Snapshots of the retarded interaction of charge carriers with ultrafast fluctuations in cuprates

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Abstract: We measure the transient reflectivity of an high-T_c superconductor with an unprecedented temporal resolution (~15fs) demonstrating that on the 20-fs timescale electron dynamics is described in terms of charge carriers interacting with short-range antiferromagnetic fluctuations.

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The origin of high temperature superconductivity in cuprates remains controversial because of the elusive nature of the bosonic excitations mediating the formation of Cooper pairs [1]. Contrary to the case of BCS superconductors, electronic correlations are thought to play a decisive role in the formation of the superconductive condensate and they are expected to dramatically speed up the electron-boson scattering processes. The time scale of these processes is strictly linked to one of the key questions about the nature of the pairing mechanism: can the pairing mechanism be mediated by bosonic excitations of electronic origin, like spin fluctuations, whose dynamics is characterized by a retarded time scale of the order of ħ/2J (2J~300 meV), where J is the high energy cutoff of the bosonic spectrum? Alternatively to this hypothesis, the superconductivity could arise from the Hubbard model and the on-site Coulomb interaction U (where U>>J) would be the only relevant energy scale [2].

In the present work we developed an ad-hoc time-resolved experimental setup in the infrared spectral region (0.75-1.5 eV) and with unprecedented temporal resolution (~15 fs) [3], in order to investigate possible retardation effects in the dynamics of electron-boson scattering in cuprates [4]. After the interaction of a pump pulse, high energy fermionic quasiparticles (holes) are impulsively injected in the system and thermalize with the bosonic degrees of freedom of the system. This exchange energy process involves a heating of the bosonic excitations and consequently a change of the optical electron-boson scattering rate γ which is monitored by the probe pulse. The broad spectral coverage combined with the high temporal resolution of the setup allows us to monitor the evolution of the dielectric function and in particular of the scattering rate. This technique was previously employed to disentangle the electronic and phononic contributions to the generalized electron-boson Eliashberg function but the low temporal resolution (~100 fs) prevented the investigation of the possible retardation effect of the electron-boson interaction [5]. Fig. 1a reports the equilibrium reflectivity $R_{eq}(\omega)$ at T=300 K of a slightly underdoped $Bi_2Sr_2Y_{0.08}Ca_{0.92}Cu_2O_{8+\delta}$ crystal (YBi2212, hole doping p=0.13, T_c=83 K). R_{eq} is dominated by a Drude-like plasma edge, whose damping, in the framework of the Extended Drude model, accounts for the coupling between quasiparticles and the spectrum of the bosonic fluctuations. Furthermore it exhibits an isosbestic point at the frequency ω_i~1.1 eV. It is for this reason that a measurement of the reflectivity variation in a broad energy window around ω_i can provide information about the temporal evolution of the scattering rate variation. Fig. 1b reports the transient reflectivity of YBi2212 in the infrared region following a 7-fs visible pulse. Pump and probe pulses are obtained from synchronized ultra-broadband optical parametric amplifiers. We used a Singular Value Decomposition (SVD) based analysis to extract the principal component which contributes to the overall spectral response for all the probe wavelengths. The first SVD component $\Psi_1(\omega)$, reported in fig. 1c, accounts for more than the 97% of the total signal. We find that $\Psi_1(\omega)$ can be solely reproduced by assuming a transient increasing of the electron-boson scattering rate (black line in fig. 1c).

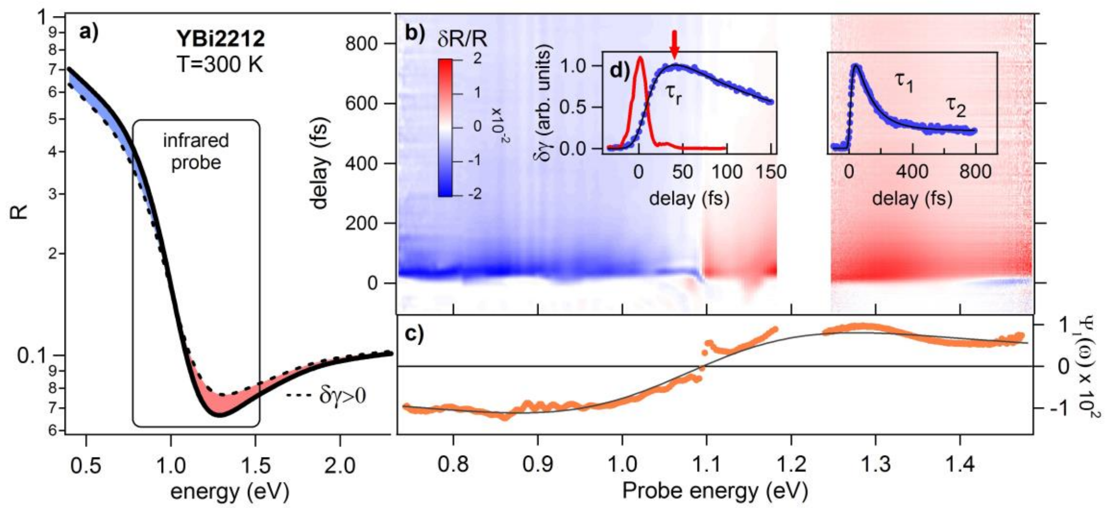

Figure 1 a) Equilibrium reflectivity spectrum (black line) of a YBi2212 crystal. The dashed line represents the reflectivity obtained by increasing the electron-boson scattering rate, in the extended Drude model. b) Transient reflectivity map $\delta R/R(\omega,t)$, measured across the isosbestic point. The measurements have been performed with an incident pump fluence of 0.7 mJ/cm^2. c) The first eigenfunction, $\Psi_1(\omega)$, obtained through the SVD is displayed (yellow dots). The fit obtained by increasing the total scattering rate is reported as a black line. d) The time-dependent scattering rate variation, $\delta\gamma(\tau)$, is reported (blue dots). The black lines are the fit to the data of the function $(1-\exp(-t/\tau_r))\cdot(I_1\exp(-t/\tau_1)+I_2\exp(-t/\tau_2))$ convoluted with a gaussian pulse accounting for the experimental resolution. The red line is the cross-correlation trace between the pump and the probe pulses which sets the temporal resolution of the experiment to 19±2 fs.

Within this interpretation the dynamics of the scattering rate corresponds to the principal SVD temporal trace $\delta\gamma(\tau)$, reported in fig. 1d. The variation of the temporal eigenvalue reaches the maximum value at a finite delay time (~40 fs) with respect to the pump excitation (red arrow in fig. 1d). The intrinsic rise time of the scattering rate (τ_r=16±3 fs) is estimated from the fit and takes into account the experimental resolution of the setup carefully characterized by XFROG and estimated to be to be 19±2 fs). The two subsequent decay processes with time constants τ_1~90 fs and τ_2~1 ps, are attributed to thermalization processes with respectively strongly coupled optical phonon modes and with the rest of the lattice vibrations. This analysis clearly indicates that the energy transfer between photoexcited holes and the bosonic bath takes a finite time to occur and the electron dynamics in the doped cuprates can be captured by an effective model in which the charge carriers exchange their energy with bosonic modes on a very fast (<20 fs) but fully resolved temporal scale.

The extremely fast time scale of this relaxation process, strongly suggests that short range antiferromagnetic excitations are involved as the mediators of the interaction between charged carriers. This physical scenario is corroborated by theoretical calculation describing the non-equilibrium dynamics of the photo-excited holes in the t-J Hamiltonian giving a very similar timescale for the electron-boson coupling[6]. Our results represent a new time-domain benchmark for realistic microscopic models of the pairing in high-temperature superconductors and pave the way to unveiling the pairing mechanisms in a wealth of intriguing systems, like different families of cuprates and iron-based superconductors.

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