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Disentangling the non-equilibrium dynamics in the Pseudogap phase of Y-Bi2212 Cuprate Superconductors, by the Time Resolved Optical Spectroscopy.

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INTRODUCTION

The pseudogap state is the most exotic and elusive phase of the phase diagram of High Critical Temperature Superconductors (HTSCs), and the comprehension of the microscopic electronic mechanisms behind is of paramount importance to reveal the nature of this intriguing phase.

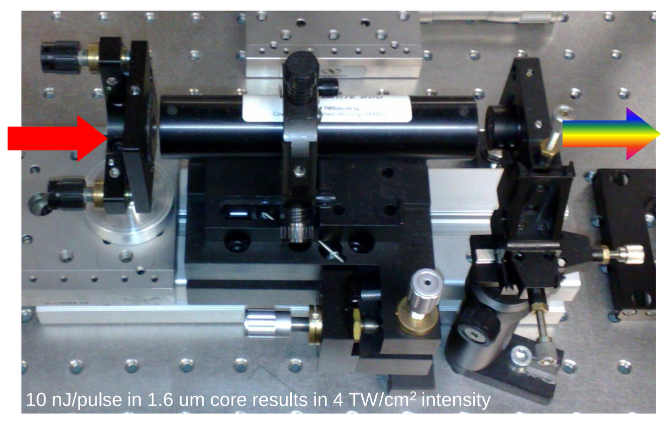
By exploiting a novel, non-equilibrium all-optical experimental technique, the time-resolved spectroscopy, I demonstrate that the non-equilibrium dynamics underlying this phase are more complex than those observed for the normal state phase, suggesting that the pseudogap in HTSCs is a state of matter where the fermionic quasiparticles and the bosonic excitations are strongly intertwined.

Several Y-Bi2212 ($Bi_2Sr_2Y_{0.08}Ca_{0.92}CuO_{8+\delta}$) superconducting samples, differing in the doping level, have been probed with the time-resolved optical spectroscopy, to map the evolution of the time-resolved optical signal through the phase diagram of the compound. In particular, for the non-equilibrium dynamics of samples in the pseudogap phase, measurements are performed at T=100 K, the pseudogap extending between T^* (~140 K for OP samples and ~200 K for UD samples) and T_c. In this region of the doping-T phase-diagram, the transient frequency resolved optical response is different from what observed in the normal state and a pure non thermal scenario does not account for the results. Exploiting the time- and frequency-resolution of our technique and the differential analysis based on a differential dielectric function approach, I demonstrate that, on the femtosecond timescale, the transient optical response at T=100 K is dominated by an excitation-dependent bosonic glue and by an impulsive quench of a gap in the density of states.

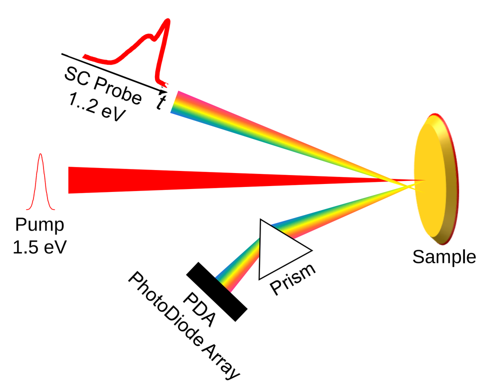
THE TIME RESOLVED SPECTROSCOPY (TRS)

The TRS joins the spectral resolution typical of equilibrium spectroscopies with the time resolution of non-equilibrium techniques, providing a unique capability of disentangling the interplay of the different degrees of freedom by their characteristic timescales and spectral responses.

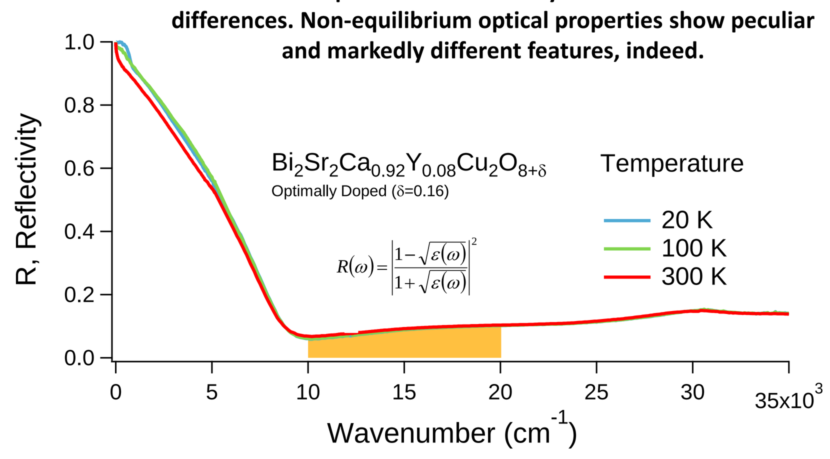
A broadband (0.5-3 eV) White Light supercontinuum spectrum is produced by a PCF, seeded by an Ti:Sa oscillator[1].



The White Light is dispersed with a prism onto a PDA: the optical properties at each wavelegth are analyzed. Temporal resolution is ~150 fs. Pump fluence is kept on the ~uJ/cm² level. High S/N ratios (>10⁴) are achieved.



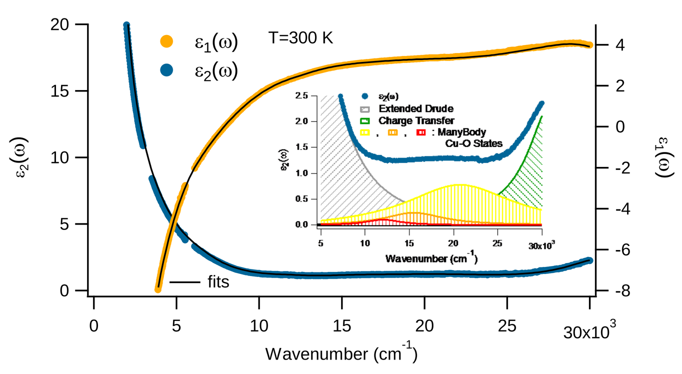
Equilibrium Optical Properties of OP Y-Bi2212 are shown for T=20 K, T=100 K, T=300 K. The spectral region probed by TRS is evidenced. Equilibrium Reflectivity doesn’t show marked differences. Non-equilibrium optical properties show peculiar and markedly different features, indeed.



THE STATIC MODEL for ε(ω)

Reproducing the experimentally determined complex dielectrc function with an Extended Drude + Lorentz phenomenologic model is the prior step for a proper differential analysis.

Ellipsometric data ($\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ at T=300 K) and their fit with the *Model Dielectric Function*.



The Model Dielectric Function is a sum of an Extended Drude term (accounting for intraband transitions) and 6 Lorentz oscillators, (accounting for interband transitions).

$$\varepsilon(\omega,T)=\varepsilon_\infty+\varepsilon_{ED}(\omega,T)+\sum_{i=1}^{6}\varepsilon_{L_i}(\omega)$$

$$\varepsilon_{L_i}(\omega)=\frac{\omega_{p_i}^2}{(\omega_{0_i}^2-\omega^2)-i\gamma_i\omega}$$

$$\varepsilon_{ED}(\omega,T)=-\frac{\omega_{p_{ED}}^2}{\omega(\omega+iM(\omega,T))}$$

M(ω,T) is the ***Memory Function***, a complex function accounting for the scattering of electrons with bosonic excitations.

Experimental ΔR/R

$$\frac{\Delta R}{R}(\omega,\tau)=\frac{R_{pumped}(\omega,\tau)-R_{unpumped}(\omega)}{R_{unpumped}(\omega)}$$

Model ΔR/R

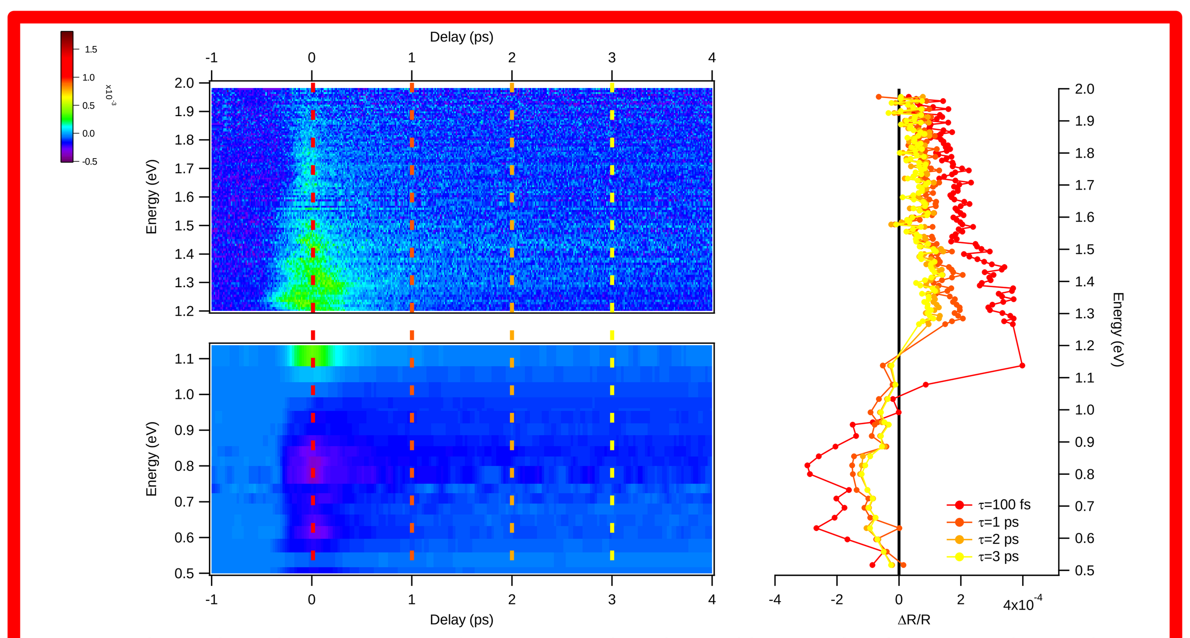
$$\frac{\Delta R}{R}(\omega,\tau)=\frac{R(\varepsilon_{excited}(\omega,\tau))-R(\varepsilon_{equilibrium}(\omega))}{R(\varepsilon_{equilibrium}(\omega))}$$

Fitting of the *differential dielectric function* model to experimental data is performed by modifying the smallest number of parameters in $\varepsilon_{excited}(\omega,t)$ w.r.t. $\varepsilon_{equilibrium}(\omega,t)$: an unambiguous physical origin can be finally assigned to the time-resolved optical signal!!

TIME RESOLVED SPECTROSCOPY on Optimally Doped $Bi_2Sr_2Y_{0.08}Ca_{0.92}CuO_{8+\delta}$ CRYSTALS

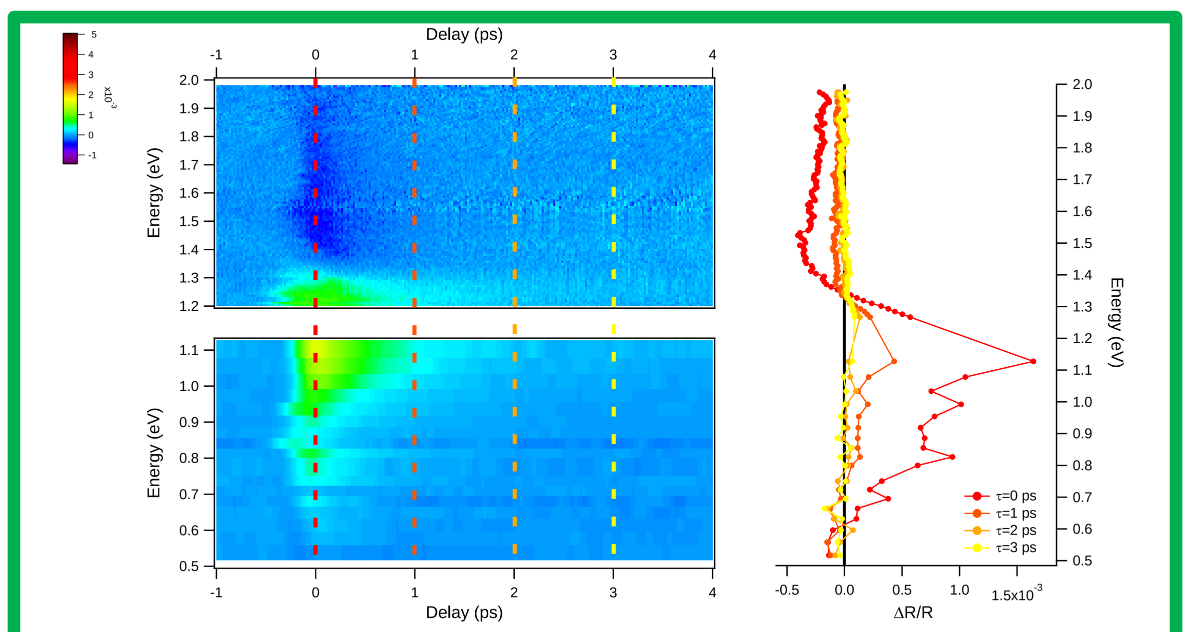
Temperature

T=300 K – Normal State



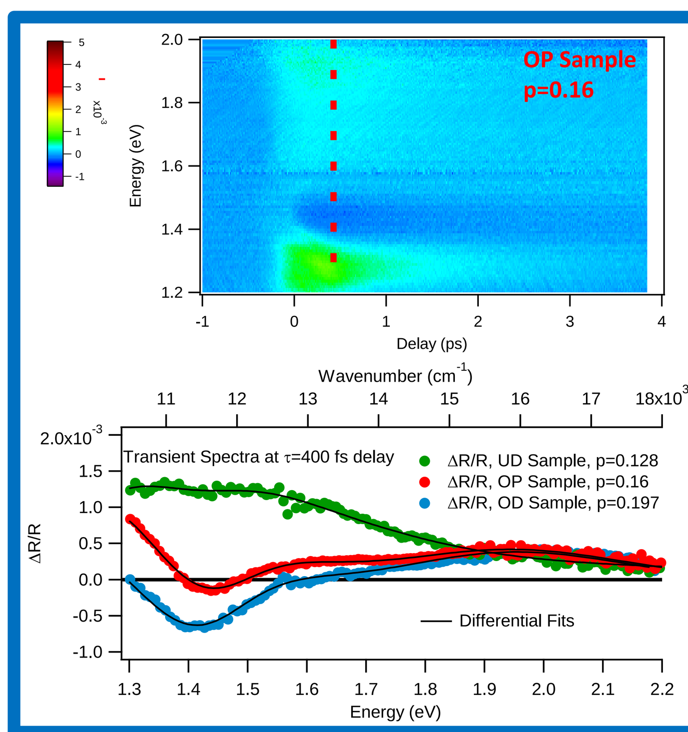
Data in the NS have been interpreted assuming only a thermal heating for the system, but with an unconventional, non-thermal character, indicating a strong coupling of electrons with bosons of electronic origin. See [3].

T=100 K – Pseudogap



Data in the PG cannot be interpreted by only assuming a purely, though peculiar, thermal heating effect. Other effects are taking place. We invoked both a photoinduced quench of the gap opened in the PG phase in the DOS, and a modification of the Bosonic Glue Π(Ω). See panel below.

T=20 K – Superconducting State



In the SC Phase, the ΔR/R is strongly structured in the visible region. A different scenario with respect to both the NS and the PG case must be invoked: the photoinduced gap closing turns out in a superconductivity induced rearrangement of two high-energy Lorentz oscillators, namely the 1.5 eV and the 2 eV ones, which is strongly doping dependent [2].

• The long-standing question regarding the interplay between high- and low- energy scale physics in cuprates have been addressed: the measured spectral weight modification involves only specific transitions.

• The doping-dependent kinetic energy variation changes sign close to the optimal doping (OP) concentration, accounting for a KE gain in the UD side of the phase diagram and a KE loss in the OD side of the phase diagram.

ULTRAFAST DYNAMICS IN THE PSEUDOGAP PHASE

EXTENDED DRUDE MODEL IN THE PRESENCE OF A NON-CONSTANT DOS

A version of the EDM has been recently formulated[4] for the case of a non constant electronic DOS, as it is the case for the system in the Pseudogap.

$$M(\omega,T)=-i\omega\left\{\int_{-\infty}^{+\infty}\frac{f(\xi,T)-f(\xi+\omega,T)}{\omega+\Sigma^*(\xi,T)-\Sigma(\xi+\omega,T)+i\gamma_{imp}}\right\}^{-1}+i\omega$$

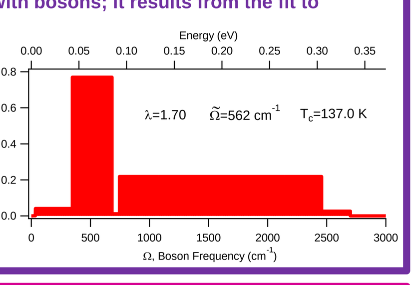
$$\Sigma_2(\omega,T)=-\pi\int_0^\infty \boxed{\alpha^2F(\Omega)}\left\{\frac{\boxed{\tilde N(\omega+\Omega,T)}}{N(0,T)}\left[n(\Omega,T)+f(\omega+\Omega,T)\right]+\frac{\boxed{\tilde N(\omega-\Omega,T)}}{N(0,T)}\left[1+n(\Omega,T)-f(\omega-\Omega,T)\right]\right\}d\Omega$$

$\alpha^2F(\Omega)$: the Glue Function (1)

Expresses the coupling of electrons with bosons; it results from the fit to equilibrium data.

The result of the fitting is in agreement with the literature [7]: an intense peak around ~70 meV is found, plus a broad continuum extending up to ~300 meV. λ and T_c have been calculated as follows:

$$\lambda=2\int_0^\infty\frac{\Pi(\Omega)}{\Omega}d\Omega \quad T_c=0.83\tilde\Omega e^{-1.04\frac{1+\lambda}{\lambda}}$$

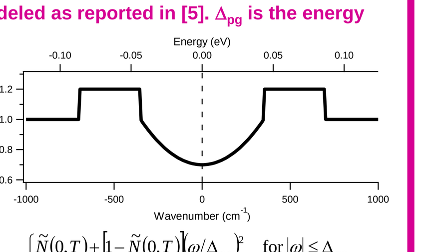


$\tilde N(\omega,T)$: the non-constant DOS (2)

The non-constant DOS has been modeled as reported in [5]. Δ_{pg} is the energy gap in the DOS in the Pseudogap.

$\tilde N(0,T)=0.7$

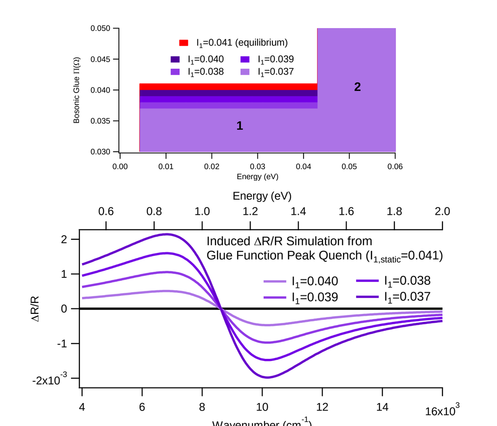
$\Delta_{pg}=350\ cm^{-1}$



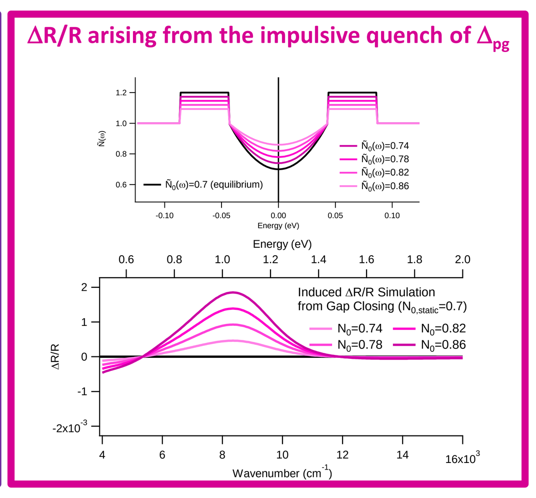
$$\tilde N(\omega,T)=\begin{cases}\tilde N(0,T)+\left[1-\tilde N(0,T)\right](\omega/\Delta_{pg})^2 & \text{for } |\omega|\le\Delta_{pg}\\ 1+2/3\left[1-\tilde N(0,T)\right] & \text{for } |\omega|\in(\Delta_{pg},2\Delta_{pg})\\ 1 & \text{for } |\omega|\ge\Delta_{pg}\end{cases}$$

SPECTRAL FEATURES ARISING FROM 1 & 2, within the DIFFERENTIAL DIELCTRIC FUNCTION APPROACH

ΔR/R arising from the impulsive quench of Π(Ω)

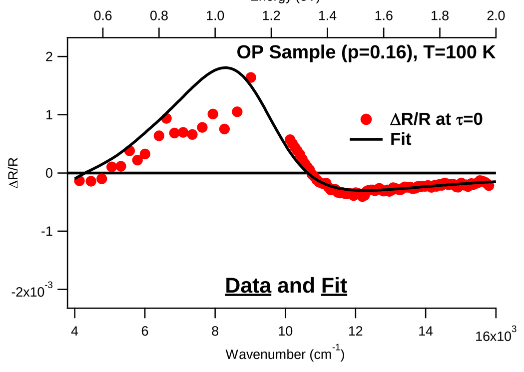


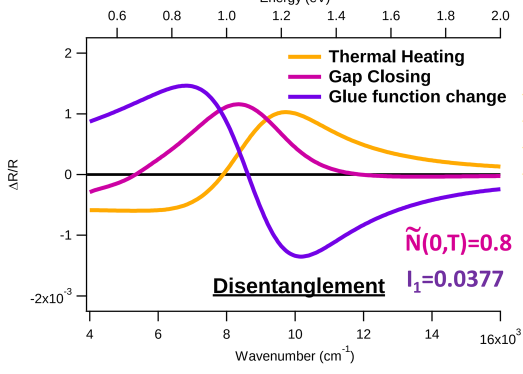
ΔR/R arising from the impulsive quench of Δ_{pg}



DISENTANGLING THE TIME-RESOLVED OPTICAL SIGNAL IN THE PSEUDOGAP

Three phenomena cooperate to give the total ΔR/R: 1) quench of one peak in Π(Ω); 2) quench of the gap in the DOS; 3) thermal heating. They can be disentangled thanks to their peculiar spectral feature.

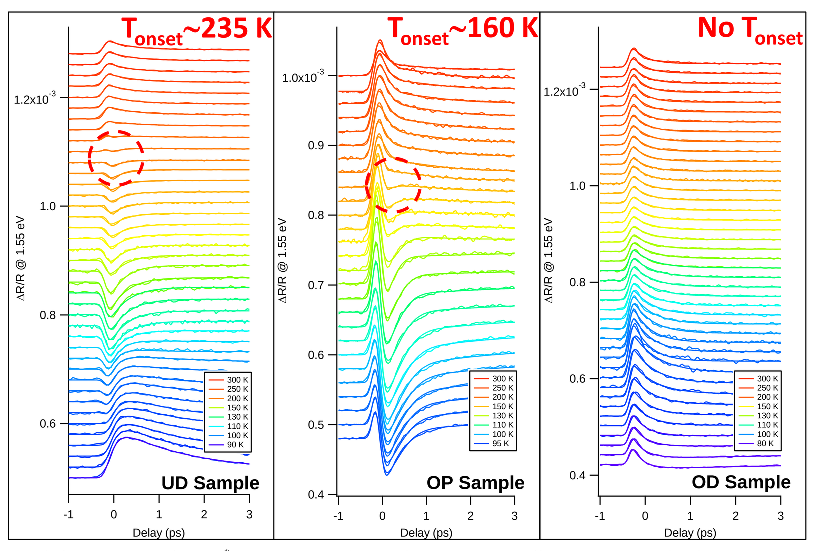




Non-thermal Heating (see [3]):
ΔT_e=9.5 K
ΔT_{be}=9.5 K
ΔT_p=1.1 K
ΔT_l=0.1 K

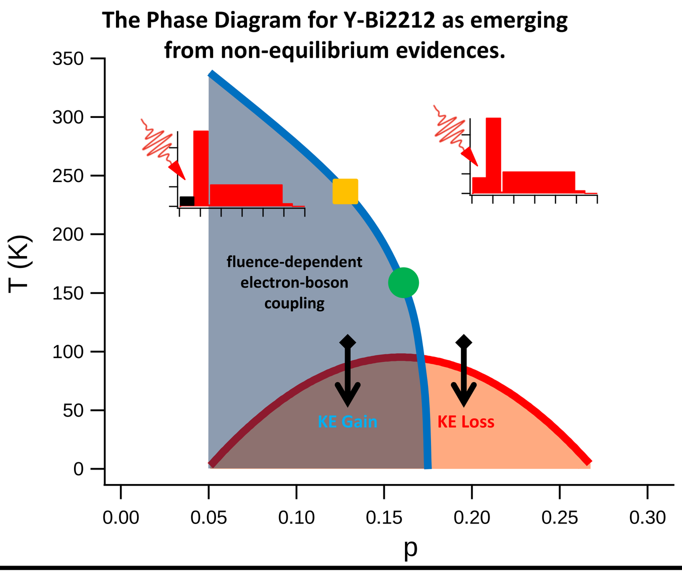
T-ONSET OF THE FLUENCE-DEPENDENT BOSONIC GLUE

To the aim of finding the temperature onset T_{onset} of contribution (1), we performed high-resolution measurements at 1.5 eV probe energy.



T_{onset} is in the range of the T^* temperature of the PG Phase. Recent findngs reported in [6] revealed the rising of a magnetic excitation mode at the T^* temperature of the sample, providing a possible origin for the effect. This new excitation mode, coupled to the electrons, is a peculiar feature of the PG Phase.

The Phase Diagram for Y-Bi2212 as emerging from non-equilibrium evidences.



CONCLUSIONS

Thanks to the TRS, which is emerging as the premier technique to unravel the elusive physics of strongly-correlated materials, the complex interplay of different phenomena in the Pseudogap Phase of a HTSC has been revealed and addressed. The major outcome is that a region of the (p,T) phase diagram of Y-Bi2212 has been found in which the Bosonic Glue acquires a fluence dependence.

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