



UNIVERSITY OF BRITISH COLUMBIA



Andrea Damascelli

Quantum Materials in the Time Domain



MP-UBC-UTokyo Centre for QM
Quantum Matter Institute

UBC-Moore Centre for Ultrafast Quantum Matter



Andrea Damascelli
David J. Jones

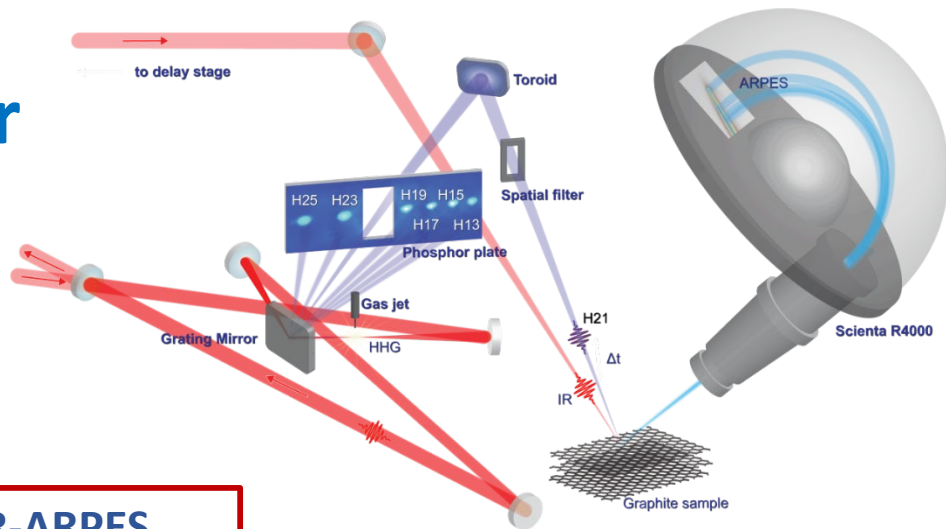


TR-ARPES

6.2 eV system

Transient Collapse high-T_c superconductivity in Bi2212
Boschini et al. Nat. Materials 17, 416 (2018)

Relation pseudogap and spin correlations in NCCO
Boschini*, Zonno*, et al. arXiv:1812.07583



10-40 eV / 20 meV / 190 fs / 60 MHz

Determination mode-projected
electron-phonon matrix element in graphite
Na*, Mills*, et al., Science 366, 1231 (2019)

Cavity-enhanced HHG for XUV TR-ARPES
Mills et al., RSI 90, 083001 (2019)

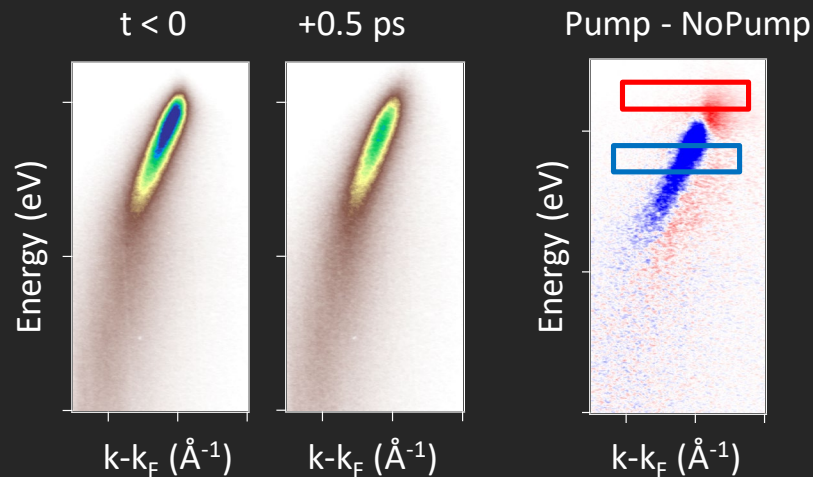
TR-ARPES Bi2212

Non-equilibrium technique (TR-ARPES)

$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ UD82

Common analysis approach:

Study **population**/**depletion** dynamics



ARPES intensity

$$I_{\text{PES}}(\mathbf{k}, \omega) = A(\mathbf{k}, \omega) \cdot |M|^2 \cdot f(\omega)$$

Electronic distribution

BUT also A (spectral function) may change:

- i) Pump-induced holes enhance the scattering rate of QPs
- ii) Pump-induced distortions modify the bare energy dispersion
- iii) **In a superconductor, the loss of coherence affects $A(\mathbf{k}, \omega)$**

Disentangle $A(\mathbf{k}, \omega, \tau)$ & $f(\omega, \tau)$ contributions

Phase stiffness in cuprates

T_C

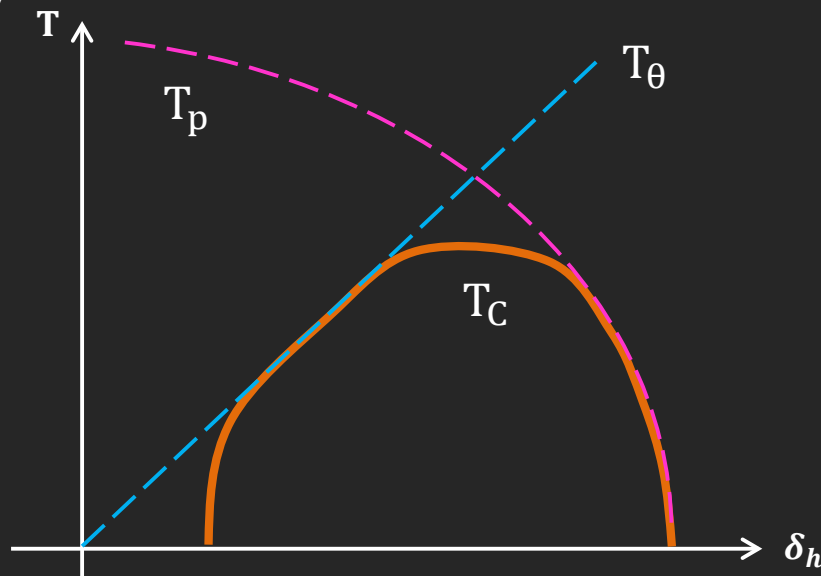
Formation pairs.
Pairing energy E_p

Onset of phase
coherence

$$V_\theta \propto n_s(0)$$

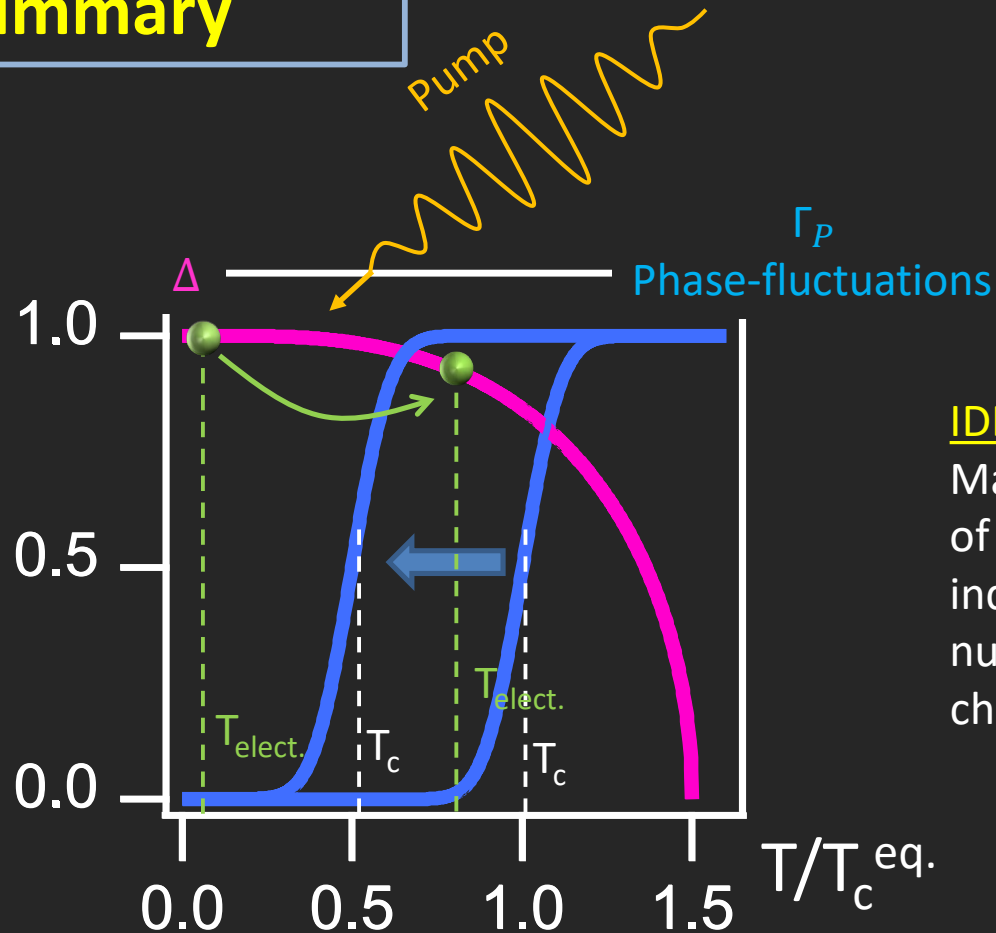
Cuprate SCs: $E_p \approx V_\theta$
(low superfluid density)

$$T_C \approx \min(T_p, T_\theta)$$



Emery and Kivelson
Nature (1995)

Idea/Summary

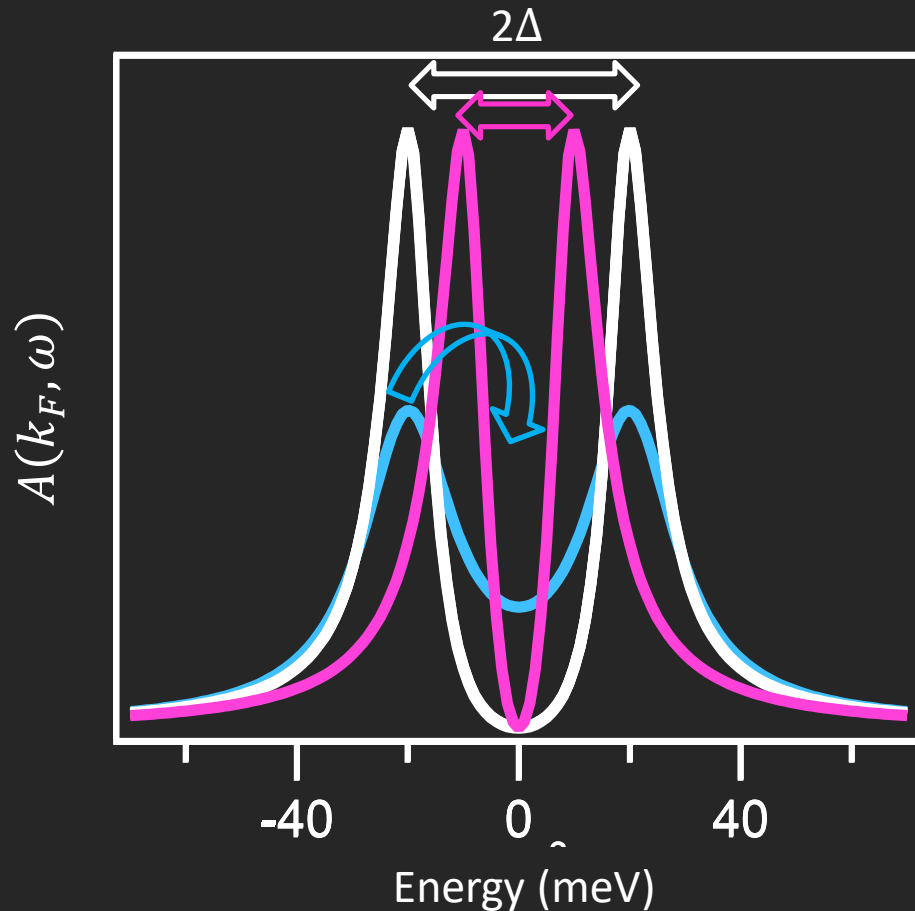


IDEA/Summary:

Manipulate the density of phase fluctuations independently of the number of across-gap charge excitations.

How detect phase-fluctuations?

We can look at the ultrafast dynamics within the superconducting gap 2Δ



Pairing strength:
Modification of 2Δ

Phase fluctuations:
Filling within the gap

$$A(k_F, \omega) = -\frac{1}{\pi} \frac{\Sigma''}{(\omega - \Sigma')^2 + (\Sigma'')^2}$$

$$\Sigma = -i \cdot \Gamma_s + \frac{\Delta^2}{\omega + i \cdot \Gamma_p}$$

Norman et al. PRB **57**, R11093 (1998)

Franz and Millis PRB **58**, 14572 (1998)

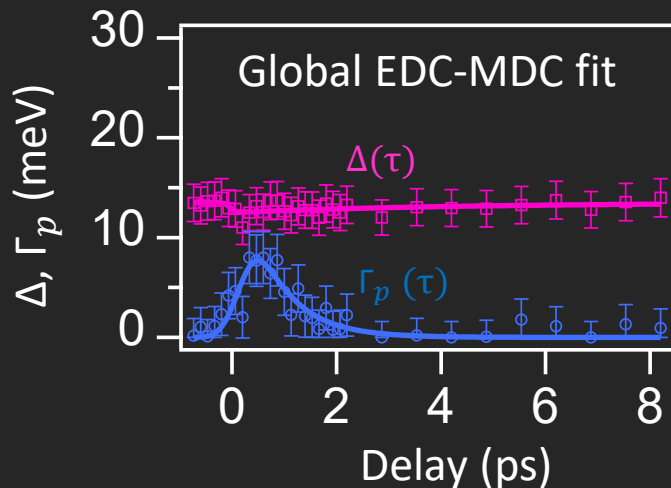
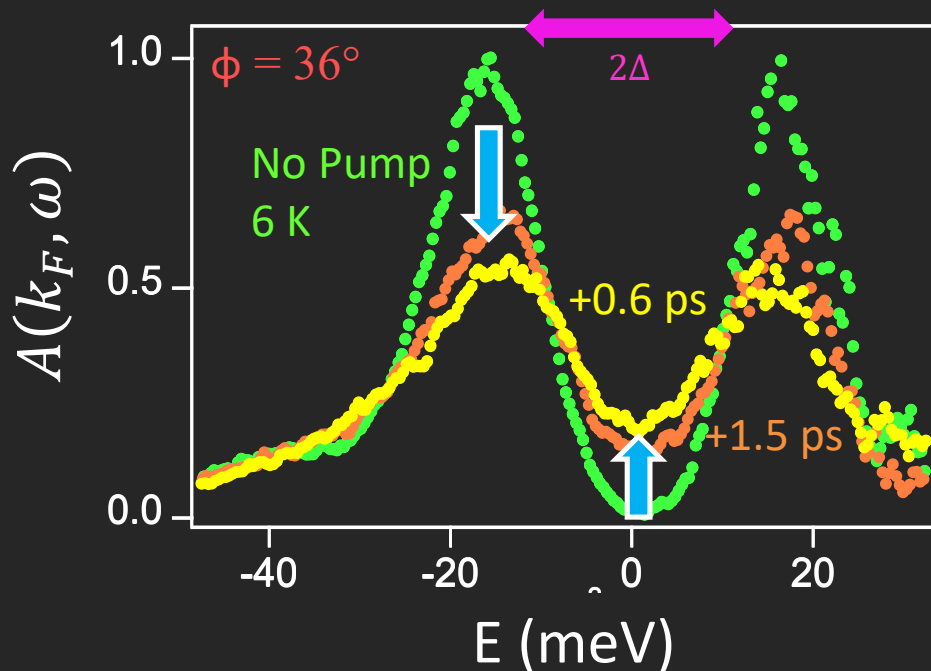
Boschini et al. Nat. Materials (2018)

Gar~~X~~closure vs. filling

$$\frac{\text{EDC}_{\text{off}}(k_F)}{\int \text{EDC}_{\text{node}}(k) dk} \propto A_{\text{off}}(k_F, \omega)$$

EDC deconvoluted from energy resol.
H.-B. Yang et al. Nature **456**, 77 (2008)

Similar to TDOS method
Reber et al. Nat. Phys. **8**, 606 (2012)



$$A(k_F, \omega) = -\frac{1}{\pi} \frac{\Sigma''}{(\omega - \Sigma')^2 + (\Sigma'')^2}$$

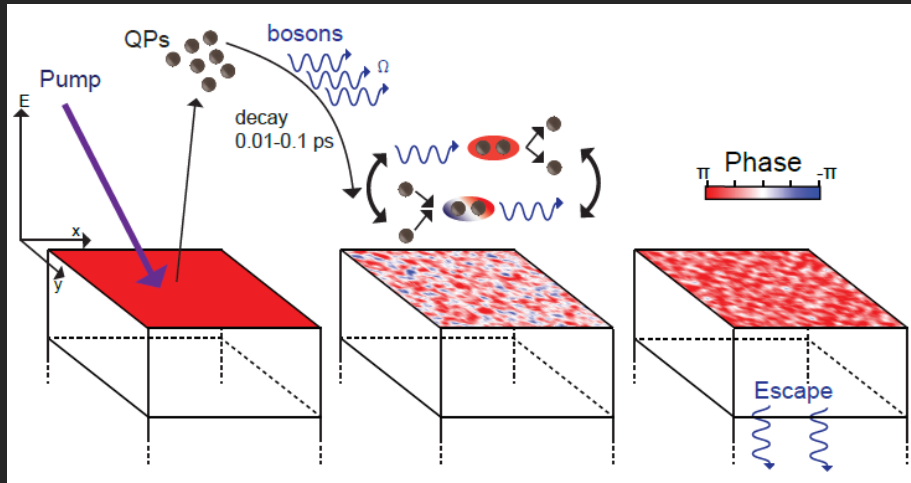
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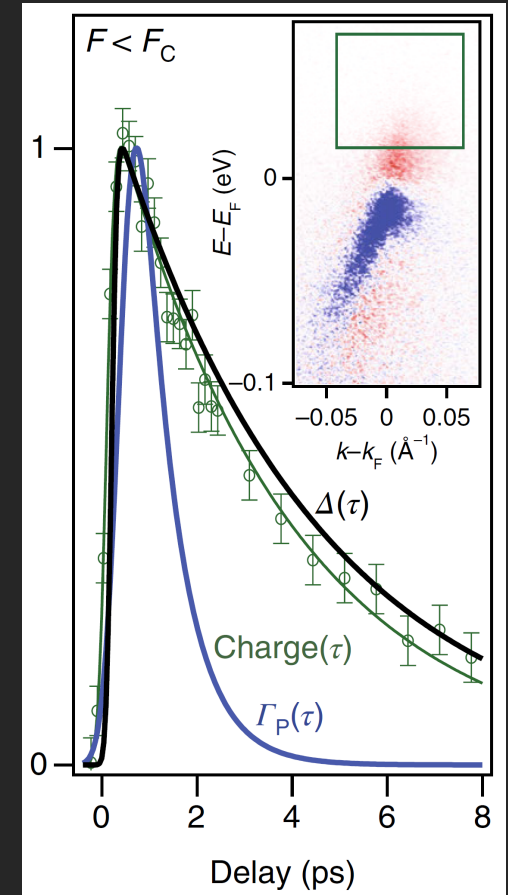
Boschini et al. Nat. Materials (2018)

Phase decoherence: mechanism

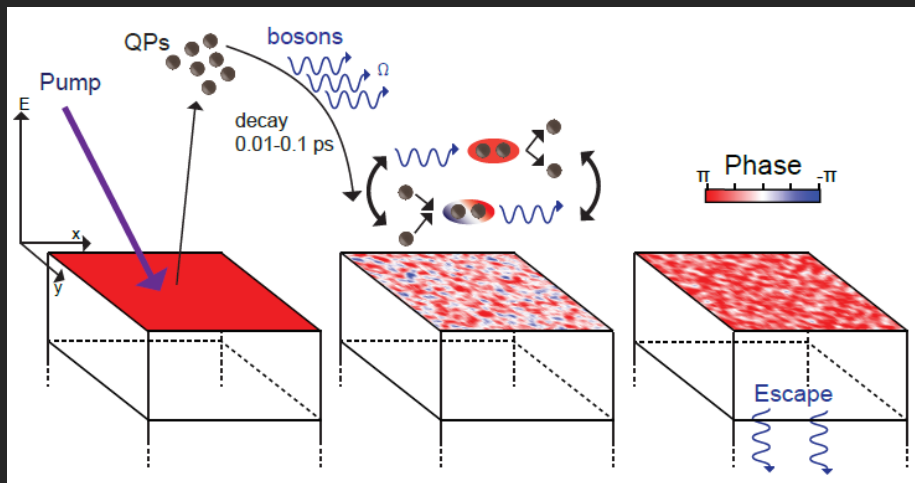


Non-thermal bosons emitted by the thermalization of hot QPs are the source of the phase fluctuations

Phase coherence plays major role in determining T_c in cuprates



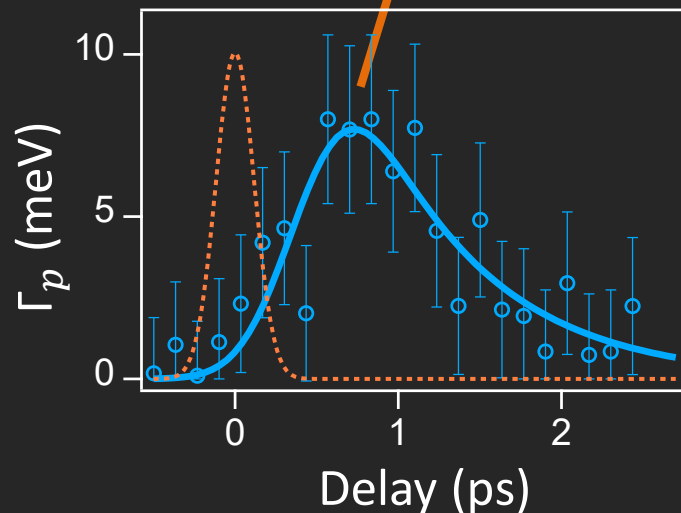
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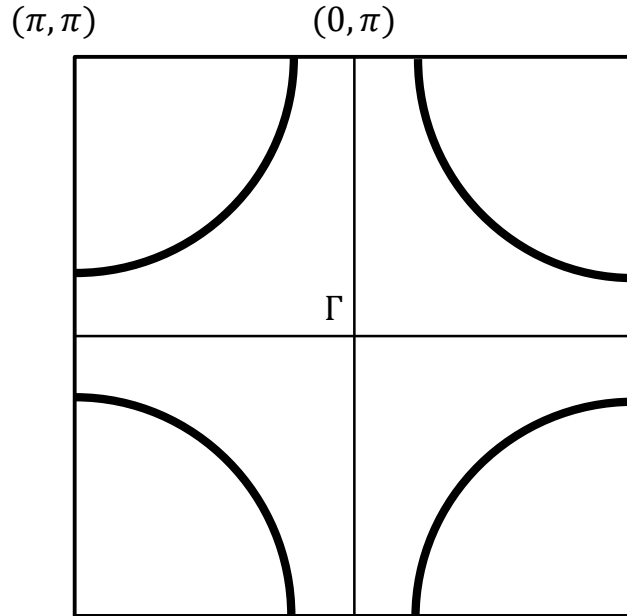
Phase coherence plays major role in determining T_c in cuprates

Build-up time >500 fs:
Formation of an out-of-equilibrium bosonic population



Pseudogap in Electron Doped Cuprates

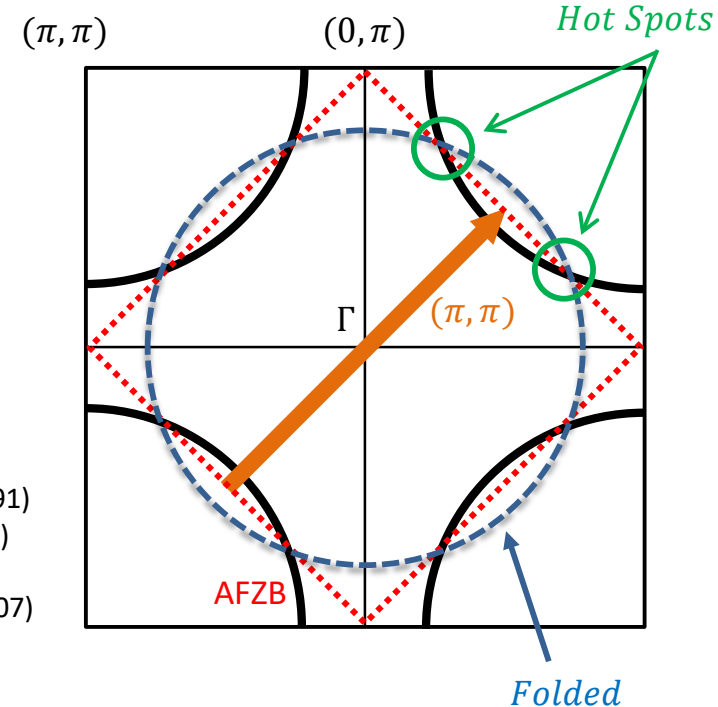
Antiferromagnetic order leads to band folding and hot spots



AFM
correlations

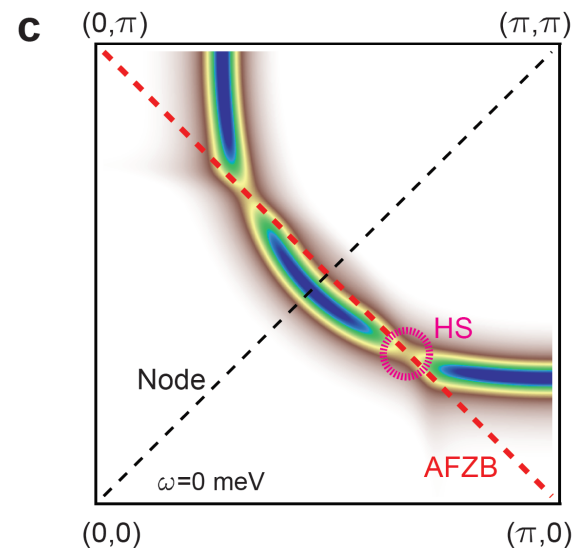
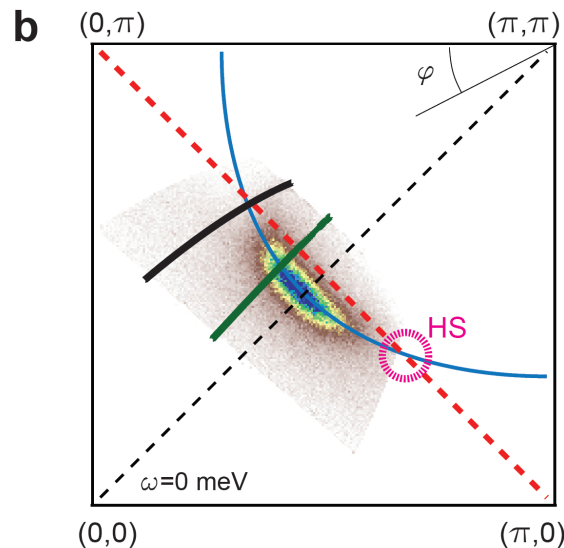
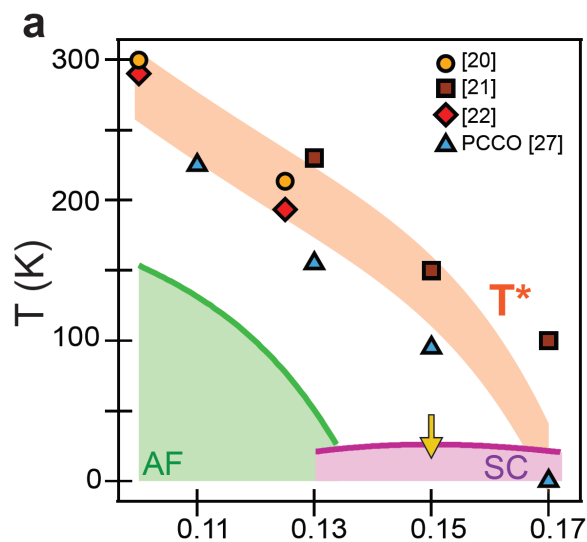


Millis et al. PRB 42, 167 (1990)
Monthoux et al., PRL 67 3448 (1991)
Vilk, Tremblay, JPI 7, 1309 (1997)
Zimmers et al. EPL 70, 225 (2005)
Norman et al. PRB 76, 174501 (2007)



Pseudogap in Electron Doped Cuprates

Antiferromagnetic order leads to band folding and hot spots

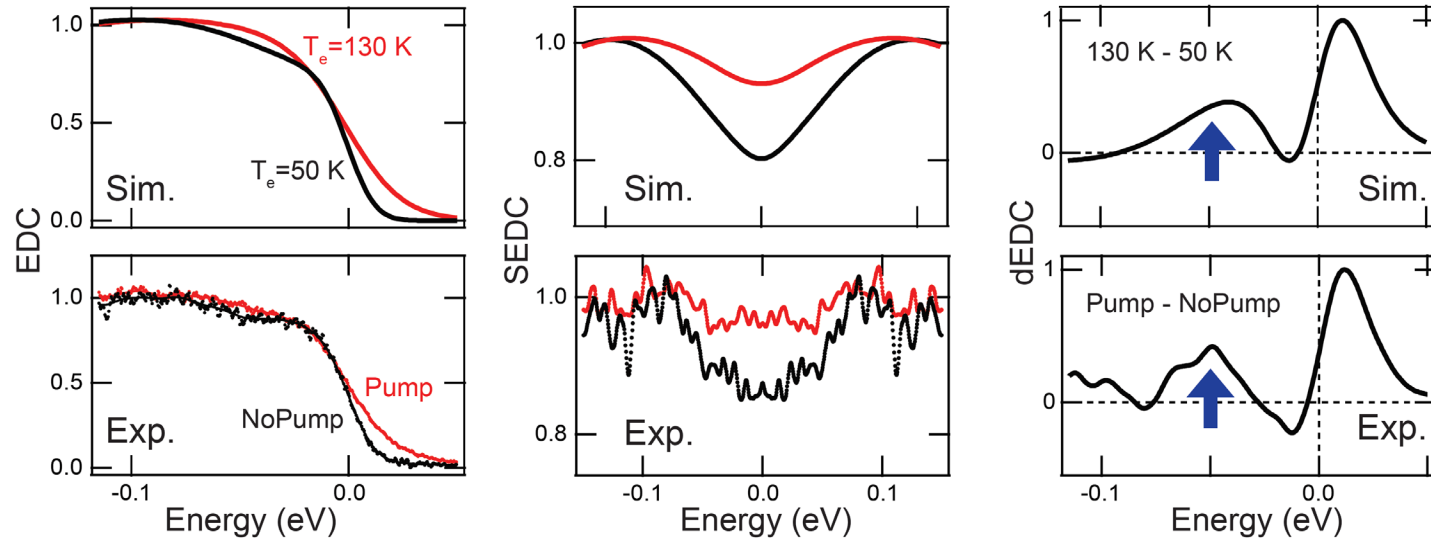


What about optimal doping, in absence of long-range AFM?

Pseudogap in Electron Doped Cuprates

$$G^{-1}(\mathbf{k}, \omega) = \omega - \epsilon_{\mathbf{k}} + i\eta - \frac{\Delta_{\text{PG}}^2}{\omega - \epsilon_{\mathbf{k}+\mathbf{q}} + i\Gamma}$$

Γ = spin-fluctuations induced

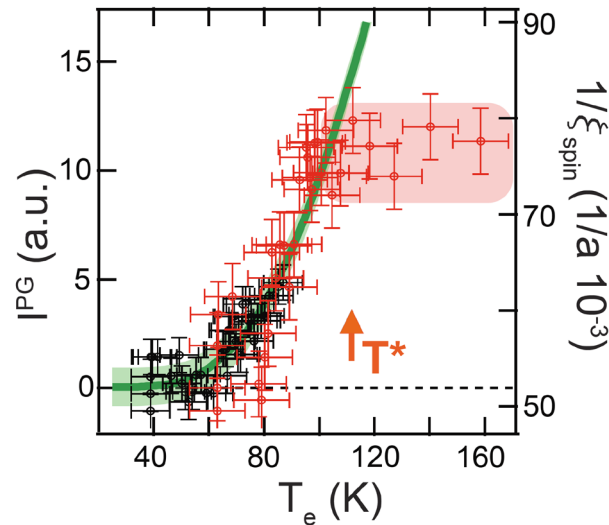
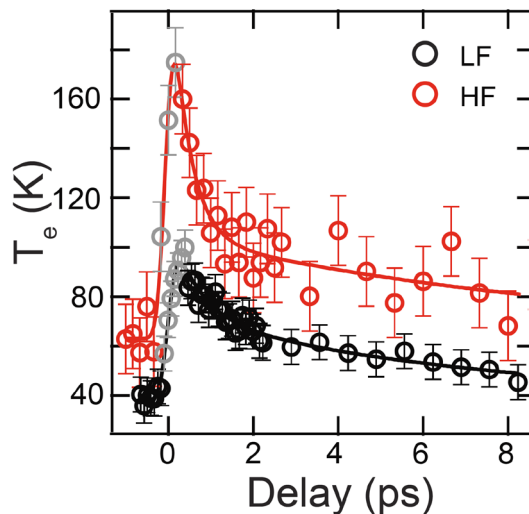
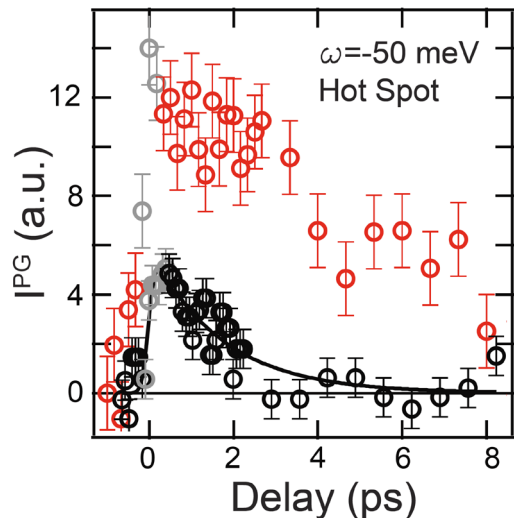


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Γ = spin-fluctuations induced



Emergence of pseudogap from short-range spin-correlations!

Pseudogap in Electron Doped Cuprates

$$G^{-1}(\mathbf{k}, \omega) = \omega - \epsilon_{\mathbf{k}} + i\eta - \frac{\Delta_{\text{PG}}^2}{\omega - \epsilon_{\mathbf{k}+\mathbf{q}} + i\Gamma}$$

Γ = spin-fluctuations induced

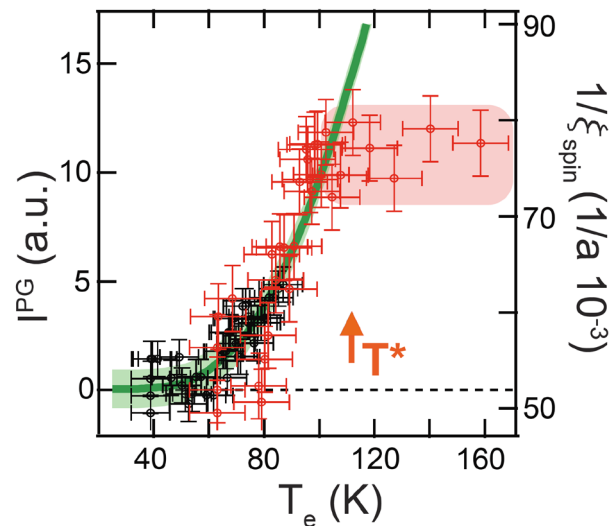
- T^* is a crossover temperature

$$T > T^* \rightarrow \xi_{\text{spin}} \sim 10-15 a$$

$$\Gamma(T^*) \sim 2\Delta_{\text{PG}} \sim 170 \text{ meV}$$

- The PG is filling, not closing

PG energy scale survives well above T^*

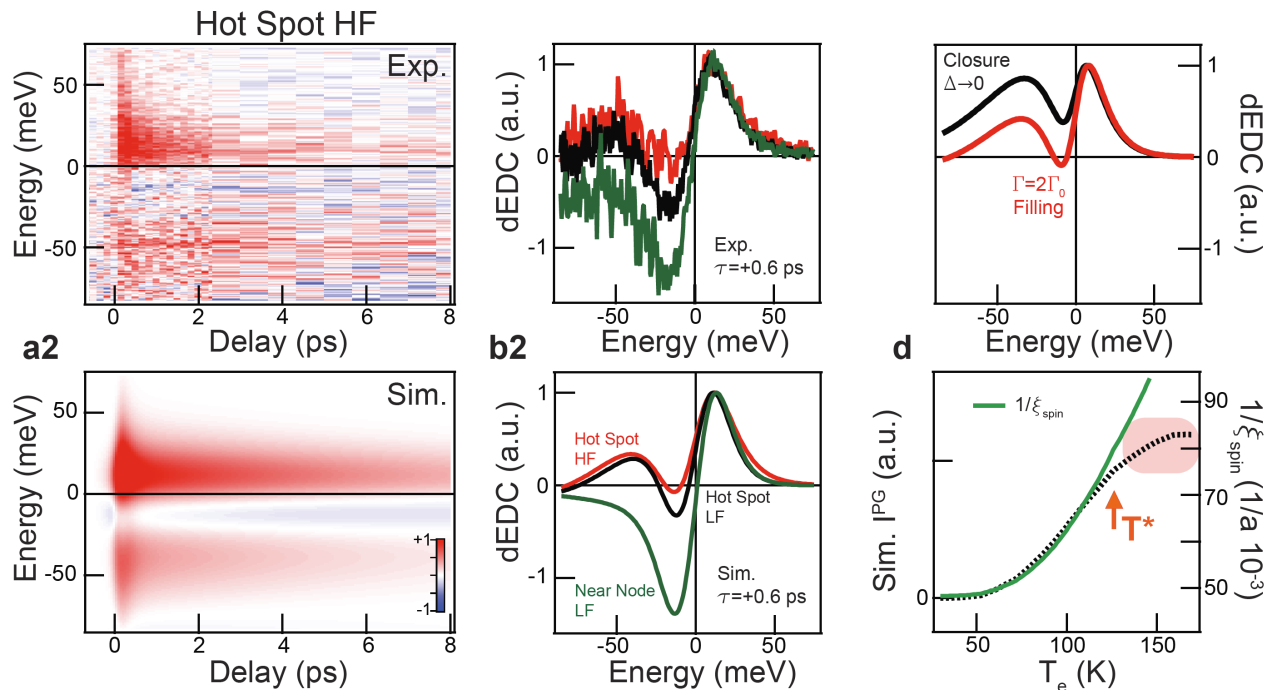


Emergence of pseudogap from short-range spin-correlations!

Pseudogap in Electron Doped Cuprates

$$G^{-1}(\mathbf{k}, \omega) = \omega - \epsilon_{\mathbf{k}} + i\eta - \frac{\Delta_{\text{PG}}^2}{\omega - \epsilon_{\mathbf{k}+\mathbf{q}} + i\Gamma}$$

Γ = spin-fluctuations induced



Direct determination of mode-projected electron-phonon coupling in the time-domain

GORDON AND BETTY
MOORE
FOUNDATION



Ketty (MengXing) Na
Fabio Boschini
Michael Schneider
Andrea Damascelli

Arthur K. Mills
Matteo Michiardi
Sergey Zhdanovich
David J. Jones



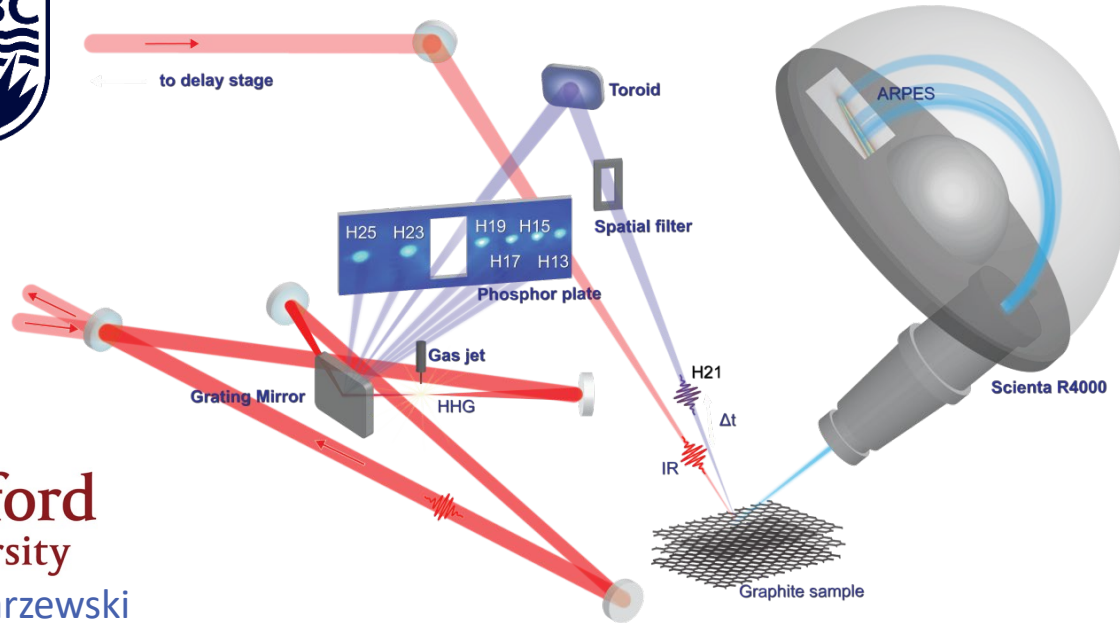
Ketty @ Flatiron



Stanford
University
Benjamin Nosarzewski
Thomas P. Devereaux

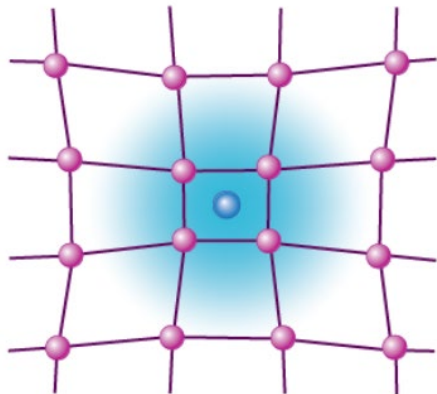
NC STATE
UNIVERSITY

Alexander F. Kemper



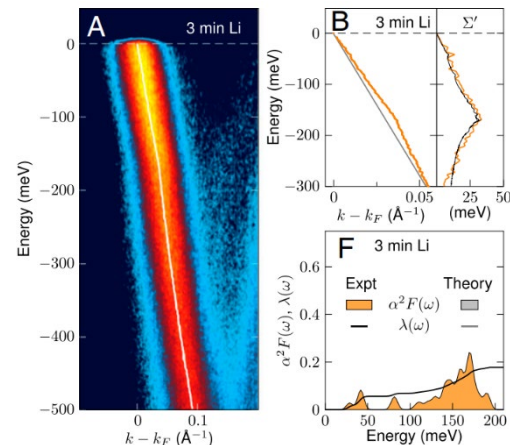
Na*, Mills*, et al.
Science **366**, 1231 (2019)

Electron-phonon coupling fundamental in many-body phenomena



Chevy et al. *Physics* 9, 86 (2016)

BCS superconductivity
or
Charge density waves



Ludbrook et al. PNAS **112** 11795 (2015)

How do we extract EPC?

- *Ab-initio* calculations
- Scattering experiments (particle or photon)
 - access EPC for specific phonon mode $\mathbf{q}$ integrated over all $\mathbf{k}$
- ARPES (kink analysis)
 - access EPC for specific $\mathbf{k}$ integrated over all modes $\mathbf{q}$

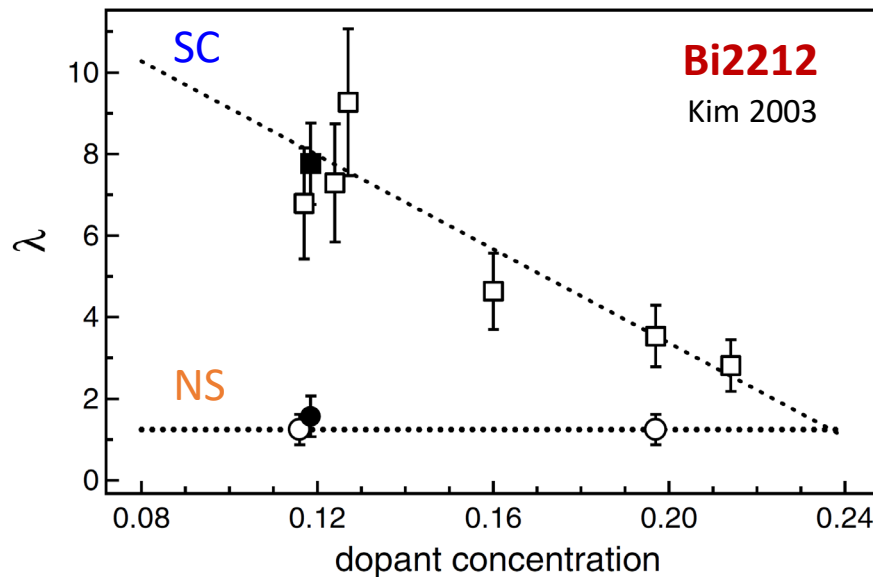
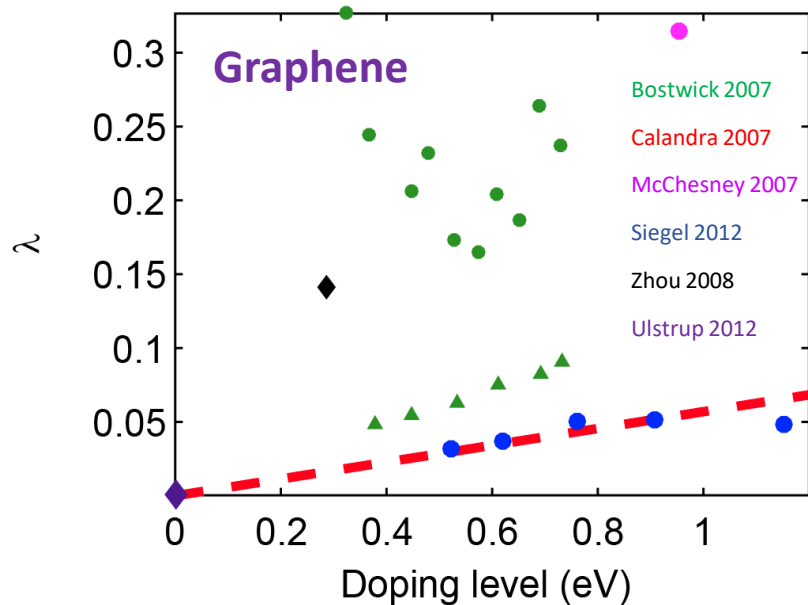
Extraction of Mass Enhancement Parameter λ from ARPES

ARPES:

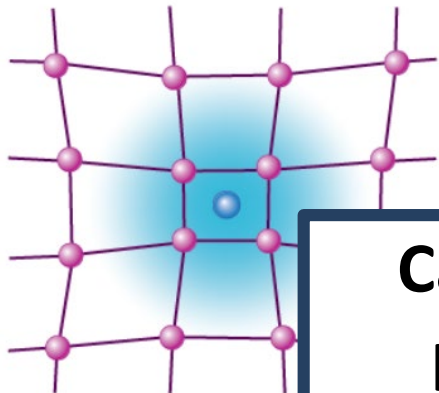
- How does system resolutions apply?
- What is the bare-band dispersion?
- How to account for contributions of electron-electron, electron-impurity?

$$\Sigma = \Sigma_{\text{el-el}} + \Sigma_{\text{el-ph}} + \Sigma_{\text{imp}}$$

- Huge doping-dependent increase in mass enhancement below T_c



Electron-phonon coupling fundamental in many-body phenomena



Chevy et al. *Physics* 9,

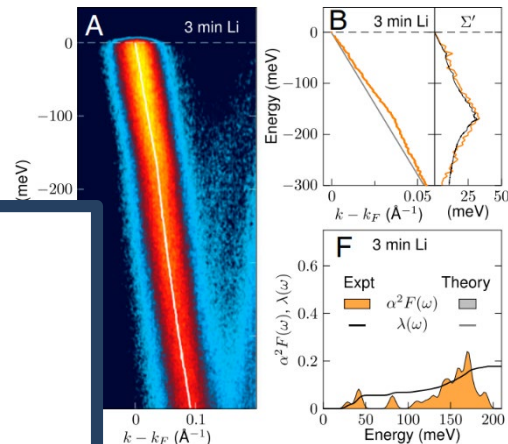
BCS superconductivity
or

Can we determine EPC for a particular phonon mode?

$$\frac{1}{\tau_q} = \frac{2\pi}{\hbar} \langle g_q^2 \rangle N$$

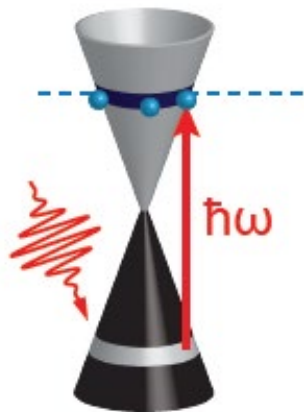
How do we e

- **Ab-initio** What about the time domain?
- **Scattering experiments (particle or photon)**
→ access EPC for specific phonon mode $\mathbf{q}$ integrated over all $\mathbf{k}$
- **ARPES (kink analysis)**
→ access EPC for specific $\mathbf{k}$ integrated over all modes $\mathbf{q}$

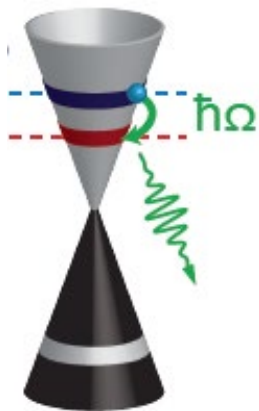


Brook et al. PNAS **112** 11795 (2015)

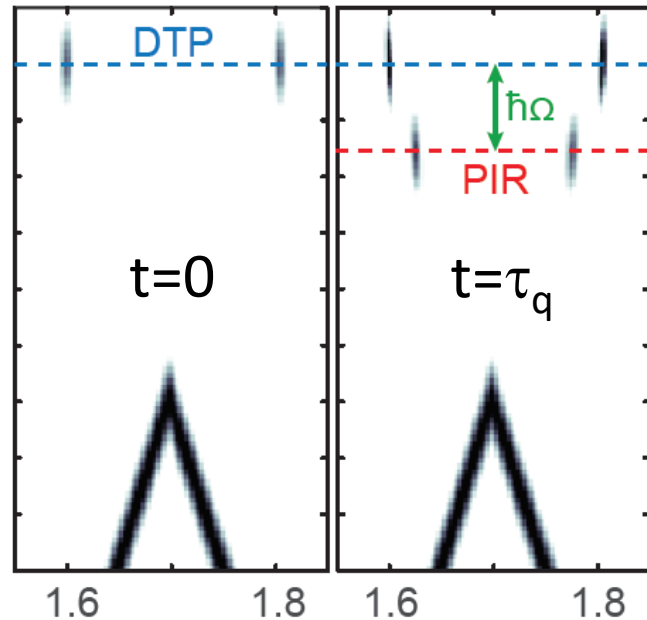
Toy-Model: Graphene Case with Coupling to Einstein Mode



Pump excitation



e-p scattering



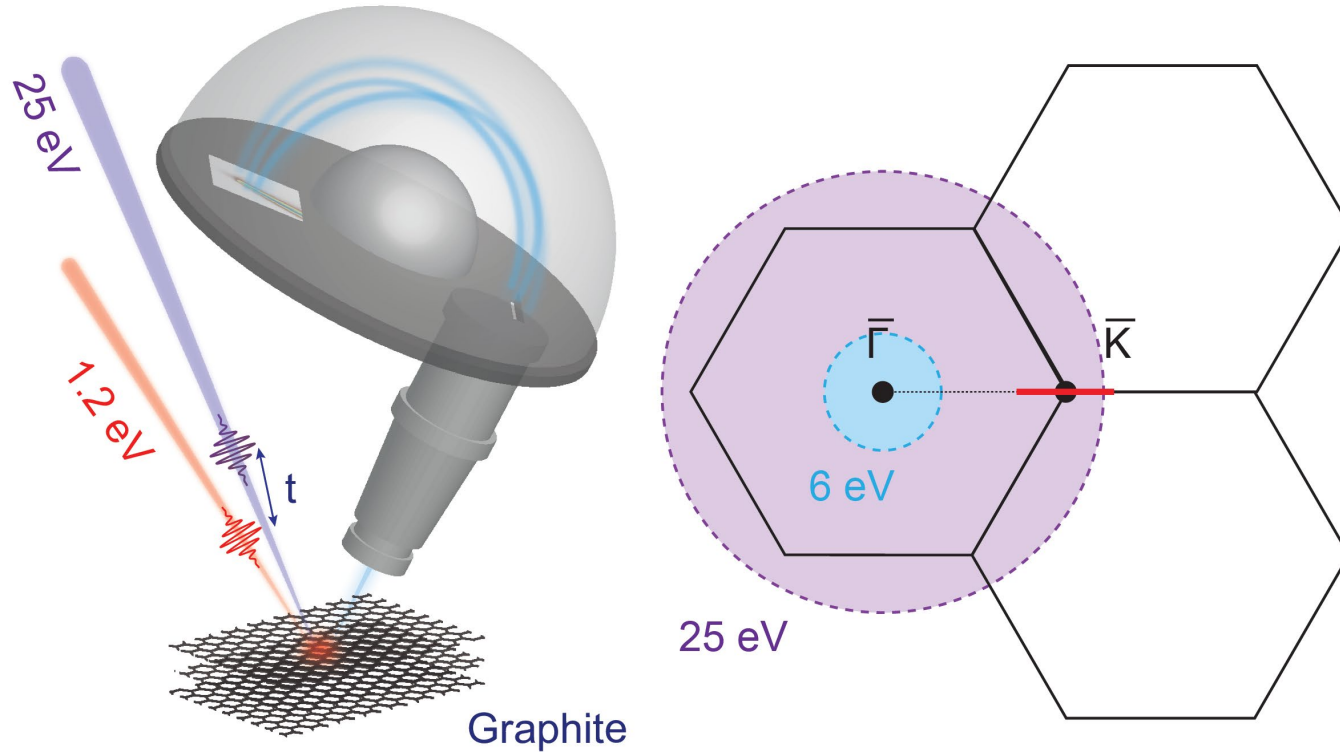
Mode-projected
e-p matrix element

Mode-projected
e-p coupling constant

For single Einstein mode $\mathbf{q}$

$$\frac{1}{\tau_q} = \frac{2\pi}{\hbar} \langle g_q^2 \rangle N = 2\pi \Omega_q \lambda_q N^*$$

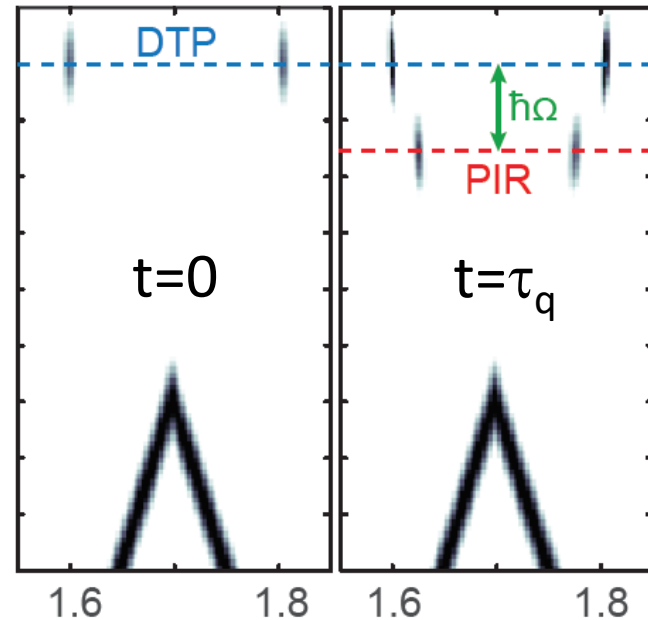
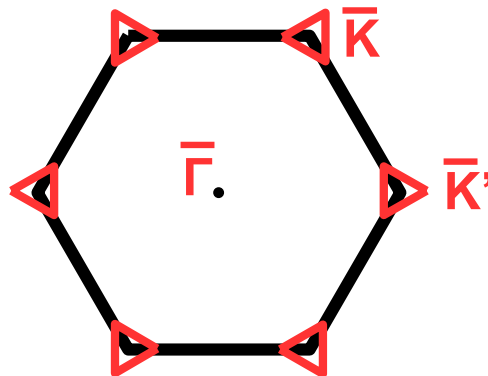
Laser Source: HHG Based on Femtosecond Enhancement Cavity



Mills et al. *SPIE Optics + Optoelectronics*. (2015)
Mills et al. *Journal of Physics B*, **45**, 142001, (2012)
Mills et al. *Rev. Sci. Instrum.* **90**, 083001 (2019)

Laser Source: HHG Based on Femtosecond Enhancement Cavity

- ➡ Photon energy >20 eV
- ➡ Phonon energy ~ 100 meV
- ➡ Scattering time ~ 100 fs



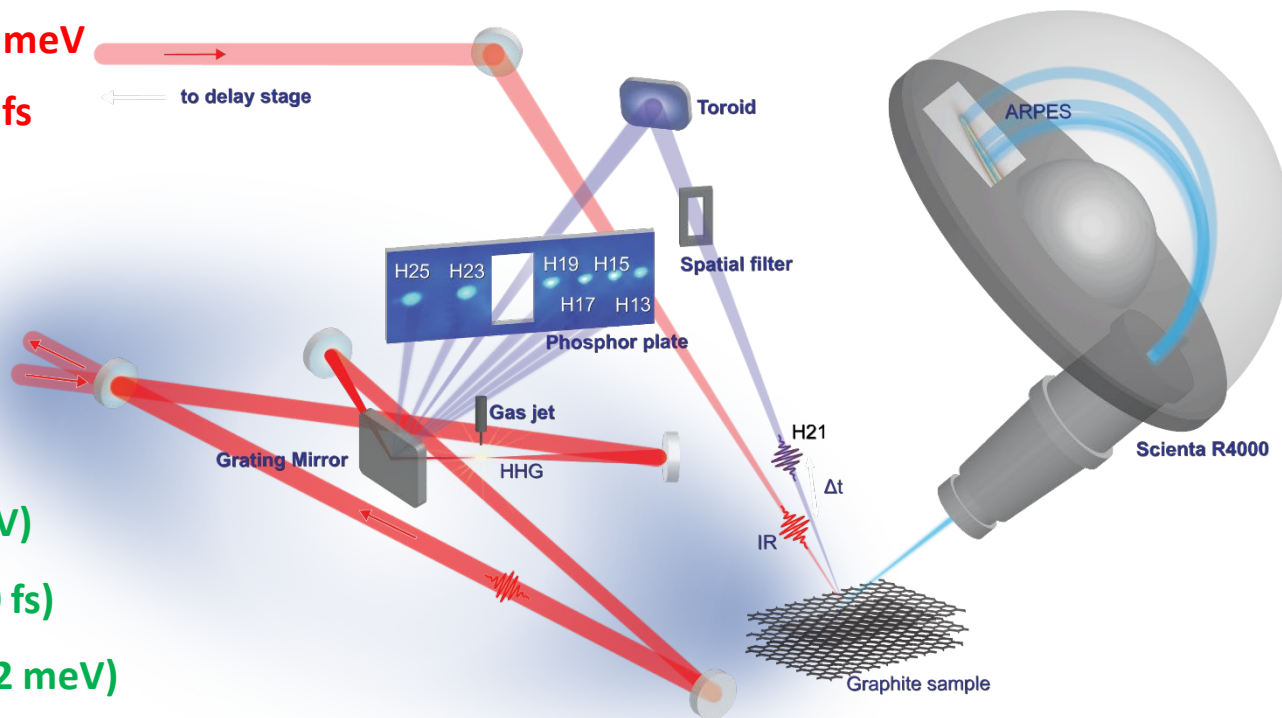
Laser Source: HHG Based on Femtosecond Enhancement Cavity

- ➡ Photon energy >20 eV
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- ➡ Scattering time ~ 100 fs

Cavity-based
high harmonic
generation

- ➡ Photon energy (25 eV)
- ➡ Time resolution (190 fs)
- ➡ Energy resolution (22 meV)
- ➡ Repetition rate (60 MHz)

➡ Needed to see small signatures!



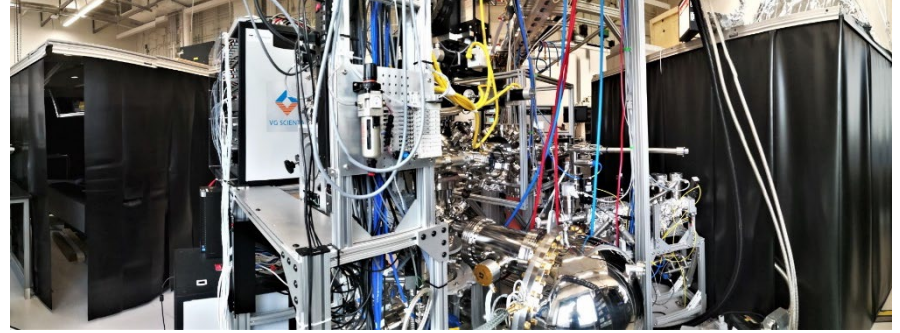
Na*, Mills*, et al.
Science **366**, 1231 (2019)

ARTEMIS at Rutherford Appleton Laboratory



- ARPES beamline endstation
- 30 fs pulse widths
- 10-80 eV photon energy
- 1 kHz with 10^9 photons/sec
- ~200 meV energy resolution

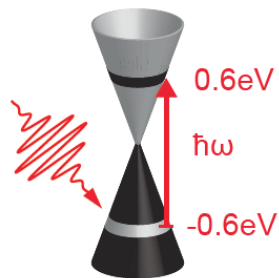
UBC – Moore Center for Ultrafast Phenomena



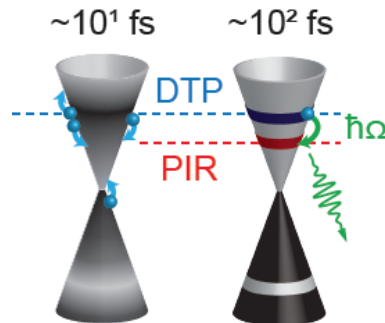
- Scienta R4000 Hemispherical Analyzer
- 190 fs time resolution
- 8-44 eV photon energy
- 60 MHz with 10^{12} photons/sec
- ~22 meV energy resolution

A different focus, a different approach!

Time-resolved ARPES Experiment on Graphite at the Zone Boundary



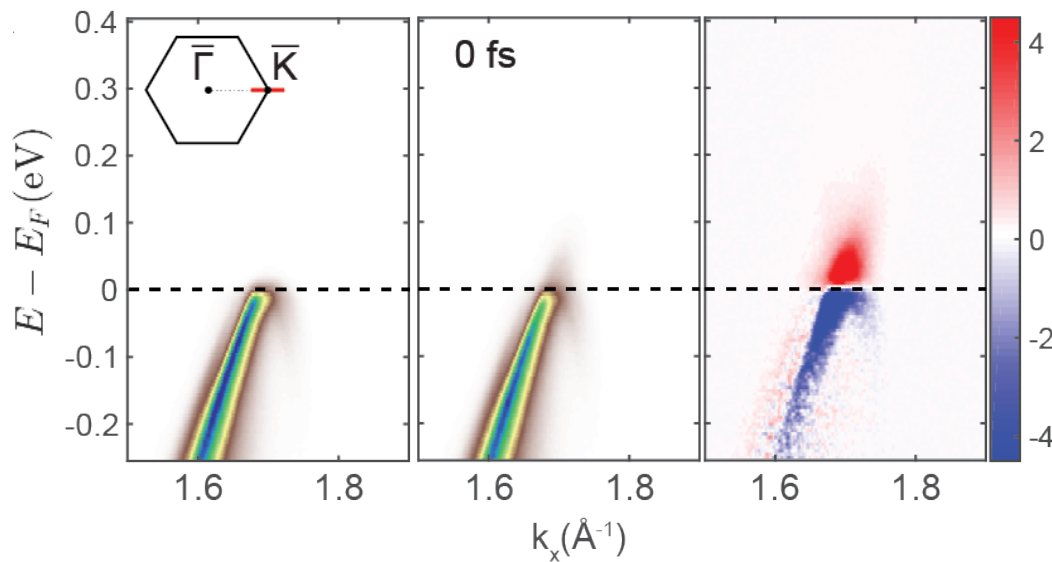
T=50K
Pump fluence: $20\mu\text{J}/\text{cm}^2$
Pump $\hbar\omega$: 1.2 eV
Pump Polarization: s
Probe Polarization: s



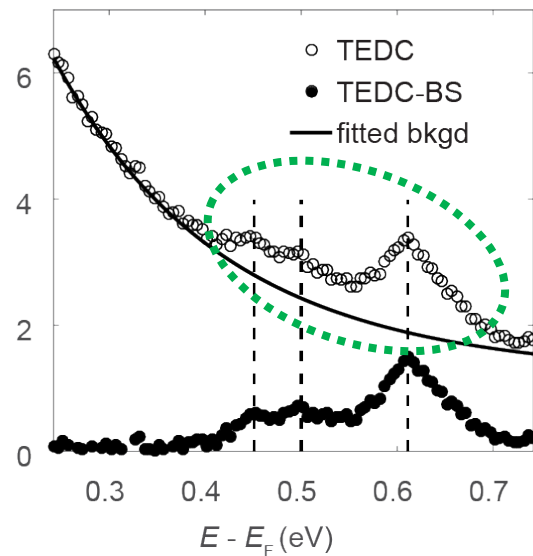
Efficient e-e covers
phonon-induced replica

Creates hot electron
background

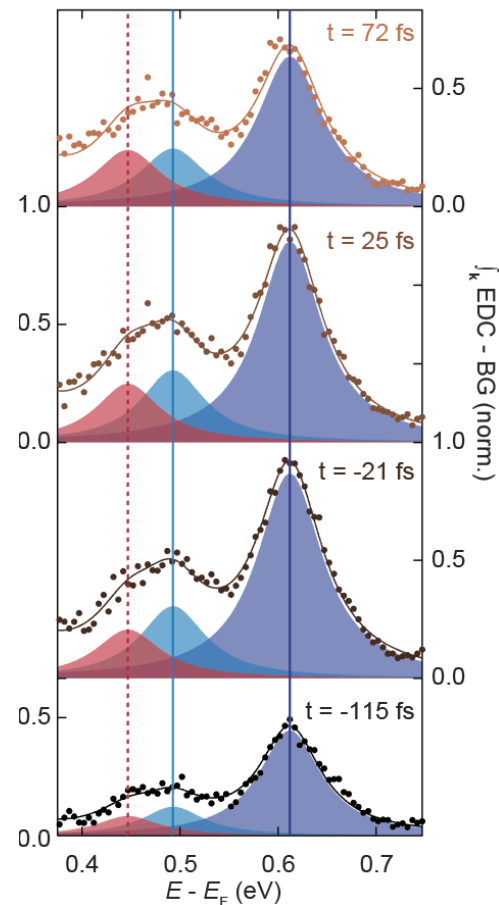
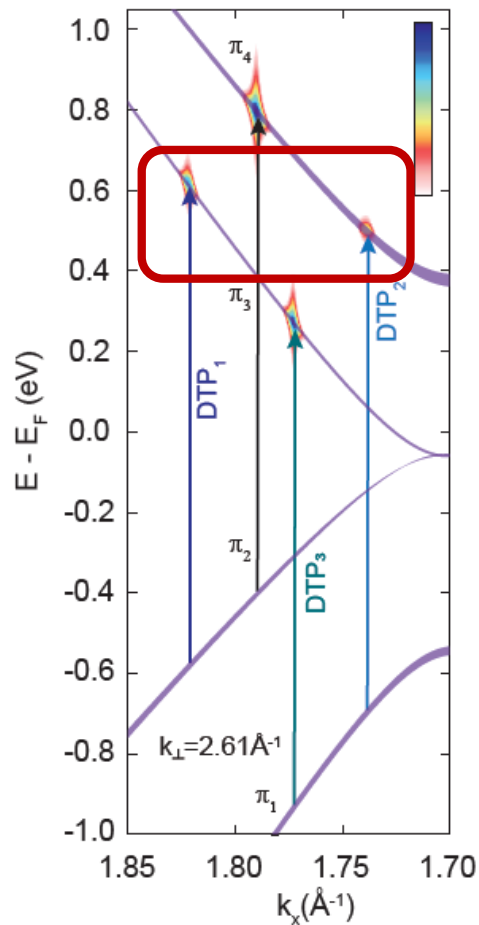
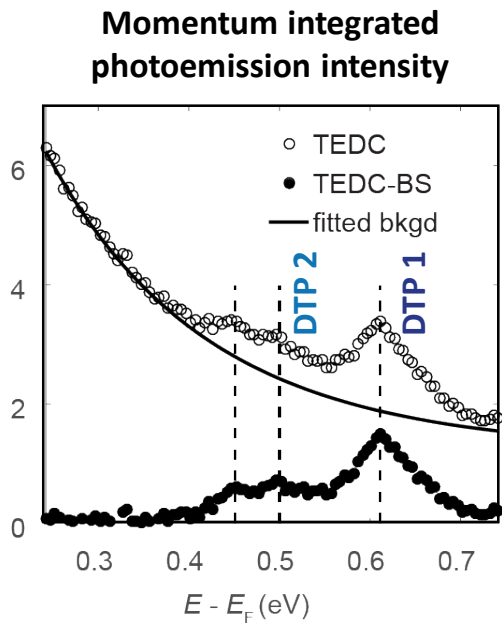
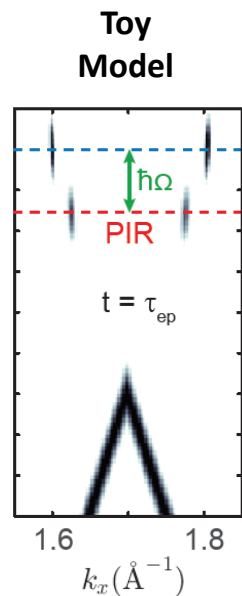
Pump excitation



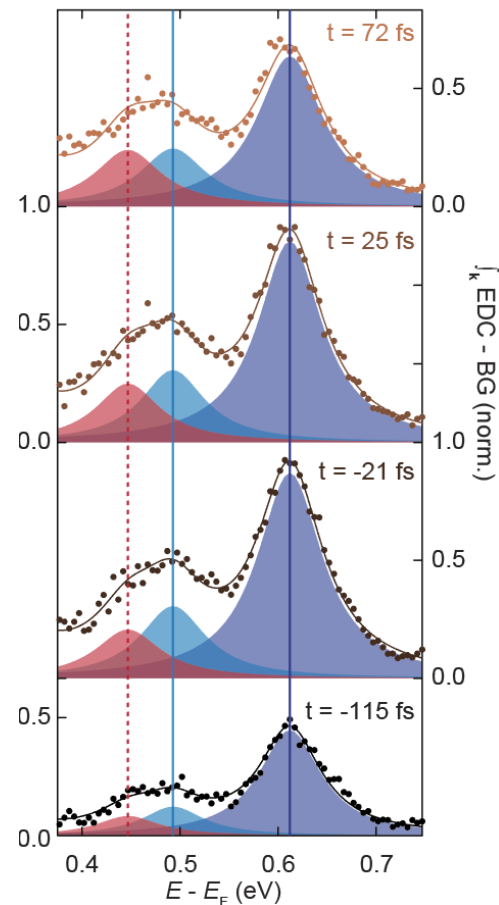
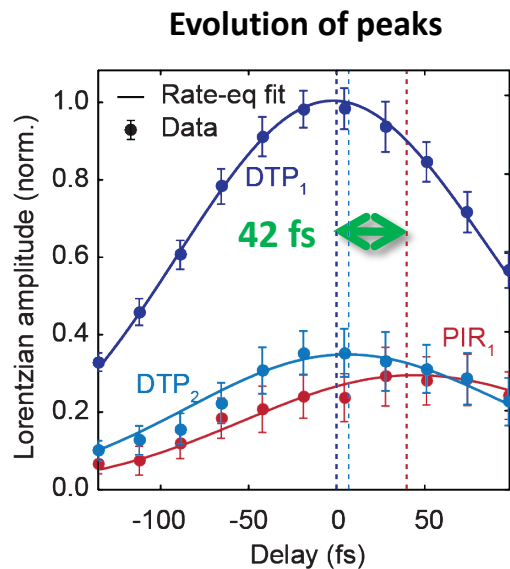
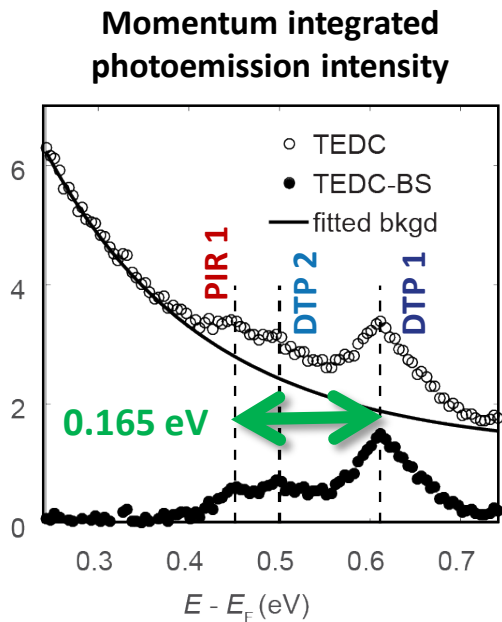
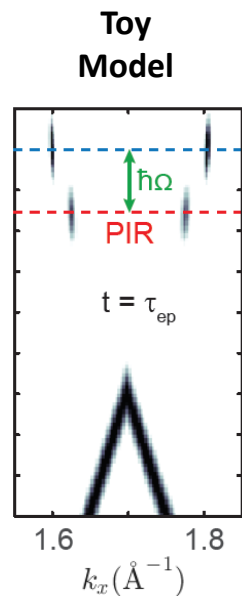
Momentum-integrated EDC



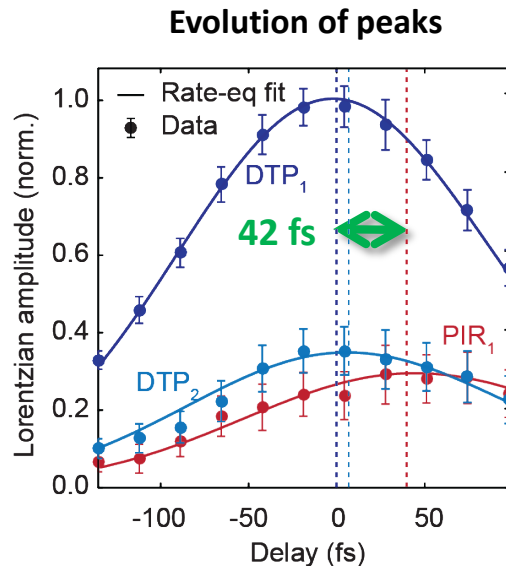
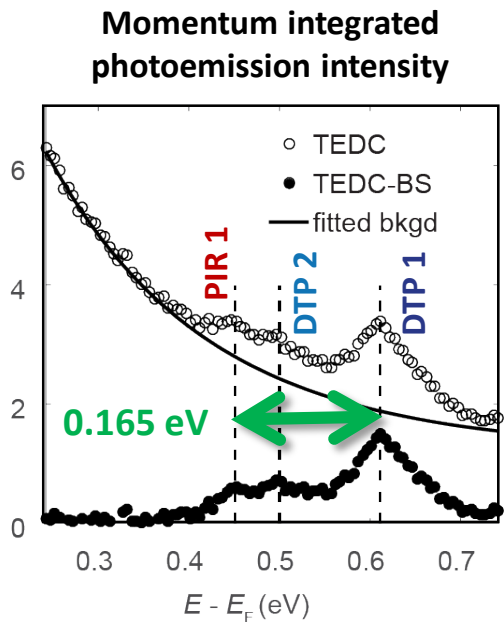
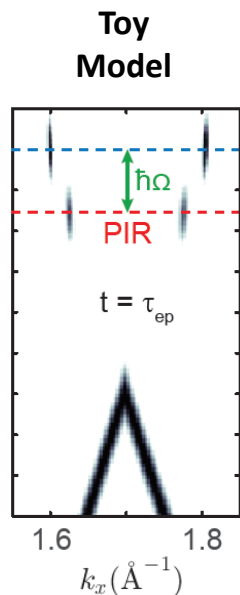
Time-resolved ARPES Experiment on Graphite at the Zone Boundary



Time-resolved ARPES Experiment on Graphite at the Zone Boundary



Time-resolved ARPES Experiment on Graphite at the Zone Boundary



Decay constants:

$$\tau_q = (174 \pm 35) \text{ fs}$$

$$\tau_{th} = (56 \pm 16) \text{ fs}$$

$$\frac{1}{\tau_q} = \frac{2\pi}{\hbar} \langle g_q^2 \rangle N(E - \hbar\Omega)$$

Measured:

$$\langle g_q^2 \rangle \cong 0.050 \pm 0.011 \text{ eV}^2$$

thermalization electron-phonon

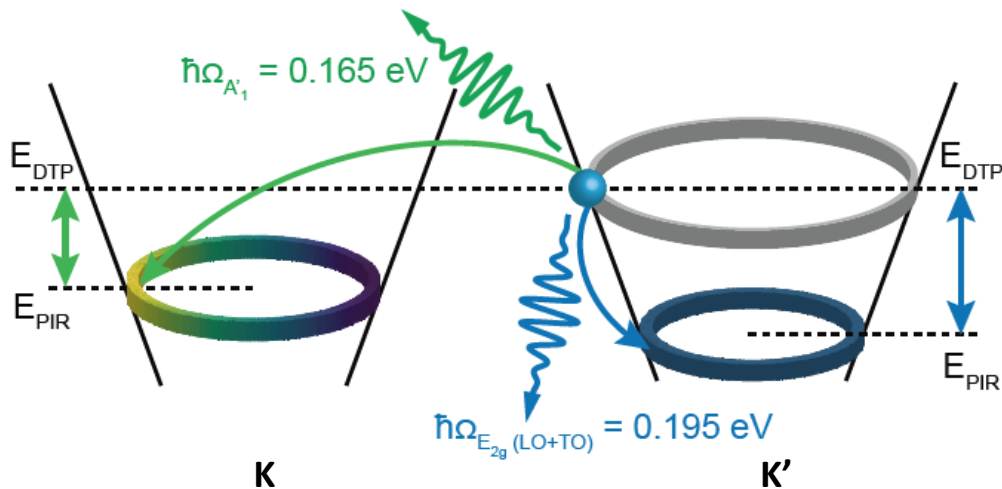
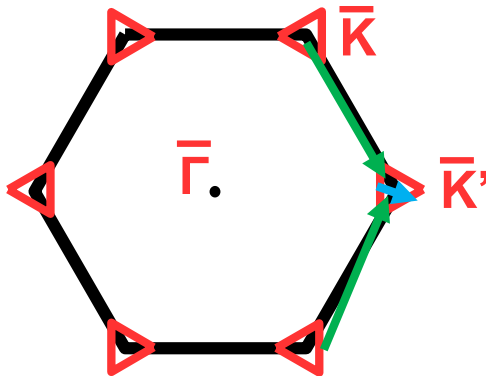
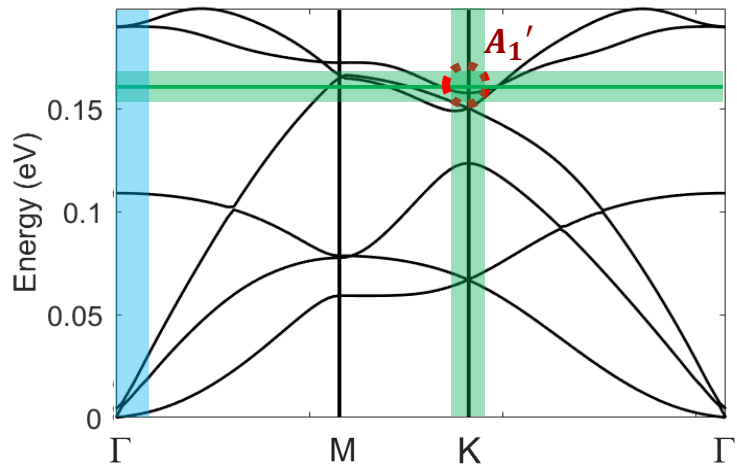
$$\begin{aligned} \frac{d}{dt} N_{\text{DTP}}(t) &= I_{\text{pump}}(t) - \frac{N_{\text{DTP}}(t)}{\tau_{\text{th}}(E_{\text{DTP}})} - \frac{N_{\text{DTP}}(t)}{\tau_q(E_{\text{DTP}})} \\ \frac{d}{dt} N_{\text{PIR}}(t) &= -\frac{N_{\text{PIR}}(t)}{\tau_{\text{th}}(E_{\text{PIR}})} - \frac{N_{\text{PIR}}(t)}{\tau_q(E_{\text{PIR}})} + \frac{N_{\text{DTP}}(t)}{\tau_q(E_{\text{DTP}})} \end{aligned}$$

Spectral weight transfer

Na*, Mills*, et al.
Science **366**, 1231 (2019)

Phonon Identification and Mode Projected Matrix Element

Phonon dispersion of Graphite



Phonon energy

$$\hbar\Omega = 0.165 \text{ eV}$$

Measured:

$$\langle g_{A_1'}^2 \rangle = 0.050 \pm 0.011 \text{ eV}^2$$

Calculated:

$$\langle g_{A_1'}^2 \rangle = 0.04 \text{ eV}^2$$

What about the Gamma mode?

Coupling is 50% that of the K mode

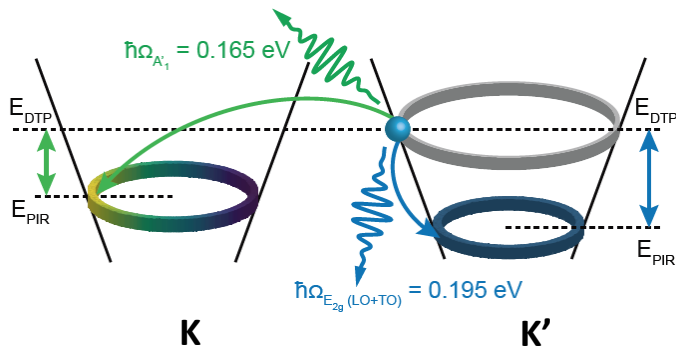
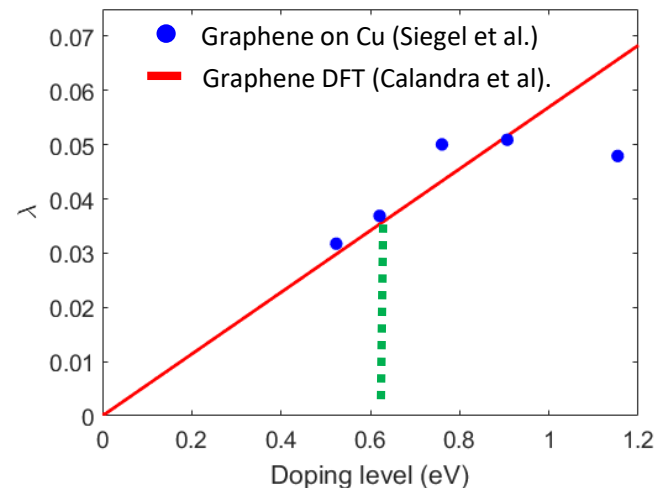
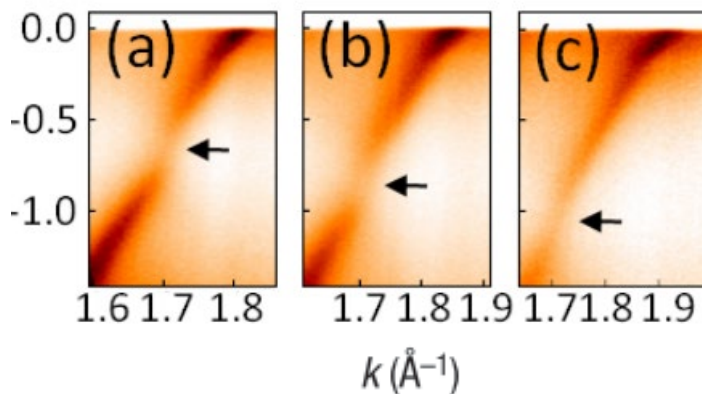
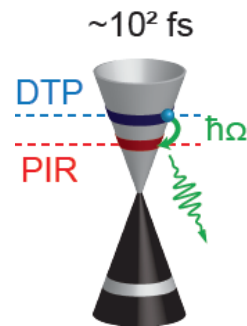
Na*, Mills*, et al.
Science 366, 1231 (2019)

Evaluation of Mode-Projected e-p Coupling Constant λ

Measured (graphite):

$$\langle g_{A_{1'}}^2 \rangle \cong 0.050 \pm 0.011 \text{ eV}^2 \rightarrow \lambda \approx 2\langle g^2 \rangle N(E_F)/(\hbar\Omega)$$

[Siegel et al. New Journal of Physics **14**, (2012)]



Comparison:

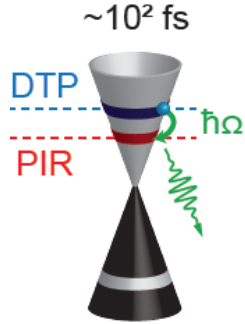
$$\lambda_{A_{1'}} \cong \lambda/2$$

We measure one of two strongly coupled modes

	Doped: 0.6 eV above CB max
Calandra et al.	$\lambda = 0.034$
Siegel et al.	$\lambda = 0.035$
Our result	$\lambda_{A_{1'}} = 0.018$

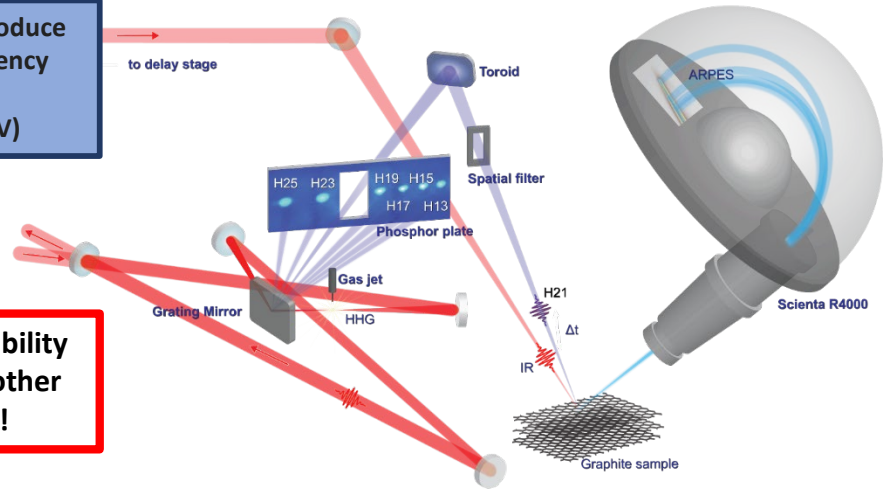
Future Directions and Novel Experiments

We've determined $\langle g_{A1'}^2 \rangle$ for the DTP at 0.6 eV,
what about other states?



OPO/DFG to produce
tunable frequency
FIR-MIR
(80-900 meV)

Increases flexibility
for studying other
materials!



Can we change **fundamental coupling constants** like $\langle g_{A1'}^2 \rangle$ by pumping? Or do we modify scattering through **fermionic and bosonic occupation**?

➡ Fluence dependence
➡ Coherent excitations of bosonic modes

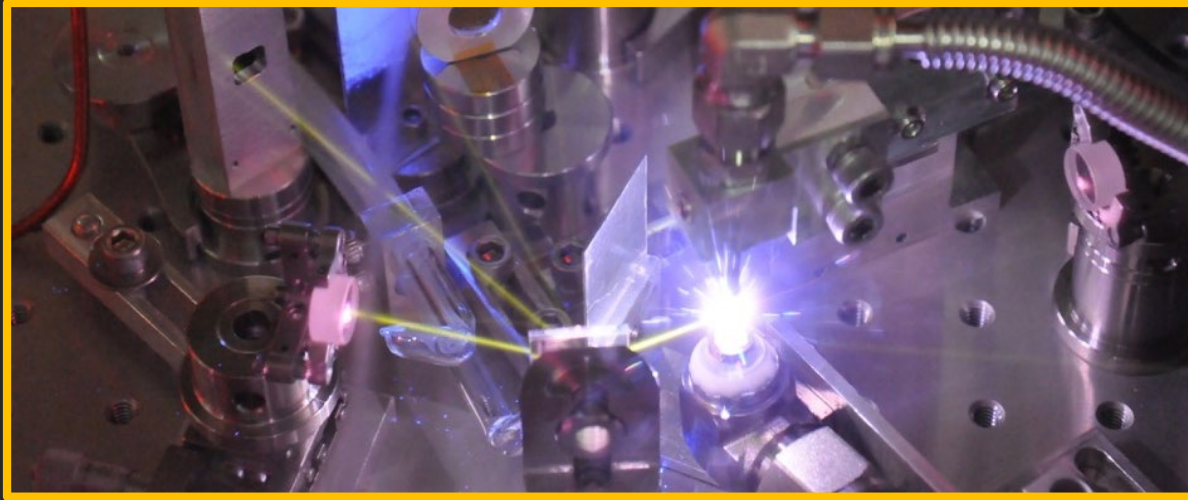
Are we sensitive to **modification of the density of states** through the scattering rate?

What are the fundamental coupling constants for superconductors?

How does pumping influence the SC gap?

$$\Gamma_{\text{emi}}(\epsilon, \Omega) = \frac{2\pi}{\hbar} \underbrace{|G_{\text{eph}}(\Omega)|^2}_{\text{green}} \underbrace{(1 + n(\Omega))}_{\text{red}} \underbrace{N(\epsilon - \Omega)}_{\text{blue}} \underbrace{(1 - f(\epsilon - \Omega))}_{\text{red}}$$

CONCLUSIONS



Exciting days lay ahead for TR-ARPES!