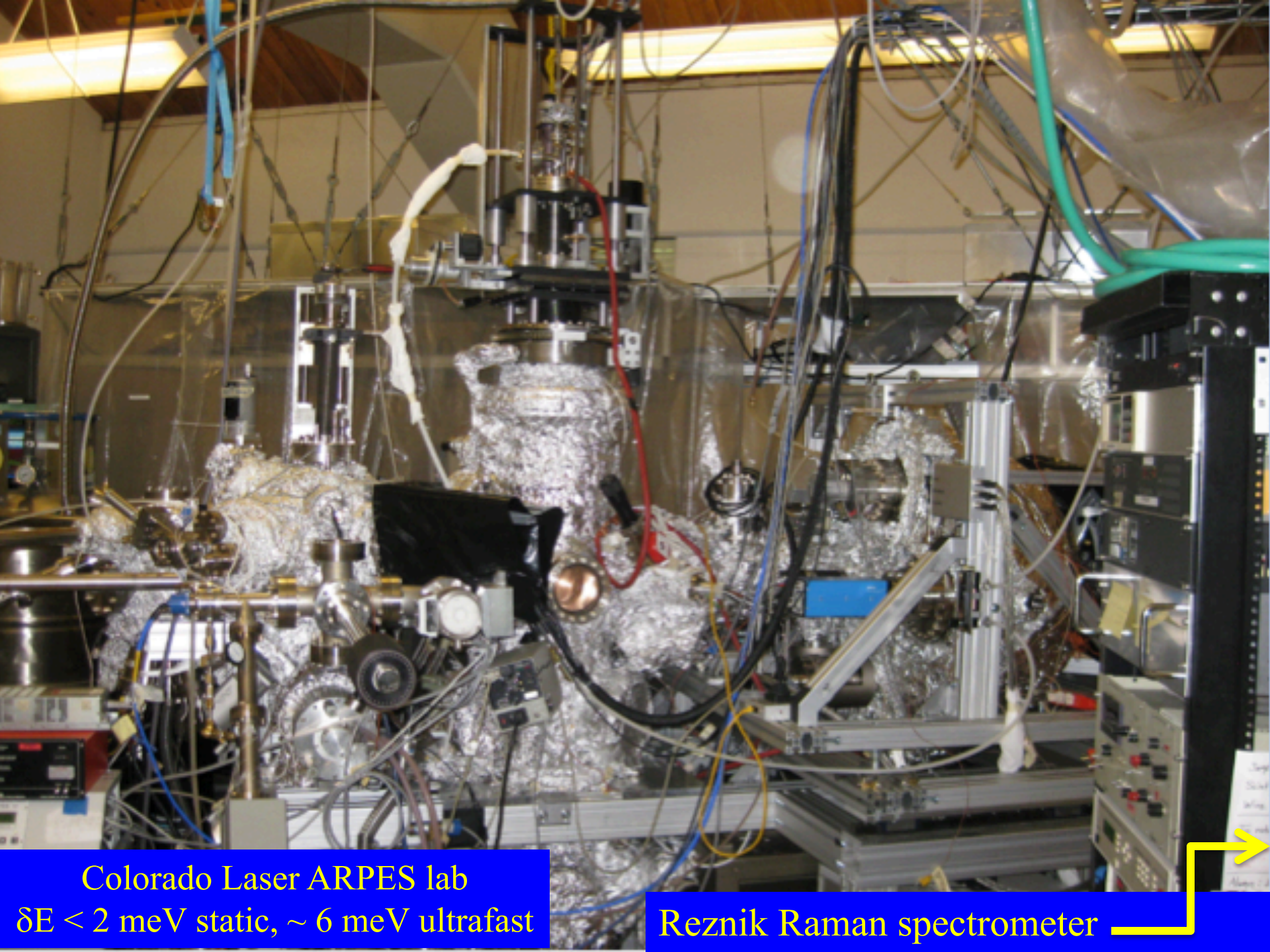


# Electron Dynamics and Electronic Interactions in Cuprates and Related Materials

Ultrafast and static ARPES.  
Evolution of electronic self-energies  
("ARPES as a Self-Energy Spectroscopy")  
(scattering rates, mass enhancements,  $++$ , vs.  $\omega$ ,  $T$ ,  $k$ ,  $x$ , etc.)

Dan Dessau  
University of Colorado, Boulder

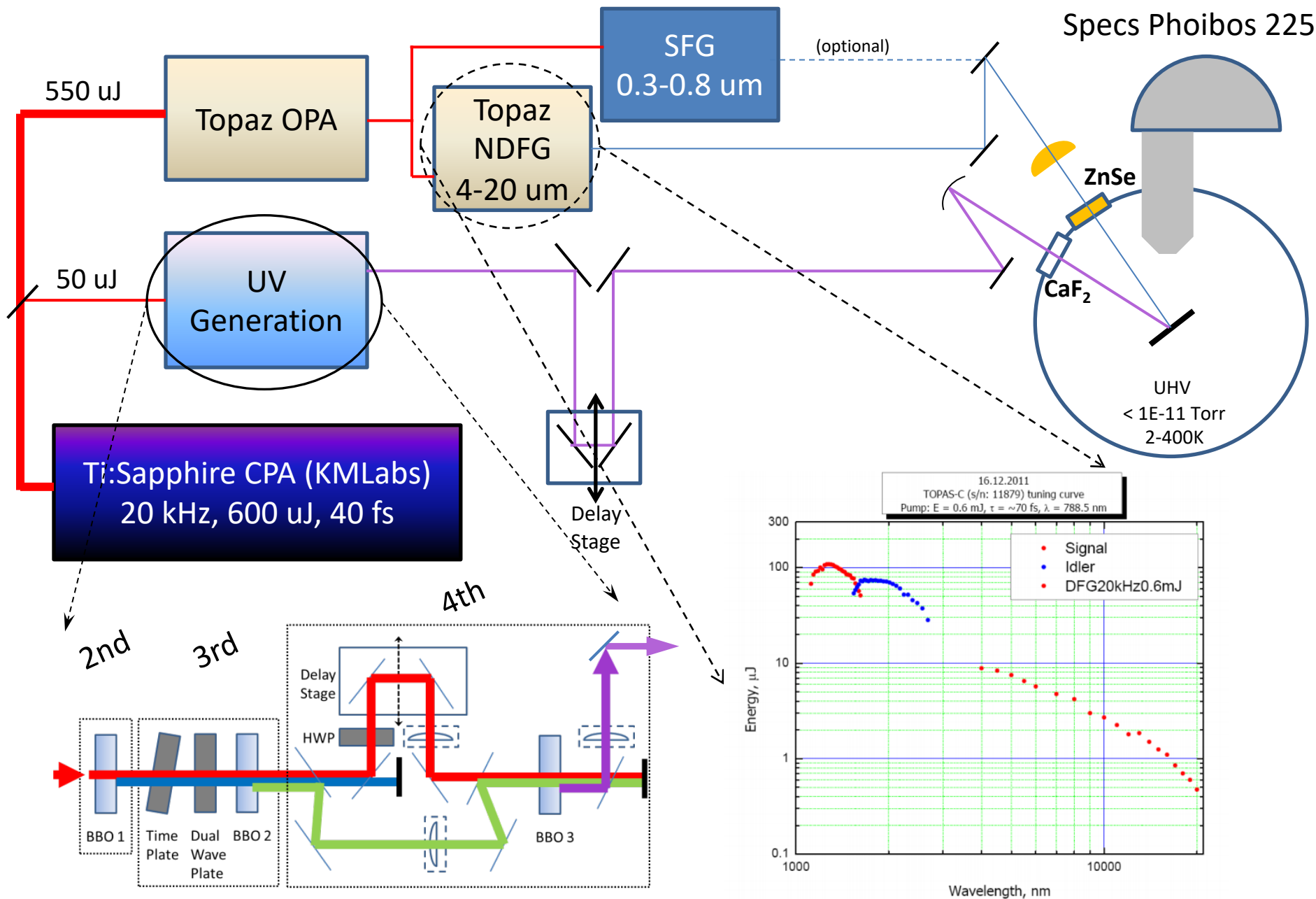


Colorado Laser ARPES lab

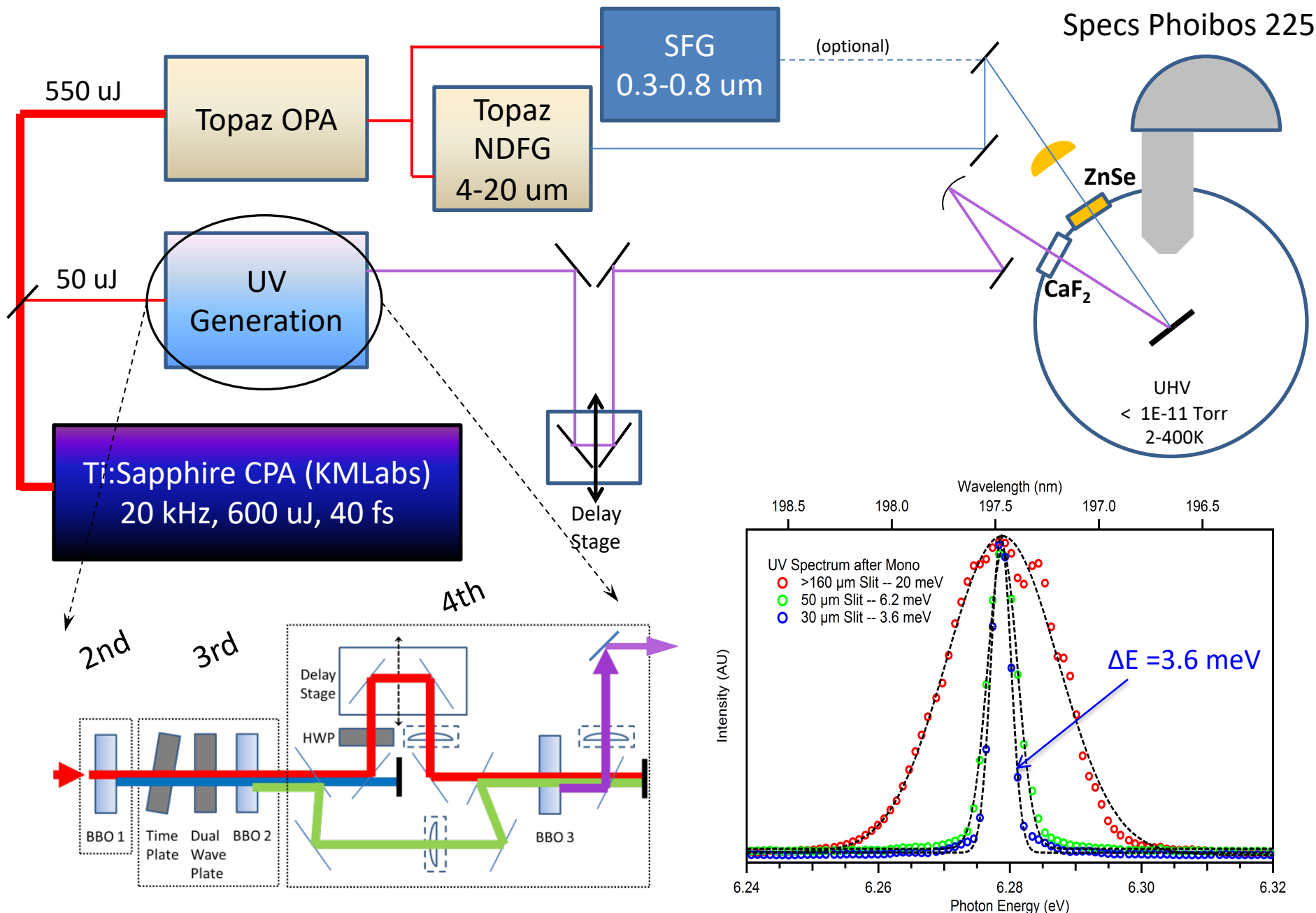
$\delta E < 2$  meV static,  $\sim 6$  meV ultrafast

Reznik Raman spectrometer

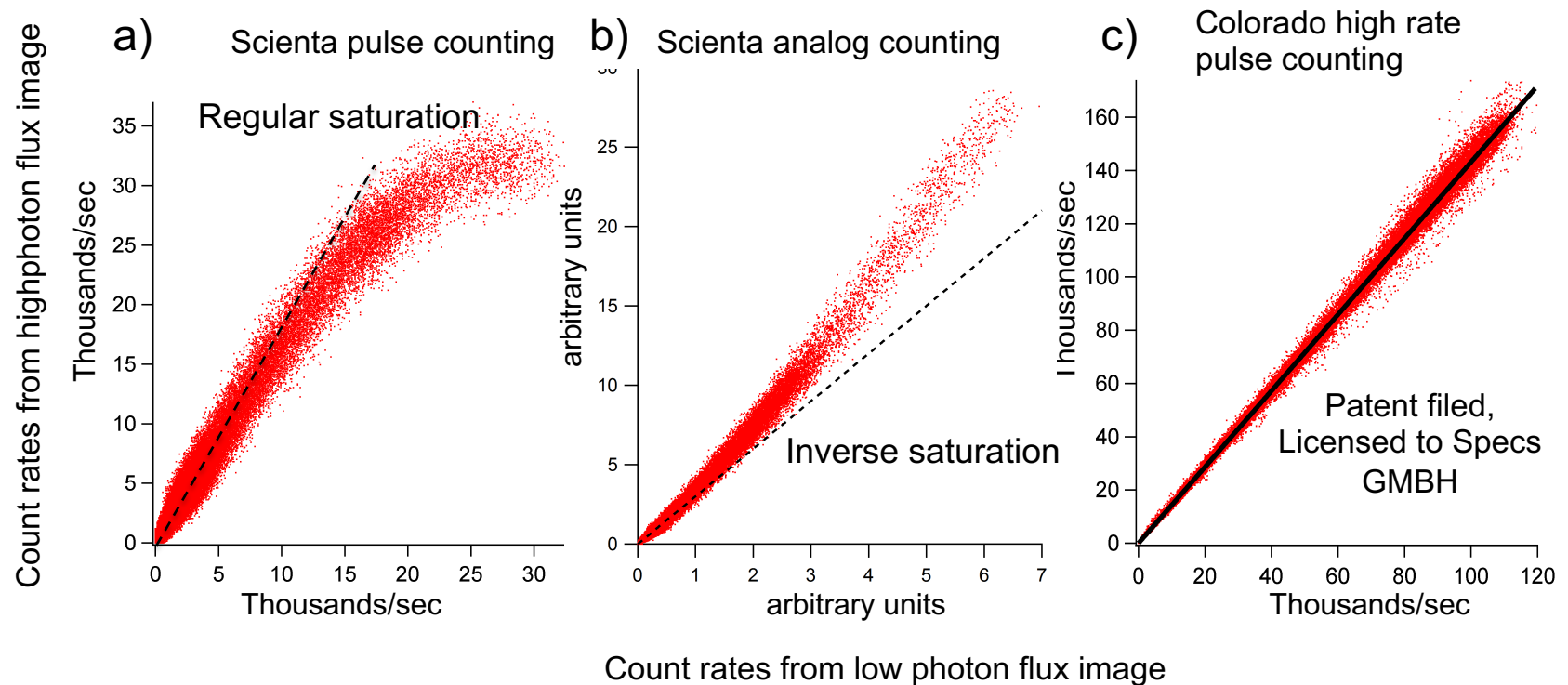
# Apparatus:



# Apparatus:



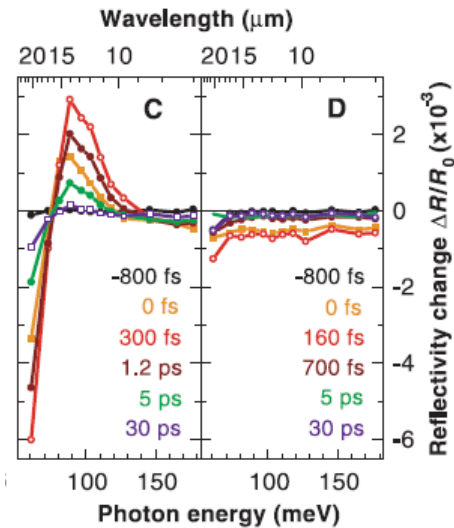
# Non-linearity of the counting in modern ARPES multichannel (camera-based) analyzers.



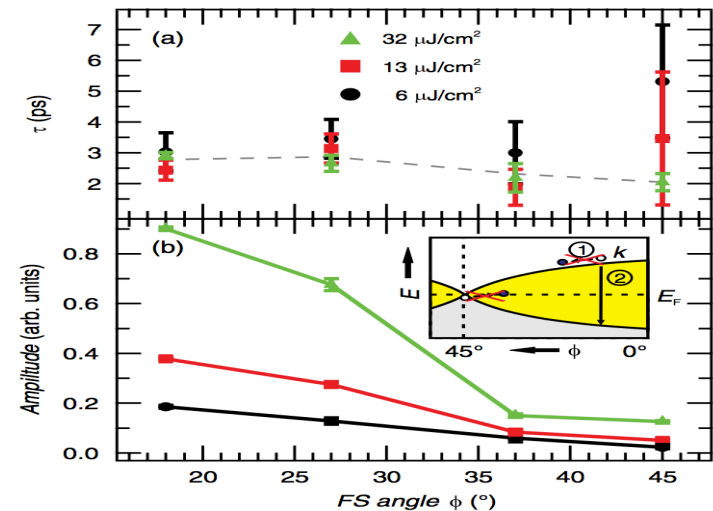
Most modern ARPES experiments are taken with CCD cameras/analog counting, which have an **inverse saturation problem**.

This nonlinearity **distorts** the spectral peaks, in general making them sharper and more beautiful.

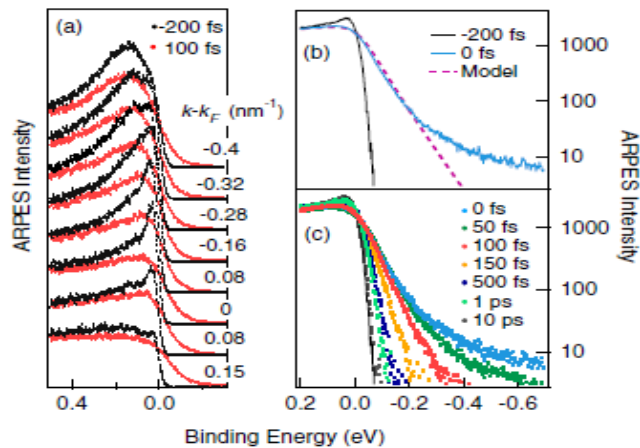
# Previous trARPES studies on cuprates – rich behavior, inconsistent interpretations.



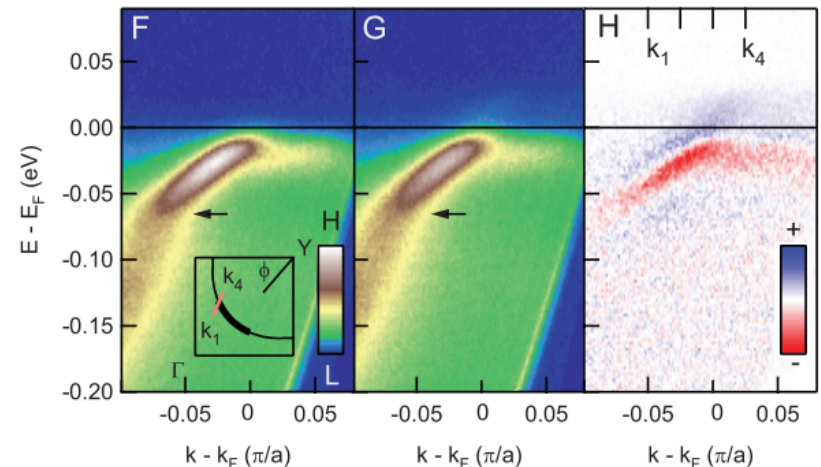
R. A. Kaindl *et al.*, *Science* **287**, 470 (2000)



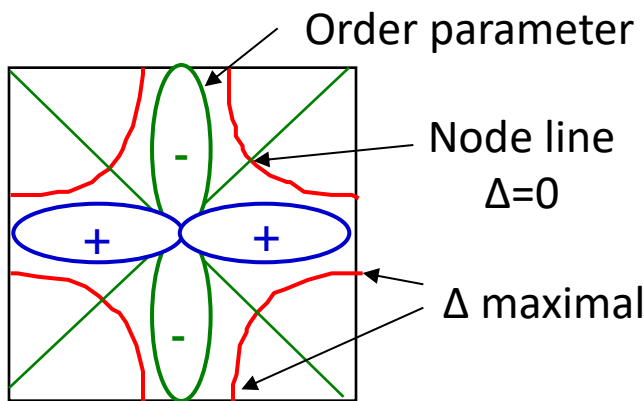
Cortes, Bovensiepen, *et al.* PRL 107, 097002 (2011)



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

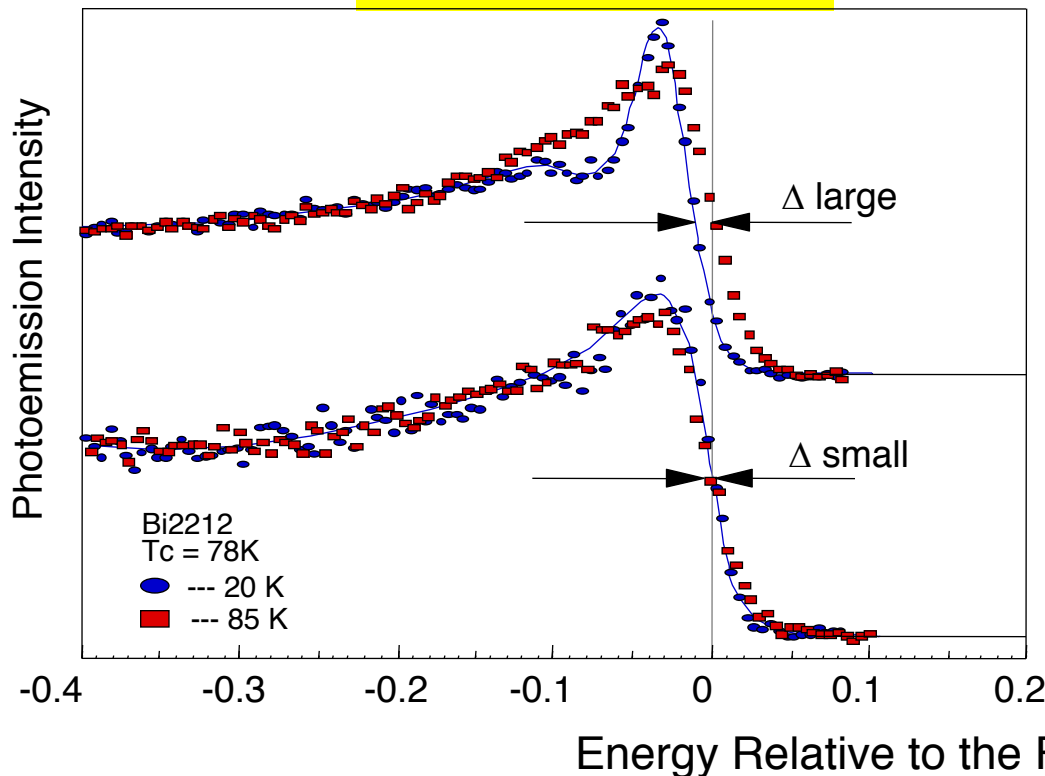


Smallwood, Lanzara, *et al.* *Science* **336**, 1137 (2012)



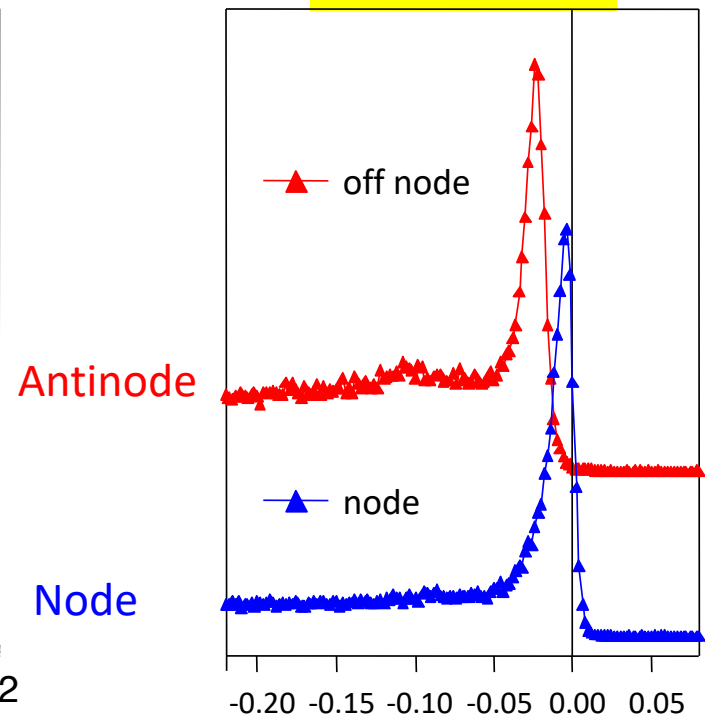
k-space dependence of superconducting energy gap  $\Delta$   
 $\rightarrow$  Symmetry of Cooper pair wavefunction is d-wave ( $\ell=2$ )

Standard (old) ARPES



Z.-X. Shen, D.S.D. et al.,  
 Phys. Rev. Lett. **70**, 1553 (1993)

Laser ARPES

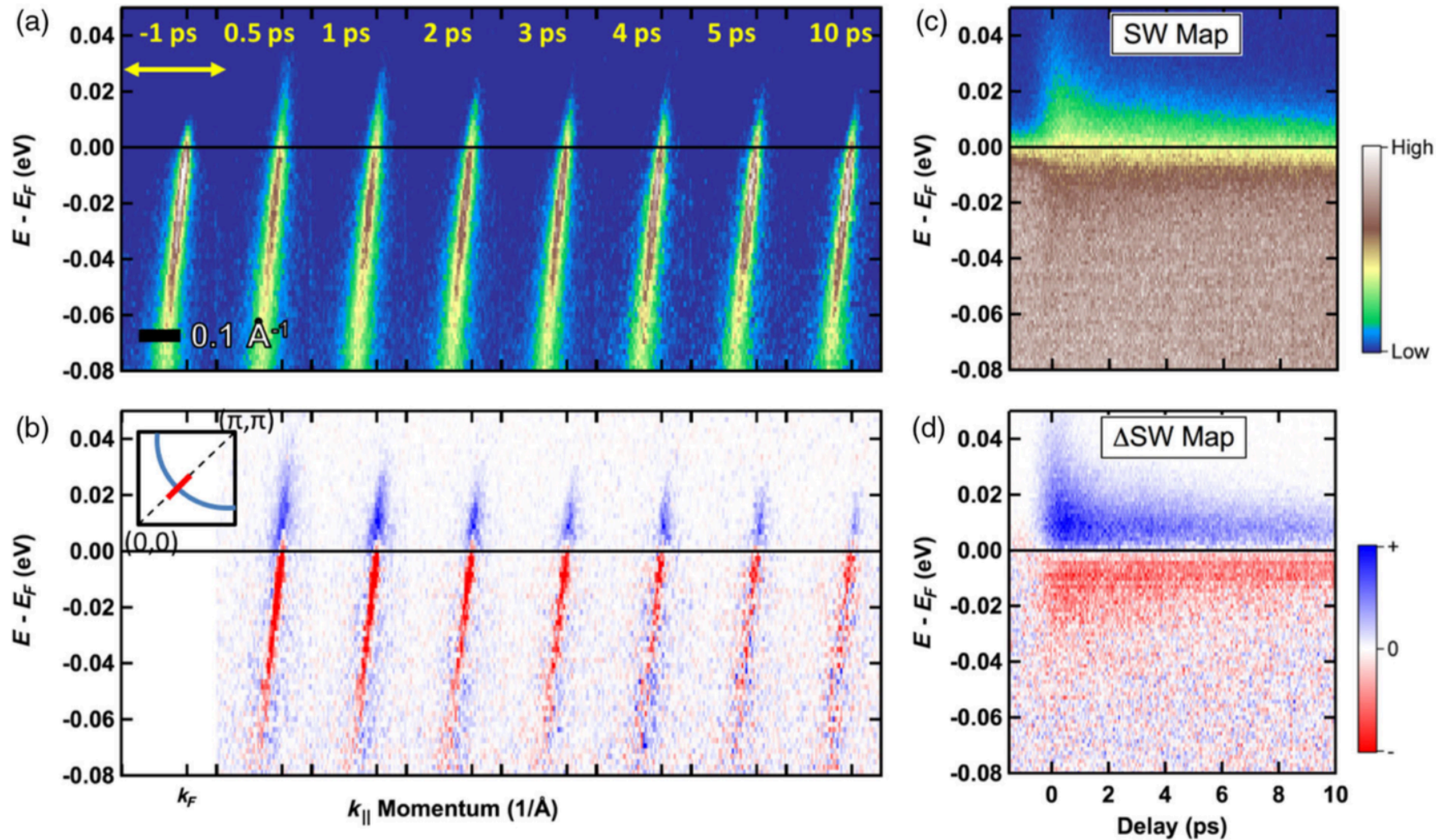


J.D. Koralek, D.S.D. et al.,  
 Phys. Rev. Lett. **96**, 017005 (2006)

# Direct measurement of electronic pseudotemperature (nodal states)

OP Bi-2212 ( $T_c=91\text{K}$ )

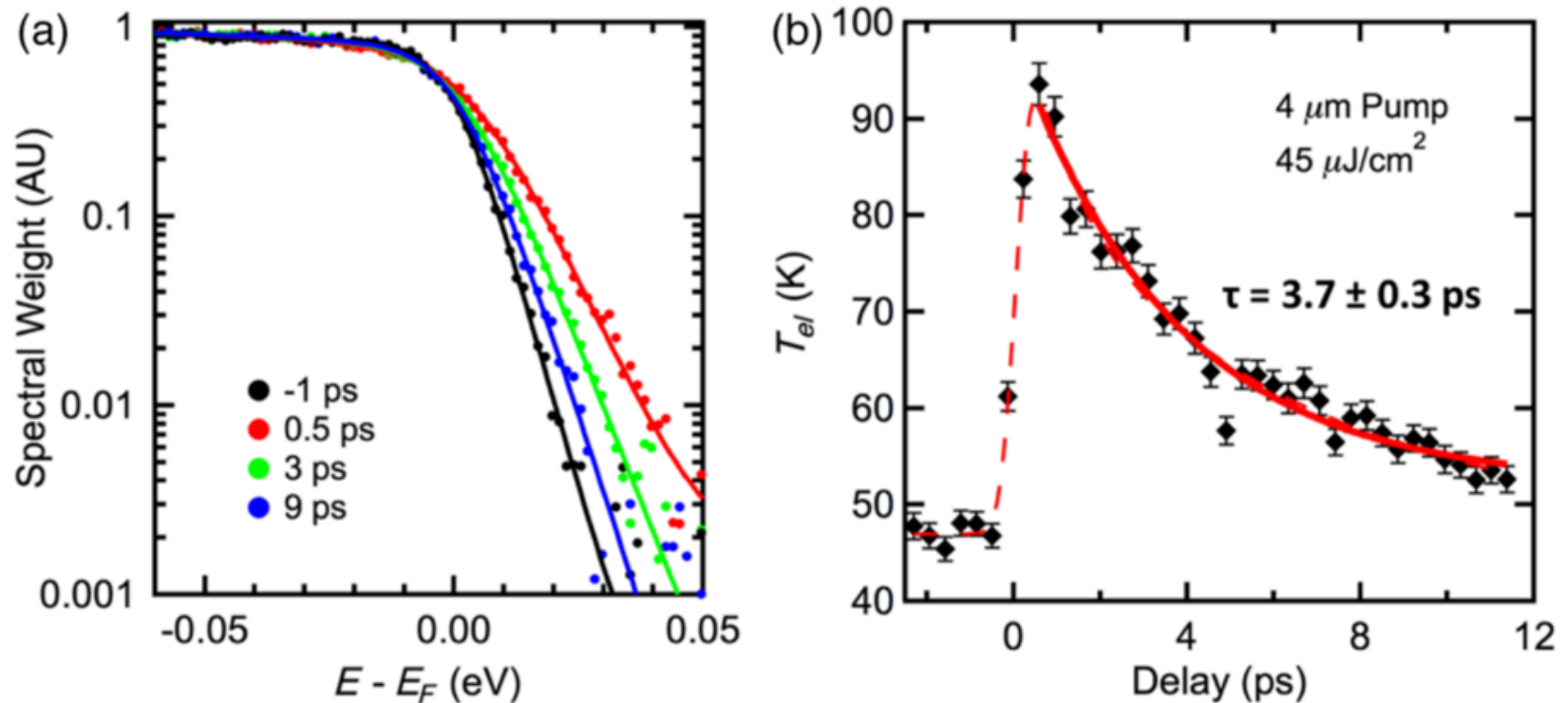
Nodal.  $T = 20\text{K}$



SW = Spectral Weight = an integration of the spectra in  $k$  along the red cut. These can be well-fit to a Fermi function.

## Nodal spectrum at $E_F$ plotted in log scale.

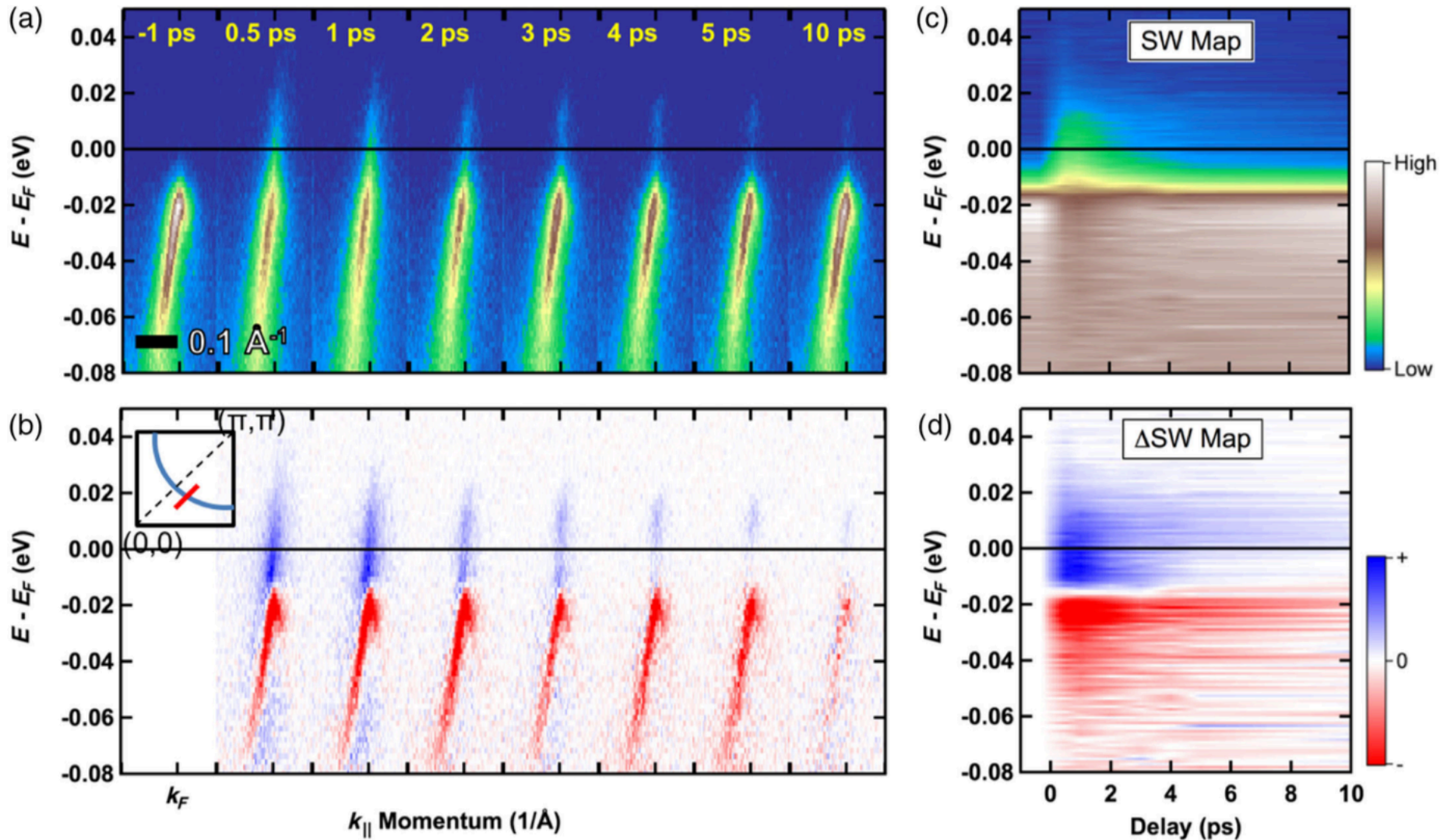
Large dynamic range allows accurate determination of deviations from a Fermi function, which would show up as a change in linearity



(weak pumping,  $T_{\text{max}} < T_c$ )

Roughly follows an exponential decay

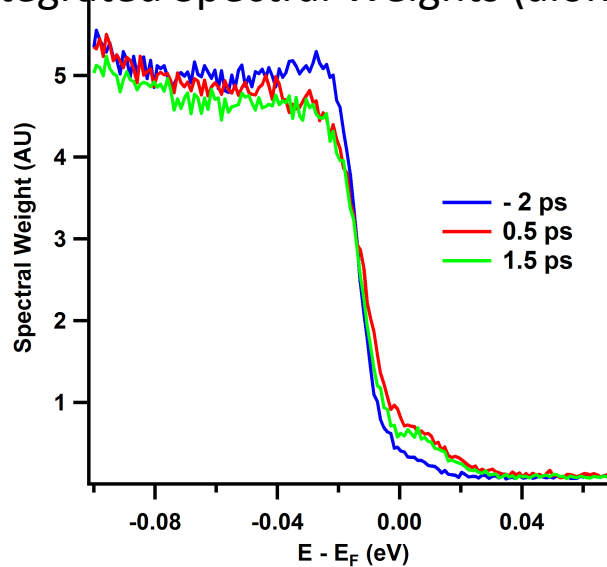
# Off-Nodal Dynamics, with SC gaps



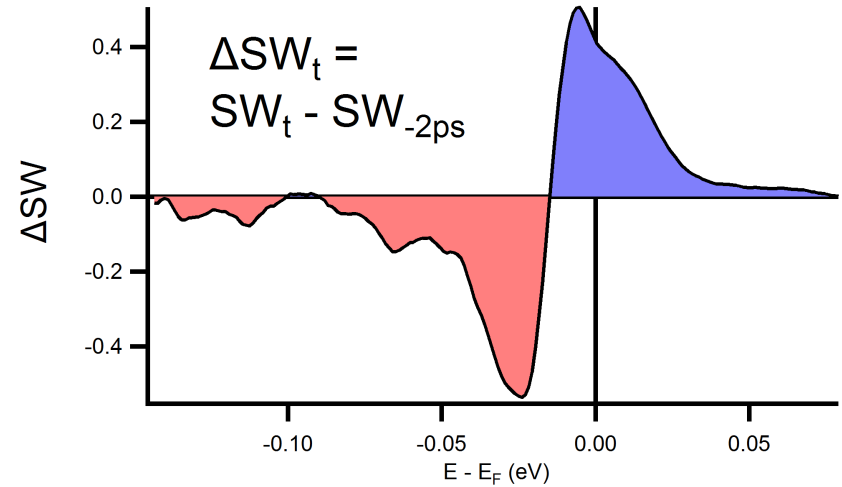
Optimally Doped BSCCO-2212  
( $T_c = 90\text{K}$ )  $T = 20\text{K}$

# Off-Nodal Dynamics, with SC gaps

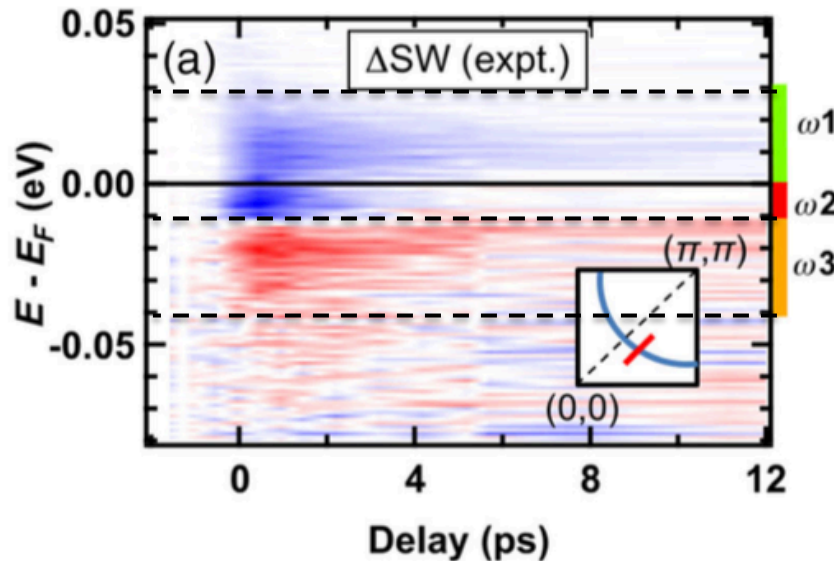
k-integrated Spectral Weights (along one cut)



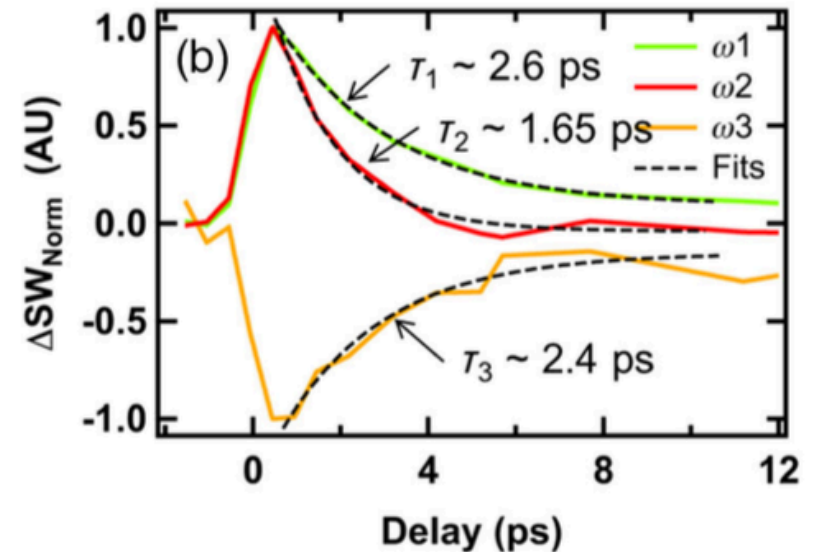
Difference in SW from starting time



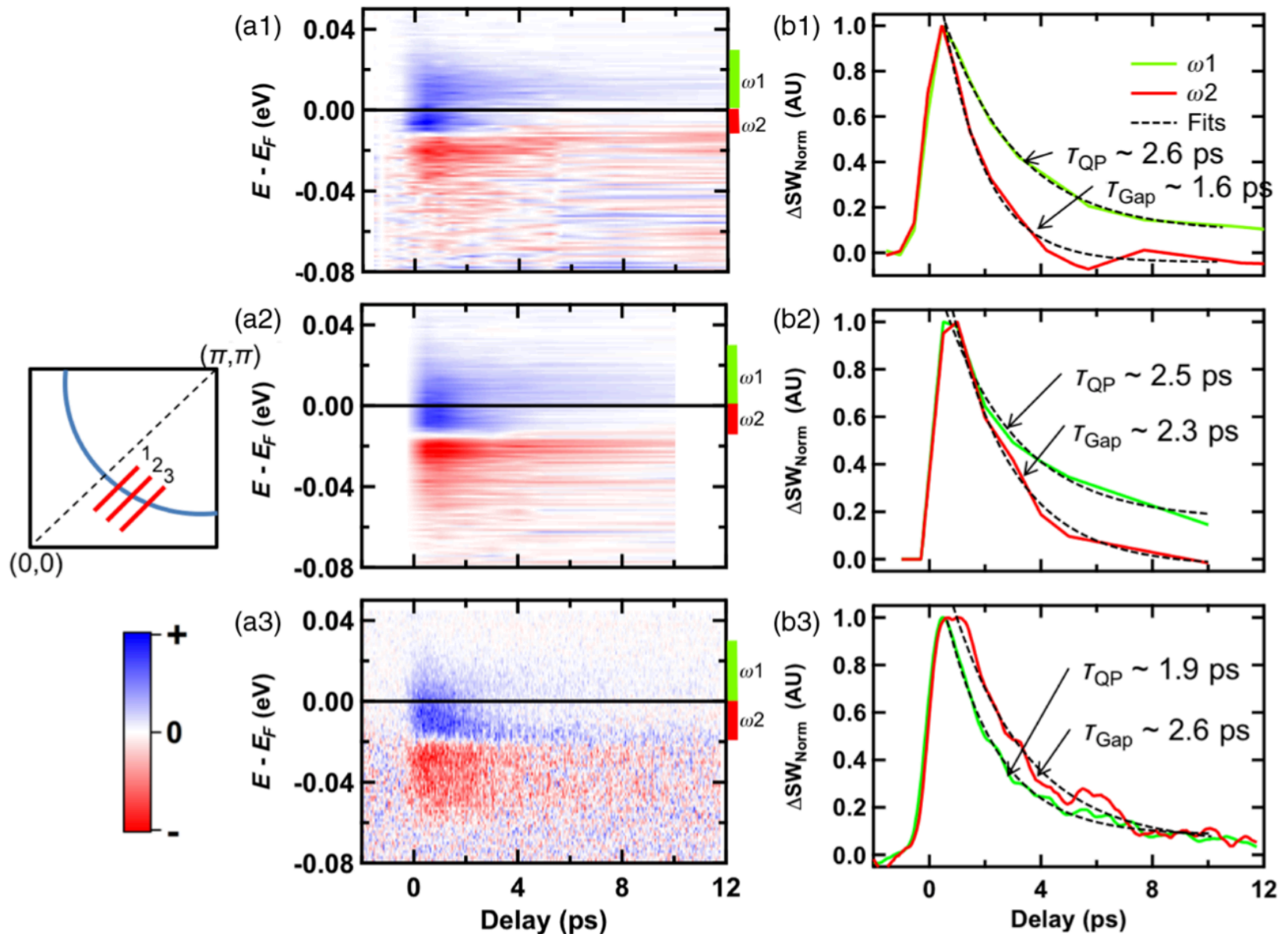
Three regions in E have different decay rates



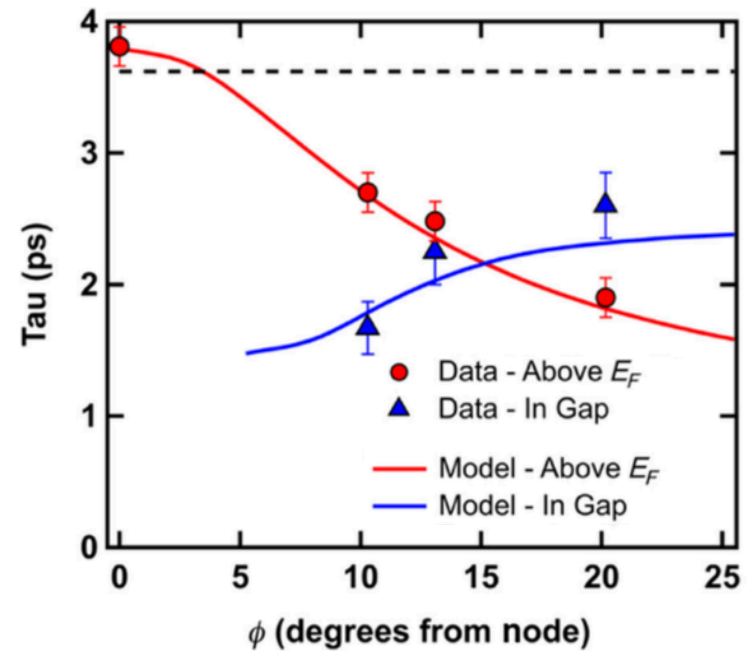
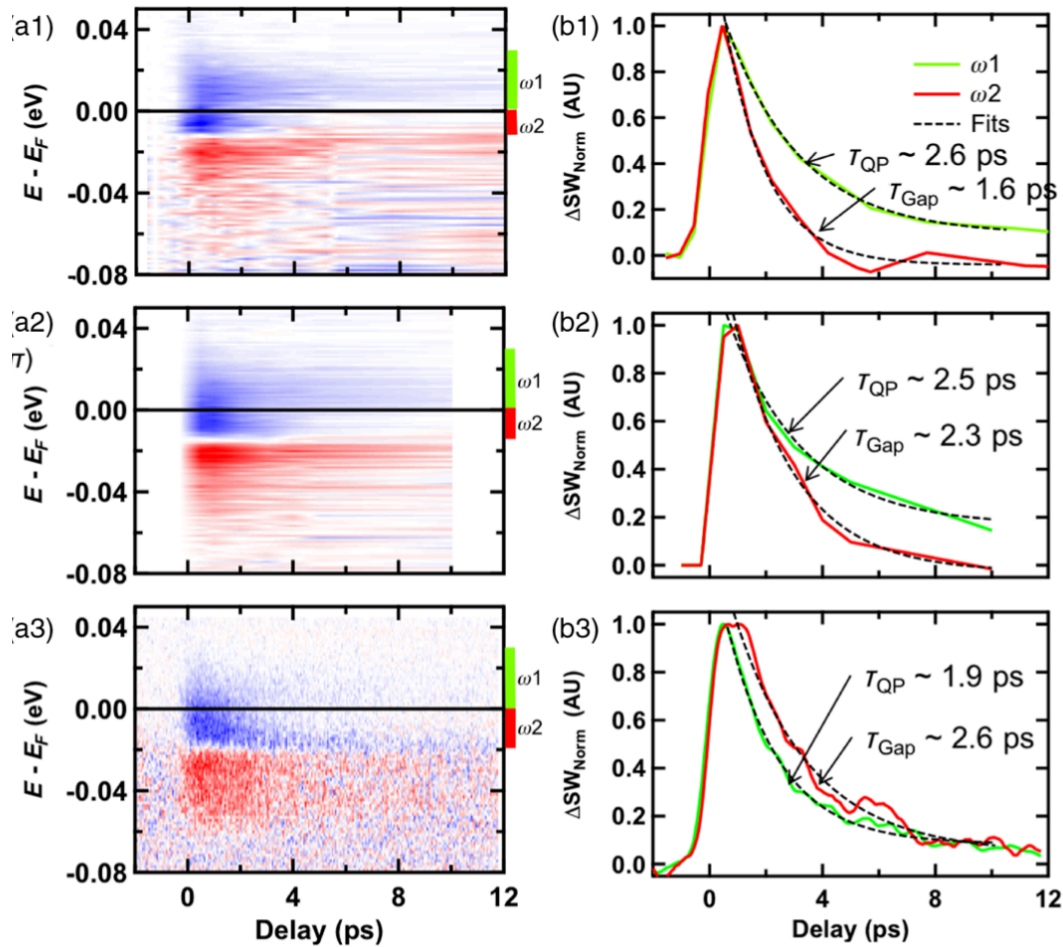
Approximate by single exponential decay



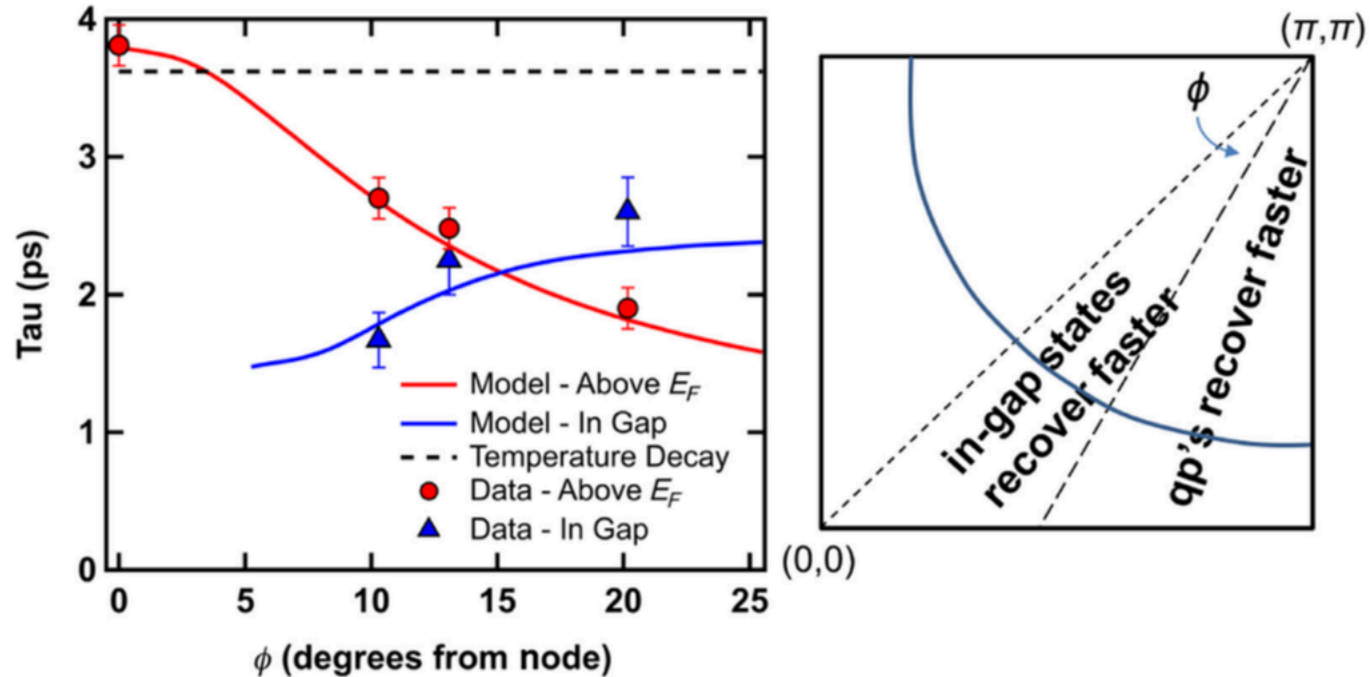
# Off-Nodal Dynamics with SC gaps, for different k-cuts



# Off-Nodal Dynamics with SC gaps, for different k-cuts



# Summary of SC state decay rate trends vs. $k$

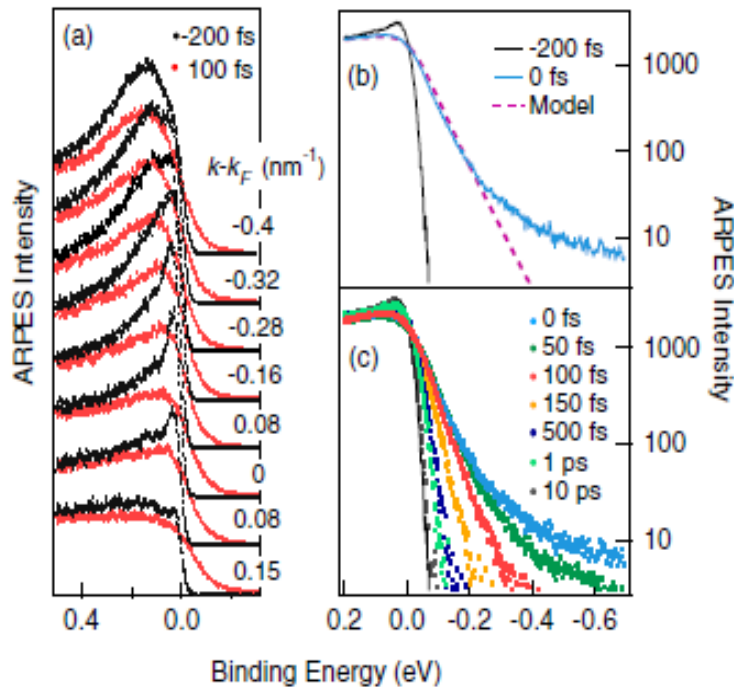


The observed behavior is very rich in  $k$ ,  $T$ , etc.

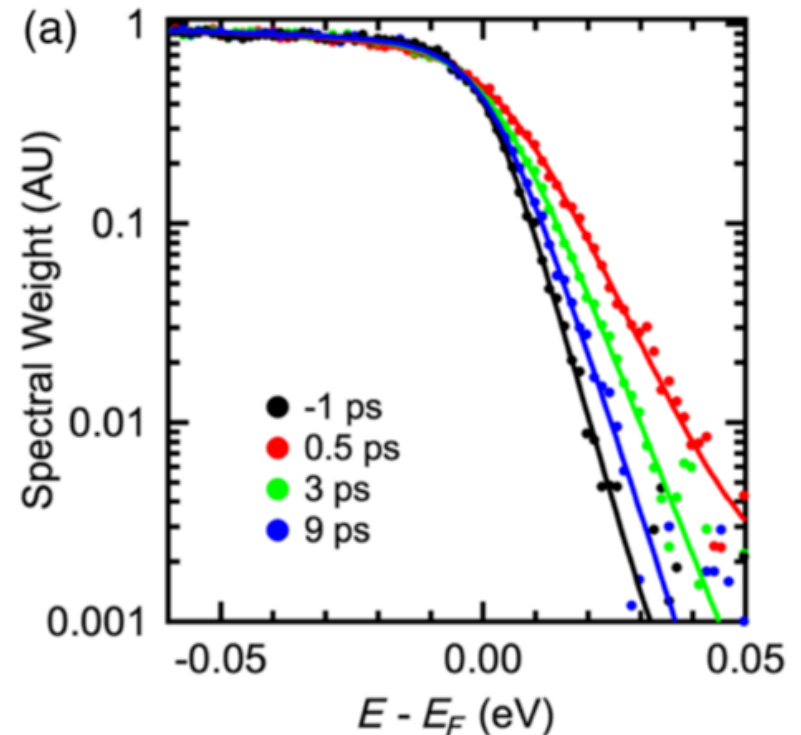
Our model (lines) captures all aspects very well.

Our model is extremely simple – essentially zero parameters!

# Thermalized Electrons



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



- Electron population thermalized within  $t_{\text{therm}} \sim 150\text{fs}$
- Appeal to equilibrium measurements vs. temperature to understand dynamics after this time period. Remember, we directly measure the electronic pseudotemperature vs. time from our nodal studies.

# Fitting to Dynes's Formula (1978)

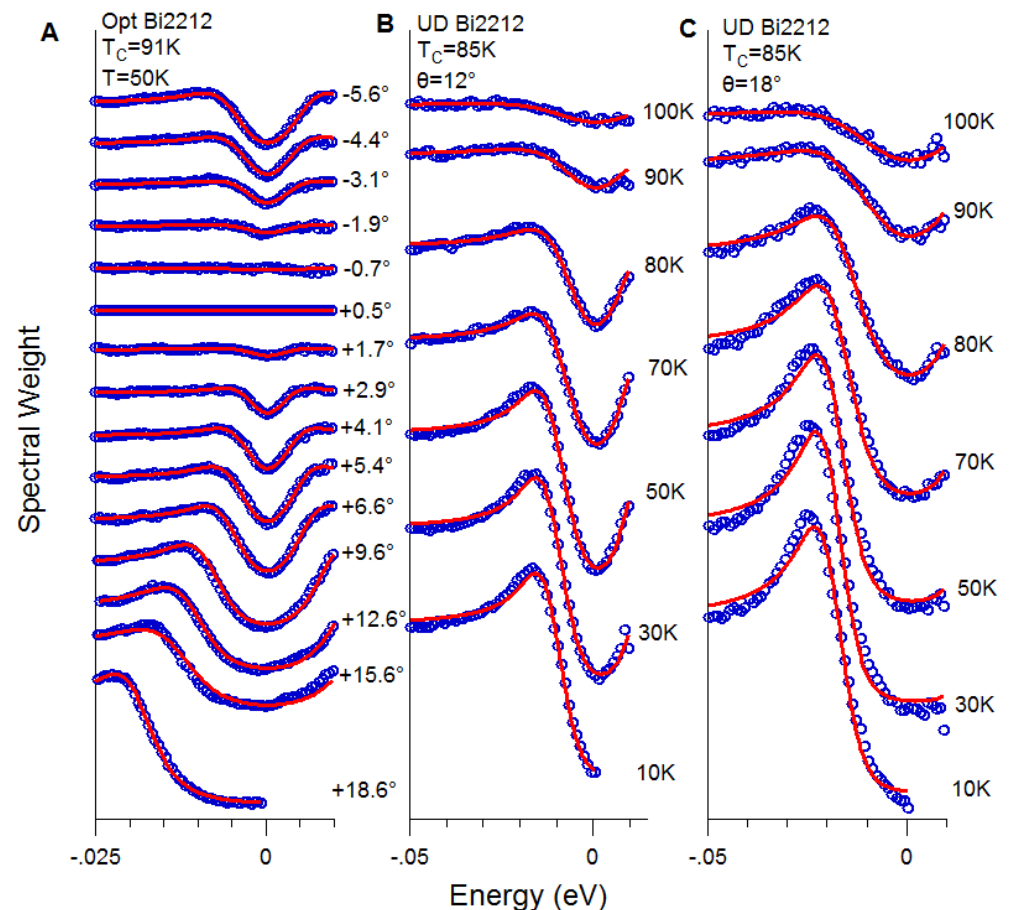
$$I_{TDoS}(\omega) = \rho_{Dynes}(\omega) = \text{Re} \frac{\omega + i\Gamma}{(\omega + i\Gamma)^2 - \Delta^2}$$

Represents a broadened BCS DOS.  
Originally empirical, later derived  
from Eliashberg theory.

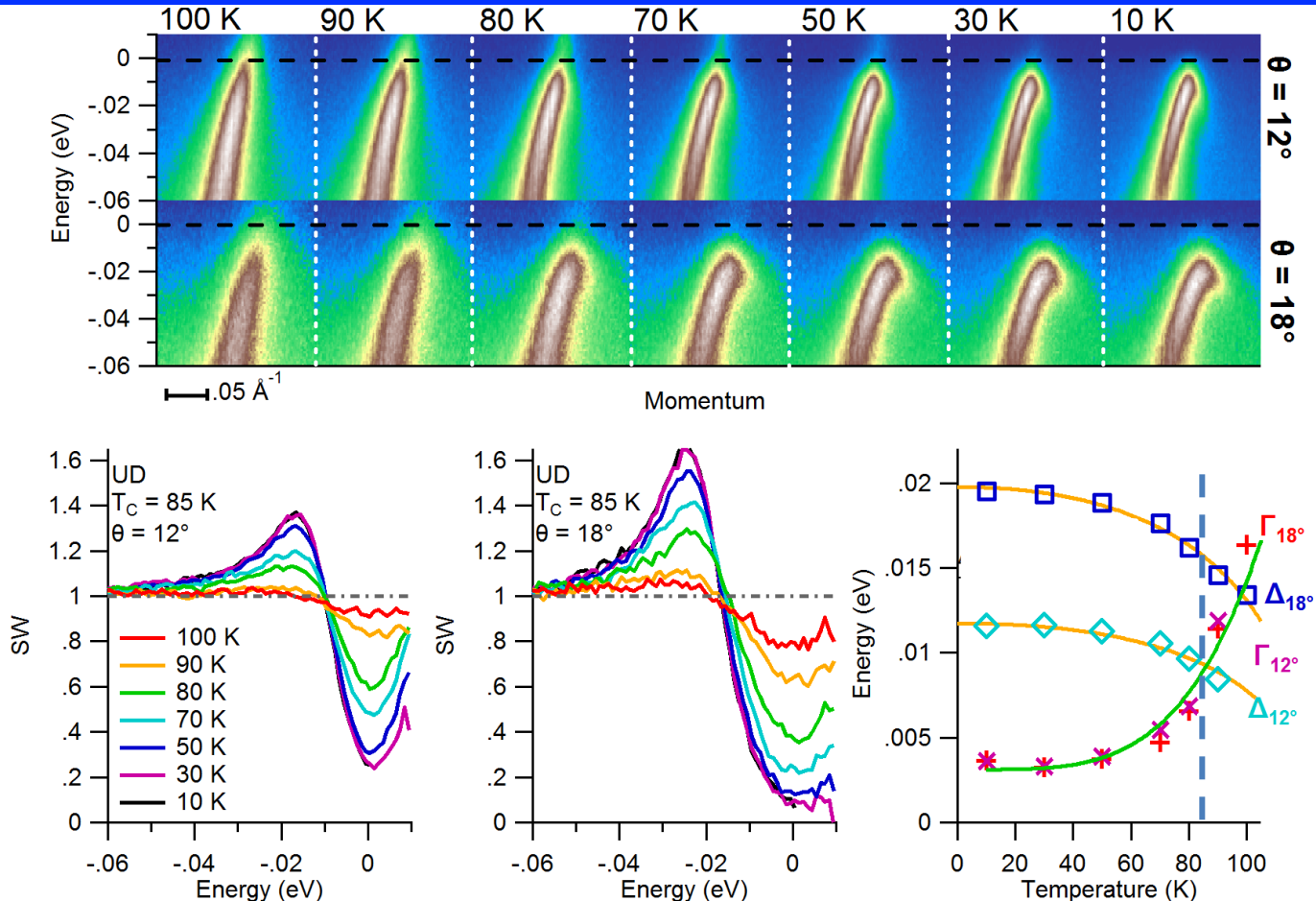
The TDoS are well fit by the  
Dynes formula over the full  
range of accessible angles and  
temperatures.

Each fit gives: pairing strength  
 $\Delta$ , and pair-breaking rate  $\Gamma$ .

Similar fits of actual tunneling in  
cuprates are not nearly as  
successful because of the d-  
wave nature of  $\Delta$ , the van-Hove  
in the DOS, etc.



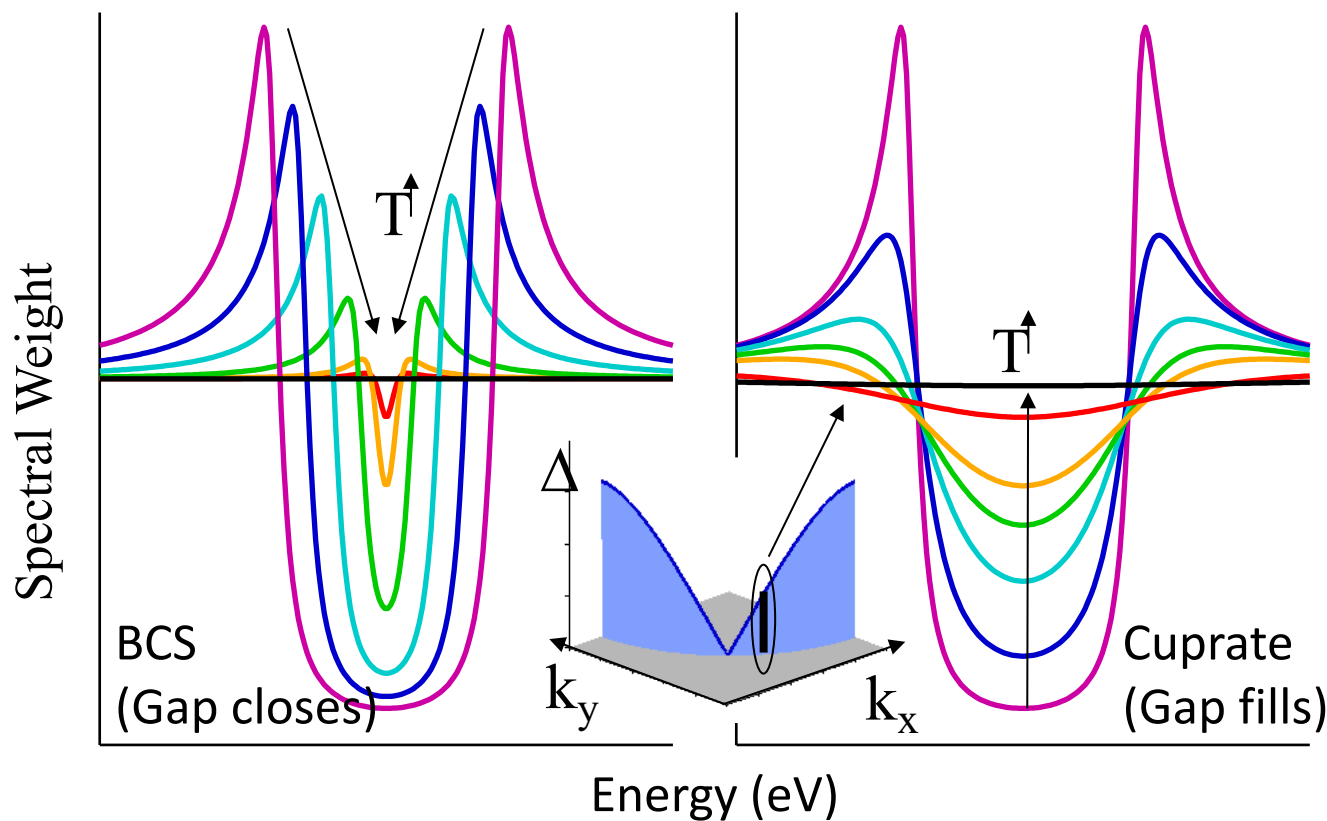
# Equilibrium measurements off-node



With increasing  $T$ , the key effect is a gap filling (due to self-energy term  $\Gamma$ ), which dominates over the gap term  $\Delta$ .

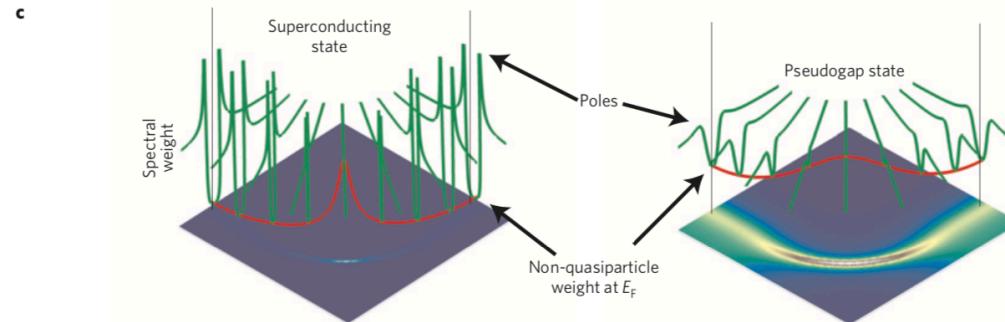
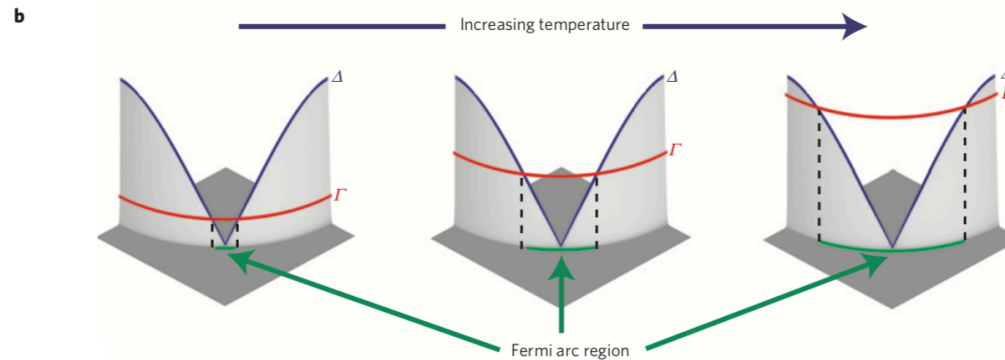
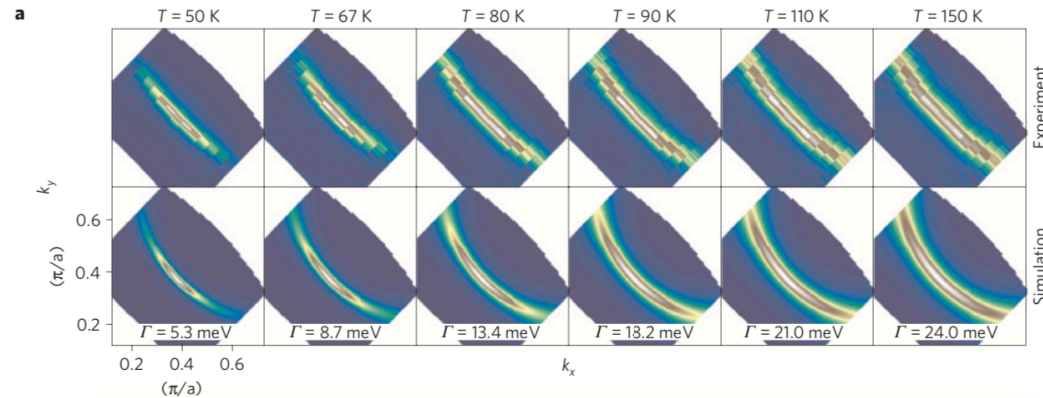
T. J. Reber, D.S.D. et al. Nature Physics **8**, 606–610 (2012);  
Phys. Rev. B **87**, 060506(R), (2013)

# Closing/Filling of the gaps with temperature

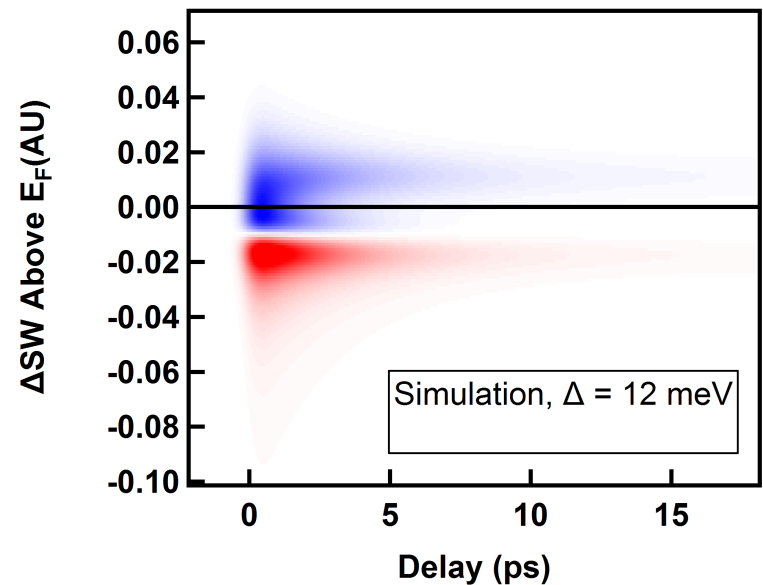
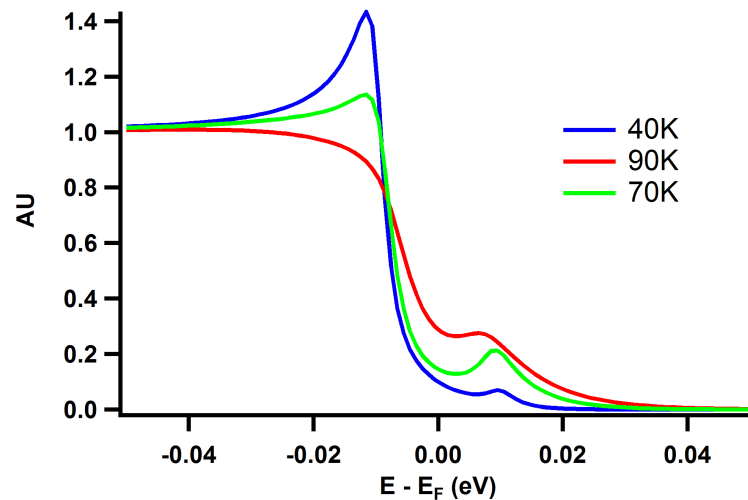
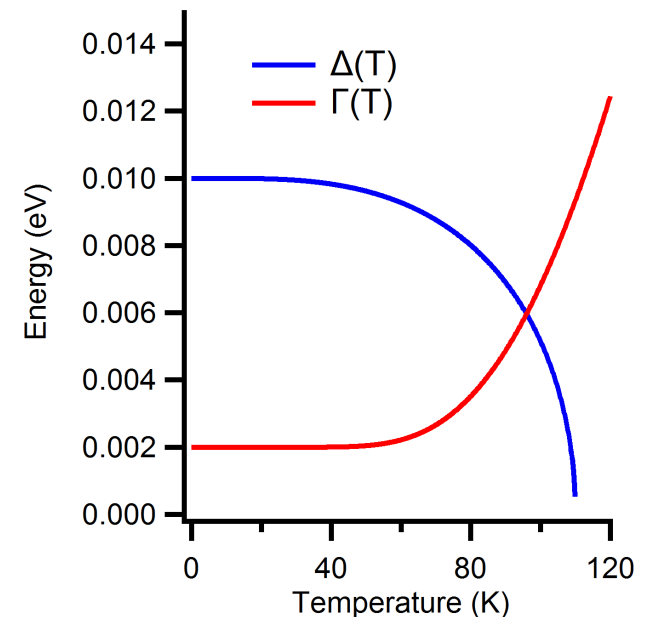
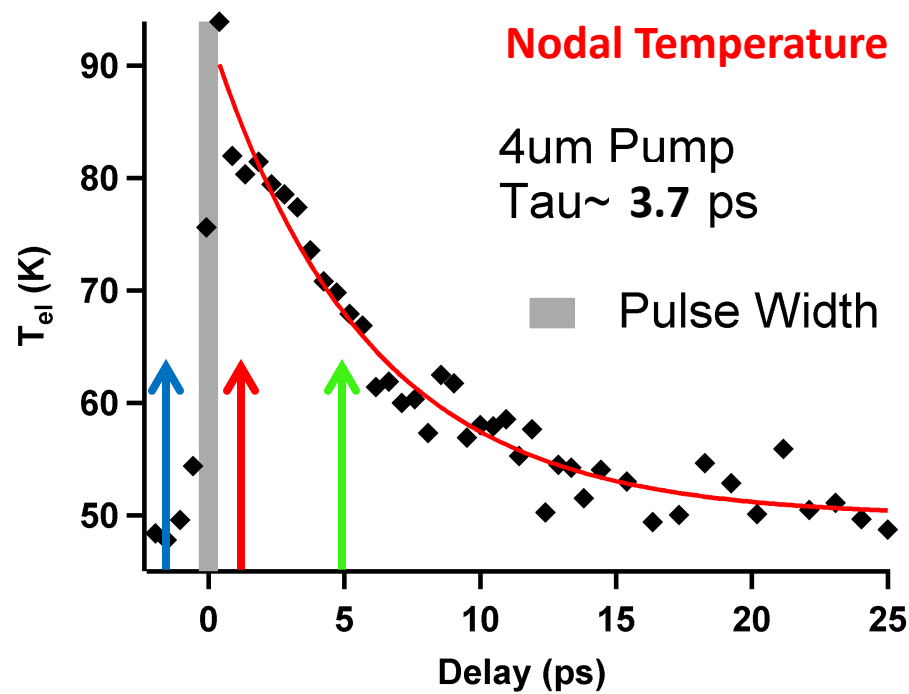


The filling of the gap in cuprates is due to the rapidly rising  $\Gamma$  (scattering rate) with temperature. This is a phenomenology observed in essentially all spectroscopies on cuprates, but has been difficult to quantify.

# “Fermi Arcs” growing with $T$ from competition between $\Delta$ and $\Gamma$ .



# Ultrafast Simulation

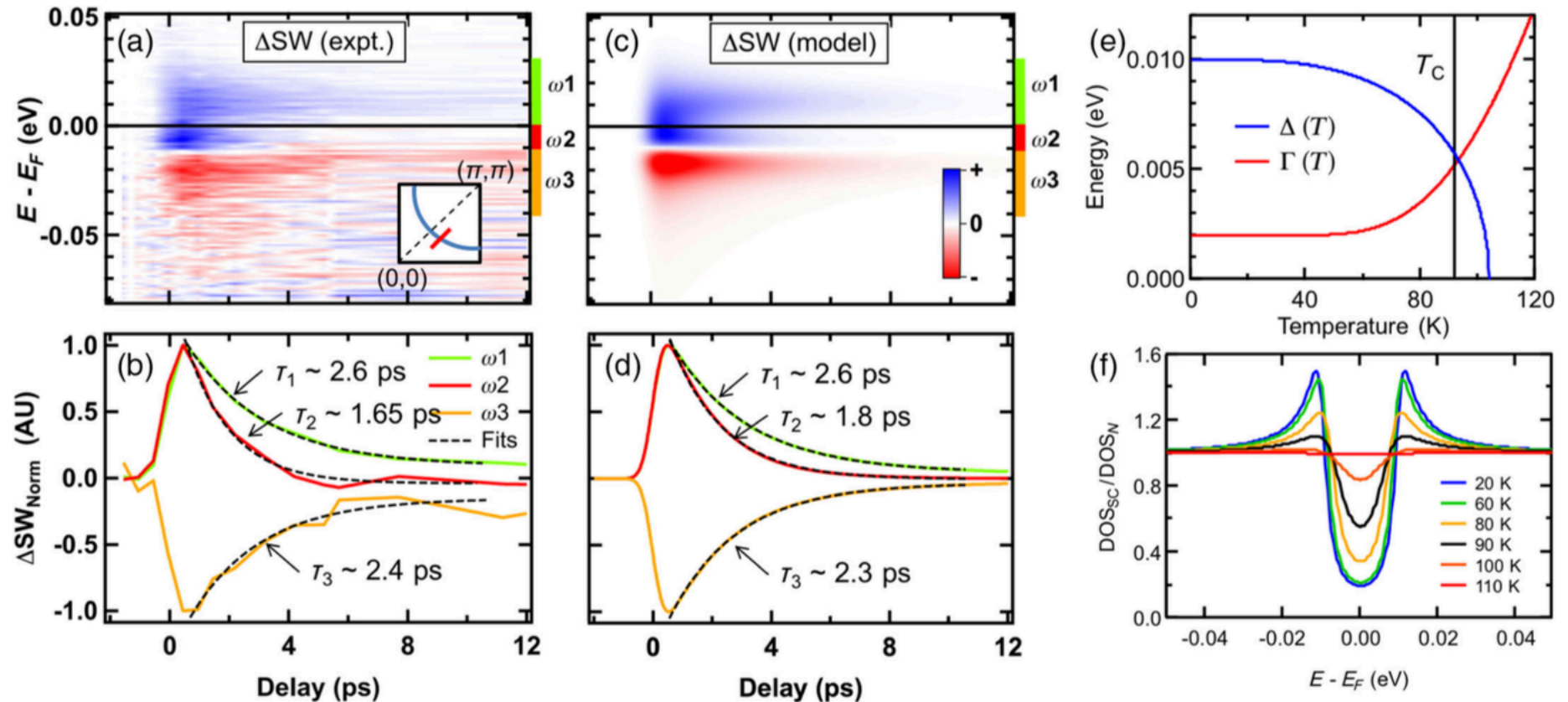


# Experiment vs. static-inspired model

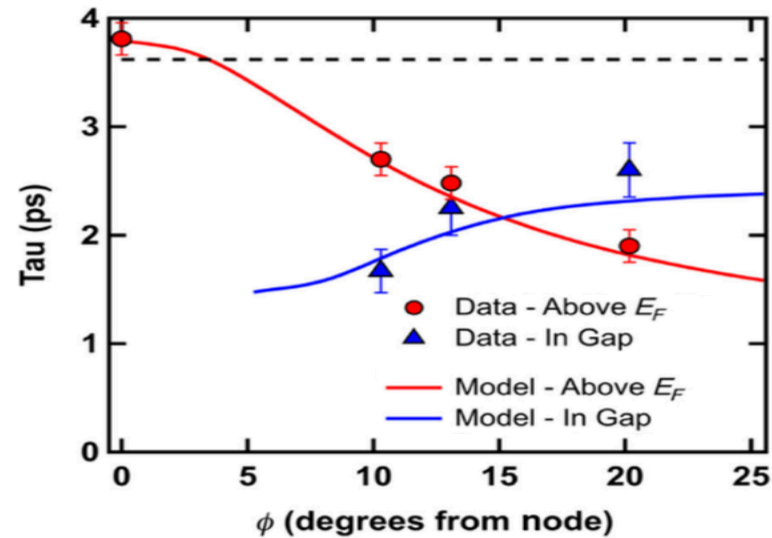
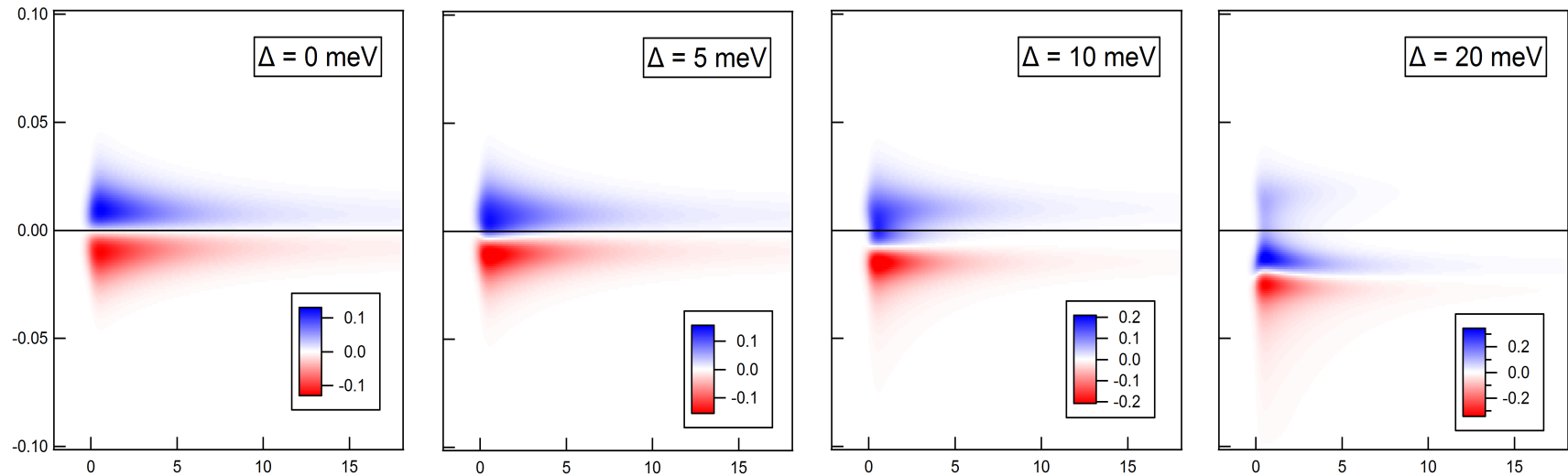
Experiment

Model

Input from Equilibrium



# Gap size dependence in the model ( $\rightarrow$ k-dependence in the experiment)



# Implications for ultrafast decay dynamics of a purely electronic system for $t > t_{\text{therm}}$

Need:

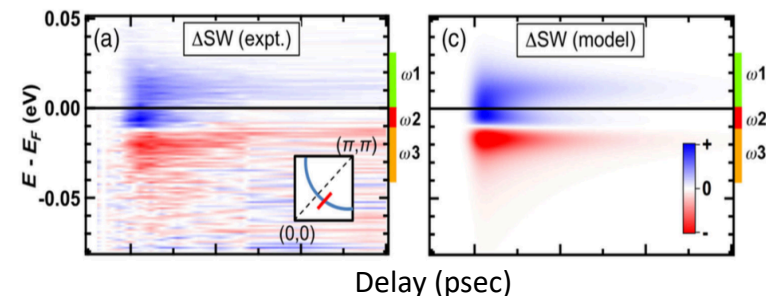
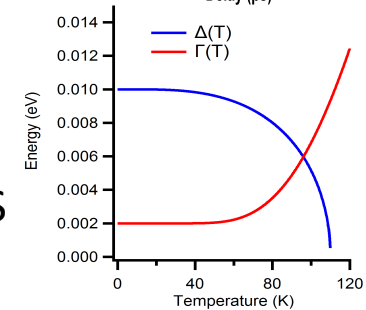
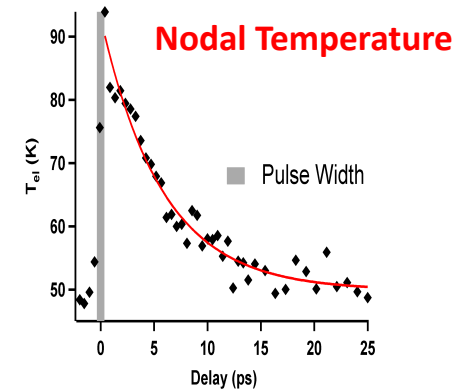
a) Electronic pseudotemperature vs. time.

- ARPES on cuprates – directly measurable from the Fermi edge along the nodal direction.
- Dominated by diffusion of heat away from the pump spot.

b) Key electronic parameters vs. equilibrium temperature

Cuprates –  $\Delta(k,T)$  and  $\Gamma(k,T)$  from equilibrium ARPES

c) Replace equilibrium temperature with electronic pseudo temperature  $\rightarrow$  All key parameters understood vs. time.

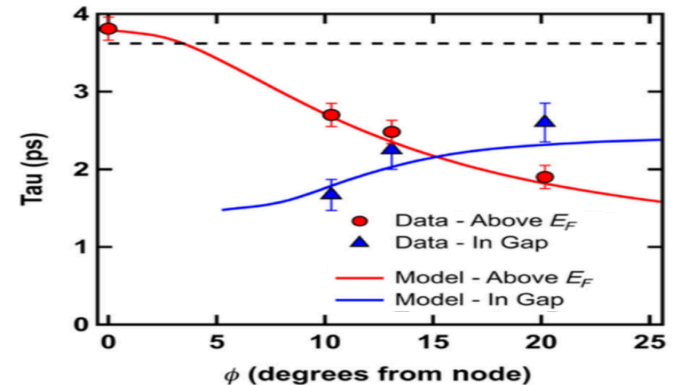


No direct information on phase stiffness.

# Implications

## Decay dynamics of an electronic system for $t > t_{\text{therm}}$

Replace equilibrium temperature with **electronic pseudo temperature** → key parameters understood vs. time.



- We expect the power of this concept to extend to many other materials and ultrafast measurements, even if the electronic pseudotemperature can not be directly accessed, as it can in the present case.
- Situation is different (richer) when system can be periodically/resonantly driven.

## Nambu-Gorkov and Self-energies

$$G^{-1} = \begin{pmatrix} \omega + i\Gamma + \varepsilon_k & \Delta \\ \Delta^* & \omega + i\Gamma - \varepsilon_k \end{pmatrix}$$

$$G_{11} = \frac{\omega + i\Gamma + \varepsilon_k}{(\omega + i\Gamma)^2 - \varepsilon_k^2 - \Delta^2}$$

$$G_{11}^{-1} = \omega + \Sigma - \varepsilon_k = \frac{(\omega + i\Gamma)^2 - \varepsilon_k^2 - \Delta^2}{\omega + i\Gamma + \varepsilon_k} = (\omega + i\Gamma) - \varepsilon_k - \frac{\Delta^2}{\omega + i\Gamma + \varepsilon_k}$$

$$\therefore \Sigma = i\Gamma - \frac{\Delta^2}{\omega + i\Gamma + \varepsilon_k}$$

*Which Norman and Randeria change to*

$$\Sigma = i\Gamma_{Single} - \frac{\Delta^2}{\omega + i\Gamma_{Pair} + \varepsilon_k}$$

They treat the two  $\Gamma$ 's fully independently, allowing them to vary separately with  $T$ ,  $\omega$ , etc.

## ARPES as a self-energy spectroscopy.

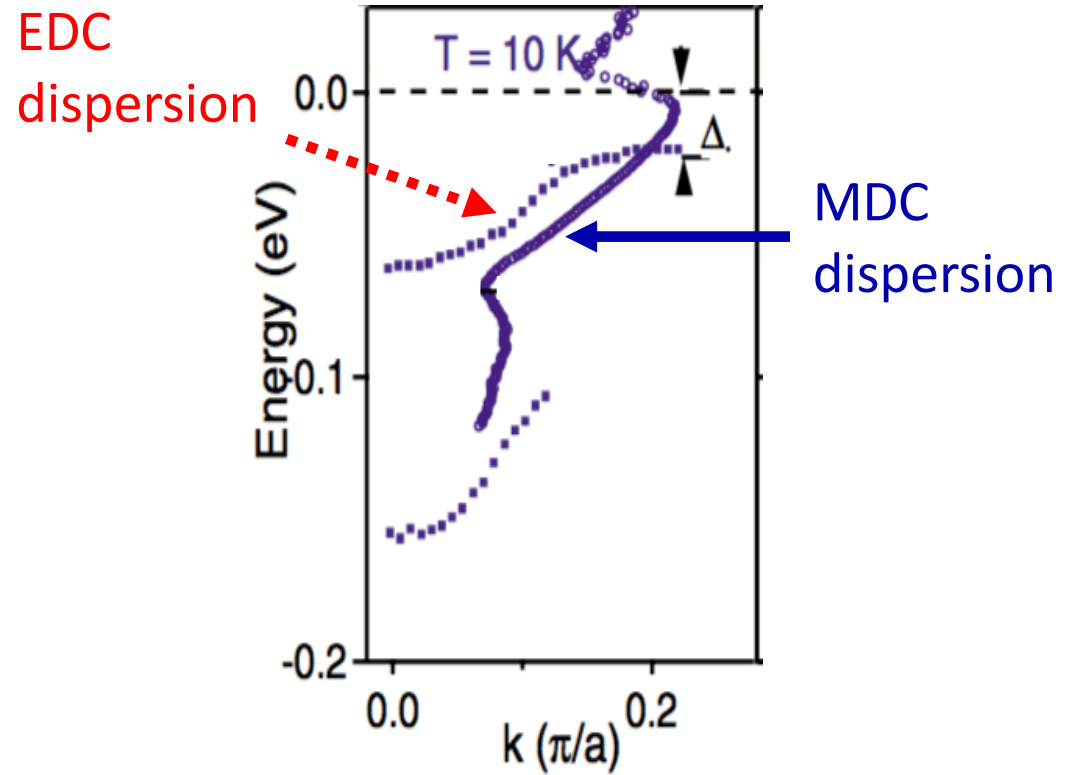
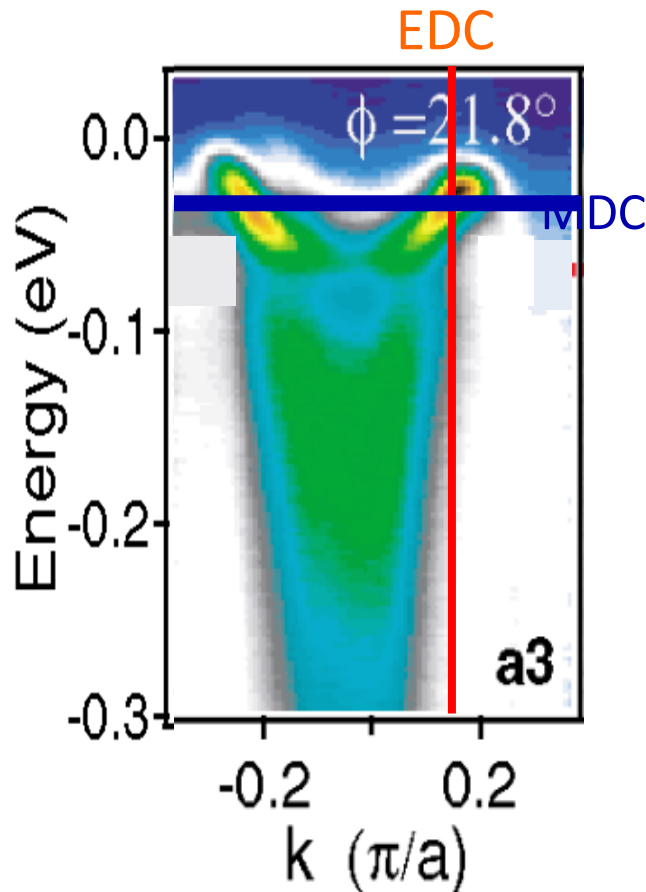
Using the “proper” or conventional Nambu-Gorkov Greens function.  
Non-conventional properties – contained in the novel behavior of  $\Sigma$ .

Fully 2-dimensional analysis (beyond 1D EDCs and MDCs)  
ARPES and self-energies in the presence of SC gaps, band curvature, etc.  
(ARPES throughout the zone).

# EDCs (Energy Distribution Curves) and MDCs (Momentum Distribution Curves).

Serious trouble for system with large broadening (large self energies) and band curvature.

No problem if spectral peaks are very sharp (self energy small).



A new step:  
True 2D fitting (EDCs and MDCs fit simultaneously).

Obtain:

- Bare-band dispersion  $\varepsilon(k)$
- Gaps  $\Delta(k, T)$
- Fully causal self energy  $\Sigma(k, \omega, T) = \Sigma'(k, \omega, T) + i\Sigma''(k, \omega, T)$   
All the details of the 1-particle electronic interactions.

Orders of magnitude fewer free parameters.

New level of accuracy  $\rightarrow$  new physics.

# Development of a 2D fitting method (EDCs and MDCs together).

Mid-zone cut, lightly underdoped T<sub>c</sub>=85K Pb-BSCCO sample.

## Nambu-Gorkov's Green's function

T=50K (UD85K)

$$G^{-1} = \begin{pmatrix} \omega + \varepsilon_k - \Sigma & \phi \\ \phi & \omega - \varepsilon_k - \Sigma \end{pmatrix}$$

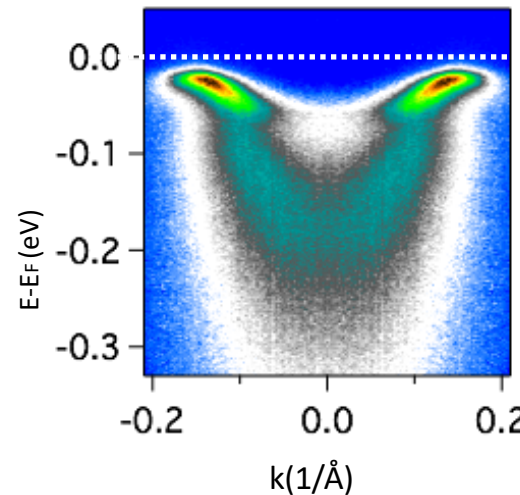
$$G_{11} = \frac{\omega + \varepsilon_k - \Sigma}{(\omega - \Sigma)^2 - \varepsilon_k^2 - \phi^2}$$

$$A(k, \omega) = -\frac{1}{\pi} \text{Im} G_{11}$$

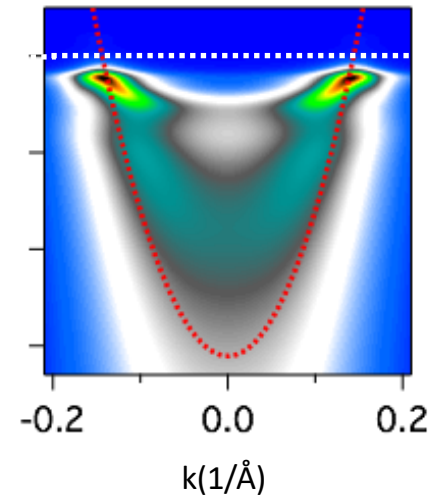
$$I(k, \omega) \propto \left( \left| \langle f | A \cdot p | i \rangle \right|^2 \times f(\omega) \times A(k, \omega) \right) \otimes R(k, \omega)$$

Note:  $\begin{cases} \phi = \Delta \cdot Z \\ Z = 1 - \frac{\Sigma'(\omega)}{\omega} \end{cases}$

ARPES Data



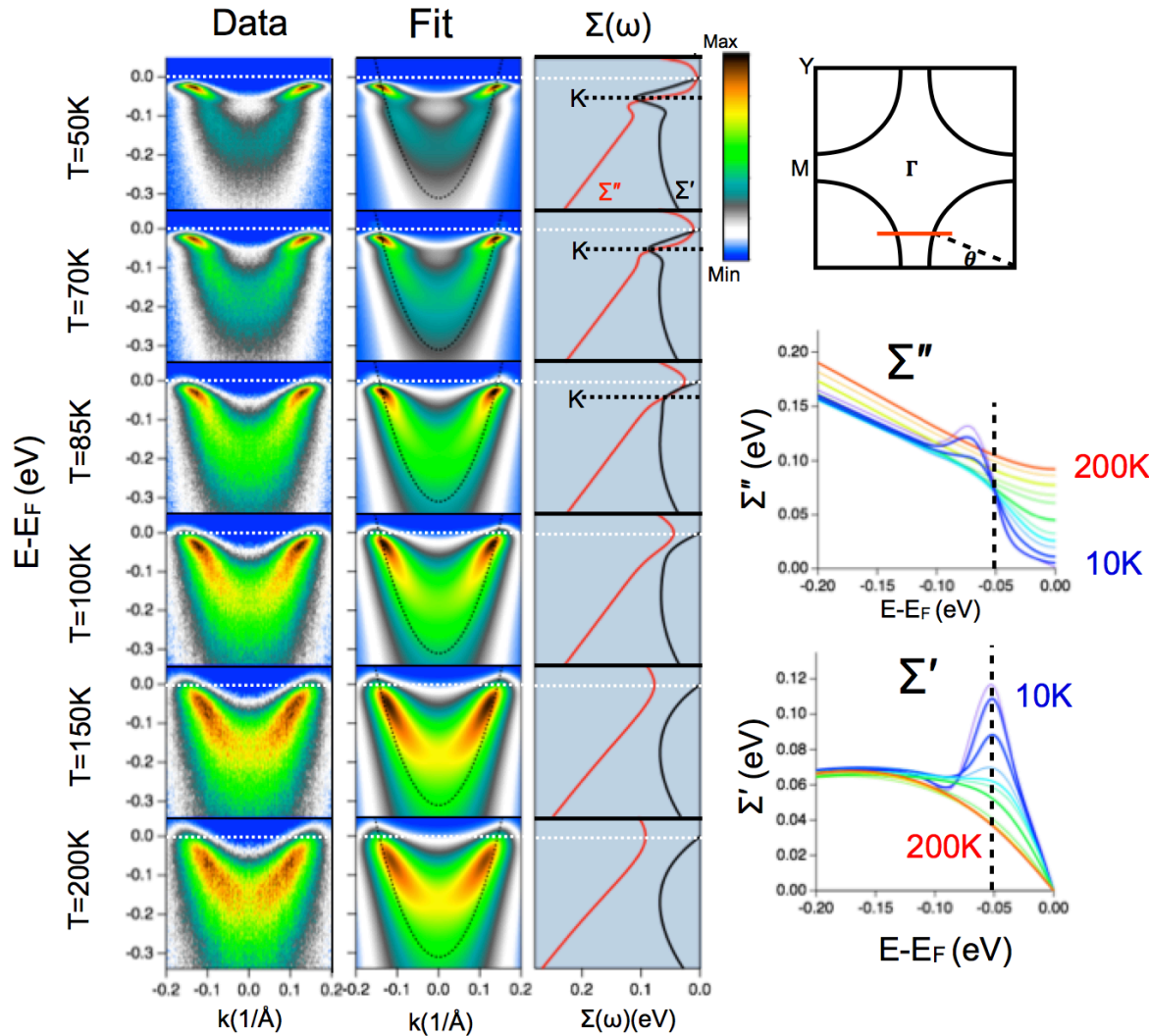
Fit



# Development of a 2D fitting method (EDCs and MDCs together).

Superstructure-free Pb-Bi2212  
T<sub>c</sub>=85K underdoped

“ARPES as a self-energy spectroscopy.”



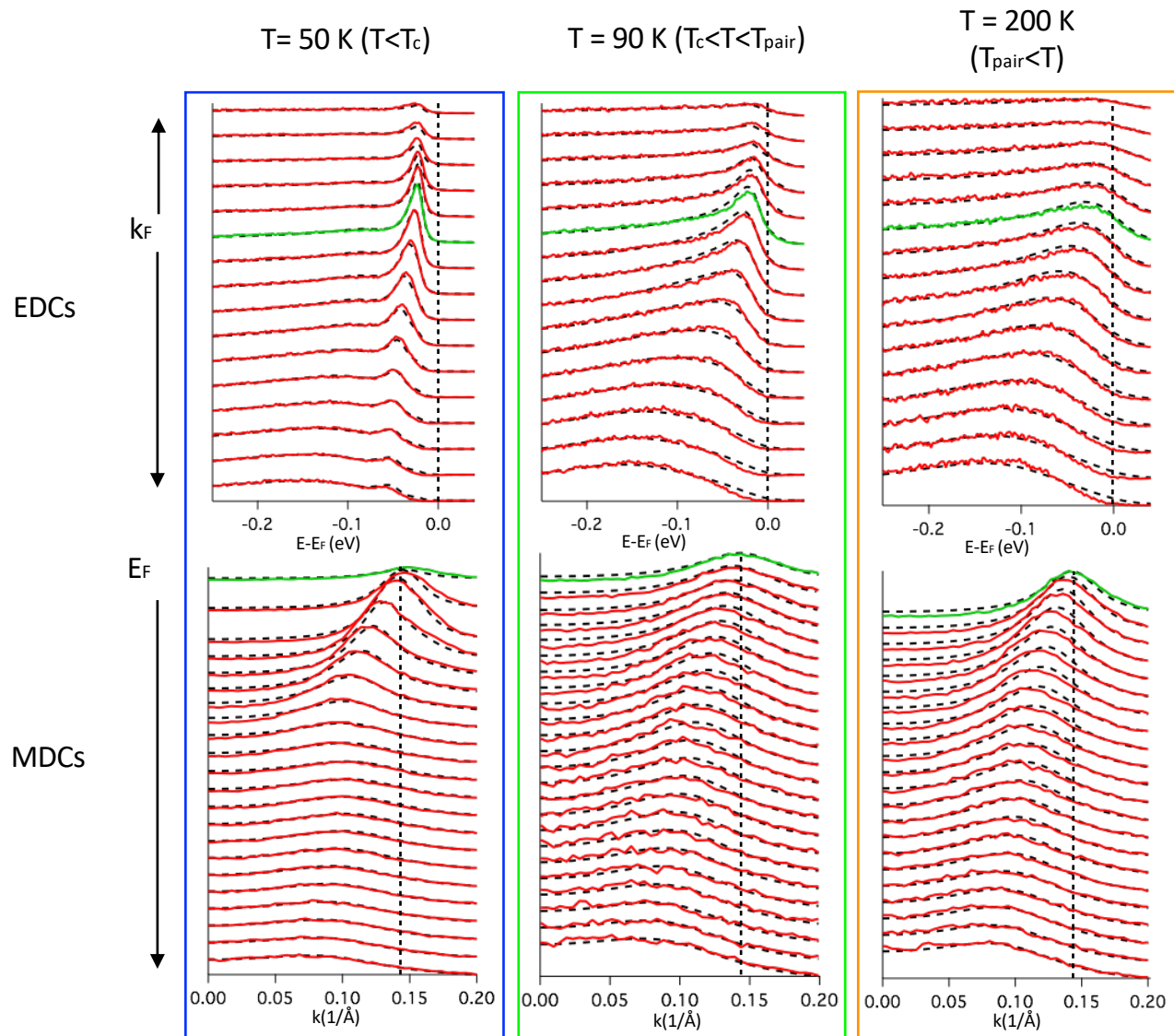
Fully causal treatment within Nambu-Gor'kov:

Simultaneous extraction of:

- SC gaps,  $\Delta(\mathbf{k}, T)$
- $\Sigma'(\mathbf{k}, \omega, T)$  and  $\Sigma''(\mathbf{k}, \omega, T)$
- Renormalization  $Z(\mathbf{k}, \omega, T)$
- Unrenormalized “bare” bands,  $v_F(\mathbf{k}, T)$ , etc.

Orders of mag. fewer free parameters compared to EDC or MDC fitting.

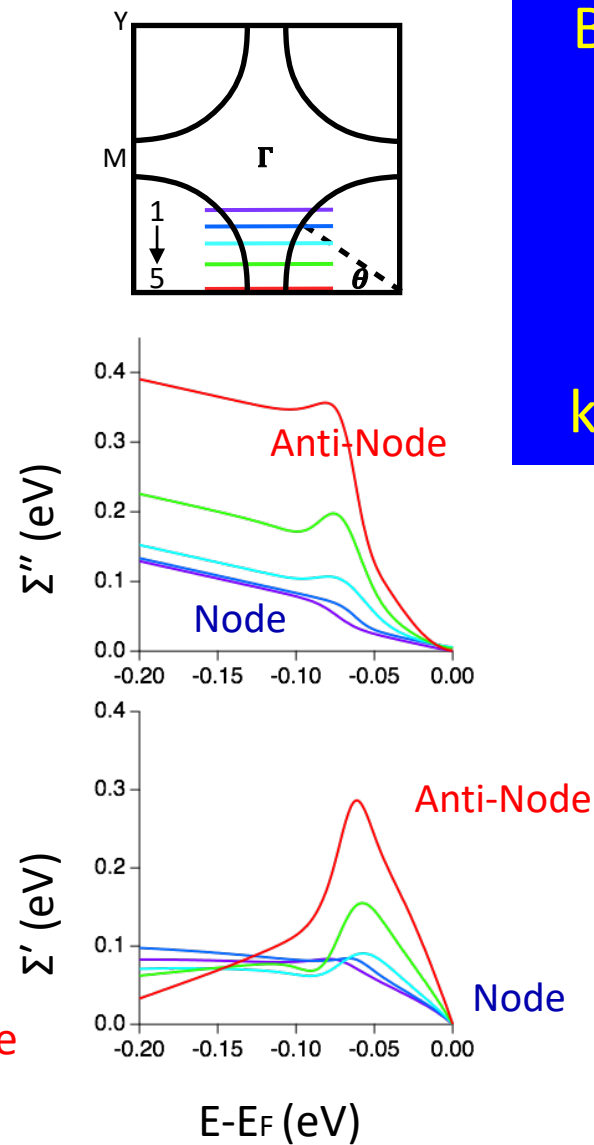
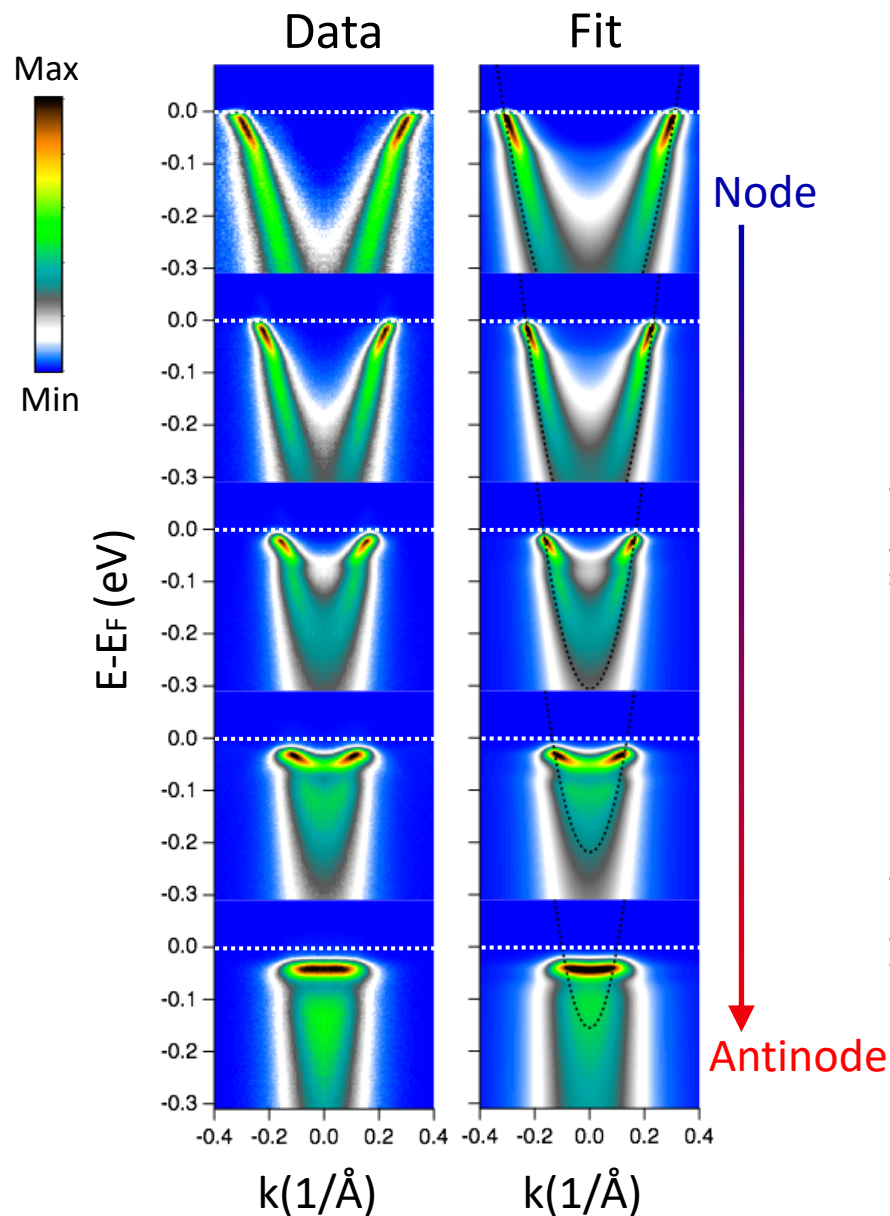
# 2D fit result shown as 1D cuts (All fits are done simultaneously)



2D fit results are dashed black lines.

Band curvature makes MDCs asymmetric – can not captured in MDC fitting.

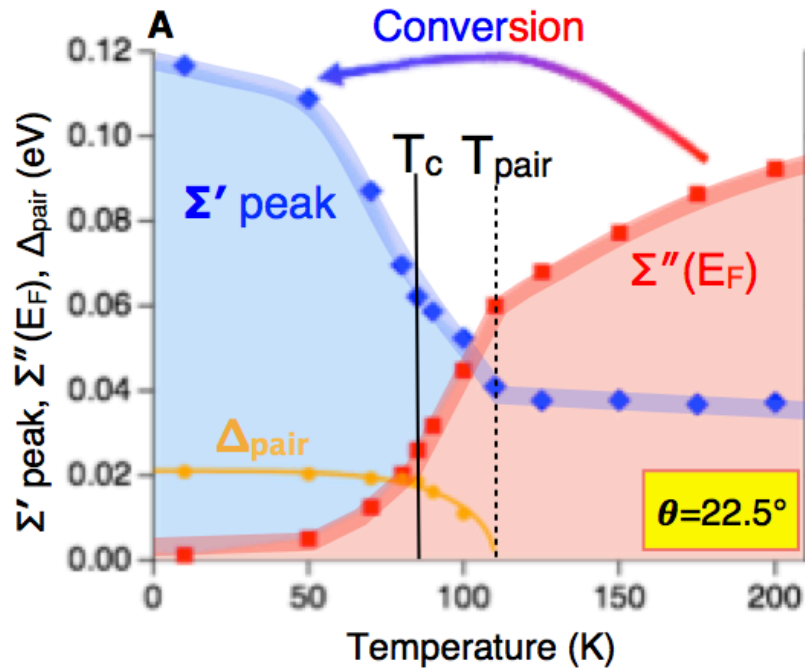
Orders of magnitude fewer free parameters than from 1D fits.



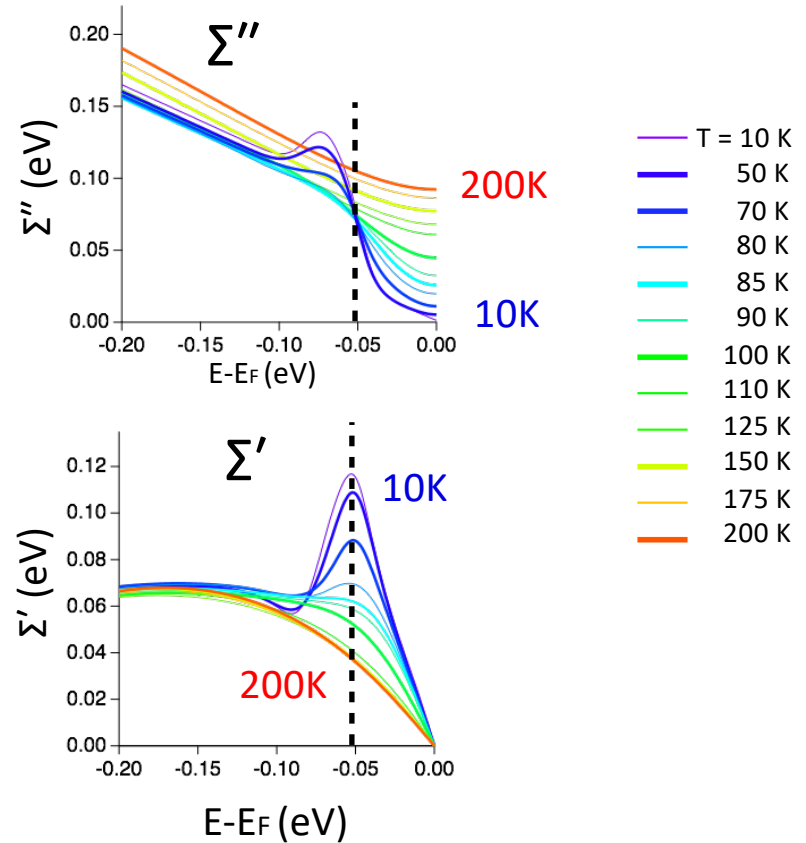
Pb-  
 $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$   
 $T_c=85\text{K}$   
Lightly UD  
Low T

k-dependence

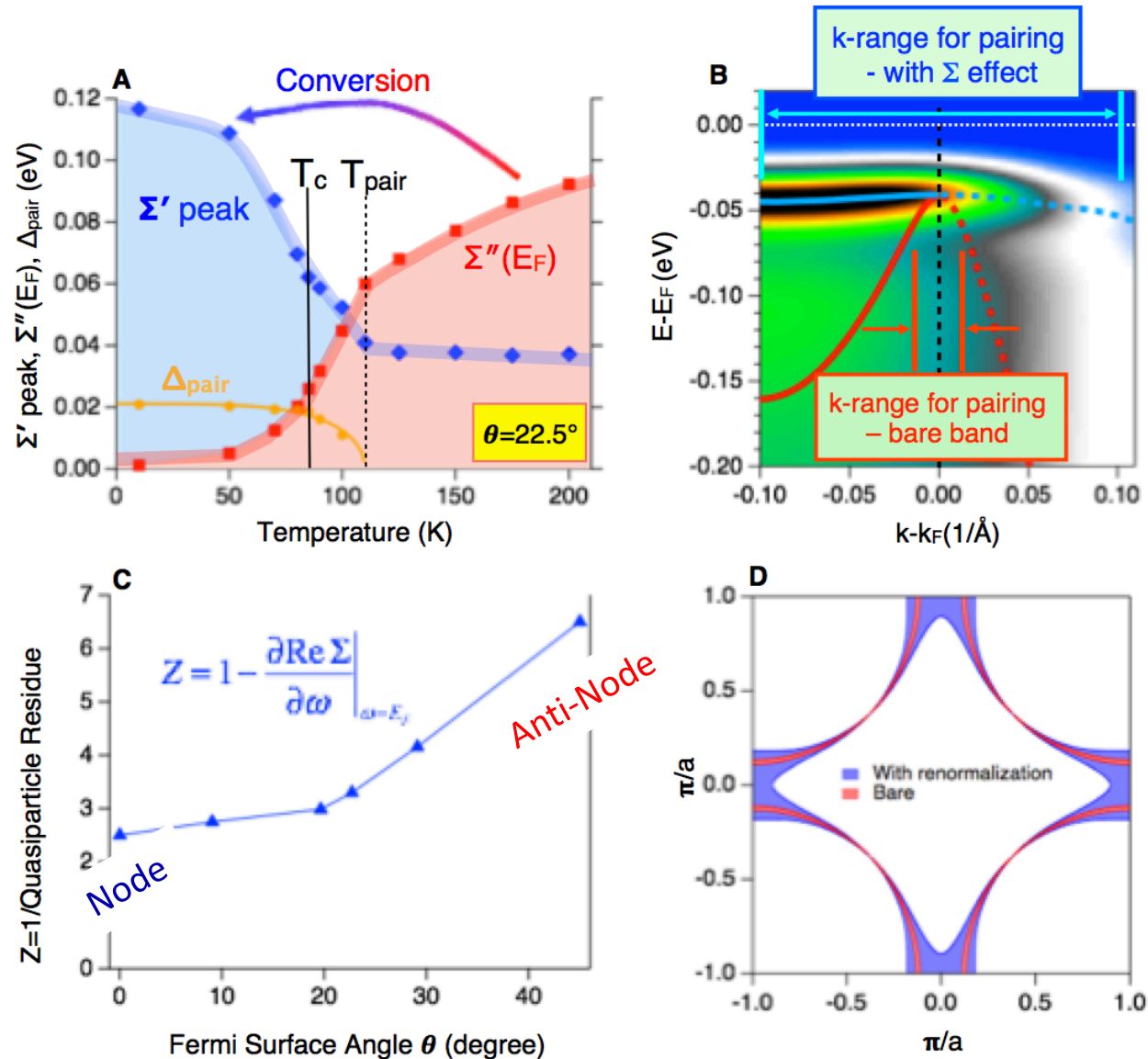
# Coherent organization (conversion) of electronic correlations



Mid-zone cut shown as an example.

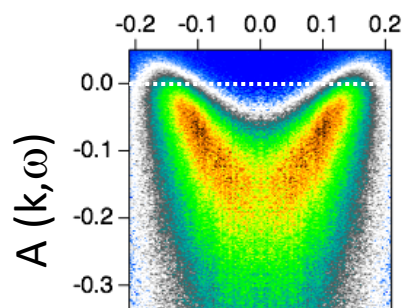


# Increased k-range for pairing due to the correlations

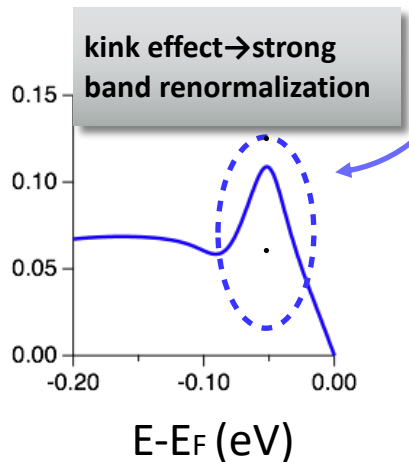
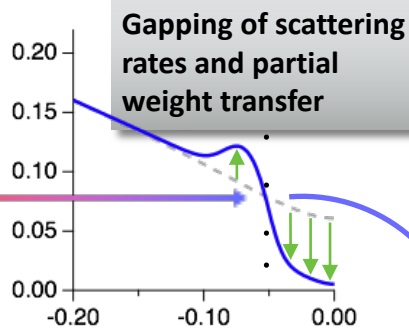
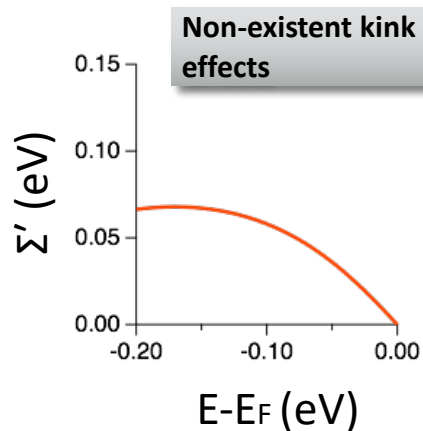
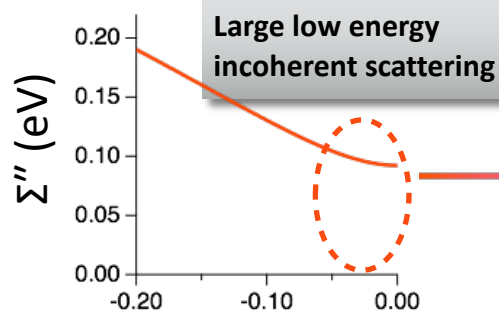
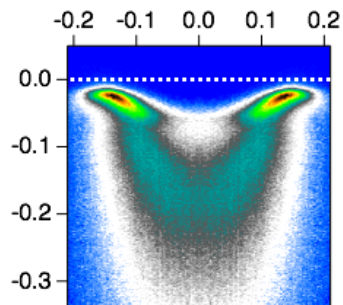


Mid-zone cut shown as an example.

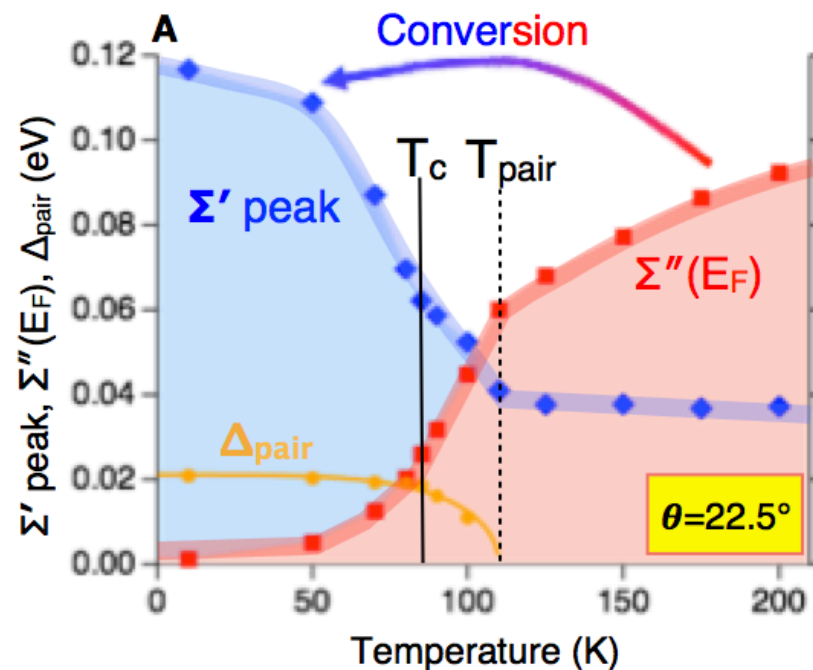
**High T normal state**



**Low T SC state**

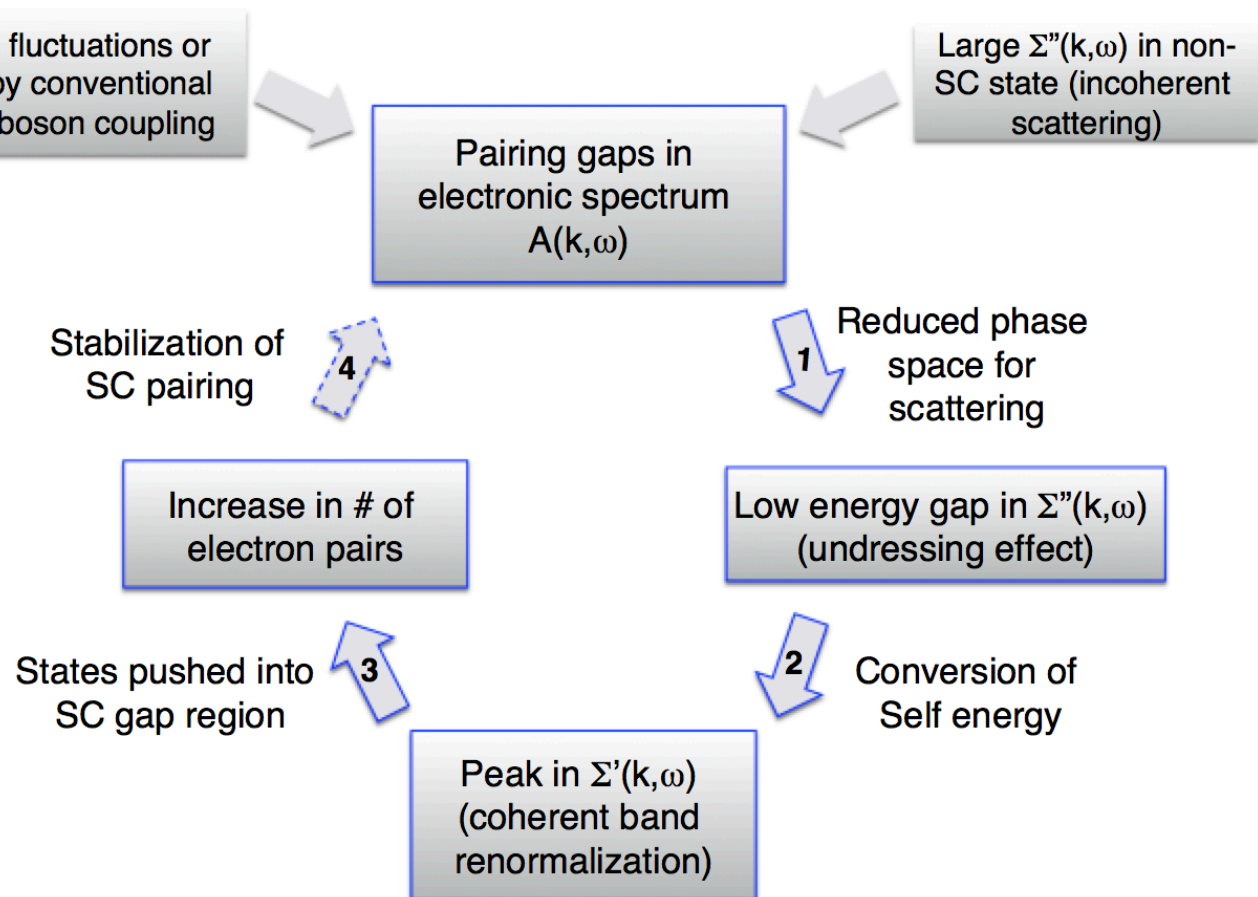


**Coherent organization  
(conversion) of electronic  
correlations**



**Stronger  $\Sigma''$  in N-state gives a  
stronger  $\Sigma'$  effect in the SC state  
(strongest at antinode).**

# Positive feedback loop due to increasing # of states for pairing.



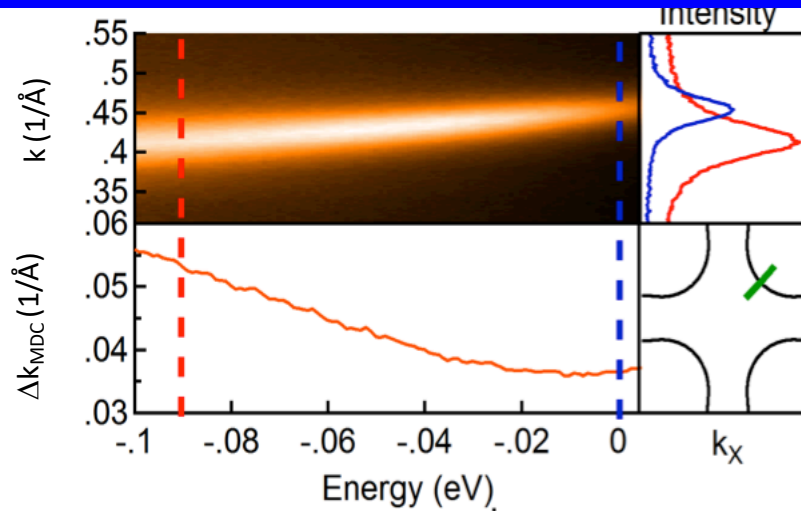
- Should strengthen any mechanism for pairing (e.g. electron-phonon).
- May support a fully-electronic mechanism, starting from fluctuations.

# Strange metal “normal state” of cuprates.

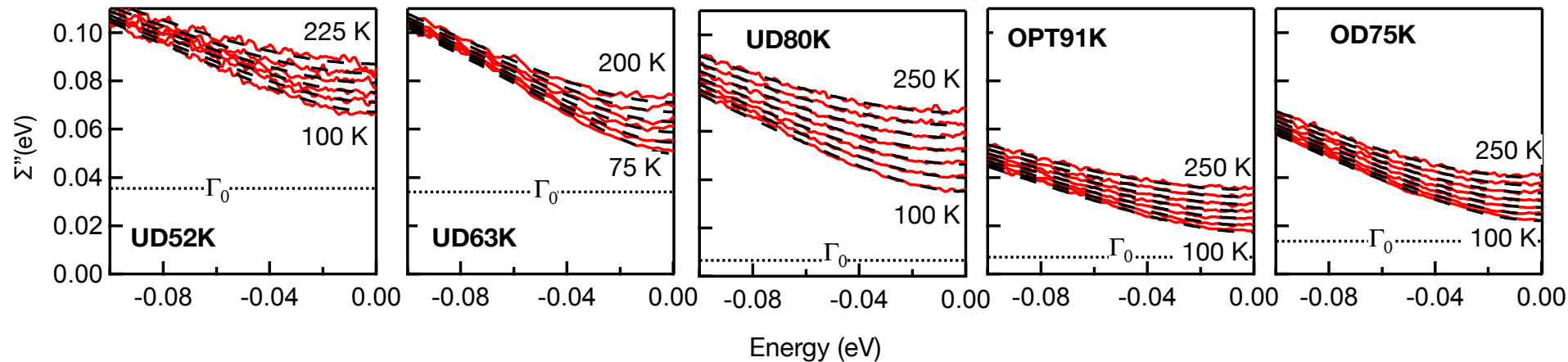
Linear-in-T resistivity (non-Fermi Liquid/non-quasiparticle).  
“Planckian” scattering. Much more....

Nodal electrons are most relevant for this.

# Normal-state nodal ARPES scattering rates ( $\Sigma''$ ) as a function of $\omega, T$ , doping (Bi2212).



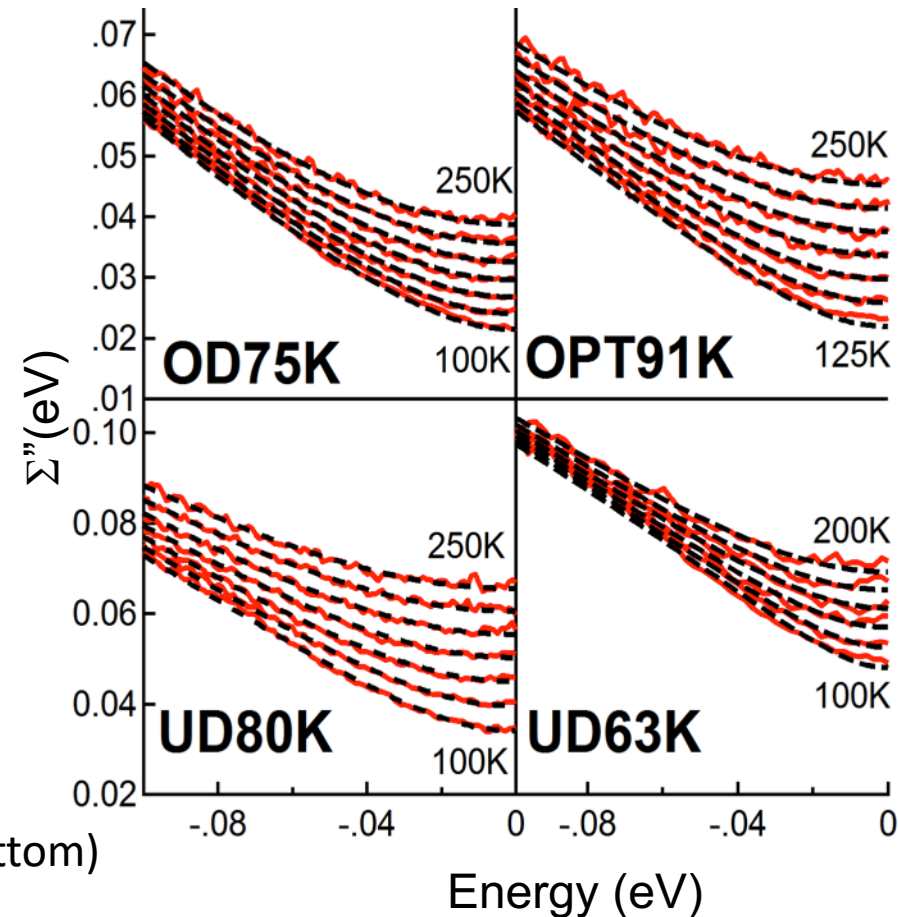
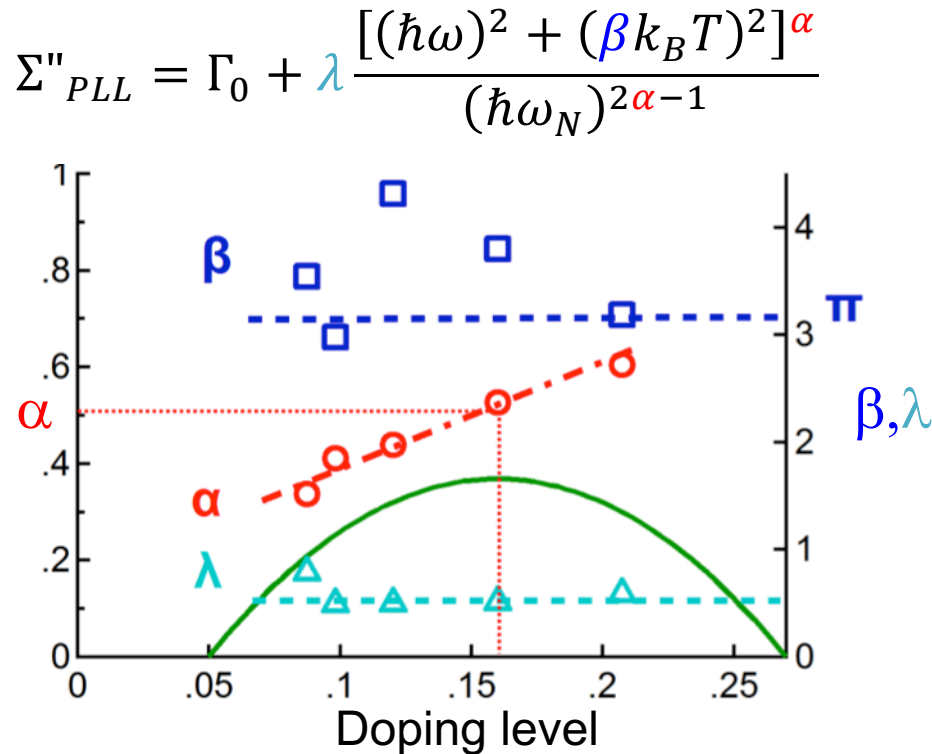
T.J. Reber, D.S.D. et al.  
(Nature Comm 2019)



All data looks quadratic at low  $\omega$  (FL-like), linear at high  $\omega$  (MFL-like).

All data for one sample (many T and E) fit simultaneously to a more general form of  $\Sigma(\omega, T)$ .

# Ansatz for normal state $\Sigma''$ - the Power Law Liquid (PLL) All data on one sample fit simultaneously.



$\omega_N$  (unit normalization) fixed at 0.5 eV ( $\sim$  band bottom)

$\Gamma_0$ =impurities+disorder (discussed later)

$\beta \sim \pi$ . Expected from Matsubara and Fermi Liquid (not previously proven).

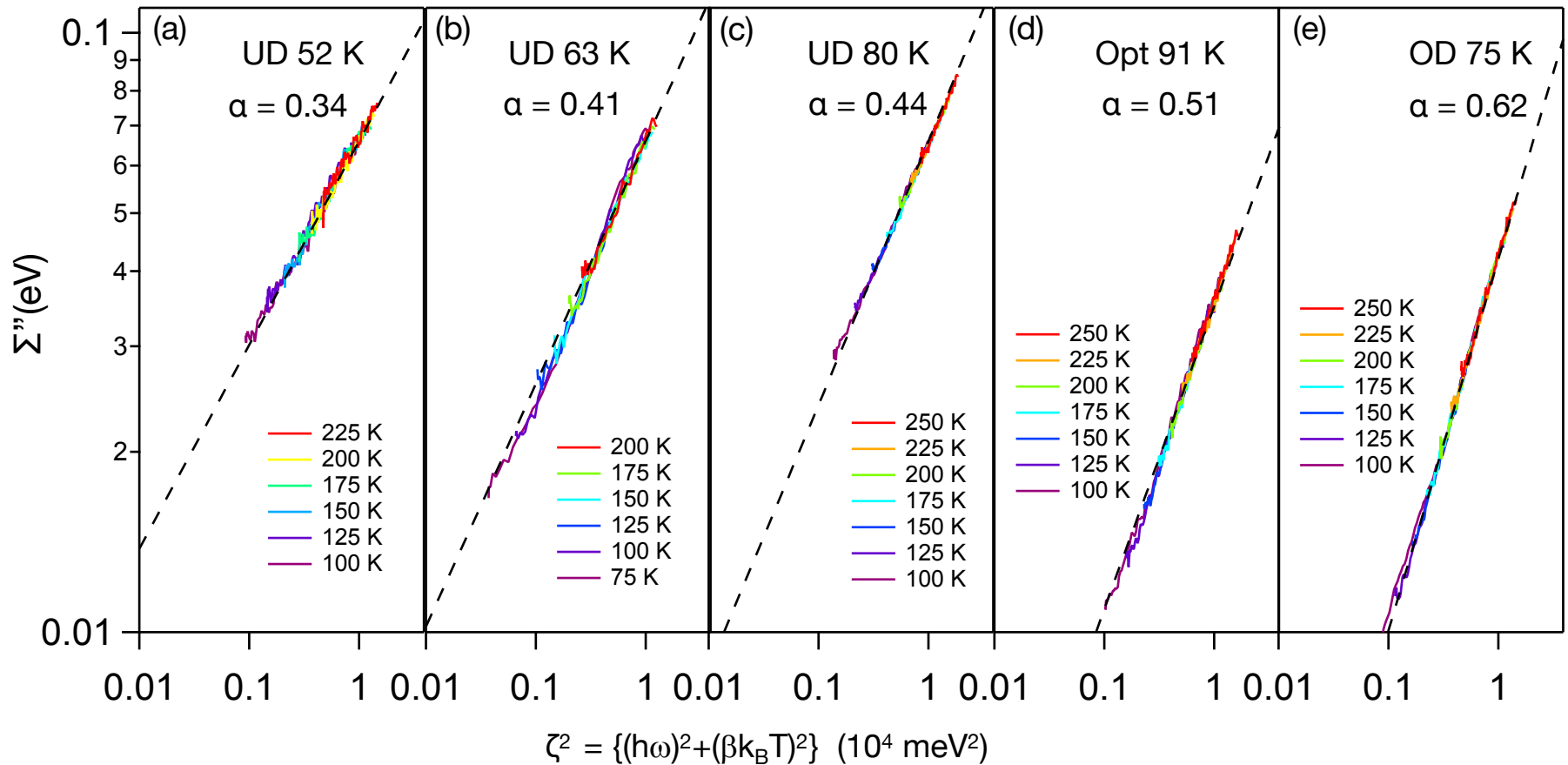
$\lambda$  = coupling strength  $\sim$  constant=0.5.

$\alpha = 0.5$  :  $\omega, T$  linear (Marginal Fermi Liquid).

$\alpha = 1$  :  $\omega, T$  quadratic (True Fermi Liquid)

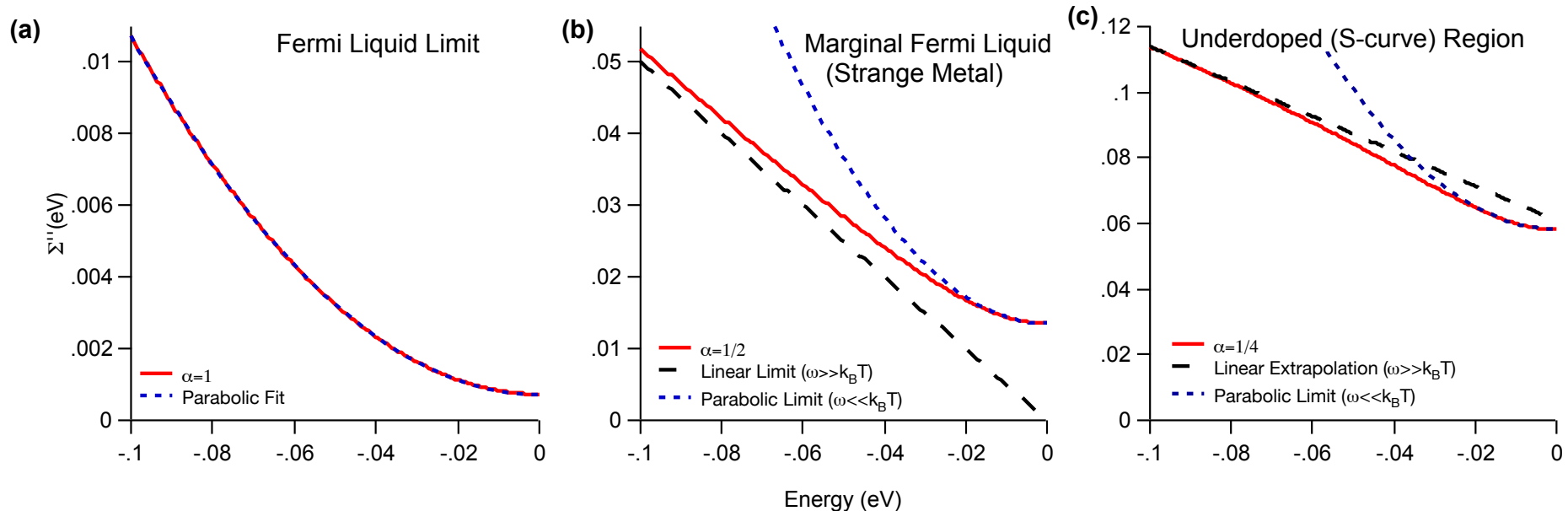
**Entire evolution essentially  
controlled by the one parameter  $\alpha$ .**

# Scaling behavior of $\Sigma''$ as a function of $T$ and $\omega$



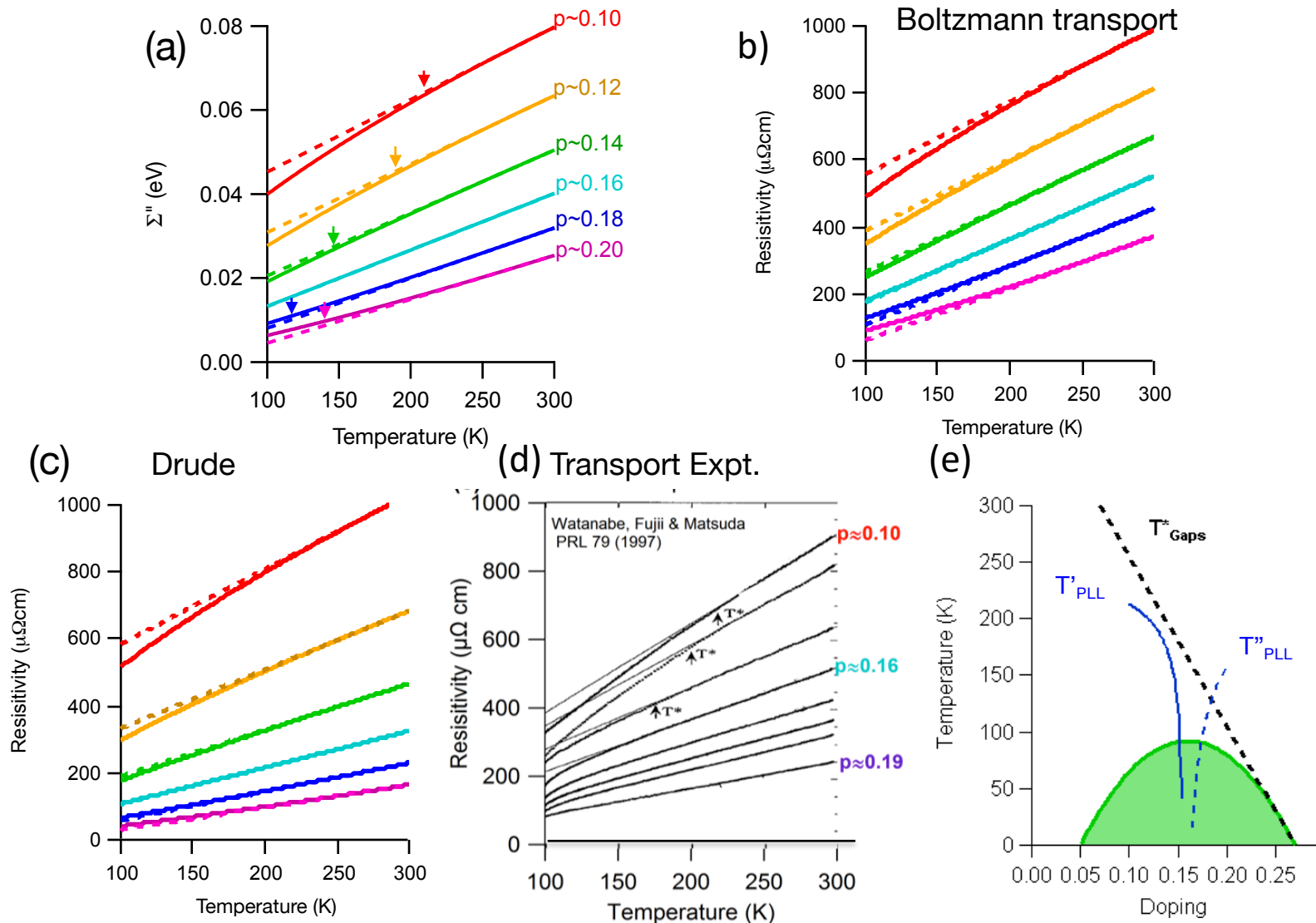
Different slopes  $\rightarrow$  different power law exponents  $\alpha$ .

# Low energy behavior of $\Sigma''$ as a function of PLL exponent $\alpha$ (T=100K)

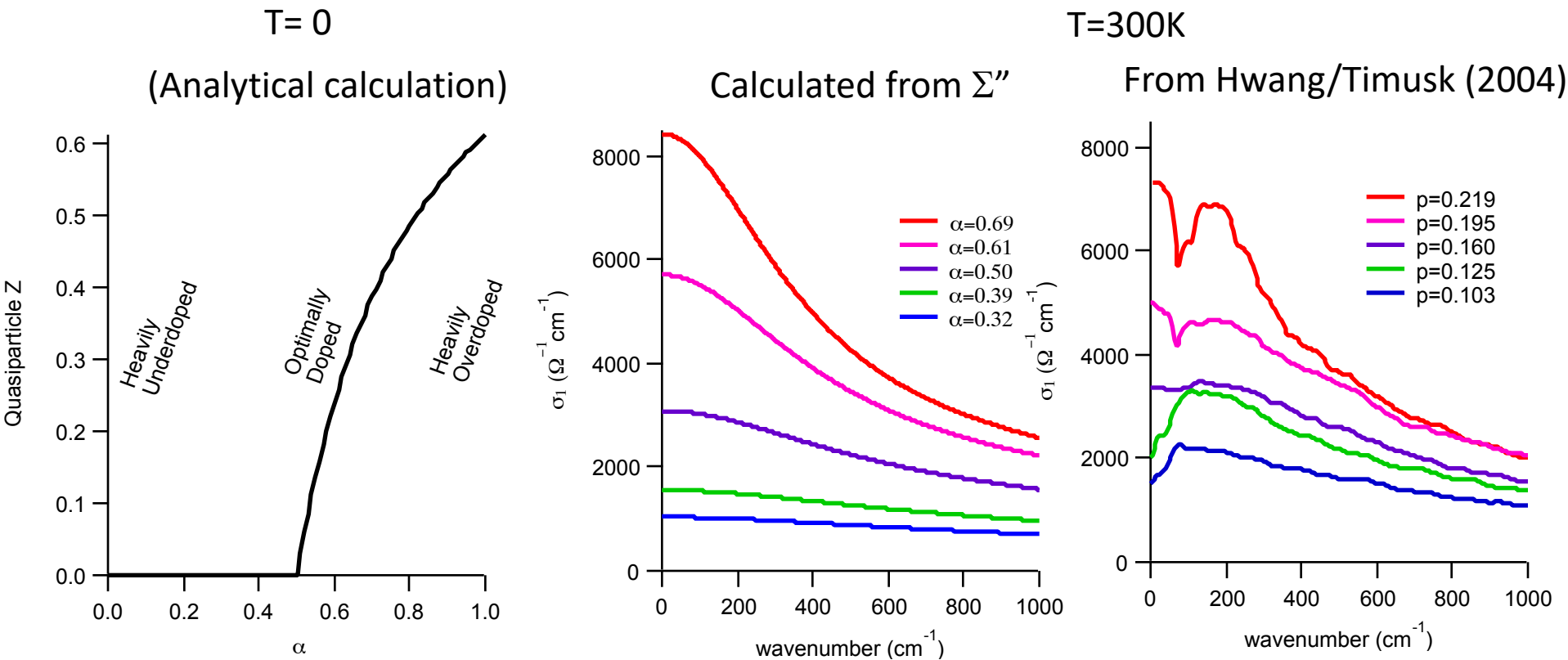


Low energy scattering rate right of middle and right spectra look quadratic.  
 Fermi Liquid? (No).  
 Indicative of quasiparticles? (No).

# Resistivity vs T from Power Law Liquid $\Sigma''$



# QP Residue and optical cond from Power Law Liquid $\Sigma''$



“Sharpish” ARPES QP-like peak and optical Drude peak even when  $T=0$  quasiparticle residue is identically zero.

# Power Law Liquid as a quantum critical phase (not quantum critical point).

Luttinger liquid physics in 1D. PLL spectra (and ARPES data) don't have the power law spectral weight that the LL physics has.

SYK (Sachdev/Ye/Kitaev) exactly solvable models.

Quantum gravity-based ideas (ADS/CFT)?

Doped Hubbard model? Si, Kotliar 1993.

The superconductivity is born out of these states. How is all this connected?

# SCIENTIFIC REPORTS

OPEN

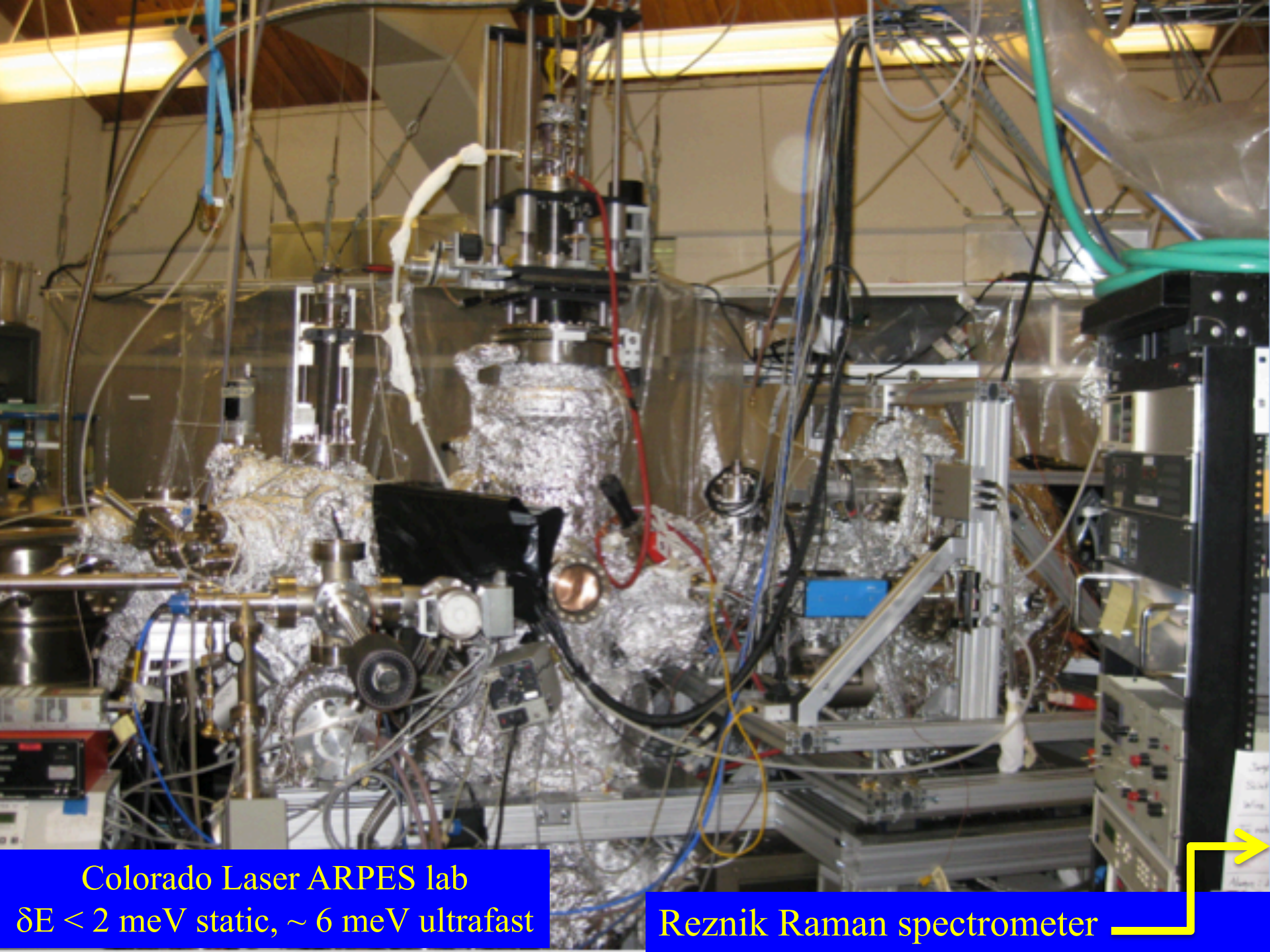
## Novel Electron-Phonon Relaxation Pathway in Graphite Revealed by Time-Resolved Raman Scattering and Angle-Resolved Photoemission Spectroscopy

Received: 11 August 2016  
Accepted: 13 December 2016  
Published: 19 January 2017

Jhih-An Yang<sup>1</sup>, Stephen Parham<sup>1</sup>, Daniel Dessau<sup>1,2</sup> & Dmitry Reznik<sup>1</sup>



Dmitry Reznik

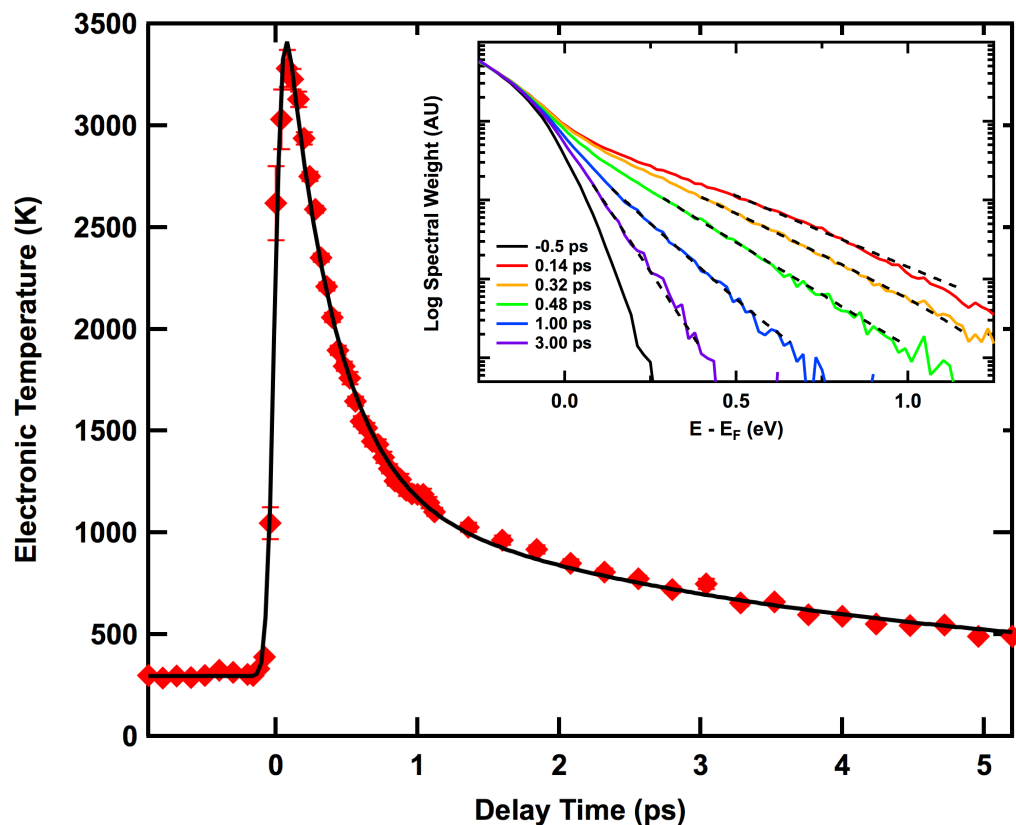


Colorado Laser ARPES lab

$\delta E < 2$  meV static,  $\sim 6$  meV ultrafast

Reznik Raman spectrometer

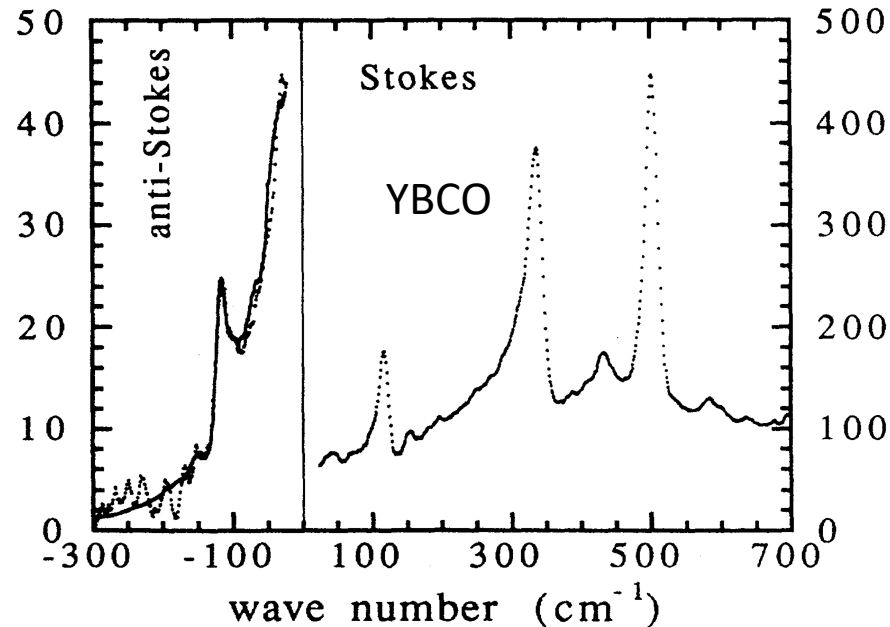
# Electronic Temperature of graphite as a function of time from tr-ARPES (pumped hard).



Good fit combining fast and slow time constants: 0.35ps and 3.5ps.

# Looking at ratio of Stokes to anti-Stokes to track phonon mode occupation

Anti-Stokes: Energy gain side (negative energy)



Stokes: Energy loss side (positive energy)

Principle of detailed balance:

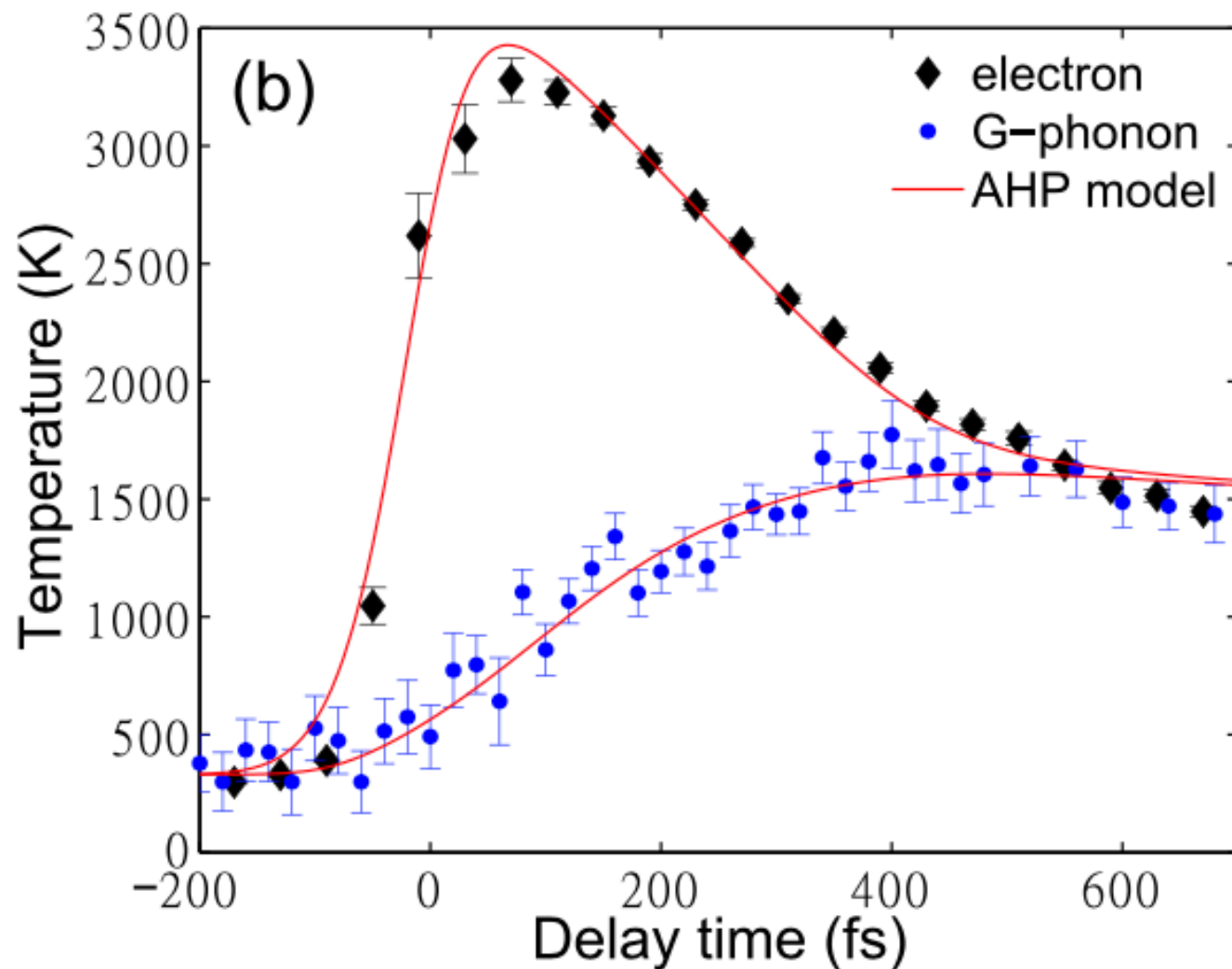
$$S(-\omega) = S(\omega)e^{-\hbar\omega/k_bT}$$

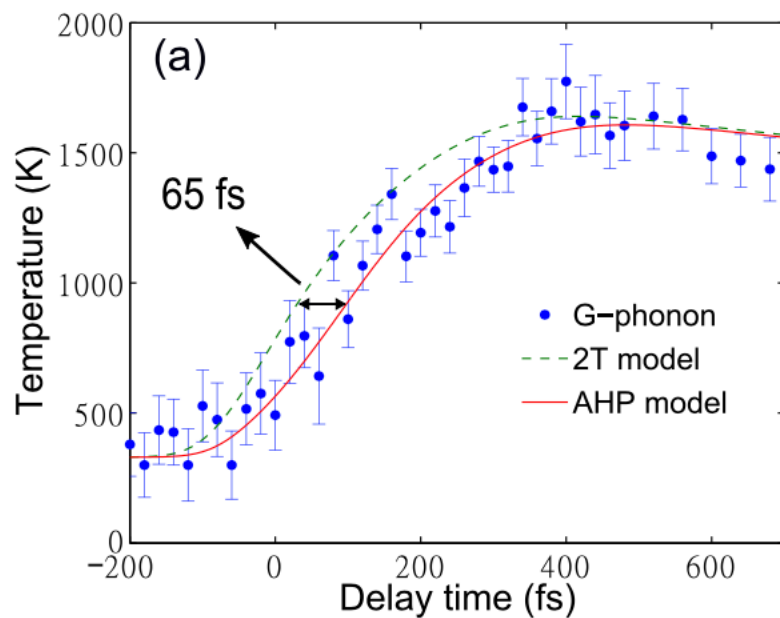
Phonon occupation number:

$$n = \left(e^{\hbar\omega/k_bT} - 1\right)^{-1}$$

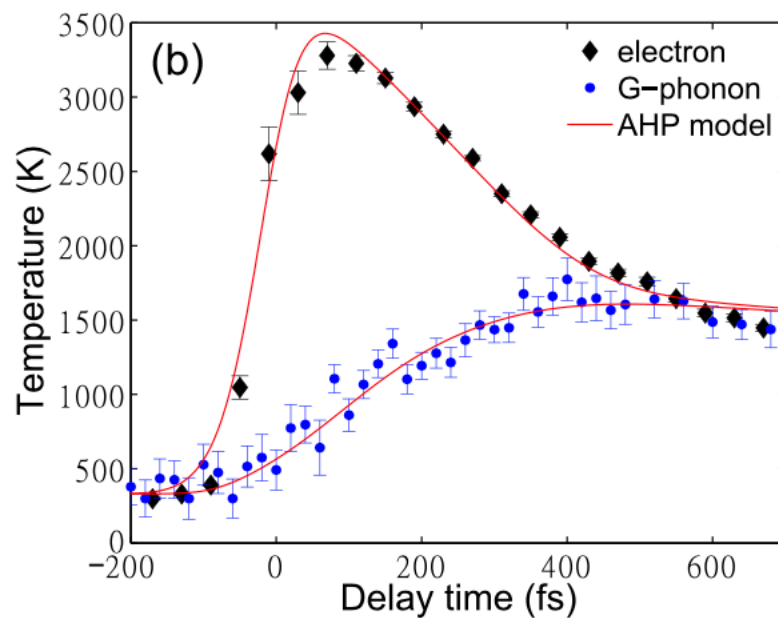
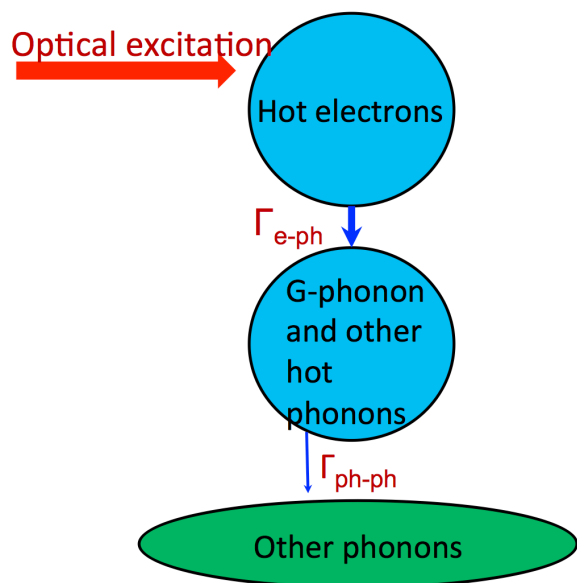
Phonon occupation number can be extracted from the ratio of intensities on the Stokes and anti-Stokes sides.

# Electronic and phonon temperatures of graphite as a function of time – combined trARPES & tr-Raman.

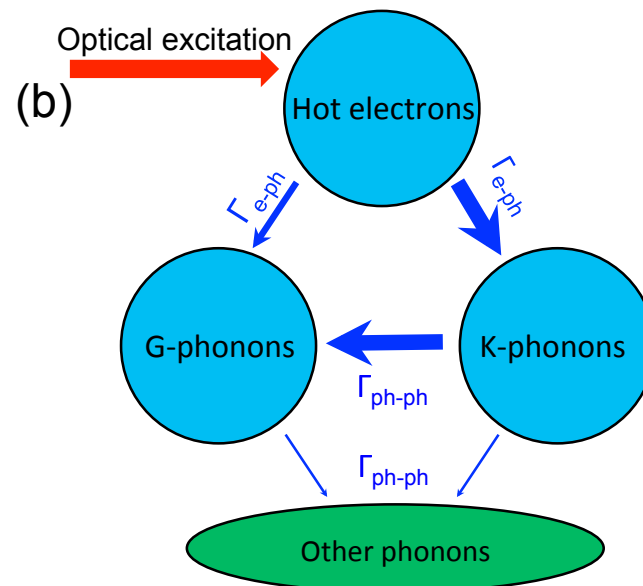




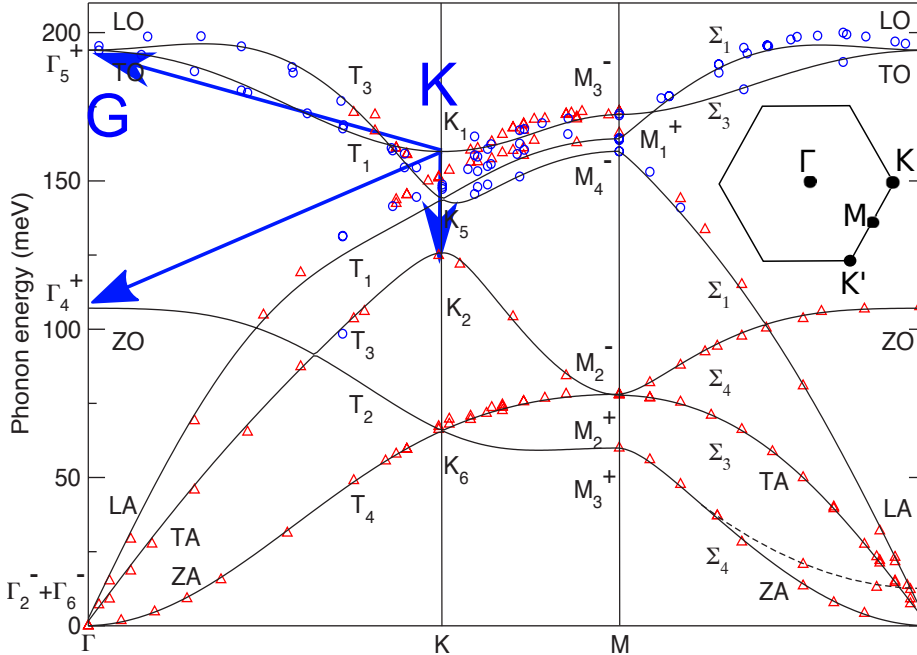
2 temperature (2T) model



Anharmonic phonon (AHP) model



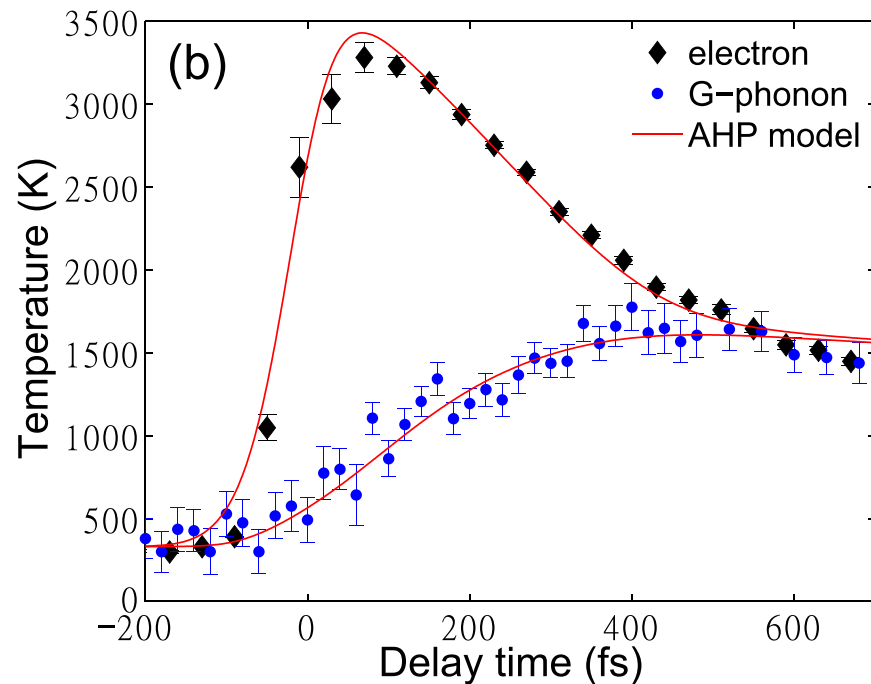
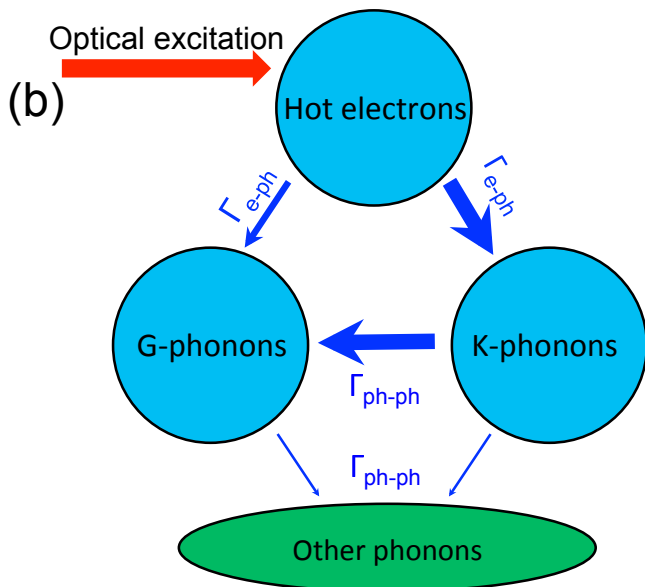
# AHP model consistent with phonon dispersions



M. Mohr et al. Phys.  
Rev. B 76, 035439  
(2007).

Anharmonic phonon model allows for hot phonons to decay into each other on very fast timescales. The resulting time-scale is consistent with ab-initio calculations and the observed time-delay

# Anharmonic phonon model



Electron-hole pair decay into K-phonon: (LDA) 176 fs

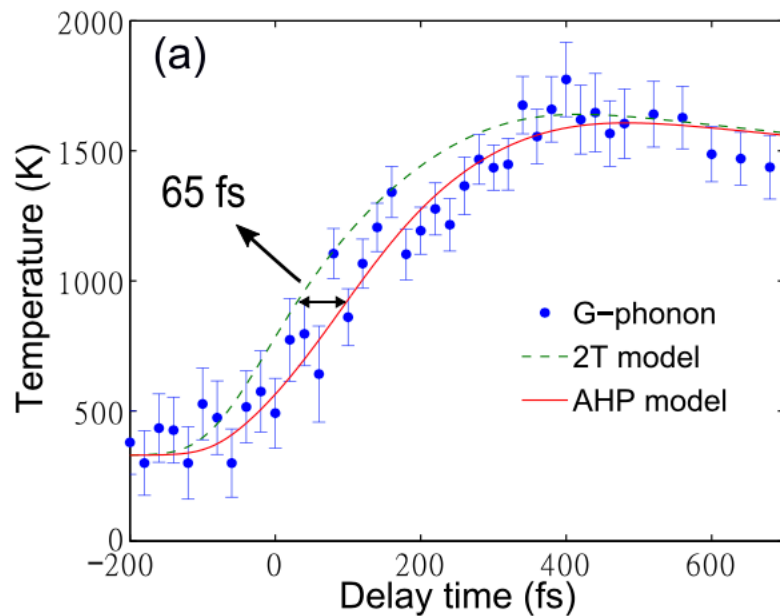
4-phonon decay involving K and G-phonons: (2400K) 132 fs

$$\frac{dT_{el}}{dt} = \frac{I(t) - \Gamma_{e-ph}(T_{el}, T_G) - \Gamma_{e-ph}(T_{el}, T_K)}{C_{el}(T_{el})},$$

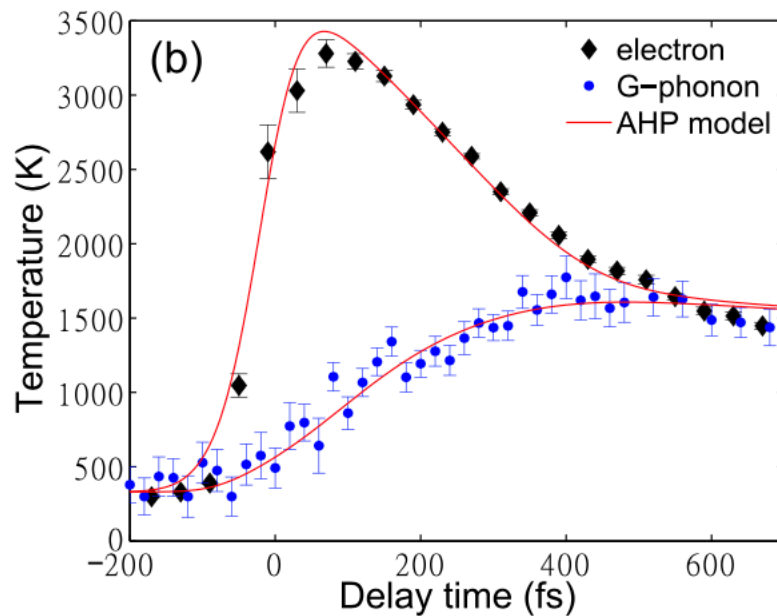
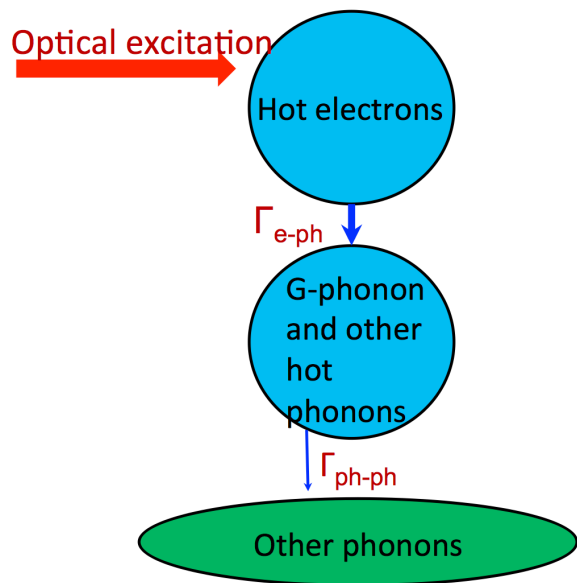
$$\frac{dT_K}{dt} = \frac{\Gamma_{e-ph}(T_{el}, T_K) - \Gamma_{ph-ph}(T_K, T_G)}{C_K(T_K)} - \frac{T_K - T_0}{\tau},$$

$$\frac{dT_G}{dt} = \frac{\Gamma_{e-ph}(T_{el}, T_G) + \Gamma_{ph-ph}(T_K, T_G)}{C_G(T_G)} - \frac{T_G - T_0}{\tau},$$

S. Piscanec et al. Phys. Rev. Lett. 93, 185503 (2004) S. Berciaud et al. PRL 104, 227401 (2010)  
 M. Lazzeri et al. Phys. Rev. B 73, 155426 (2006) G. Montagnac et al. Carbon 54, 68–75 (2013).



2 temperature (2T) model



Anharmonic phonon (AHP) model

