Separating pairing from quantum phase coherence dynamics above the superconducting transition by femtosecond spectroscopy

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In classical superconductors an energy gap and phase coherence appear simultaneously with pairing at the transition to the superconducting state. In high-temperature superconductors, the possibility that pairing and phase coherence are distinct and independent processes has led to intense experimental search of their separate manifestations. Using femtosecond spectroscopy methods we now show that it is possible to clearly separate fluctuation dynamics of the superconducting pairing amplitude from the phase relaxation above the critical transition temperature. Empirically establishing a close correspondence between the superfluid density measured by THz spectroscopy and superconducting optical pump-probe response over a wide region of temperature, we find that in differently doped Bi2Sr2CaCu2O81dcrystals the pairing gap amplitude monotonically extends well beyond Tc, while the phase coherence shows a pronounced power-law divergence as T → Tc, thus showing that phase coherence and gap formation are distinct processes which occur on different timescales.

A nomalous normal state behavior above the critical temperature appears to be a hallmark of unconventional superconductivity and is present in many different classes of materials. A pseudogap (PG) state has been suggested to be associated with a wide range of possible phenomena preceding the onset of macroscopic phase coherence at the superconducting (SC) critical transition temperature at Tc: pre-formed pairs1–9, a spin-gap10, the formation of a Bose metal11, a Fermi or Bose glass, or a state composed of “dirty bosons”12–14, and more recently a charge-density-wave state15,16.

In addition to the PG response below the temperature designated as T*, the response attributed to “superconducting fluctuations” above Tc has been observed in a number of experiments17–27. The temperature region Tc < T < Tonset where such fluctuations are observable is significantly wider than in conventional superconductors, but smaller than T*. The open and obvious question is whether the pseudogap, or the superconducting fluctuations can be attributed to pairing.

The problem in separating the response due to superconducting fluctuations from the PG is that so far, inevitably, one has had to make extrapolations, or assumptions about the response functions underlying temperature dependences and line shapes in transport17,18,28–30, magnetic susceptibility26,31, specific heat20,21 or photoemission (ARPES)22, which may at best introduce inaccuracies in the temperature scales, and at worst lead to erroneous conclusions. Alternatively one can suppress superconductivity by high magnetic fields up to 60 T32, although there exists a risk of inducing new states by such a high field33. Thus, so far it has not been possible to satisfactorily characterize superconducting fluctuations and discriminate between fluctuations of the amplitude δψ (related to the pairing gap) and phase δφ of the complex order parameter Ψ = ψ e iφ.

In pump-probe experiments three relaxation components shown in Fig. 1 a) are typically observed44: 1) the quasiparticle (QP) recombination in the SC state, 2) pseudogap state response below T* and 3) energy relaxation of hot electrons. The QP dynamics has been shown to be described very well by the Rothwarf-Taylor (R-T) model45,56, and the response related to the presence of non-equilibrium QPs is thus unambiguous. Importantly, the presence of the QP response is directly related to the presence of a pairing gap for QC excitations.

Pump-probe experiments have already shown the coexistence of the pseudogap excitations with superconductivity below Tc over the entire range of phase diagram37–41. However, little attention has been paid to the region of superconducting fluctuation. In this paper we present measurements by a 3-pulse technique which allows us to...
single out the response of superconducting gap fluctuations. The approach is based on selective destruction of the superconducting state by a femtosecond laser pulse\(^4\) which allows us to discriminate pseudogap excitations from superconducting fluctuation seen in transient reflectivity signals, thus avoiding the necessity of making extrapolations or assumptions in separating the different contributions. We then compare these data with a.c. conductivity (THz) measurements from\(^3\) and establish proportionality of the amplitude of superconducting component in pump-probe experiment to the bare phase stiffness \(\rho_0\), measured by THz experiments. This is directly proportional to the bare pair density \(n_{23,41}\), which in turn coincides with the superfluid density when the latter is measured on a timescale on which changes of the order parameter due to either depairing or movement of the vortices can be neglected. Comparison of the critical behavior of the amplitude and phase correlation times above \(T_c\) leads us to the conclusion that two quantities arise from different microscopic processes.

We perform measurements on under- (UD), near optimally- (OP) and over- (OD) doped \(Bi_2Sr_2CaCu_2O_{8+\delta}\) (Bi2212) with \(T_c\) of 81, 85 and 80 K respectively. In the discussion we focus on the underdoped sample, and discuss comparisons with the optimally and overdoped samples, where applicable.

**Results**

**Measurements of pairing amplitude above \(T_c\).** To separate the SC component from the PG component we use a 3 pulse technique described in Refs. 44–47, and schematically represented in Fig. 1f).

A pulse train of 800 nm 50 fs pulses produced by a 250 kHz regenerative amplifier is divided into three beams with variable delays. First a relatively strong “destruction” (D) pulse, with fluence just above the superconducting state photodestruction threshold, \(F_{th}^{SC} = 13 \mu J/cm^2\)\(^1\), destroys the superconducting condensate\(^6\). The ensuing recovery of the signal is measured by means of the 2 pulse Pump-probe (P-pr) response at a variable delay \(\tau_{DP}\) between D and P pulses. There is no effect of the D pulse on the PG response within the noise level of \(\sigma_{DPR} < 0.4 \times 10^{-5}\) indicated by measurements at 120 K shown in Fig. 1e). A typical result of the 3 pulse experiment is presented in Fig. 1g). In the absence of the D pulse the signal consists of a positive SC and a negative PG component. After the arrival of the D pulse we see a disappearance of the SC part and only the PG component is present (dark-red line in Fig. 1c)). With increasing \(\tau_{DP}\) the superconducting response gradually re-emerges (blue line on Fig. 1c)). As most of the condensate in the probe volume is “destroyed” by the D pulse we can extract the superconducting component by subtracting the signal remanent after the destruction (measured 200 fs after the D pulse) from the signal obtained in the absence of the D pulse. Such an extracted superconducting component is plotted in Fig. 1b). The temperature dependence of the amplitude of the superconducting component \(A_{SC}\) is shown in Fig. 2b).
explained by an incomplete destruction of fluctuating superconducting state in the 3 pulse experiment and errors in the PG subtraction.

In Fig. 2a) we see that the QP relaxation time \( \tau_Q^{Pulse} \) obtained by fitting an exponential function to the data Fig. 1b) decreases rather gradually with increasing \( T \) above \( T_c \) and nearly coincide with the QP relaxation time obtained by the pseudogap subtraction procedure \( \tau_Q^{Subtraction} \). The recovery time \( \tau_{Rec} \) obtained from exponential fits to the recovery of the SC response above \( T_c \) (Fig. 1h) shows a similar \( T \)-dependence. The experiments thus show that the recovery of the SC and the QP relaxation show very similar dynamics above \( T_c \).

**Comparison of optical and a.c. conductivity measurements.** We now compare these data with THz measurements of the order parameter correlation time and bare phase stiffness. The agreement between \( \rho_0 \) and \( \Delta SC \) shown in Fig. 2b) is seen to be remarkably good over the entire range of measurements \( 0.8 T_c < T < 1.3 T_c \). This agreement is important because, taking into account \( \Psi \), it validates the approximation that the pump-induced changes in the reflectivity or dielectric constant \( \epsilon \) for small \( \Psi \) are related to the order parameter \( \Psi \) as \( \delta R \sim \delta \epsilon \sim |\Psi|^2 \).

In contrast to \( \Delta SC \) and \( \rho_0 \), remarkable differences are seen in the temperature dependences of the characteristic lifetimes shown in Fig. 2a) obtained by optical techniques and THz conductivity measurements. The phase correlation time \( \tau_{THz} \) determined from the THz conductivity dies out very rapidly with increasing temperature, while the \( T \)-dependence of the amplitude relaxation \( \tau_{Pulse} \) or \( \tau_{Subtraction} \) is much more gradual.

Measurements on an optimally doped sample (Fig. 3c, d)) show qualitatively the same results with \( T_{onset} \sim 102 \text{ K} \), which is \( 17 \text{ K} \) above \( T_c \), and slightly faster decrease of both amplitude and QP relaxation time with temperature. For the overdoped sample, \( F_{opt} \) becomes comparable to \( F_{SC} \), so the superconducting component cannot be significantly suppressed without affecting the pseudogap. Nevertheless the superconducting component is clearly observable in the 2-pulse response up to \( T_{onset} \sim 93 \text{ K} \), i.e. 13 K above \( T_c \). Comparison of 2-pulse data for different doping levels is shown in Fig. 3 e–g), showing qualitatively similar behavior of the SC amplitude above \( T_c \).

**Discussion**

The co-existence and distinct dynamics of the PG and SC excitations above \( T_c \) highlights the highly unconventional nature of these states. A possible explanation for the coexistence of the SC and PG excitations is that the SC and PG quasiparticles which are giving rise to the observed processes are associated with relaxation at different regions on the Fermi surface. Recent Raman and cellular dynamical mean-

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**Figure 3 | Doping dependence.** Comparison of the QP recombination time \( \tau_Q^{Pulse} \) (a) and (c) and the amplitude of the SC signal (b) and (d) measured by the three pulse technique for under-(UD) and optimally(OP) doped samples, respectively. (e)–(g) \( T \)-dependence of 2-pulse response for under-(UD), optimally(OP) and overdoped(OD) samples. Blue dashed and cyan solid lines marks \( T_{onset} \) and \( T_c \), respectively. \( T_{onset} \) shows gradual decrease with doping. The values of the color bars indicate \( \Delta R/R \times 10^{-4} \).
While percolation dynamics is associated with the time dynamics of Josephson tunneling between fluctuating pairs or superconducting patches. The percolation timescale $\tilde{\tau}_p$ is given by the Josephson energy $E_J = I_0 \phi_0 / 2 \pi$, where $I_0$ is the critical current and $\phi_0$ is the magnetic flux quantum. In cuprates, $\tilde{\tau}_p = \hbar / E_J \approx 300 $ fs, which is compatible with the dynamics of phase shown in Fig. 2a).

A picture highlighted by Fig. 2 thus emerges in these materials where the relaxation of the phase $\theta$ is faster than relaxation of the amplitude $\psi$ of the complex order parameter $\Psi = \psi e^{i \theta}$. The dynamics of $\psi$ and $\theta$ being governed by microscopically different processes. It is worth remarking here that the opposite situation is found in charge density wave systems, where the phase relaxation is slow compared to the amplitude relaxation, and the dynamics can be described by TDGL equations for the amplitude $\psi$, neglecting phase relaxation $\theta$.

**Methods**

**Samples.** The samples used in this work were under-, near optimally- and over-doped Bi2212 with $T_c$s of 81, 85 and 80 K respectively. Samples were grown by the traveling solvent floating zone method. Critical temperatures were obtained from susceptibility measurements (e.g. inset in Fig. 1a) for the underdoped sample.

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