Ultrafast infrared nano-spectroscopy and nano-imaging of unconventional superconductivity in cuprate and pnictide high-T_c systems

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High-T_c superconductivity is surpassed by few, if any, other unsolved problems in contemporary physics in terms of its richness, complexity, impact on other fields, and potential technological importance. Before 2008, the term "high-T_c superconductivity" was reserved for copper oxides or cuprates (maximum T_c~160 K). The discovery of superconductivity in Fe-based pnictides compounds with T_c almost as high as 60 K has prompted a renewed surge of research activity. Systematic studies have revealed both common and contrasting trends in the cuprate and pnictide superconductors. In both classes of materials, superconductivity occurs in close proximity with other electronic phases: an antiferomagnetic (AF) Mott insulator state in the case of the (hole-doped) cuprates and metallic spin density wave (SDW) phase in the case of the pnictides. One common aspect of Fe- and Cu-based systems is a propensity towards electronic and magnetic self-organization, leading to dynamic inhomogeneities at nano-to-mesoscopic length scales. The inhomogeneities have so far been documented with quasi-static probes such as nuclear magnetic resonance, neutron and x-ray scattering along with scanning tunneling microscopies. In contrast, ultrafast infrared/optical studies enable characterization of dynamics and fluctuations in superconductors. However, such experiments have been carried out using diffraction-limited optics and therefore probe length scales that inform us of a "mixed" response involving contributions from the multiple electronic, chemical and structural phases occurring in real materials while averaging over nanoscale heterogeneities. As a result, the interpretation of area-averaged ultrafast optics data has often remained both complicated and ambiguous.

Our team proposed an effort to investigate the intrinsic spatio-temporal electromagnetic response of pnictides and cuprates. The goal of the proposed work was to carry out pump-probe infrared (IR) spectroscopy and imaging studies with 10-nm spatial resolution and a temporal resolution of tens of femtoseconds.

Summary of published work. Co-PIs have published 10 peer reviewed publications including: one article in *Nature Photonics*[1], one article in *Nature Materials*[2] and two articles in *Nature Physics*[3,4], one paper in *Physical Review Letters*[5], articles in *Science*[6] and *Reports on Progress in Physics*[7] and several other[8,9,10]. Most publications are co-authored by more than one co-investigator.

Significant accomplishments.

Pump-probe spectroscopy: ultra-high optical fields and ultra-high stability. Developmental work by *Averitt* and *Basov* has resulted in experimental ultrafast optical capabilities using a 1KHz amplifier for mid-infrared pumping (1.2–17 μm → 75 meV–1.0 eV) achieving fluence of ~5 mJ/cm^2 and high-field terahertz pulse generation (0.1–2 THz) with ultra-high peak fields in excess of 400 kV/cm. The dynamics initiated with phonon pumping or electric field driving can be probed at THz, mid-IR, and visible frequencies. Initial phonon pumping studies have been performed on nickelates. Experiments investigating photo-enhanced superconductivity of $La_{2-x}Ba_xCuO_4$ (x =11.5%) are underway to corroborate and extend the measurements of the Hamburg group. In parallel, *Averitt* has developed a high repetition rate laser beamline (50KHz–1MHz) to obtained intense fields (up to 50 kV/cm) with ultra-high stability needed for high finesse measurements including experiments on FeSe/SrTiO3 interfaces.

Advanced nano-infrared spectroscopy and imaging. Basov and *Averitt* demonstrated for the first time pump-probe nano-imaging using amplified lasers offering ultra-intense electric fields utilizing a high repetition rate beamline to provide ultra-high stability. These innovations facilitated the discovery of novel hidden phases in correlated oxides including manganites[2] and V_2O_3[11]. These scanning probe experiments are performed in the setting of a unique cryogenic nano-infrared apparatus with ultra-high stability of the suspended scanner developed by *Basov*. *Basov, Hone* and *Fogler* reported for the first time on hyper-spectral pump-probe nano-imaging[1]. This novel experimental technique combines broad-band infrared spectroscopy with 10-nm spatial resolution imaging. This new novel system is essential for the proposed experiments on local pump-probe studies of transient superconductivity.

Superconductors integrated with meta-materials and van der Waals heterostructures. Simulation, synthesis, and terahertz characterization by *Averitt, Basov,* and *Hone* has enabled sculpting, localization, and enhancement of electromagnetic fields in superconductors using metamaterial resonator devices. These capabilities are crucial for nonlinear far-field and near-field studies providing the means to achieve localized phonon pumping or field-driven effects in superconductors. The initial work has focused on THz frequencies and we have developed metamaterial "tapes" that can be easily applied (and removed) to single crystals samples. For $La_{2-x}Sr_xCuO_4$ (x =15%) single crystals we have investigated coupling between the c-axis Josephson Plasmon Resonance (JPR) and split ring resonators (SRRs). By creating a series of tapes with differing resonator dimensions, we were able to sweep the SRR mode across the JPR to investigate mode coupling. THz spectroscopy data reveal a strong interaction between the superconducting condensate and the SRRs, providing a new approach to interrogate and potentially augment superconductivity. In a parallel development *Hone* has designed and fabricated vdW heterostructures where few monolayers of superconductor are integrated in a gated device with hBN gate insulators and graphene transparent electrodes. Encapsulation with hBN capping layers allows one to preserve air-sensitive specimens of FeSe.

Non-equilibrium phase diagram of correlated electron materials. Theoretical work of *Millis*[12] demonstrated that the dominant symmetry-allowed coupling between electron density and dipole active phonon modes implies an electron density-dependent squeezing of the phonon state and provides an attractive contribution to the electron-electron interaction. This effect is independent of the sign of the bare electron-phonon coupling whereas the magnitude of this effect is proportional to the degree of laser-induced phonon excitation. Reasonable excitation amplitudes lead to non-negligible attractive interactions that may cause significant transient changes in electronic properties including superconductivity. The mechanism is generically applicable to a wide range of systems, offering a promising route to manipulating and controlling electronic phase behavior in novel materials.

Plasmon amplification. Theoretical work of *Fogler*, which is motivated experimental results from our team[13], studies dynamical evolution of the collective modes of a photoexcited electron-hole plasma. The key idea is that the gradual change of the instantaneous frequency of the mode is analogous to the adiabatic variation of parameters of a harmonic oscillator. Based on this analogy, *Fogler* showed that depending on the magnitude of the loss, the mode amplitude may increase with time. This amplification pathway does not require any gain media; it occurs during the natural relaxation of the plasma. The principle of adiabatic amplification is general; in Ref.[13] we discuss a specific case of thin-film superconductors, which are expected to be low-loss plasmonic media. We also analyzed the detection of amplified plasmons via pump-probe nano-optical techniques.

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