Ultrafast Dynamics of Strongly-Correlated Electron Materials investigated by Time-Resolved Table-Top Spectroscopies

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A PhD Project at the T-ReX Laboratory at Elettra-Sincrotrone Trieste is available. The topics of this project comprise both the commissioning of innovative ultrafast photon sources and the use of time-resolved optical and electron spectroscopies for the study of the functional properties of complex materials. In particular, the advanced characterization of novel photon sources delivering ultrashort (100 fs) photon pulses in the Vacuum-Ultra-Violet (VUV) and Extreme-Ultra-Violet (EUV) spectral regions, seeded by ultrafast table-top laser sources, and devoted to time-resolved photoemission experiments, will provide the candidate with a deep expertise about highly-nonlinear optical phenomena. Such developments, together with the present spectroscopies, will permit the study of the out-of-equilibrium femtosecond electron dynamics measured by Time&Angle Resolved PhotoElectron Spectroscopy (TR-ARPES) [1] of strongly correlated materials, as well as their time-resolved optical properties.

Two innovative sources of EUV photons are under commissioning at T-ReX, at present. A High Harmonic Generation (HHG) setup is providing tunable photon energies from 15 to 50 eV. The setup produces the odd-order harmonics of the output wavelength at 1030 nm (1.2 eV) from a Yb:KGW laser source, in argon gas. The setup will be soon upgraded to improve the overall time-resolution attainable in time-resolved experiments. Furthermore, a setup for 9HG (ninth harmonic generation) producing 10.8 eV photon pulses of the laser fundamental at 1.2 eV in xenon gas is under development. It is working at 1 to 2 MHz repetition rate. Here the goal will be to introduce the possibility to obtain circularly-polarized pulses.

These advancements will pave the way to a number of novel experiments that will be the core of the PhD project. In particular, the 10.8 eV source and the HHG beamline, both operating at high-repetition rate (200-2000 kHz), are particularly well suited for TR-ARPES studies with high energy and momentum resolution, high signal statistics and low space charge, allowing to access the entire Fermi surface of correlated materials. The scientific themes that will be covered during the proposed PhD project will strongly benefit from these novel sources. The common thread of the proposed investigations concerns the study of materials with strong electronic correlations and complex (anisotropic) Fermi surfaces. Electrons occupying different regions of the Fermi surface brings complementary information about ordered phases (for example charge density wave, superconductivity), competing phenomena (for example superconducting gap and pseudogap) or a different degree of electronic correlation (the so-called 'Selected Mottness'). Hence, high-resolution TR-ARPES at large momenta is the best suited technique in order to tackle these themes: i) on the one side, momentum resolution allows to disentangle phenomena localized at precise momentum regions; ii) the non-equilibrium approach allows decoupling ordered phases or degrees of freedom that at equilibrium coexist on similar energy scales. Thanks to the non-equilibrium approach, the ground state of the material can be manipulated and photoinduced phase transitions between ordered phases can be triggered. The focus of the project will be the study of the interplay among these ordered phases connected through photoinduced phase transitions, in order to determine their mutual interactions (competing, coexisting, …) by their non-equilibrium dynamics.

In detail, the topics that will be tackled during the PhD course are described below. Other topics will likely be explored, depending on the evolution of the community and the availability of new materials in the next few years.

• **Studies of cuprate superconductors**

In the field of copper-based high critical temperature superconductors, a large number of fundamental issues are left opened, despite the intense efforts of the past >30 years. The non-equilibrium approach and in particular Time-Resolved ARPES are specifically best-suited to tackle the following open issues: i) the understanding of the interplay between the (d-wave) superconducting gap and the pseudogap phase (having its fingerprint localized in the antinodal region of the Brillouin zone [2]); ii) the interplay between superconductivity and charge density wave in cuprates (the signature of the charge density wave is thought to be hidden at the 'hot spots' of the Fermi arc [3]). These issues will take advantage of the possibility of photoinducing phase transitions between the superconducting state to the pseudogap state and by the possibility to melt the charge-density-wave order, as allowed by the pump-probe technique.

• **Studies of Alkali-metal-doped fullerenes**

Very recently alkali-metal-doped fullerenes, well known for they can sustain superconductivity up to ≈40 K, have been brought back to the attention of the physics community after the discovery of a possible light-induced superconducting state far above their critical temperature [4], after strong infrared irradiation. However, this finding is highly controversial and debated [5]. In order to try to shed light in this field, the idea is to use TR-ARPES to directly measure the light-induced modification of the electronic states at the Fermi level, where superconductivity manifests with the opening of the superconducting gap. Very briefly, the debate about the development of this 'more metallic' photoinduced state sees on one side the idea that it is induced by a coherent and resonant optical excitation of molecular vibrations, that enhance the conductivity. On the other side, arguing that the phenomenon is observed in a broad pumping frequency range that coincides with the mid-infrared electronic absorption peak still of unclear origin, a different mechanism was proposed. Since the broad absorption peak represents a "super-exciton" involving the promotion of one electron from the t_{1u} half-filled state to a higher-energy empty t_{1g} state, the IR-induced excitons could act as a sort of cooling mechanism that permits transient superconducting signals to persist up to temperatures much larger than T_C. TR-ARPES can contribute to this debate, by measuring directly the nature of the photoinduced metallic state at the Fermi level.

• **Studies of iron-based superconductors**

Iron-based superconductors have a Fermi surface composed by all the 5 Iron d-orbitals [6,7]. All these electronic states become gapped in the superconducting state, and the natural question is whether the superconducting transition is driven by a conventional BCS mechanism or it is driven by electronic correlations. It was predicted that the hole- or electron-states at the Fermi surface have a different degree of electronic correlation, also termed 'Selective Mottness' [8], that can point toward a purely electronic mediated coupling. This model will be analyzed by TR-ARPES experiments, that can directly measure the correlation strength of each orbital thanks to the measurement of its single-particle lifetime, and also how the photo-induced superconducting-to-metal phase transition is affecting the multiple gaps at the Fermi level. This work will be performed in collaboration with SISSA, where researchers are performing calculations about the 'Selective Mottness' and will provide theoretical support to interpret results by TR-ARPES [9,10].

References

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