Femtosecond Pump-Probe Polarization Dependent Investigation of Relaxation Dynamics in YBa$_2$Cu$_4$O$_8$

D. Dvorsek,1 V. V. Kabanov,1 J. Demsar,1,2 J. Karpinski,3 S. M. Kazakov,3 and D. Mihailovic1

Received 10 August 2003

Femtosecond pump-probe experiments are reported on quasiparticle relaxation and recombination in YBa$_2$Cu$_4$O$_8$, as a function of temperature and polarization. We compare our results with the data obtained on YBa$_2$Cu$_3$O$_{7-x}$ and show that similar two-component relaxation is present in both cases. A strong polarization anisotropy of the picosecond response is observed below $T_c$ and interpreted with the aid of a simple model which considers the anisotropy of the probe transition matrix elements.

KEY WORDS: high-temperature superconductivity; femtosecond spectroscopy; carrier relaxation dynamics.

Femtosecond time–domain spectroscopy gave in recent years a lot of information on the quasiparticle excitations and a low-energy structure of correlated electron systems in different high-$T_c$ superconducting cuprates (HTSC). Measurements in differently doped YBa$_2$Cu$_3$O$_{7-x}$ (YBCO123) compounds have shown the simultaneous existence of two gaps in optimally doped and overdoped regions [1], though no anisotropy in measured response was observed due to the high degree of twinning present in the samples. The effects of anisotropy were investigated on untwinned YBCO123 single crystal [2], where a response parallel and perpendicular to the Cu--O chains was separately probed and found to be different. The authors explained the difference in response by proposing different mechanisms for the observed signals along the $a$ and $b$ axis. The response along $a$ axis was attributed to the quasiparticle excitations across a temperature-dependent gap, whereas the response along $b$ axis was associated with the pseudogap. Another highly anisotropic compound from the YBaCuO family is YBa$_2$Cu$_4$O$_8$ (YBCO124), which contains double Cu--O chains instead of single chains as in YBCO123. A fixed oxygen content and a well-defined untwinned orthorhombic structure are properties, which make YBCO124 a very suitable system for more detailed investigation of polarization dependence in femtosecond spectroscopy.

In this paper we wish to show that the two-component relaxation behavior observed in YBCO123 [1] is similar also in YBCO124, where simultaneous existence of two gaps is present in all polarizations. Particular attention is given to the analysis of the polarization dependence. The response attributed to the existence of temperature-dependent gap shows a polarization dependence, whereas the pseudogap response is found to be isotropic. We propose a simple model from which the anisotropy of the photoinduced signal with respect to the probe pulse polarization arises.

A detailed description of the experimental technique and the theory of excitation and relaxation of the photoexcited carriers in superconductors with different gap structures can be found in Refs. [3,4]. The YBCO124 sample used for this investigation was prepared in Zürich by a nonstoichiometric flux-growth technique, using a BaO–CuO eutectic mixture as the flux. The crystal had a thickness of approximately 100 $\mu$m in the $c$-axis direction and dimensions of 0.4 $\times$ 0.2 mm$^2$ in the $a$-$b$ plane. The crystal axis were determined by X-ray analysis. Details of the growth

1Jozef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia.
2Los Alamos National Laboratory, Los Alamos, New Mexico 87545.
3Institut für Festkörperphysik, ETH Zurich, Switzerland.
Fig. 1. The photoinduced reflection $\Delta R/R$ from Y$_1$Ba$_2$Cu$_4$O$_8$ at different temperatures above and below $T_c$ as a function of time, measured (a) with the polarization of probe pulse in the direction parallel to the crystal axis $a$ and (b) parallel to (the direction of chains) the crystal axis $b$. The inset shows the data at 28 K and 74 K already presented in (a) on the logarithmic scale, so that two-component decay can be easily observed. In (c) we show photoinduced reflection $\Delta R/R$ from Y$_1$Ba$_2$Cu$_3$O$_7$ at different temperatures and a presence of the two components is again shown with the logarithmic plot.

method and the techniques used for characterization of the sample are given in Ref. [5].

The photoinduced (PI) reflection $\Delta R/R$ as a function of time at different temperatures is shown in Fig. 1 for the direction of polarization of probe pulse along the $a$ axis and $b$ axis of YBCO124. The chains are parallel to the $b$ axis and the PI response in this directions clearly shows a presence of at least two relaxation processes with different sign of $\Delta R/R$. At temperatures $T > T_c$ we see a signal with a positive $\Delta R/R$ and with a relatively fast relaxation time ($\tau_p \approx 0.2$ ps). As the temperature is lowered below $T_c$ a second component with a longer relaxation time ($\tau_d \approx 2$ ps) and negative sign starts to appear. This kind of behavior of the two components is also present in PI response with the probe polarization along the $a$ axis, which can be clearly seen, if the data is presented on a logarithmic scale as in the inset of Fig 1. The relaxation of $\Delta R/R$ after 200 fs can be modeled with a function of the form $\Delta R(t, T)/R = G(T)\exp(-t/\tau_G) + P(T)\exp(-t/\tau_P)$, where the temperature-dependent amplitudes $G(T)$ ($G(T) = 0$ for $T \geq T_c$) and $P(T)$ can have the same, or opposite sign below $T_c$, depending on the polarization direction of the probe pulse. Fits, which are presented by a continuous line in Fig. 1, are in a good agreement with the data. We note that above $T_c$ the data shows a slight departure from a simple exponential decay. Similar behavior above $T_c$ has also been reported in Ref. [6], where two components of opposite sign below $T_c$ and one component slightly nonexponential above $T_c$ were observed on Tl$_2$Ba$_2$Cu$_2$O$_{y+6}$. Whether a stretch exponential decay is relevant or the presence of an additional component is the reason for the discrepancy, we cannot assert with a good degree of certainty from the present data.

In Fig. 1c, we compare PI response obtained on YBCO123 at different temperatures, which shows a qualitatively same behavior as YBCO124 along the $a$ axis. The existence of two relaxation processes is again seen on a logarithmic plot.

We analyze the temperature dependences of relaxation time $\tau$ and of both amplitudes $G(T)$, $P(T)$ in the same way as was done previously for YBCO123 [1]. The relaxation time $\tau_p$ was approximately constant. In Fig. 2, we show the divergent behavior near $T_c$ in the temperature dependence of $\tau_G$, predicted by the model [4] (solid curve). The data points were obtained from the fits of the time evolution of $\Delta R/R$ for YBCO123, and for YBCO124 along the $a$ and $b$ axis.

Fig. 2. The relaxation times $\tau_G$ as a function of temperature (a) for Y$_1$Ba$_2$Cu$_4$O$_8$, with the direction of polarization of probe pulse along $a$ axis (open triangles) and $b$ axis (squares), and (b) for YBa$_2$Cu$_3$O$_{y+6}$ (squares).
Femtosecond Pump-Probe Polarization Dependent Investigation

Fig. 3. (a) The temperature dependence of photoinduced amplitude for Y$_{1}$Ba$_{2}$Cu$_{3}$O$_{7}$, measured with the polarization of probe pulse along a axis; (b) The temperature dependence of photoinduced amplitude for Y$_{1}$Ba$_{2}$Cu$_{3}$O$_{7-δ}$. The values of $\Delta_0(0)$ and $\Delta_F$ obtained from the fits are also shown.

In Fig. 3, we compare the measured temperature dependence of the signal amplitude for YBCO124 along the $a$ axis and for YBCO123 with the theoretical predictions [4] for the sum of responses from BCS temperature-independent gap $\Delta_F$. The agreement between the data and the theory is seen to be good. The values for the temperature-dependent gap $\Delta_0(0)$ at $T = 0$ and $\Delta_F$, which are stated in the Fig. 3a, b, were obtained from the fits.

In Fig. 4, we show a polarization dependence of both amplitudes $G(T, \theta)$, $P(T, \theta)$ for YBCO124 at $T = 45$ K. $P(T)$ does not show any polarization dependence, which was also confirmed by polarization measurements above $T_c$. In contrast, the polarization dependence of $G(T)$ shows a significant angular dependence, exhibiting a change of sign as the polarization direction is changed from parallel to perpendicular to the Cu--O chains.

This angular dependence can be understood in terms of the anisotropy of the probe transition matrix elements. We first note that in spite of the fact that YBCO124 is orthorhombic, the main contribution to the probe signal comes from interband resonance transitions [7] which involves atomic wave functions in which the dipolar matrix elements have a four fold rotational symmetry around the $c$ axis. Without specifying exactly which transitions are involved, we can write the absorption coefficient in terms of the Fermi golden rule:

$$\alpha \propto \int d\epsilon N(\epsilon)N(\epsilon + \omega)|M(\epsilon, \omega)|^2 f(\epsilon)(1 - f(\epsilon + \omega))$$

Here $N(\epsilon)$ is the density of electronic states, $f(\epsilon)$ is the distribution function for holes, $\hbar\omega$ is the energy of the probe photons and $M(\epsilon, \omega)$ is the dipole matrix element for the transition. For small perturbations, $\Delta R$ is proportional to the photoinduced absorption $\Delta\alpha$, so:

$$\Delta R \propto \Delta\alpha \propto \int d\epsilon N(\epsilon)M(\epsilon, \omega)|^2 (f'(\epsilon) - f(\epsilon))$$  (1)

where $f'(\epsilon)$ is the nonequilibrium distribution function of the charge carriers. The integral is taken in the vicinity of the Fermi energy $E_F$ over the width of the resonance [7] and we assumed for simplicity that $N(\epsilon + \Omega)$ is constant within the resonance width.

If $M(\epsilon, \omega)$ is constant over the whole range of energies $-\hbar\omega < \epsilon - E_F < \hbar\omega$, then $\Delta R = \Delta\alpha = 0$ because of the conservation of particles. In other words, the photoinduced increase in absorption due to probe transitions 1 (see Fig. 5) originating from the photexcited electron states at $\epsilon - E_F \geq \Delta$ just above the gap to unoccupied hole states at $\epsilon - E_F \sim \hbar\omega + \Delta$ exactly cancel the decrease in absorption due to transitions 2 originating from occupied electronic states at $\epsilon - E_F \leq -\Delta$ just below the gap to unoccupied states at $\epsilon - E_F \sim \hbar\omega - \Delta$. The same is true for the hole transitions 3 and 4 (see Fig. 5).

Turning to the YBCO124 data we assume that $M(\epsilon, \omega)$ is not constant and proceed to derive an expression for the polarization anisotropy of the probe absorption. The general expression for the square of
the dipolar matrix element can be written as:

\[ |M(\epsilon)|^2 = M_x^2(\epsilon) \sin^2(\theta) + M_y^2(\epsilon) \cos^2(\theta) \]

where \( \theta \) is the angle between polarization of light and the \( b \) axis. For an orthorhombic structure \( M_x \neq M_y \). On the other hand, the main contribution to \( M \) which comes from atomic wave functions is independent of \( \theta \), so we can expand \( M \) in the vicinity of the Fermi energy and express matrix element in the form:

\[ M_{\alpha\beta}(\epsilon) = M_0 + \gamma_{\alpha\beta} \epsilon \]  \hspace{1cm} (2)

where \( \gamma_{\alpha\beta} = dM_{\alpha\beta}/d\epsilon \) and the derivative is taken at the Fermi energy. Substituting this expression into Eq. (1) we obtain a qualitative description of the angular dependence of the probe signal \( G(\theta) \) and \( P(\theta) \):

\[ \Delta R \propto \Delta \alpha \propto M_0(\gamma_x \sin^2(\theta) + \gamma_y \cos^2(\theta)) \Delta n \]  \hspace{1cm} (3)

Here \( \Delta n \) is the number of photoexcited quasiparticles, as described in the Ref. [4]. The values of \( \gamma_{\alpha\beta} \) can be depending on Doppler and can easily have different sign, depending strongly on \( \omega \) and the material’s band structure in the vicinity of the resonance \( \epsilon - E_F \approx \hbar \omega \pm \Delta \). However, since the probe polarization anisotropy is a consequence of the anisotropy of the probe transition matrix elements, Eq. (3) unfortunately does not give any direct information regarding the anisotropy of the low-energy electronic gap structure. In Fig. 3c, we plot the polarization dependence of photoinduced reflectivity amplitude given by Eq. (3) using \( \gamma_x/\gamma_y = 55/(1 - 10) \), which is shown to describe the main features of the data quite well.

In conclusion, the femtosecond relaxation dynamics in YBCO124 is found to be very similar to that reported previously in YBCO123 [1]. The presence of the two-component relaxation dynamics, which has been thus far observed in several HTSC [6,8–11], is confirmed in YBCO124 for different polarizations of probe pulses. The observed probe polarization dependence and sign change of the transient signal below \( T_c \) is described well by a model which considers the anisotropy of the probe transition matrix elements in YBCO124. These results also show that photoinduced changes in reflectivity can be explained by relatively straightforward physics [4], which is well understood.

ACKNOWLEDGMENTS

The authors would like to acknowledge A. Mironov of the Chemical Department Moscow State University for single-crystal X-ray investigation of the samples.

REFERENCES