LEES 2010 – *Low Energy Electrodynamics in Solids* – Les Diablerets, 5-10 July 2010

Broadband Ultrafast Time-Resolved Spectroscopy: a new technique to investigate the electron dynamics in strongly-correlated systems

Federico Cilento[1,2,*], Claudio Giannetti[3], Gabriele Ferrini[3], Stefano Dal Conte[4], Tommaso Sala[3], Giacomo Coslovich[1,2], Matteo Rini[5], Andrea Cavalleri[6,7] and Fulvio Parmigiani[1,8]

1. Department of Physics, Università degli Studi di Trieste, Trieste I-34127, Italy; **2**. Laboratorio Nazionale TASC, AREA Science Park, Basovizza, Trieste I-34012, Italy; **3**. Department of Physics, Università Cattolica del Sacro Cuore, Brescia I-25121, Italy

4. Department of Physics A. Volta, Università degli Studi di Pavia, Pavia I-27100, Italy; **5**. Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA; **6**. Condensed Matter Division, Max Planck Research Department for Structural Dynamics, University of Hamburg, 22607 Hamburg, Germany; **7**. Department of Physics, Clarendon Laboratory, University of Oxford, OX1 3PU Oxford, United Kingdom; **8**. Sincrotrone Trieste S.C.p.A., Basovizza, Trieste I-34012, Italy

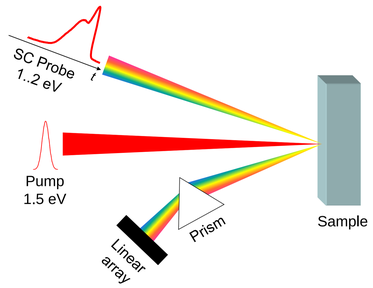
*. for correspondence and requests: federico.cilento@elettra.trieste.it

INTRODUCTION

We have developed a novel technique to overcome the main limit of the conventional pump-probe technique, which allow only to study the non-equilibrium optical properties of a system at a fixed probe energy (typically 1.5 eV). Introducing a supercontinuum (broadband) probe pulse this limitation is overcome: a true non-equilibrium spectroscopy with high spectral resolution (5 nm) can be achieved. An experimental apparatus which combines the peculiarities of the ultrafast time-resolved approach (high temporal resolution: 120 fs in our setup, high signal to noise ratio, sensitivity to the electronic dynamics) with the ones of the spectroscopic approach (energy resolution) allowed us to study – through the nonequilibrium dielectric function – the transient electronic dynamics in several strongly correlated materials. The use of a cavity-dumped oscillator as a laser source allow us to explore different excitation regimes, opting for a high statistic or a low average heating.

TIME-RESOLVED BROADBAND SPECTROSCOPY

This technique allow to study the transient dynamics of the dielectric function $\varepsilon(\omega)=\varepsilon_1(\omega)+i\varepsilon_2(\omega)$ of several solid state systems. The supercontinuum (SC) probe pulse is generated with a photonic crystal fiber seeded with 120 fs - 800 nm pulses.



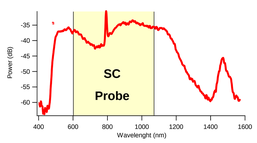
The setup works in reflectivity and measures the *differential optical properties* (difference between the pump-excited system's optical properties and the unexcited one's):

$$\frac{\Delta R}{R}=\frac{R_{excited}-R_{un-excited}}{R_{un-excited}}$$

The dielectric function $\varepsilon(\omega)$ establishes the connection between the measured optical properties and the material's electronic structure:

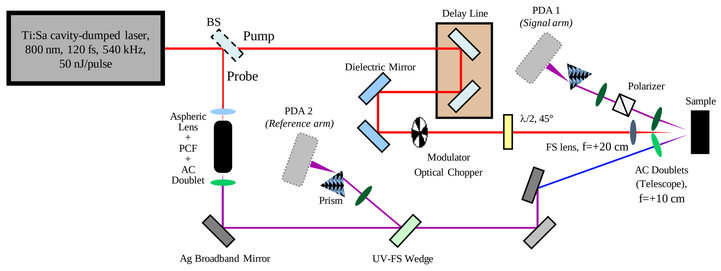
$$R(\omega)=\left|\frac{1-\sqrt{\varepsilon(\omega)}}{1+\sqrt{\varepsilon(\omega)}}\right|^2$$

Spectrum produced by the PCF.



PUMP-SUPERCONTINUUM PROBE SETUP

Our setup in detail: the SC spectrum is dispersed with equilateral prisms and imaged onto linear arrays: at each pixel correspond about 5 nm of spectral content. Taking a reference for the SC intensity before the interaction with the sample, and normalizing the reflectivity signal, a s/n ratio better than 10^4 is reached.



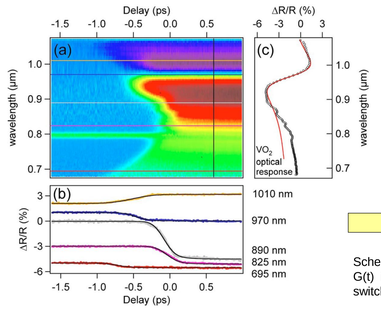
THE CHARACTERIZATION OF A COLOURED LIGHT PULSE

The generation of a white-light continuum with a few nJ/pulse is only achievable with photonic crystal fibers. The wavelengths of these pulses are distributed in time following the PCF dispersion characteristic: the use of this pulse as a probe in time-resolved experiment thus requires a careful characterization.

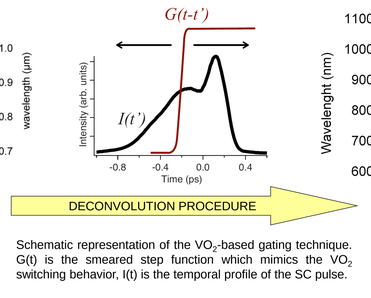
We adopt three techniques to obtain a *spectrogram* (time-energy map) of the pulse:

* Optical switching, exploiting the photoinduced phase transition of VO_2
* SFG (sum-frequency-generation) in an angle-dithered NL BBO crystal
* Two-photon-absorption in a ZnSe window

OPTICAL SWITCHING (GATING) WITH VO_2 FILM

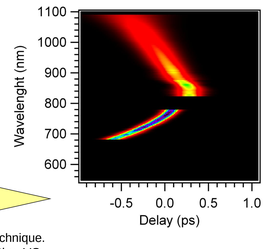


Time-resolved traces on VO_2 evidence the switching behavior of the material, due to the photo-induced structural and electronic phase transition, which manifests in an abrupt change of the optical properties.

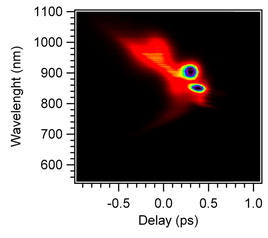


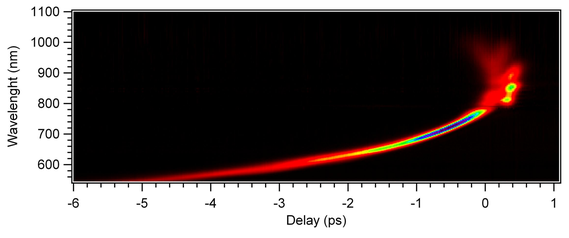
Schematic representation of the VO_2-based gating technique. G(t) is the smeared step function which mimics the VO_2 switching behavior, I(t) is the temporal profile of the SC pulse.

XFROG (BBO NL CRYSTAL)



TWO PHOTON ABSORPTION IN ZnSe





The plots represent the spectrograms of the SC pulse, as obtained with the different techniques. The VO_2-based ***switching technique*** is the only working in reflectivity, and its spectral response covers the entire spectral range. Due to the switching behavior, it can be used for pulse diagnostics. On the other hand, it requires a deconvolution procedure and has a lower temporal resolution, because of the VO_2 finite switching time (~80 fs) [1]. XFROG and two-photon absorption, being ***gating tecnhiques*** (based on extremely fast electronic processes), offer higher temporal resolution, which allow to distinguish the solitons present in the pulse.

These techniques are complementary and provide equivalent results about the temporal duration of the pulse: 120 fs in the UV-visible region, and up to 200 fs in the IR region.

TIME-RESOLVED BROADBAND SPECTROSCOPY

Time-resolved reflectivity in the spectral region between 1 and 2 eV has been measured on cuprate superconductors (Y-Bi2212 and Hg1201) and on HOPG, with respect to the delay τ between excitation (pump pulse) and probing (probe pulse).

THE DRUDE-LORENTZ MODEL

Transient reflectivity is interpreted with a differential Drude-Lorentz model: a static model is constructed for the static optical properties; considering the difference with a slightly-modified model allow to take into account the effect of excitation.

$$\varepsilon(\omega)=1+i\frac{4\pi}{\omega}\sigma(\omega)=\varepsilon_{\inf}+\sum_j\frac{\omega_{p\ j}^2}{\omega_{0\ j}^2-\omega^2-i\omega\gamma_{\ j}}\qquad \omega_{p\ j}^2=\frac{4\pi n_j e^2}{m^*}$$

where: ω_{pj}: plasma frequency – oscillator strength of each oscillator

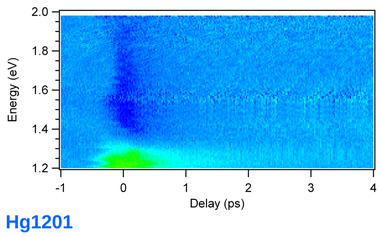
ω_{0j}: central frequency of each oscillator (zero for Drude term(s))

γ_i: damping term: oscillator width

Y-Bi2212

Optimally Doped Y-Bi2212 ($Bi_2Sr_2Ca_{0.92}Y_{0.08}Cu_2O_{8+\delta}$, δ=0.16), T_c=96 K

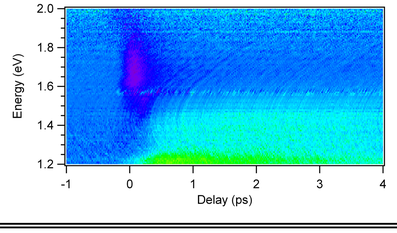
T=100 K (pseudogap state), $\mathcal{F}$=10 μJ/cm², rep. rate 540 kHz



Hg1201

Optimally Doped Hg1201 ($HgBa_2CuO_{4+\delta}$), T_c=94.5 K

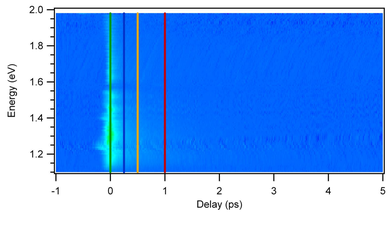
T=15 K (superconducting state), $\mathcal{F}$=20 μJ/cm², rep. rate 540 kHz



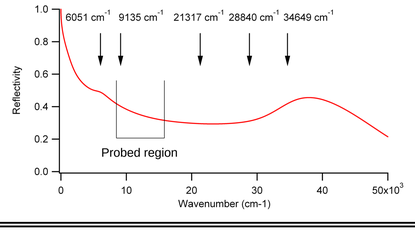
HOPG

Highly-Oriented-Pyrolitic-Graphite

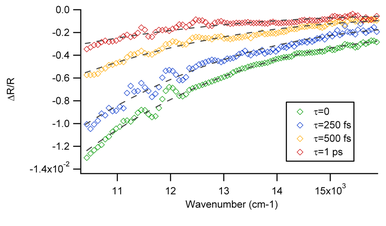
T=300 K, $\mathcal{F}$=500 μJ/cm², rep. rate 1 MHz



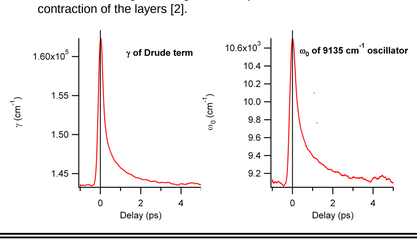
Static reflectivity model of HOPG. The central frequencies of Lorentz oscillators are indicated.



Traces at selected delays – and their fit – are reported.



Temporal evolution of the free parameters of the fitting procedure, for the trace at delay zero. The fact that the 9135 cm^{-1} oscillator moves toward higher energies is compatible with a laser-induced contraction of the layers [2].



CONCLUDING

* A pump-probe apparatus with the peculiarity of exploiting a broadband supercontinuum pulse as a probe, has been developed and fully characterized.

* The use of a cavity-dumped oscillator as laser system allowed us to explore different excitation regimes, while maintaining a high statistic. This system allowed us to study the low energy electronic dynamics (in the energy range 1 .. 2 eV) in strongly correlated systems.

FUTURE

* Extension of the probed spectral region toward the mid-IR (1000-1600 nm), with InGaAs linear sensors.

* The possibility to generate far-IR radiation (3-10 μm) to be used as a probe, exploiting a NL DFG (difference-frequency generation) process, is being explored.

[1] A. Cavalleri et al., Phys. Rev. B **80**, 161102 (2004)

[2] F. Carbone et al., Phys. Rev. Lett. **100**, 035501 (2008)

Further details on: Appl. Phys. Lett. **96**, 021102 (2010)