Research Framework, Prof. Daniele Fausti

Quantum spectroscopies for quantum materials (Q4Q)

The holy grail of controlling material's properties on sub-picosecond timescales led, in the last decade, to a large number of experiments exploring phase transformations photo-induced by ultrashort light pulses in a large set of materials [1, 2, 3, 4, 5]. The basic idea of these experiments is to drive the phase transformation by means of ultrashort light pulses "*impulsively*" injecting a large number of excitations. The photo-excitation within time windows shorter than the characteristic times of the relaxation processes drives matter into highly off-equilibrium transient regimes characterized by anomalous energy distribution between electrons, ions, and spins[6, 7, 8]. This can strongly perturb the interaction among the different degrees of freedom and thereby results in the formation of meta-stable "phases" not always reachable under quasi-equilibrium adiabatic transformations.

In order to achieve such a control, the most commonly explored schemes are based on sudden photo-injection of an excess of high energy ($h\nu$>1eV) electronic excitations by ultrashort light pulses, eventually leading to photo-induced-phase-transitions (PIPT).[9] The major emerging limitation of this approach, hampering bi-directional optical control, lies in the fact that while the optical switch between different phases can be driven within picoseconds, the recovery is generally limited to much longer times and is determined by slower relaxation processes, ruled by the thermodynamic characteristic of the sample.

These limitations can in principle be overcome using different excitation schemes allowing for a resonant excitation of low energy modes in solids such as vibrations and magnetic excitations. **In contrast to light-induced phase transitions, phase transitions driven by the coherent control of low lying excitation are directly due to the large-amplitude low-frequency excitation of the bosonic modes produced by the electromagnetic field and are not related to hot-carrier injection.** This means that thermal relaxation processes do not limit the coherently controlled phase transformation and bi-directional changes of material properties at rates exceeding the thermodynamic restrictions become conceivable.

In spite of the infancy of this approach, some important results have already been achieved in manganese-based oxides where the resonant excitation of a Mn-O mode can trigger a transition in the electronic properties from insulator to metal [10] and orbitally-order to disordered [11]. With a similar approach in the past years I have been focussing on using pulsed infrared electromagnetic fields to control the formation and relaxation of superconducting phases on ultrashort timescales [5, 12, 13, 14] and, more recently, evidences of light driven quantum coherent transport up to ambient temperature have been reported in YBCO [15,16]. **The possibility of controlling quantum coherent states in matter, by means of pulsed electromagnetic fields, unlocks a new regime of physics where thermodynamic constraints can be beaten and quantum coherence might be sustainable at ambient temperatures.**

The mechanism leading to such spectacular effects remain elusive: Can the coherence of the resonantly excited phonon modes be "transferred" to the electronic bath? In addition to this, major questions remains concerning the nature of the photo-induced states induced both with high- and low-photon energy: To what extend (in which systems) the transient states observed are thermodynamic phases? Should they be considered quasi-equilibrium system with well defined thermodynamical quantities (chemical potential, temperature, etc…) or should they rather be described as a non-equilibrium «tangle» between the different material constituents whose properties are mostly determined by the dynamical coherent coupling between the different degrees of freedom? Further, are the transient states spatially uniform or dynamic phase separation occurs? Can we control the dynamical formation of transient phases?

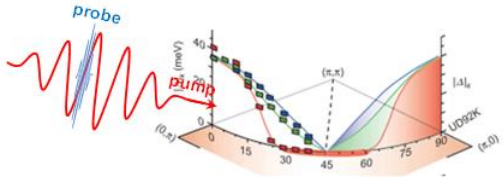

Fig.1 : The idea of the research project proposed is to combine the excitation with long wavelength pump pulses (red) with high frequency short probe pulses (blue). The probe pulses shorter than the wavelength of the pump field will allow for the measurements of responses of the SC with amplitude and phase selectivity. In addition to this (see text), non classical statistical features of the probe pulses will be measured.

In order to achieve a full control of transient non-equilibrium responses, we need new spectroscopic tools capable of addressing the time evolution of the fluctuations of different degrees of freedom in quantum materials. The leading idea of our research is that **through the light-matter interaction, the quantum state of the probe light pulse will carry information on the transient pump-induced state of the material and, therefore, the measurement of the** statistical properties of light provide the mean to retrieve detailed information on the thermodynamical quantities leading the non-equilibrium response. In this framework, we have demonstrated in the last 3 years that photonic fluctuations (of both classical and quantum nature) contain viable spectroscopic information which are lost in standard time domain measurements and provide a unique and powerful asset to address fluctuations of different low energy degrees of freedom in real materials [17, 18, 19]. While in standard time domain experiments the evolution of the response of a material is typically averaged over the illuminated area as well as over many pump and probe measurements repeated stroboscopically, we have recently developed a new generation of time resolved spectroscopy that combines quantum optic techniques with the traditional pump and probe approach.

Title of the PHD project:

Imprinting multimode optical correlations in ultrashort light pulses by means of superconducting currents

In the research framework described above which involve activities accross the fields of condensed matter, ultrafast optics and quantum information, we propose a PhD research project dedicated to undestand the interaction of ultrashort light pulses (10s of femtoseconds long) with superconducting currents in high temperature superconductors. One of the major open question in cuprate superconductors is to understand what is the role of localized electronic excitation of «molecular nature» (such as the Charge transfer excitation or onsite localized dd-electronic transition) in the formation of the superconducting phase. What is the interaction between the itinerant charges described by a collective superconducting wave function with localized electronic excitation on the Cu-O bonds? Can localized electronic excitation provide a «glue» for pairing electrons to form Cupper pairs? Can we use optically driven local electronic excitation to control the collective electronic properties of the condensate?

The PhD project proposed will consist of a research program to address those questions by means of the unique set of time resolved spectroscopied available within Q4Q. Through the different research projects carried out in the last years we developed a unique set of spectroscopic tools combining standard quantum optics techniques with time domain measurements and we are now capable of revealing how the interaction with a photo-excited sample can modify the photon number statistic (integrated in frequency) and introduce quantum correlations between the different spectral component of the radiation (frequency resolved)[14, 17, 18, 19, 20]. In particular, as depicted in Fig. 1, by performing non-equilibrium experiments based on long wavelength pump pulses (in the mid-IR or THz) and short wavelength probes (visible near-IR) we can drive low frequency superconducting currents and measure the effects of such a pump on the optical properties associated to localized electronic excitation of Charge transfer and dd-transition nature. Further, by combining such mid-IR excitation to the measurements of photon number fluctuation in low photon-number probe pulses we can possibly unveil the quantum correlations between localized excitations which are mediated by the coupling to itinerant superconducting charges. We stress that such detailed information about local behaviour in the material is obscured in standard time domain experiments based on mean value response.

The candidate interested in such a position will therefore have the unique opportunity to build a solid experience accross the different research fields of condensed matter, laser physics, ultrafast non-linear spectroscopies and quantum information. Should you be interested please contact me for further information at: dfausti@units.it or Tel. +390403758449.

Example References (Bold Fausti's publications)

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