Coherent Charge Fluctuation Spectroscopy: A New Tool to Investigate the Pairing Wave Function

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Raman Scattering

\[ E = E_0 e^{-i\omega_L t} \]

\[ P = \chi E \quad \varepsilon = 1 - 4\pi\chi \]

\[ \chi = \frac{\sigma}{i\omega} \]

\[ \chi = \chi_0 + \frac{\partial \chi}{\partial \xi} \xi + \frac{\partial \chi}{\partial \xi^*} \xi^* \]

Raman tensor

\[ P = \left( \chi_0 e^{-i\omega_L t} + \frac{\partial \chi}{\partial \xi} \xi e^{-i(\omega_L + \omega_{ph}) t} + \frac{\partial \chi}{\partial \xi^*} \xi^* e^{-i(\omega_L - \omega_{ph}) t} \right) E_0 \]
Raman Scattering

\[ H = H_{ph} - \frac{1}{2} \mathbf{E}(t) \cdot \frac{\partial \chi(\omega_L)}{\partial \xi} \cdot \mathbf{E}(t) \hat{\xi} \]

\[ \hat{\rho} = \frac{\partial \chi}{\partial \xi}(\omega_L) \hat{\xi} \]

\[ \frac{d\sigma}{d\Omega d\omega} = \frac{\omega_s^4 V^2}{(4\pi)^2 c^4} \sum_\nu \left| \langle 0 | \hat{e}_s \cdot \hat{\rho} \cdot \hat{e}_l | \nu \rangle \right|^2 \delta(\omega - \omega_\nu) \]

FIG. 10. Incident wavelength dependence of the Raman spectra in La$_2$CuO$_4$ at 30 K.
Coherent Excitation of phonons by Impulsive Stimulated Raman Scattering

Merlin ssc 1997, Stevens, Kuhl, Merlin PRB 2002

\[ H = H_{ph} - \frac{1}{2} \mathbf{E}(t) \cdot \frac{\partial \chi(\omega_L)}{\partial \xi} \cdot \mathbf{E}(t) \hat{\xi} = H_{ph} - F(t) \hat{\xi} \]

\[ H_{ph} = \frac{1}{2} \Pi^2 + \frac{1}{2} \omega^2_{ph} \xi^2 \]

\[ \dddot{\xi} + \omega^2_{ph} \xi = F(t) \]

\[ \xi(t) = \int_{-\infty}^{t} dt' \frac{\sin[\omega_{ph}(t - t')] F(t)}{\omega_{ph}} \]

pump \parallel [100] 1.55eV
probe \parallel [001] broad band
La\textsubscript{2}-x Sr\textsubscript{x}CuO\textsubscript{4} x=0.15

Okamoto (Yesterday), Demsar (this morning)
Detection of Excitations

\[ \xi(t) = \int_{-\infty}^{t} dt' \frac{\sin[\omega_{ph}(t - t')]}{\omega_{ph}} F(t) \]

\[ \varepsilon = 1 - 4\pi \chi \]

\[ \delta \epsilon_{\mu\nu}(t) = -4\pi \frac{d \chi_{\mu\nu}}{d\xi} \xi(t) \]

pump \parallel [100]
probe \parallel [001]
Swiss Knife Matrix Element

\[ \frac{d\chi_{\mu\nu}}{d\xi} = \]

Spontaneous Raman scattering

Coherent generation of excitations

Detection of excitations

Coherent control of excitations
Real time Raman vs Frequency Domain

- Phase sensitive information
- Raman profile in one shot
- Coherent control of excitations
A new coherent excitation

$A_{1g} + B_{2g}$

10 K

$E = 0.15$

B

Energy (eV)

0 2000 4000

Delay (fs)

$\Delta R/R$

2.45 eV

4x10^{-3}

4 3 2 1 0

FFT amplitude (arb. un.)

Frequency (THz)

0 5 10 15 20

Energy (meV)

0 20 40 60 80 100

FFT

Raman

pump $\parallel [110]$

probe $\parallel [110]$

x = 0.15
Coherent generation of excitations

**Phonons**

\[
H = H_{ph} - \frac{1}{2} \mathbf{E}(t) \cdot \frac{\partial \chi(\omega_L)}{\partial \xi} \cdot \mathbf{E}(t) \hat{\xi} \\
= H_{ph} - F(t) \hat{\xi}
\]

\[
\xi(t) = \int_{-\infty}^{t} dt' \frac{\sin[\omega_{ph}(t - t')]}{\omega_{ph}} F(t)
\]

**Charge Fluctuations**

\[
H = H_{BCS} - \frac{1}{2} \mathbf{E}(t) \cdot \frac{\partial \chi(\omega_L)}{\partial N_X} \cdot \mathbf{E}(t) \hat{N}_X \\
= H_{BCS} + \nu_X(t) \hat{N}_X
\]

\[
\delta N_X(t) = -i \int_{-\infty}^{t} dt' \langle [\hat{N}_X(t), \hat{N}_X(t')] \rangle \nu_X(t)
\]
Magnetism and Superconductivity

\[ H_{BCS} = \sum_k \xi_k c_{k\sigma}^\dagger c_{k\sigma} - \sum_k (\Delta_k^* c_{-k\downarrow}^\dagger c_{k\uparrow}^\dagger + h.c.) \]

\[ \sigma_k^x = (c_{k\uparrow} c_{-k\downarrow} + h.c.), \sigma_k^y = (c_{k\uparrow} c_{-k\downarrow} - h.c.), \sigma_k^z = 1 - n_{k\uparrow} - n_{-k\downarrow} \]

\[ |BCS\rangle = \prod_k (u_k + v_k c_{-k\downarrow}^+ c_{k\uparrow}^+)|0\rangle \]

Diagram: Transition from Normal to Superconducting state with energy level movement and quasiparticle creation.
Magnetism and Superconductivity


dt \frac{\partial \sigma_k}{\partial t} = -2[b_k^0 + \delta b_k(t)] \times \sigma_k.

Larmor at Ef \quad \omega_k = 2\Delta_k

NMR like!
NMR in Charge Space
NMR in Charge Space
Coherent Charge Fluctuation Spectroscopy

$$\delta \epsilon(\omega, t) = -4\pi \sum_X \frac{\partial \chi}{\partial N_X}(\omega) \langle N_X \rangle(t)$$

Very specific!
Glue Debate

Anderson, Scalapino, Science 2007

Scalapino: Numerics support a retarded interaction scenario. Paramagnons are the glue.

Anderson: RVB is the true. Do not search for phonons, magnons, etc. There is no need to be a glue-sniffer.

Maier, Poilblanc, D. J. Scalapino, PRL 2008

~20% of the attraction from coupling to high energy states
B1g Symmetry and Fluency Dependence

A1g + B1g pump \parallel [100] \quad \text{probe} \parallel [100]

A1g - B1g pump \parallel [100] \quad \text{probe} \parallel [010]
Conclusions

• Superconducting charge fluctuations generated and detected by light pulses for the first time.
• Raman profiles carry precious information on the coupling between low energy excitations and high energy excitations.
• Strong analogy with NMR opens the possibility of coherent control of the superconducting wave function.
• Enables Coherent Charge Fluctuation Spectroscopy, a new technique that allows to answer the question: Which excitations are coupled to the superconducting quasiparticles. High specificity like Isotope effect.
• In our system: Excitations at the scale of the Hubbard U are coupled to low energy charge fluctuations? Signature of Mottness in the superconducting wave function.
Polarization Analysis

\[ A_{lg} \]

\[ B_{lg} \]
Raman Scattering: the $A_{1g}$ La Phonon

\[ \Delta = \Delta_0 + Z e^2 \left( \frac{1}{d_{LaO}} - \frac{1}{d_{LaCu}} \right) \]

\[ \text{el-ph} \]

\[ \frac{d\chi}{d\xi} = \frac{d\chi}{d\Delta} \frac{d\Delta}{dz} \frac{dz}{d\xi} \]

\[ \frac{d\chi}{d\Delta} \approx -\frac{d\chi}{d\omega} \]

\[ \sqrt{M_{La}} u_{La} = e_{La} \xi e^{-i\omega t} + e_{La}^* \xi^* e^{i\omega t} \]
Raman profile as a fingerprint of excitations

\[
\delta \epsilon_{xx}(\omega, t) = -4\pi \frac{\partial \chi_{xx}}{\partial n_{CT}}(\omega) \delta n_{CT}(t)
\]

pump $|| [110]$
probe $|| [110]$
$X=0.15$

\[10K\]
$A_{tg}+B_{2g}$
NMR in Charge Space

Carbone_movies2.gif
NMR in Charge Space
High-Tc Cuprates

Correlated Insulator

Fermi Liquid

Left Revolutionaries

Right Conservatives/Reformists

Temperature, $T$ (K)

Hole concentration, $p$

$T_N$, $T^*$, $T_c$, $p_s$, $p^*$, $p_c$
Raman Scattering

$|f\rangle$

$\omega_{0f}$

$|0\rangle$

$\omega_L \pm \omega_{ph}$
Raman Scattering

\[ \xi(t) = \xi e^{-i \omega_{ph} t} \]

\[ \sqrt{M_i u_i} = e_i \xi e^{-i \omega_{ph} t} + e_i^* \xi^* e^{i \omega_{ph} t} \]

\[ \alpha = \frac{e^2}{m \omega_0^2 f - \omega^2 - i \omega \gamma} \]

\[ \alpha = \alpha_0 + \frac{\partial \alpha}{\partial \xi} \xi e^{-i \omega_{ph} t} + \frac{\partial \alpha}{\partial \xi^*} \xi^* e^{i \omega_{ph} t} \]

\[ p = \alpha E \]

\[ E = E_0 e^{-i \omega_L t} \]

\[ p = \left( \alpha_0 e^{-i \omega_L t} + \frac{\partial \alpha}{\partial \xi} \xi e^{-i (\omega_L + \omega_{ph}) t} + \frac{\partial \alpha}{\partial \xi^*} \xi^* e^{-i (\omega_L - \omega_{ph}) t} \right) E_0 \]
Conventional Superconductors

Moving charge

\[ r \]
Conservative/Reformist View

\[ V_{\text{tot}}(\mathbf{r},t) = V_{\text{dir}}(\mathbf{r},t) + V_{\text{ind}}(\mathbf{r},t) \]

\[ V_{\text{ind}}(\mathbf{r},t) = -e e' g_n^2 \chi_n(\mathbf{r},t) - s \cdot s' g_m^2 \chi_m(\mathbf{r},t) \]

\(^3\text{He}: \text{Leggett, RMP 1975} \)
\(\text{Superconductors: Monthoux, Pines, Lonzarich, Nature 2007} \)
“Left Revolutionary”

PW Anderson

RVB
Scales U and J
Important
No retardation
Raman Scattering

Example: The A1g La Phonon

\[
\Delta = \Delta_0 + Ze^2 \left( \frac{1}{d_{LaO}} - \frac{1}{d_{LaCu}} \right)
\]

\[
\sqrt{M_{La}} u_{La} = e_{La} \xi \exp(-i\omega t) + e_{La}^* \xi^* \exp(i\omega t)
\]

Increase in \(\Delta\)

\[
\frac{d\chi}{d\xi} = \frac{d\chi}{d\Delta} \frac{d\Delta}{dz} \frac{dz}{d\xi}
\]

\[
\frac{d\chi}{d\Delta} \approx -\frac{d\chi}{d\omega}
\]

\[
\frac{d\sigma}{d\Omega d\omega} (\omega_L)
\]
Raman in Solids

\[ P = np \quad \chi = n\alpha \quad P = \chi E \quad \epsilon = 1 - 4\pi\chi \quad \chi = \frac{\sigma}{i\omega} \]

\[ \chi = \chi_0 + \frac{\partial \chi}{\partial \xi} \xi e^{-i\omega_{ph}t} + \frac{\partial \chi}{\partial \xi^*} \xi^* e^{i\omega_{ph}t} \]

\[ \omega_R = \omega_L - \omega_s \]

\[ \frac{d\sigma}{d\Omega d\omega} = \frac{\omega_s^4 V^2}{(4\pi)^2 c^4} \sum_{\nu} |\langle 0|\hat{e}_s, \hat{\rho}\cdot\hat{e}_l|\nu\rangle|^2 \delta(\omega_R - \omega_{\nu}) \]

\[ \hat{\rho} = \frac{\partial \chi}{\partial \xi}(\omega_L)\xi \quad \frac{1}{\pi} \text{Im}\Pi(\omega_R) \]

\[ \Pi(\omega) = i \int_{-\infty}^{t} dt' e^{i\omega t'} \langle [\hat{e}_L\cdot\hat{\rho}(t)\cdot\hat{e}_s, \hat{e}_L\cdot\hat{\rho}(t')\cdot\hat{e}_s] \rangle \]

Cardona, Light Scattering in Solids II
Heavy Fermions

[Diagram of phase diagram for CeCu$_2$Si$_{2-x}$Ge$_x$ showing magnetic metal, heavy fermion, superconductivity, and intermediate valence regions.]
Coherent Excitation of charge fluctuations by Impulsive Stimulated Raman Scattering

\[ H_R(t) = -\frac{1}{2} \sum_k \mathbf{E}(t) \cdot \chi_{el}^R \mathbf{E}(t) f_k^X (n_{k\uparrow} + n_{-k\downarrow}) \]

\[ H_R(t) = \sum_k \nu_k^X(t)(n_{k\uparrow} + n_{-k\downarrow}) \]

\[ \nu_k^X(t) = -\frac{1}{2} \mathbf{E}(t) \cdot \chi_{el}^R \mathbf{E}(t) f_k^X \]
Optical Conductivity and Fluctuations

\[ \sigma(\omega) = \frac{1}{1+\frac{i\omega}{\tau}} \]

Uchida et al. PRB'91


\[ \delta n(r_i) \propto c \rho Q + \delta \]

\[ Q = (\pi / 2, 0) \]
Unconventional Superconductors

\[ V_{\text{ind}}(r,t) = - e e' g_n^2 \chi_n(r, t) - s \cdot s' g_m^2 \chi_m(r, t) \]

Close to instabilities the susceptibility is large at small energies (long times)
Raman Scattering

\[ H = H_{ph} - \frac{1}{2} \mathbf{E}(t) \cdot \frac{\partial \chi(\omega_L)}{\partial \xi} \cdot \mathbf{E}(t) \xi = H_{ph} - F(t) \xi \]

\[ \frac{d\sigma}{d\Omega d\omega} = \frac{\omega_s^4 V^2}{(4\pi)^2 c^4} \sum_{\nu} |\langle 0 | \hat{\rho} \cdot \hat{\rho} | \nu \rangle|^2 \delta(\omega - \omega_\nu) \]

\[ \hat{\rho} = \frac{\partial \chi}{\partial \xi}(\omega_L) \xi \]

\[ \frac{1}{\pi} \text{Im} \Pi(\omega_R) \]

\[ \Pi(\omega) = i \int_{-\infty}^{t} dt' e^{i\omega t'} \langle [\hat{\rho}_L(t') \cdot \hat{\rho}_s (t), \hat{\rho}_L(t) \cdot \hat{\rho}_s (t')] \rangle \]

FIG. 9. Raman spectra of (La$_{1-x}$Sr$_x$)$_2$CuO$_4$ with $x = 0.035$ at 30 and 337 K in the (z,z) (solid curves) and (x,z) (dotted curves) polarization configurations.

FIG. 10. Incident wavelength dependence of the Raman spectra in La$_2$CuO$_4$ at 30 K.

Sugai PRB '89