



# Coupling of a high-energy excitation to superconducting quasiparticles in a high-T<sub>c</sub> cuprate from coherent charge fluctuation spectroscopy



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## Pairing interactions in high-T<sub>c</sub> cuprates



What is the glue (if any) maintaining electrons of Cooper pairs together?

Low-energy theories (Eliashberg formalism)

High-energy theories

## Pairing interactions in high-T<sub>c</sub> cuprates

Low-energy theories (Eliashberg formalism)  $H_{BCS} = \sum_{\mathbf{k},\sigma} \xi_k c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}\sigma} - \frac{V}{2\Omega} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q},\sigma\sigma'} c_{\mathbf{k}+\mathbf{q},\sigma}^{\dagger} c_{\mathbf{k},\sigma} c_{\mathbf{k}',\mathbf{q},\sigma'}^{\dagger} c_{\mathbf{k}',\mathbf{q},\sigma'} c_{\mathbf{k}',\sigma'}$  $\Psi = \prod_{\mathbf{k}} (u_{\mathbf{k}} + v_{\mathbf{k}} c_{\mathbf{k}\uparrow}^{\dagger} c_{\mathbf{k}\downarrow}^{\dagger}) \left| 0 \right\rangle$ Electrons bind in Cooper pairs through attractive effective potential V>0 **BCS:** phonons Cuprates: spin fluctuations (?) Energy scale: meV Time-scale: picosecond **RETARDED INTERACTIONS** 

Scalapino et al., PRB 34 (1986)

High-energy theories

$$H_{t-J} = P \sum_{\mathbf{k},\sigma} \xi_k c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}\sigma} P + J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j \qquad J \propto t^2 / U$$
$$P\Psi = P \prod_{\mathbf{k}} (u_{\mathbf{k}} + v_{\mathbf{k}} c_{\mathbf{k}\uparrow}^{\dagger} c_{-\mathbf{k}\downarrow}^{\dagger}) |0\rangle \qquad P = \prod_i (1 - n_i\uparrow n_i\downarrow)$$

Formation of a Resonating-Valence-Bonds liquid of Cooper pairs due to strong electronic correlations



0

Energy scale: U: eV  $r = \frac{|t_r t_{r'} \rangle - |t_r t_{r'} \rangle}{\sqrt{2}}$ Time-scale: sub-femtosecond

#### **NON-RETARDED INTERACTIONS**

Anderson, Science 235 (1987)

Determination of the pairing mechanism: study of the energy-scale and/or the time-scale of the pairing interactions

### Experimental setup: broadband pump-probe reflectivity



B. Mansart et al, PNAS 109 (2012)

### Static optical properties of La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub>, x=0.15



Lorenzana et al., PRL 90 (2003)

- Static optical properties isotropic in (a,b) plane
- Broadband measurements: access to the relation between high-energy excitations and superconducting condensate

### Effect of laser excitation



Coherent generation of elementary excitations: charge fluctuations



### Effect of laser excitation in a superconductor

 $H = H_{BCS} + H_R$ 



Bogoliubon: superposition of an  $e^{-}$ excitation and a hole-excitation in a SC

Excited state wavefunction

$$\Psi(t)\rangle = |BCS\rangle + \sum_{k} \epsilon_{k} e^{-i2E_{k}t} \gamma_{k\uparrow}^{+} \gamma_{-k\downarrow}^{+} |BCS\rangle$$
$$E_{k} = \sqrt{(\epsilon_{k} - \mu)^{2} + |\Delta_{k}|^{2}}$$

Pump: Prepare the system on a wave function that is the SC GS + a given charge-fluctuation

Describe the GS with PS formalism, the fluctuations induce precession of the PS.

$$H_{BCS} = -2\sum_{k}^{\mathsf{Ekin}} \xi_k \sigma_k^z - \sum_{k,k'}^{\mathsf{CP}} (\sigma_k^x \sigma_{k'}^x + \sigma_k^y \sigma_{k'}^y) + cste \qquad \begin{cases} \sigma_k^x = \frac{1}{2}(b_k^\dagger + b_k) \\ \sigma_k^y = \frac{1}{2}i(b_k^\dagger - b_k) \end{cases} \begin{cases} b_k = c_{-k\downarrow}c_{k\uparrow} & \text{Anderson,} \\ b_k^\dagger = c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger & \text{Hys. Rev.} \\ 112 (1958) \end{cases}$$
  
BCS Hamiltonian -> ferromagnetism problem

BCS Hamiltonian —>ferromagnetism problem

Pump effect  $\rightarrow$  time-dependent impulsive potential  $\rightarrow$  fictitious magnetic field  $\delta b_k$  which makes the pseudospins precessing

 $\hbar \frac{\partial \boldsymbol{\sigma}_{\mathbf{k}}}{\partial t} = -2[\mathbf{b}_{\mathbf{k}}^{0} + \delta \mathbf{b}_{\mathbf{k}}(t)] \times \boldsymbol{\sigma}_{\mathbf{k}}. \qquad \text{PS motion} \quad 2|\mathbf{b}_{\mathbf{k}}^{0}|/\hbar, \ |\mathbf{b}_{\mathbf{k}}^{0}| = \sqrt{\xi_{\mathbf{k}}^{2} + \Delta_{\mathbf{k}}^{2}} \quad \text{Angular freq.}$ 

### Effect of laser excitation in a superconductor



B. Mansart, et al. PNAS **110** 4539 (2013).

# Transient A<sub>1g</sub>+B<sub>2g</sub> response



 $E_{pump}//(110), E_{probe}//(110), k//001$ F ~ 300 µJ/cm<sup>2</sup>

A<sub>1g</sub>+B<sub>2g</sub> Raman excitation

The signal changes sign as a function of wavelength

4000

B. Mansart, et al.

PNAS 110 4539 (2013).



# Transient A<sub>1g</sub>+B<sub>2g</sub> response



 $E_{pump}//(110), E_{probe}//(110), k//001$ F ~ 300 µJ/cm<sup>2</sup>

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

The signal changes sign as a function of wavelength

The coherent oscillations vanish above T<sub>c</sub>



## Transient A<sub>1g</sub>+B<sub>1g</sub> response



 $E_{pump}//(100), E_{probe}//(100), k//001$ F ~ 300 µJ/cm<sup>2</sup>

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# Generating and probing coherent superconducting condensate oscillations



Ultrafast experiments compared to static Raman scattering



B. Mansart, et al. PNAS **110** 4539 (2013).

# Generating and probing coherent superconducting condensate oscillations



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### **Coherent Charge Fluctuations: pseudospins description**



Trieste, October 2014

## Superconducting condensate oscillations energy scale



Probe-energy dependence of B<sub>2g</sub> fluctuation in frequency-domain

Clear resonance at 2.6 eV Charge-transfer energy of the parent compound

Evidence for non-retarded contribution to the pairing mechanism

> B. Mansart, et al. PNAS **110** 4539 (2013).

## Conclusion



- Evidence for instantaneous pair-breaking by charge-transfer excitation
- New technique, Coherent Charge Fluctuation Spectroscopy, which has a high degree of specificity (similar to the isotope effect for BCS systems)
- Evidence for non-retarded (high energy) contribution to the pairing mechanism (Anderson scenario)
- Polarization, spectroscopy and time resolution are mandatory

B. Mansart, et al. PNAS **110** 4539 (2013).

# **Broken symmetry and collective modes**

 Below Tc (100K/250GeV) amplitude and phase fluctuations lead to welldefined collective excitations

Phase and amplitude fluctuations are decoupled



massive:  $\omega^2 = m^2 + (v_s q)^2$ 



#### Goldstone

For a neutral superconductor massless:  $\omega = v_s q$ 

Adapted from a slide by Lara Benfatto

# Anderson-Higgs mechanism

In a superconductor:

D. Van der Marel

Journal of Superconductivity: Incorporating Novel Magnetism, Vol. 17, No. 5, October 2004 (© 2004)



# **Condensed Matter vs High Energy Physics**

Amplitude mode Bogoliubov mode Anderson mechanism plasmon Neutral phase oscillations Higgs Boson Nambu-Goldstone Higgs-mechanism (phase and amplitude coupled) W particle Z particle mediate the weak interaction



### $A_{1g}$ Bogoliubov or Nanbu-Goldstone mode made massive by Anderson-Higgs



# B<sub>19</sub> Mode



# How to excite the Higgs mode?



A transient change in the pairing interaction may excite the amplitude mode.

Also pumping in the gap region in a dirty superconductor may excite the Higgs in a similar way to a Raman process.

$$H=\chi^{\prime}(\omega) \to \Delta$$

# Higgs mechanism and TH spectroscopy

The condensate SW changes have a direct impact on the spectrum at  $2\Delta$ 





# It was such a good idea.....



B. Mansart, et al. PNAS **110** 4539 (2013).

Arxiv 2011.....

#### Matsunaga et al., Science, 2014



ÉCOLE POLYTECHNIQUE Fédérale de Lausanne

# Higgs mechanism and visible light spectroscopy

#### In BCS dirty limit

In High Tcs





The superconducting condensate changes influence the SW at  $2\Delta$  AND at the CT energy

PHYSICAL REVIEW B 74. 024502 (2006) PHYSICAL REVIEW B 74, 064510 (2006)

Disentangling the origin of the condensate's coherent oscillations is not as straightforward as It seems......

# Take-home messages

- Light excitation can excite coherent oscillations of the Cooper pairs condensate
- These oscillations resonate at the CT energy (High-energy scale, suggestive of RVB type of pairing mechanism)
- These oscillations may be driven by Higgs modes (under debate)



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