Conference on the *Ultrafast Dynamics of Correlated Materials* – ICTP, Trieste, 14-18 October 2013

Elettra Sincrotrone Trieste

Drawing a Phase Diagram for High-Tc Cuprates by out-of-equilibrium spectroscopies

F. Cilento[1], S. Dal Conte[2,3,†], G. Coslovich[4,‡], S. Peli[2,5], N. Nembrini[2,5], S. Mor[2], F. Banfi[2,3], G. Ferrini[2,3], H. Eisaki[6], M.K. Chan[7], C. Dorow[7], M. Veit[7], M. Greven[7], D. van der Marel[8], R. Comin[9], A. Damascelli[9,10], L. Rettig[11,§], U. Bovensiepen[11], M. Capone[12], C. Giannetti[2,3]*, F. Parmigiani[1,4].

1. Elettra – Sincrotrone Trieste S.C.p.A., Basovizza I-34149, Italy; **2.** Department of Physics, Università Cattolica del Sacro Cuore, Brescia I-25121, Italy; **3.** i-Lamp (Interdisciplinary Laboratories for Advanced Materials Physics (i-LAMP), Università Cattolica del Sacro Cuore, Brescia I-25121, Italy; **4.** Department of Physics, Università degli Studi di Trieste, Trieste I-34127, Italy; **5.** Department of Physics, Università degli Studi di Milano, Italy; **6.** Nanoelectronics Research Institute, National Institute of Advanced Industrial Science and Technology, Tsukuba, Ibaraki 305-8568, Japan; **7.** School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455, USA; **8.** Département de Physique de la Matière Condensée, Université de Genève, Switzerland; **9.** Department of Physics and Astronomy, University of British Columbia, Vancouver, BC V6T 1Z1, Canada; **10.** Quantum Matter Institute, University of British Columbia, Vancouver, BC V6T 1Z4, Canada; **11.** Fakultaet fuer Physik and Zentrum für Nanointegration (CENIDE), Universitaet Duisburg-Essen, 47048 Duisburg, Germany; **12.** CNR-IOM Democritos National Simulation Center and Scuola Internazionale Superiore di Studi Avanzati (SISSA), Via Bonomea 265, 34136 Trieste, Italy; **†.** Present address: Department of Physics, Politecnico di Milano, I-20100 Milano, Italy; **‡.** Present address: Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA; **§.** Present address: Swiss Light Source, Paul Scherrer Institute, Villigen, Switzerland.

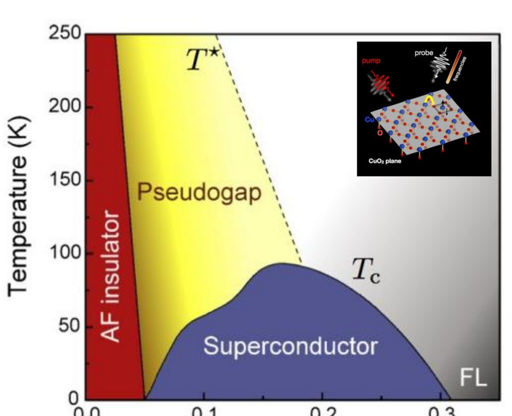
For correspondence and requests: federico.cilento@elettra.eu; claudio.giannetti@unicatt.it

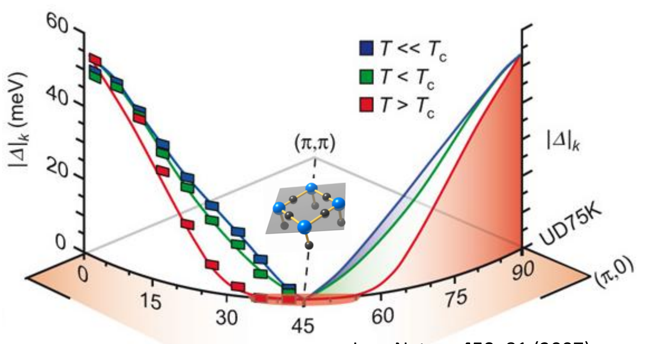
Introduction

The pseudogap pheonomenon in cuprate High-Tc superconductors is among the most elusive, though ubiquitous aspects of the physics of strongly correlated materials. The comprehension of the nature of this phase is fundamental in order to understand the microscopic mechanism determining the phase diagram of cuprates.

Here we tackle this problem by exploiting out-of-equilibrium spectroscopies, that allowed us to draw an advanced phase diagram for these materials. In particular, we revealed that the pseudogap phase is a state of matter where strong electronic correlations (that we call Mottness) affecting the antinodal excitations are at play. This result was obtained by combining optical and photoemission time-resolved spectroscopies with the solution of the 2D Hubbard model by CDMFT.

The emergence of long range ordered phases in the pseudogap phase of cuprates is a consequence of this correlated ground state, though the actual connection among the pseudogap with its associated Mottness and the broken symmetries remains to be understood.





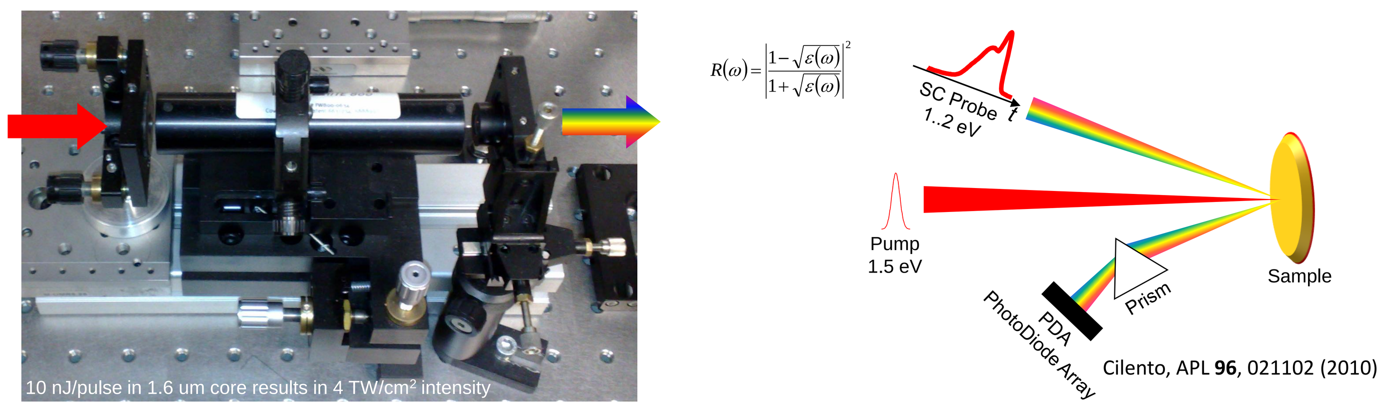
Lee, Nature **450**, 81 (2007)

Non-Equilibrium Spectroscopies: Experimental Approach

Out-of-Equilibrium spectroscopies with temporal resolution of ~100 fs allow to disentangle the intertwined degrees of freedom in strongly correlated materials thanks to their different spectral features and dynamics in the non-equilibrium signal.

Time Resolved Optical Spectroscopy (TROS)

Probes the dielectric function in a wide energy range, through reflectivity or transmittivity measurements

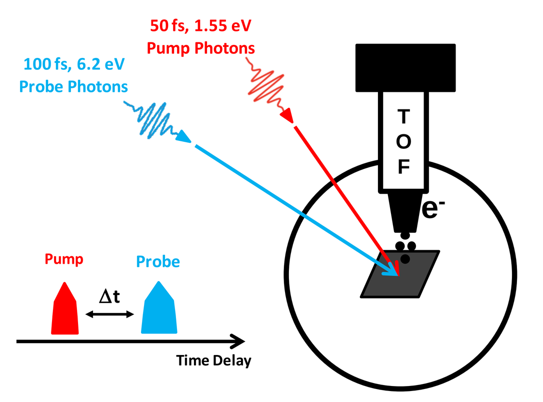


$$R(\omega)=\left|\frac{1-\sqrt{\varepsilon(\omega)}}{1+\sqrt{\varepsilon(\omega)}}\right|^2$$

Cilento, APL **96**, 021102 (2010)

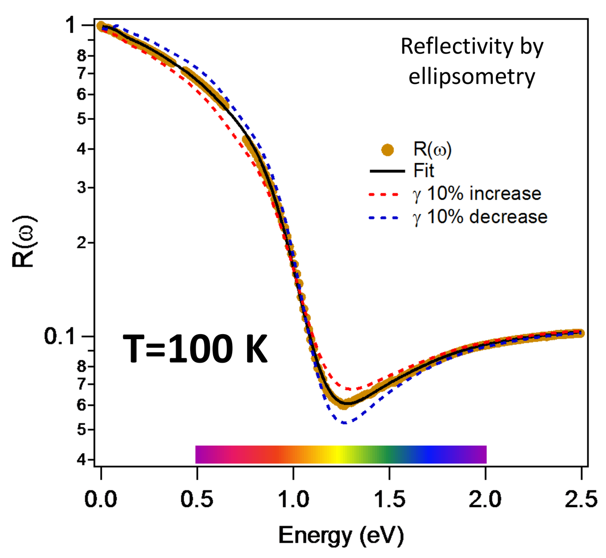
Time and Angle Resolved Photoelectron Spectroscopy (TR-ARPES)

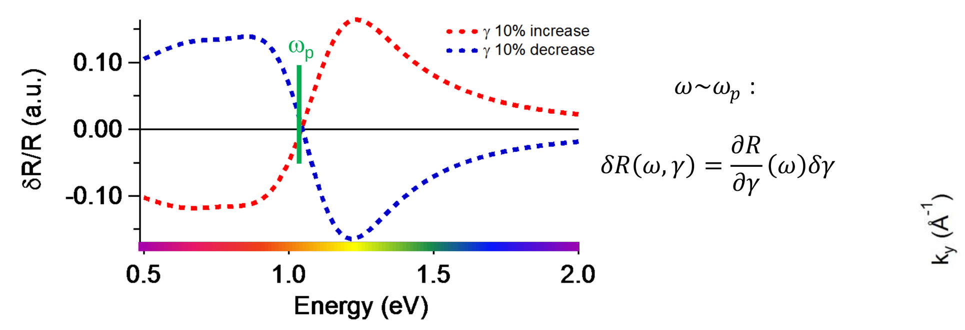
Directly probes electronic band structure



Data at Equilibrium

Equilibrium data constitute the playground to understand the variations of specific quantities revealed by non-equilibrium spectroscopies.



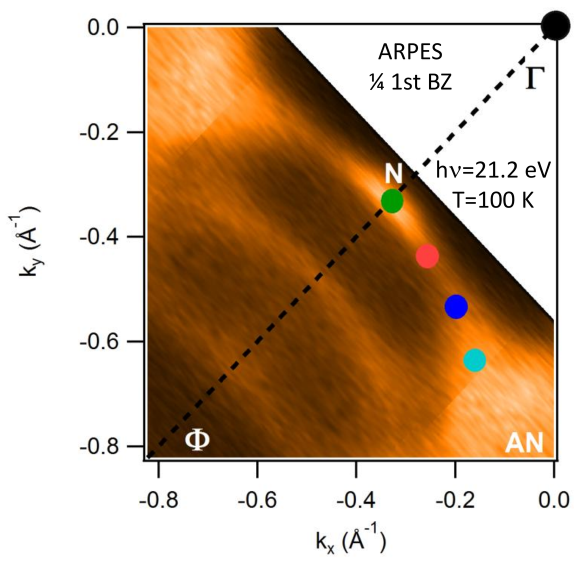


$\omega \sim \omega_p$:

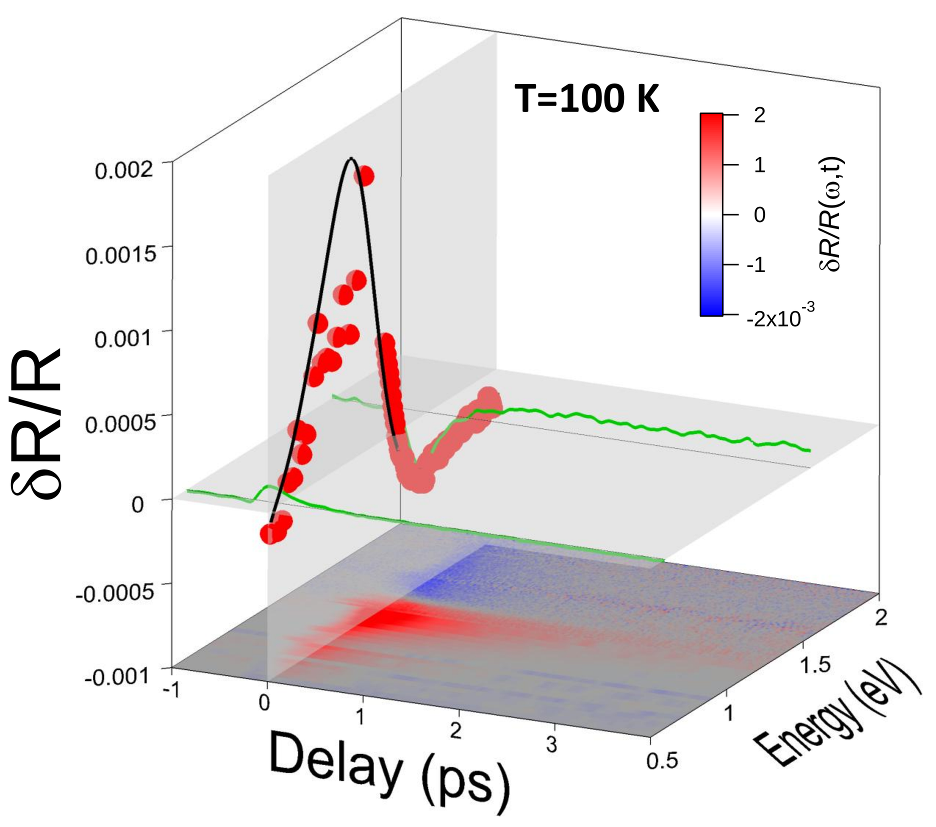
$$\delta R(\omega,\gamma)=\frac{\partial R}{\partial\gamma}(\omega)\delta\gamma$$

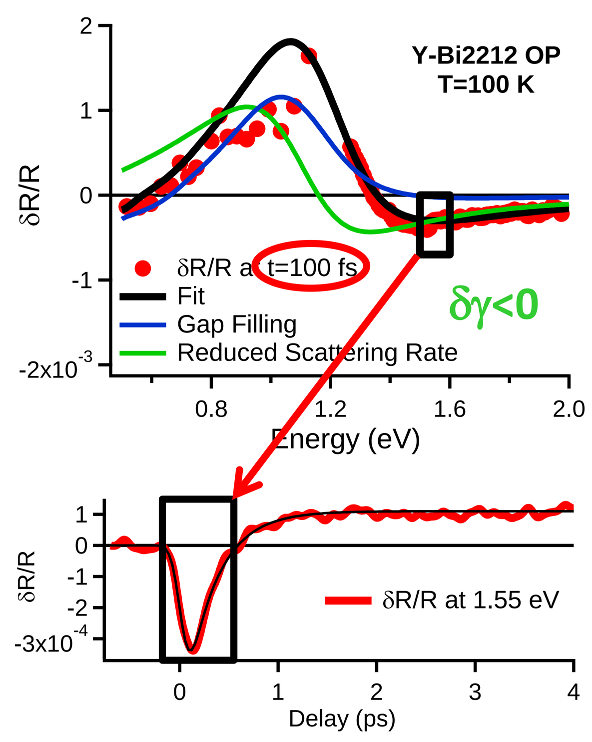
Applying a differential model to the dielectric function, it is possible to assign unambiguously a spectral feature in the δR/R signal to a precise origin at the microscopic level.

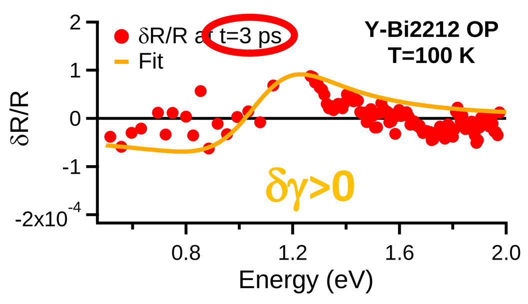
In particular, a modification of the scattering rate γ induces a peculiar modification of the reflectivity. R(ω) exhibits an isosbestic (fixed) point, around which the variation of reflectivity is proportional to the change of γ.



TR-Optical and TR-Photoemission Spectroscopic Data on Optimally Doped $Bi_2Sr_2Y_{0.08}Ca_{0.92}CuO_{8+\delta}$

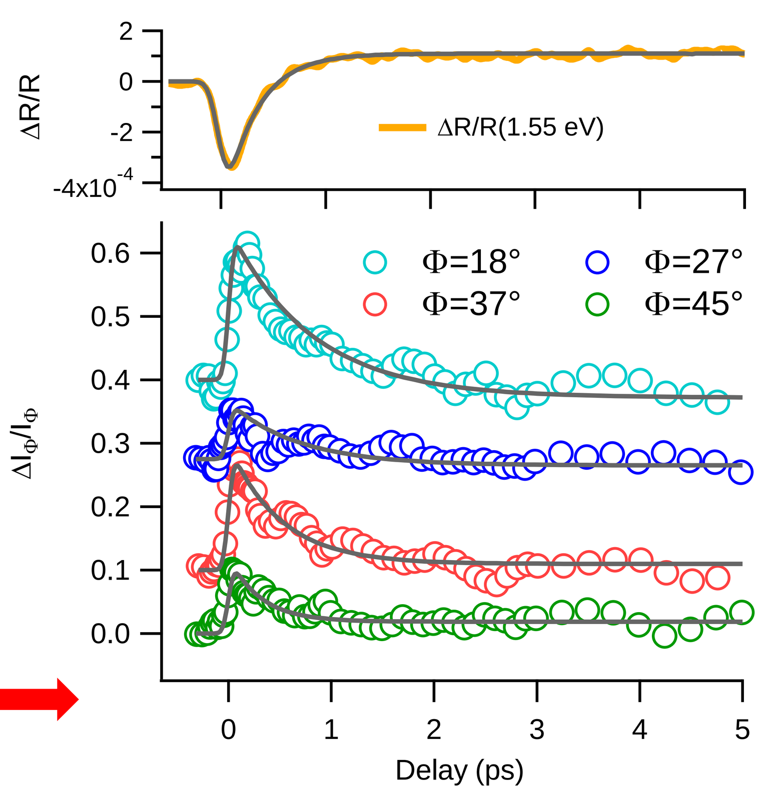


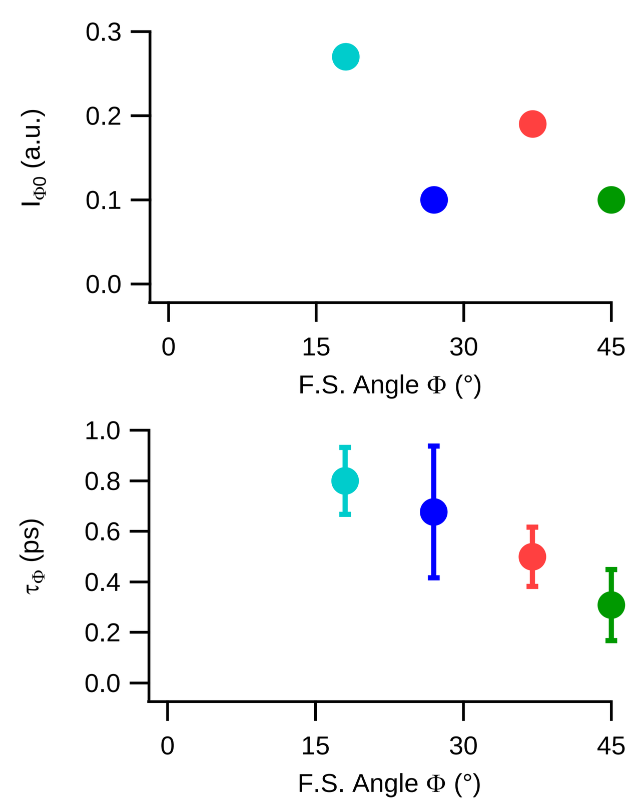




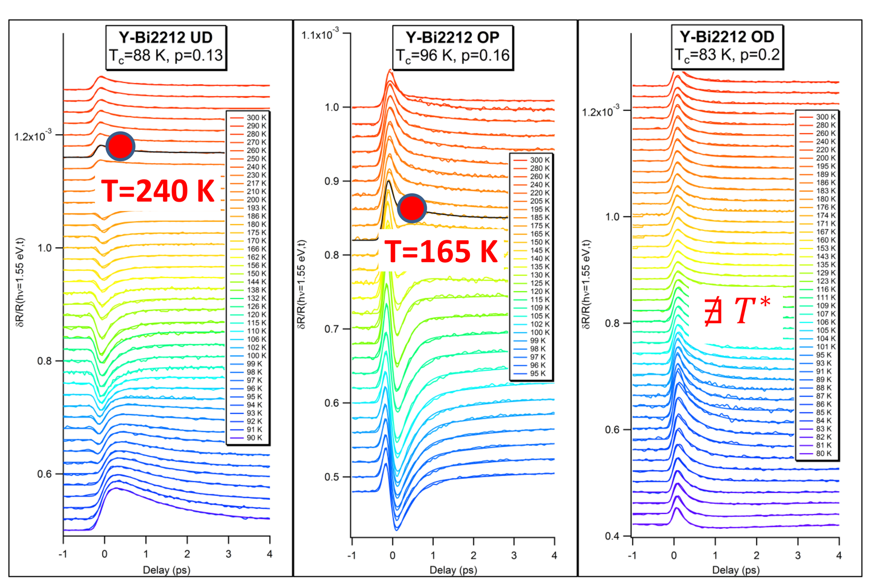
- **t < 1 ps:** $\delta\gamma < 0$ transient *decrease* of scattering rate (impulsive effect)
- **t > 3 ps:** $\delta\gamma > 0$ transient *increase* of scattering rate (thermic effect)

Connection between the impulsive effect and the non-equilibrium population





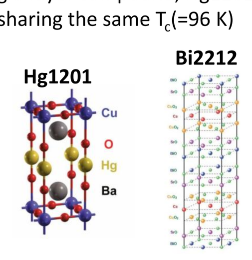
Drawing the Phase Diagram

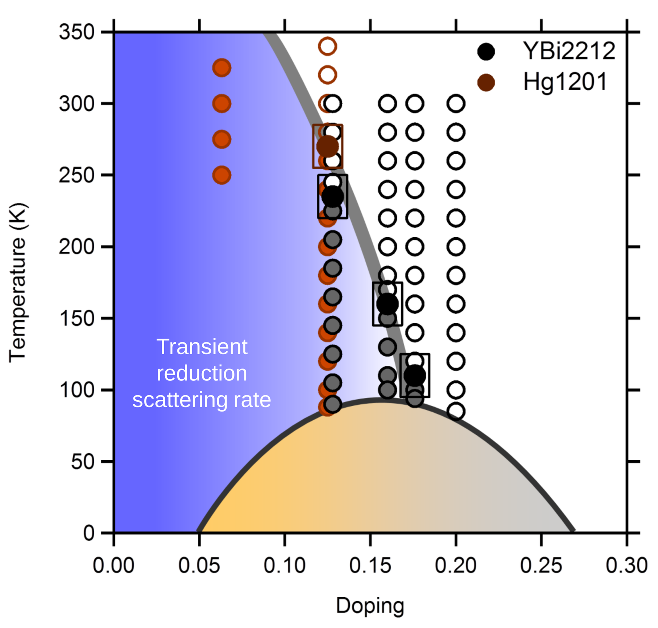


High resolution temperature scan at 1.55 eV probe photon energy

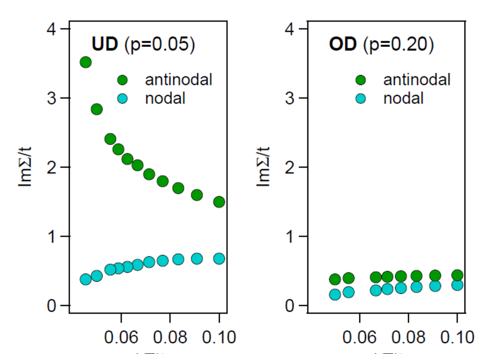
A negative component in the δR/R optical signal at 1.55 eV is the fingerprint of a transient reduction of the scattering rate. By following the evolution of this feature with temperature, we draw the phase diagram.

More generality is achieved by repeating the procedure for a single-layer compound, Hg1201, sharing the same T_c(=96 K)

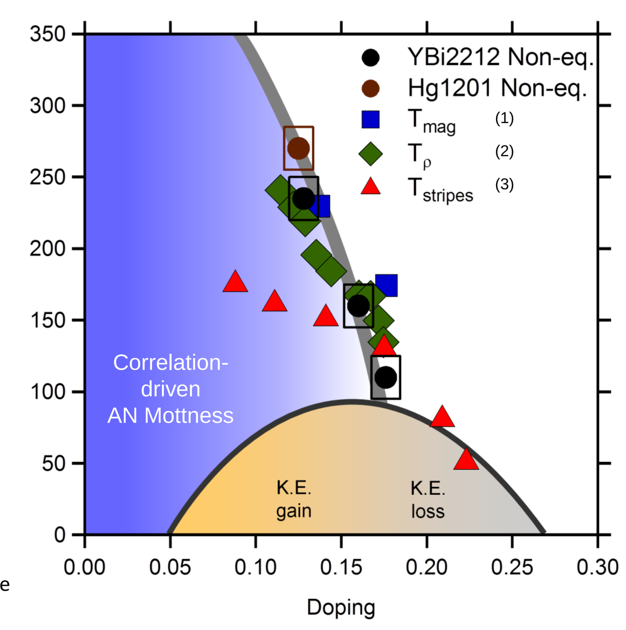




Electronic scattering rate (proportional to $Im\Sigma$) for N and AN QPs, obtained from the CDMFT solution of the 2D Hubbard model.

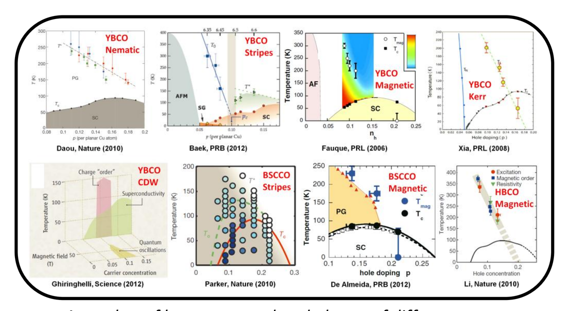


Providing energy to the gapped AN quasiparticles, they evolve from localized states to more mobile and metallic-like states. This explains our experimental findings, assuming a strongly correlated ground state



1) De Almeida-Didry, PRB **86**, 020504(R) (2012)
2) Raffy, Physica C **460**, 851 (2007)
3) Parker, Nature **468**, 677 (2010)

The Pseudogap T* line is reproduced by the CDMFT solution of the 2D Hubbard model, neglecting long range ordered phases while keeping the electronic correlations in a 2x2 Cu plaquette. Our conclusion is that the various broken symmetry phases recently detected in the Pseudogap emerge from a strongly correlated ground state, but are not its cause. They depend on the chemical and structural properties of the compound, as well as on the details of its Fermi surface. It remains to be clarified how these long range orders develop. It is worth to be noted that the anomaly in the resistivity follows intead the T* line.



A number of long range ordered phases of different nature has been detected in several families of cuprates.

Models and Methods

Extended Drude Model in the case of a non-constant DOS

A version of the EDM has been recently formulated for the case of a non constant electronic DOS, as it is the case for the system in the Pseudogap phase [cfr. Sharapov, PRB **72**, 134506 (2005)]

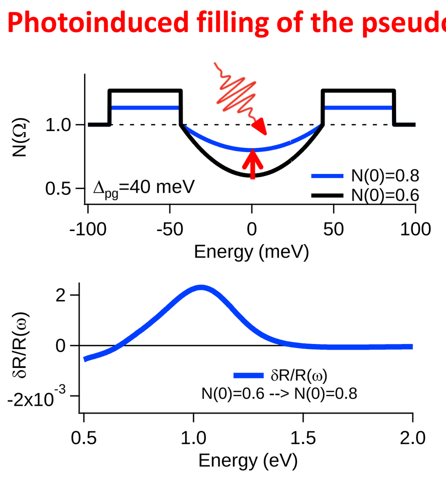
$$M(\omega,T)=\omega\left\{\int_{-\infty}^{+\infty}\frac{f(\xi,T)-f(\xi+\omega,T)}{\omega+\Sigma^*(\xi,T)-\Sigma(\xi+\omega,T)+i\gamma_{imp}}d\xi\right\}^{-1}-\omega$$

$$Im\Sigma(\omega,T)=-\pi\int_0^\infty \Pi(\Omega)\left\{\begin{matrix}\tilde{N}(\omega+\Omega,T)[n(\Omega,T)+f(\omega+\Omega,T)]+\\+\tilde{N}(\omega-\Omega,T)[1+n(\Omega,T)-f(\omega-\Omega,T)]\end{matrix}\right\}d\Omega$$

$$\varepsilon_D(\omega,T)=1-\frac{\omega_p^2}{\omega(\omega+M(\omega,T))}$$

R(ω) → ε(ω) → M(ω) → Σ(ζ) → Π(Ω), N(Ω)

Photoinduced filling of the pseudogap

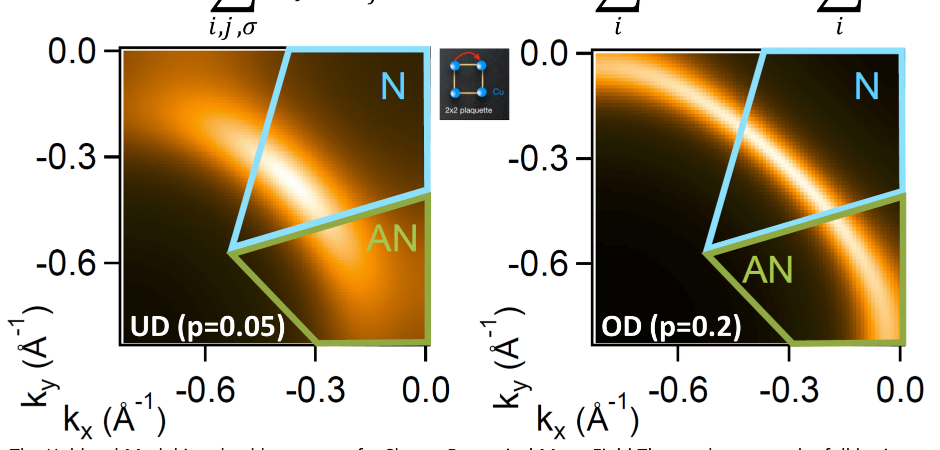


$$\tilde{N}(\Omega,T)=\begin{cases}\tilde{N}(0,T)+[1-\tilde{N}(0,T)](\Omega/\Delta_{PG})^2 & \Omega\le\Delta_{PG}\\ 1+2/3[1-\tilde{N}(0,T)] & \Omega\in(\Delta_{PG},2\Delta_{PG})\\ 1 & \Omega\le\Delta_{PG}\end{cases}$$

Hwang, PRB **83**, 014507 (2011)

2D Hubbard Hamiltonian

$$\hat{H}=-\sum_{i,j,\sigma}\left(t_{ij}\hat{c}^\dagger_{i\sigma}\hat{c}^\dagger_{j\sigma}+c.c.\right)+U\sum_i \hat{n}_{i,\uparrow}\hat{n}_{i,\downarrow}-\mu\sum_i\hat{n}_i$$



The Hubbard Model is solved by means of a Cluster Dynamical Mean Field Theory that maps the full lattice model onto a finite small cluster (here a 4-site cluster) embedded in an effective medium which is self-consistently determined as in standard mean-field theory. Doing so we fully account for the short-range quantum correlations inside the cluster. The anisotropy of the Fermi surface is correctly accounted for for both UD and OD doping levels.

Conclusions

Non equilibrium spectroscopies are fundamental in order to disentangle the intertwined degrees of freedom in strongly correlated materials: the onset of the pseudogap phase of cuprates is accompanied by a transient reduction of the optical scattering rate, whose observation is precluded to conventional spectroscopies at equilibrium. The solution of the 2D Hubbard model within CDMFT allowed us to interpret this peculiar behavior as originating from a strongly correlated ground state, in which the AN excitations are promoted by the the absorption of the pump pulse toward more metallic and delocalized states. The long range orders characterizing the pseudogap phase of cuprates emerge as a consequence of this strongly correlated ground state.