Low-density optical pump – THz probe analysis of high-temperature superconductors

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Abstract. Using time-resolved THz probing we analyze the dynamics of low-density optically excited, optimally doped $YBa_2Cu_3O_{7-\delta}$. A previously unknown additional relaxation component is found, tentatively indicating the separate relaxation of charge and spin excitations.

1. Introduction

Dynamic analysis of the relaxation dynamics of high-temperature superconductors (HTSC) after optical excitation offer a detailed insight into the superconductivity mechanism of these materials. Spectrally similar effects which would be superposed in stationary experiments can readily be distinguished in the time domain. In the past, most time-resolved experiments on HTSC have probed high energy states at photon energies around 1.0- 2.0 eV [1]. However, under these conditions, superconductivity related pairing dynamics cannot be distinguished from energy relaxation dynamics, as the probing energy is too far above the superconductivity gap. Recent experiments, probing pairing dynamics at lower energies e.g. MIR [2] or THz [3], have yielded interesting observations. However, these experiments were performed under very high optical excitation densities, which significantly modify the superconducting state [3]. Here we present a time-resolved optical pump-THz probe analysis with a drastically improved resolution better than $\Delta T/T_0=10^{-9}$, which allows to perform analysis at excitation densities 2 - 3 orders of magnitudes below previous approaches. Thus density dependent recombination dynamics in optimally doped $YBa_2Cu_3O_{7-\delta}$ probed at low energies are isolated for the first time. In contrast to a previously seen single relaxation component, two components are observed, tentatively indicating the separate relaxation of spin and charge excitations.

2. Experimental Methods

The experiments are performed with a time-domain THz transmission setup using a 76 MHz repetition rate 100 fs laser. Its great advantage is the possibility to directly measure the complex conductivity $\sigma = \sigma_1 - i\sigma_2$ of HTSC [4]. Based on a Two-Fluid-Model [5], one can simultaneously and separately analyze the behavior of quasiparticle x_n and superconducting condensate x_s densities. The samples used consist of optimally doped 20 nm $YBa_2Cu_3O_{7-\delta}$ films on $NdGaO_3$ substrates mounted in a cryostat. A stationary, temperature dependent evaluation of the complex conductivity delivers a critical temperature of T_C = 88 K confirming a good sample quality despite its thin thickness. For dynamical analysis a further optical laser pulse is focused, collinear to the THz beam, onto the sample with a 800 µm focus (= THz focus). Primary excitation densities in the range of $3{\cdot}10^{16}$ to $4.5{\cdot}10^{17}cm^{-3}$ are induced. The optical excitation breaks Cooper-pairs resulting in a decrease of x_s and increase of x_n, accordingly an increase of σ_1 and decrease of σ_2. The following relaxation process is dynamically analyzed by probing the THz transmission. A detailed numerical calculation, considering the optically altered conductivity due to the changed fractions of x_n and x_s, leads to expected THz transmission intensity modifications of maximally $\Delta T/T_0 \approx 10^{-8}$ which have to be resolved, compared to a free-space THz transmission T_0. To avoid long data acquisition measurements we do not analyze the full dependence of the THz transmission as function of optical and THz time delays but concentrate only on characteristic fixed delay points of the THz transient, and evaluate the dependence of the optical time delay. As indicated on the THz transmission transient in the inset of Fig. 1, we concentrate on the conductivity dynamics at the marked peak and zero crossing positions. Our numeric calculation reveals, that the signal increase measured at the peak of the THz-transient is dominated by the decrease of σ_2, whereas the signal at the marked zero crossing increases, depends equally on the σ_1 increase and the σ_2 decrease.

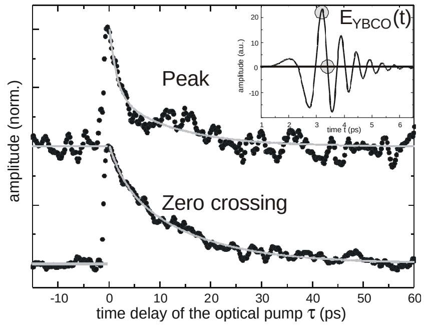

Fig 1. The inset shows a THz transient and the characteristic positions used for dynamical analysis. The main figure shows the respective relaxation signals after optical excitation.

3. Results and Discussion

Two exemplary relaxation transients taken at 10K are shown in Fig. 1. One can directly see different relaxation times of both transients indicating that at least two different components are necessary to explain the relaxation process. As the zero crossing signal has a better signal to noise ratio and contains both dynamics, we restrict in the following to such analysis and evaluate these transients via two component exponential fits with decay times τ_1 and τ_2. Fig. 2 shows the analysis of the density and temperature dependencies of both components. The left plot indicates a fast component τ_2 in the region of 1-3 ps that is in agreement with previous high-density observations [2,3]. This fast component exhibits a characteristic Rothwarf-Taylor density dependence. For the first time we observe an additional slow relaxation component τ_1 which decreases for increasing excitation density. We think that this additional slow component was not previously observed, as pre-

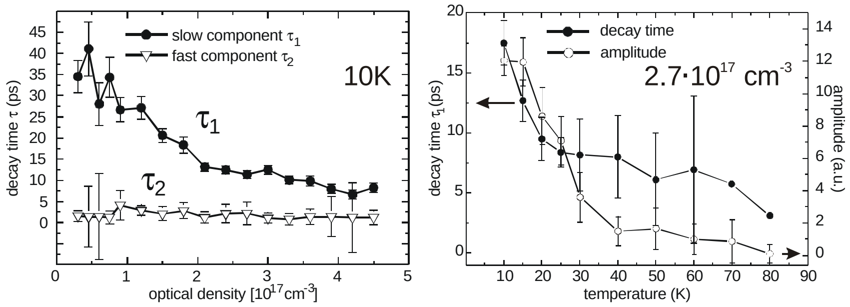

Fig2. Decay time density dependence of both components τ_2 and τ_2 at 10 K (left) and temperature dependence of amplitude and decay time of the slow component τ_1 (right).

vious experiments were performed at much higher excitation densities where both relaxation channels τ_1 and τ_2 become restricted by phonon dissipation. The temperature dependence of the amplitude and relaxation time of τ_1 are shown in the right part of Fig. 2. The decay time τ_1 decreases for higher temperatures. While the amplitude of the fast component τ_2 has a typical BCS type behavior with a sharp drop close to T_C (data not shown), the τ_1 component decays significantly already much below T_C.

4. Conclusions

The presence of two markedly different relaxation components proofs that the simple assumption of a single type of excitations is clearly not sufficient to explain the observed dynamics. We tentatively ascribe both components to the separate relaxation of charge and spin excitations (antiferromagnetic spin fluctuations), supporting the notion of independent charge and spin excitations within a collective state, the "quantum protectorate" [6]. The decrease of the respective relaxation times with increasing excitation density would then reflect the increasing probability for the coincidence of charge or spinon pairs, respectively. Further experimental and theoretical analysis will however be necessary to further support this conjecture and to assign the respective components.

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