Quasiparticle dynamics and gap structure in $HgBa_2Ca_2Cu_3O_{8+\delta}$ investigated with femtosecond spectroscopy

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Measurements of the temperature dependence of quasiparticle (QP) dynamics in $Hg_1Ba_2Ca_2Cu_3O_{8+\delta}$ with femtosecond time-resolved optical spectroscopy are reported. From the temperature dependence of the amplitude of the photoinduced reflection, the existence of two gaps is deduced, one temperature-dependent $\Delta_c(T)$ that closes at T_c, and another temperature-independent ''pseudogap'' Δ^p. The zero-temperature magnitudes of the two gaps are $\Delta_c(0)/k_BT_c=6\pm0.5$ and $\Delta^p/k_BT_c=6.4\pm0.5$, respectively. The quasiparticle lifetime is found to exhibit a divergence as $T\rightarrow T_c$ from below, which is attributed to the existence of a superconducting gap that closes at T_c. Above T_c the relaxation time is longer than expected for metallic relaxation, which is attributed to the presence of the pseudogap. The QP relaxation time is found to increase significantly at low temperatures. This behavior is explained assuming that at low temperatures the relaxation of photoexcited quasiparticles is governed by a biparticle recombination process.

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I. INTRODUCTION

Time-resolved measurements of quasiparticle dynamics in cuprate superconductors were shown recently to give significant new information about single-particle excitations and the low-energy structure of correlated electron systems such as high-T_c superconducting cuprates[1–3] and charge-density wave systems.[4] Systematic measurements on $YBa_2Cu_3O_{7-\delta}$ (YBCO) have shown the existence of two simultaneous gaps in the optimally doped and overdoped regions,[2] but only one (pseudo)gap in the underdoped region.[1] The observation of two energy scales with different temperature dependences were in apparent agreement with frequency-domain measurements like angle-resolved photoemission on $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212),[5] as well as Raman spectroscopy on both YBCO and Bi2212.[6] In this paper we report a series of measurements of quasiparticle dynamics on $Hg_1Ba_2Ca_2Cu_3O_{8+\delta}$ (Hg1223) with a T_c of 120 K and find some similarities with femtosecond spectroscopy measurements on YBCO, but also some differences, particularly the temperature dependence of the quasiparticle (QP) lifetime at low temperatures.

The femtosecond time-resolved pump-probe technique involves the measurement of small photoinduced changes in the optical reflectivity or transmittance of a sample caused by photoexcitation. After excitation with a high-energy photon (1.5 eV), the electrons and holes rapidly relax towards equilibrium; they scatter amongst themselves and subsequently with lattice phonons in a process described theoretically by Kaganov *et al.*[7] and Allen.[8] As the carriers reach low energies, the presence of a gap in the spectrum presents a bottleneck for further relaxation and the QP's accumulate at the band edge, waiting to recombine. A second suitably delayed probe laser pulse measures the change in reflectivity of the sample by excited-state absorption, with these QP's occupying initial states of the probe transition. Because the QP dynamics critically depends on the presence of a gap, the technique gives direct information about the temperature dependence of the QP lifetime and the T dependence of the gap magnitude. The details of the experimental technique as well as the theory describing how the gap magnitude is obtained from the data were described in detail elsewhere.[1,9]

II. EXPERIMENTAL DETAILS

The samples used in this investigation were single crystals of $Hg_1Ba_2Ca_2Cu_3O_{8+\delta}$ with a T_c of 120 K as determined from ac susceptibility measurements. The samples were prepared in Zürich by a high-gas-pressure synthesis (Ar pressure of 10–11 kbar and crystallization temperatures in the range of 995 °C$<T<$1025 °C), with $BaCuO_2$-CuO-Ag_2O eutectic mixture as a flux. Details of the preparation method are given in Ref. 10. As a result, platelike single crystals of Hg1223 with c-axis thickness of $\sim$10 μm and 0.3×0.3-mm^2 area were obtained. Great care was exercised to chose a sample that was without inclusions and was as much as possible single phase.

The photoinduced reflectivity measurements were performed using a standard pump-probe technique,[9] with a Ti:sapphire laser producing 70-fs pulses at approximately 800 nm (1.5 eV) as the source of both pump and probe optical pulses. The pump and probe pulses were cross polarized with polarizations parallel to the a-b plane of the sample. The experiments were performed at typical pump-pulse fluence $\mathcal{E}_0=1.3$ μJ/cm^2 (taking a typical pump-pulse energy of 0.1 nJ, and a spot diameter of $\sim$100 μm). The probe intensity was approximately 100 times lower. Estimating that each absorbed photon with energy $E=1.5$ eV excites $N_{QP}=$30-40 quasiparticles ($N_{QP}=E/2\Delta$, where Δ is the magnitude of the superconducting gap) and taking the optical absorption length to be approximately 100 nm, we find the number of photogenerated QP's due to excitation with a pump pulse to be of the order of $n_{pe}\lesssim 3\times10^{-3}$/unit cell. On the other hand, the typical carrier concentration relevant for superconductivity is $n_0=2N(0)\Delta\simeq$0.2-0.4/unit cell, where

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$N(0)$ is the density of states at E_F. From the ratio $n_{pe}/n_0 \lesssim 10^{-2}$ we can see that we are dealing with weak perturbations of the electronic system and therefore the pump-probe experiments are probing the equilibrium properties of the system.

Another important experimental detail that needs to be further discussed is the sample heating, which takes place due to excitation of the sample with the train of pump pulses (the heating due to the probe pulse train can be neglected). In general there are two effects that need to be considered: (i) transient heating due to absorption of the single pulse and (ii) steady-state heating, which results in a steady-state temperature increase of the probed area (in this case the train of pulses separated by 12 ns equals the continuous wave (CW) laser beam with the same average power). It can be shown that in this low photoexcitation regime the transient heating is of the order of 0.1 K and can be neglected.[11] Steady-state heating can be quite substantial ($\sim$10 K) and should be taken into consideration[9,11] since the temperature of the probed spot T_s may differ substantially from the temperature of the cold finger T_{cf}, which is directly recorded.

The steady-state temperature increase $\Delta T_{cw} = T_s - T_{cf}$ can be accurately determined at temperatures close to T_c in several ways.[9,11] In the analysis of the data taken on Hg1223 single crystals we used the so-called ''scaling'' method. Namely, in Hg1223 both amplitude and relaxation time of the photoinduced picosecond reflectivity transient show anomalies near T_c associated with the opening of the superconducting gap.[1] These have thus far already been observed in various cuprates near optimal doping.[11–14] When the data obtained with different average powers of the pump beam are systematically compared, anomalies appear at different cold finger temperatures. Since the temperature increase is linearly proportional to the power of the pump beam, one can scale the data and determine the temperature increase near T_c quite accurately.[9] With typical experimental conditions $\Delta T_{cw}(T_c) = 9$ K was determined, in close agreement with the calculation using a heat-diffusion model.[11] Since ΔT_{cw} is inversely proportional to the thermal conductivity κ of the sample, one can estimate the temperature increase in the whole temperature range[15] providing $\Delta T_{cw}(T_c)$ is known and the thermal conductivity data are available. In our analysis the thermal conductivity data from Ref. 16 were used. In all the presented data, temperature increases due to heating of the probed spot were accounted for, with about ± 2 K uncertainty over the whole temperature range.

III. EXPERIMENTAL RESULTS AND DATA ANALYSIS

In Fig. 1 we show the time dependence of the photoinduced signal at a number of temperatures below and above T_c. The time evolution of the photoinduced reflection $\Delta R/R$ first shows a rapid rise time (of the order of the pump pulse length) and a subsequent picosecond decay. These data can be fit quite well using a single exponential decay (see Fig. 1) over the whole temperature range.

In addition to the picosecond decay, we also observe the ubiquitous near-constant background,[17] which has a lifetime longer than 12 ns.

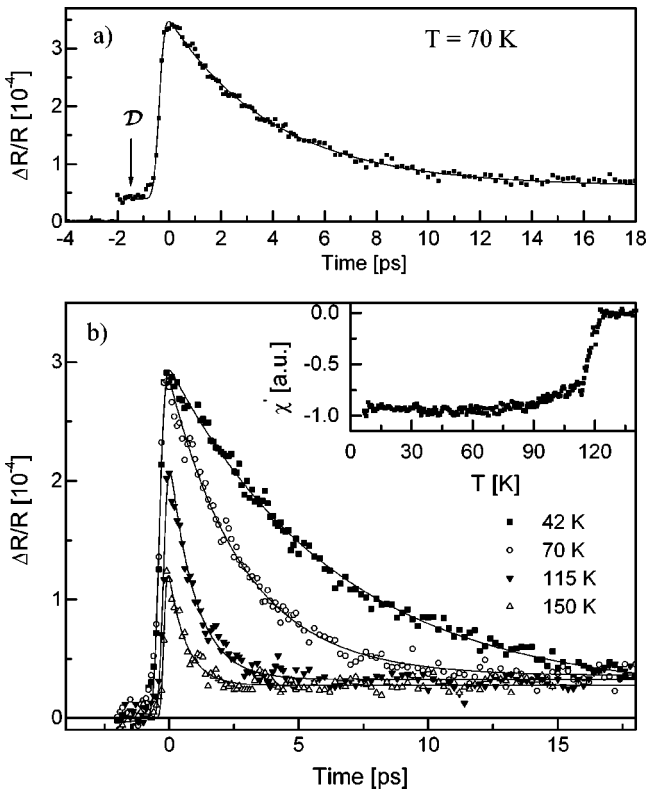

FIG. 1. (a) $\Delta R/R$ taken on Hg1223 at 70 K. The fast rise time is followed by a picosecond decay. Some long-lived photoinduced signal (difference between signal at point $\mathcal{D}$ and zero signal, when pump beam is blocked) persists up to 12 ns (difference between two successive pump pulses), resulting in a temperature-dependent offset. (b) $\Delta R/R$ at various temperatures below and above T_c. In these traces the T dependent background, $\mathcal{D}$, is subtracted. (Its T dependence is analyzed separately.) Inset: Real part of the ac susceptibility taken on one of the samples, showing a T_c onset at 120 K.

A. Amplitude of the picosecond relaxation component

The temperature dependence of amplitude $|\Delta R/R|$ and relaxation time τ_R of the picosecond component in photoinduced reflectivity are shown in Fig. 2. The picosecond component amplitude $|\Delta R/R|$ is almost constant at low temperatures, followed by a rapid decrease as T_c is approached. At T_c there appears to be a break in the T-dependence and above T_c the amplitude decreases much more gradually, falling asymptotically to zero at higher temperatures. (At temperatures above $\sim$230 K it is difficult to extract the value of the amplitude of the picosecond relaxation component, because some very fast sub-100 fs relaxation becomes evident, which we attribute to metallic relaxation.[8,7,18]) It is worth mentioning here that the T-dependence of $|\Delta R/R|$ [Fig. 2(a)] shows almost perfect agreement with the data on near-optimally doped YBCO.[1,2,12]

To analyze the temperature dependence of $|\Delta R/R|$ quantitatively, we use the model by Kabanov *et al.*,[1] where the T-dependence of the photoexcited QP density n_{pe} was derived for different gaps. The model, which was tested also on quasi-one-dimensional semiconductor $K_{0.3}MoO_3$,[4] is based on the relation

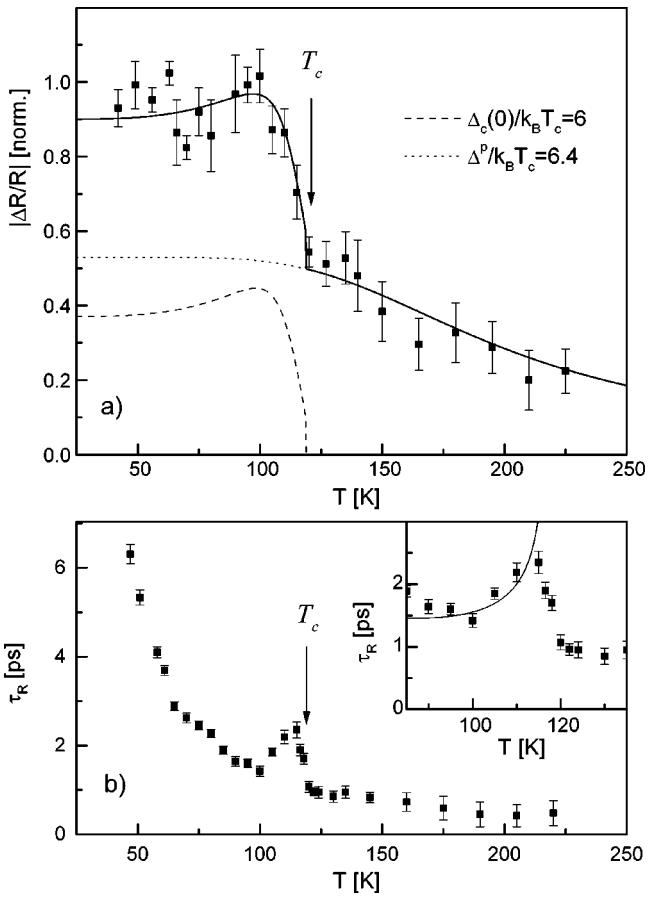

FIG. 2. (a) Amplitude of the picosecond component, $|\Delta R/R|$, fit with the sum of two components (solid line) given by Eq. (2) (dashed), and Eq. (3) (dotted). Values of $\Delta_c(0)$ and Δ^p from the fit are shown. (b) The temperature dependence of relaxation time τ_R. Inset: the divergence at T_c is compared to the $1/\Delta_c(T)$, with $\Delta_c(T)$ having BCS T dependence.

$$\Delta R = \frac{\partial R}{\partial n} n_{pe}. \tag{1}$$

The above relation is valid if n_{pe} is small with respect to the number of thermally excited QP's, which is the case in our experimental configuration, see Refs. 1 and 9. In other words the perturbation is small and hence linear response is sufficient. Moreover, it is known that the reflectivity R in high-T_c superconductors (HTSC's) is a very weak function of temperature in that particular energy range (1.5 eV), as observed experimentally by Holcomb *et al.*[19] using the thermal-difference reflectivity data on a variety of high-T_c superconductors. Therefore we expect that the derivative $\partial R/\partial n$ taken in the equilibrium limit (since we are dealing with linear response) is a weak function of T, and in the first approximation we can consider it to be constant. It should be pointed out that calculation of the constants $\partial R/\partial n$ is the subject of the microscopic theory and probably very model dependent. Our approach is more phenomenological and requires only that $\partial R/\partial n$ is weakly temperature dependent.

From the T dependence of the reflectivity amplitude (which is proportional to n_{pe}) and relaxation time one can, using the analytical expressions connecting n_{pe} and magnitude and T dependence of the gap $\Delta(T)$, extract the magnitude and T dependence of the gap.

In the limit of small photoexcited carrier density n_{pe}, we can assume that all possible contributions to $\Delta R/R$—arising from excited-state absorption or photoinduced band-gap changes, for example—are linear in the photoexcited carrier density n_{pe}. For an isotropic T-dependent gap $\Delta_c(T)$, the temperature dependence of the amplitude of the photoinduced reflectivity $|\Delta R/R|$ is given by[1]:

$$|\Delta R/R| \propto n_{pe} = \frac{\mathcal{E}/[\Delta_c(T) + k_B T/2]}{1 + \dfrac{2\nu}{N(0)\hbar\Omega_c}\sqrt{\dfrac{2k_BT}{\pi\Delta_c(T)}}\, e^{-\Delta_c(T)/k_BT}}, \tag{2}$$

where $\mathcal{E}$ is the incident energy density of the pump pulse per unit cell, ν is the effective number of phonon modes interacting with the QP's, $N(0)$ is the density of states (DOS) at E_F, and Ω_c is a typical phonon cutoff frequency. A similar expression gives the amplitude for an isotropic T-independent gap Δ^p,[1]

$$|\Delta R/R| \propto n'_{pe} = \frac{\mathcal{E}/\Delta^p}{1 + \dfrac{2\nu}{N(0)\hbar\Omega_c} e^{-\Delta_p/k_BT}}. \tag{3}$$

In Fig. 2(a) we fit the temperature dependence of $|\Delta R/R|$ with the sum of Eqs. (2) and (3) using $\nu=10$, $\Omega_c=0.1$ eV and $N(0)=5\ \mathrm{eV}^{-1}\,\mathrm{cell}^{-1}\,\mathrm{spin}^{-1}$. It is evident from the plots that the total amplitude $|\Delta R/R|$ cannot be described by either component separately. However, assuming the coexistence of two gaps, one of which is T dependent with a BCS-like temperature dependence and one T independent, we can obtain a good fit to the data as shown in Fig. 2(a). The values of $\Delta_c(0)$ and Δ^p obtained from the best fit are $6\pm0.5k_BT_c$ and $6.4\pm0.5k_BT_c$, respectively.

At this point we should state that a simple prediction for the case of the *d*-wave gap (gapless DOS) cannot account for the observed data. Namely, as soon as we assume that the density of states is gapless, then after the initial relaxation, QP's would accumulate in the nodal regions and the number of photoexcited QP's can be approximated as $n_{pe}=E_i/T^*$, where T^* is their effective temperature. This means that sublinear dependence of the photoinduced signal amplitude as a function of photoexcitation intensity E_i should be observed in case of gapless density of states. This is due to the temperature dependence of the electronic specific heat in case of a *d*-wave superconductor, which goes as $C_{el}\propto T^2$. This clearly contradicts our experiments, since linear dependence of photoinduced reflectivity amplitude on E_i was observed over a wide range of photoexcitation energies, see Refs. 1 and 20. Moreover, for the *d*-wave case the model (see Ref. 1 for details) predicts nonexponential relaxation, no anomaly at T_c in the relaxation time, and different T dependence of the picosecond component amplitude, all of which are not consistent with the data.

B. Relaxation time

The T dependence of τ_R obtained by the single exponential decay fit to the data is shown in Fig. 2(b). There are two noticeable features in the observed temperature dependence. First, at temperatures above $\sim$80 K the T dependence is quite similar to the behavior seen in optimally doped and overdoped YBCO (Refs. 1,2,12) and Tl2223 (Ref. 13). Upon increasing the temperature through T_c, the relaxation time [see inset to Fig. 2(b)] shows an anomaly. Such an anomaly is expected to occur in the relaxation time in the presence of a gap that closes at T_c.[1] Near T_c the relaxation is governed by the anharmonic decay time of high-frequency phonons[1] given by

$$\tau_{ph}=\frac{\hbar\omega^2 \ln\{1/[\mathcal{E}/2N(0)\Delta_c(0)^2+e^{-\Delta_c(T)/k_BT}]\}}{12\Gamma_\omega\Delta_c(T)^2}, \quad (4)$$

$\Delta_c(T)$ being the magnitude of the T-dependent gap, ω is the phonon frequency (typically $\omega\simeq 500\ \mathrm{cm}^{-1}$), and Γ_ω is the optical phonon linewidth, typically $10\mathrm{cm}^{-1}$. Near T_c, $\tau_{ph}\propto 1/\Delta_c(T)$ as plotted in the inset to Fig. 2(b). Divergence below T_c is followed by a rapid drop to a lower value, and above T_c the relaxation time shows only a weak T dependence. It needs to be mentioned that above T_c, τ_R is much longer than expected for metallic relaxation,[1,18] implying the presence of a pseudogap in the density of states as already observed in YBCO (Ref. 2) and consistent with the observed T dependence of $|\Delta R/R|$. At temperatures above $\sim$230 K an additional, very fast relaxation with sub-100-fs relaxation time becomes evident, which we attributed to metallic relaxation. At temperatures below $\sim$80 K, unlike in YBCO,[1] τ_R shows a strong T dependence—increasing rapidly as the temperature is decreased. A similar T dependence has been reported also in Bi2212 (Ref. 21) and $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201).[3] The possible origin of the different low-temperature behaviors in various HTSC's will be discussed in the Discussion section.

C. The amplitude of the nanosecond component

In Fig. 3 we show the T dependence of the long-lived photoinduced signal amplitude $\mathcal{D}$ (see Fig. 1). The lifetime appears to be much longer than the distance between two successive pump laser pulses, so its relaxation time cannot be measured directly. However, from the comparison of the amplitude at negative time delays ($\sim$12 ns after photoexcitation) with the photoinduced signal at 100 ps (when there is no picosecond relaxation signal left), one can estimate the relaxation time to be of the order of 100 ns or longer.[17] Indeed, a similar photoinduced signal was observed also on the 100-μs timescale,[22] which is close to the measured μs dynamics in the photoinduced absorption of mm waves.[23] Taken all together it seems we are dealing with glasslike relaxation dynamics with no well-defined timescale.[24]

At low temperatures the signal amplitude increases upon increasing temperature, which is contrary to the expected behavior due to heating.[11] At T_c the amplitude drops substantially followed by a gradual decrease at higher temperatures. The nanosecond relaxation component was previously

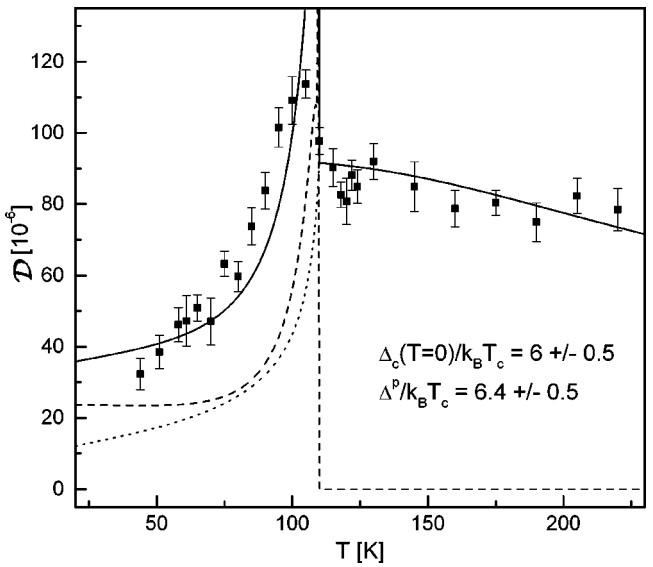

FIG. 3. The T dependence of the slow component amplitude $\mathcal{D}$ [see Fig. 1(a)], together with the fit using theoretical model for photoinduced absorption from in-gap localized states, see text.

observed also on YBCO (Ref. 14) and the quasi-1D-CDW (charge-density wave) semiconductor $K_{0.3}MoO_3$.[4] Its lifetime and T dependence suggested an explanation for the long-lived signal in terms of in-gap localized states.[17] The model gives the T dependence of slow component amplitude for different T dependencies of the gap. In case the gap is mean-field-like, closing at T_c the T dependence of $\mathcal{D}$ is given by[17]

$$\mathcal{D}\propto\sqrt{\eta n_{pe}(T)}\,\frac{\Omega_c}{\Delta_c(T)}, \quad (5)$$

where $\eta=\gamma\tau_{ph}\propto\eta'/\Delta_c(T)$ is the probability of trapping a QP into a localized state, $n_{pe}(T)$ is the number of photoinduced QP's at temperature T created by each laser pulse given by Eq. (2), Ω_c is the phonon cutoff energy, and $\Delta_c(T)$ is the T-dependent gap magnitude. In case the gap Δ^p is T independent, the model gives[17]

$$\mathcal{D}\propto\sqrt{\frac{\eta n'_{pe}(T)}{\alpha[1-(T/T_c)]+\Gamma}},$$

$$T<T_c, \quad \alpha>0, \quad T>T_c, \quad \alpha=0. \quad (6)$$

Here n'_{pe} is given by Eq. (3) and Γ is the T-independent biparticle recombination time present also at temperatures above T_c since Δ^p is T independent. Below T_c, the presence of the condensate may also have an effect on the recombination of localized excitations, which makes the first term proportional to the square of the order parameter in the denominator.[17]

The data (see Fig. 3) show substantial long-lived photoinduced signal present also at temperatures above T_c, suggesting the presence of a gap (and in-gap localized states) also at high temperatures, similar to that deduced from the picosecond relaxation data, see Fig. 2. We thus fit the data

with the model[17] assuming the coexistence of two gaps, a T-dependent gap $\Delta_c(T)$, and a T independent (pseudo)gap Δ^p, with the two contributions to the signal $\mathcal{D}$ given by Eqs. (5) and (6), respectively. Substituting $\Delta_c(0)$ and Δ^p from fits to the picosecond decay components into Eqs. (5) and (6) we obtain the solid line fit in Fig. 3. The two components are plotted separately by dashed [Eq. (5)] and dotted [Eq. (6)] lines, respectively.

IV. DISCUSSION

The present measurements appear to show very similar two-gap behavior as in overdoped and optimally doped YBCO.[2] In particular, both show an apparent coexistence of two gaps for QP excitations, that is, a pseudogap Δ^p coexisting with a collective temperature-dependent gap $\Delta_c(T)$ that closes at T_c. Unlike in YBCO, where the two relaxation components (one present also above T_c with T-independent τ_1, and the other present only at $T<T_c$ with τ_2 diverging at T_c) (Ref. 2) are clearly distinguishable in the decay because of their very different lifetimes, on Hg1223 the relaxation is well reproduced by single exponential decay. However the presence of a picosecond timescale relaxation above T_c, together with an asymptotic decrease in $|\Delta R/R|$ at high temperatures, suggests similar two-component behavior, with the two relaxation times too close to be resolved. This is supported by the fact that the relaxation time at $T>T_c$ in Hg1223 is nearly the same as at $T<T_c$ ($\sim$100 K), whereas in YBCO it is found to be almost an order of magnitude lower.[2] Two such distinct picosecond relaxation components with opposite signs were observed also on $Tl_2Ba_2Ca_2Cu_3O_{10}$ (Tl2223),[13] Bi2212,[21] and Tl2201,[3] suggesting that the two-component behavior is quite general in high-T_c superconductors near optimal doping. Furthermore, the two-gap behavior is clearly apparent in the slow component T dependence as discussed in the previous section. We note that the apparent similarity in all these HTSC materials is very important from the theoretical point of view of universality of the low-energy excitations in cuprate superconductors. More specifically, the apparently universal coexistence of two components (two gaps) in YBCO, Bi2212, Hg1223, Tl2201, and Tl2223 appears to impose some quite stringent restrictions on the theoretical framework for the solution of the high-T_c problem.

The main difference between the data on Hg1223 and optimally doped YBCO is in the behavior of the relaxation time at low temperatures. While in YBCO, τ_R is almost T independent at low temperatures,[12,1] showing only slight upturn at very low temperatures,[12,2] in Hg1223, τ_R increases significantly upon lowering the temperature from $\sim$80 K. A similar T dependence was observed also in Tl2201 (Ref. 3 and Bi2212,[21] suggesting that this low-T behavior of τ_R is not universal.

In order to understand the low-temperature behavior of τ_R in Hg1223, and account for the difference in behavior between YBCO and Hg1223, we have analyzed the processes governing the relaxation of photoexcited quasiparticles in more detail.

We first consider the theoretical model for quasiparticle relaxation,[1] which quantitatively described the T dependence of QP relaxation in YBCO (Ref. 1) as well as in the quasi-1D-CDW semiconductor $K_{0.3}MoO_3$ (Ref. 4) over a wide range of temperatures. The model assumes that after excitation with a high-energy photon, the electrons and holes rapidly relax towards equilibrium; they scatter amongst themselves (quasiparticle avalanche multiplication due to electron-electron collisions) and subsequently with lattice phonons reaching states just above the band edge within $\tau \ll 100$ fs.[8,7,18] The photoexcited QP's recombine with the creation of high-frequency phonons with $\hbar\omega > 2\Delta$. High-frequency phonons, on the other hand, get reabsorbed creating new pairs of QP's, or anharmonically decay into low-energy phonons ($\hbar\omega < 2\Delta$), which cannot excite new QP's because of energy conservation. In case the recombination and reabsorption processes are fast compared to the anharmonic phonon decay (typically a few ps) a near-steady-state distribution of the QP's and high-frequency phonons is established on a sub-100-fs timescale, described by common temperature. The relaxation rate of the photoinduced QPs is then dominated by the energy transfer from high-frequency phonons with $\hbar\omega > 2\Delta$ to phonons with $\hbar\omega < 2\Delta$ (Ref. 1) given by Eq. (4).

The assumption that recombination is fast compared to the anharmonic phonon decay, however, can be violated at low temperatures, when the gap is large and the number of thermally excited QP's is small. It can lead to a situation when the recombination time becomes longer than the anharmonic phonon decay time. In this case the relaxation time of the photoexcited QP density is governed by biparticle recombination process, and QP's and high-energy phonons can be described by quasiequilibrium distribution functions with different temperatures T_{qp} and T_{ph}. (Note that both temperatures are higher than the equilibrium lattice temperature T, which is also the temperature of the low-energy phonons.[1])

To estimate the temperature dependence of the relaxation time of photoexcited QP density (governed by biparticle recombination process) at low temperatures, we consider the kinetic equation for QP's.[25] The collision integral describing the kinetics of the QP's has two different terms. The first one describes inelastic scattering of QP's (with creation or absorption of a phonon) and the second describes the recombination (or creation) of two QP's with creation (or absorption) of a high-frequency phonon ($\hbar\omega > 2\Delta$). The ratio of these two terms is determined by coherence factors, and when $T \ll T_c$, the first term is small as $\sim(T/\Delta)^2$. In this case the rate equation for the total density of QP's, $n(T_{qp})$, can be reduced to the equation (see also Ref. 26)

$$\frac{\partial n(T_{qp})}{\partial t} = \frac{8\pi\lambda\Delta^2}{\hbar\Omega_c N(0)}[n^2(T_{qp}) - n^2(T_{ph})]. \tag{7}$$

As can be seen from this equation when $T_{qp} \gg T_{ph}$ the relaxation is nonexponential. On the other hand, when $T_{qp} \approx T_{ph}$, we can linearize the right-hand side of Eq. (7) to obtain

$$\frac{\partial n(T_{qp})}{\partial t} = \frac{1}{\tau_{rec}}[n(T_{qp}) - n(T_{ph})], \tag{8}$$

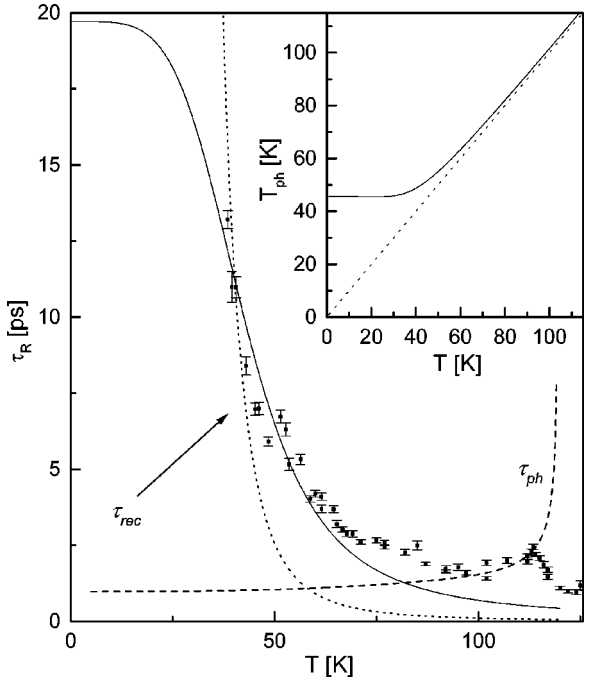

FIG. 4. The relaxation-time data compared to the theoretical predictions. Equation (4) is plotted by dashed curve, whereas expression (9) with $T_{ph}=T$ and T_{ph} given by Eq. (10) is plotted with dotted and solid line, respectively. At temperatures below ~30 K Eq. (9) is expected to fail, and the relaxation becomes nonexponential. Inset: The T dependence of the high-energy phonon temperature T_{ph} for $T_{ph}=T$ (dotted) and T_{ph} given by Eq. (10) (solid).

with

$$\tau_{rec}=\frac{\hbar\Omega_c^2}{32\pi\lambda\Delta^2\sqrt{\pi\Delta k_B T_{ph}/2}}\exp(\Delta/k_B T_{ph}). \qquad (9)$$

Here λ is the dimensionless electron-phonon coupling constant for HTSC's typically of the order of 1.[18] As a result, τ_{rec} shows an exponential increase at low temperatures, whereas the relaxation time of the high-frequency phonons is constant at low T.[1] It means that at some temperature $T<T_c$ we should expect a crossover from high-temperature relaxation behavior near T_c, as described previously in Ref. 1, to low-temperature recombination that shows different behavior and is described by Eq. (9).

In Fig. 4 we plot the relaxation time data, together with expressions (4) and (9) describing the T dependence of the relaxation times τ_{ph} and τ_{rec}, respectively. At temperatures close to T_c the divergence in relaxation time τ_R is well reproduced by τ_{ph}, whereas at temperatures below ~70 K τ_R becomes larger than predicted τ_{ph}.

To be able to compare the low-temperature relaxation time data on Hg1223 with Eq. (9), there is an important detail that needs further discussion. Namely, the temperatures T_{qp} and T_{ph} entering Eqs. (7) and (9) depend on T and Δ.[1] Near T_c, when the gap is small, the number of photoexcited carriers is small compared to the number of thermally excited QP's (the same goes also for densities of high-energy phonons); therefore T_{qp} and T_{ph} are very close to the equilibrium lattice temperature T. At low temperatures, on the other hand, the situation is changed and even weak photoexcitation strongly increases T_{qp} (and T_{ph}) with respect to the equilibrium temperature T. Assuming that all the absorbed energy goes to the quasiparticle system one obtains[1]

$$k_B T_{qp}\simeq\Delta(T)/\ln(1/\{\mathcal{E}/2N(0)\Delta^2(0)+\exp[-\Delta(T)/k_B T]\}) \qquad (10)$$

giving $T_{qp}\simeq T_c/2$ for the limiting case when $T\to 0$ using the above experimental configuration.[1] In case the anharmonic phonon decay is faster than the biparticle recombination time, T_{ph} is expected to be lower than T_{qp}, expression (10) giving an upper limit for T_{ph}. Since the main T dependence of τ_{rec} [Eq. (9)] comes from $\exp(\Delta/k_B T_{ph})$, small changes in T_{ph} bring substantial change in τ_{rec}; therefore this surely is an important issue. In Fig. 4 we plot Eq. (9) using two extreme cases: the dotted line represents expression (9) with $T_{ph}=T$, whereas the solid line represents Eq. (9) where T_{ph} is given by Eq. (10) and plotted in inset to Fig. 4. As can be seen both fits account reasonably well for the data, giving $\Delta/k_B T_c\approx 2-4$ depending strongly on $T_{ph}(T)$. At temperatures below ~30 K τ_{rec} is expected to saturate. However at a low temperature, T_{qp} is expected to be substantially higher than T_{ph} leading to nonexponential relaxation. Indeed the crossover to nonexponential relaxation was reported at very low temperatures in Bi2212 (Ref. 21) and Tl2201.[3] Considering that the T dependence of τ_{rec} is governed by $\exp(\Delta/k_B T_{ph})$, a crossover from high-temperature *relaxation* to low-temperature *recombination* picture is expected to highly depend on the magnitude of the superconducting gap Δ. Since the gap value in YBCO, determined from tunneling data is lower than that of Bi2212 (and Hg1223) the crossover is expected to be lower in temperature.

V. CONCLUSIONS

We have performed measurements of the temperature dependence of quasiparticle dynamics in $HgBa_2Ca_2Cu_3O_{8+\delta}$ with femtosecond time-resolved optical spectroscopy. From the temperature dependence of the amplitude, and the relaxation time of the photoinduced reflection, the existence of two gaps is deduced, one temperature-dependent $\Delta_s(T)$ that closes at T_c, and a temperature-independent pseudogap Δ^p. The zero-temperature magnitudes of the two gaps obtained from the fit to the data, using the theoretical model by Kabanov *et al.*[1] are $\Delta_s(0)/k_B T_c=6\pm 0.5$ and $\Delta^p/k_B T_c=6.4\pm 0.5$, respectively. In addition to the picosecond quasiparticle relaxation component a long-lived nanosecond component was observed, whose dynamics is described with the model for photoexcited localized in-gap state relaxation.[17]

Unlike in YBCO (Ref. 2) the relaxation of the picosecond transient is found to be single exponential over the whole T region, showing a significant increase at low temperatures. From the model analysis we suggest that at low temperatures the relaxation time is dominated by biparticle recombination in this material and find good agreement between the model and the data. The fact that the relaxation times for the pseudogap relaxation and collective-gap relaxation are nearly the same in Hg1223, while in YBCO they differ by almost an order of magnitude still needs to be understood.

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