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Current Perspectives

# Ultrafast spectroscopy of quasiparticle dynamics in cuprate superconductors

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ABSTRACT

Ultrafast pump-probe spectroscopy is a powerful tool to study the nonequilibrium dynamics in high-Tc cuprate superconductors. The photo-induced quasiparticle (QP) dynamics revealed by pump-probe spectroscopy are sensitive to the near-Fermi level electronic structures. Here we review several selected examples to illustrate the enduring challenges including pairing glue, phase separation, and phase transitions in cuprate superconductors. We also present the data obtained on thin films of $YBa_2Cu_3O_{7-\delta}$ in connection to these issues.

## 1. Introduction

The nature of high-temperature superconductivity (HTSC) in cuprate compounds has been a central challenge in modern condensed-matter physics [1]. The past two decades of vigorous research has uncovered a plethora of exciting phenomena mapped on to the complex and surprisingly asymmetric (electron vs. hole doping) phase diagrams. To understand the microscopic mechanisms, it is essential to elucidate the physics behind the exotic properties and at the same time to identify those features that are quintessential to the phenomena of HTSC. Despite great successes in unraveling the electronic and magnetic properties of cuprates by time-integrated techniques, such as electric and thermal transport [2], tunneling [3], angle-resolved photoemission (ARPES) [4], and conventional optical spectroscopies [5], there are still unresolved challenges in disentangling the interplay between the elementary excitations. To this end, ultrafast optical spectroscopy has been extensively exploited as a unique probe with the potential to explore the physics of elementary excitations on a temporal scale by elucidating spectrotemporal features for specific physical processes [6–8].

Historically, since the early 80s, the technique of pump-probe femtosecond spectroscopy has been applied as a tool for investigating nonequilibrium dynamics in conventional metals and semiconductors [9,10]. As an example, it was able to provide the earliest reliable experimental evaluation of the electron–phonon renormalization parameter [9]. This extraordinary achievement stimulated rapid interest in using the femtosecond laser as a probe of high temperature superconductivity [11–17] and more generally correlated physics in complex oxides materials [6,18,19]. Towards this end, in the early 1990s, some pioneering works began on high-Tc cuprate superconductors [11–17]. In the past three decades, pump-probe spectroscopy has been successfully applied to study the QP dynamics in many cuprate superconductors [20–22].

From the experimental point of view, the technique relies on a fractional change in reflectivity or transmission monitored as a function of pump-probe delay time. The photo-induced reflectivity change ($\Delta R/R(t)$) is assumed to be proportional to the density of photo-excited quasiparticles (QPs) ($\Delta N(t)$) [12,23], i.e., $\Delta R/R(t) \propto \Delta N(t)$, since the change in dielectric constant ($\Delta\varepsilon$) is linearly dependent on the QP density as $\Delta\varepsilon = (1+i/\omega\tau_s)\omega_p^2\Delta N_0/N_t\omega^3$, where $\omega$ is the frequency of probe photon, $\omega_p$ is the plasma frequency, $\tau_s$ is the scattering time, and $N_t$ is the total electron density [12,24]. When temperature surpasses the superconducting transition temperature ($T_C$), the magnitude of $\Delta R/R(t)$ exhibits an abrupt drop [25]. The recovery traces can be analyzed as multiple exponential decay components with each decay channel assigned to a specific physical process [8,26–28].

Recently, thanks to the rapid advances in laser technology, significant progresses have been achieved in ultrafast spectroscopic studies of superconductors [8,27,29–34]. Here, we want to specially mention the development of Ti:Sapphire lasers which has enabled the pump-probe method at 800/400 nm and resulted in fruitful results on superconductors [35–39]. At the same time, the optical response at 800 nm can only give indirect information

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due to the probe photon energy being much larger than the energy scale specific to order parameters and elementary excitations in superconductors [5,24]. To address this issue, broadband pump-probe spectroscopy based on the super-continuum generation and optical parametric amplification has been employed [8,27,40,41]. This approach is able to provide dynamical information in both time and frequency domains, connecting the reflectivity change at high-energy electronic excitation with the resonant spectral function of Boson modes [8]. Furthermore, it is now feasible to detune ultrashort pulses to the extreme wavelength regime. For example, with different nonlinear optical methods on a lab bench, the spectral coverage from deep ultraviolet to terahertz (THz) bands is now available, leading to the rapid developments of time-resolved infrared (IR) – THz spectroscopy [6,42], time-resolved ARPES [30,31], and time-resolved Raman spectroscopy [43]. These new probes have dramatically improved the capabilities of the ultrafast optical techniques, shifting interests to resonant excitations and probes. To illustrate, the technique of high harmonic generation provides deep UV pulses that have been integrated into ARPES; such time-resolved ARPES enables mapping of the transient density of states dynamically in cuprates, which may answer some enduring issues in such interesting systems [30,31].

Unlike the rapid development in ultrafast experimentation on superconductors, to date the progress in microscopic theory has been somewhat limited. Early on, the non-equilibrium phenomena in conventional superconductors were described by Rothwarf and Taylor in 1967 [44]. Such a phenomenological model, expressed in the set of the Rothwarf–Taylor equations (RTE), has been successfully applied for BCS superconductors and is frequently employed to interpret QP dynamics in cuprates. Later on, Kabanov and co-workers have suggested to treat the ground state with two types of gaps: the temperature-dependent gap (superconducting gap) and the temperature-independent gap (pseudogap) [25]. Their model has successfully explained the temperature dependence of signal amplitude and decay lifetime. Nevertheless, the dynamic behavior in cuprates was found to be more consistent with the model of an isotropic gap [25], which is in contrast to the general argument of a d-wave symmetry gap in cuprates. Nicol and Carbotte attempted to model the QP dynamics in cuprates with two nonequilibrium models (the $\mu^*$ and $T^*$ models) [45], but they found that neither s-wave nor d-wave gap models can produce a quantitative explanation to the observed behaviors. Recently, there have been new proposals to describe the nonequilibrium phenomena in high temperature superconductors [46–48]. For experimental scientists, phenomenological descriptions, such as two/three/four-temperature models, in the view of thermodynamics have been widely used to fit the experimental data [8,10,49].

The QP dynamic behaviors revealed by pump-probe experiments in cuprates are tightly associated with the spectral weight transfer due to the photo-induced modification of ground state. The recovery dynamics have been studied to investigate glue for Cooper pairing including phononic and electronic contributions. Initially, ultrafast spectroscopy has been performed on $YBa_2Cu_3O_{7-\delta}$ (YBCO) [11–15,21,50,51]; ever since, the study has been expanded to include a variety of compounds in the cuprate family [8,17,20,27,29,39,40,49,52–55]. The normal state has also been at the center of focus to investigate the QP dynamics related to the pseudogap phase and other competing orders [34,37,38]. The ultrafast optical spectroscopy is expected to provide important information on this long-standing issue by searching for distinct spectrotemporal features of superconductivity and the pseudogap. These features have been captured in reflection change, spectral dispersion, polarization dependence and coherent phonon anomalies [20,25,40,56,57].

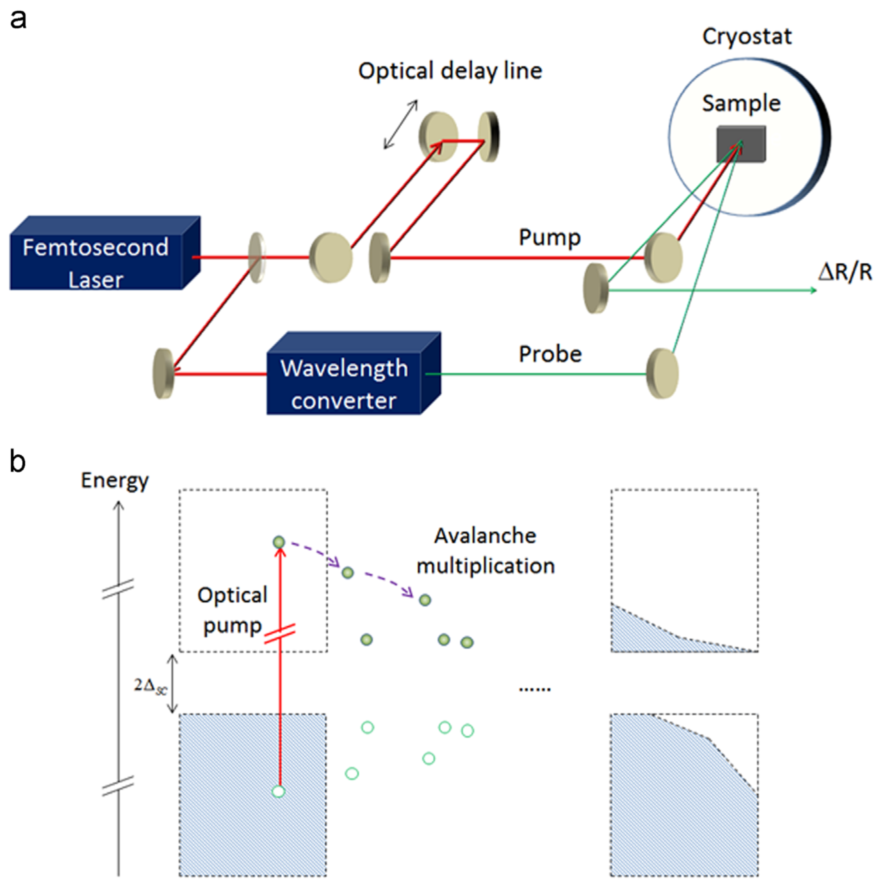

**Fig. 1.** (a) A schematic diagram of the pump-probe spectroscopy. (b) A schematic illustration of the ultrafast photo-excitation processes in cuprates. One absorbed photon generates a QP with high energy which loses its energy by creating multiple QPs through the process of avalanche multiplication.