Electronic Raman scattering in high Tc cuprate superconductors
Toward an understanding of the phase diagram

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Courtesy: M. Le Tacon, Squap, MPQ
Outline

Part 3

• Exploration of the superconducting state of cuprates
  - the symmetry of a superconducting gap
  - the emergence of 2 energy scales in the SC state
  - the question of one or two gaps?
  - the controversy on photo-emission data

• Density of Cooper pairs and fraction of coherent Fermi Surface:
  - Do we really need to invoke 2 gaps to explain the 2 energy scales?
  - How to reconcile transport and spectroscopy measurements: thermal conductivity, heat capacity

• Exploration of the normal state of cuprates:
  - electronic Raman response function in the normal state
  - observation of the pseudo gap state in cuprates
Outline

Part 4

• An attempt to give a global view of the cuprate phase diagram

• Differences and analogies between cuprates and pnictides superconductors (Yann Gallais)
  - introduction to iron pnictides superconductors
  - superconducting gap symmetry
  - superconductivity and spin density wave order
s-wave gap **versus** d-wave gap

**s-wave gap:** full gap

- **T = 0**
- **T ≠ 0 below Tc**

**d-wave gap:** nodes

- **Nodes**
- **d-wave more excited electrons only around nodes**

**s-wave few excited electrons**
Thermal conductivity

Note the exponential drop at low temp

\[ \kappa(T) \propto C_v(T) I_m(T) v_F \]

S-wave  
\[ \text{YBCO} \]

Yu, PRL 1992
Tunneling in Al/AlO/Al

From Nobel Prize lecture in 1973

Ivar Giaever 1973

From Nobel Prize lecture in 1973
Tunneling experiments Nb vs Cuprates

Pan et al. 1998
Renner and Fisher 1998
Maggio Aprile 1995

For a review:
See RMP 79, 2007, O. Fischer
Electronic Raman scattering process

Photon in

Photon out

Electron-hole pair created

energy of the excited electron,
Momentum area
Photo-emission (ARPES)

Extraction of one electron energy of the excited electron momentum

Excited electronic states from the Fermi sea
Temperature-dependent photoemission spectra from optimally doped Bi-2212 ($T_c=91$ K), angle integrated over a narrow cut at $(\pi,0)$. Inset: superconducting-peak intensity vs temperature. After Fedorov et al., 1999.
ARPES measurements on Bi-2212

Ding, Norman, et al., 1996
What is Electronic Raman response?
Mercurate family

Tetragonal structure

Hg-1201
Tc = 95 K

Hg-1212
Tc = 120 K

Hg-1223
Tc = 135 K

Tc = 165 K (30 GPa)
-108 °C

D. Colson, CEA

Hg
Ba
Ca
CuO₂ plane
Oxygen
diamond past polishing (one tenth microns)
Electronic Raman scattering

Raman shift \[ \omega_R = \omega_i - \omega_s \]

Polarizations \( E_i \) et \( E_s \)

Energy probe
resolution < 1 meV

Momentum probe
\( k_x, k_x=\pm k_y \)
Raman momentum probe in cuprate structure

Real space

\[ \gamma_{B_{1g}} \]
\[ x^2-y^2 \]

\[ \gamma_{B_{2g}} \]
\[ xy \]

Cu

O

k space

symmetry

\[ \gamma_{B_{1g}} \]
\[ x^2-y^2 \]

\[ \gamma_{B_{2g}} \]
\[ xy \]

Probe (\( \pi,0 \))

Anti-Nodes

Probe (\( \pi,\pi \))

Nodes

\( \Gamma \)

X

M
Electronic Raman Response

$d$ - wave superconducting gap

$\Delta = 1.2$
Probing the Superconducting state vs doping, $p$

- Strange metal
- SC state
- UD state
- OD state

$T_c^M = 95 \text{ K}$

AF state

Pseudo gap

Fermi liquid ?

Temperature

doping level

Doping level

Temperature

10 K

0
Probing the Superconducting state vs doping, $p$.

- **Strange metal**
- **Pseudo gap**
- **AF state**
- **SC state**
- $T_c^M = 95 \, K$ (Transition temperature)

Temperature vs doping level diagram:
- UD (Undoped)
- OD (Over-doped)

Diagram shows regions corresponding to different states and phase transitions.
Raman response in a superconductor

\[ \chi''_{AN,N}(0, \omega) \propto \frac{2\pi N_F}{\omega} \text{Re} \left( \frac{(Z\Lambda)_k \gamma_{AN,N}^2 \Delta_k^2}{(\omega^2 - 4\Delta_k^2)^{1/2}} \right) \]

\[ \Delta_k \approx \omega \gamma \chi''(\omega) \]

\[ 2\Delta_k \]

M. V. Klein, S.B. Dierker PRB 29(84)
Mercurate family

**Tetragonal structure**

Hg-1201

Tc = 95 K

Hg-1212

Tc = 120 K

Hg-1223

Tc = 135 K

Tc = 165 K (30 GPa)

-108 °C

D. Colson, CEA
Anti-Nodal Response
In the superconducting state

\[ \Gamma (\pi,0) (\pi,\pi) \]

\[ B_{1g} \]

doping decrease

\[ \chi''(\omega) \text{ (a.u)} \]

M. Le Tacon
Nature Physics
2, 537, 2006

Raman shift (cm\(^{-1}\))
Electronic Raman Response

$d$ - wave superconducting gap

\[ \Delta_{AN}/\Delta_N = 1.2 \]

M. V. Klein, S. B. Dierker (84)
T. P. Devereaux, D. Einzel (95)
Nodal Response
In the Superconducting state

\((\pi,0)\) \((\pi,\pi)\)

\(B_{2g}\)

(doping decrease)

10 K

2 qsp dynamics

Raman shift \((cm^{-1})\)

\(0\) \(200\) \(400\) \(600\) \(800\)

Und. 89 K

89 K

297 cm\(^{-1}\)

Und. 86 K

80 K

370 cm\(^{-1}\)

Und. 78 K

100K

360 cm\(^{-1}\)

Und. 63 K

100K

230 cm\(^{-1}\)

Ov. 42 K

50 K

170 cm\(^{-1}\)

Ov. 70 K

80 K

297 cm\(^{-1}\)

Ov. 92 K

100K

400 cm\(^{-1}\)

Opt. 95 K

70K

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2 Energy Scales in the Superconducting state of Hg-1201

W. Guyard et al. PRB 77, 24524, 2008
Do not make confusion between the energy and the temperature phase diagram!
Bi-2212 single crystal

Bi - 2212
\[ T^{\text{max}_c} = 92 \text{ K} \]

G. Gu, BNL, USA
2 energy scales in the SC state of UD cuprates

Bi-2212

\[ \chi''(\omega, T) \text{ (cts.s}^{-1}.\text{mW}^{-1}) \]

\[ \omega \text{ (cm}^{-1}) \]

\[ p = 0.10 \]  \[ T_c = 87K \]
\[ p = 0.12 \]  \[ T_c = 75K \]
\[ p = 0.14 \]  \[ T_c = 87K \]
\[ p = 0.19 \]  \[ T_c = 75K \]
\[ p = 0.21 \]  \[ T_c = 75K \]

\[ \omega \text{ (cm}^{-1}) \]

\[ B_{1g} \]
\[ B_{2g} \]
2 energy scales in the SC state of UD cuprates by ERS

S. Blanc et al. PRB 89 2009
W. Guyard et al. PRB 77 2008
Two energy scales story

G. Deutscher, Nature 99

- Giaever scale
- ASJ scale

- Insulating barrier
- $\Delta_p$: first excited state
- $\Delta_c$: excitation gap
- $\Delta$: energy range where single particle states are missing
- $\Delta$: energy range beyond which the pair amplitude return to zero
Observations of two energy scales by several techniques

\[ \frac{\omega_{AN}}{\omega_N / T_c^\text{max}} \]

\[ T < < T_c \]

Huefner, Damacelli Rep. Prog. Phys. 08

M. Le Tacon et al. Nat.Phys.06

**HC**: Loram, Tallon , **ERS**: R.Hackl, Sugai, Le Tacon.. **ARPES**: Mesot, Tanaka, Kondo/Kaminski, Borisenko, **STM**: Boyer, Gomes, Hanaguri/Davis…
“Two Gaps Make a High Temperature Superconductor?”

Huefner, Damacelli  Rep. Prog. Phys. 08

A. Millis, Science 314, 1888, 06
Arpes in Bi-compounds: no consensus...


U. Chatterjee et al. , Nat. Phys. 2010
films: Tc = 33K(UD), 80K(OPT)
crystals: 69K (UD).
2 gaps against 1 gap

La$_{2-x}$Sr$_x$CuO$_4$

$X = 0.145$

$T_c = 36$ K

Slightly UD

Nodes

La$_{2-x}$Sr$_x$CuO$_4$

$X = 0.15$

$T_c = 37$ K

Opt. Doped

PRL, 101, 2007 M. Shi et al.

PRL 99, 2007, Terashima
Probing the Superconducting state vs doping, $p$
Electronic Raman scattering in a Superconductor

- \((u_k v_k)^2\) condensation probability of the Cooper Pairs (de Gennes, Superconductivity of metals and Alloys, 66)

- \(\sum (u_k v_k)^2\) density of Cooper Pairs around \(E_F\) as a gap is opening (Legett, Quantum Liquids in Condensed Matter systems, 2006)
Raman response function

\[ \chi''_R(q, \omega, T) \propto \int \frac{dt}{2\pi} e^{i\omega t} \langle \hat{\rho}_q(t)\hat{\rho}^+_q(0) \rangle_T \]

In the BCS framework:

\[ \chi''(\omega) = \pi \sum_k (\gamma^\mu_k)^2 \tanh \left( \frac{E_k}{2k_B T} \right) \frac{|\Delta_k|^2}{E_k^2} \delta(\omega - 2E_k) \]

As T tends to zero,

\[ \int \chi''(\omega) d\omega = \pi \sum_k (\gamma_k^\mu)^2 \frac{|\Delta_k|^2}{E_k^2} \quad \omega = 2\Delta_k \]

\[ \frac{|\Delta_k|^2}{E_k^2} = 4(u_k v_k)^2 \]
Area of the peak : density of Cooper pairs

\[ \chi''(\omega) \propto \sum_k (u_k v_k)^2 \]
Temperature dependence of $B_1g$ and $B_2g$ peaks

$\chi''(\omega, T)$ (cts.s$^{-1}$.mW$^{-1}$)

Normalized Peak Area

B1g & B2g peaks are coherent excitations of the SC state

$\text{Bi-2212}$

Normalized Peak Area

$T/T_c$
Loss of AN coherent superconducting peak

Bi-2212

\[ \Gamma \]

\( B_{1g} \)

\( B_{2g} \)

Anti-Nodes

Over-doped

Nodes

Under-doped
Quantitative Raman measurements

- Easily cleaved
- Large homogeneous surface (mm$^2$)
- Optical constants (ellipsometry)

Bi-2212

$T_{\text{max}} = 92 \text{ K}$

G. Gu, BNL, USA

S. Blanc et al., 09

Change in intensity less than 5% for two samples with the same doping level

Raman response $\chi''(\omega)$ (cts/s.mW)

Optical response

Bi-2212

$293 \text{ K}$

$514.52 \text{ nm}$

B$_{2g}$
Loss of coherent AN Peak

S. Blanc et al. PRB 80, 094504 Rapid Comm. 2009

Superconductivity is robust at the nodes

Under-doped
Loss of coherent Anti-Nodal superconducting peak

Summary of our experimental findings:

- 2 energy scales in the SC state of UD cuprates varying in opposite manners with U-doping.
- Both are coherent excitations of the SC state and disappear at Tc.
- Coherent Bogoliubov quasiparticles are reduced in the A-N region with U-doping.

Are you able to explain the 2 energy scales in a one-gap scheme?
Loss of coherent quasi-particles in a one gap scheme

QPSW decrease

QPSW is preserved

Fermi surface

Strongly overdoped

\[ \phi = 45^\circ \]

\[ v_\Delta \propto \Delta_{\text{max}} \]

\[ \Omega_{B1g} \quad \Omega_{B2g} \]

\[ \gamma_{B2g} \]

\[ Z(\phi), \Delta(\phi) \]

\[ \phi \text{ in degrees} \]

\[ \omega / \Delta_0 \]
Loss of coherent quasi-particles in a one gap scheme

\[ \chi_{AN,N}(0,\omega) \propto \frac{2\pi N_F}{\omega} \text{Re} \left( \frac{(Z_{\phi})_{k\phi}^{AN,N} \Delta_k^2}{(\omega^2 - 4\Delta_k^2)^{1/2}} \right) \]
Loss of coherent quasi-particles in a one gap scheme

![Fermi surface diagram]

- QPSW decrease
- QPSW is preserved

Strongly overdoped
Weakly underdoped
Underdoped

\[ \chi_{AN,N}^{\alpha} \propto \frac{2\pi N_F}{\omega} \text{Re} \left( \frac{|Z^\alpha|^2}{(\omega^2 - 4\Delta^2)^{1/2}} \right) \]

- \( \Omega_{B1g} \)
- \( \Omega_{B2g} \)

\( p=0.16 \)
\( p=0.12 \)
Loss of coherent quasi-particles in a one gap scheme

QPSW decrease

QPSW is preserved

Fermi surface

\[ \Phi = 45^\circ \]

Strongly overdoped
Weakly underdoped
Underdoped
Strongly underdoped

\[ \chi_{AN,N}^{\omega}(0,\omega) \propto \frac{2\pi N_F}{\omega} \text{Re} \left\langle \frac{\Delta^2}{\omega^2} \right\rangle_{SF} \]

\[ 2\Delta(\phi)/\Delta_0 \]

\[ Z(\phi), \Lambda(\phi) \]

\[ \phi \text{ in degrees} \]

\[ \omega / \Delta_0 \]
Doping Evolution of the Slope of the B$_{2g}$ peak

$\alpha \propto \frac{(Z\Lambda)^2}{N\nu_{\Delta}} \approx Cste$

in the doping

Range of $p \sim 0.1 - 0.16$

S. Blanc et al. PRB 80, 094504 Rapid Com. 2009
Loss of coherent quasi-particles in a one gap scheme

Strongly overdoped
Weakly underdoped
Underdoped
Strongly underdoped
Fractions of coherent Fermi surface

\[ f_c \propto \Delta \propto \Delta_{\text{max}} \] (Blanc et al.)

\[ f_c^{\text{Arpes}} \] (Kanigel et al.)

\[ f_c^{\text{HC}} \] (H.H.Wen et al.)

\[ \hbar \Omega_{B2g} \propto f_c \nu_{\Delta} \]

\[ \hbar \Omega_{B2g} \propto k_B T_c, \nu_{\Delta} \propto \Delta_{\text{max}} \]

\[ k_B T_c \propto f_c \Delta_{\text{max}} \]

S. Blanc et al.
Antinodal quasi-particles

H. Ding in PRL 2001
« the so called Coherent and Excitation gaps »

only one gap in reality

G. Deutscher, Nature 99

Giaever scale

Insulating barriere

first excited state

Pair Breaking Peak energy

energy range where single particle states are missing

G. Deutscher, Nature 99

energy range on which the Cooper pairs flow

ε

Δc

Δp

B_{1g}

f_c

Δ
Fractions of FS and a single d-wave gap

\[ \frac{dI}{dV} \propto N_F \left( Z(\phi) \text{Re} \frac{eV - i\Gamma}{(eV - i\Gamma)^2 - \Delta^2(\phi)} \right)_{FS} \]

S. Blanc et al.

K. McElroy et al. PRL 2005
CONCLUSION

Fraction of coherent Fermi Surface

- explains the existence of the 2 energy scales in the SC state

- controls $T_c$ such as $T_c \propto f_c \Delta_m$

- Consistent with: Raman, « Arpes », Tunneling and transport measurements
Cuprates phase diagram hole doped

- \( T_N \)
- \( T^* \)
- \( T_C \)
- PseudoGap
- AF Mott insulator
- 2 distinct regimes
  - In the SC states

Hole doping

\( p^* \)

\( 0 \) \( 0.05 \) \( 0.16 \) \( 0.27 \)
Cuprates phase diagram hole doped

Temperature

AF Mott insulator

$T_N$

Hole doping

2 scales

d-wave superconductor

1 scale

$T^*$

$T_c$

$p^*$

$p$
Cuprates phase diagram hole doped

Temperature

Hole doping

$T_N$

Superconductivity developps
Partially on the Fermi surface

$T_c$

d-wave superconductor

Superconductivity developps
on full Fermi surface

$p^*$
Normal state
and Pseudo-gap
Cuprates phase diagram hole doped

Temperature

AF Mott insulator

Normal Metal $\rho \propto T^2$

$T_N$

$T_C$

$T^*$

Pseudogap

$\rho \propto T$

$d$-wave superconductor

Hole doping $p^*$

$0$ $0.05$ $0.16$ $0.27$ $p$
Raman observation of the Pseudo-Gap

Comparison between the Raman PG and others technics

ERS

\[ \chi''(\omega) \text{ (u.a.)} \]

Symmetrized Raman shift (cm\(^{-1}\))

-1000 -500 0 500 1000

B\(_{1g}\) A-Nodal
Hg-1201

\( p = 0.16 \)

UD 95 K

S

N

RT

Bi-2201

\( T_c = 29\text{K} \)

ARPES

Energy (meV)

0 20 40

T. Kondo et al. Nature 457, 09

W. Guyard et al. not yet published, 09
Analogy between Raman response function and optical conductivity

Shastry-Shraiman relation (PRL 65, 1997)
(from Kubo formula)

\[
\text{Im } \chi(\Omega) \propto \Omega \text{ Re } \sigma(\Omega)
\]

\[
\varepsilon_0 \omega_{pl}^2 \tau \frac{1}{1 + \left(\Omega \tau\right)^2}
\]

\(\tau\) Life time of qsp

\(\times \Omega\)
The out-of-plane hopping which is modulated by the in-plane momentum in such a way that it is strongly governed by qps located along the BZ axes

\[ t_{\perp}(k) = t_{\parallel}0 \left[ \cos(k_x) - \cos(k_y) \right] \]