# Snapshots of the retarded interaction of charge carriers with ultrafast fluctuations in cuprates


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i-*Lamp*
*(Interdisciplinary laboratories for advanced materials physics)*


GO FAST
*GOverning ultraFAST the conductivity of correlated material*

SEVENTH FRAMEWORK PROGRAMME

# Outline

- Ultrafast spectroscopies of quantum materials

J. Orenstein, *Phys. Today* 65, 44 (2012)

- The “glue” and the retarded-interaction problem in high-$T_c$ superconductors

P.W. Anderson, *Science* **317**, 1705 (2007) and reply from D.J. Scalapino

- Snapshots of the retarded interaction with ultrafast fluctuations via 10 fs pulses

S. Dal Conte et al., *Science* **335**, 1600 (2012)
S. Dal Conte et al. *submitted to Nature Physics*

- Towards a non-equilibrium phase diagram of cuprates

F. Cilento et al., *Nature Communications* **5:4353** (2014)

# Outline

•Ultrafast spectroscopies of quantum materials
J. Orenstein, *Phys. Today* 65, 44 (2012)

•The “glue” and retarded-interaction problem in high-$T_c$ superconductors
P.W. Anderson, *Science* **317**, 1705 (2007) and reply from D.J. Scalapino

•The “glue” problem in (time-resolved) optics

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# Hunting for fast details

A movie in 4 frames:

http://moviesinframes.tumblr.com

# Hunting for fast details

A movie in 4 frames:

http://moviesinframes.tumblr.com


Which sequence is

*Inglorious Basterds* from Q. Tarantino?

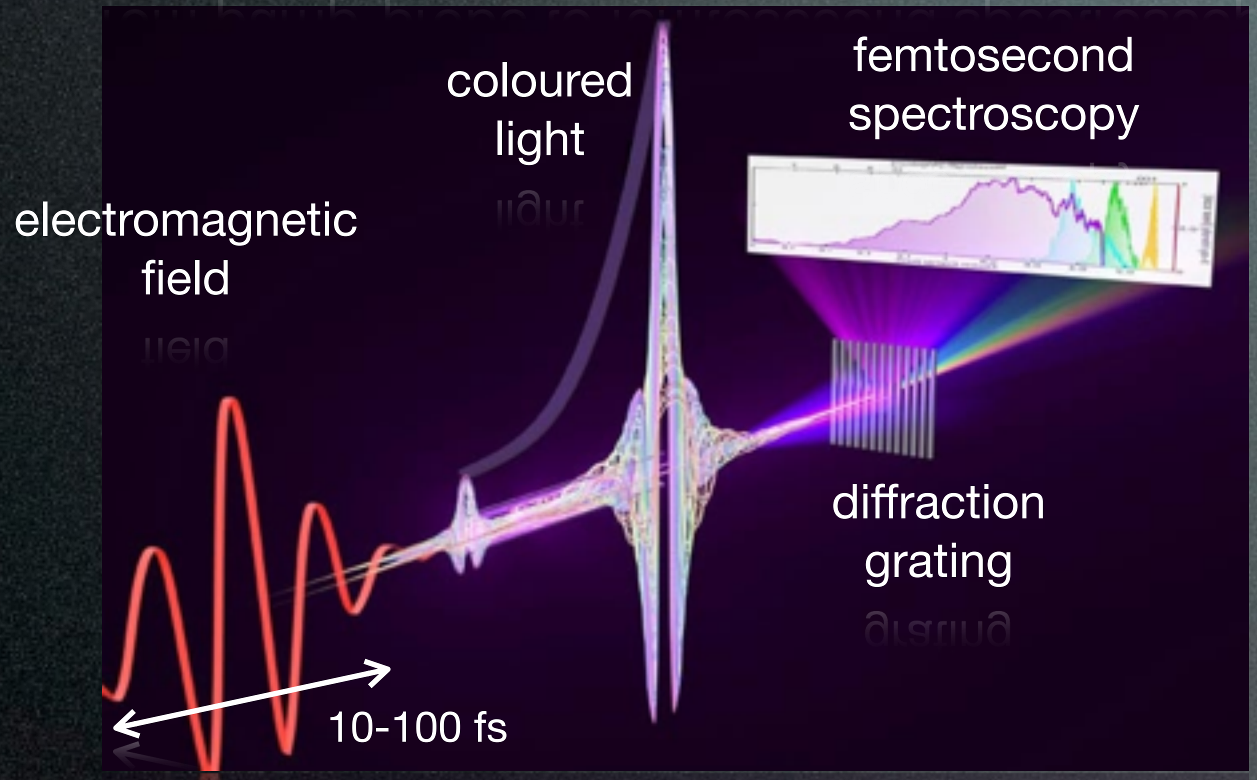
# Time-resolved optics

A movie in 1000 frames:

ultrafast shutter for ultrafast details...

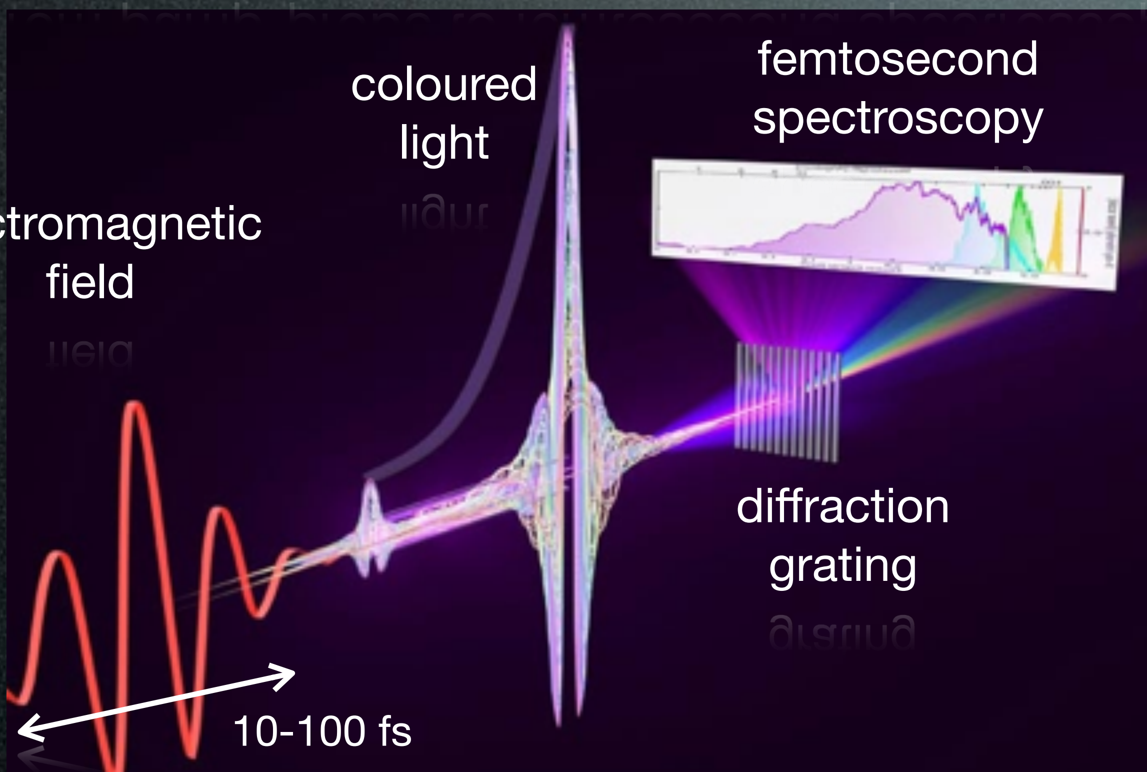
# Time-resolved optical spectroscopy

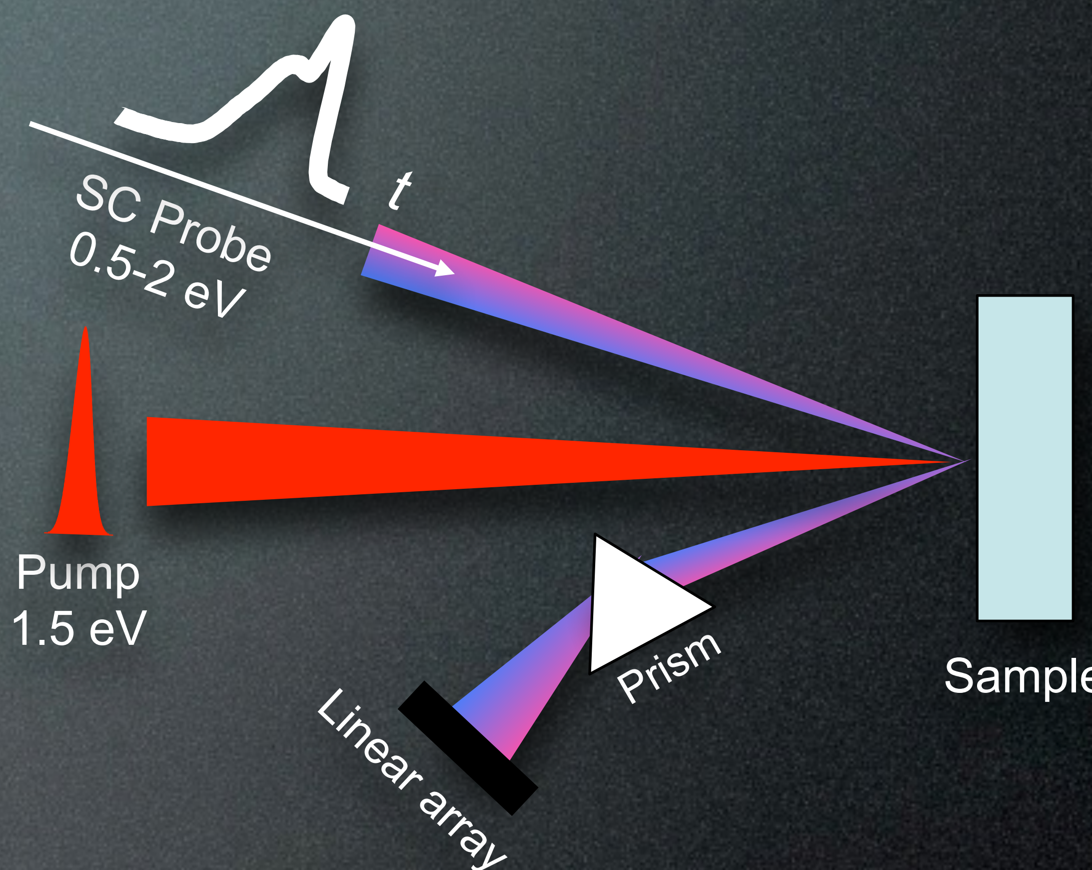
from pump-probe to femtosecond spectroscopy



# Time-resolved optical spectroscopy

from pump-probe to femtosecond spectroscopy



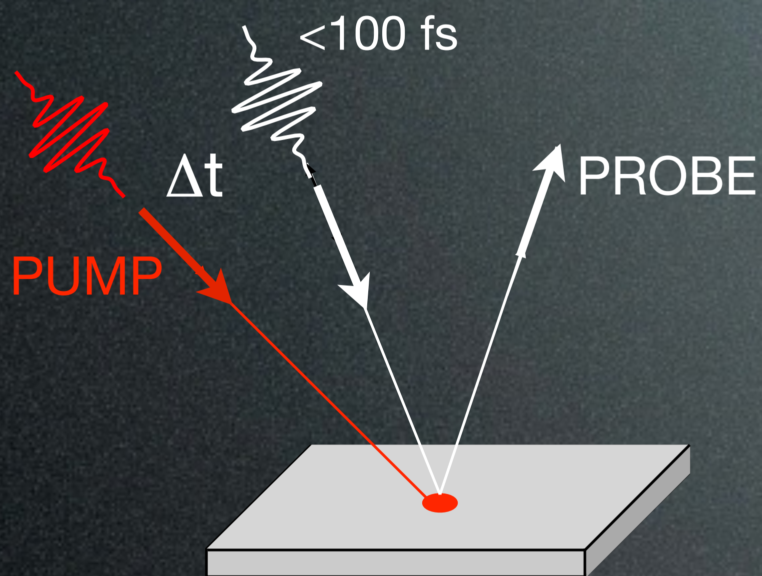




$$\frac{\delta R}{R}(\omega, t) = \frac{R_{exc}(\omega, t) - R_{eq}(\omega)}{R_{eq}(\omega)}$$

time+frequency information

# Time-resolved optical spectroscopy

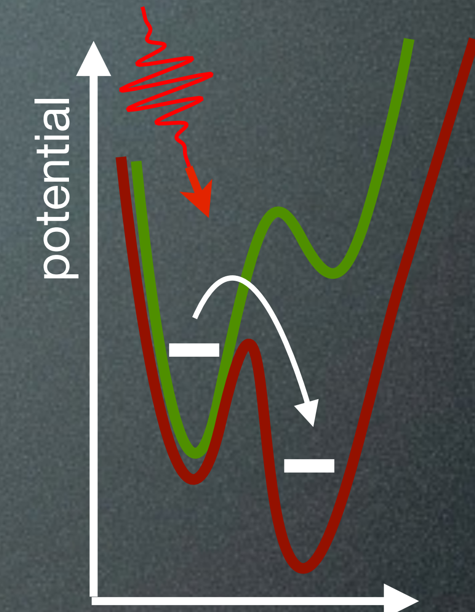
pump probe on quantum materials




J. Orenstein, *Phys. Today* 65, 44 (2012)

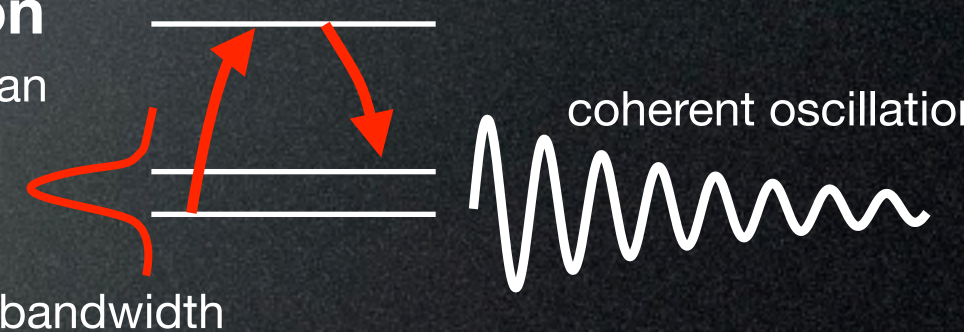
**optical control**

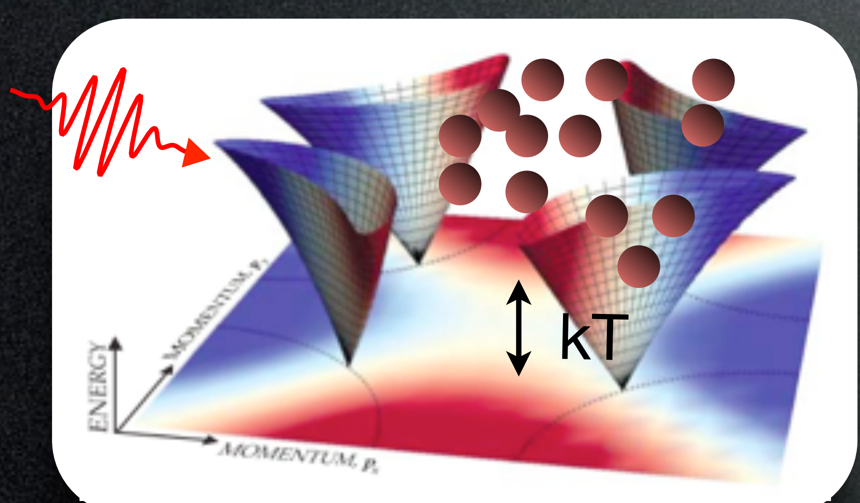
new transient ground state




**coherent excitation**

impulsive Raman

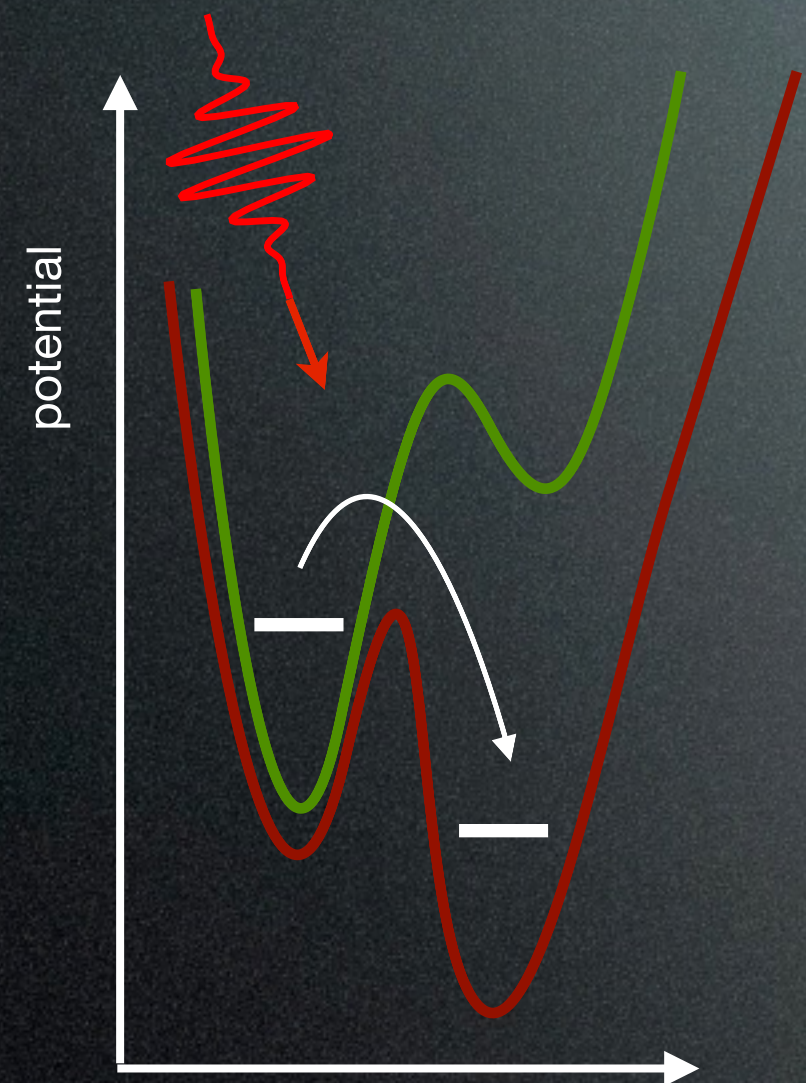


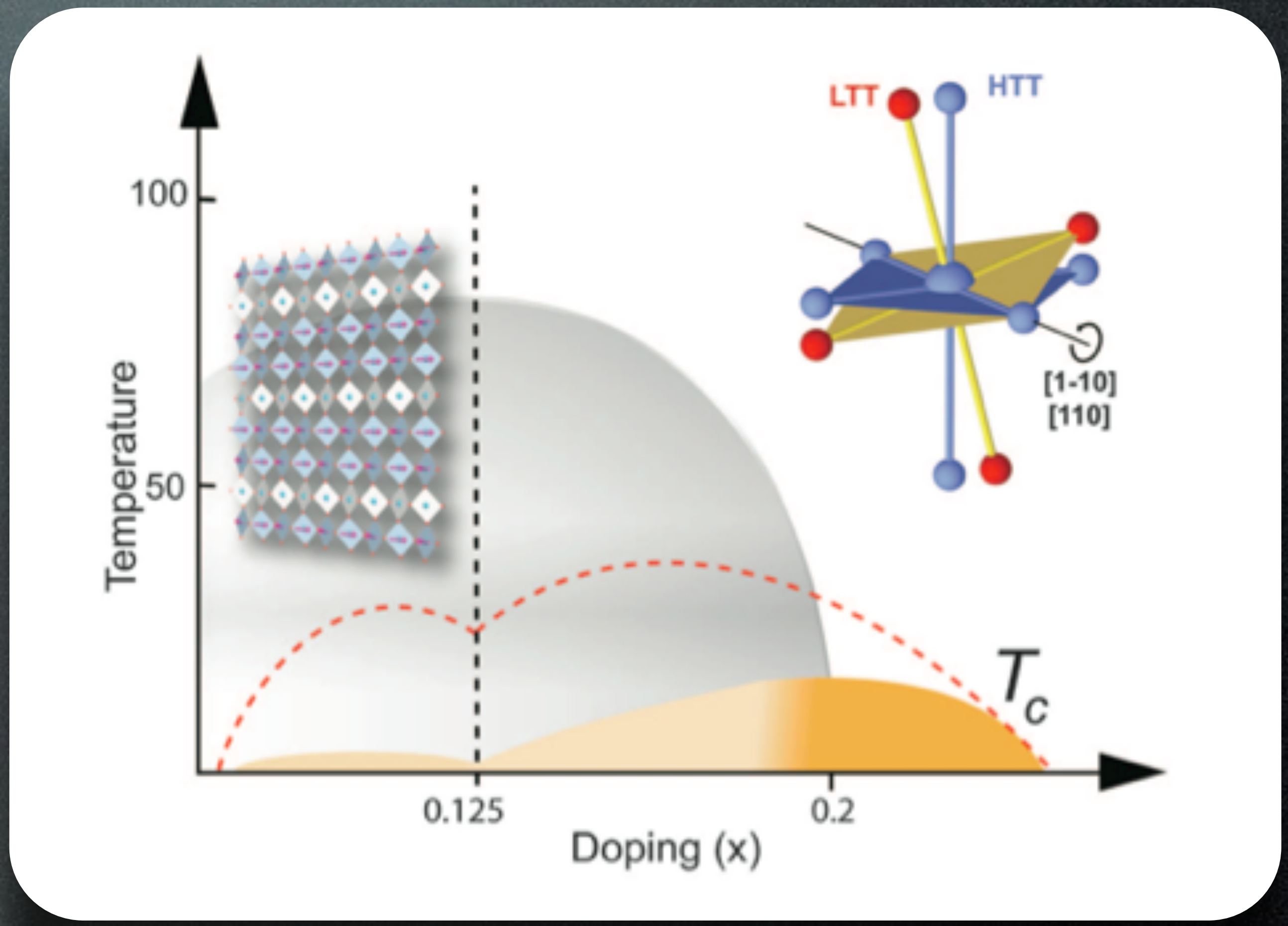

**non-equilibrium spectroscopy**

recovery of the ground state

# Optical control



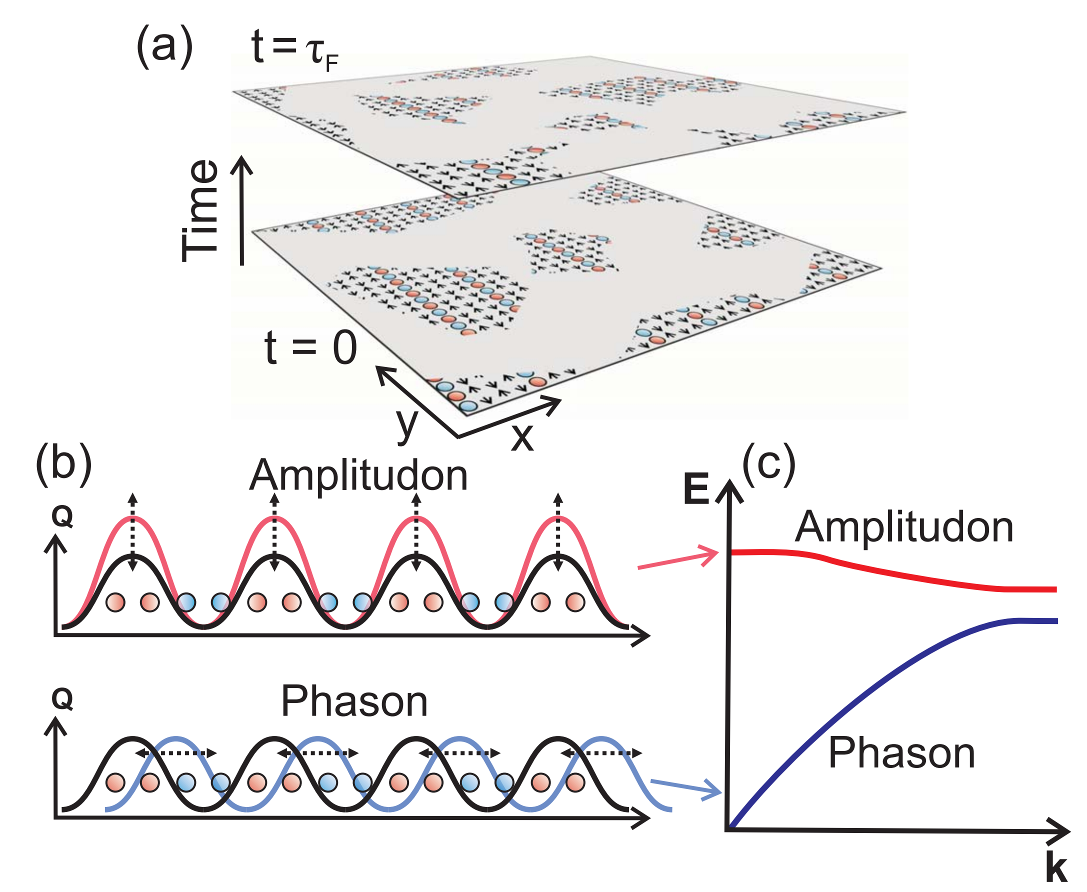

Transient superconductivity by removing a competing order



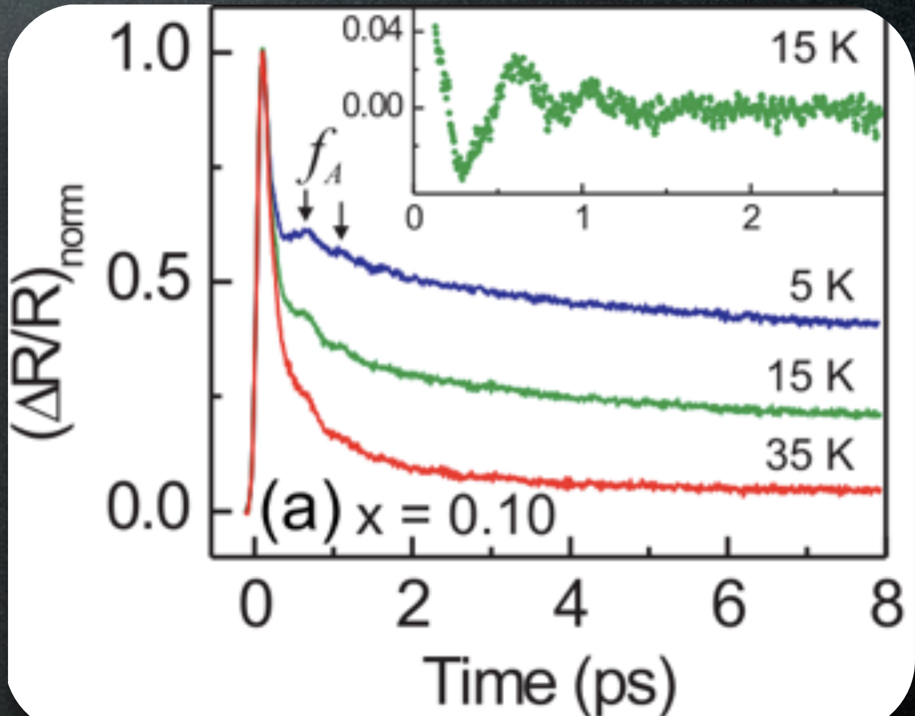

D. Fausti et al., *Science* **331**, 189 (2011)

# Coherent excitation

**coherent excitation**
impulsive Raman


Fluctuating Charge-Density-Wave in UD $La_{1.9}Sr_{0.1}CuO_4$




D. Torchinsky et al, *Nature Materials* 2013

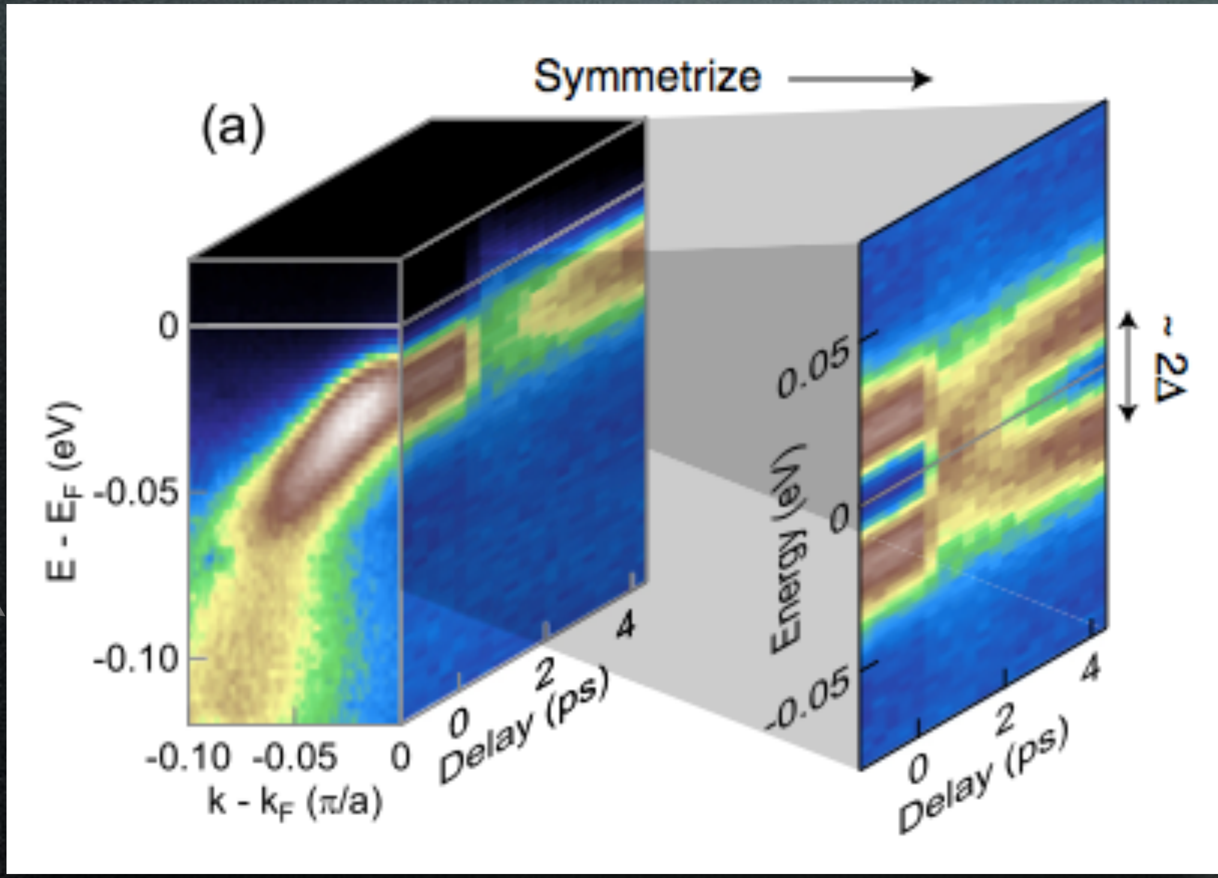
see talks: N. Gedik and F. Carbone

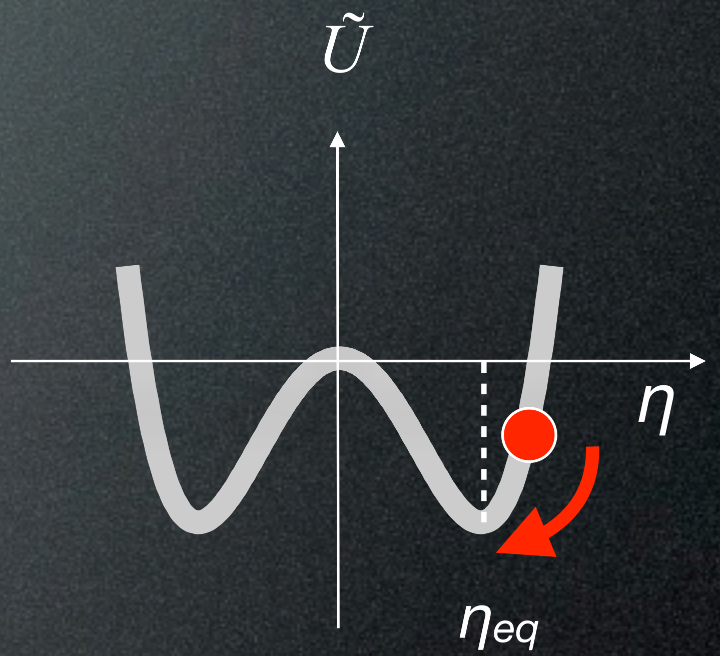
# Non-equilibrium spectroscopy

disentangling intertwined degrees of freedom by their dynamics

Gap dynamics in superconducting copper oxides

Time-resolved photoemission spectroscopy




C. Smallwood et al. et al, *Phys. Rev. B* **89,** 115126 (2014)

see A. Lanzara's talk

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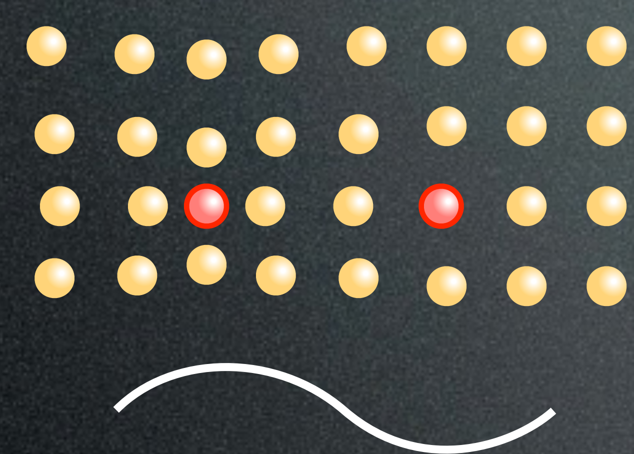
# retarded vs instantaneous interaction

## BCS

$$\hat{H} = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} \hat{c}^{\dagger}_{\mathbf{k}} \hat{c}_{\mathbf{k}} + \sum_{\mathbf{k},\mathbf{Q}} M_Q \hat{c}^{\dagger}_{\mathbf{k}+\mathbf{Q}} \hat{c}_{\mathbf{k}} (\hat{b}_{\mathbf{Q}} + \hat{b}^{\dagger}_{-\mathbf{Q}})$$

boson

glue: lattice distortion

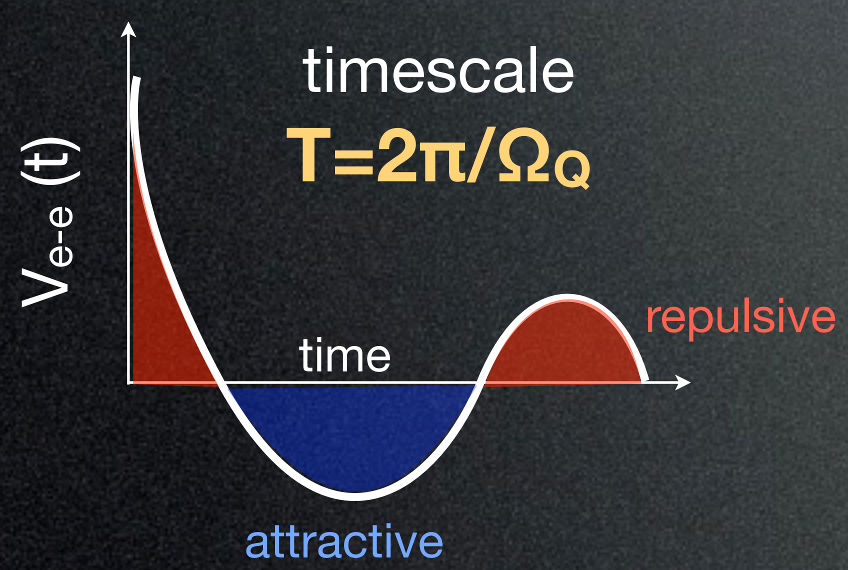


space

λ=2π/|Q|

timescale

**T=2π/Ω$_Q$**



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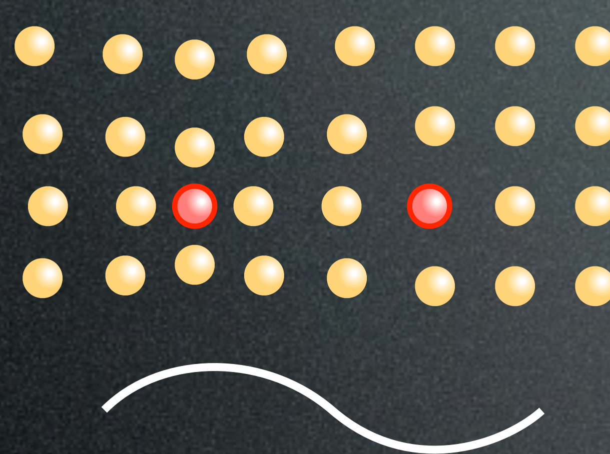
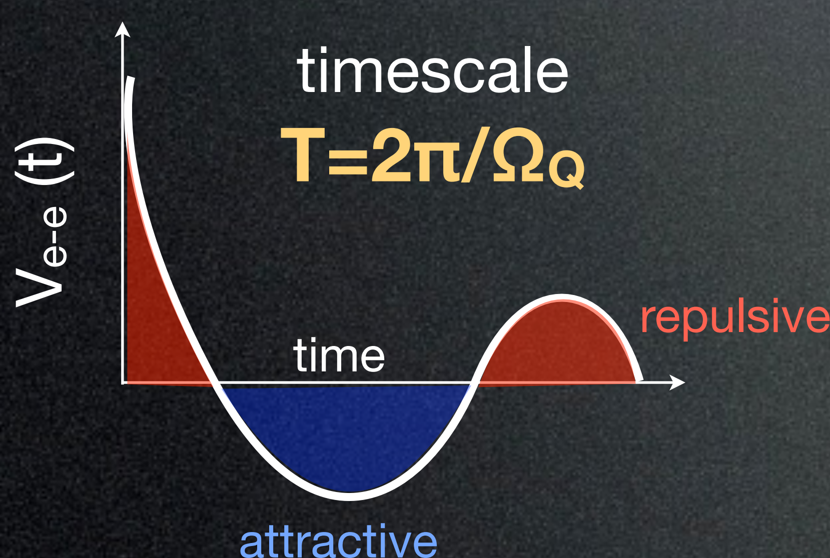


space

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

timescale

**$T = 2\pi/\Omega_Q$**



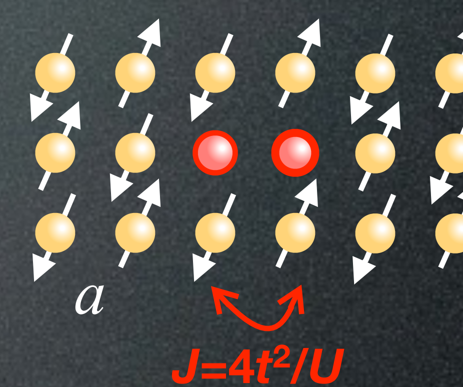

## Hubbard model

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

space

*d*-wave: other glue?

$\lambda = a$ ($\cong$4 Å)




timescales

**$\hbar/t_{ij}$ ($\cong$2 fs)**

vs

**$\hbar/J$ ($\cong$6 fs)**

vs

$\hbar/U$ ($\cong$0.3 fs)

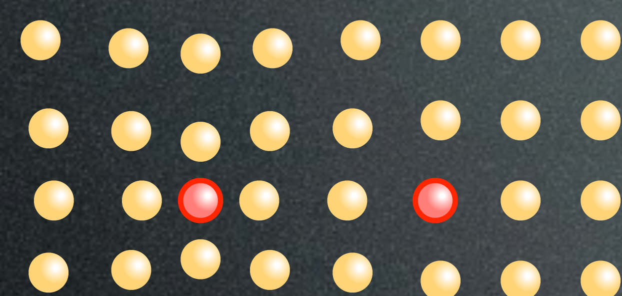
# retarded vs instantaneous interaction

## BCS

$$\hat{H} = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} \hat{c}^{\dagger}_{\mathbf{k}} \hat{c}_{\mathbf{k}} + \sum_{\mathbf{k},\mathbf{Q}} M_Q \hat{c}^{\dagger}_{\mathbf{k}+\mathbf{Q}} \hat{c}_{\mathbf{k}} (\hat{b}_{\mathbf{Q}} + \hat{b}^{\dagger}_{-\mathbf{Q}})$$

boson

glue: lattice distortion

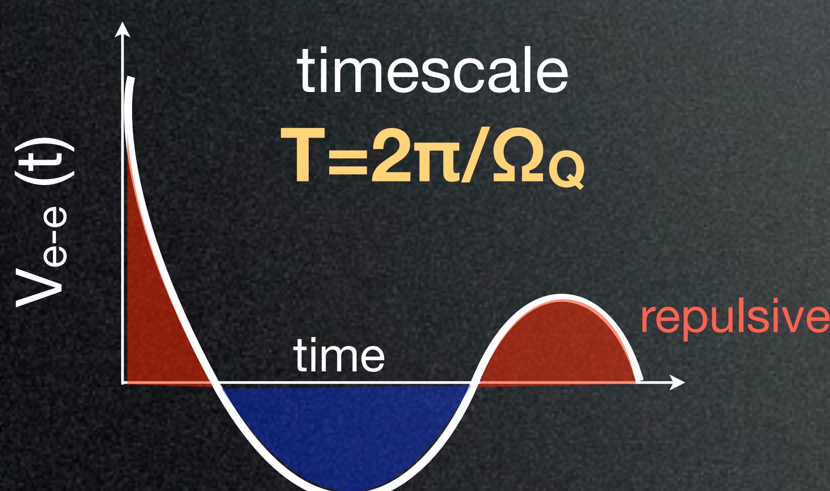


space

λ=2π/|Q|

timescale

**T=2π/$\Omega_Q$**

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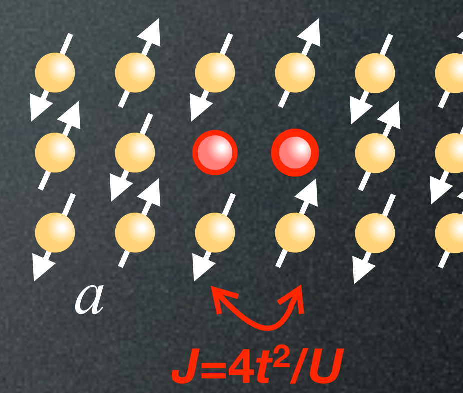
space

*d*-wave: other glue?

λ=$a$ (≅4 Å)




timescales

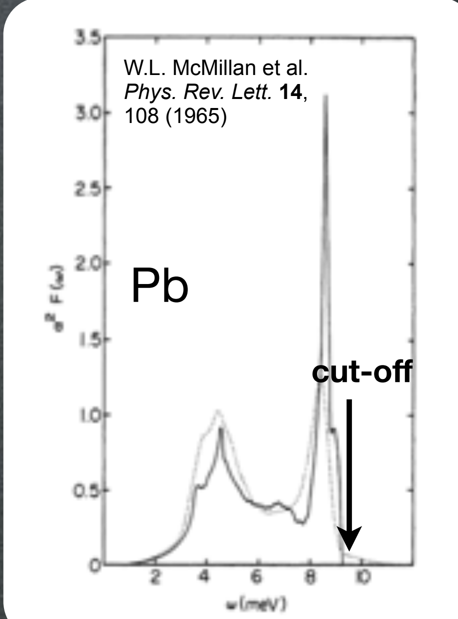
**ħ/$t_{ij}$ (≅2 fs)**

vs

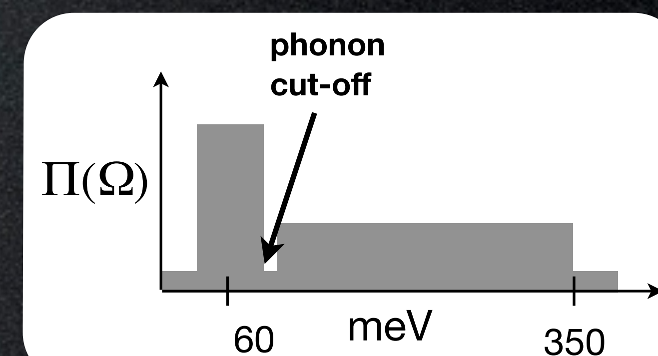
**ħ/J (≅6 fs)**

vs

ħ/U (≅0.3 fs)




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



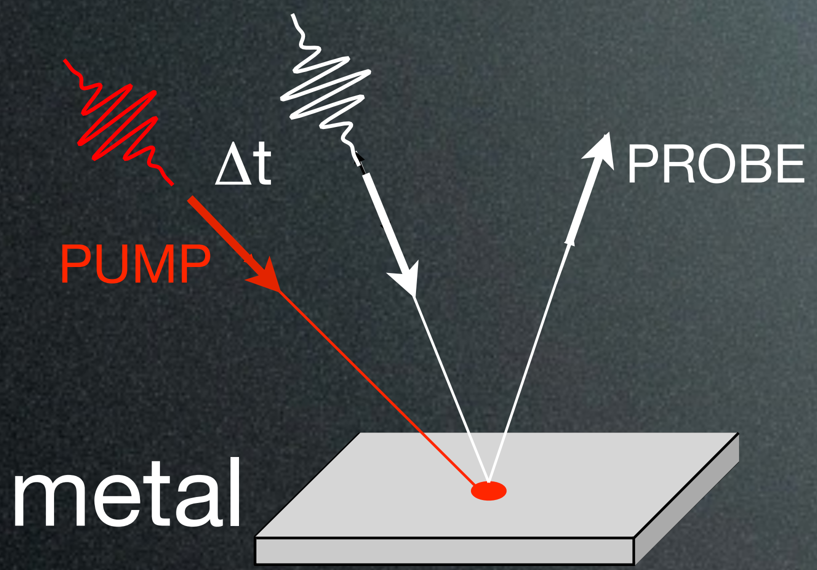

J. Carbotte et al. *Rep. Prog. Phys.* **74**, 066501 (2011)

*C. Giannetti*

**Single-colour** pump-probe experiments and electron-phonon coupling

# non-equilibrium reflectivity and e-ph coupling

pump probe on metals




2-temperature model

PUMP

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

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

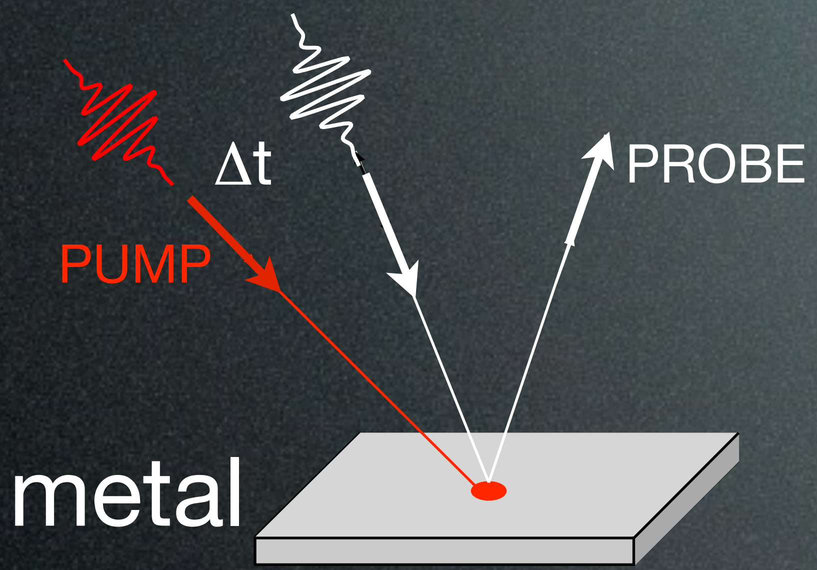
$G/C$

determines the dynamics in the time domain

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

# non-equilibrium reflectivity and e-ph coupling

pump probe on metals




2-temperature model

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

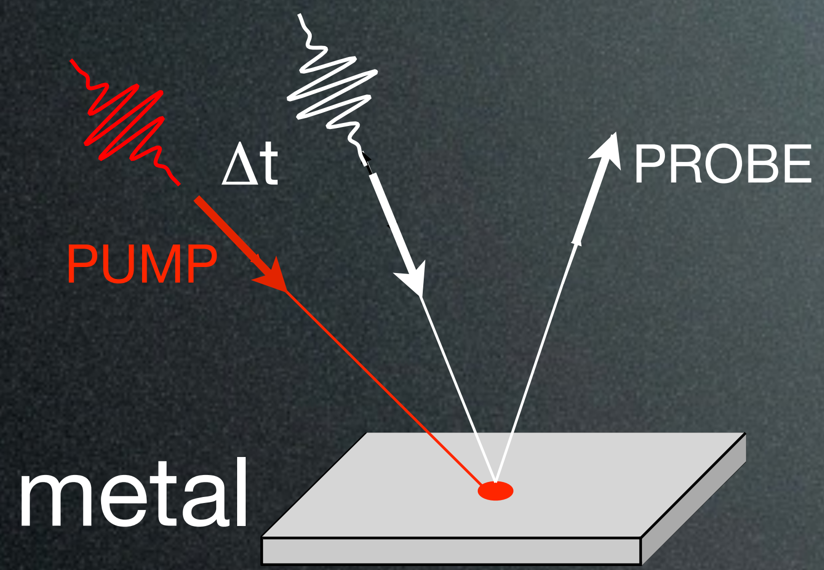
$$\frac{\partial T_{lat}}{\partial t} = -\frac{G(\Pi_{lat}, T_{lat}, T_e)}{C_{lat}}$$

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

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

# non-equilibrium reflectivity and e-ph coupling

pump probe on metals




2-temperature model

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

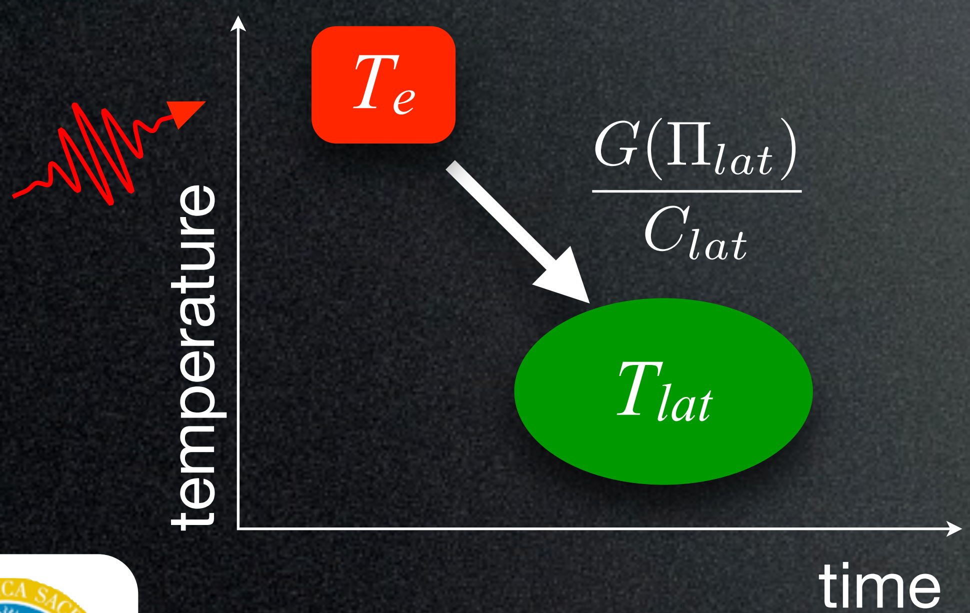
PUMP

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

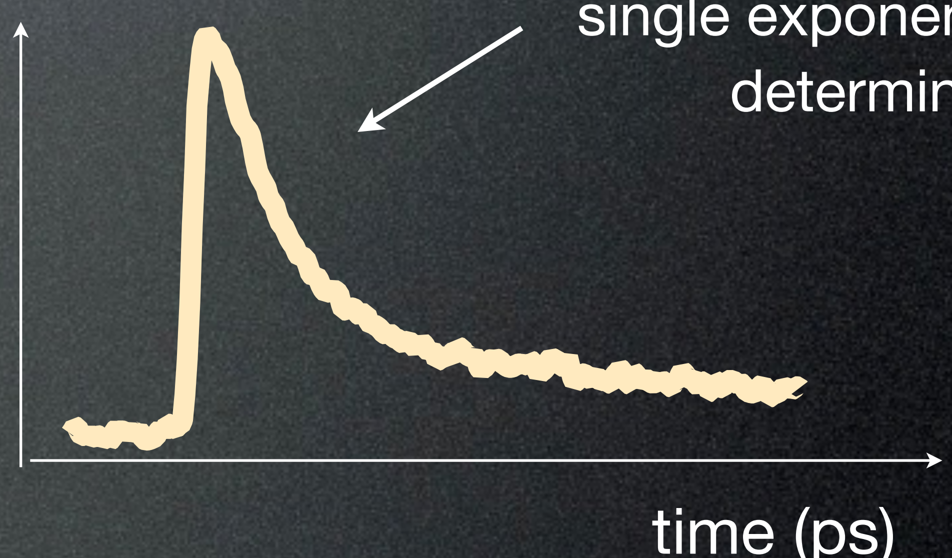
$G/C$

determines the dynamics in the time domain

hierarchy in the dynamics



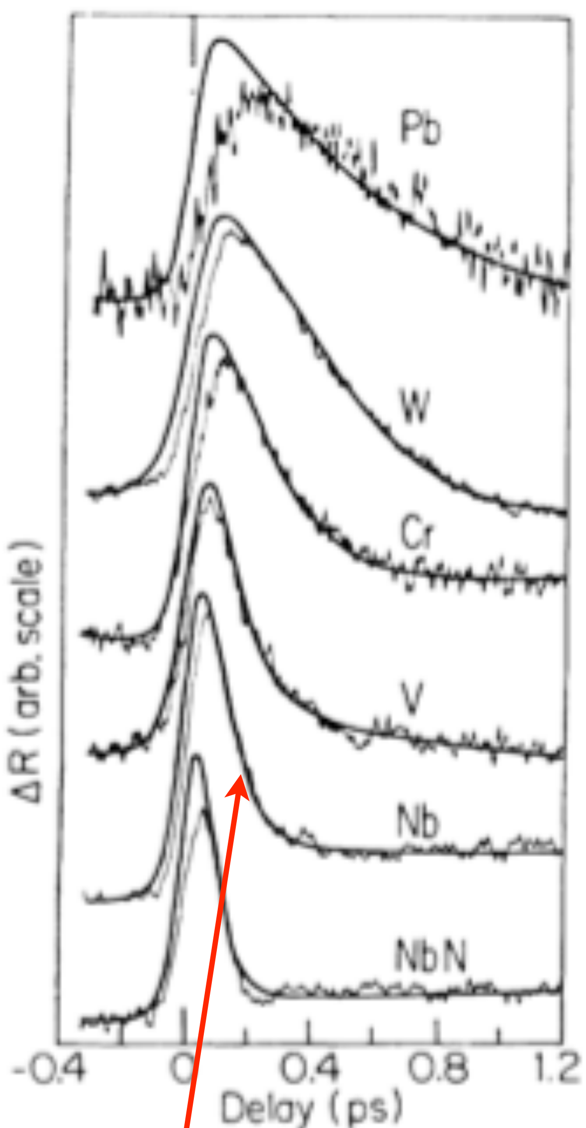

$\delta R/R \propto \delta T_e/T_e$




in conventional metals: single exponential decay determines λ

# non-equilibrium spectroscopy on metals

## electron-phonon coupling in metals




single exponential decay

| | $T_c(0)$ (K)[a] | $\lambda_{exp}\langle\omega^2\rangle$ (meV$^2$) | $\langle\omega^2\rangle$ (meV$^2$) | $\lambda_{exp}$ | $\lambda_{th}$ |
|---|---|---|---|---|---|
| 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] |
| $V_3Ga$ | 1110 | $370 \pm 60$ | 448[k] | $0.83 \pm 0.13$ | 1.12[k] |

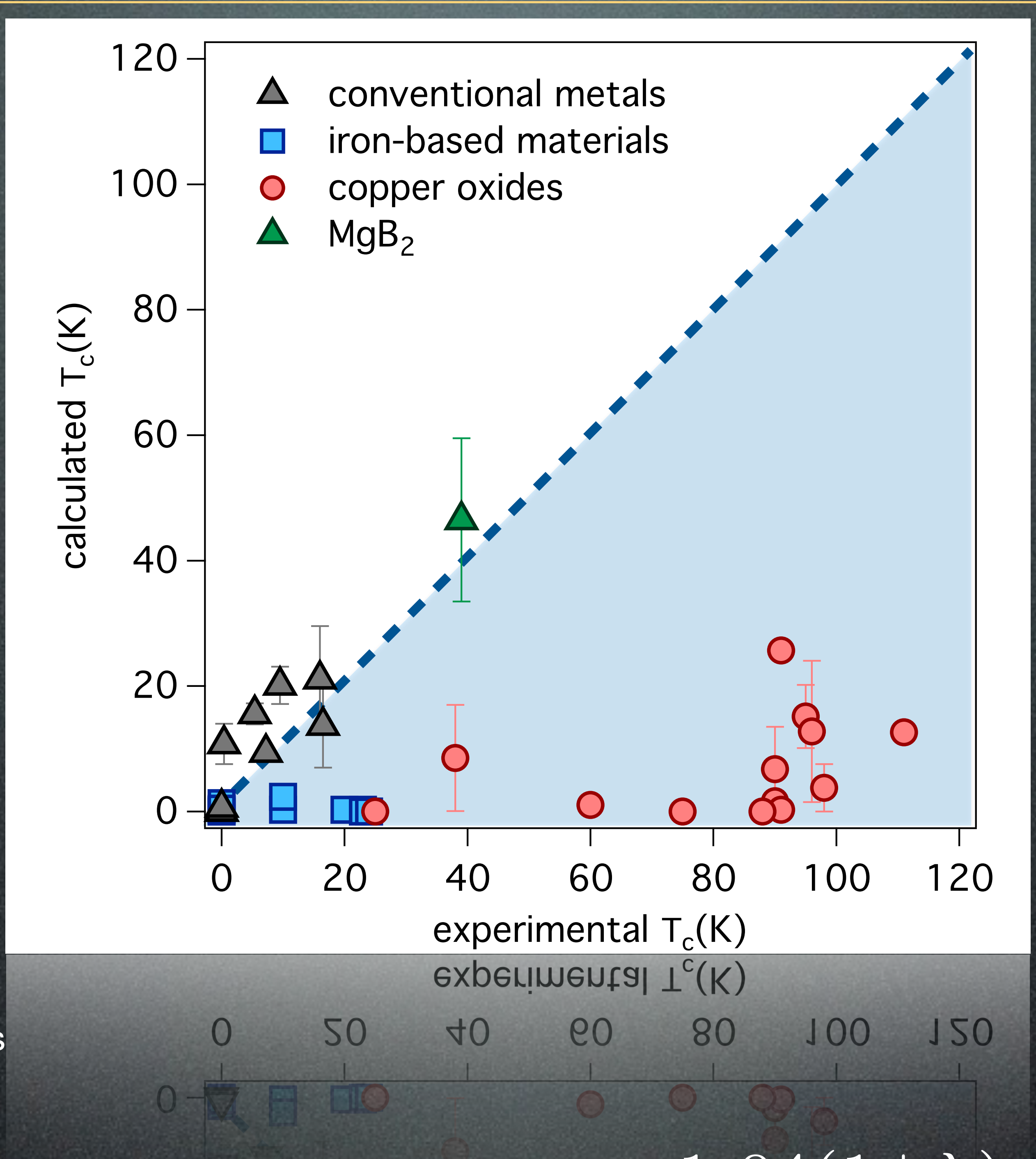
S.D. Brorson et al. *Phys. Rev. Lett.* **64,** 2172 (1990)

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

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

# electron-phonon coupling in superconductors

electron-phonon coupling λ obtained from time-resolved techniques




CUPRATES:
the glue is not in the electron-phonon coupling!!

μ*: effective Coulomb repulsion
g=1 for s-wave BCS superconductors
g<1 for d-wave superconductors

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

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

P.B. Allen and R.C. Dynes, *Phys. Rev B* **12** 905 (1975)
A. J. Millis, S. Sachdev, C. M. Varma, Phys. Rev. B **37**, 4975 (1988).

*C. Giannetti*

Beyond single-colour experiments to access the ultrafast dynamics

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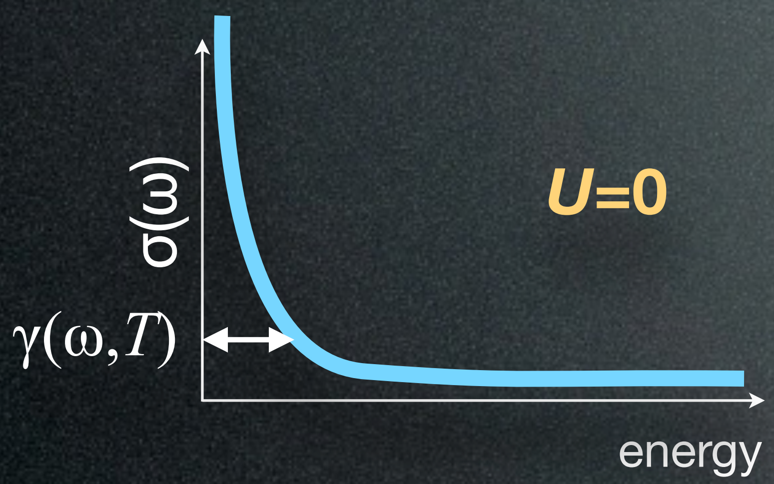
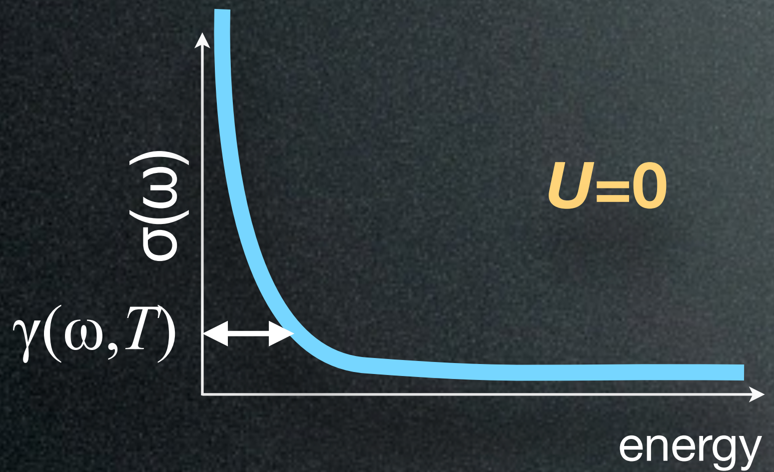
# Extended Drude Model

equilibrium optical conductivity



# Extended Drude Model

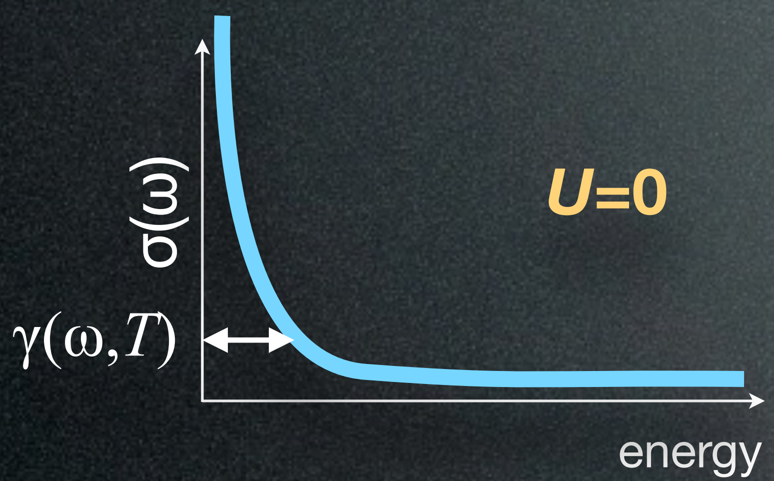
equilibrium optical conductivity




Drude dielectric function $\epsilon_D(\omega) = 1 - \dfrac{{\omega_p}^2}{\omega(\omega + i\tau^{-1})}$ τ constant

# Extended Drude Model

## equilibrium optical conductivity




Drude dielectric function

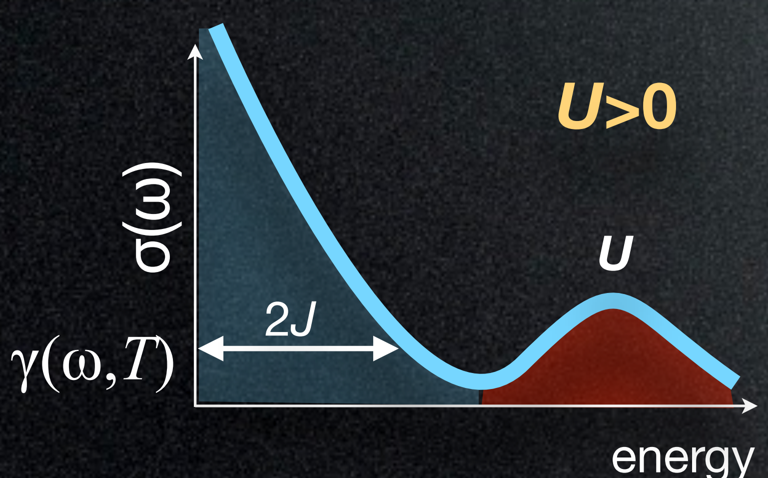
$$\epsilon_D(\omega) = 1 - \frac{{\omega_p}^2}{\omega(\omega + i\tau^{-1})}$$

τ constant

Extended Drude dielectric function

$$\epsilon_D(\omega, T) = 1 - \frac{{\omega_p}^2}{\omega(\omega + M(\omega, T))}$$

Memory function

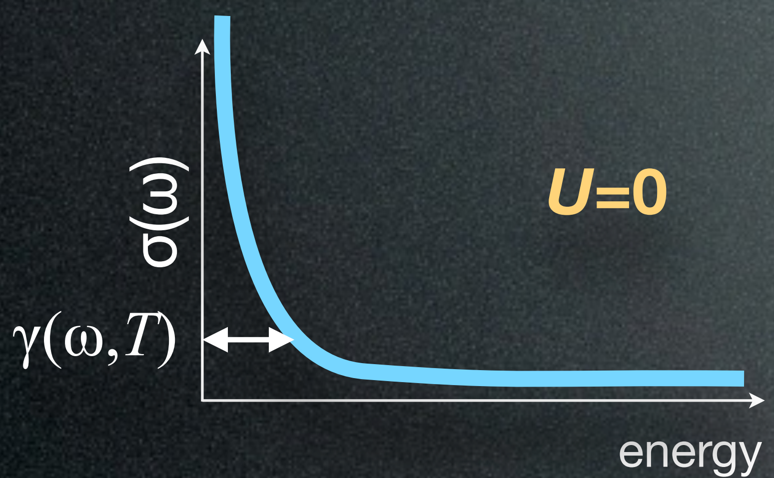



Fermi-Dirac distributions

$$\frac{M(\omega)}{\omega} = \left\{ \int \frac{f(\xi) - f(\xi + \omega)}{\omega + \Sigma^*(\xi) - \Sigma(\xi + \omega)} d\xi \right\}^{-1} - 1$$

# Extended Drude Model

## equilibrium optical conductivity

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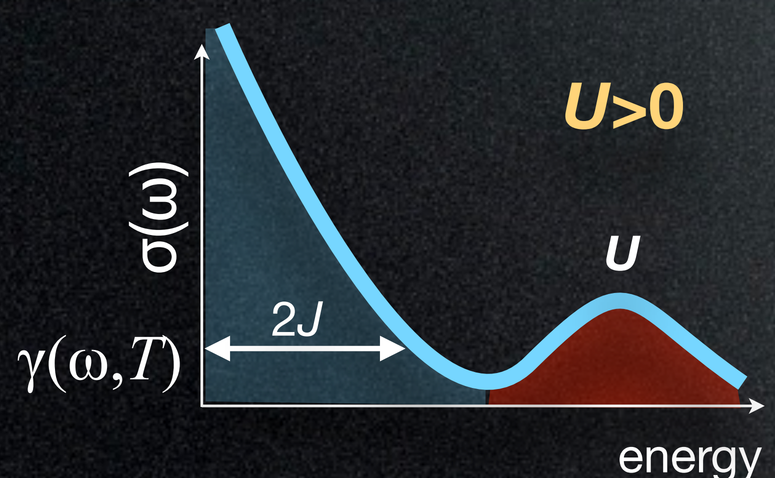



single particle self-energy $\Sigma(\xi) = \int_0^\infty d\Omega L(\xi, \Omega; T)\Pi(\Omega)$

T-dep. kernel function $L(\xi,\Omega;T_e) + L(\xi,\Omega;T_b)$

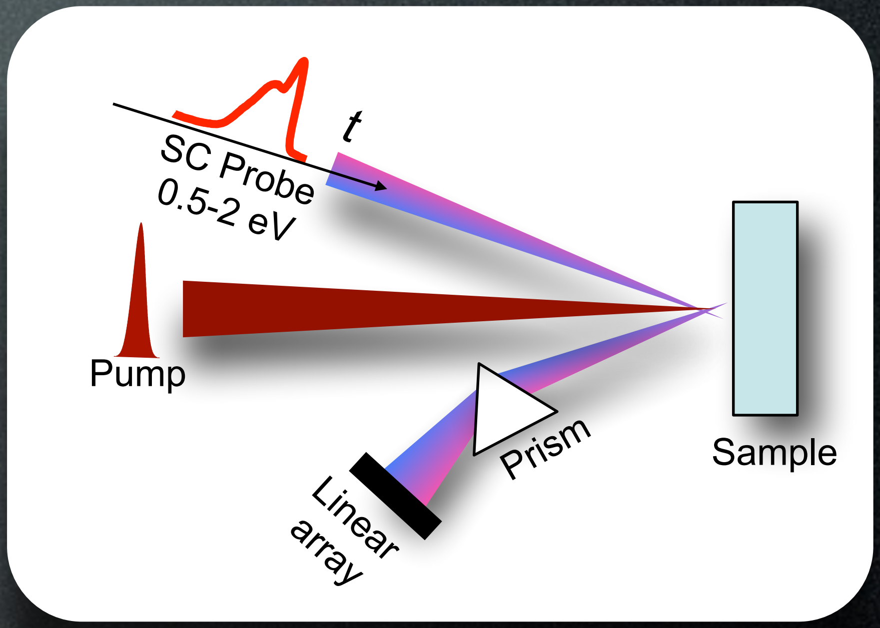
bosonic function $\alpha^2 F(\Omega)$ in conventional metals

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

# The “glue” problem in time-resolved optics

A) Is it really possible to describe the electron dynamics of doped cuprates in terms of charge carriers interacting with bosonic fluctuations? (energy resolution)

B) On which timescale the charge carriers can exchange energy with bosons? (temporal resolution)




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

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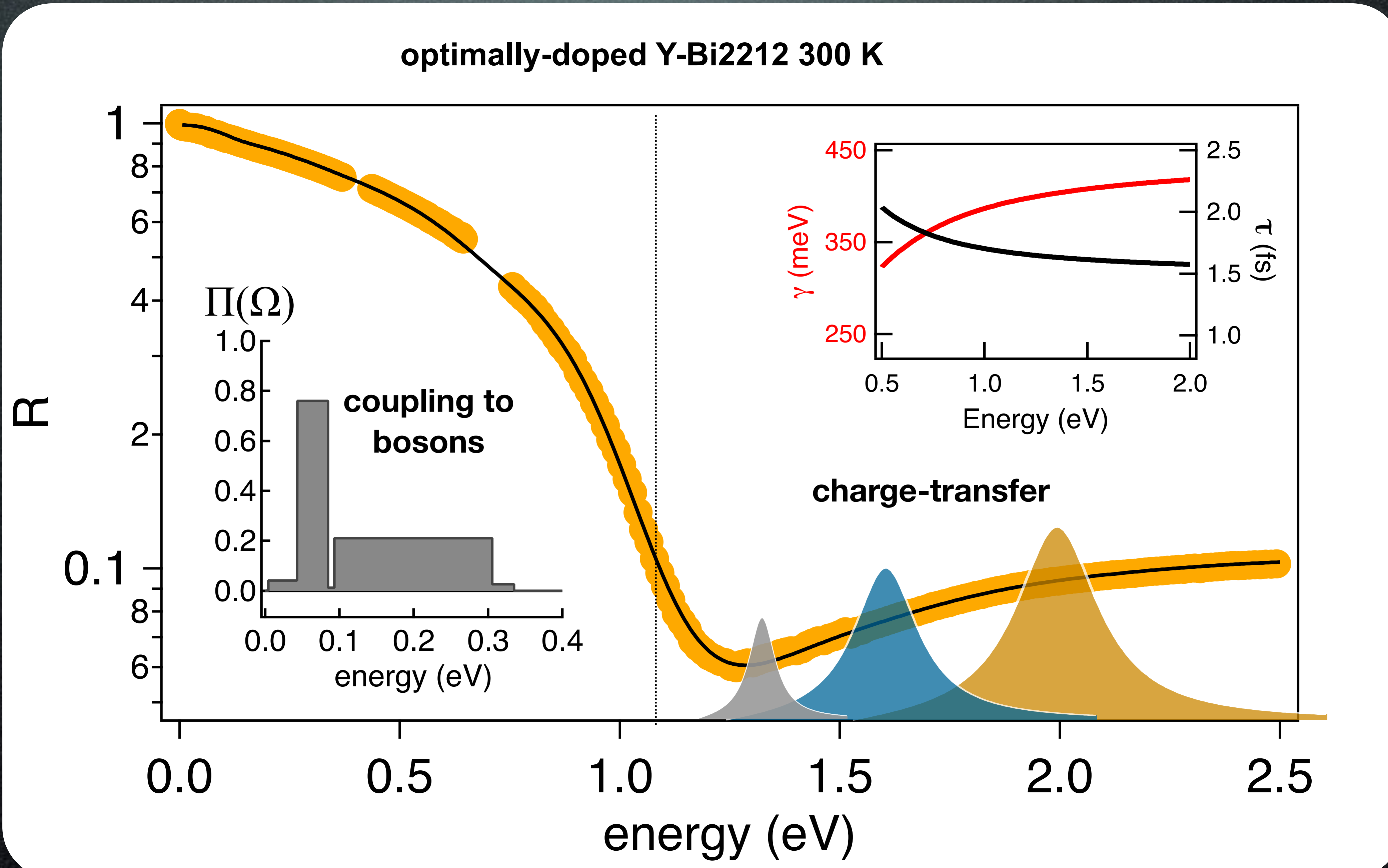
# Equilibrium optical properties of cuprates

scattering rate $ImM(\omega, T) = \hbar/\tau(\omega, T)$

D.N. Basov et al.
*Rev. Mod. Phys.* **85**, 471 (2011)

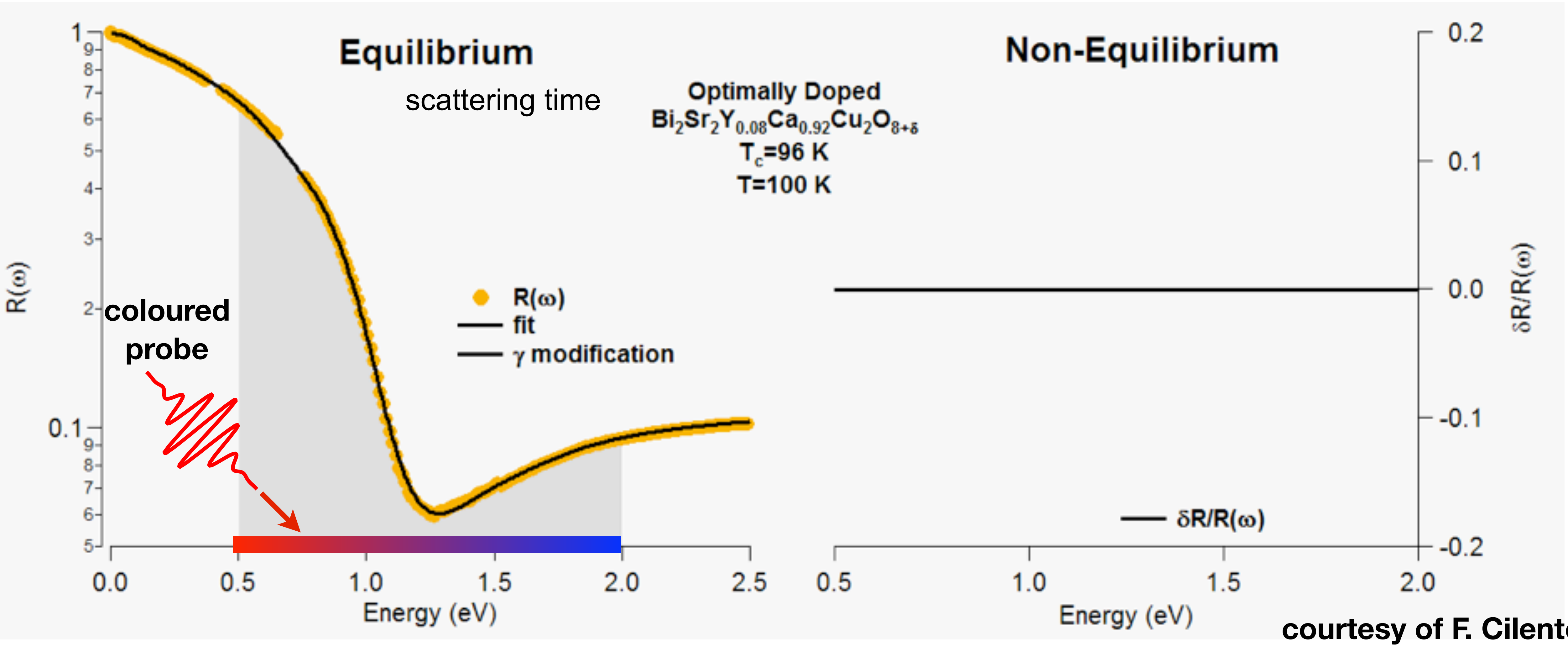
$R(\omega)$
↓
$M(\omega)$
↓
$\Sigma(\xi)$
↓
$\Pi(\Omega)$




E. van Heumen et al., *Phys. Rev. B* **79**, 184512 (2009)

# NON-equilibrium optical properties of cuprates

effective e-bos scenario: change of scattering rate $\gamma(\omega,T)$




courtesy of F. Cilento

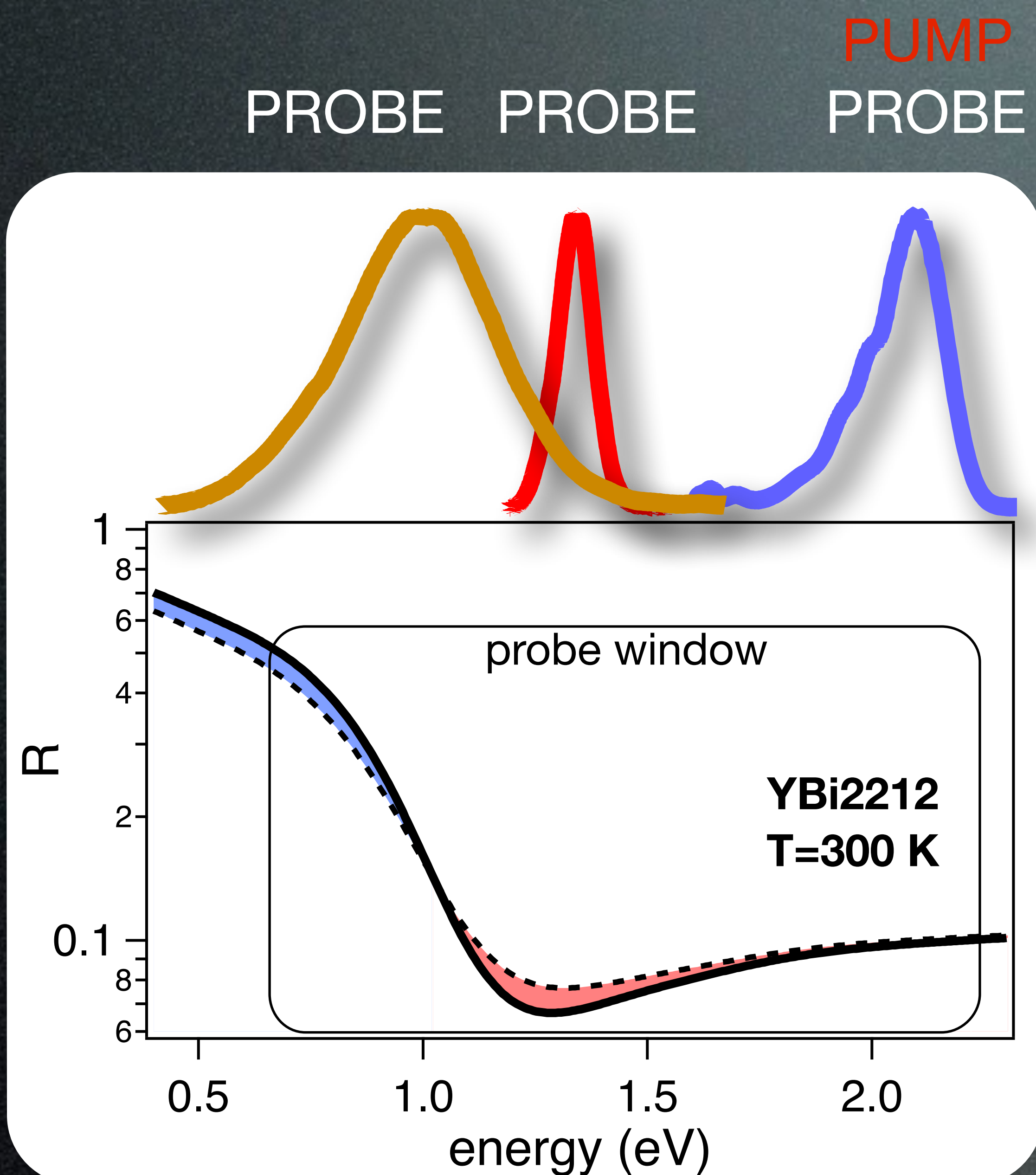
isosbestic point at 1.1 eV

$$\delta R(\omega,\gamma) = \frac{\partial R}{\partial \gamma}(\omega)\delta\gamma$$

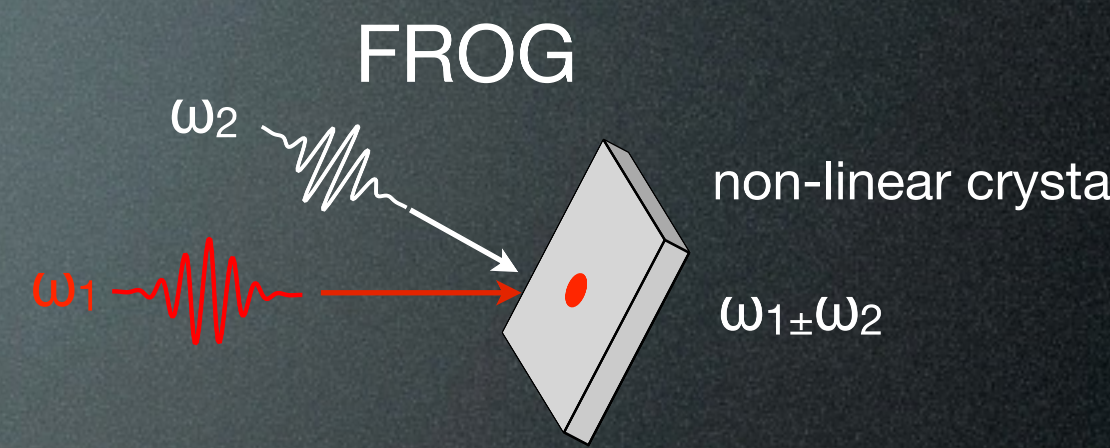
$$\frac{\partial R}{\partial \gamma}(1.1) = 0$$

M. Greger et al. *Phys. Rev. B* **87,** 195140 (2013)

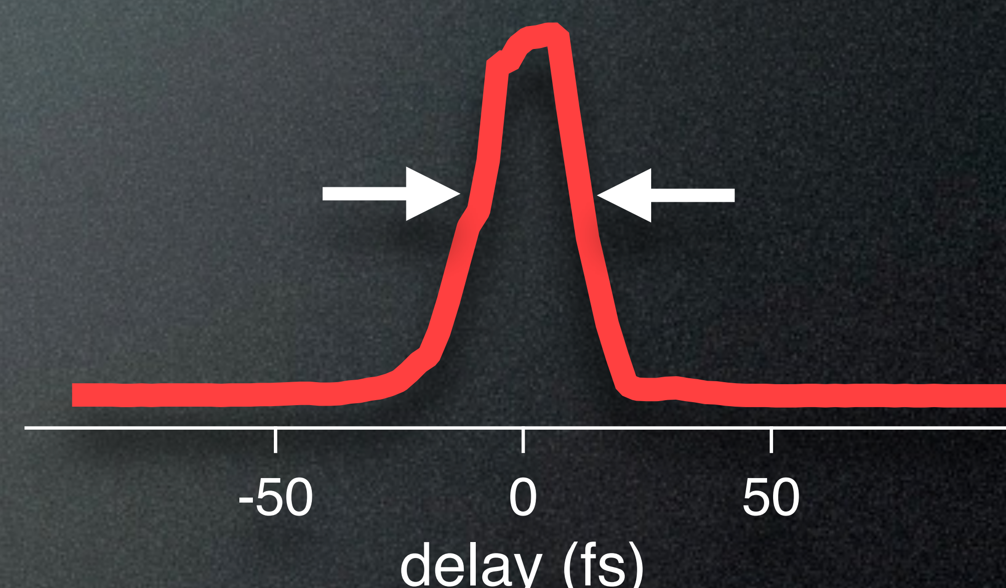
# ultra high temporal resolution




G. Cerullo's group (Politecnico of Milan)

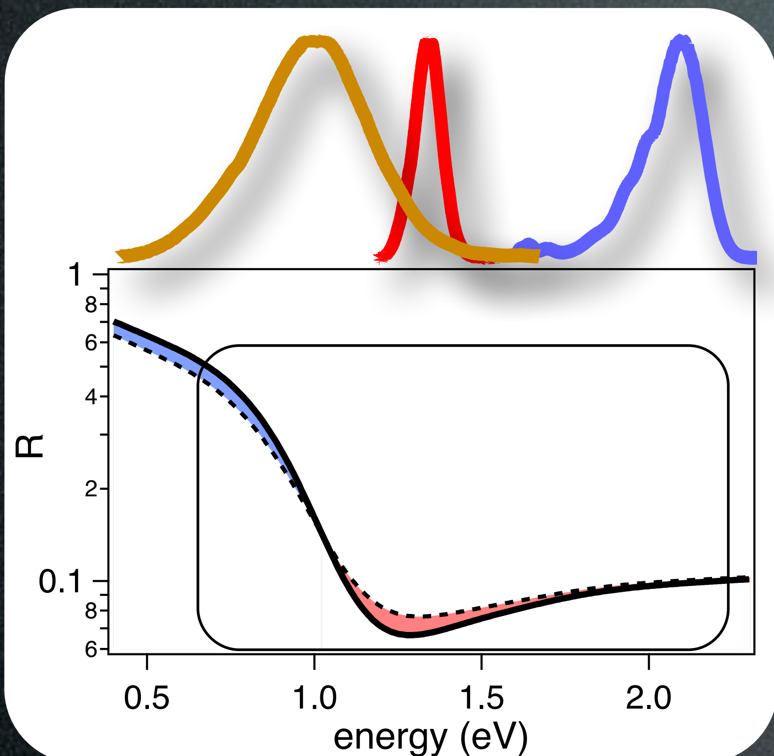


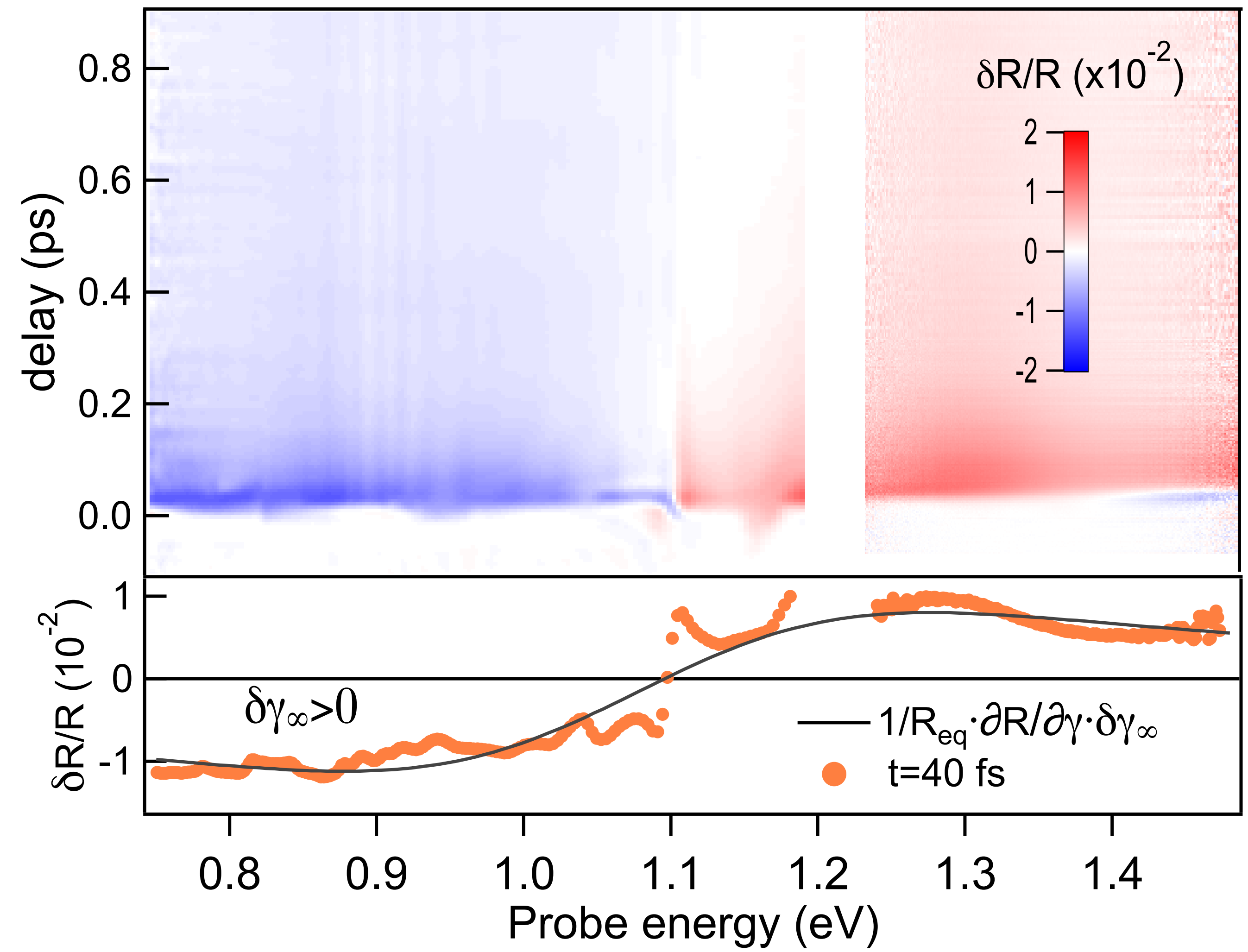

temporal resolution
fwhm: **18±2 fs**

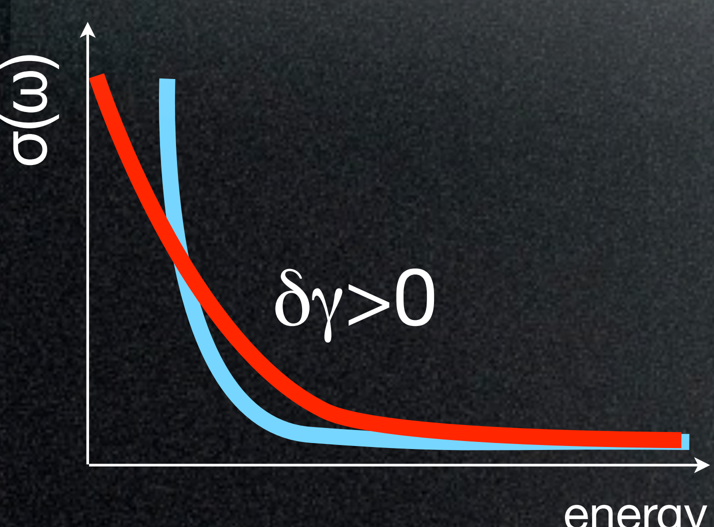



optimally doped $Bi_2Sr_2Y_{0.08}Ca_{0.92}Cu_2O_{8+\delta}$ (YBi2212)
$T_c$=96 K

# results on YBi2212 $T$=300 K



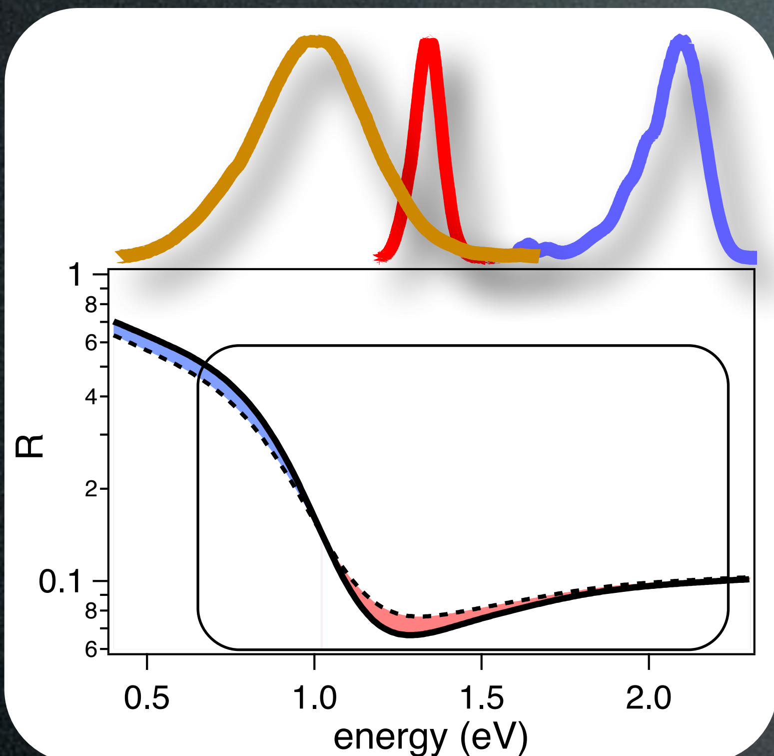



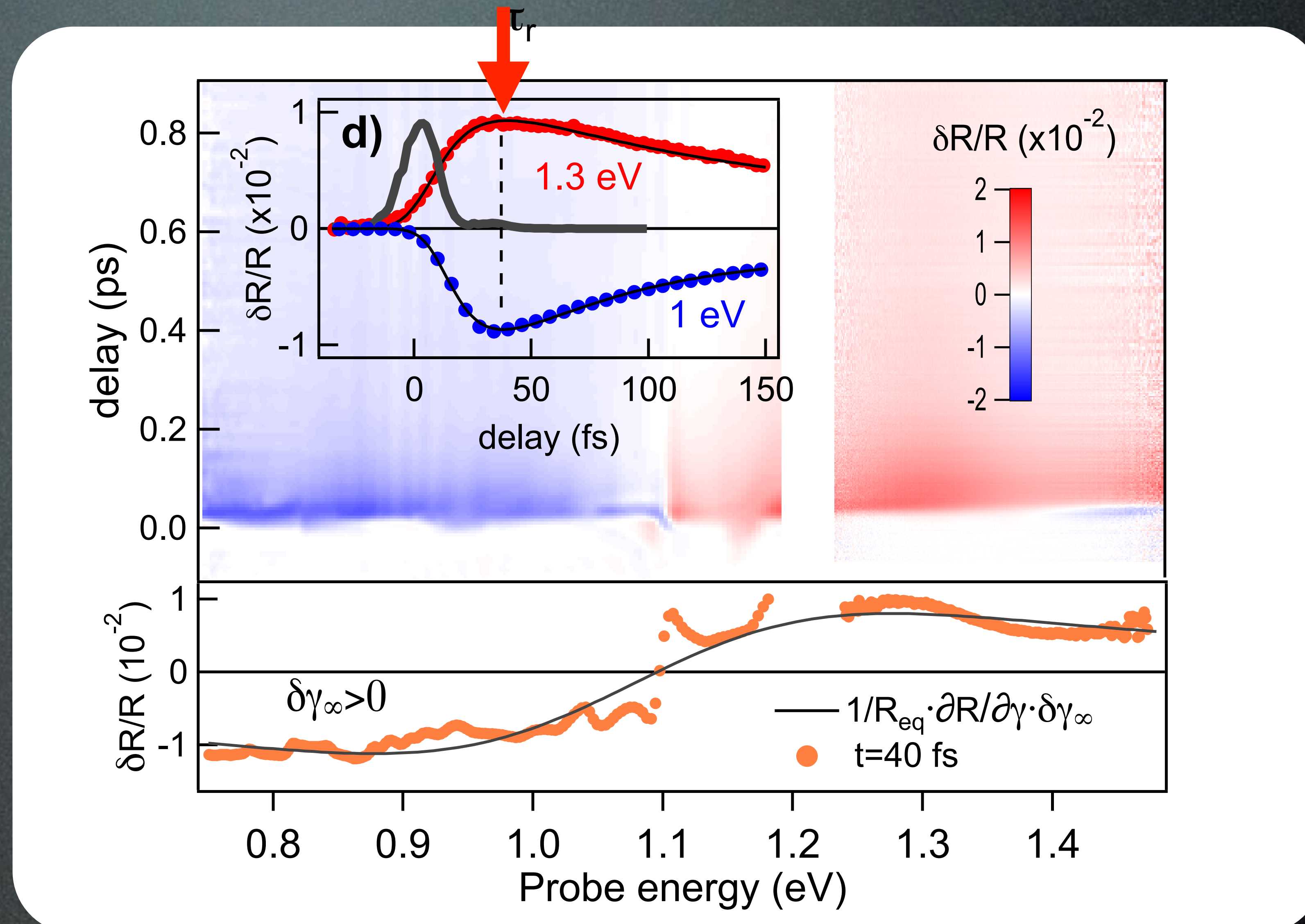



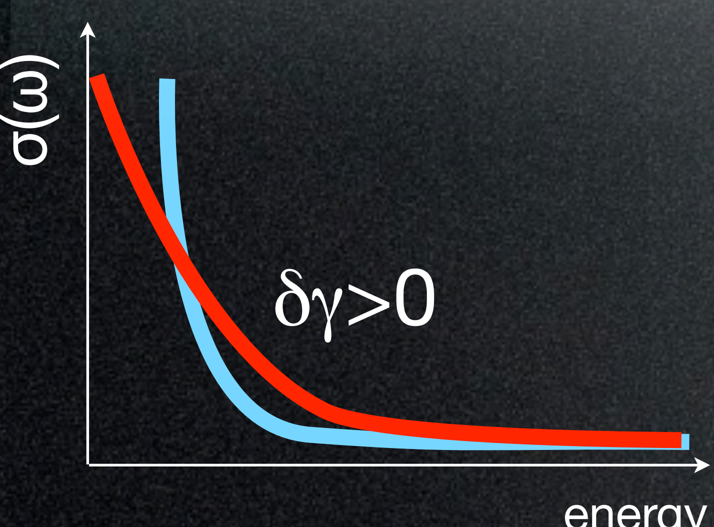

no variation at the energy scale $U$! → $\delta R(\omega) \propto \delta T_b$

A) In 10-20 fs it is really possible to describe the dynamics of doped cuprates in terms of charge carriers interacting with effective bosonic fluctuations ✔

# results on YBi2212 $T$=300 K

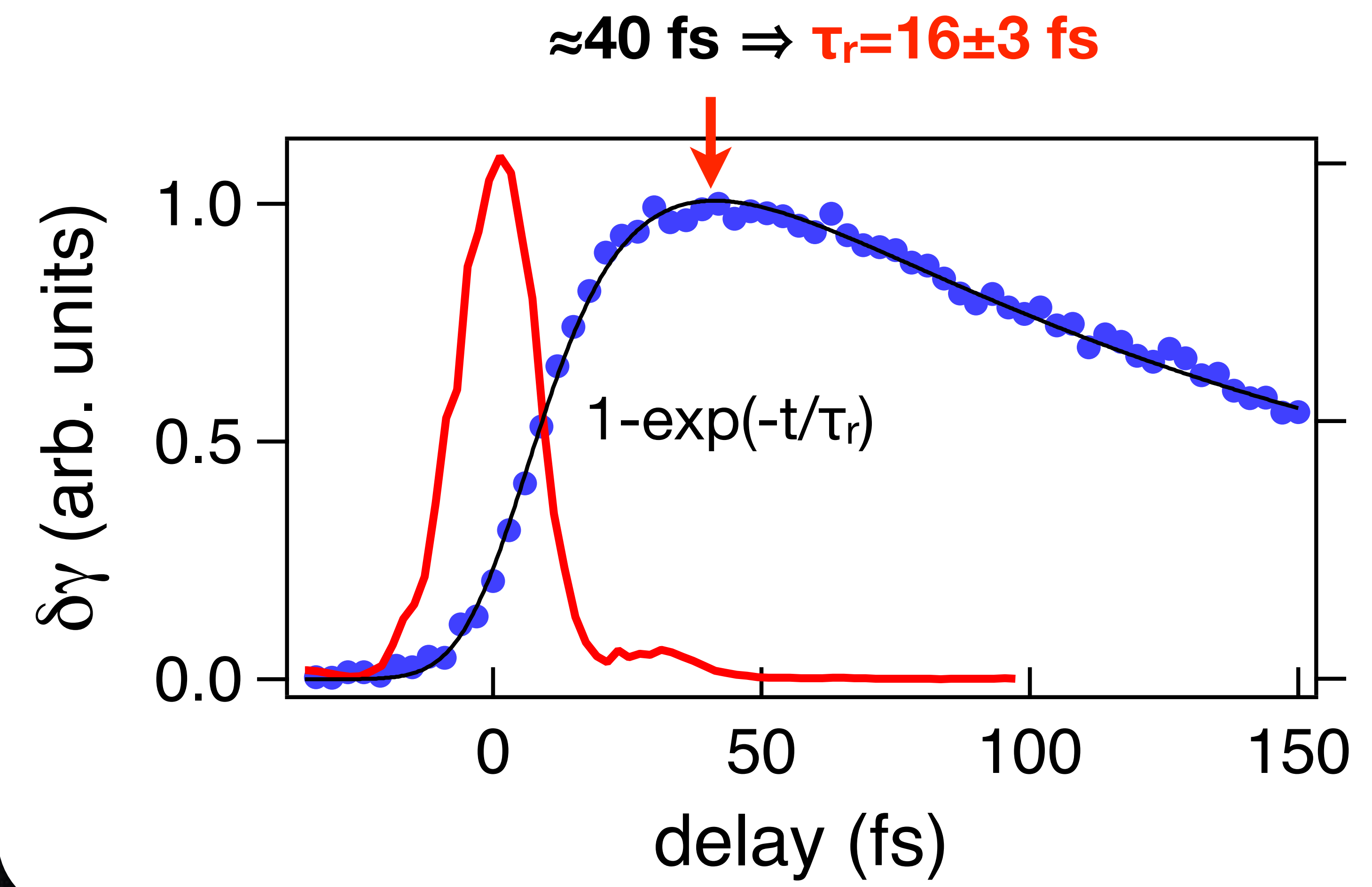


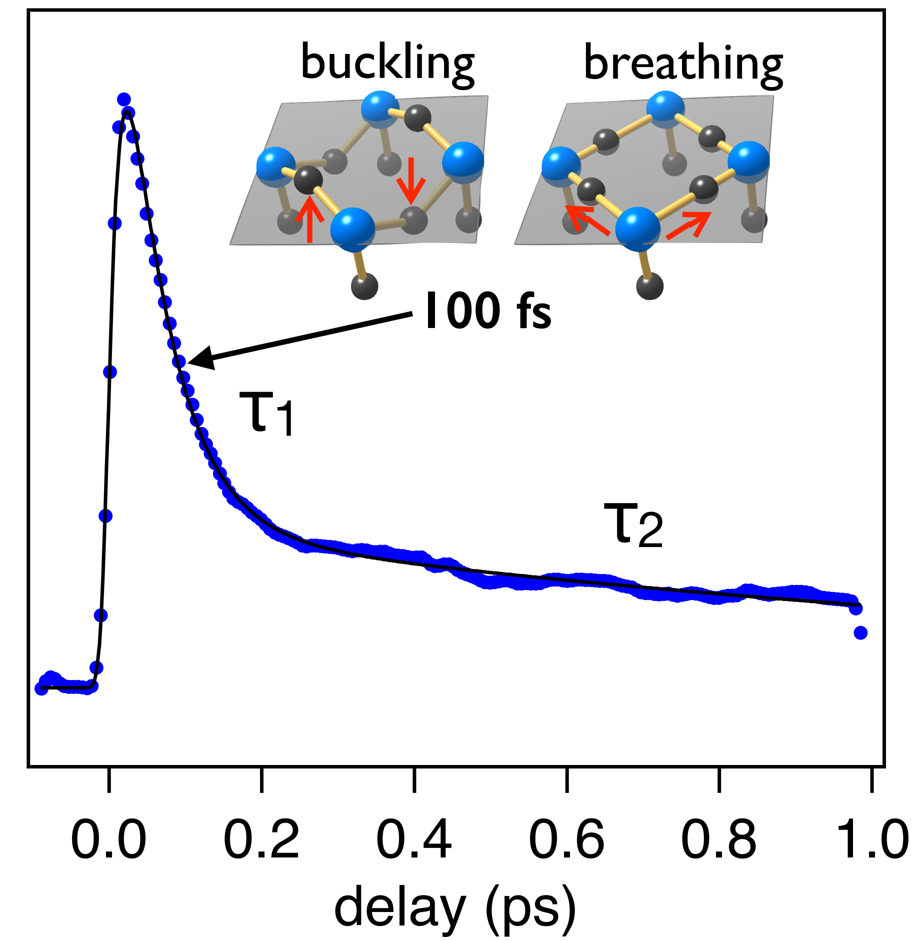




A) In 10-20 fs it is really possible to describe the dynamics of doped cuprates in terms of charge carriers interacting with effective bosonic fluctuations ✔

# retarded e-boson interaction

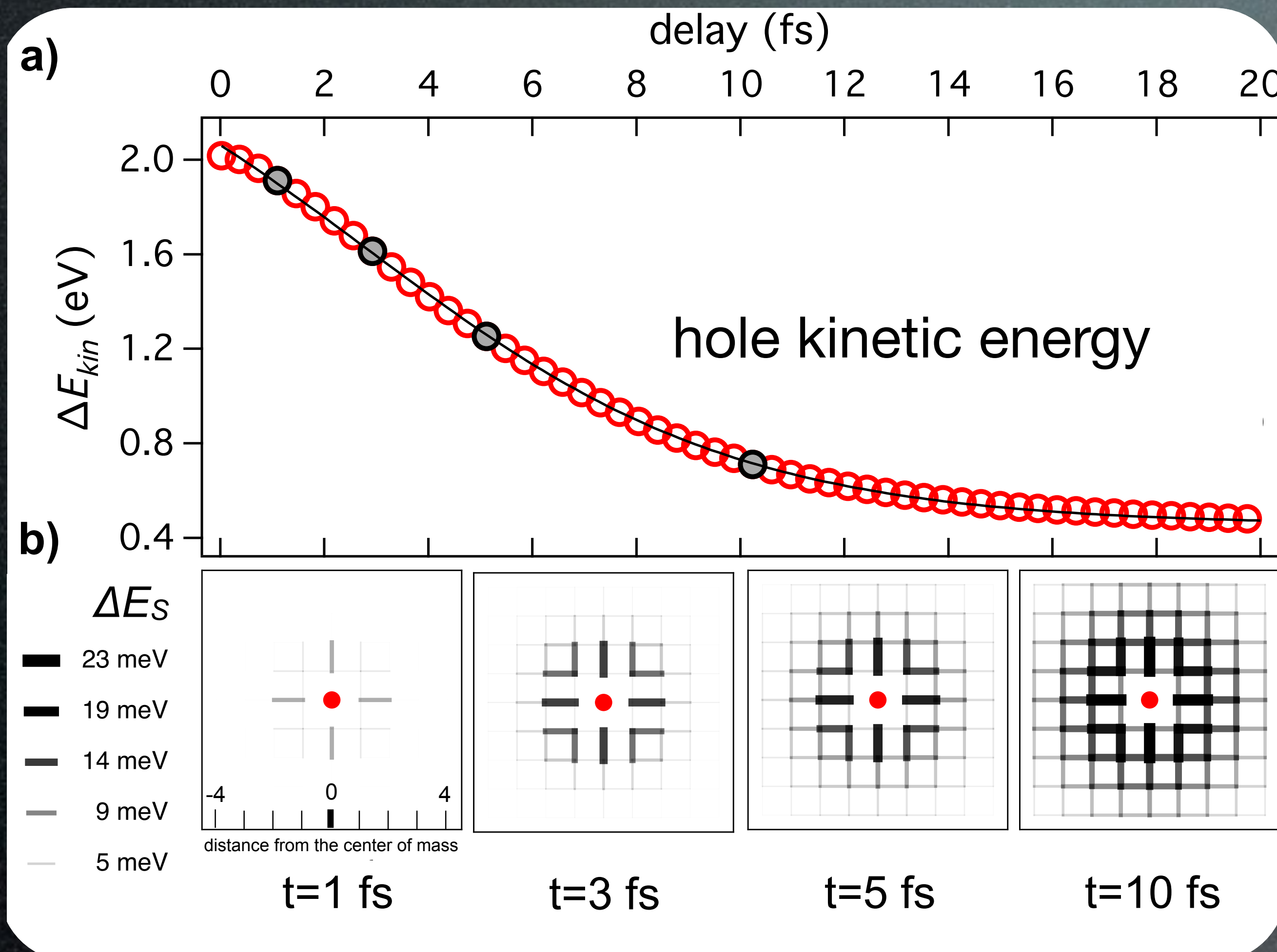



$\delta R(\omega) \propto \delta T_b$

B) In 16 fs photoexcited carriers can exchange energy with bosons ✔

# 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

D. Golez, M. Mierzejewski, J. Bonca, L. Vidmar

$$H = -t_h \sum_{\langle \mathbf{lj} \rangle, \sigma} (c^\dagger_{\mathbf{l},\sigma} \tilde{c}_{\mathbf{j},\sigma} + \mathrm{h.c.}) + J \sum_{\langle \mathbf{lj} \rangle} \mathbf{S_l} \cdot \mathbf{S_j}$$

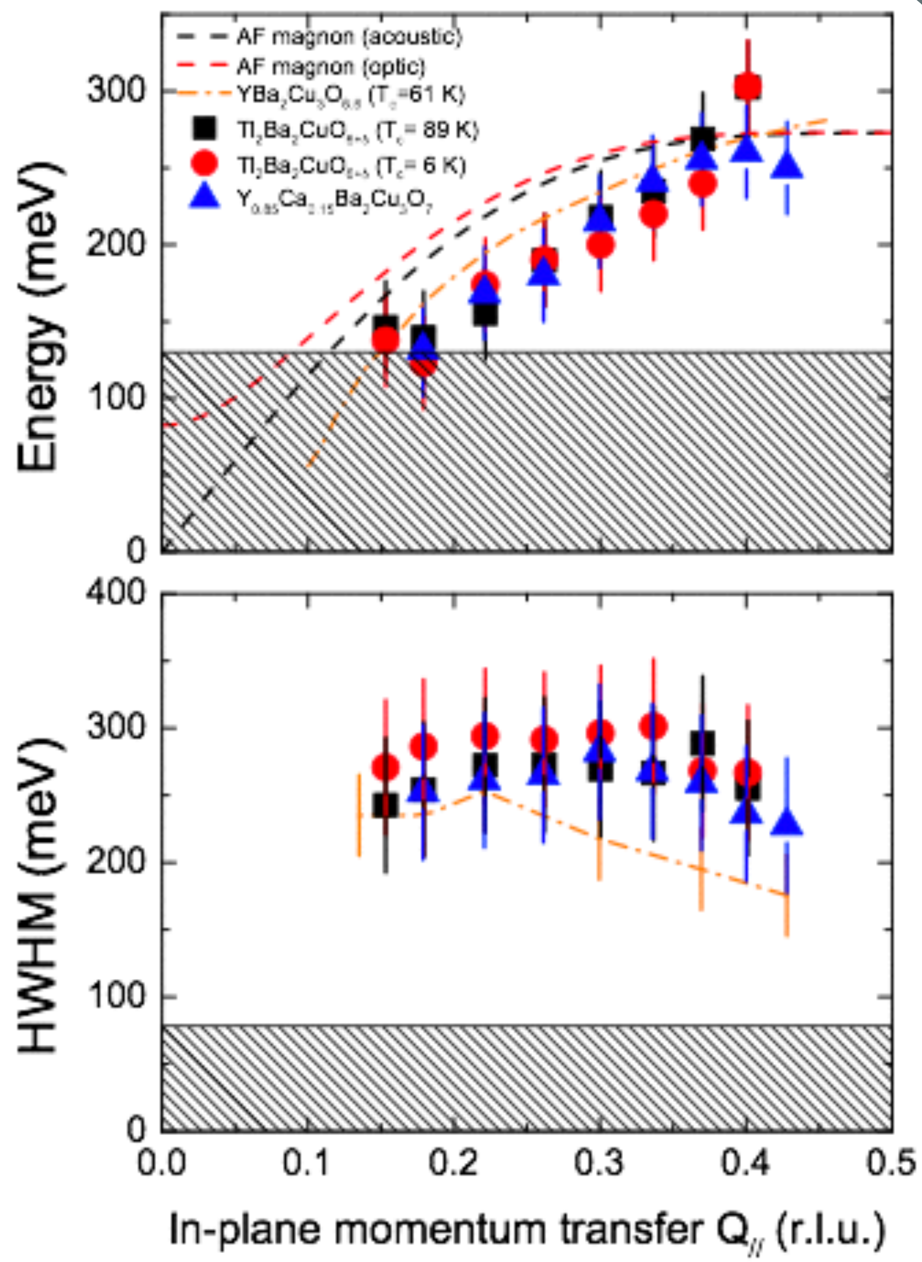
$t$=360 meV  $J$=120 meV

B) In 16 fs photoexcited carriers can exchange energy with bosons ✔

in agreement with non-eq. DMFT (M. Eckstein & P. Werner *arXiv:1410.3956*)

# short-range antiferromagnetic fluctuations

RIXS: Dispersion of paramagnons in cuprates at all dopings




M. Le Tacon et al, *Nature Physics* **7**, 725 (2011)
M. Dean et al, *Nature Materials* **12**, 1019 (2013)

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

- ~2 fs optical scattering rate
- 2-3 fs the inverse of paramagnon width (2-300 meV)

$$\tau = \frac{\hbar}{\gamma}$$

$\hbar$=658 meV fs

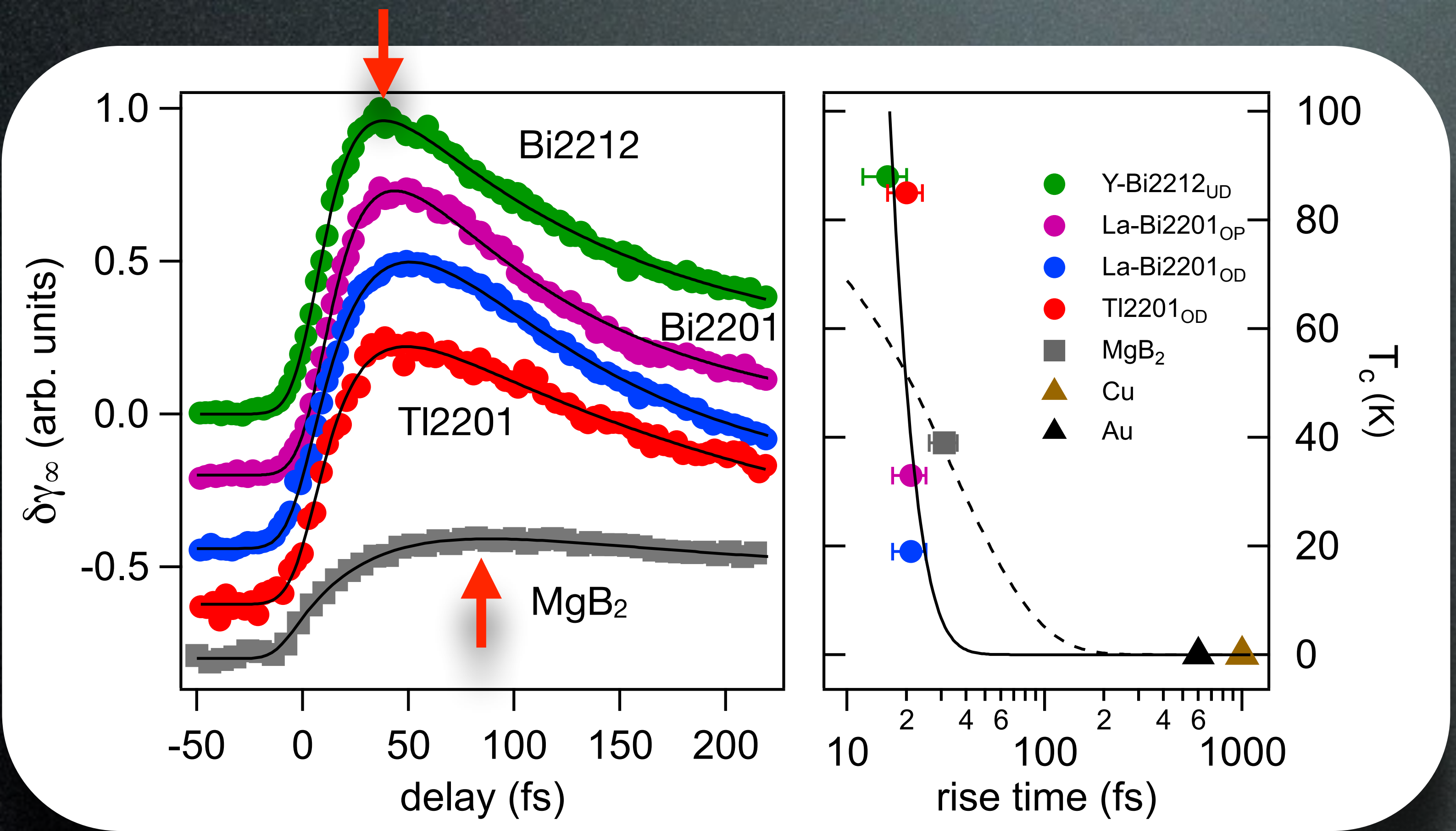
# comparing to conventional superconductors ($MgB_2$)

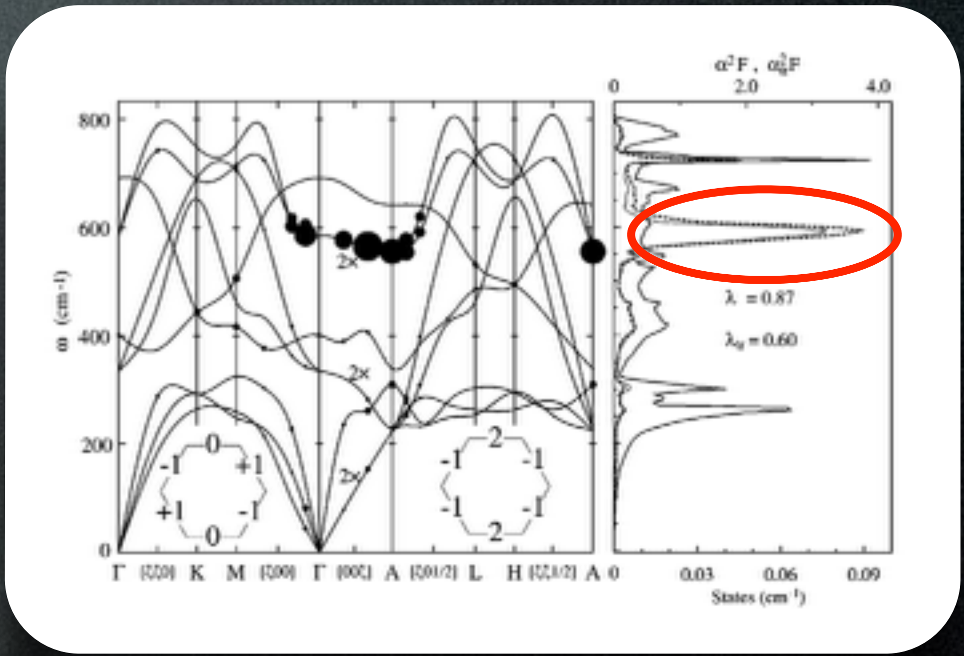
**$MgB_2$ :** conventional BCS system with $T_c$=40 K

**≈100 fs ⇒ $\tau_r$=32±3 fs**

coupling with optical mode at 70 meV

**$\tau_r \sim 2\pi/\Omega_Q$**



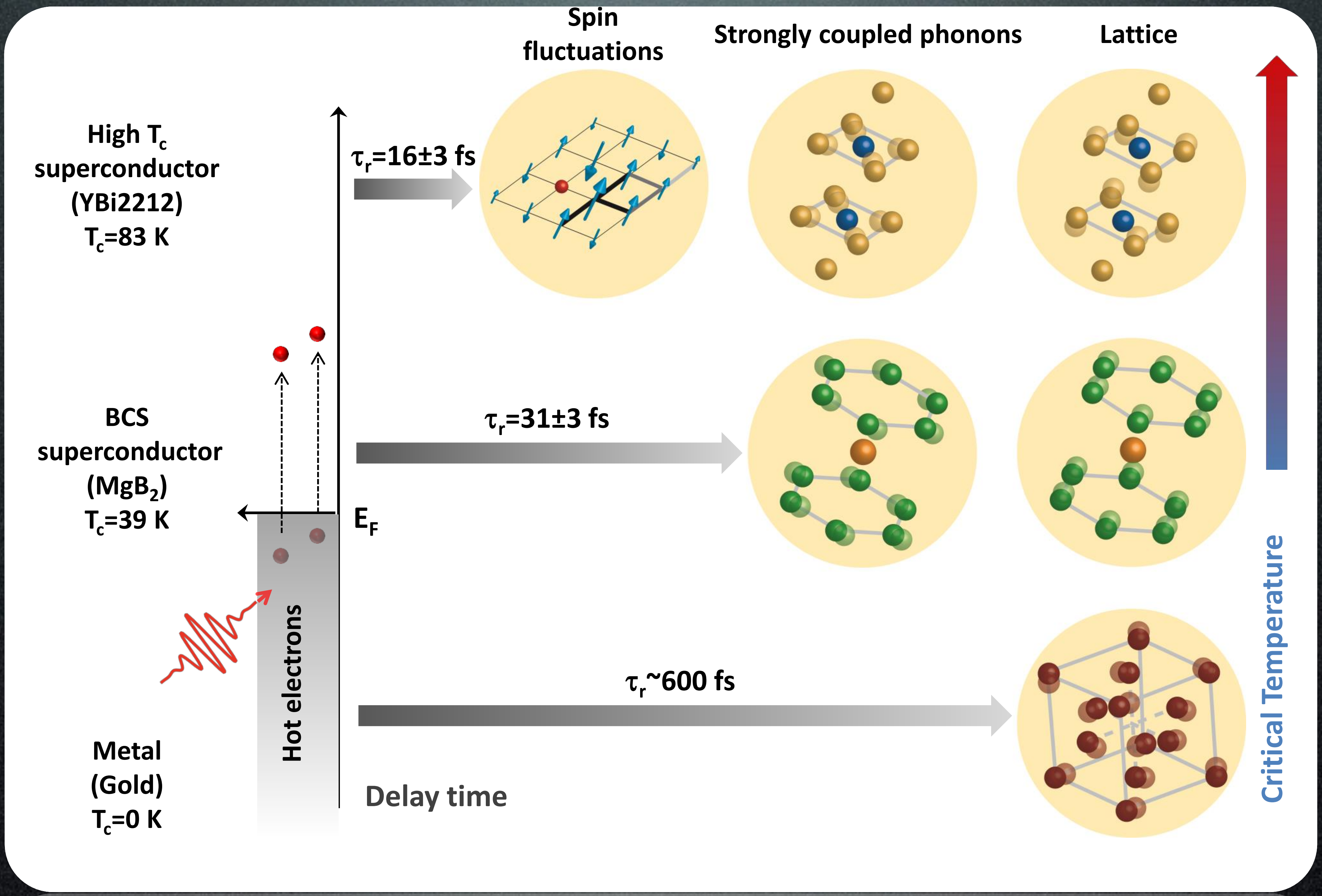



Y. Kong et al., *Phys. Rev. B* **64**, 020501(R) (2001)

P.B. Allen and R.C. Dynes, *Phys. Rev B* **12** 905 (1975)
A. J. Millis et al., Phys. Rev. B **37**, 4975 (1988).

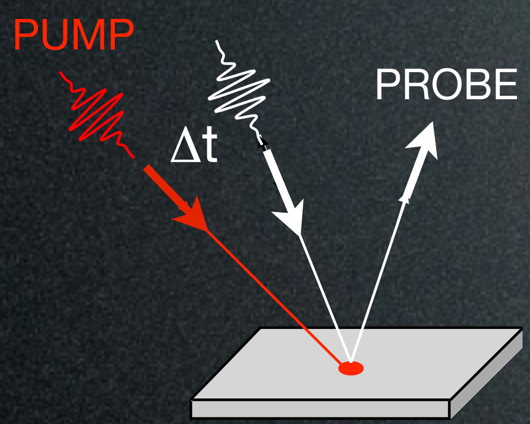
$$T_c = 0.83\tilde{\Omega}e^{-\frac{1.04(1+\lambda)}{g(\lambda-\mu^*(1+0.62\lambda))}}$$

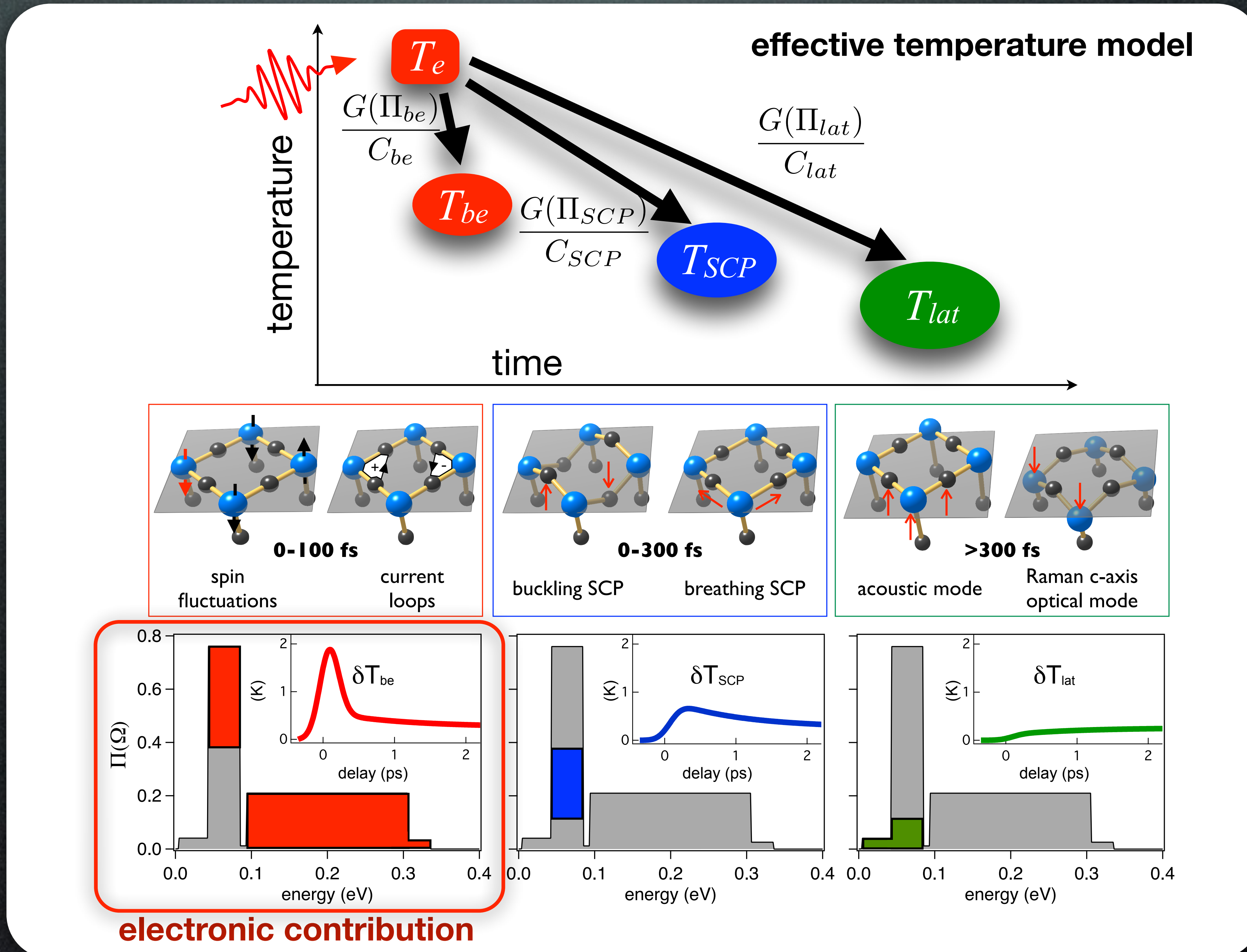
# Summary



Spin fluctuations
Strongly coupled phonons
Lattice
High $T_c$ superconductor (YBi2212) $T_c$=83 K
$\tau_r$=16±3 fs
BCS superconductor ($MgB_2$) $T_c$=39 K
$\tau_r$=31±3 fs
$E_F$
Hot electrons
Metal (Gold) $T_c$=0 K
$\tau_r$~600 fs
Delay time
Critical Temperature

# electron-boson coupling in CUPRATES




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

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

E. van Heumen et al., *Phys. Rev. B* **79**, 184512 (2009)

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

in agreement with the glue extracted from the Hubbard model

E. Gull & A.J. Millis *arXiv:1407.0704* (2014) → see talk: E. Gull

# Summary

- Non-equilibrium spectroscopy for disentangling different degrees of freedom
- Snapshots of the electron-boson coupling in correlated materials
- Coupling of the charge-carriers with AF fluctuations on the 15 fs timescale
- Effective glue in optimally and overdoped cuprates

# People and Collaborations

•Ultrafast optics group (Università Cattolica, Brescia)
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•Ultrafast optics group (Università degli Studi di Trieste)
F. Cilento, G. Coslovich, D. Fausti, F. Parmigiani

•Ultrafast optics group (Politecnico di Milano)
S. Dal Conte, D. Brida, G. Cerullo

•Equilibrium spectroscopies
R. Comin, B. Ludbrook, A. Damascelli (University of British Columbia, Vancouver)
M. Greven (University of Minnesota & Stanford University)
L. Chauviere, B. Keimer (MPG-UBC center for QM)

•Non-equilibrium models of correlated materials
L. Vidmar (LMU Munich), M. Mierzejewski (Katowice), D. Golez, J. Bonca (Ljubljana)
M. Capone, M. Fabrizio (SISSA, Trieste)

•Equilibrium optical properties of HTSC
D. van der Marel (Université de Genève)

Thank you!