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Analysis of the photoresponse of Y-Ba-Cu-O thin films on ps to μs timescales

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Abstract. Femtosecond time-resolved optical spectroscopy and transient photoconductivity measurements have been used to investigate the time-dynamics of electronic excitations in $YBa_2Cu_3O_{7-\delta}$. Whereas the picosecond response is due to energy relaxation of carriers to phonons, the slow response, which is observed in both experiments, cannot be explained by a bolometric mechanism. The proposed alternative explanation involves localised states (polarons) near E_F giving rise to both an optical absorption signal and a kinetic inductance on the microsecond timescale. The bolometric response, which is also present, is shown to agree well with theoretical model calculations.

1. Introduction

The photoresponse of high-T_C cuprate superconductors have been studied over the past few years by variety of techniques. One of them involves illumination of a current biased microbridge structures at temperatures below T_C with short laser pulses. The measured electrical transients were in most cases found to have two components: a fast one on the picosecond timescale, and slow one, with longer decay time, typically ns to μs [1]. Slow component has been usually interpreted as a bolometric effect where optical illumination results in simple heating of the film into a resistive region. In this case electron and phonon subsystems are described by the same temperature shift, which is in turn determined by thermal properties of the film and substrate [2]. The nature of fast component (1.5 ps decay time at 100 fs laser pulse has been reported using electro-optic sampling) is on other hand still under debate and several mechanisms have been proposed.

Although the slow component is generally attributed to the bolometric effect, recent measurements[3,4] indicate, that situation is more complex, since localised electronic states in the vicinity of E_F should be taken into consideration. Picosecond resonant Raman spectroscopy[3] for example gives unambiguous signature of localised electronic states in the normal state of optimally doped superconducting YBCO and LaSCO samples. Time-resolved optical absorption spectroscopy, on the other hand, gives evidence of localised states near E_F, with life-time as high as several tens of microseconds in YBCO and BiSCO[5]. To pin down the origin of this excitations we have performed time-resolved differential transmission measurements on different $YBa_2Cu_3O_{7-\delta}$ samples and complemented the data by measurements of the photoinduced transient signals due to the illumination of the current biased microbridge by laser pulses, with special care devoted to the slow component.

2. Sample preparation and experimental details

The samples used in this investigation were prepared by high-pressure DC sputtering method. Approximately 100 nm thick films were deposited on SrTiO and MgO substrates. Typical transition temperatures were around 90 K for pristine films. Low resistivity contacts to the film have been made by sputtering approximately 500 nm thick gold pads on the YBCO surface. Contacts were subsequently annealed in oxygen atmosphere at 400 ^{o}C for half an hour. Following the annealing microbridge structures were produced using non-invasive laser patterning technique[6]. Microbridges produced in this way showed practically no shift in critical temperature, narrow transitions (ΔT ~ 1 K, 0%-90%) and a critical current density of about 2×10^{6} A/cm^{2} at 77 K.

The time-resolved differential transmission measurements were performed using mode-locked Ti:sapphire laser producing 150 fs pulses at 88 MHz repetition rate. Pump and probe beams (λ=800 nm) were focused to a 100 μm diameter spot, with average pump power of 40 mW. High-frequency modulation (200 kHz) with phase sensitive detection was used to provide detection sensitivity of ~10^{-6}.

The measurements of photoinduced voltage transients were performed using an acusto-optically modulated CW Ar-ion laser. In this way 200 ns (FWHM) pulses at 1kHz repetition were produced. A 50 Ω semi-rigid coaxial cable and wire bonding technique was used to connect microbridge structures with read-out electronics. The bridge was biased with DC current through bias-tee. The response was recorded with wide-band amplifier (Miteq AM-1305, 55 dB, 10 kHz -1 GHZ) and subsequently analised using Tek TDS 544 A scope.

3. Results and discussion

The time-resolved femtosecond measurements on YBCO and the dependence of the amplitude on temperature has been recently described[4], with two clearly identifiable relaxation components being observed, one exhibiting band-like picosecond timescale properties and other operative on microsecond timescale. In this paper we shall focus our attention on the slow component in time-resolved and transient photoconductivity data.

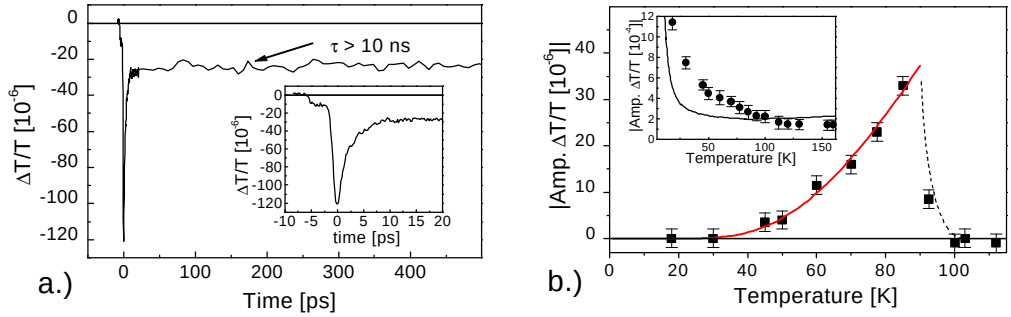

Figure 1.: a.) The differential transmission of a near optimally doped YBCO sample (δ ~ 0.1) as a function of time after photoexcitation. The insert shows the behavior at short times. b.) The temperature dependence of of the slow component amplitude. The insert shows the bolometric signal measured out-of-phase together with the expected bolometric signal calculated using thermal and optical constants of the film and substrate[2].

Figure 1a.) presents the temporal evolution of measured differential transmission (*DT/T*) at temperature close to T_C showing two distinct carrier relaxation processes. The fast relaxing component is typical of band-carrier relaxation determined by electron-phonon coupling, whereas the slow one displays non-exponential relaxation with a time constant far greater than the time between the two succeeding laser pulses (11.4 ns). The amplitude of the slow component (Figure 1 b.)) at temperatures below T_C follows thermally activated behavior

across the superconducting gap of the form $\Delta T/T \approx Exp\left(-2\Delta(\mathrm{T})/k\mathrm{T}\right)$, where $2\Delta_0 = 3.5 \pm 0.5 k\mathrm{T}$ and drops to zero above $\mathrm{T_C}$. The strong decrease in signal at $\mathrm{T_C}$ can be understood as arising from the rapid decrease in the density of unpaired hole states close to the gap as the system reverts to the normal Fermi liquid behavior. This cannot be described by thermal effects, where temperature dependence of thermal and optical constants for film on substrate are taken into consideration. In contrast, the bolometric signal increases with decreasing temperature and can be observed in time-resolved data as the amplitude of the out-of-phase signal (with no temporal dependence on the nanosecond timescale) on the lock-in amplifier as shown in insert to Figure 1 b.).

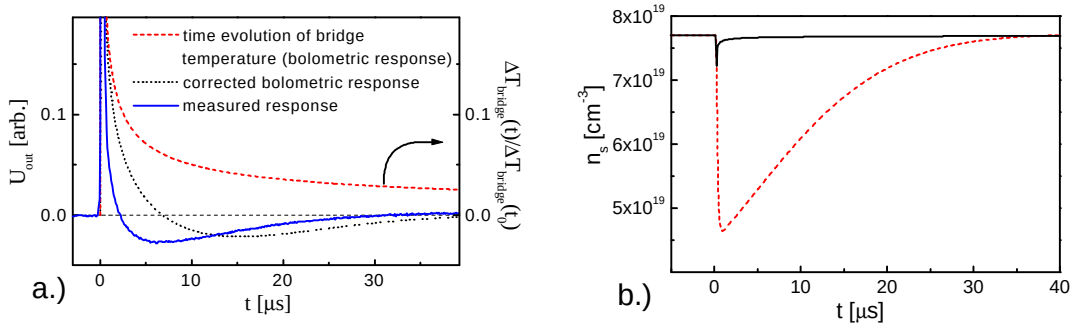

Figure 2: a.) Photoresponse following a laser pulse incident on a microstrip (solid line) compared with time evolution of the microbridge temperature (dashed line) and corresponding calculated bolometric response at the output of our sampling electronics. b.) Temporal evolution of the superconducting pair density (dashed) obtained from analysis of the data, and the change in density due to bolometric contribution (solid).

The time-resolved data are complemented by measurements of the photoconducting signal recorded as the transient voltage drop across a current biased YBCO microbridge followed the excitation by a laser pulse. In this case the laser pulse drives the superconductor into the resistive state which results in the appearance of a voltage transient across the microbridge. The response observable from approximately 4 K to 1 K below $\mathrm{T_C}^*$ ($\mathrm{T_C}$ depending on I_{bias}) is shown in figure 2 a.) with solid line. Careful evaluation of the experimental data, based on measured $R(\mathrm{T},I_{bias})$ characteristics, dependence of the signal on pulse energy, temperature and bias current, has shown the signal to be predominantly bolometric in origin. However, there is an additional feature in the observed photoresponse, manifested as a drop to a negative values and unusually long relaxation, that cannot be described by bolometric mechanism. In order to evaluate the bolometric contribution to the signal we have used the simple heat transfer model[7], wherein the optical pulse supplies heat (at a rate corresponding to the temporal profile of the laser pulse) to the very thin superconducting bridge lying on a semi-infinite substrate. The modeled time evolution, using thermal and optical properties of film and substrate[2], of bolometric response is shown in figure 2 a.) as a dashed line. In order to compare the modeled response with measurement, one has to account for the bridge response to the output of the amplifier using carefully measured transfer function of all the links in the read-out chain, including connections, bias-tee and amplifier. As expected, the calculated signal (dotted line on figure 2 a.)) undergoes the transition to the negative voltages, which is the result of the slight differentiation of the input signal at a lower frequency margin of the read-out chain. However, the difference between the measured signal and signal calculated on purely bolometric model is still substantial.

The point at which the calculated signal crosses the zero line is determined by the (measured) read-out chain transfer function and can not be shifted to shorter times to make better agreement with the data, even if the shape of the input signal is drastically changed. This was experimentally verified by substituting the microbridge with passive electronics

and measuring the amplifiers response to various input signals. The reason for discrepancy between the microbridge response and bolometric response is therefore to be found elsewhere.

We should mention that although the dynamics of the resistive vortex flow on these time scales is still not understood in detail, we estimate that this contribution to be too small and of the wrong sign to explain the signal observed. The same arguments apply to bolometric kinetic inductance mechanism, which was proposed to explain the negative transients measured on the ps timescale[8]. This model is based on two-fluid model where the number of super-carriers is being determined by the lattice temperature. As the temperature is increased (due to heating by the laser pulse) the number of super-carriers is decreased (solid line in figure 2 b.)) and the remaining pairs should be accelerated in order to sustain constant supercurrent. This results in voltage transient ($U(t) \propto -(1/n_S^2)\, dn_S/dt$) due to changing kinetic inductance. This mechanism could explain the negative signal, but the estimated amplitude is two orders of magnitude lower than observed.

Finally, taking into account the existence of localized states near E_F, the physical picture can be significantly altered. In this way the system remains out of equilibrium due to slow quasiparticle relaxation, with longer time constants than for band relaxation, and the electron and phonon sub-systems cannot be described by the same temperature. Taking this into account we have used the non-bolometric kinetic inductance model to describe the discrepancy between measured signal and modelled bolometric response. By using the initial super-carrier density as calculated within two-fluid model and the decrease in super-carrier number estimated from experimental data we have calculated the temporal dependence of the super-carrier density (dashed line in figure 2 b.)). From the calculation we estimated the quasiparticle recombination time constant τ to be $13 \pm 3 ms$, which is in agreement with optical data[5].

4. Conclusions

The general conclusion from these experiments is that there exists two different types of quasiparticle relaxation processes in these materials. The existence of two components in the relaxation do not necessarily mean that we are probing two independent carrier bands, but rather, the data give an indication that the super-carriers have a dual character. Moreover the two different relaxation experiments (optical and transport) compare rather well regarding quasiparticle lifetimes, in spite of the fact that the details of the relaxation processes in the two cases are necessarily different.

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