Low-energy electronic structure in \( Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta} \): comparison of time-resolved optical spectroscopy, NMR, neutron and tunneling data.

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Abstract. Time-resolved optical measurements give information on the quasiparticle relaxation dynamics in YBCO, from which the evolution of the gap with doping and temperature can be systematically deduced. In this paper these optical charge-channel "pseudogap" data are compared with the "pseudogap" obtained from the NMR Knight shift \( K_s \), spin polarized neutron scattering (SPNS) and single particle tunneling measurements. A simple energy level diagram is proposed to explain the different "gap" magnitudes observed by different spectroscopies in \( YBa_2Cu_3O_{7-\delta} \), whereby the spin gap \( \Delta_s \) in NMR and SPNS corresponds to a triplet local pair state, while \( \Delta_p \) in the charge excitation spectrum corresponds to the pair dissociation energy. At optimum doping and in the overdoped state, an additional \( T \)-dependent gap becomes evident, which closes at \( T_c \), suggesting a cross-over to a more conventional BCS-like superconductivity scenario.

I INTRODUCTION

Spectroscopic studies of cuprates over the years have shown that at low energies these materials exhibit quite a complex spectral structure, which changes with temperature and doping in a complicated way. Often there appears to be reasonable agreement regarding some of the main features between various experimental techniques. For example, optical femtosecond quasiparticle relaxation measurements and single-particle (Giever) tunneling show a similar size pseudogap in the spectrum over a large portion of the phase diagram, and the latter is remarkably similar to the spectral features measured by angle-resolved photoemission. However, the pseudogap \( \Delta_s \) as observed by spectroscopies like NMR and spin-polarized neutron scattering appears to be smaller than the optical and tunneling pseudogap \( \Delta_p \) by approximately a factor of 2, for which there is as yet no accepted theoretical explanation. Thus in spite of the availability of spectral data over a large range of doping in many materials, the interpretation of the low-energy excitation spectrum is still highly controversial. In this paper we summarize some of the results of
time-resolved quasiparticle recombination spectroscopy as a function of doping and temperature in YBa$_2$Cu$_3$O$_{7-\delta}$, which appear to give qualitative new insight into the origin of the low-energy spectral features of this material and its phase diagram. We compare the doping-evolution of the pseudogap $\Delta_p$ obtained from time-resolved spectroscopy with a quantitative analysis of the "spin-gap" from the NMR knight shift $K_s$ in the underdoped state using the same type of $T$-independent gap as deduced from the quasiparticle relaxation data. We find that the time-resolved QP relaxation data are quite consistent with Giever tunneling and ARPES data and suggest that all the observations together can be explained by the existence of a pair-breaking pseudogap $\Delta_p$ and a triplet-state local pair excitation $\Delta_s$ at $E \approx \Delta_p/2$.

II TIME-RESOLVED SPECTROSCOPY RESULTS

The time-resolved optical spectroscopy applied to superconductors and other materials with a gap has been given in detail elsewhere [1,2], so here we shall discuss only the results. The equations describing the temperature dependence of photoexcited QP density and their lifetime as a function of temperature are given by Kabanov et al [1]. Using these equations, the properties of the gap can be investigated as a function of doping and temperature. Systematic experiments on

![Graph showing the photoinduced reflectivity in YBa$_2$Cu$_3$O$_{6.95}$ showing the two-component relaxation below $T_c$ attributed to the two gaps $\Delta_p$ and $\Delta_{BCS}$](image)

YBaCuO as a function of O concentration over a wide range of $\delta$ have shown very clear and systematic behavior of the QP dynamics [1]. Particularly important is that the results are quite insensitive to surface quality and have been repeated after a period of a year with the same results. Single crystals were also studied and no
significant differences were found in comparison with the thin film data. These measurements have recently been extended to the overdoped state using calcium-doped (Y,Ca)Ba$_2$Cu$_3$O$_{7-\delta}$ crystals by Demsar et al [2], giving data on the whole phase diagram from insulating and underdoped to overdoped YBCO. A typical time-resolved signal in the optimally doped phase is shown in Figure 1. Above $T_c$, one exponential short-lived decay is observed whose amplitude is decreasing as $T$ is increased (Figure 2), while below $T_c$, two exponentials are clearly seen, one of which has a longer lifetime which is temperature-dependent and diverges as $T \to T_c$. Such behaviour is typical also for overdoped samples, but not for underdoped samples, where only a single exponential decay is observed, with a $T$-independent time constant $\tau_p \sim 0.4$ ps showing no anomaly at $T_c$. From these experiments we can deduce that in underdoped state the evolution of the QP dynamics with temperature and doping is dominated by a temperature-independent pseudogap $\Delta_p$. Near optimum doping an additional BCS-like temperature-independent gap $\Delta_{BCS}$ appears below $T_c$, which is present also in all the overdoped samples [2] and gives rise to the additional temperature-dependent relaxation process below $T_c$.

![Figure 2](image.png)

**FIGURE 2.** The amplitude of the photoinduced absorption (PIA) as a function of temperature for optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$ ($\delta \sim 0.05$). Overdoped samples with substituted Ca for Y show similar 2-component $T$-dependence for all Ca concentrations. b) The temperature dependence of the PIA for underdoped YBa$_2$Cu$_3$O$_{7-\delta}$ ($\delta > 0.15$). The lines are the theoretical fits to the data [Ref. 1] with $\Delta_p$ or $\Delta_{BCS}(T = 0K)$ as the only fitting parameter.

The fact the $\tau$ diverges near $T_c$ unambiguously signifies that $\Delta_{BCS} \to 0$ at $T_c$. Thus close to optimum doping and in the overdoped state two gaps appear to be present simultaneously, a feature consistent with the spatially inhomogeneous
ground state [3] in which the pair-breaking excitation is represented by $\Delta_p$, while the $\Delta_{BCS}$ is a gap associated with the collective behaviour of high-carrier density stripes or clusters which start to form near optimum doping. The values of the gaps $\Delta_p$ and $\Delta_{BCS}$ as a function of doping obtained by fitting the temperature dependence of the photoinduced signal amplitude as shown in Fig. 2 with the model calculation of Kabanov et al [1] are plotted in Figure 4.

### III COMPARISON WITH OTHER SPECTROSCOPIES

From the time-resolved data on the underdoped YBCO we have deduced that the low-energy spectrum can be described by a single temperature-independent gap $\Delta_p$. Starting from this observation we decided to analyse the temperature dependence of the NMR Knight shift $K_s$ available from the literature using the same $T$-independed $\Delta_p$. The aim is a) to see if the simple model can describe the $T$-dependence of $K_s$ and b) to see if the values of the pseudogap obtained for the NMR $K_s$ agree with the optical values. The NMR knight shift for such a case can be written as [4] $K_s = K_0 + AT^{-\beta} \exp[-\Delta / k_B T]$, where $K_0$ is the value of $K_s$ at zero temperature, $\beta$ is an exponent which depends on the shape of the singularity of the DOS at the gap and $A$ is a constant depending on the NMR nucleus.

**FIGURE 3.** The $^{89}$Y NMR Knight shift $K_s$ as a function of temperature in YBa$_2$Cu$_3$O$_{7-\delta}$ for different $\delta$ [5]. b) $K_s$ as a function of temperature in YBa$_2$Cu$_4$O$_8$ ($T_c=81$ K). The solid squares data are for $^{63}$Cu from Curro et al [6] and the open circles for $^{17}$O from Williams et al [7].

The results of fits to published data in YBCO 123 and 124 on $^{89}$Y [5], $^{63}$Cu [6] and $^{17}$O [7] are shown in Figure 3 using $\beta = 1/2$. In spite of its simplicity, the
model appears to describe the data very well. The gap values $\Delta_s$ with $\beta = 1/2$ obtained for YBa$_2$Cu$_3$O$_{7-\delta}$ are shown in Figure 4. $\Delta_s$ appears consistently lower than the $\Delta_p$ by approximately a factor of 2.

Apart from NMR $K_s$, spin-polarized neutron scattering (SPNS) also shows a spin-excitation peak at 34 meV (~390 K) in underdoped YBa$_2$Cu$_3$O$_{6.6}$, which is smaller than $\Delta_p$ by approximately a factor of 2. However, the anomalies in the phonons signifying the presence of charge excitations occur near 70 meV (~810K), also approximately twice the spin excitation energy and very close to the $\Delta_p$ in Fig. 4 (see Mook et al, this volume).

To explain the two different gaps $\Delta_p$ and $\Delta_s$, we propose a rather straightforward electronic structure in YBCO. A schematic diagram is shown in the insert to Fig. 4. The ground state is composed of local $S=0$ singlet Cooper pairs. Since $\Delta_p$ is a QP charge excitation it is clearly associated with pair breaking. However, if $J < \Delta_p$, the triplet local pair state also exists and lies within the gap. It should be visible by spin-flip spectroscopies like NMR and SPNS, but not by optical spectroscopy.
or SP tunneling which are charge excitations. We therefore propose that the $\Delta_s$ observed by NMR and SPNS is the S=1 local pair triplet excited state. From the experimental data in Fig. 4, both $\Delta_p$ and $\Delta_s$ decrease with increasing doping, more or less as $1/x$, where $x$ is the hole density. For low doping, at $\delta \sim 0.6$, $J \approx 800\text{K}$, consistent with Raman and neutron measurements. We note that possibly the situation might be different in La$_{2-x}$Sr$_x$CuO$_4$, where the optical gap and the NMR gap appear to have the same energy scale [8], suggesting that either $J \gtrsim \Delta_p$ in this material or that the triplet pair state is not a bound state in this material.

In underdoped YBCO, at $T_c$ there is no anomaly in either the QP relaxation [1] or the NMR $K_s$ [5, 6]. Similarly the SPNS intensity at 34 meV shows no anomaly at $T_c$ itself, but only a gradual drop with increasing $T$. From this we can deduce that there is no change in the DOS at $T_c$ in the underdoped state, and that all changes of the DOS are gradual with $T$. This is equivalent to there being no condensation energy associated with the superconducting transition itself which is consistent with the Bose condensation of local pairs scenario, where at $T_c$ macroscopic phase coherence is established with no change in pairing amplitude. A 3D superconducting state forms when phase coherence percolates through the entire sample resulting in a transition to a coherent macroscopic state at $T_c$. In contrast, in optimally doped and overdoped YBCO as the carrier density increases, the pairs begin to overlap, collective effects become important and a transition to a more conventional BCS-like scenario takes place [1]. QP relaxation, NMR $K_s$, SPNS and tunneling data all show an abrupt anomaly at $T_c$ signifying that pairing and phase coherence occur at the same (or nearly the same) temperature.

The most important feature of the QP relaxation data not available from other spectroscopies is the simultaneous unambiguous observation of 2 gaps in optimally doped and overdoped samples, one $T$-independent $\Delta_p$ and the other $T$-dependent $\Delta_{BCS}$, with a BCS-like $T$ dependence.

REFERENCES