UBC-Moore Centre for Ultrafast Quantum Matter

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6.2 eV system

Transient Collapse high-Tc superconductivity in Bi2212

Relation pseudogap and spin correlations in NCCO

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

Determination mode-projected electron-phonon matrix element in graphite

Cavity-enhanced HHG for XUV TR-ARPES
Mills et al., RSI 90, 083001 (2019)
**TR-ARPES Bi2212**

Non-equilibrium technique (TR-ARPES)

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

**Common analysis approach:**
Study population/depletion dynamics

**ARPES intensity**

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

**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 \(A(k, \omega)\)

**Disentangle** \(A(k, \omega, \tau)\) & \(f(\omega, \tau)\) contributions
Phase stiffness in cuprates

Formation pairs. Pairing energy $E_p$

Onset of phase coherence

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

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

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$V_\theta \propto n_s(0)$

Emery and Kivelson
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 $2\Delta$

### Pairing strength:
Modification of $2\Delta$

### 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)  
Gar×osure vs. filling

\[ \frac{EDC_{\text{off}}(k_F)}{\int EDC_{\text{node}}(k)dk} \propto A_{\text{off}}(k_F, \omega) \]


Similar to TDOS method

\[ \phi = 36^\circ \]

No Pump
6 K

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

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

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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

Phase decoherence: mechanism

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

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

Antiferromagnetic order leads to band folding and hot spots

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?

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\[ G^{-1}(k, \omega) = \omega - \epsilon_k + i\eta - \frac{\Delta_{PG}^2}{\omega - \epsilon_{k+q} + i\Gamma} \]

\[ \Gamma = \text{spin-fluctuations induced} \]

Emergence of pseudogap from short-range spin-correlations!

Pseudogap in Electron Doped Cuprates

\[ G^{-1}(k, \omega) = \omega - \epsilon_k + i\eta - \frac{\Delta_{PG}^2}{\omega - \epsilon_{k+q} + i\Gamma} \]

\[ \Gamma = \text{spin-fluctuations induced} \]

- \( T^* \) is a crossover temperature
  
  \[ T > T^* \rightarrow \xi_{spin} \sim 10-15a \]
  
  \[ \Gamma(T^*) \sim 2\Delta_{PG} \sim 170\text{meV} \]

- The PG is filling, not closing
  
  PG energy scale survives well above \( T^* \)

Emergence of pseudogap from short-range spin-correlations!

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

$\Gamma = \text{spin-fluctuations induced}$
Direct determination of mode-projected electron-phonon coupling in the time-domain

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Michael Schneider
Andrea Damascelli

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

Electron-phonon coupling fundamental in many-body phenomena

How do we extract EPC?

- **Ab-initio calculations**
- **Scattering experiments (particle or photon)**
  \[ \rightarrow \text{access EPC for specific phonon mode } \mathbf{q} \text{ integrated over all } \mathbf{k} \]
- **ARPES (kink analysis)**
  \[ \rightarrow \text{access EPC for specific } \mathbf{k} \text{ integrated over all modes } \mathbf{q} \]
Extraction of Mass Enhancement Parameter $\lambda$ from ARPES

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

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

- Huge doping-dependent increase in mass enhancement below $T_c$

**Graphene**
- Bostwick 2007
- Calandra 2007
- McChesney 2007
- Siegel 2012
- Zhou 2008
- Ulstrup 2012

**Bi2212**
- Kim 2003

**SC**

**NS**

**Doping level (eV)**

**Dopant concentration**
Electron-phonon coupling fundamental in many-body phenomena

How do we extract EPC?

• Ab-initio calculations
• Scattering experiments (particle or photon)
  → access EPC for specific phonon mode $q$ integrated over all $k$

BCS superconductivity or Charge density waves

Can we determine EPC for a particular phonon mode?

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

What about the time domain?

$\tau_q \approx 2\pi \hbar g^2_q N$
For single Einstein mode $q$

$$\frac{1}{\tau_q} = \frac{2\pi}{\hbar} \langle g_q^2 \rangle N = 2\pi \Omega_q \lambda_q N^*$$
Laser Source: HHG Based on Femtosecond Enhancement Cavity

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

Na*, Mills*, et al.  
Science 366, 1231 (2019)
Laser Source: HHG Based on Femtosecond Enhancement Cavity

- Photon energy $>20$ eV
- Phonon energy $\sim 100$ meV
- Scattering time $\sim 100$ 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!

ARTEMIS at Rutherford Appleton Laboratory

- ARPES beamline endstation
- 30 fs pulse widths
- 10-80 eV photon energy
- 1 kHz with $10^9$ photons/sec
- $\sim 200$ 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
- $\sim 22$ meV energy resolution

A different focus, a different approach!
Momentum-integrated EDC

\( T = 50\, {\text{K}} \)

Pump fluence: 20\( \mu \text{J/cm}^2 \)

Pump \( \hbar \omega \): 1.2 eV

Pump Polarization: s

Probe Polarization: s

Efficient e-e covers phonon-induced replica

Creates hot electron background

Time-resolved ARPES Experiment on Graphite at the Zone Boundary
Time-resolved ARPES Experiment on Graphite at the Zone Boundary

Toy Model

Momentum integrated photoemission intensity

- $\hbar \Omega$
- PIR
- $t = \tau_{ep}$

$E - E_F$ (eV)

$E - E_F$ (eV)

$k_x (\text{Å}^{-1})$

$k_x (\text{Å}^{-1})$

$E - E_F$ (eV)

$f$, EDC - BG (norm.)

$E - E_F$ (eV)

$k_x = 2.61 \text{Å}^{-1}$

$\pi_1$

$\pi_2$

$\pi_3$

$\pi_4$
Time-resolved ARPES Experiment on Graphite at the Zone Boundary

**Toy Model**

- $\hbar \omega$
- PIR
- $t = \tau_{ep}$

**Momentum integrated photoemission intensity**

- DTP 1
- DTP 2
- PIR 1
- 0.165 eV

**Evolution of peaks**

- T1EDC
- T1EDC-BS
- Fitted bkgd
- DTP 1
- DTP 2
- Evolution at $t = 42$ fs

**Graphs**

- Lorentzian amplitude (norm.)
  - Rate-eq fit
  - Data
  - DTP 1
  - PIR 1

**Images**

- $k_z(\text{Å}^{-1})$
- $E - E_r$ (eV)
- Delay (fs)
- $E - E_r$ (eV)
- $t = 72$ fs
- $t = 25$ fs
- $t = -21$ fs
- $t = -115$ fs
Time-resolved ARPES Experiment on Graphite at the Zone Boundary

Toy Model

Momentum integrated photoemission intensity

Evolution of peaks

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:

\[ \langle g_q^2 \rangle \approx 0.050 \pm 0.011 \text{ eV}^2 \]

\[ \frac{d}{dt} N_{\text{DTP}}(t) = I_{\text{pump}}(t) - \frac{N_{\text{DTP}}(t)}{\tau_{th}(E_{\text{DTP}})} - \frac{N_{\text{DTP}}(t)}{\tau_q(E_{\text{DTP}})} \]

\[ \frac{d}{dt} N_{\text{PIR}}(t) = -\frac{N_{\text{PIR}}(t)}{\tau_{th}(E_{\text{PIR}})} - \frac{N_{\text{PIR}}(t)}{\tau_q(E_{\text{PIR}})} + \frac{N_{\text{DTP}}(t)}{\tau_q(E_{\text{DTP}})} \]

Spectral weight transfer

Phonon dispersion of Graphite

Phonon energy
\[ \hbar \Omega = 0.165 \text{ eV} \]

Measured:
\[ g_{A_1'}^2 = 0.050 \pm 0.011 \text{ eV}^2 \]

Calculated:
\[ g_{A_1'}^2 = 0.04 \text{ eV}^2 \]

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 $\lambda$

Measured (graphite): \[
\langle g^2_{A1} \rangle \cong 0.050 \pm 0.011 \text{eV}^2 \quad \Rightarrow \quad \lambda \approx 2\langle g^2 \rangle N(E_F) / (\hbar\Omega)
\]

Calandra et al. and Siegel et al. have reported the following values:

- Calandra et al. $\lambda = 0.034$
- Siegel et al. $\lambda = 0.035$

Our result is:

$\lambda_{A1'} \cong \lambda / 2$

We measure one of two strongly coupled modes.

Comparison:

<table>
<thead>
<tr>
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<th>Doped: 0.6 eV above CB max</th>
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<tbody>
<tr>
<td>Calandra et al.</td>
<td>$\lambda = 0.034$</td>
</tr>
<tr>
<td>Siegel et al.</td>
<td>$\lambda = 0.035$</td>
</tr>
<tr>
<td>Our result</td>
<td>$\lambda_{A1'} = 0.018$</td>
</tr>
</tbody>
</table>
We’ve determined $\langle g_{A1}^2 \rangle$ for the DTP at 0.6 eV, what about other states? 

$\sim 10^2$ fs

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?
CONCLUSIONS

Exciting days lay ahead for TR-ARPES!