# FEMTOSECOND DYNAMICS OF QUASI-PARTICLES IN $YBa_2Cu_3O_{7-\delta}$ SUPERCONDUCTOR FILMS

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## Abstract

We have studied the transient electronic response of $YBa_2Cu_3O_{7-\delta}$ epitaxialy grown $HT_c$ superconductor thin films in the femtosecond time domain, using transient photoinduced reflectivity ($\Delta R$) with 60 fsec time resolution. For temperatures $T>T_c$ only a bolometric signal was observed with $\Delta R>0$. For $T<T_c$, $\Delta R<0$ with a temperature dependent rise time of order 300 fsec followed by a relaxation (of order 3 psec) into a state with $\Delta R>0$. The results for $T<T_c$ are explained in terms of quasi-particle (QP) electronic response giving $\Delta R<0$. Thus the femtosecond rise time is interpreted as avalanche multiplication of QP across the gap $2\Delta$ and the subsequent picosecond relaxation as QP recombination. The QP optical response is explained within the two fluid model.

## Introduction

Since the discovery of $HT_c$ superconductors, many efforts have been made to explain the transient optical reponse of $YBa_2Cu_3O_{7-\delta}$ (YBCO) films[1-6], for the purpose of understanding the nature of superconductivity and for applications in high speed optical detectors. However none of the previous experiments gave a clear picture of the photoexcited QP dynamics due to the limited time resolution of the measuring systems.

In the present study, using femtosecond time resolved spectoscopy , we have clearly observed two main QP processes; (1) avalanche multiplication of QP following photon absorption. This process takes about 300 fsec with QP production gain of order 100, thus making it possible,with our 60 fsec resolution to study electron-Cooper pair(CP) inelastic scattering processes. (2)A nonlinear recombination of the photogenerated QP .

## Experimental technique

Photoinduced changes in reflectivity were measured between 10 fsec to 3 nsec using the polarized pump and probe technique[7](Fig. 1), where both beams were derived from a CPM ring dye-laser[8] which produces 60 fsec pulse duration at a center wavelength of 625 nm, with an energy per pulse of 100 pJ at 80 MHz repetition rate.

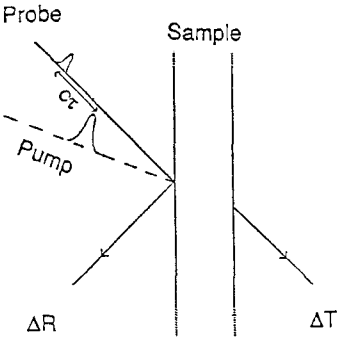


Fig. 1 The pump and probe experimental set up. The delay time $\tau$ between the pulses is introduced by a translation stage.

The pump beam used to excite the sample had about 4 times higher intensity than that of the probe beam. The probe pulse was delayed with respect to the pump pulse by a translational stage that had 6 fsec resolution. The pump beam was modulated at 4 MHz using an acousto-optic modulator and the photoinduced change in refectivity($\Delta R(t)$) of the probe beam was measured using a moderately fast Si photodiode and a fast lock-in amplifier. Both pump and probe beams were focused on the same spot on the sample with approximately 30 μm diameter; this

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corresponds to initial photoexcited carrier density $\Delta N$ of $10^{18}$ $cm^{-3}$. The overall sensitivity of our system for $\Delta R/R$ was $10^{-6}$. The YBCO films were deposited on (100) $SrTiO_3$ substrates by the UV laser ablation technique[9]. They were oriented perpendicular to the c axis with thickness between 100 to 500 nm. The pump and probe polarizations were parallel to the film surface, thus only $\Delta R_{a,b}$ was studied.

## Results and discussion

Fig. 2 shows a typical transient $\Delta R$ response(bolometric) of a $YBCO_7$ sc film ($T_c$~93K) at 300K. At 300K as well as for all T>Tc, we observed a step function reponse $\Delta R>0$ which decays through heat diffusion with a 3 nsec typical time constant; it is slower at lower temperature. The $\Delta R>0$ response can readily be shown to be bolometric since we measure $\partial R/\partial T>0$ for all T.

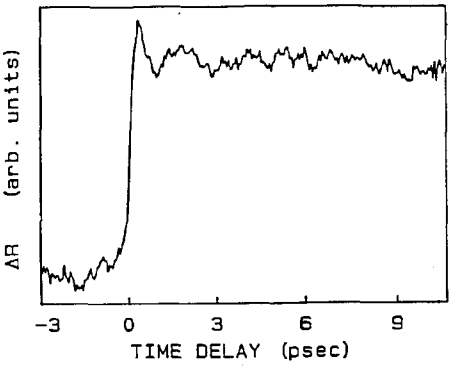


Fig. 2 $\Delta R/R$ transient response of $YBa_2Cu_3O_7$ at 300K.

For $T<T_c$, Fig. 3, $\Delta R(t)$ shows a completely different behavior. $\Delta R$ is negative immediately after t=0 and it reaches a maximum negative value, $-(\Delta R/R)_{max}$, with a characteristic time of 300 fsec. To determine the correct zero time delay of the probe pulse with respect to the pump pulse, we used an a-Si;H sample where t=0 is well known[10], without changing the geometry of the system(Fig. 4); we see a clear delay in the $YBCO_7$ $\Delta R$ response. After reaching $-(\Delta R/R)_{max}$, $\Delta R$ comes back and it crosses the zero level into a plateau of $\Delta R>0$.

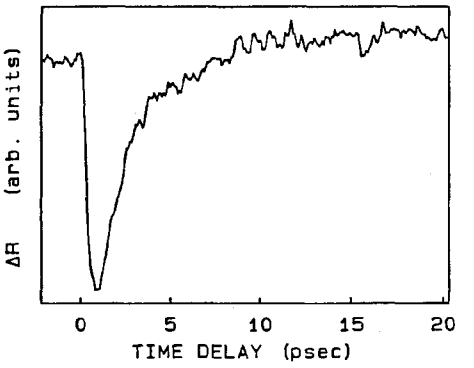


Fig. 3 $\Delta R/R$ transient response of $YBa_2Cu_3O_7$ at 20K (<Tc~93K).

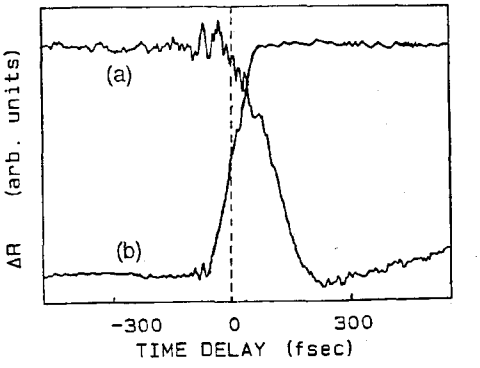


Fig. 4 The transient $\Delta R$ response of (a) $YBCO_7$ at 20K compared to that of (b) a-Si:H. The delay t=0 can be readily determined.

We measure R(T) at 635 nm; it is an increasing function of T, thus the initial rapid $\Delta R<0$ cannot be bolometric, but it is due to QP response.

Fig. 5 shows the peak values of $-(\Delta R/R)_{max}$ vs. temperature. The data can be best fit within the two fluid model[11]: $[1-(T/T_c)^4]$, which gives the relative density of the QP and superconducting electrons; an attempt to fit the data using the BCS model does not give as good a fit(Fig. 5).

We interpret the fast build up in $\Delta R<0$ as an avalanche multiplication of QP. Hot electrons which are created immediately following photon absorption thermalize by about 100 scattering events ( $g = 2h\omega/2\Delta(0)$~100) into optical phonons which in turn break Cooper pairs and this explains the finite rise time.

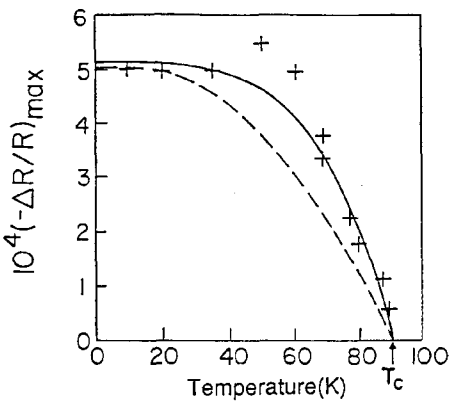


Fig. 5 The maximum QP response $(-\Delta R/R)_{max}$ of the $YBCO_7$ film vs. temperature. The lines are fits to the data: full line-two fluid model; dashed line-BCS.

The QP response can be observed only if the system is not saturated. The saturation QP level[6] is related to the density of electrons in the ceramic materials which is $N_s=1.6\times10^{21}$ $cm^{-3}$. We note that the peak density of the photoinduced QP, $\Delta N_{max}=g\Delta N(0)=10^{20}$ $cm^{-3}$, is less than $N_s$ in our experiment.

The origin of the QP response can be explained within the two fluid model (clean limit), where the optical conductivity $\sigma$ is written as;

$$\sigma = N_n\sigma_n + N_s\sigma_s \qquad (1)$$

where $N_n$ and $N_s$ are the relative densities of the normal and the superconducting electrons and $\sigma_n$(Drude) and $\sigma_s$ are their respective optical conductivities. Indeed, infrared studies of $YBCO_7$ show that the Drude($\sigma_n$) oscillator strength gradually disappears at low temperatures[12-14] consistent with the decrease in $N_n$. Generation of QP increases $N_n$ at the expense of $N_s$[15]. The changes $\Delta\varepsilon$ in the dielectric constant $\varepsilon=\varepsilon_1+\varepsilon_2$ can be thus readily calculated;

$$\Delta\varepsilon_1 = \delta N\,\omega_p^2 / \omega^2(1+\omega^2\tau^2) \qquad (2)$$

$$\Delta\varepsilon_2 = \delta N\,\omega_p^2/\omega^3\tau \qquad (3)$$

where, $\omega_p$ is the plasma frequency ($\sim$1eV)[13,14] and $\tau$ is the scattering time.[13,14] For $\omega\tau=150$ and $\delta N=10^{20}cm^{-3}$, we calculate $\Delta\varepsilon_2$ to be of order $10^{-4}$, which is in agreement with the value of our signal $\Delta R/R$ (Fig. 3).

Fig. 6 shows typical $\Delta R(t)$ response of a non superconducting ceramic film. There is no delay in $\Delta R(t)$. The initial fast decay , roughly 3 ps, is due to hot electrons[16] , and

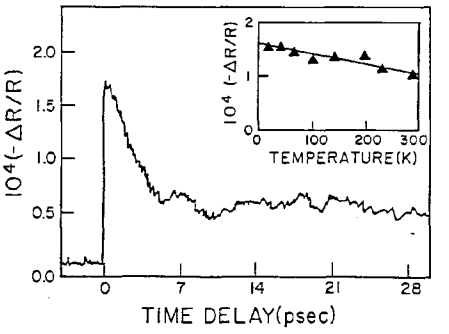


Fig. 6 $\Delta R/R$ transient response for a $YBa_2Cu_3O_6$ (non-superconductor) film at 20K. The inset shows the maximum value of $\Delta R/R$ vs. temperature.

the plateau is bolometric in origin. Also we plotted $(\Delta R/R)_{max}$ as a function of temperature; the dramatic changes observed in $YBCO_7$ (Fig. 5) are not observed in $YBCO_6$ .

We interpret the picosecond relaxation of $\Delta R<0$ back into a plateau of $\Delta R>0$ in $YBCO_7$ (Fig. 3) as due to QP recombination which creates excess phonons;these eventually increase the sample's temperature and this temperature is manifested as a $\Delta R>0$ plateau, as is the case for $T>T_c$ (Fig. 2). We found, however, that the recombination rate $\upsilon$ is a strong function of temperature as shown in Fig. 7. $\upsilon$ is almost constant at low temperature but decrases dramatically at $T> T_c/2$. This behaviour is surprisingly similar to the temperature dependence of the linewidth $\Gamma$ of the $A_g$ and $B_{1g}$ optical phonons in $YBCO_7$, as recently measured by Raman scattering[17]. $\Gamma$ also increases dramatically at low temperature ($T<T_c/2$) and this was explained as due to a strong resonance interaction with the gap $2\Delta$. Since the energy released by QP recombination is given up to phonons, the resonance with the optical modes may influence the recombination rate in the same way as for $\Gamma$. We claim therefore that the decrease in $\upsilon$ with T is the manifestation of the decrease in $\Delta$ which in turn relaxes the resonance interaction with the optical phonons.

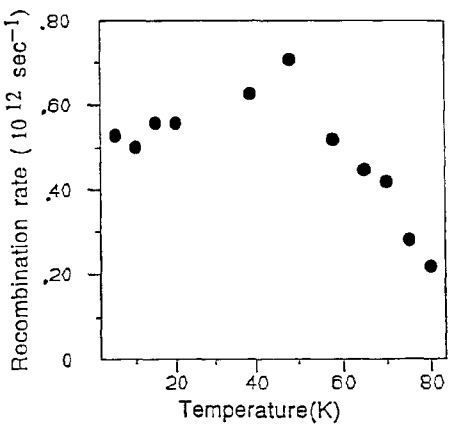


Fig. 7 QP recombination rate $\upsilon$ as a function of temperature, as determined from the QP transient response ($\Delta R(t)<0$).

## Conclusions.

In conclusion , we have separated the bolometric and nonbolometric transient optical responses of YBCO thin films. We have resolved the QP dynamics for the first time; it is composed of two parts. Fast thermalization of QP by inelastic scattering which eventually produces additional QP, and a relatively slow recombination into Cooper pairs, with a strong temperature dependence.

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*Supported by BP America