Quasiparticle Dynamics from the Bose–Einstein Condensation to the BCS-Like Regions of the Phase Diagram in YBa$_2$Cu$_3$O$_{7-\delta}$

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In the underdoped region of the phase diagram, time-resolved optical experiments on quasiparticle recombination dynamics show that the state is characterized by a temperature independent energy gap $E_g$ which exists at all temperatures and has a magnitude which appears inversely proportional to doping. Close to optimum doping another, collective gap $\Delta$ becomes visible simultaneously, which has a BCS-like temperature dependence and closes at $T_c$.

1. INTRODUCTION

The study of nonequilibrium quasiparticle dynamics using ultrafast time-domain optical spectroscopy introduces the possibility of distinguishing between different types of excitations in a superconductor on the basis of their relaxation dynamics. This enables qualitatively new information to be obtained about the low-energy structure of these materials. The technique is complementary to frequency domain spectroscopies like infrared reflectivity, tunneling, photoemission and Raman. In high-temperature superconducting cuprates, there has been a great deal of controversy regarding the low-energy excitation spectrum. The results of infrared spectroscopy, Raman and angle-resolved photoemission (ARPES) have been often very controversial and have shown that the gap structure is not easily interpreted unambiguously. Since ultrafast time-resolved photoinduced absorption (TRPA) spectroscopy measures the excited state absorption due to photoexcited quasiparticle (QP) initial states, the temperature and doping dependence of the recombination dynamics gives direct information about the gap, its anisotropy and its $T$-dependence in addition to direct information on the QP recombination kinetics and excitation lifetimes.

When an electron and a hole are first photoexcited by a laser pulse, they immediately start to cool by phonon emission. Once they have lost sufficient kinetic energy and approach the Fermi level (within 10–40 fs) [1], it becomes reasonable to discuss them in terms of dressed particles (polarons or quasiparticles) sensitive to the low-energy electronic fine structure. It is easy to show that the final relaxation step, namely the recombination of these quasiparticles into pairs, is qualitatively different, depending on whether the pairing mechanism is local, e.g., bipolaronic, or collective, as in the BCS case. In the former case, the energy gap $E_g$ between single polarons and bipolarons is determined by the microscopic bipolaron binding energy and is expected to be more or less independent of temperature, its existence becoming apparent as $k_B T \lesssim E_g$. On the other hand, a collective gap which is a result of many-body interactions displays mean-field like closure at the critical temperature $T_c$, which in turn gives a qualitatively different $T$-dependence of the QP recombination dynamics. In this paper, we analyze the time-resolved data on underdoped YBa$_2$Cu$_3$O$_{7-\delta}$ in terms of a polaron–bipolaron band structure and compare this with the results one obtains near optimum doping, where a BCS-like superconducting gap structure is clearly

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apparent. The experimental details and a theoretical model calculation of the QP dynamics are given in Ref. 1.

2. EXPERIMENTS

The experimental results are summarized in Figs. 1 and 2. In Fig. 1 we have plotted the amplitude of the time-resolved absorption (negative transmission) as a function of temperature for underdoped samples (Fig. 1a) and near-optimally doped samples (Fig. 1b) scaled to \( T^* \) and \( T_c \), respectively, \( T^* \) being temperature where the signal drops to 5% of the low-\( T \) value. We also show the theoretical s-wave gap fits to the data for the case of a \( T \)-independent gap \( E_g \) (Fig. 1a) and a temperature-dependent gap \( \Delta \) (Fig. 1b). Distinctly different behavior is observed in the underdoped state and the optimally doped state. The \( d \)-wave prediction for both 2-D and 3-D DOS is also plotted and cannot be reconciled with the data (Fig. 1c). (A discussion of the symmetry is given in Ref. 2.)

A particularly important observation is the appearance of a divergence in the QP relaxation time \( \tau_R \) at \( T_c \) which is observed in the time-resolved absorption near optimum doping [3]. Theory predicts that \( \tau_R \propto 1/\Delta \), so the experimental observation of a divergence in \( \tau_R^* \) is a clear and unambiguous sign of a gap which closes at \( T_c \). The temperature where the divergence appears is plotted in the phase diagram in Fig. 2. In the same plot we also show the pseudogap temperature \( T^* \) as well as \( T_c \), for many different samples with \( 0.48 < \delta < 0.08 \).

3. DISCUSSION

There are a number of distinct features in the data in the underdoped state of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \)
(δ > 0.15). The first is a photoinduced absorption amplitude whose temperature dependence clearly shows asymptotic behavior extending to high temperatures and no visible anomalies at \( T_c \) (Fig. 1a). The data are shown to fit very well a model with a temperature-independent gap. The second important feature of the underdoped state is the absence of any divergence or anomaly of the recombination time \( \tau_R \) at \( T_c \) (or \( T^* \)). In fact, the recombination time \( \tau_R \) is much the same below and above \( T_c \) [3], again consistent with a temperature independent gap. The present data clearly suggest that in the underdoped state only phase coherence is established at \( T_c \), and no changes are observed in the DOS at this temperature in accordance with the Bose–Einstein condensation (BEC) scenario. The argument for the existence of preformed pairs in the underdoped state is fairly unambiguous: It is an experimental fact that there are pairs (bosons) below \( T_c \). From our measurements of \( \tau \) and \( |AT/T| \) there is no observable change in the DOS near \( E_F \) at \( T_c \) or below \( T_c \). Whatever causes the pairing, it is charges that carry the supercurrent, and thus charge carriers must be paired at some point above \( T_c \). To obey the sum rule, pairing of charge must involve a change in the DOS. It follows that since they do not pair at \( T_c \) or below, they must form pairs above \( T_c \), and it is reasonable to assume that \( T^* = T_{\text{pairing}} \), which is the point where we do observe a change in the DOS.

Near optimum doping (δ ~ 0.1), the data are distinctly different. The TRPA amplitude drops rapidly near \( T_c \), and the relaxation time shows a clear divergence anomaly at \( T_c \) [1]. Both of these features can only be reconciled if the gap is assumed to close at \( T_c \) such that \( \Delta(T) \rightarrow 0 \) as \( T \rightarrow T_c \) as shown by the theoretical fit to the data.

In the TRPA experiments we are dealing exclusively with interband optical excitations of electrons from occupied states near \( E_F \) to states well above the plasma frequency, and it is therefore implicit that the redistribution of the DOS which we are observing with temperature is in the charge channel and not the spin channel, the latter being forbidden for optical dipole transitions. The possibility of only a spin gap opening at \( T^* \) can thus be effectively excluded. Since in the TRPA data there is no background to be subtracted, a pseudogap “onset” \( T^* \) can in principle be determined much more accurately than in specific heat or NMR measurements for example. However it should be emphasized that the temperature dependence of the TRPA signal is asymptotic at high temperatures in the underdoped state, so it is quite arbitrary where to assign the “pseudogap temperature” which will unavoidably lead to discrepancies between different experimental techniques. A more appropriate quantity to be discussed is the energy gap \( E_g \) obtained by fitting the temperature dependence over the whole temperature range [1].

In the Bose–Einstein condensation scenario of the underdoped state [4], a polaron band exists at an energy \( E_g \) above the bipolaronic ground state. The splitting is essentially the polaron binding energy \( E_B \), which is predicted to be temperature independent and inversely proportional to the carrier density because of increasing screening. Both of these predictions appear to be consistent with the presented data. The scenario requires that the only carriers relevant to superconductivity are the doped holes which form local bosons (bipolarons). If one assumes that polarons are on adjacent lattice sites, local pairs are fairly easily formed, which can be shown by considering the Coulomb repulsion between two carriers a distance \( r \) from each other. The Coulomb energy \( V = e^2/4\pi\varepsilon_0\varepsilon_r \) with \( r - 30 \) Å is \( V = 0.4 \) eV/e. For a more extreme case of holes on adjacent lattice sites, \( V = 3.6 \) eV/e. The dielectric constant \( \varepsilon_r \) at the typical relevant frequencies (\( E_F \sim 0.05–0.2 \) eV) measured with infrared reflectivity [5] or ellipsometry [6] ranges from 100 to 200 and similar values have been experimentally obtained also at lower frequencies [7,8]. Thus the Coulomb repulsion for carriers on adjacent lattice sites is only \( V = 0.002–0.04 \) eV, which can be clearly overcome by a bipolaron or spin-mediated binding energy with characteristic energy scales of \( \hbar\omega_p \sim 0.1 \) eV and \( J \sim 0.1 \) eV, respectively. Counting only doped holes, the number of carriers in the CuO planes is 0.1–0.2/Cu, so the number of pairs in a coherence volume within the CuO plane of (1.8 nm)³ is of the order of 1–2 or less.

A key question from the theoretical point of view remains whether one should only consider the doped holes \( x \) as relevant for the pairing interaction, or the AFM background spins should also be included, starting the count from a half-filled band in such a way that the total number of particles is \( 1 + x \) [9]. The dominant energy relaxation channel in the QP recombination processes discussed here is determined by the ratio of specific heats of the different subsystems which carry off the excess energy (e.g., phonon \( C_p \) or spin \( C_s \), etc.). At temperatures comparable to \( T^* \) or \( T_c \), \( C_p > C_s \), so the phonon subsystem is the dominant relaxation channel, and the AF spin system can be safely ignored. In other words, the recombination (pairing) of two QPs takes place with (primarily) an accompanying emission of phonons.
For describing the pairing interaction, it should then be sufficient to consider only the doped holes and the arguments for the existence of a BEC can be applied.

The conclusion from the ultrafast transient absorption measurements is that the underdoped state can be described within the BEC scenario of performed pairs. Near optimum doping however, the situation becomes more complicated by the presence of a distinct collective gap with a BCS-like $T$-dependence. The simultaneous presence of two gaps can be understood if we assume that the state is inhomogeneous. In the case of stripes, the high-density areas may act to increase the DOS, leading to apparent van Hove bands and also increasing $T_c$. An alternative (but not incompatible) view is that near optimum doping, the state is characterized by ever larger phase-coherent one-dimensional bipolaron trains (or possibly two-dimensional clusters [10]) whose dimensions are comparable to the stripes inferred from other measurements (see other contributions in this volume). The comparable magnitude of the collective gap and the $T$-independent gap near optimum doping, which we deduce from our experiments [1,2], means that since momentum scattering is very fast compared to the QP recombination time, the regions should appear indistinguishable in the ARPES spectra and only one Fermi surface (FS) is seen, albeit with a gap which will become apparent at different temperatures for different $k$-points [11].

REFERENCES