

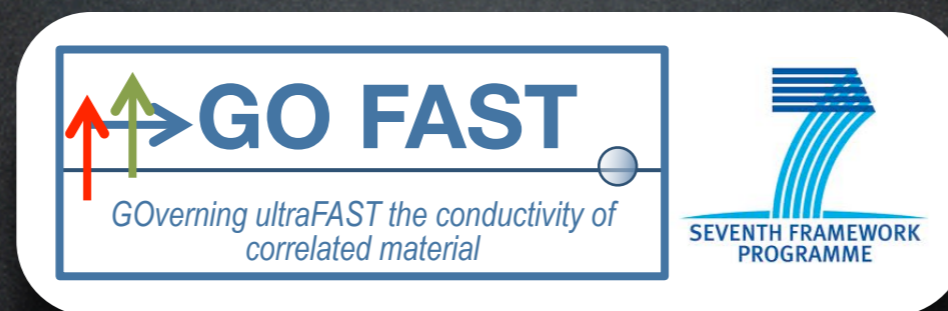
Recent advances in ultrafast spectroscopies

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i-Lamp

(Interdisciplinary laboratories for advanced materials physics)



People and Collaborations

- **Ultrafast optics group** (Università Cattolica, Brescia)
S. Dal Conte, **S. Peli**, F. Banfi, G. Ferrini, C. Giannetti
- **Ultrafast optics group** (Università degli Studi di Trieste)
G. Coslovich, F. Cilento, D. Fausti, F. Parmigiani
- **Ultrafast optics group** (Politecnico di Milano)
D. Brida, G. Cerullo
- **Equilibrium optical properties of HTSC**
D. van der Marel (Université de Genève)
- **Samples**
A. Damascelli (University of British Columbia, Vancouver)
M. Greven (University of Minnesota & Stanford University)
H. Eisaki (NIST, Tsukuba, Japan)



Outline

- Time-resolved **spectroscopies** to disentangle the intertwined degrees of freedom (low-intensity regime)
- optical control of the electronic properties of correlated materials (high-intensity regime)
- broadband ultrafast optical spectroscopy on cuprates



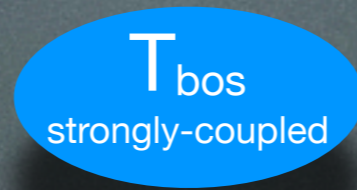
Quasi-particle dynamics in HTSC (Bi2212)

equilibrium spectroscopy

optical properties



occupation of el-states $f(E, T_e)$



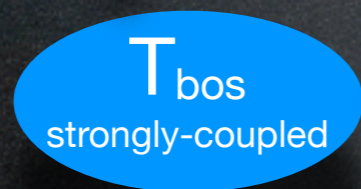
Drude broadening $n(E, T_{bos})$



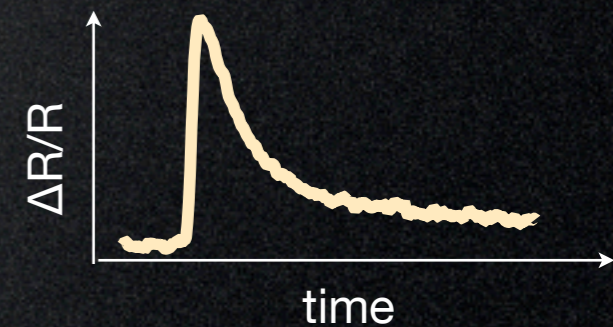
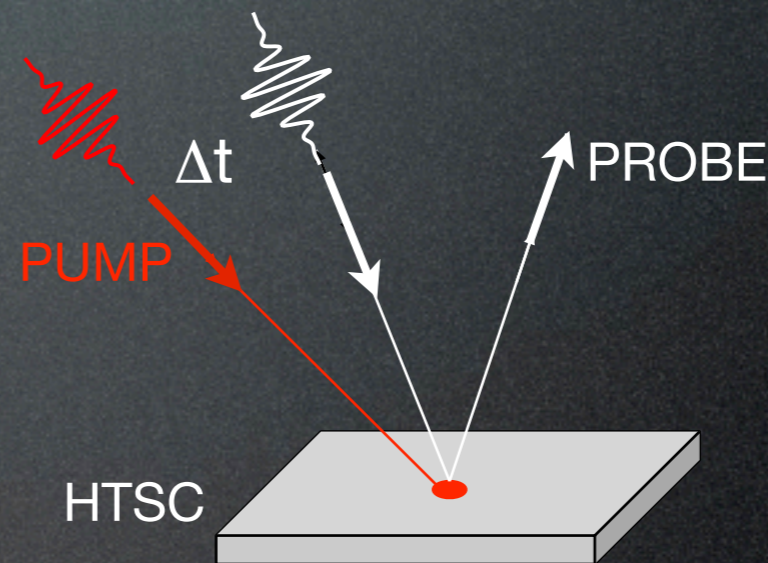
non-equilibrium spectroscopy



occupation of el-states $f(E, T_e)$



Drude broadening $n(E, T_e)$



different timescales



electron-phonon coupling and electron dynamics

METALS

VOLUME 59, NUMBER 13

PHYSICAL REVIEW LETTERS

28 SEPTEMBER 1987

Theory of Thermal Relaxation of Electrons in Metals

Philip B. Allen^(a)

Condensed Matter Physics Branch, Naval Research Laboratory, Washington, D.C. 20375

(Received 6 July 1987)

If electrons in a metal are heated to a temperature T_e greater than the lattice temperature T_L , the electron-phonon interaction causes temperature relaxation $dT_e/dt = \gamma_T(T_L - T_e)$ which is rapid for $T_L > \theta_D$. A formula $\gamma_T = 3\hbar\lambda(\omega^2)/\pi k_B T_e$ is derived, where $\lambda(\omega^2) = \eta/M$ is an important parameter in the theory of superconductivity. Quantitative agreement with recent experiments is good.

PACS numbers: 72.15.Lh, 63.20.Kr, 71.38.+i, 79.20.Ds

2-temperature model

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

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

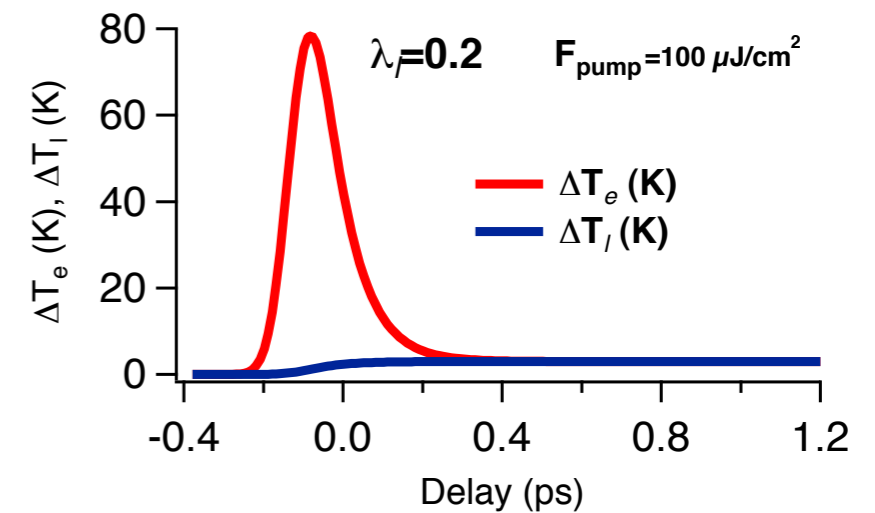
G/C_{lat}

determines the dynamics in the time domain

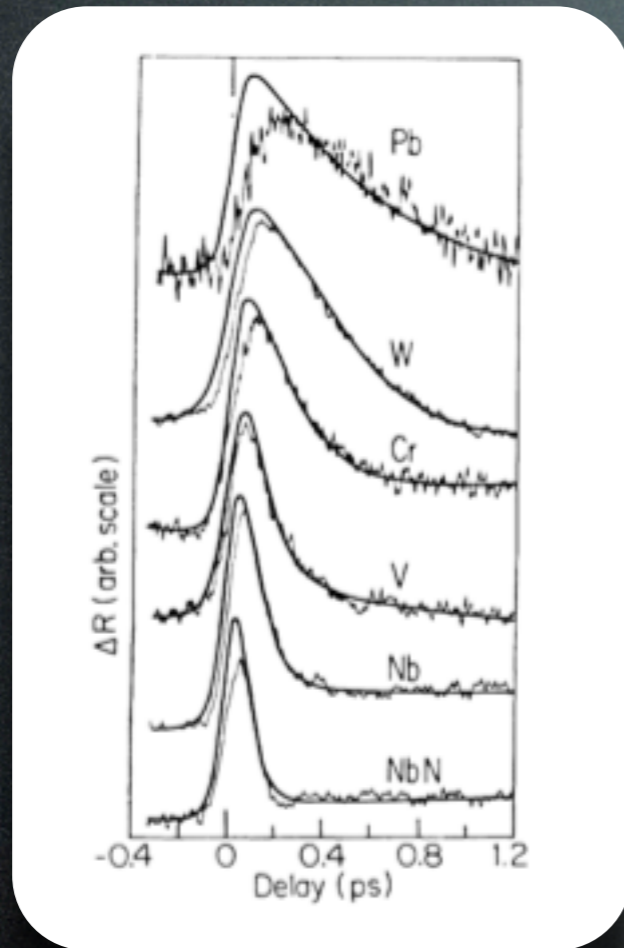
$$G(\Pi_b, T_b, T_e) = \frac{3\gamma_e}{\pi \hbar k_b^2} \int_0^\infty d\Omega \Pi_b(\Omega) \Omega^2 [N(\Omega, T_b) - N(\Omega, T_e)]$$

e-ph coupling: bosonic function

temperature evolution



electron-phonon coupling in metals



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

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

e-ph coupling

$$\lambda = 2 \int \Pi(\Omega) / \Omega d\Omega$$

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

Considering non-thermal distribution ($\tau_{e-e} > \tau_{e-ph}$):

$$\lambda \langle \omega^2 \rangle = \frac{2\pi}{3} \frac{k_B T_l}{\hbar \tau_{e-ph}}$$

V.V. Kabanov and A.S. Alexandrov, *Phys. Rev. B* **78**, 174514 (2008)

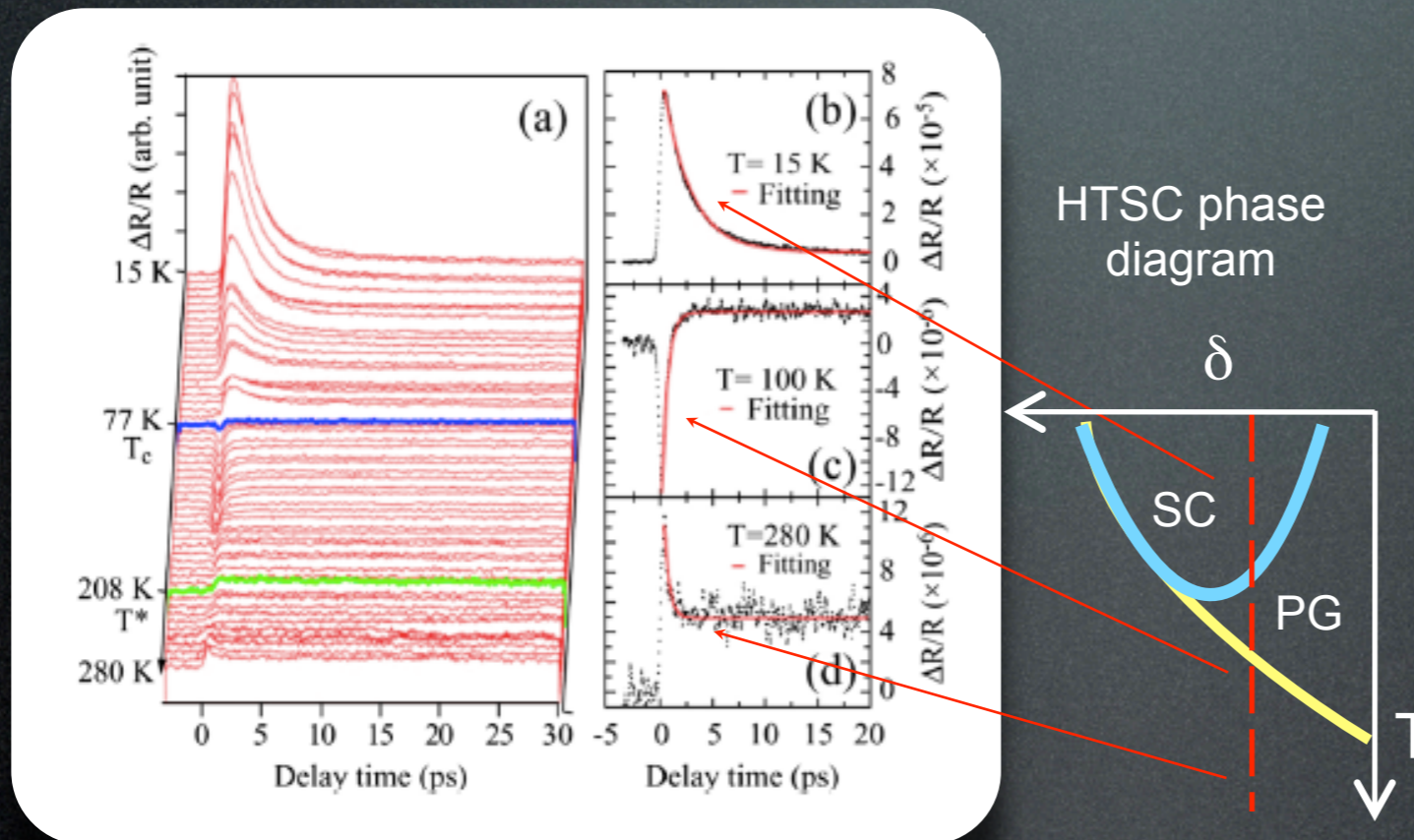
C. Gadermaier et al. *Phys. Rev. Lett.* **105**, 257001 (2010)



Quasi-particle dynamics in HTSC (Bi2212)

single-color experiments on superconducting cuprates at low temperature

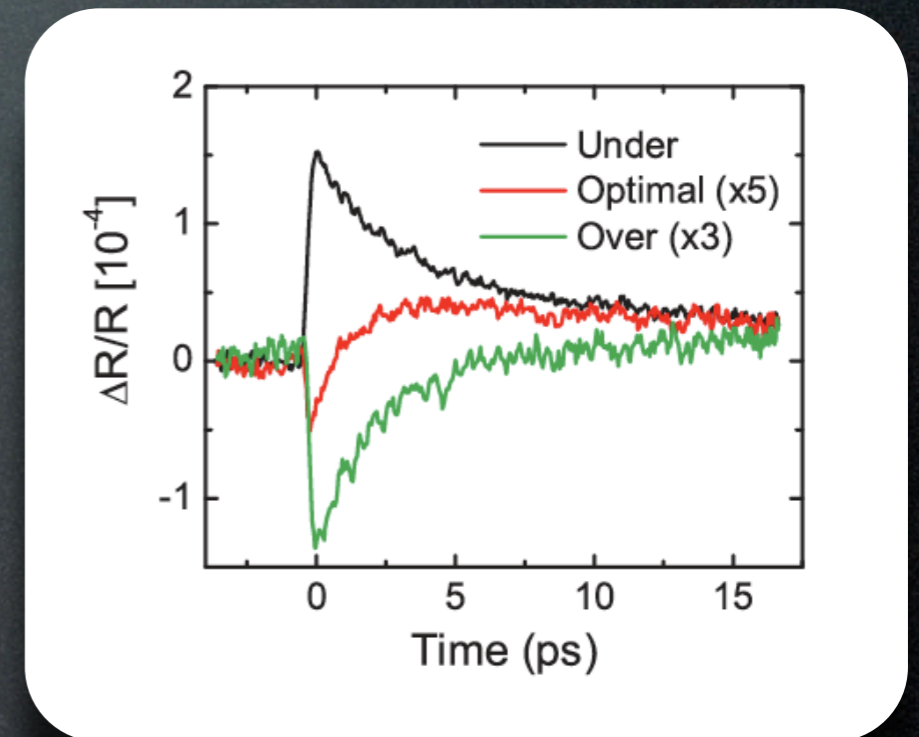
a lot of works from the Ljubljana group...



Y.H. Liu et al., *Phys. Rev. Lett.* **101**, 137003 (2008)

for $T < T_c$ slow dynamics described by Rothwarf-Taylor equations

N. Gedik et al., *Phys. Rev. Lett.* **95**, 117005 (2005)

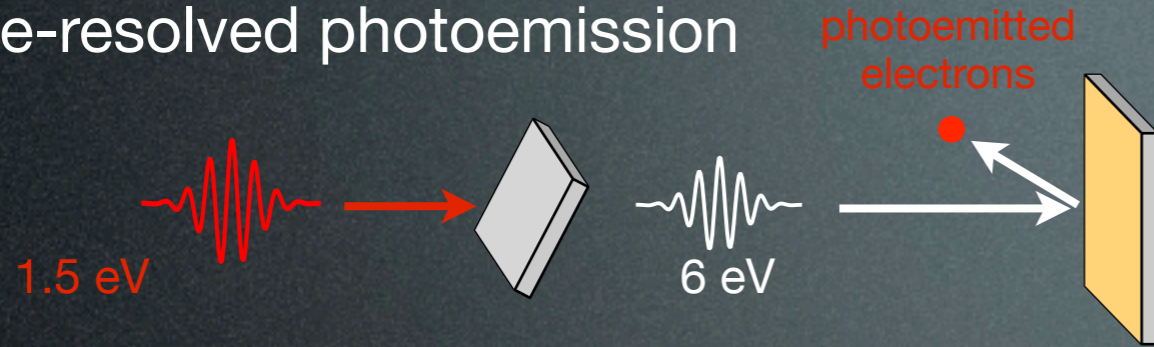


Which is the origin of the reflectivity variations at 1.55 eV?

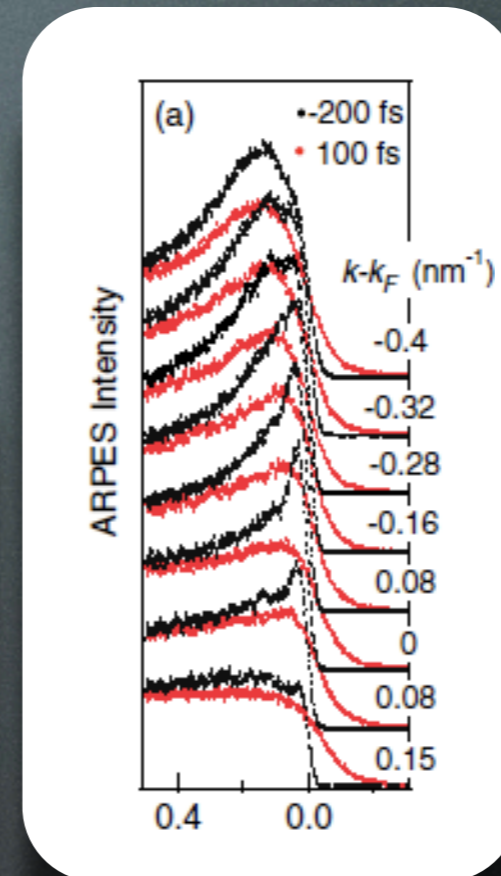
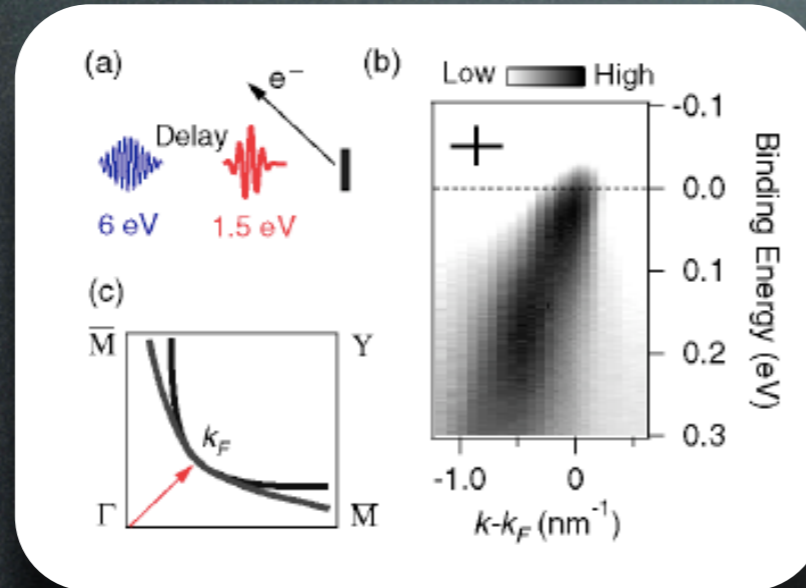


Other time-resolved techniques (Bi2212)

Time-resolved photoemission

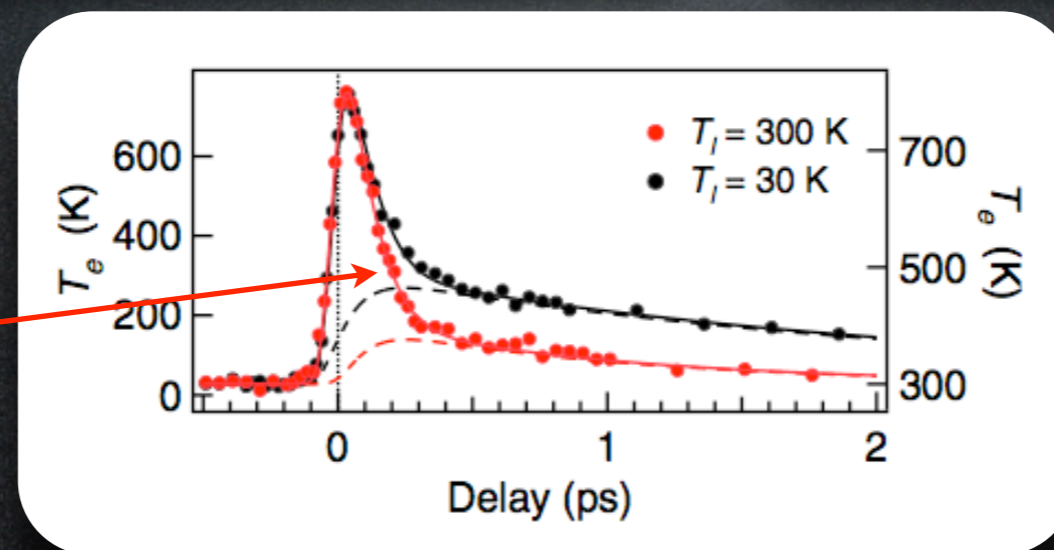


L. Perfetti et al.,
Phys. Rev. Lett. **99**,
197001 (2007)



- direct probe of the electron distribution
- probe limited to the gap-node region, due to the small photon energy

2 dynamics



- beyond the 2-temp. model: strong coupling with a subset of phonons



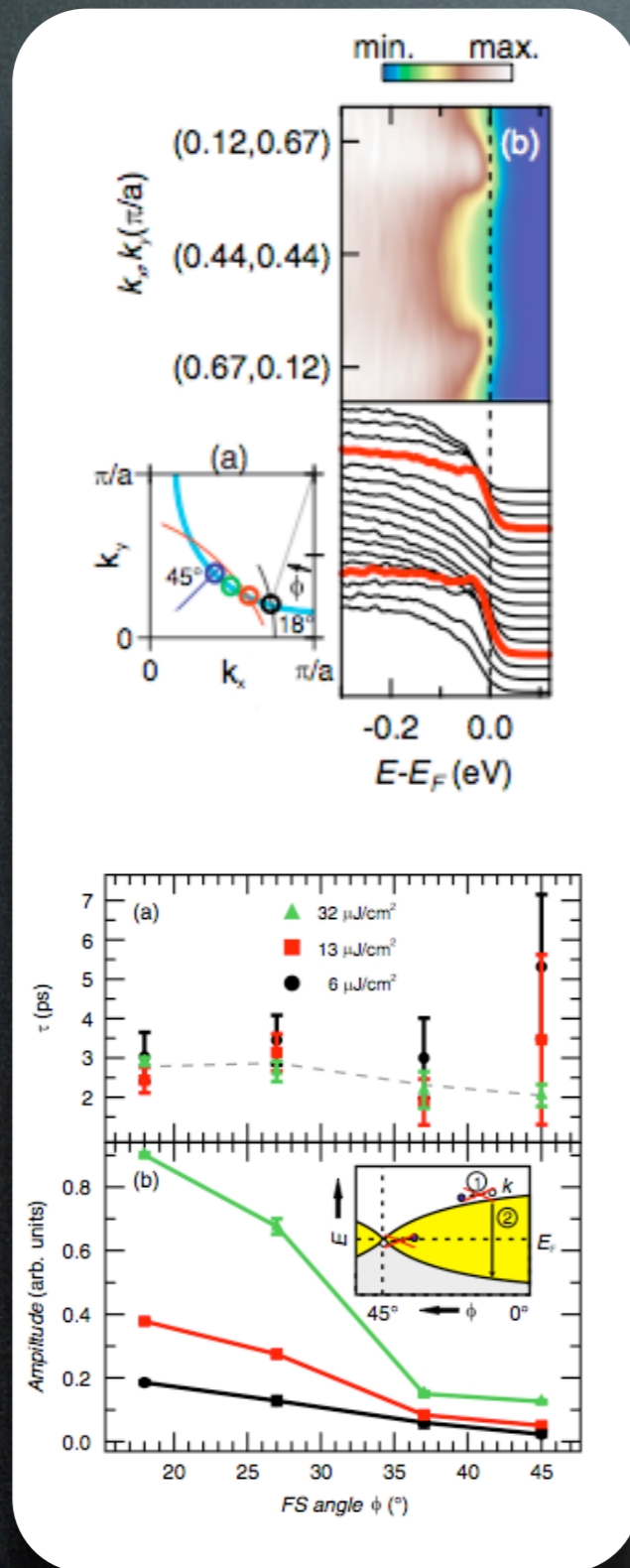
Other time-resolved techniques (Bi2212)

Exploring the entire Brillouin zone

R. Cortés et al.
Phys. Rev. Lett. **107**,
097002 (2011)

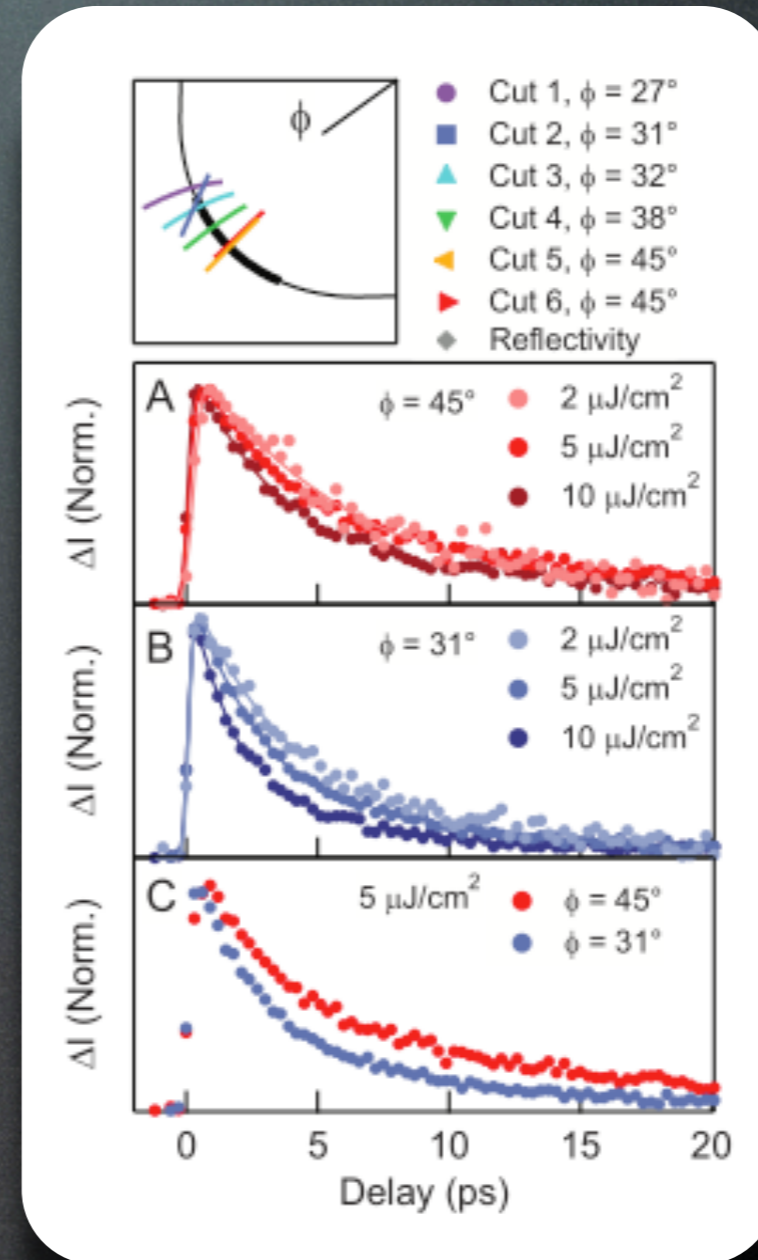
- relaxation INDEPENDENT of the k-space position

- non-thermal electron distribution



J. Graf et al. *Nature Physics* **7**, 805 (2011)

C.L. Smallwood et al. *Science* **336**, 1137 (2012)

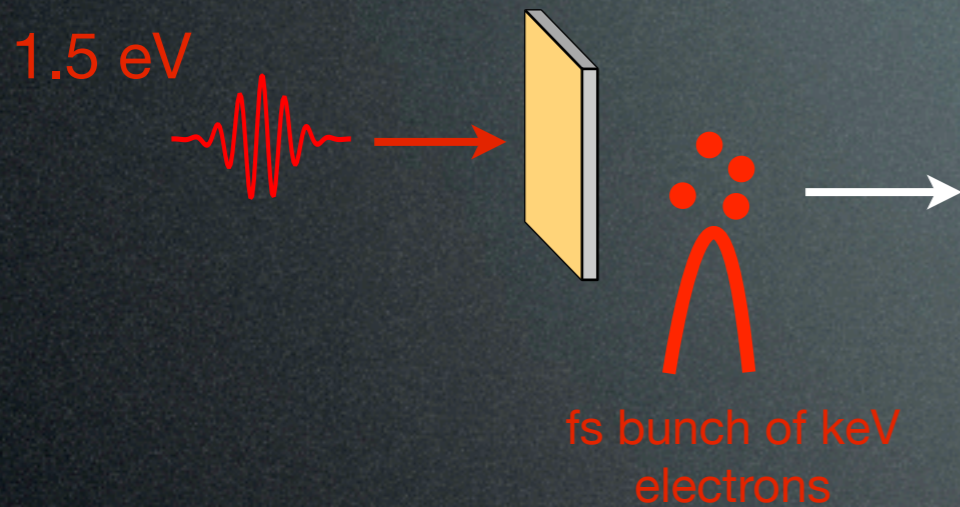


- relaxation DEPENDENT on the k-space position

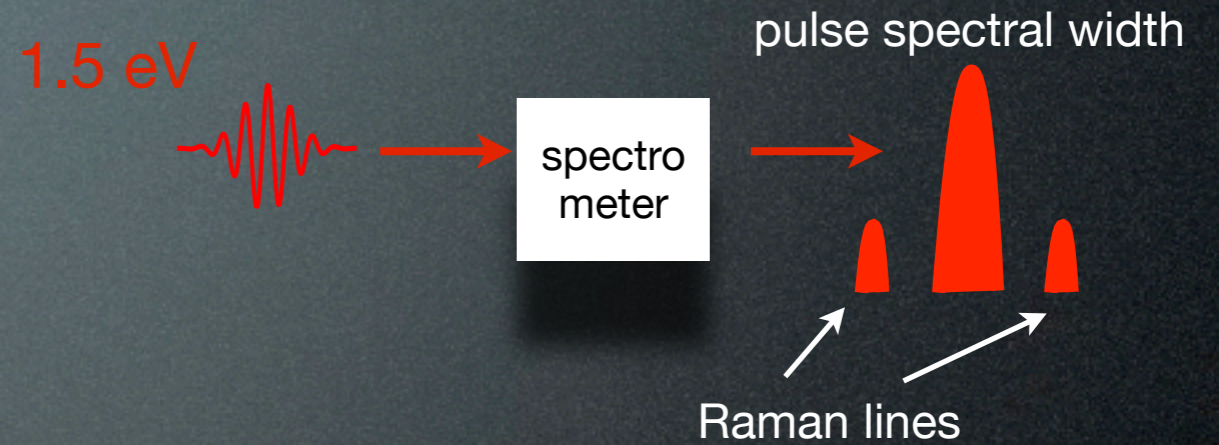


Other time-resolved techniques (Bi2212)

Time-resolved electron diffraction

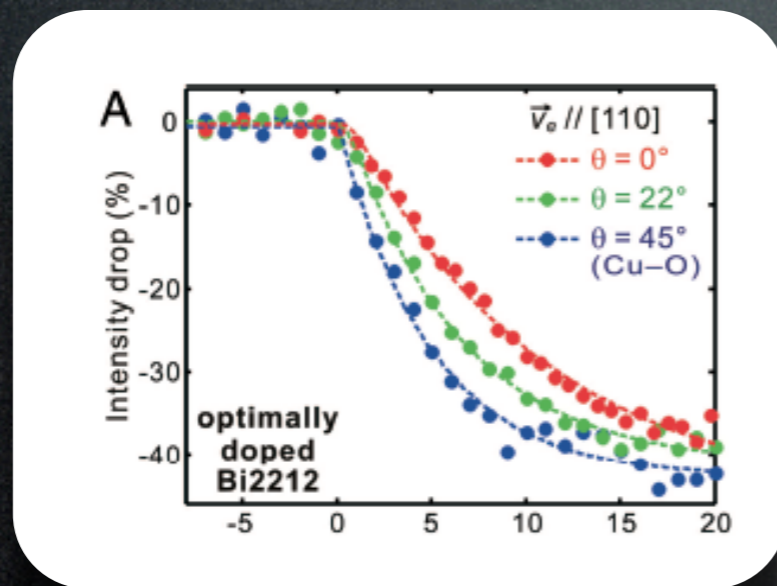


Time-resolved Raman scattering



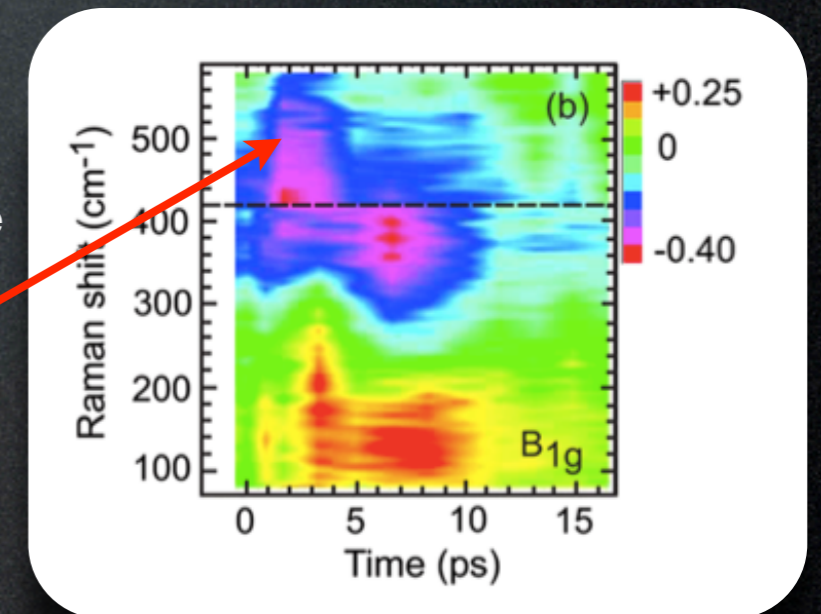
Anisotropic electron-phonon coupling

- $\lambda_{e-ph} = 0.08 - 0.55$
- fluence $> 1 \text{ mJ/cm}^2$



F. Carbone et al., *P.N.A.S.* **105**, 20161 (2008)

emission of in-plane oxygen breathing modes on the ps timescale



R.P. Saichu et al., *Phys. Rev. Lett.* **102**, 177004 (2009)



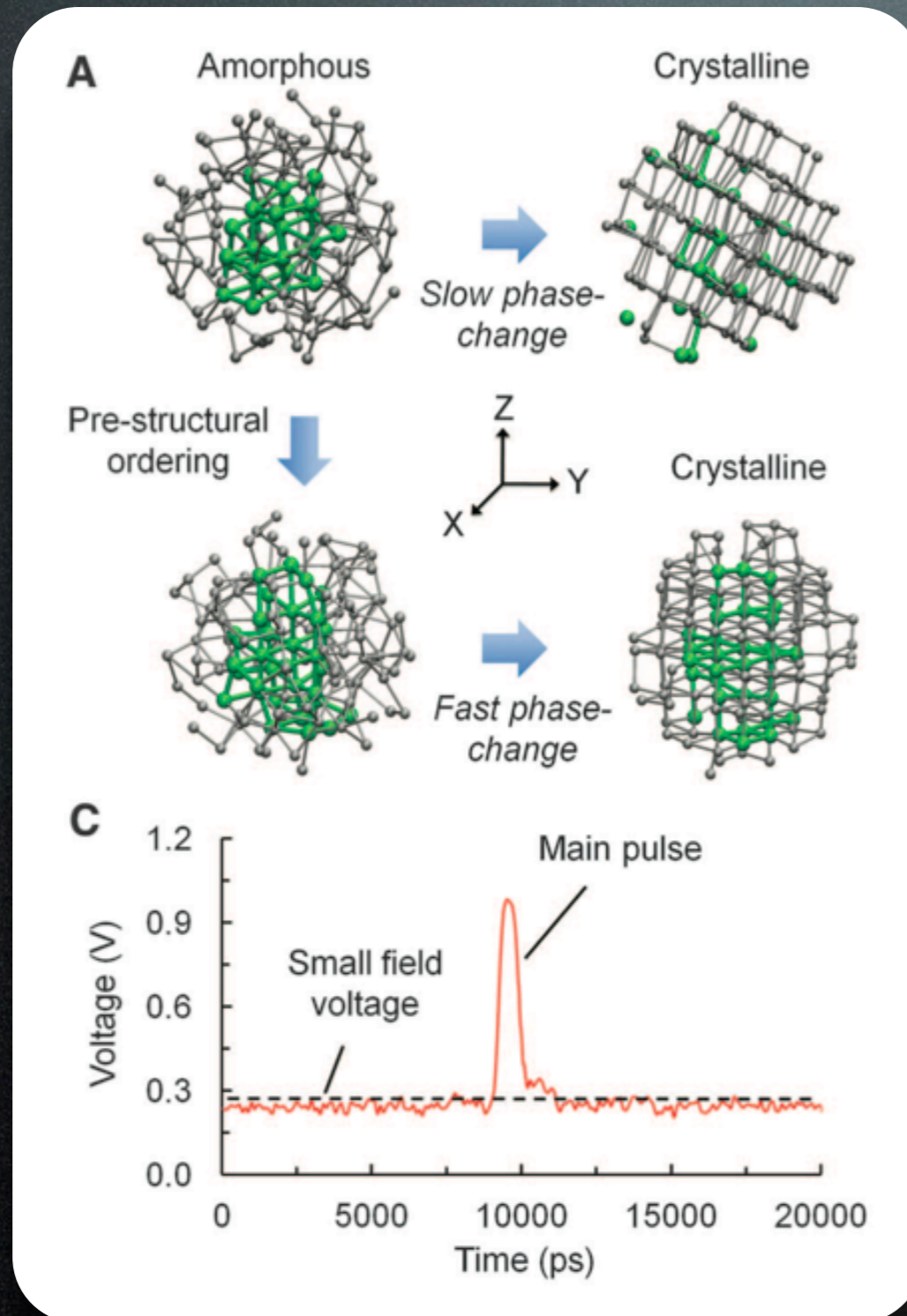
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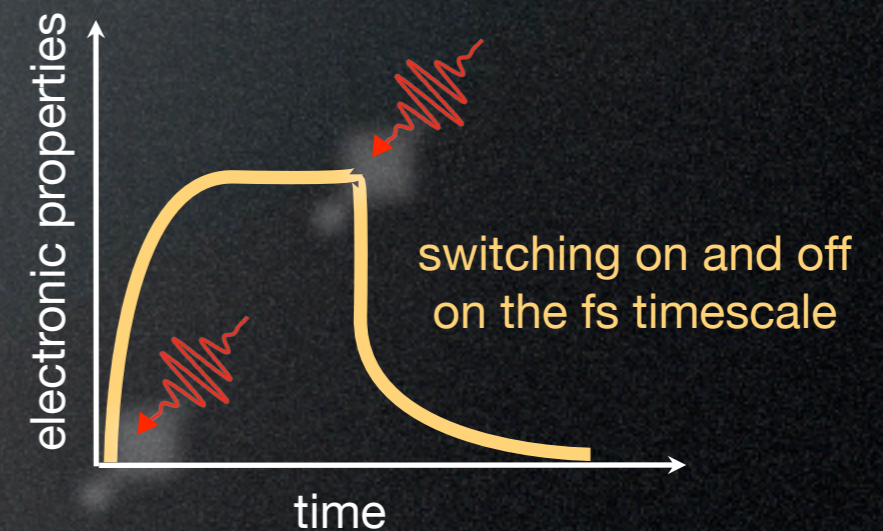
Breaking the Speed Limits of Phase-Change Memory

$\text{Ge}_2\text{Sb}_2\text{Te}_5$



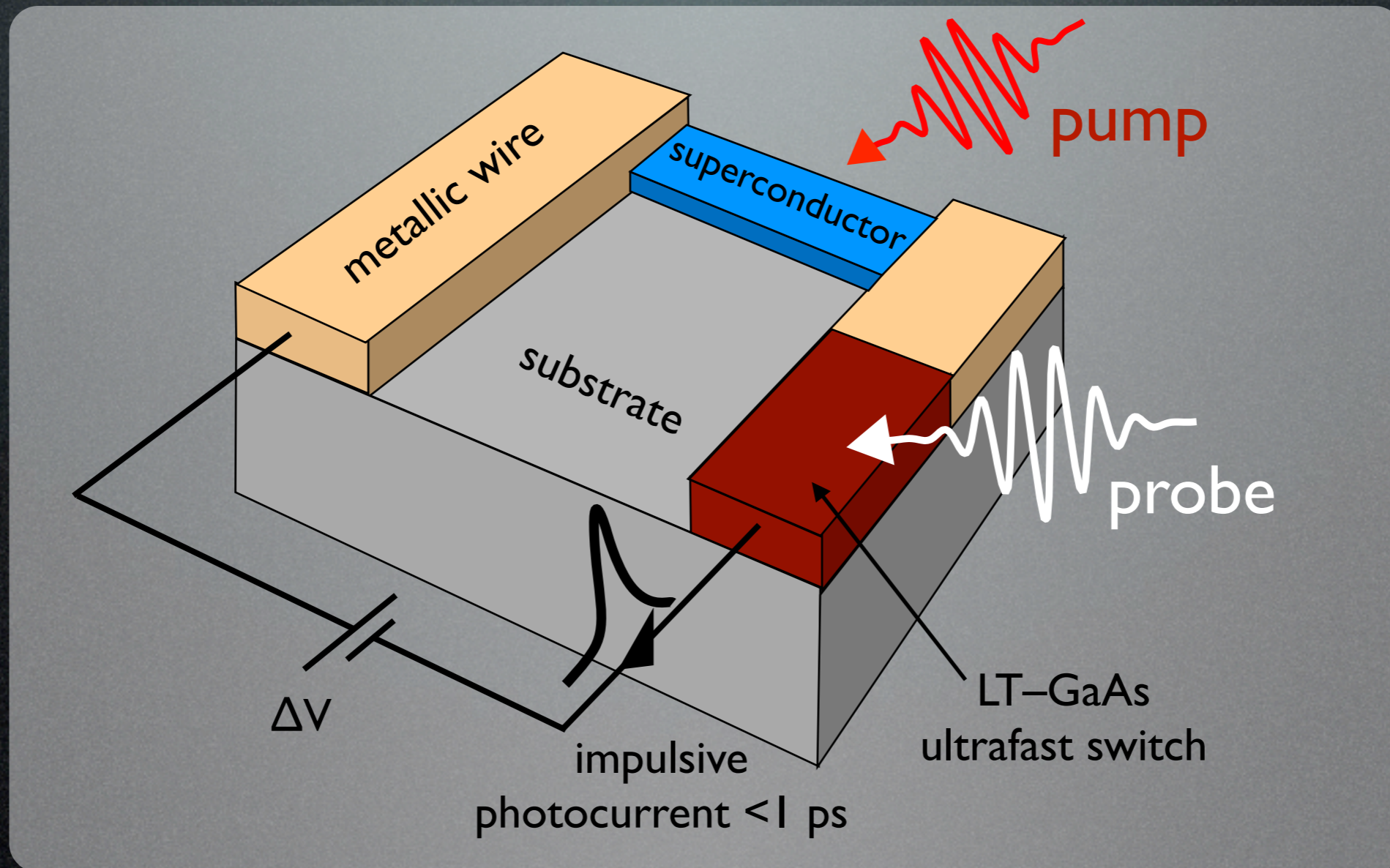
D. Loke et al. *Science* **336**, 1566 (2012)

bottleneck for structural rearrangement in PCRAM:
500 ps



Breaking the Speed Limits of Phase-Change Memory

using optical pulses to control superconductivity in a realistic device



Governing ultrafast the conductivity of correlated materials

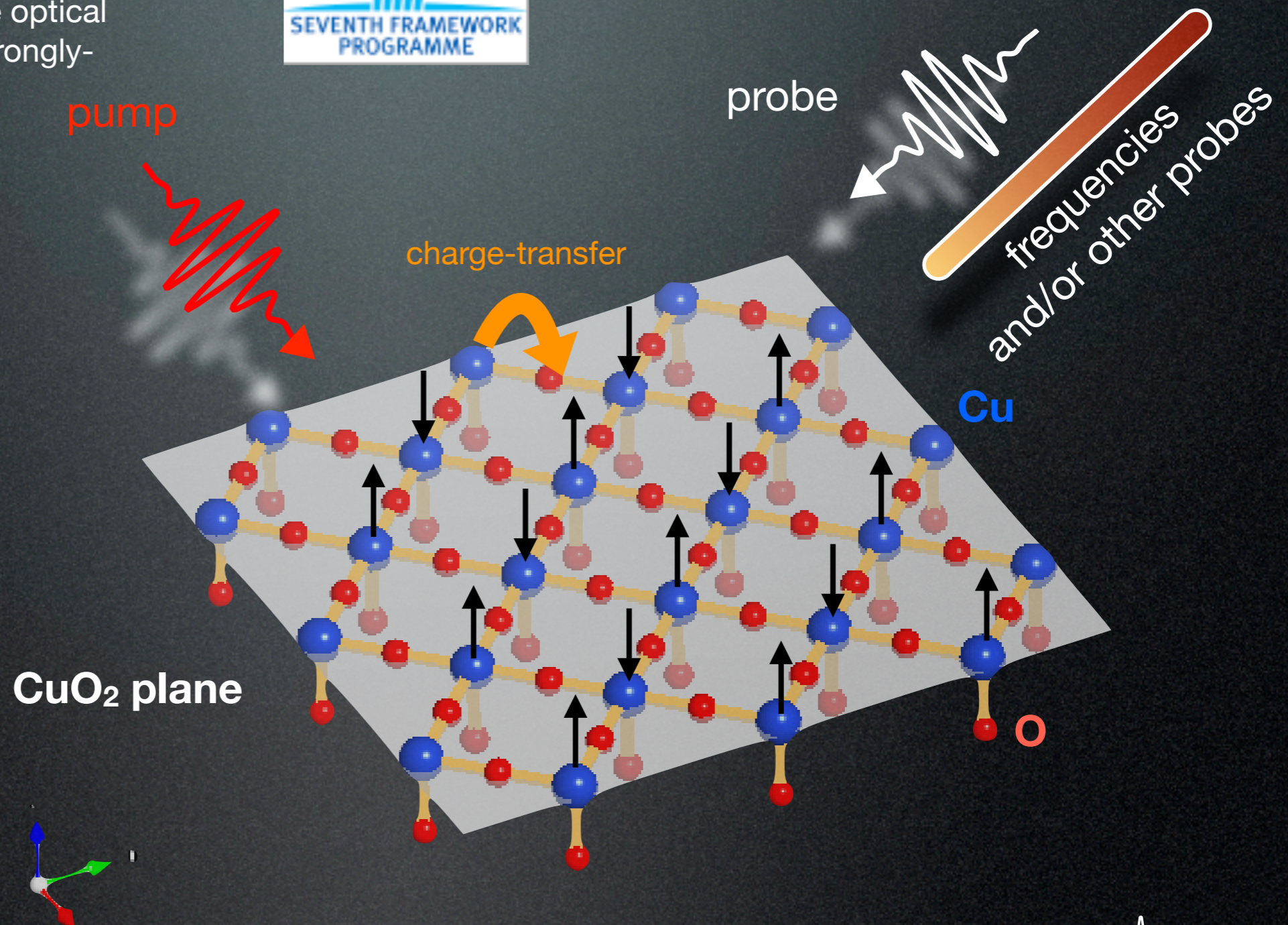


GO FAST

GOAL: Development of theoretical and experimental tools to achieve the optical control of electronic phases in strongly-correlated materials

- Time-dependent DMFT (SISSA)
- Time-resolved broadband spectroscopies (THz-visible) (Brescia, Trieste, Nijmegen)
- Time-resolved photoemission (Duisburg, Orsay)
- Time-resolved X-ray diffraction, electron diffraction (Duisburg)

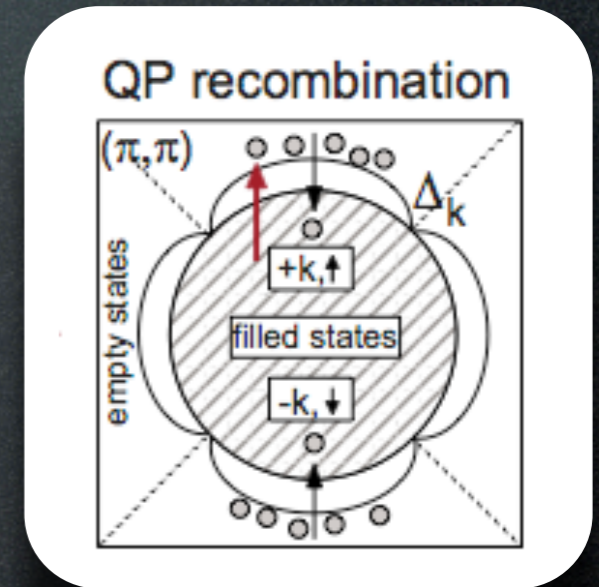
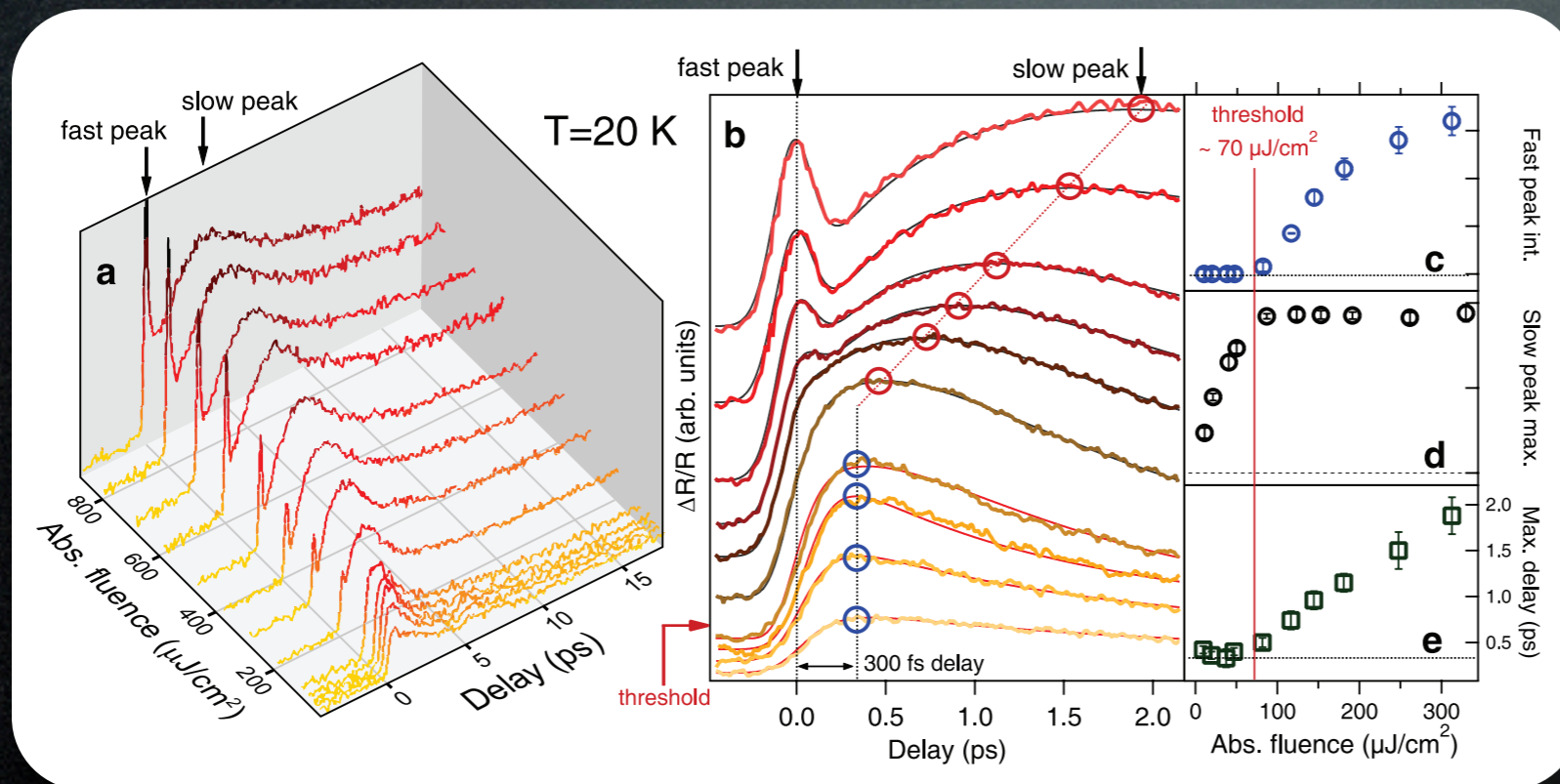
+
Micron Technology, Inc.
MenloSystems GmbH



Non-thermal quenching of superconductivity

Non-thermal photo-induced phase transition (Bi2212)

- fluence constraint for experiments $< 70 \mu\text{J}/\text{cm}^2$ (underdoped, $T=20\text{K}$)



first or second order?
in non-equilibrium conditions

$$F_{\text{SC}}(P, T, n) = F_{\text{N}}(P, T, n)$$

before complete closing of $\Delta_{\text{SC}}(T)$

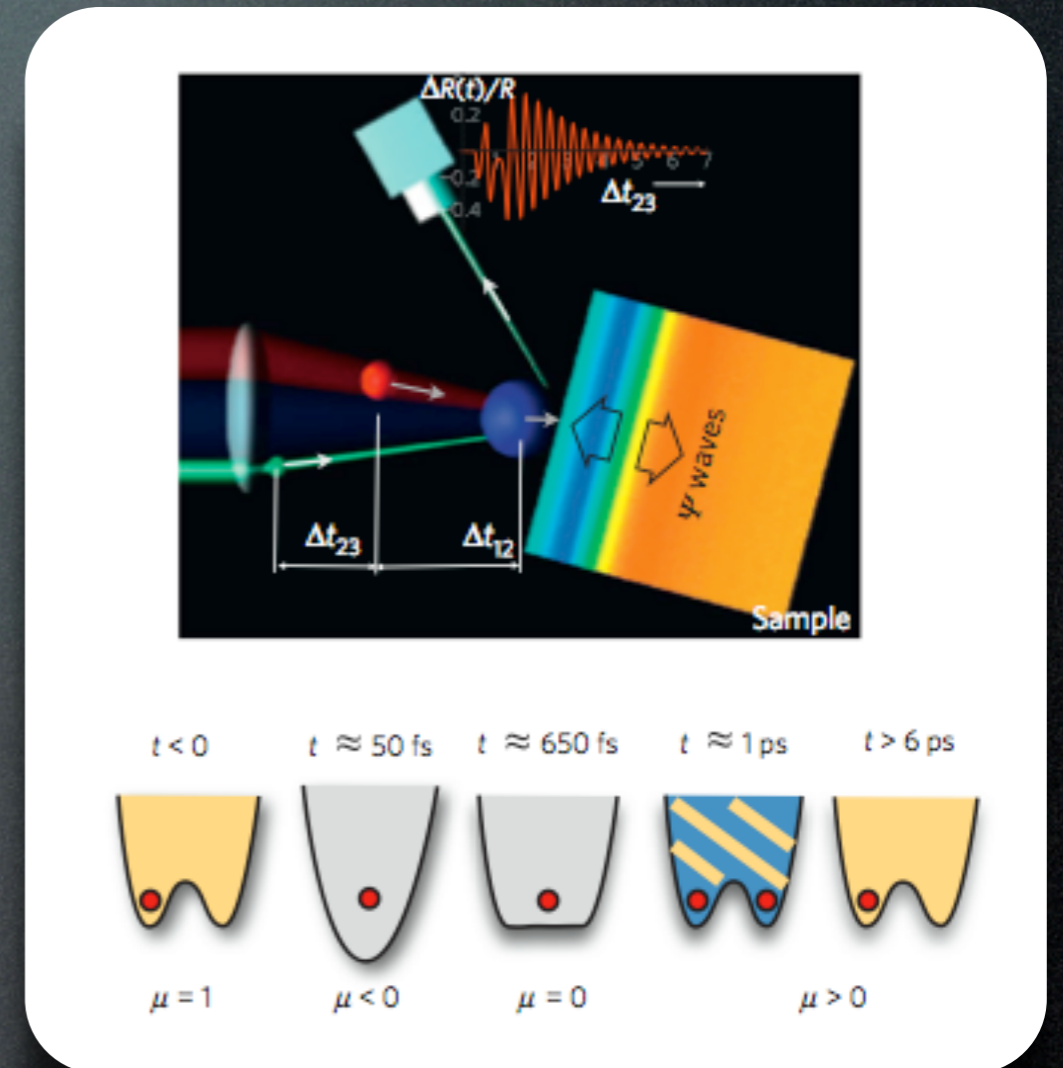
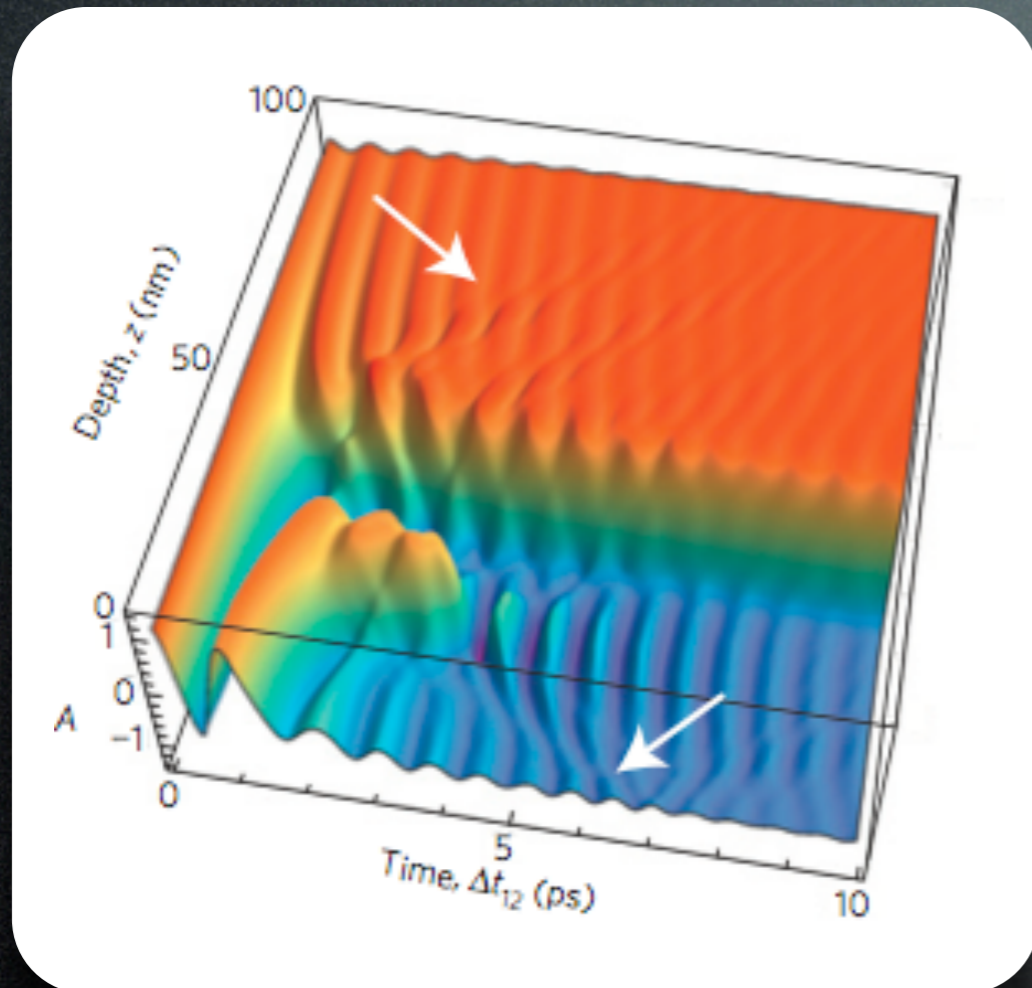
E.J Nicol and J.P. Carbotte
Phys. Rev. B **67**, 214506 (2003)

- R.A. Kaindl et al. *Phys. Rev. B* **72**, 060510R (2005)
 P. Kusar et al. *Phys. Rev. Lett.* **101**, 227001 (2008)
 C. Giannetti et al., *Phys. Rev. B* **79**, 224502 (2009)
 G. Coslovich et al., *Phys. Rev. B* **83**, 064519 (2011)
 L. Stojchevska et al. *Phys. Rev. B* **84**, 180507 (2011)



Role of the inhomogeneities in the excitation process

Time-evolution of the order parameter within Ginzburg-Landau model in CDW materials



$$U = \int dz \left(-\frac{1}{2}(1 - \eta) \frac{\Delta(t, z)^2}{\Delta_0^2} + \frac{1}{4} \frac{\Delta(t, z)^4}{\Delta_0^4} + \frac{1}{2} \frac{\xi^2}{\Delta_0^2} \left(\frac{\partial \Delta(t, z)}{\partial z} \right)^2 \right)$$

R. Yusupov et al., *Nature Physics* **6**, 681 (2010)

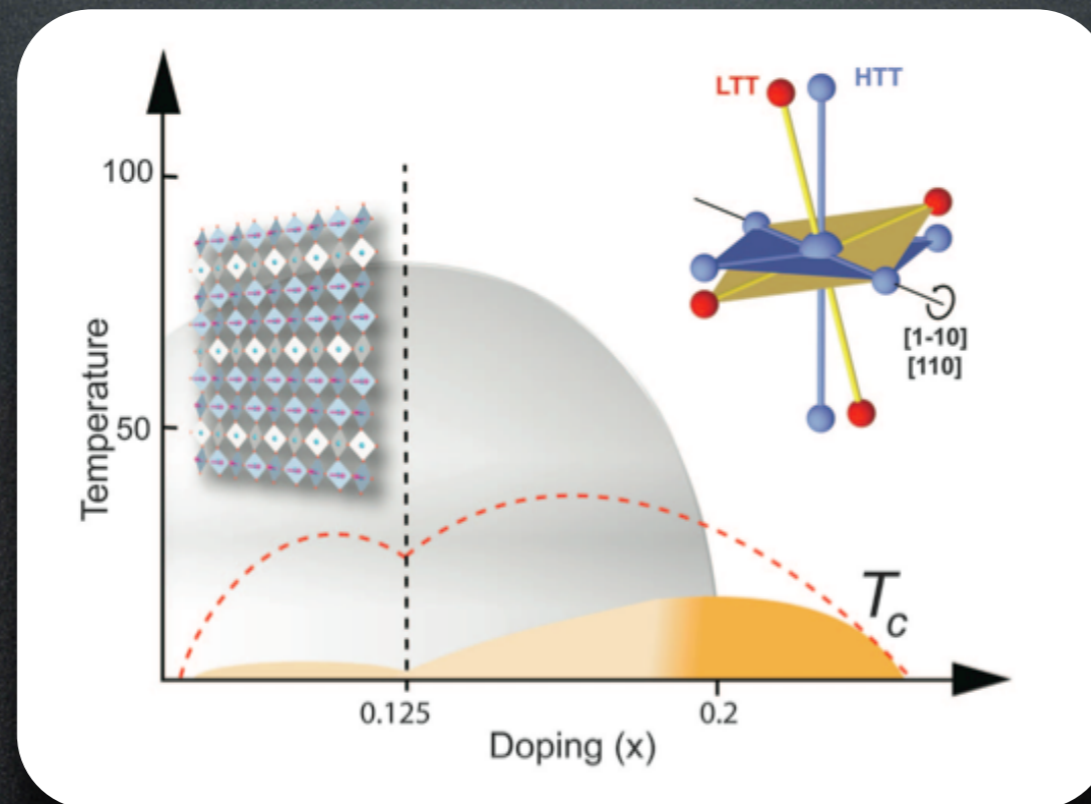


Photoinducing transient superconductivity

possible routes to control superconductivity

- Optically removing competing orders
- Effective cooling of low-energy excitations through THz pumping (Tinkham)
- Photoinducing changes in the electronic structure of correlated materials

THz
PUMP



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



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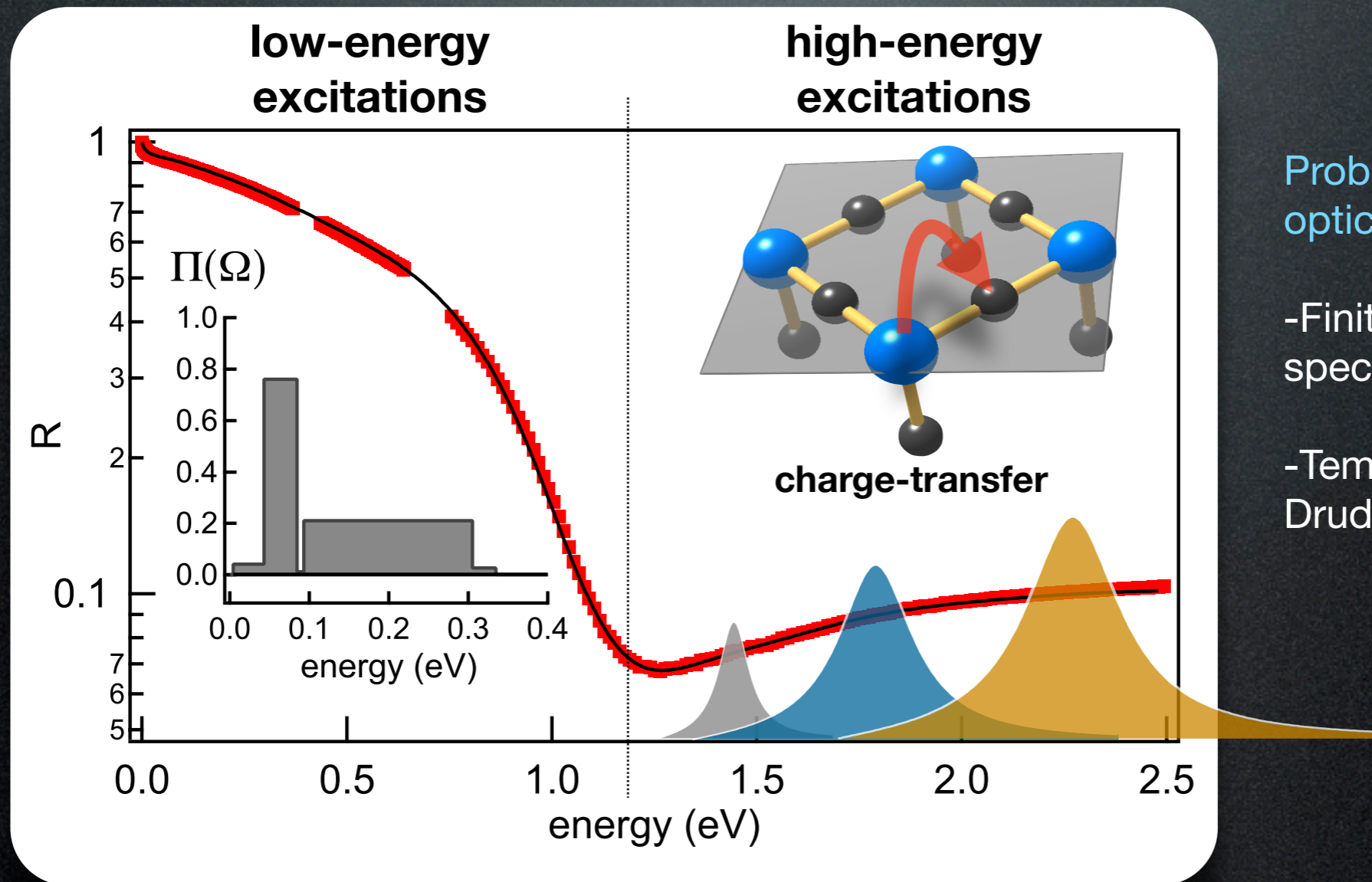


Optical properties of a cuprate superconductor

extended Drude

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

Lorentz oscillators



Problems of equilibrium optical spectroscopy:

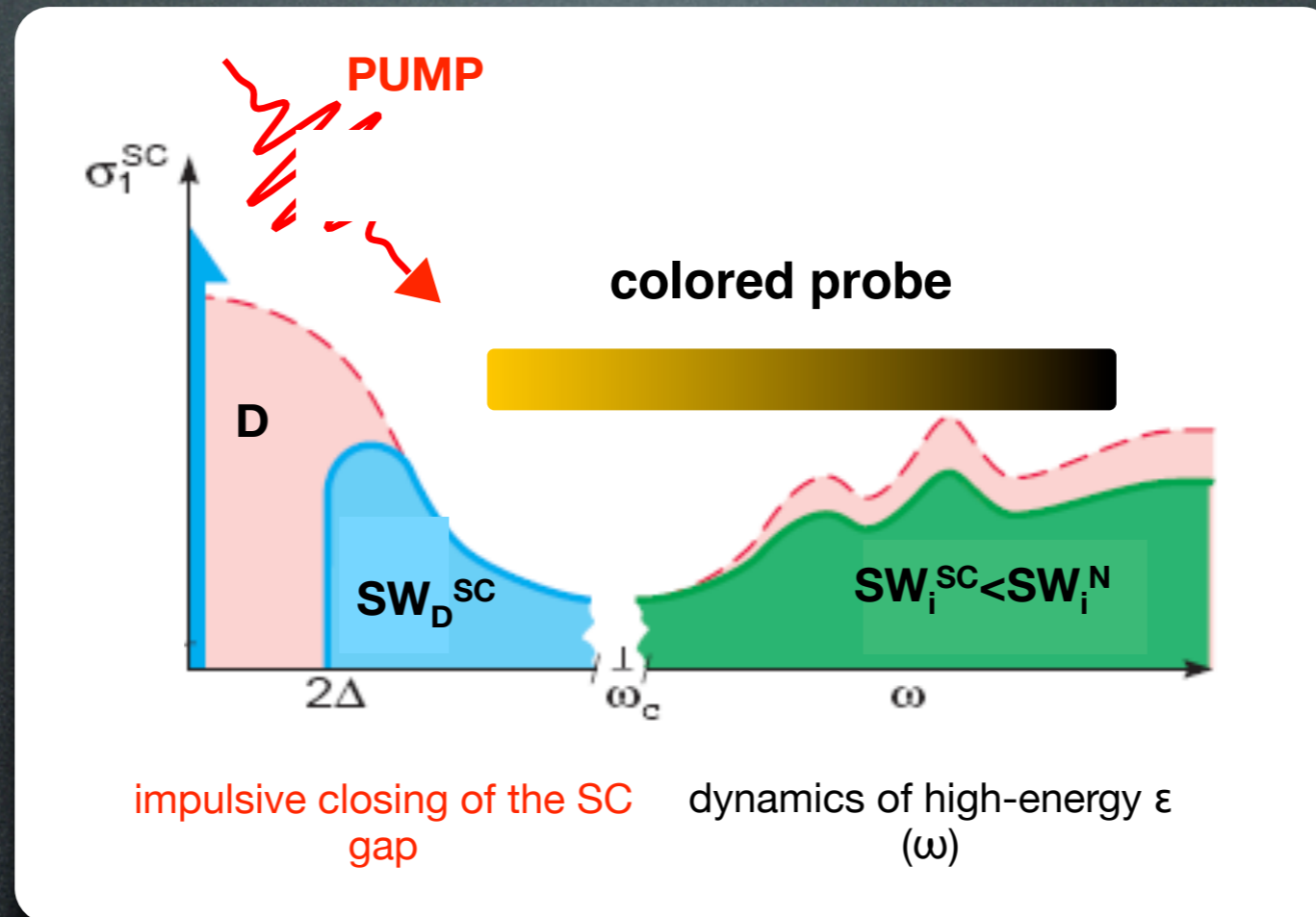
- Finite cut-off for calculating spectral weight shifts
- Temperature dependent Drude broadening

optimally-doped Bi2212. Data from van der Marel's group



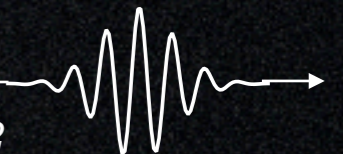
time-resolved broadband spectroscopy

time+spectral resolution



differential model

Fit function: $\delta\epsilon = \epsilon_{exc} - \epsilon_{eq} \longrightarrow \frac{\delta R}{R}(\omega, t) = \frac{R_{exc}(\omega, t) - R_{eq}(\omega)}{R_{eq}(\omega)}$



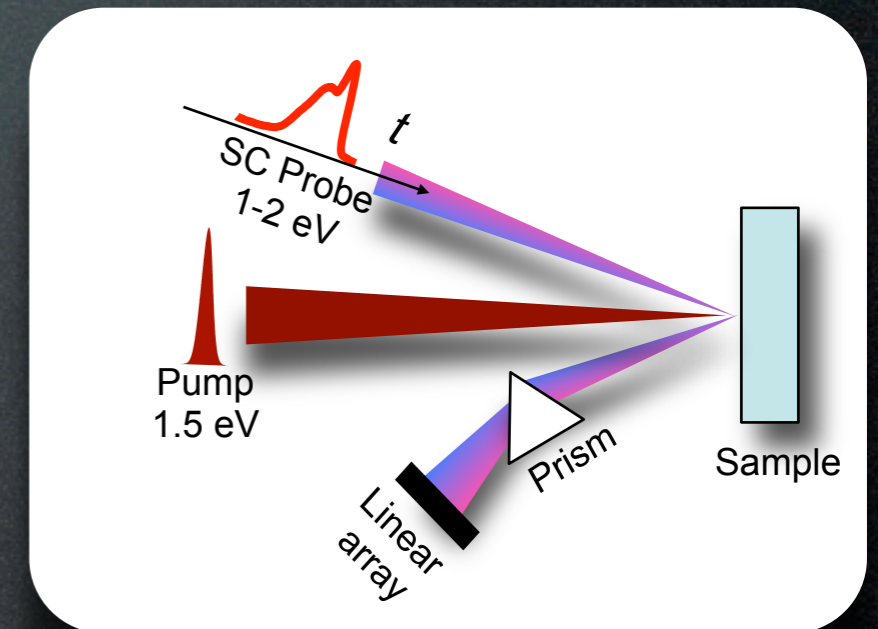
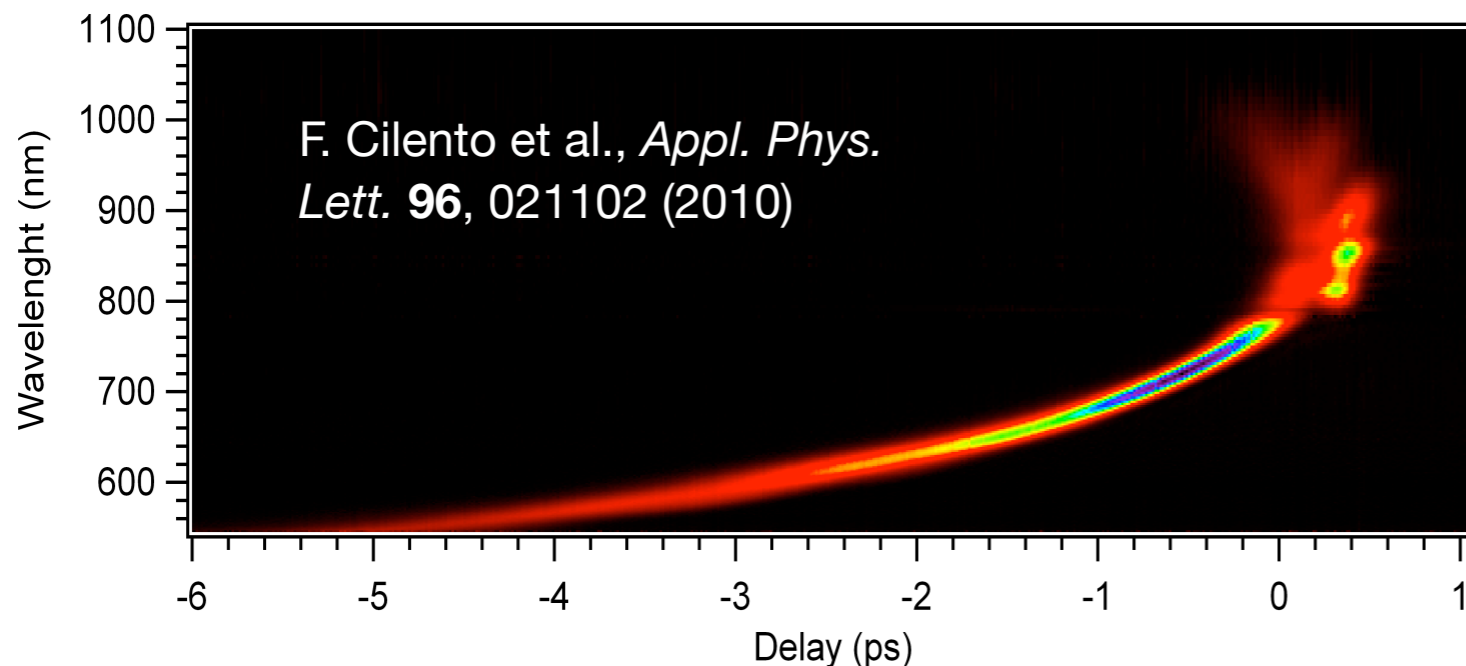
Time-resolved optical spectroscopy

problem:

avoid non-thermal destruction of the superconducting phase transition

Low-fluence ($<20 \mu\text{J}/\text{cm}^2$) and high rep.rate \rightarrow supercontinuum by a photonic fiber

Spectrogram of a supercontinuum pulse



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

- R.A. Kaindl et al. *Phys. Rev. B* **72**, 060510R (2005)
- P. Kusar et al. *Phys. Rev. Lett.* **101**, 227001 (2008)
- C. Giannetti et al., *Phys. Rev. B* **79**, 224502 (2009)
- G. Coslovich et al., *Phys. Rev. B* **83**, 064519 (2011)
- M. Beyer et al., *Phys. Rev. B* **83**, 214515 (2011)
- L. Stojchevska et al. *Phys. Rev. B* **84**, 180507 (2011)



Time-resolved optical spectroscopy on Y-Bi2212

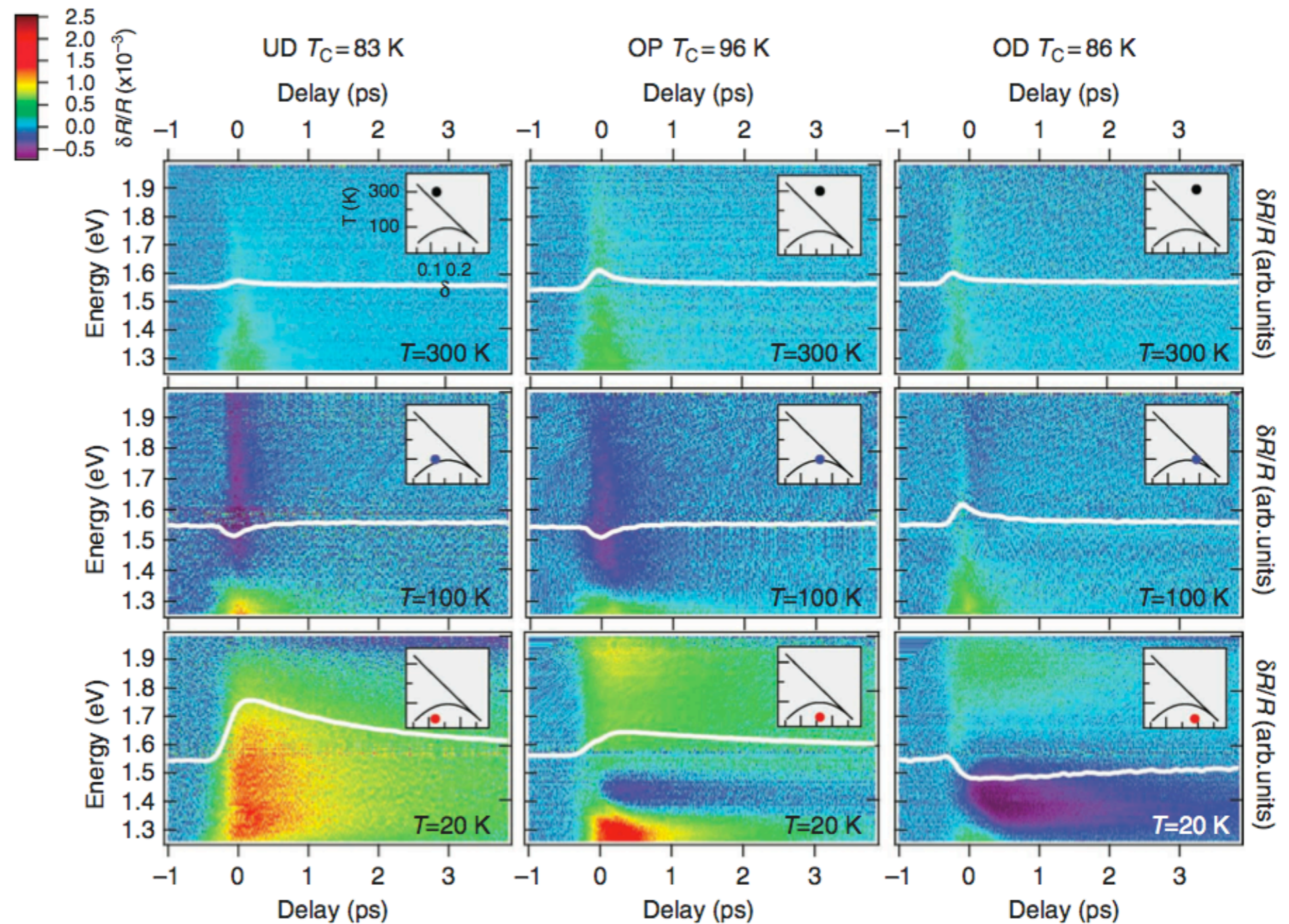
$$\frac{\delta R}{R}(\omega, t)$$

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

Normal
state

Pseudogap
state

Superconductive
state



$\text{Bi}_2\text{Sr}_2\text{Ca}_{0.92}\text{Y}_{0.08}\text{Cu}_2\text{O}_{8+\delta}$

C. Giannetti et al., *Nature Commun.* 2:353 (2011)

C. Giannetti

NGSCES, Portoroz 24-29 June 2012



Conclusions

- Broadband optical spectroscopy is a novel tool to investigate the ultrafast dynamics in correlated materials and high-temperature superconductors
- Ultrashort light pulses can be used to optically control the electronic properties of correlated superconductors on the fs timescale

