  
the condensate without heating it up



## Open problems:

- what drives the ultrafast dynamics of the superconducting condensate?

gap closing (BCS)

VS

loss of phase coherence

fragility of low-density ( $n_S$ ) superconductors  
to phase fluctuations:

$$\hbar\Omega_{\Theta} = \frac{\hbar^2 n_S(0)a}{2m^*} \simeq k_B T_C$$

V.J. Emery and S.A. Kivelson. *Nature* **374**, 434 (1995)

# superconducting to normal state transition

GAP closing or filling?

what drives the non-thermal melting?

## Open problems:

- what drives the ultrafast dynamics of the superconducting condensate?

gap closing (BCS)

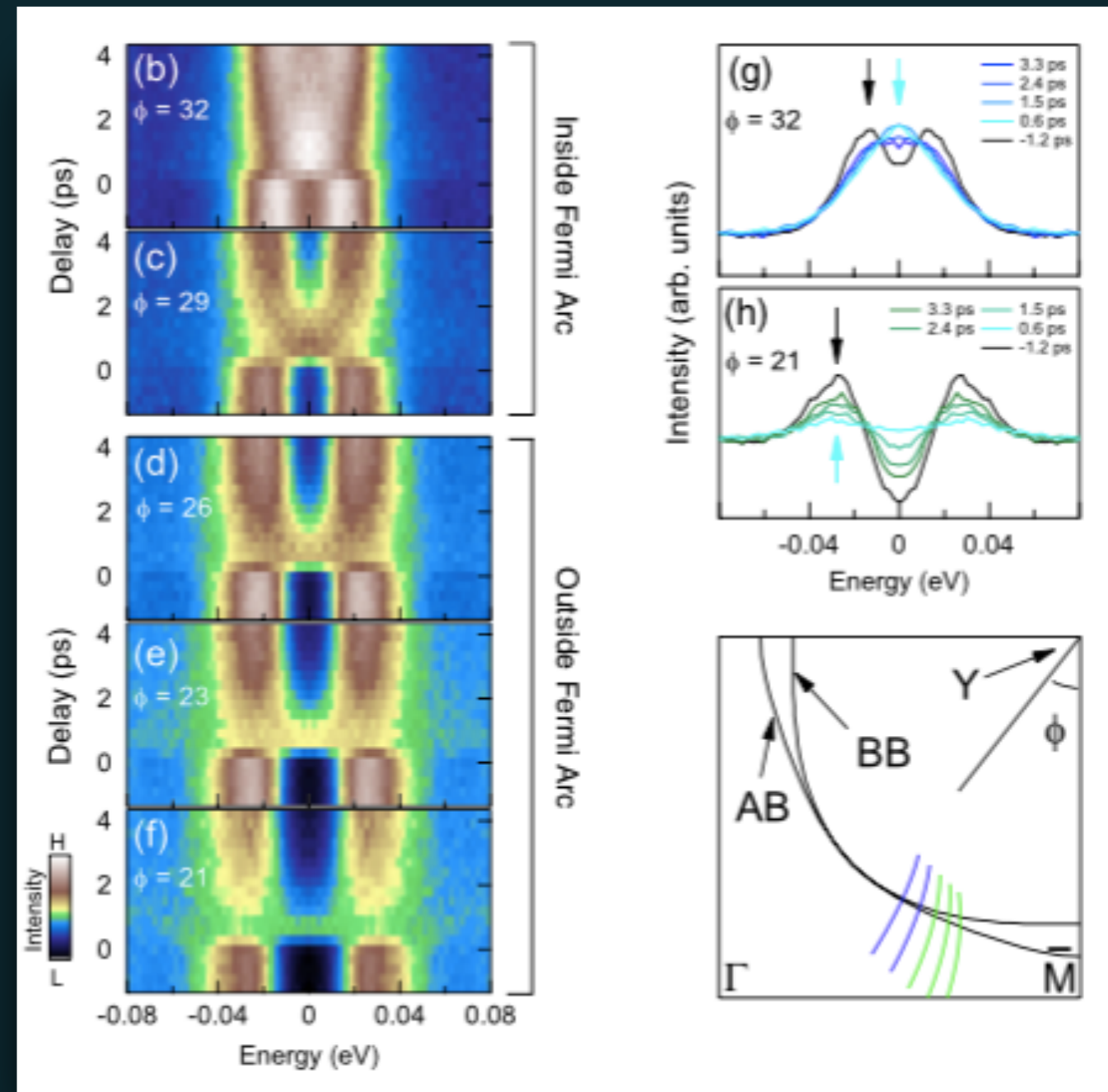
VS

loss of phase coherence

fragility of low-density ( $n_S$ ) superconductors to phase fluctuations:

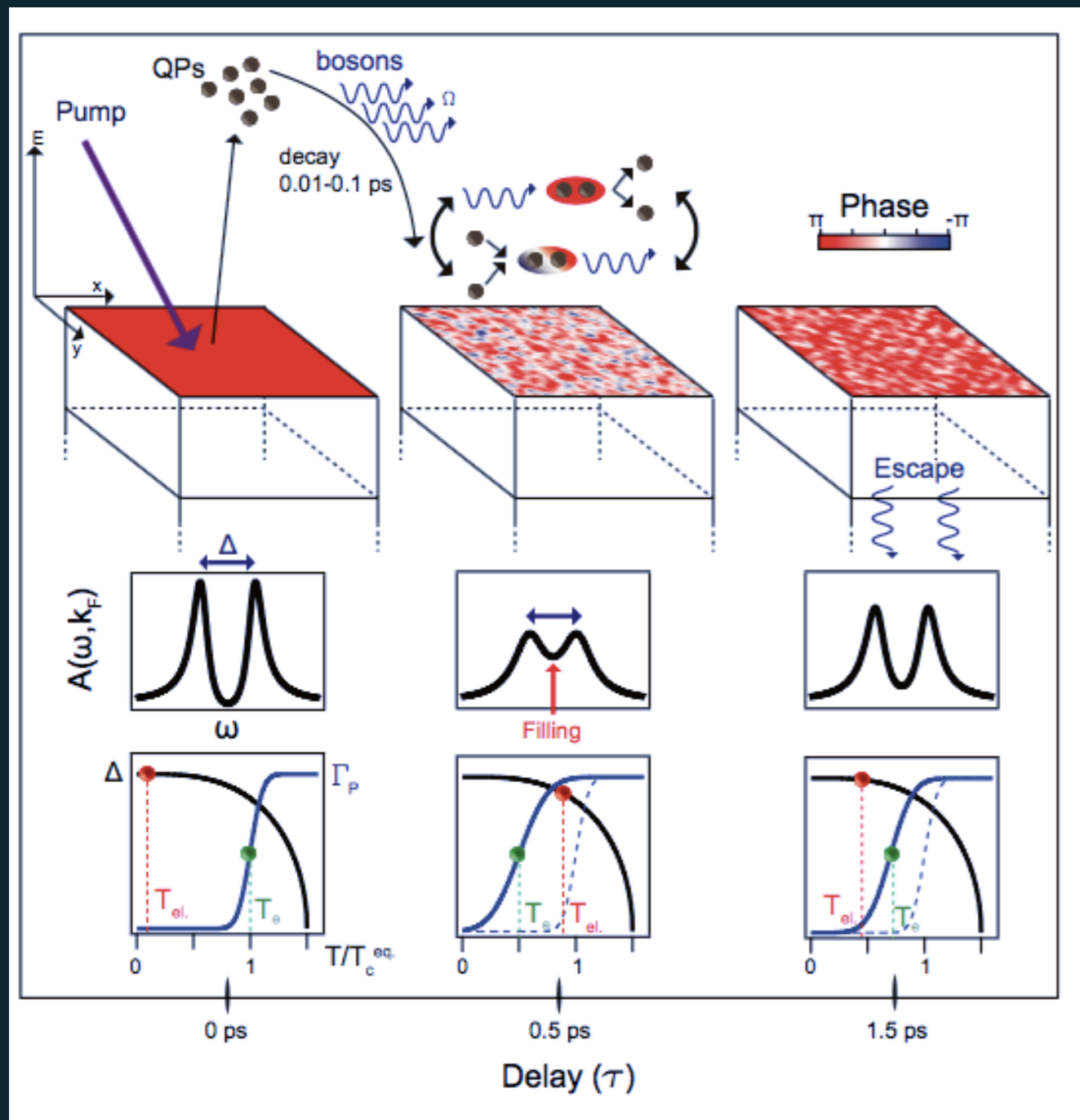
$$\hbar\Omega_{\Theta} = \frac{\hbar^2 n_S(0)a}{2m^*} \simeq k_B T_C$$

V.J. Emery and S.A. Kivelson. *Nature* **374**, 434 (1995)



# destroying phase coherence with light

Collapse of high- $T_C$  superconductivity via ultrafast quenching of the phase coherence in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$



→ fragility of low-density ( $n_s$ ) superconductors to phase fluctuations:

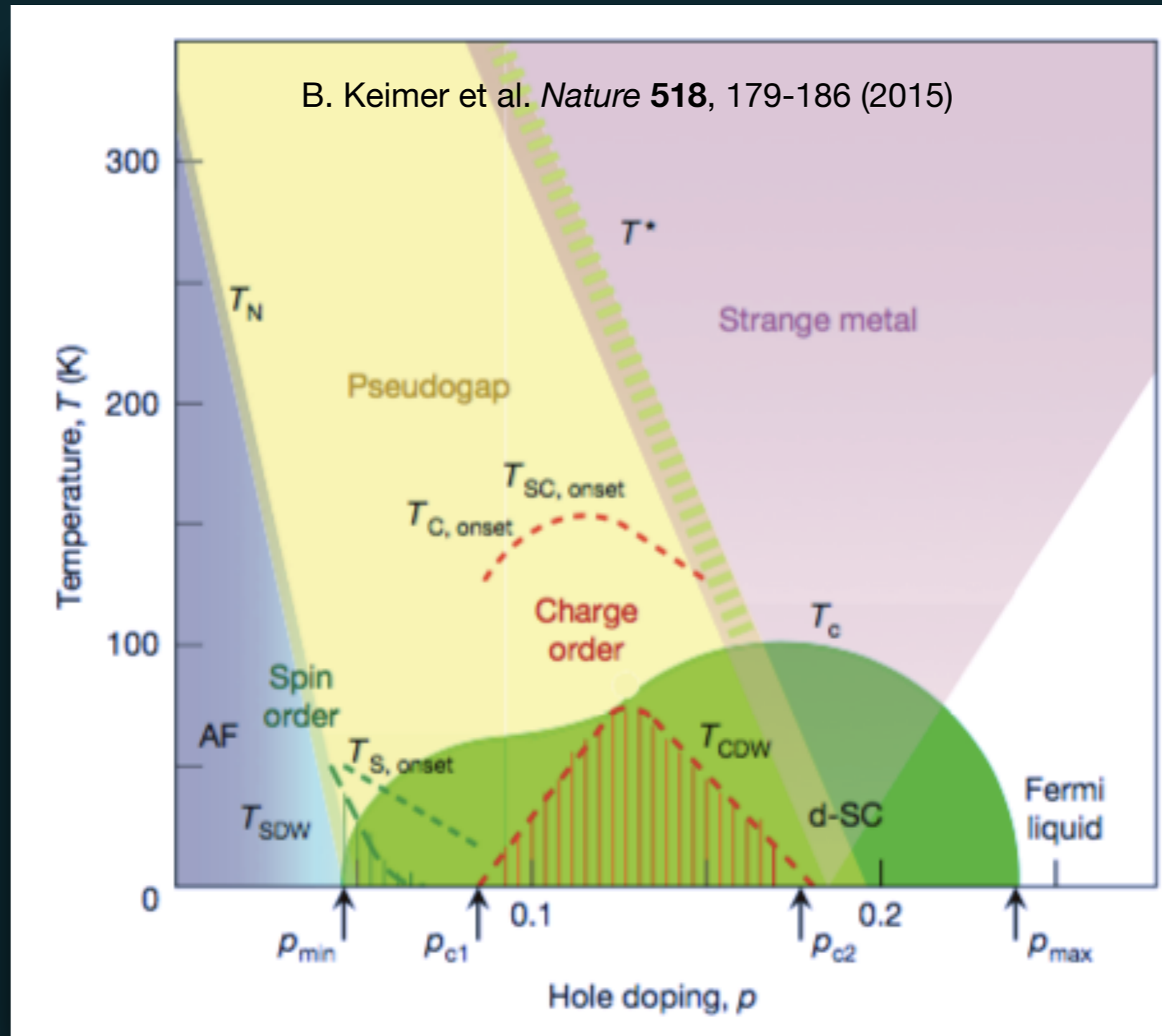
$$\hbar\Omega_{\Theta} = \frac{\hbar^2 n_s(0) a}{2m^*} \simeq k_B T_C$$

V.J. Emery and S.A. Kivelson. *Nature* **374**, 434 (1995)

On the ultrafast timescale it is possible to quench the macroscopic phase coherence without changing the gap and the pairing strength!

⇒ new time-window for exploring quantum phase transitions

# the phase diagram of copper oxides



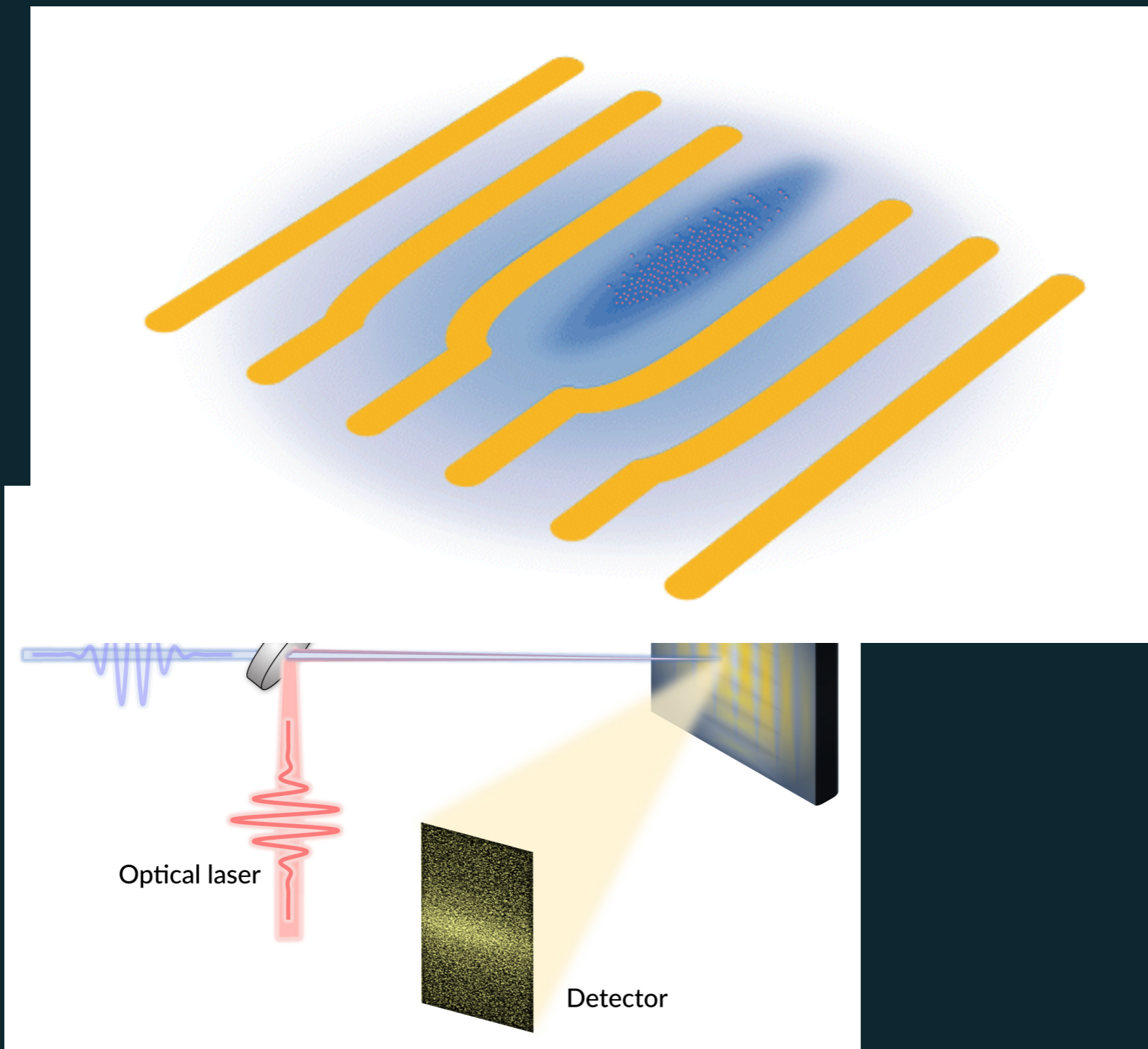
what is the interplay between Charge Density Waves and superconductivity?

---

light pulses to **MANIPULATE**  
materials properties

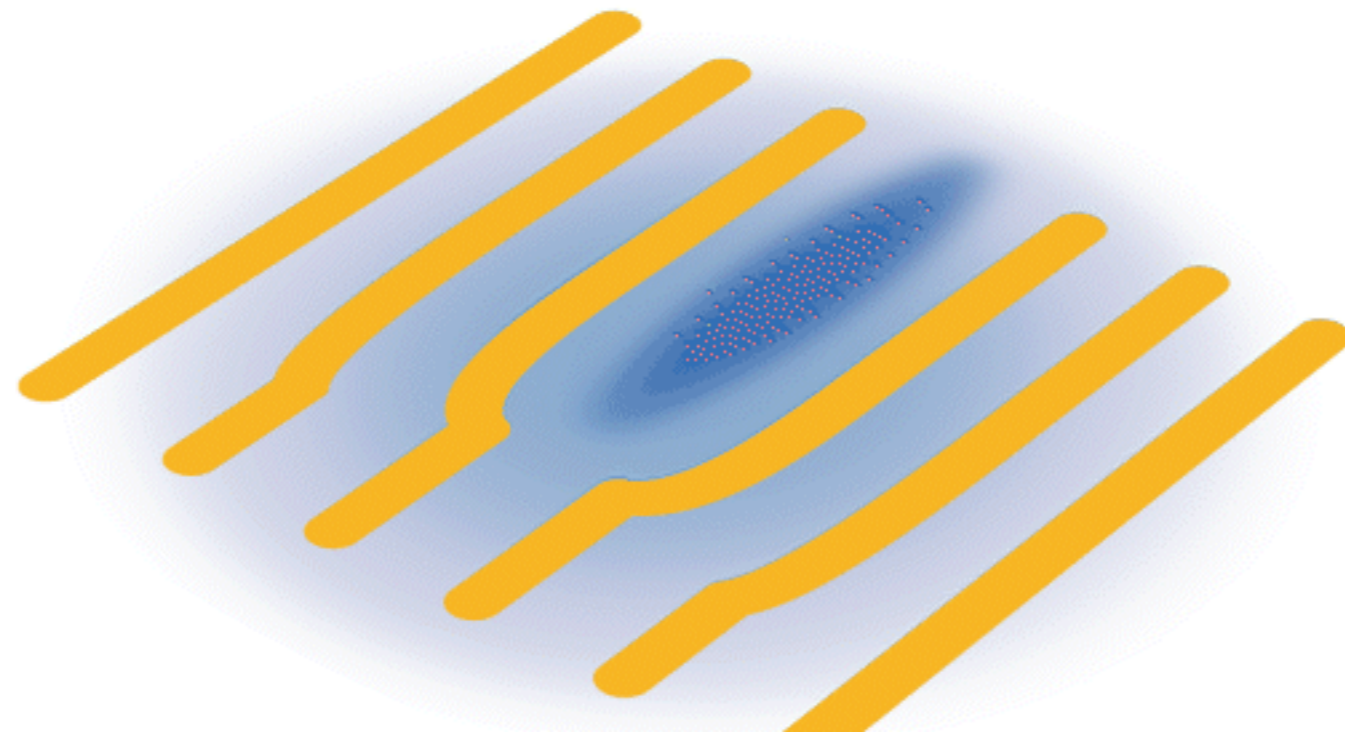
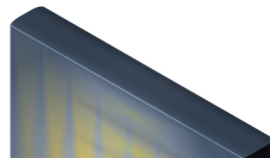
⇒ competition between charge-  
order and superconductivity



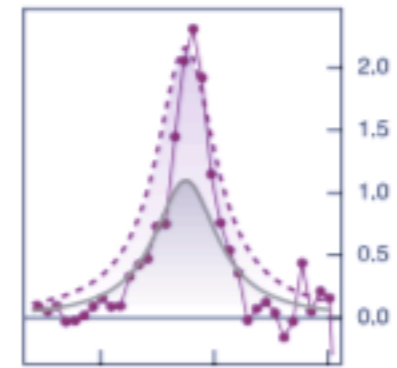
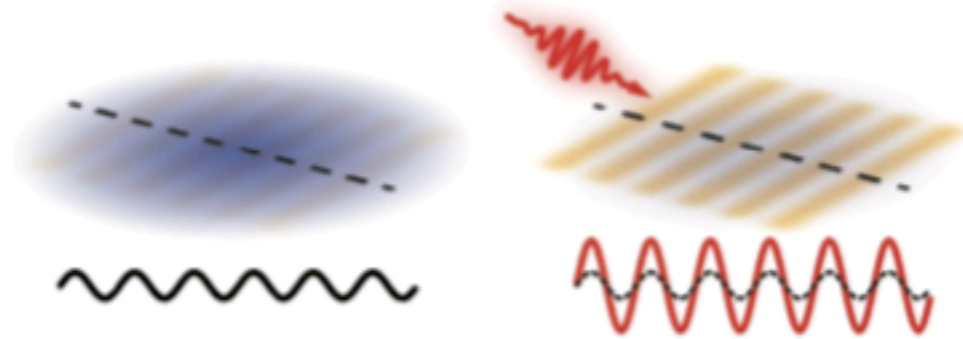


X-ray laser

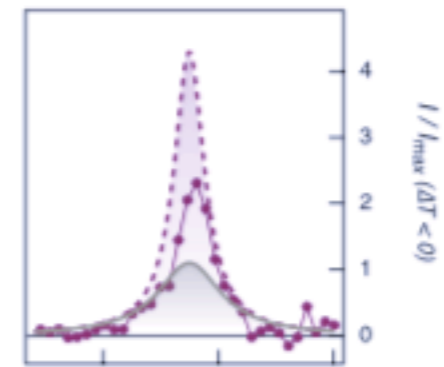
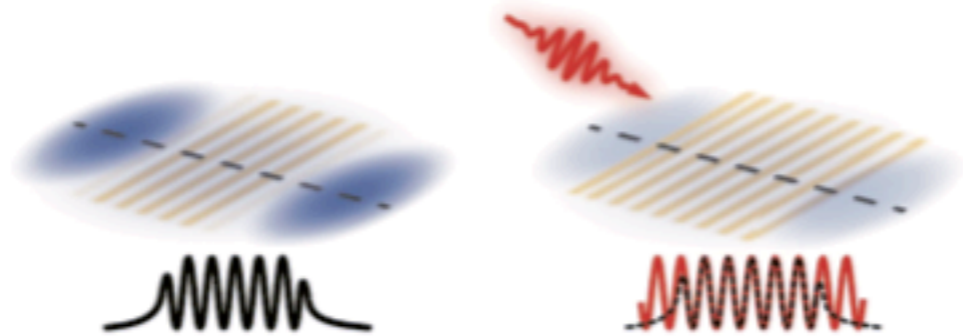
Sample



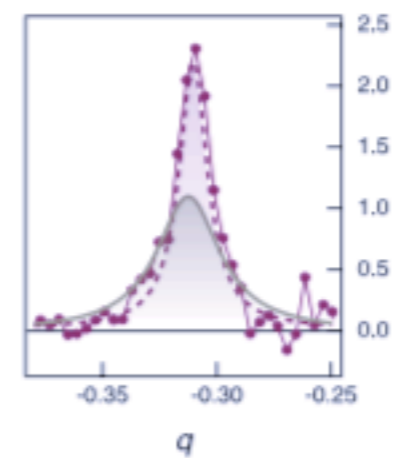
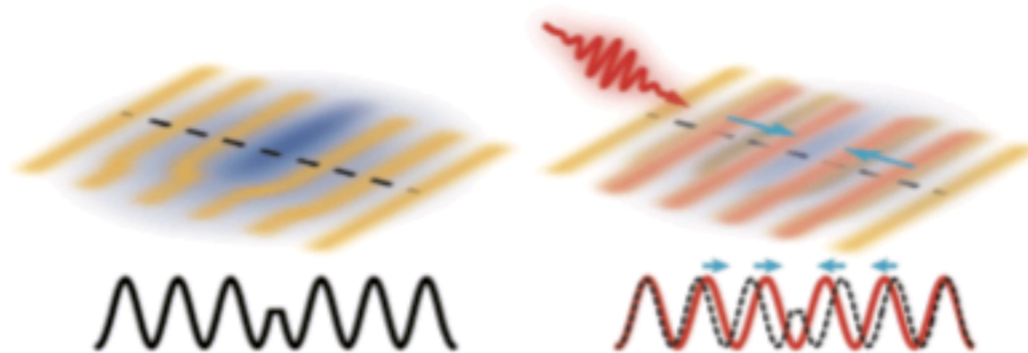
**A** Homogeneous SC-CDW



**B** SC-CDW phase separation



**C** Inhomogeneous SC-induced defect



---

light pulses to **EXCITE**  
specific degrees of freedom

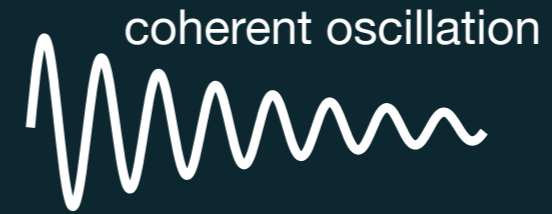
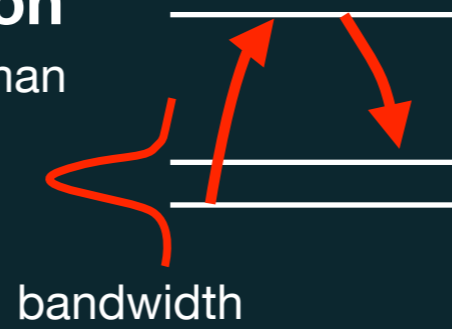
⇒ coherent lattice oscillations  
and Higgs modes

# coherent lattice driving

---

## coherent excitation

impulsive Raman

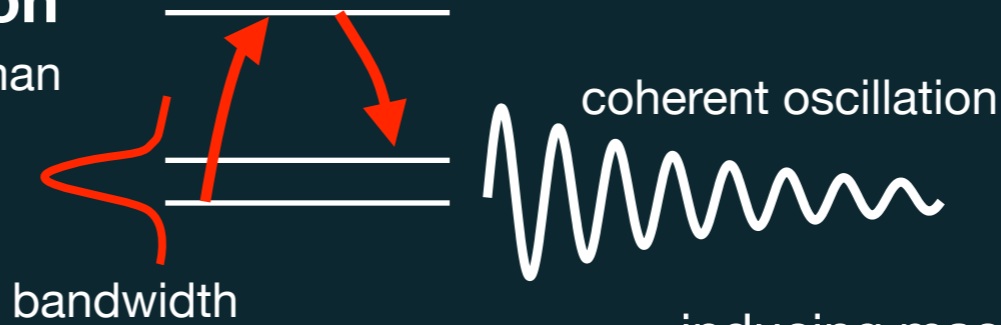




# coherent lattice driving

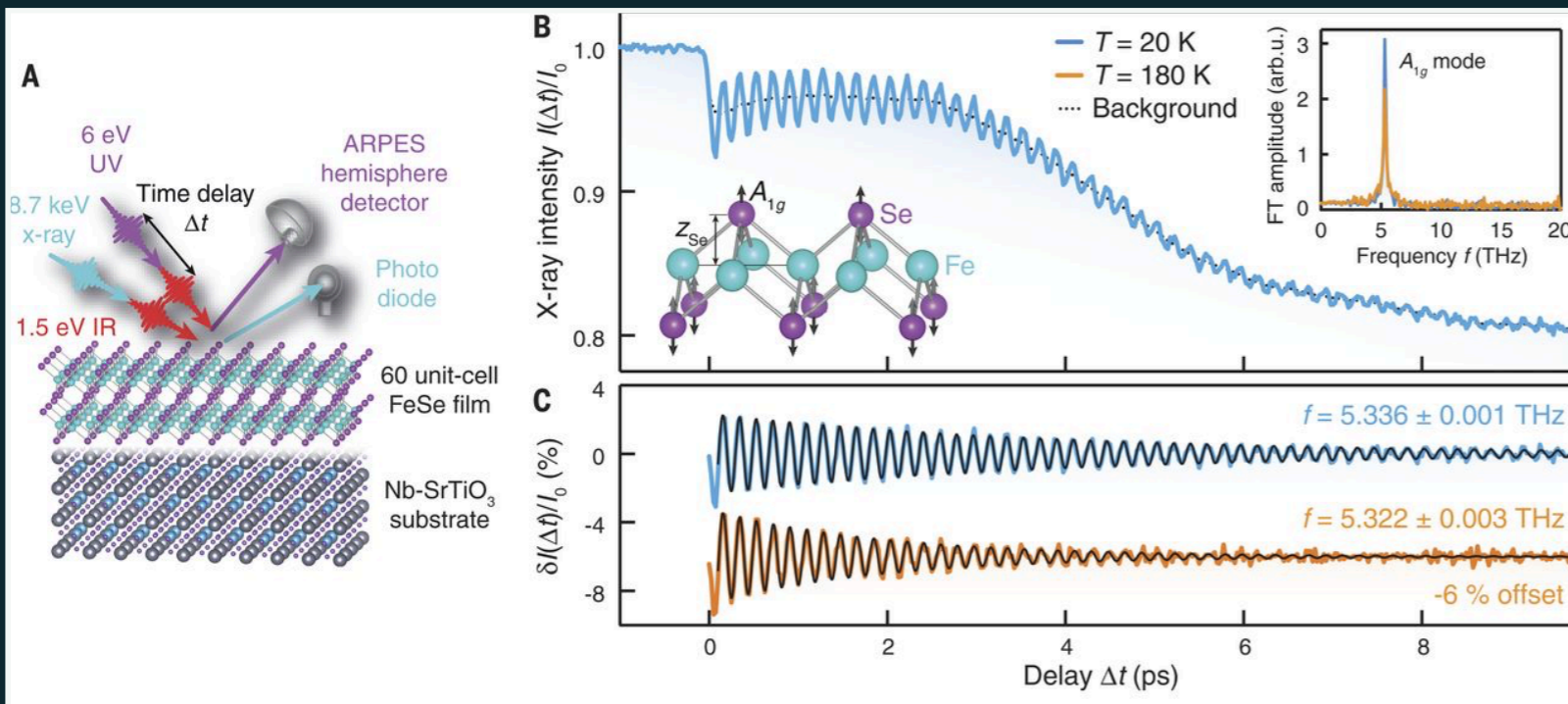
## coherent excitation

impulsive Raman



inducing macroscopic magnetic ordered via coherent phonons in  $\text{BaFe}_2\text{As}_2$

## Coherent phonon oscillation in FeSe



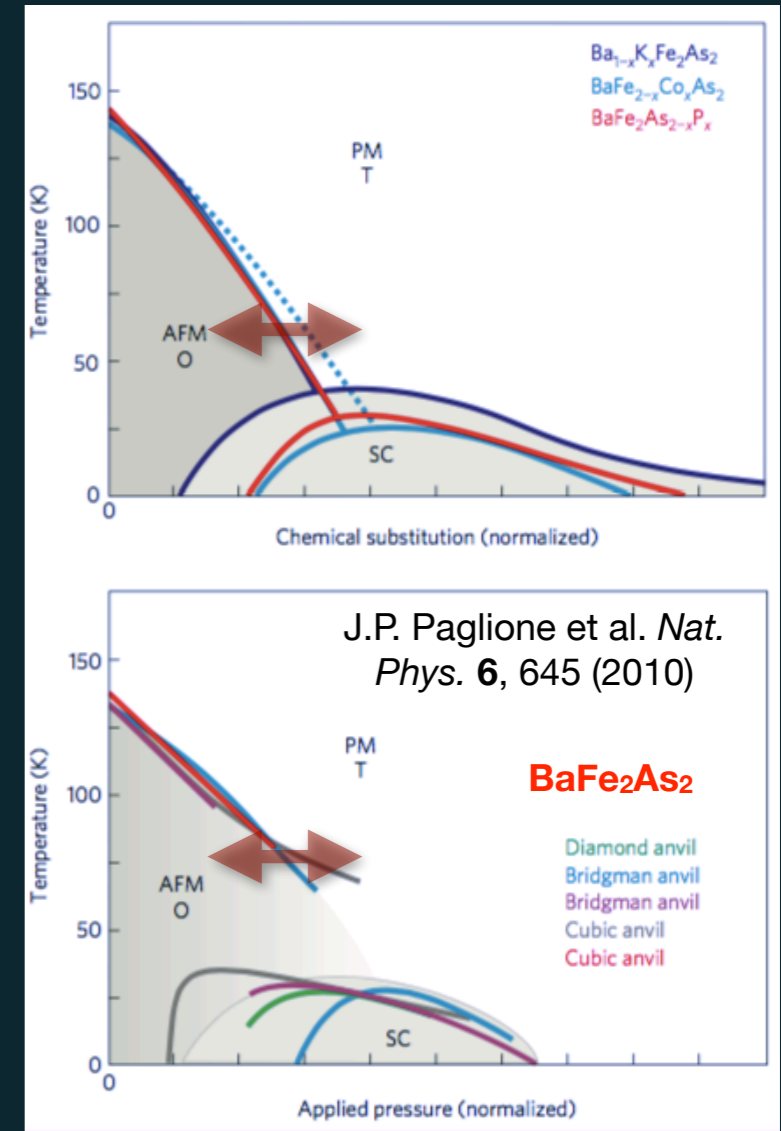
S. Gerber et al. *Science* **357**, 71-75 (2017)

X-rays: lattice oscillation

ARPES: band distortion



deformation potential & e-ph coupling



J.P. Paglione et al. *Nat. Phys.* **6**, 645 (2010)

K.W. Kim et al. *Nature Mater.* **11**, 497-501 (2012)

# order parameter in a symmetry-broken phase

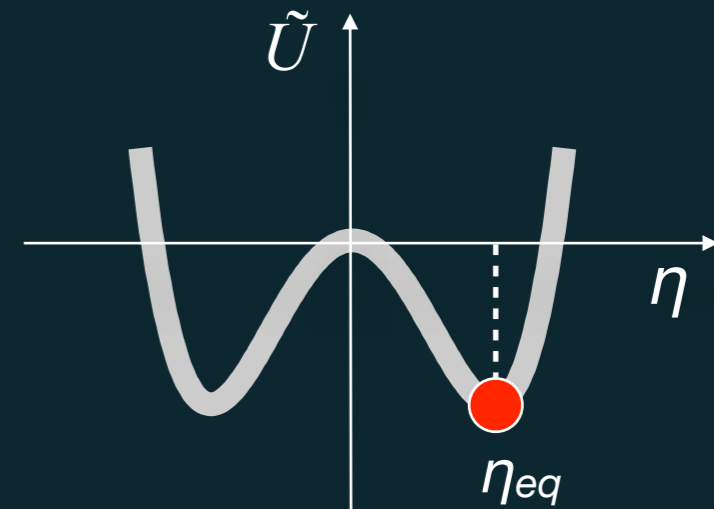
potential landscape based on Ginzburg-Landau functionals

free energy of a  
symmetry-broken  
phase

$\eta$ : order parameter

$$\tilde{U} = \int (\tilde{u}_0 + \frac{A_0(P, T)}{2} |\eta|^2 + \frac{B(P)}{4} |\eta|^4) dv$$

$A_0 = a(P) \cdot (T - T_c)$



# order parameter in a symmetry-broken phase

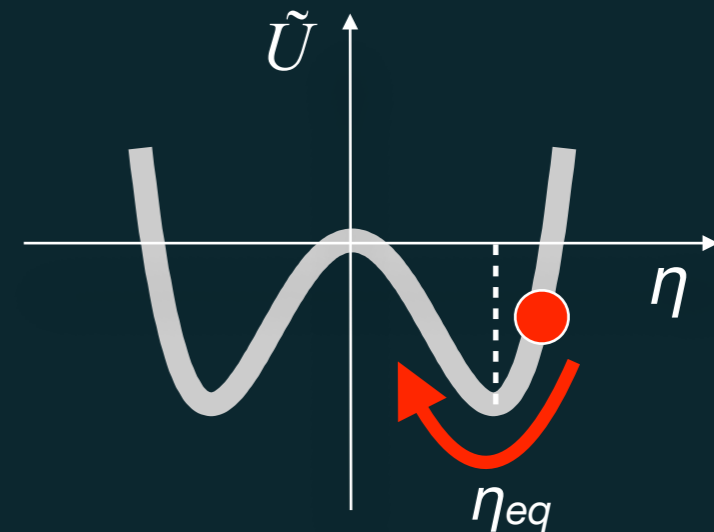
potential landscape based on Ginzburg-Landau functionals

free energy of a  
symmetry-broken  
phase

$$\tilde{U} = \int (\tilde{u}_0 + \frac{A_0(P, T)}{2} |\eta|^2 + \frac{B(P)}{4} |\eta|^4) dv$$

$$A_0 = a(P) \cdot (T - T_c)$$

$\eta$ : order parameter



motion equation for  
time-dependent  
perturbation

$$\frac{d^2 \eta}{dt^2} + \alpha \omega_0 \frac{d\eta}{dt} = -\omega_0^2 \frac{\partial \tilde{U}}{\partial \eta}$$

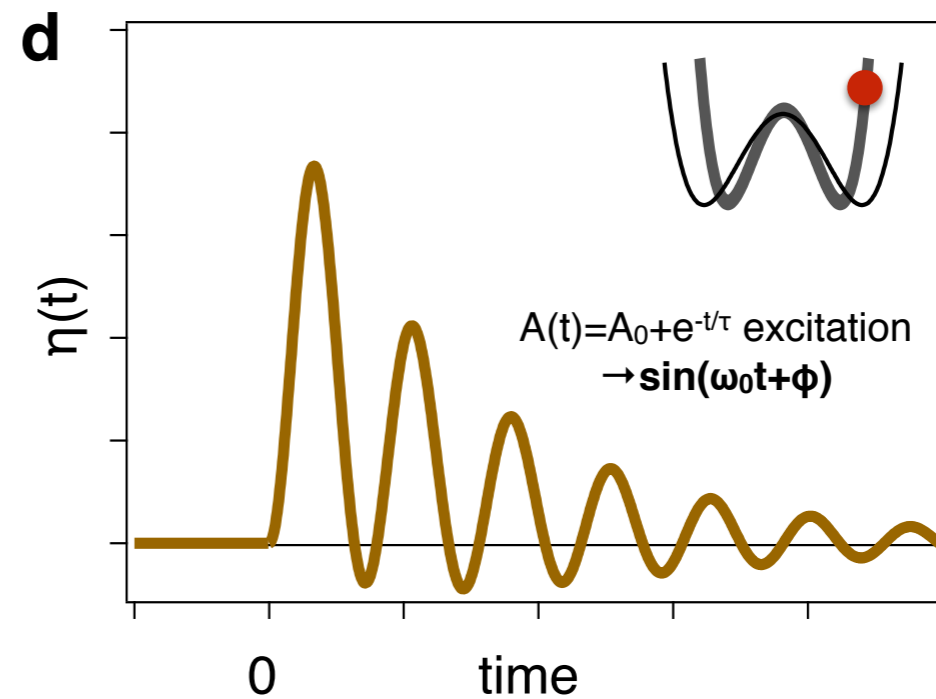
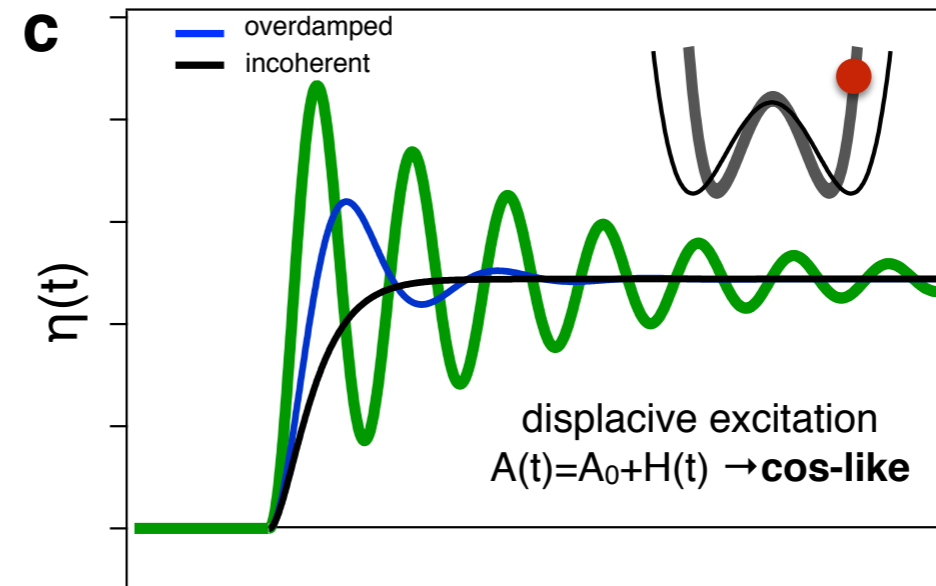
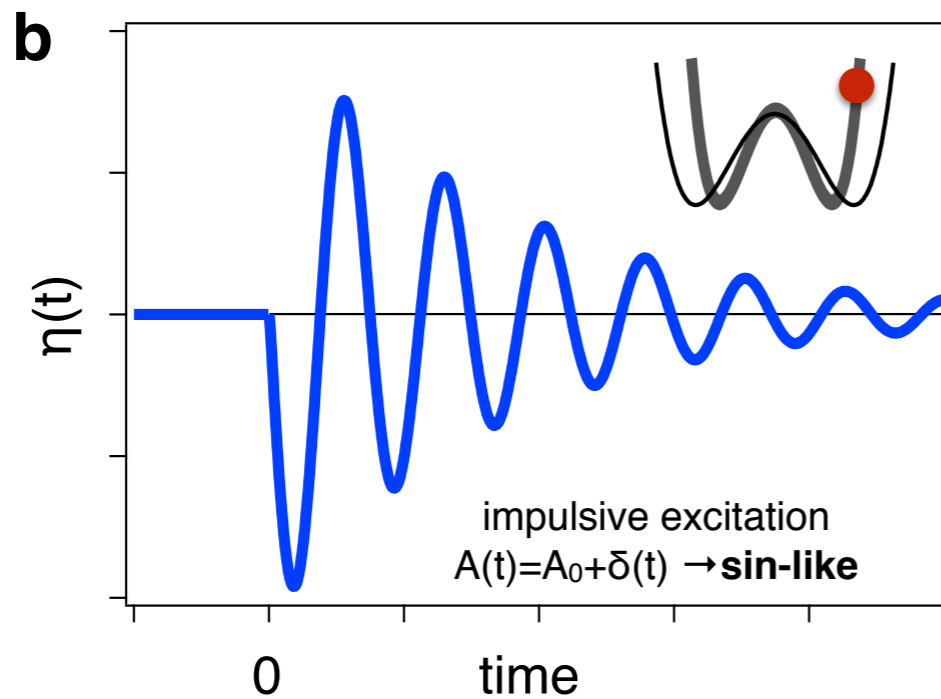
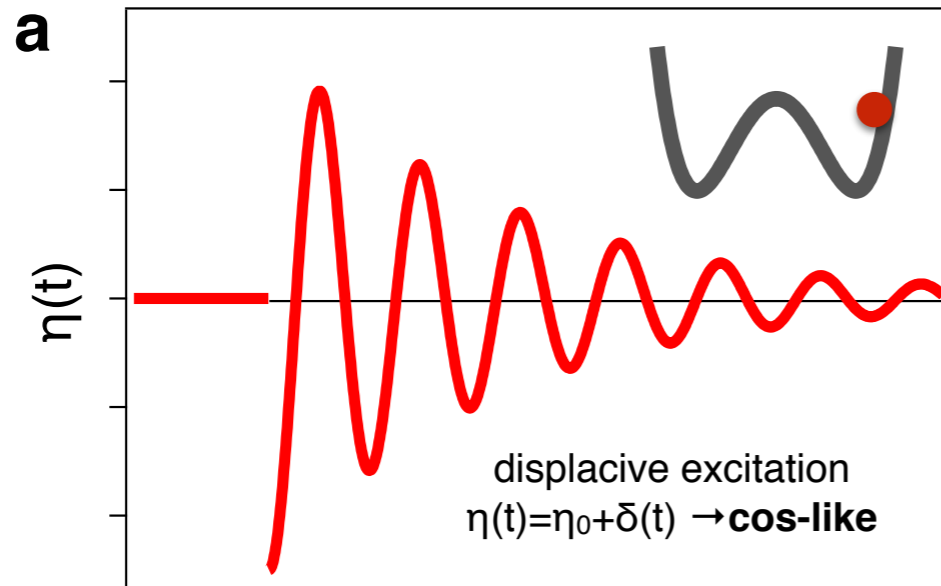
damping  
(decoherence)

intrinsic  
frequency of  
the mode

perturbation of the  
potential

# order parameter in a symmetry-broken phase

## different excitation schemes

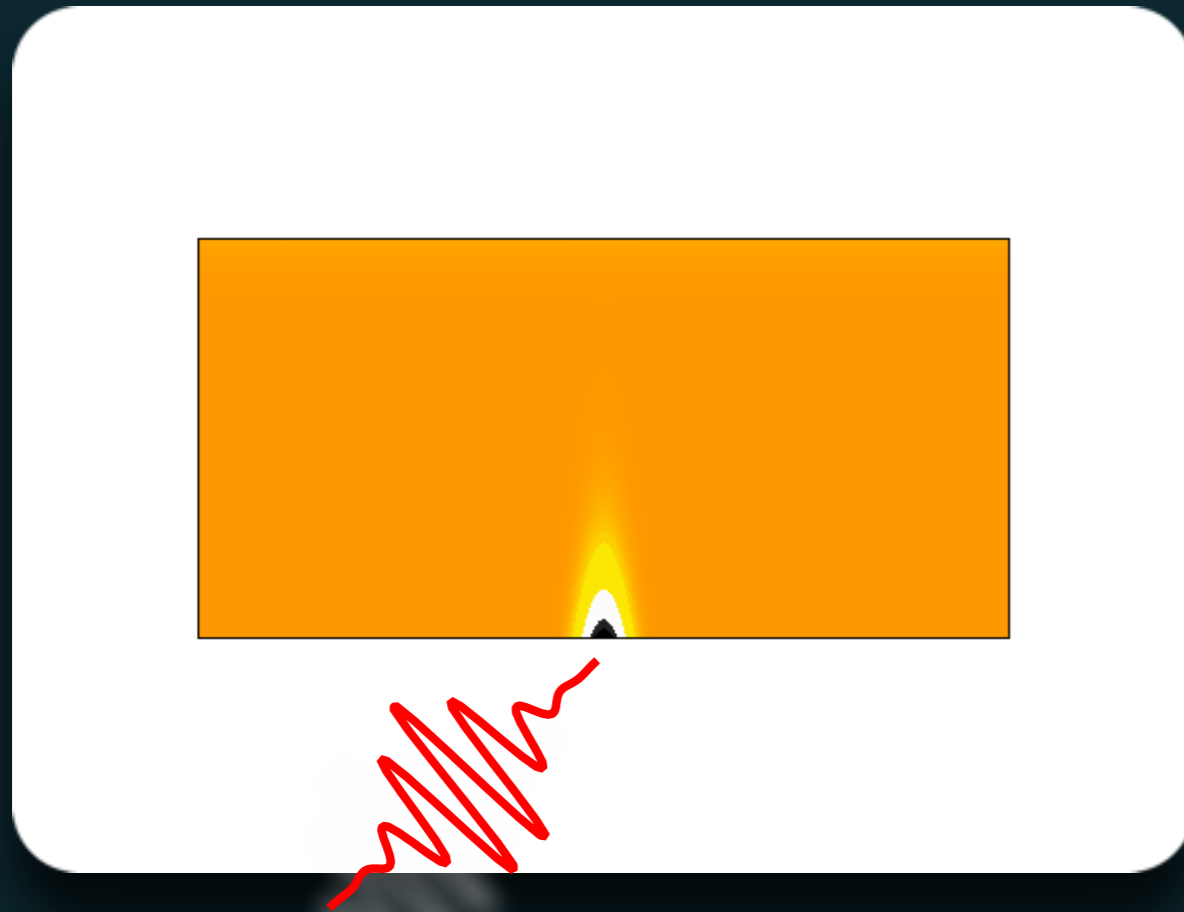


# coherent excitation of amplitude modes with THz

---

looking for Higgs modes (amplitude modes) in symmetry-broken phases

wave-behaviour of the  
amplitude mode



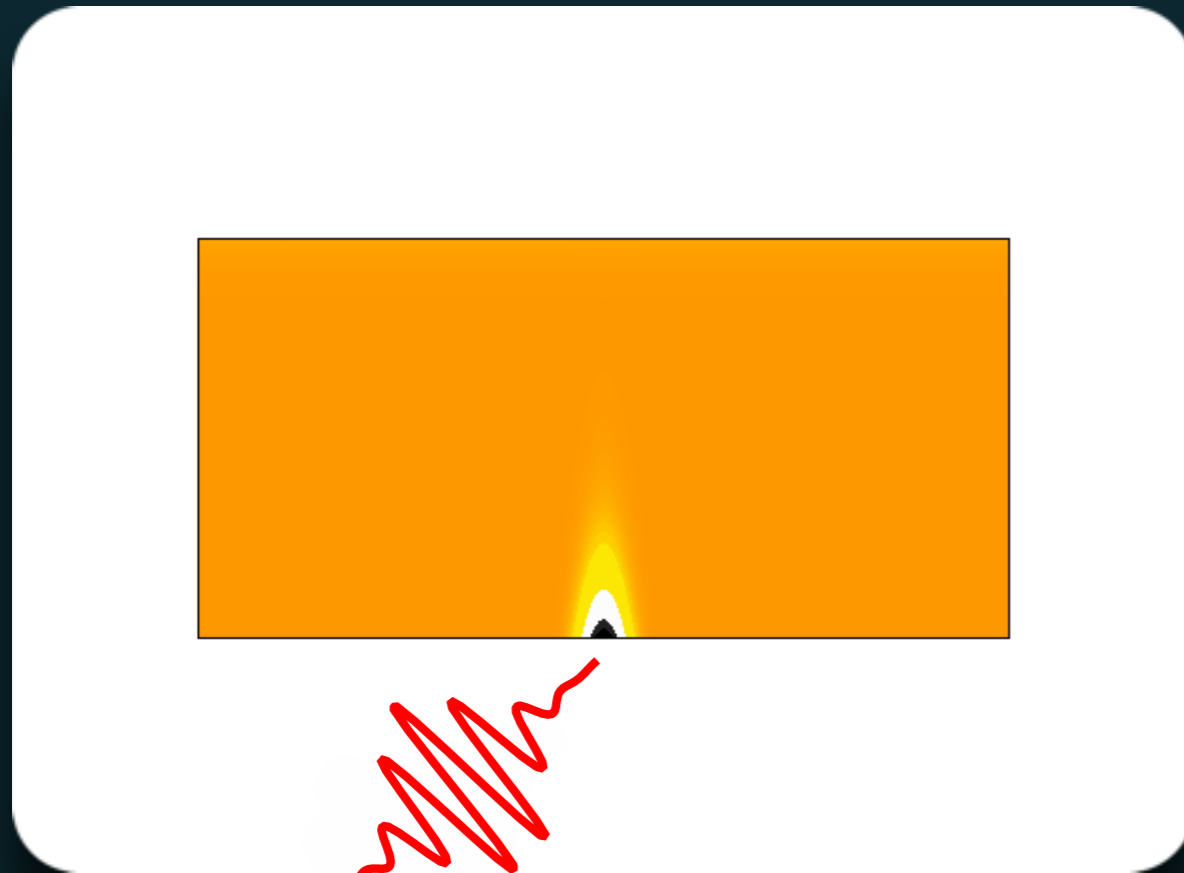
THz excitation of the  
amplitude mode



# coherent excitation of amplitude modes with THz

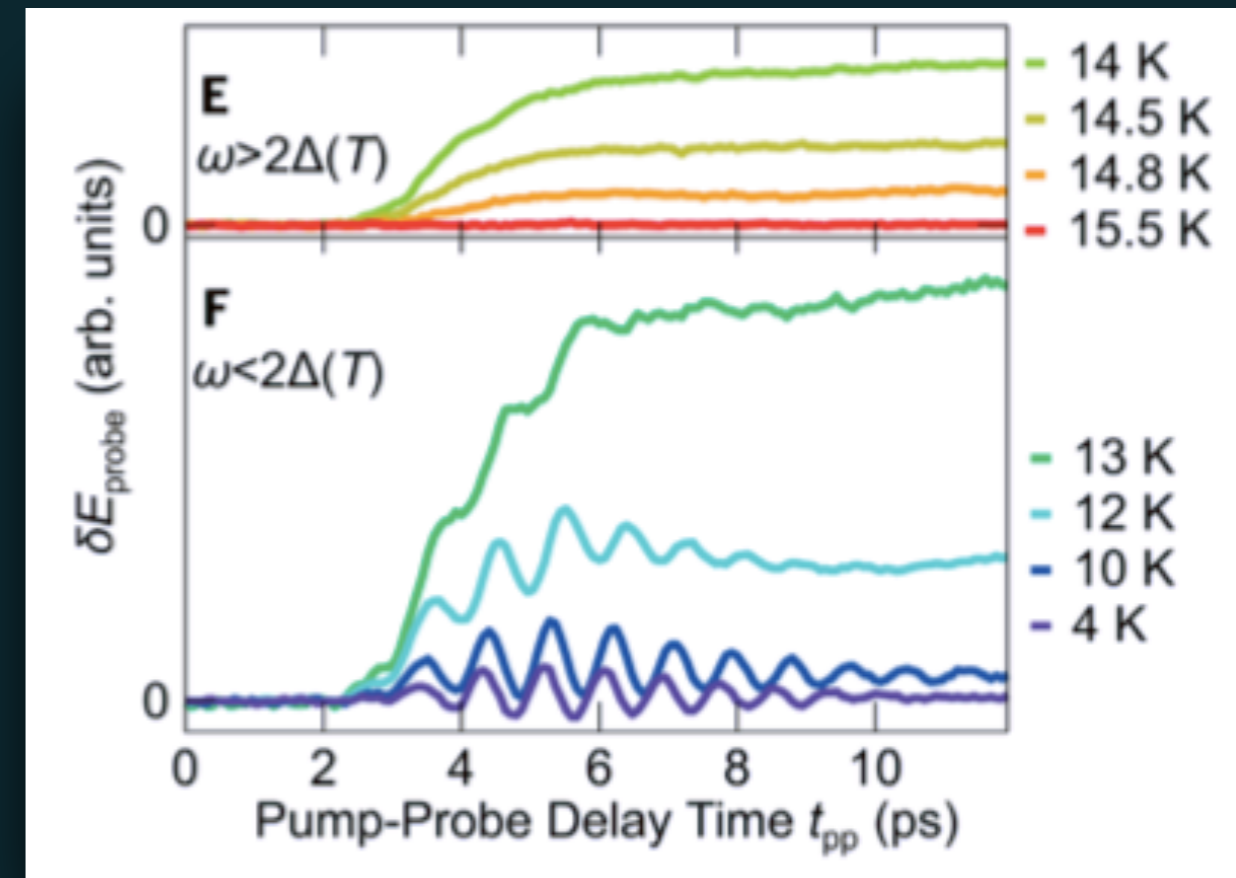
looking for Higgs modes (amplitude modes) in symmetry-broken phases

wave-behaviour of the amplitude mode



THz excitation of the amplitude mode

Higgs mode (amplitude mode of the superconducting order parameter) in  $\text{Nb}_{1-x}\text{Ti}_x\text{N}$



R. Matsunaga et al, *Phys. Rev. Lett.* **111**, 057002 (2013)

R. Matsunaga et al, *Science* **345**, 1145-1149 (2014)

T. Cea et al, *Phys. Rev. Lett.* **115**, 157002 (2015)

THz pump-THz probe  
(below-gap excitation)


# outline

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- Ultrafast spectroscopies to investigate the electron dynamics in materials  
C. Giannetti et al. *Advances in Physics* **65**:2, 58-238 (2016)
- light pulses to UNDERSTAND equilibrium properties  
⇒ pairing glue in high- $T_c$  superconductors
- light pulses to EXCITE specific degrees of freedom  
⇒ coherent lattice oscillations and Higgs modes
- light pulses to MANIPULATE materials properties  
⇒ hidden states, transient photo-enhanced superconductivity

# Looking for a PhD position?

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PH.D. SCHOOL 

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RESEARCH PROJECTS 

ADMISSION

FACULTY


NEWS

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**KU Leuven, Belgium (KU)**  
**Notre Dame, USA (ND)**  
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Thank you!