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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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals the pairing gap amplitude monotonically extends well beyond T_c, while the phase coherence shows a pronounced power-law divergence as $T \rightarrow T_c$, thus showing that phase coherence and gap formation are distinct processes which occur on different timescales.

Anomalous 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 T_c: pre-formed pairs[1–9], a spin-gap[10], the formation of a Bose metal[11], a Fermi or Bose glass, or a state composed of "dirty bosons"[12–14], and more recently a charge-density-wave state[15,16].

In addition to the PG response below the temperature designated as T^*, the response attributed to "superconducting fluctuations" above T_c has been observed in a number of experiments[17–27]. The temperature region $T_c < T < T_{onset}$ 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 transport[17,18,28–30], magnetic susceptibility[26,31], specific heat[20,21] 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 T[32], although there exists a risk of inducing new states by such a high field[33]. Thus, so far it has not been possible to satisfactorily characterize superconducting fluctuations and discriminate between fluctuations of the amplitude $\delta\psi$ (related to the pairing gap) and phase $\delta\theta$ of the complex order parameter $\Psi = \psi e^{i\theta}$.

In pump-probe experiments three relaxation components shown in Fig. 1 a) are typically observed[34]: 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) model[35,36], 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 QP excitations.

Pump-probe experiments have already shown the coexistence of the pseudogap excitations with superconductivity below T_c over the entire range of phase diagram[37–41]. 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

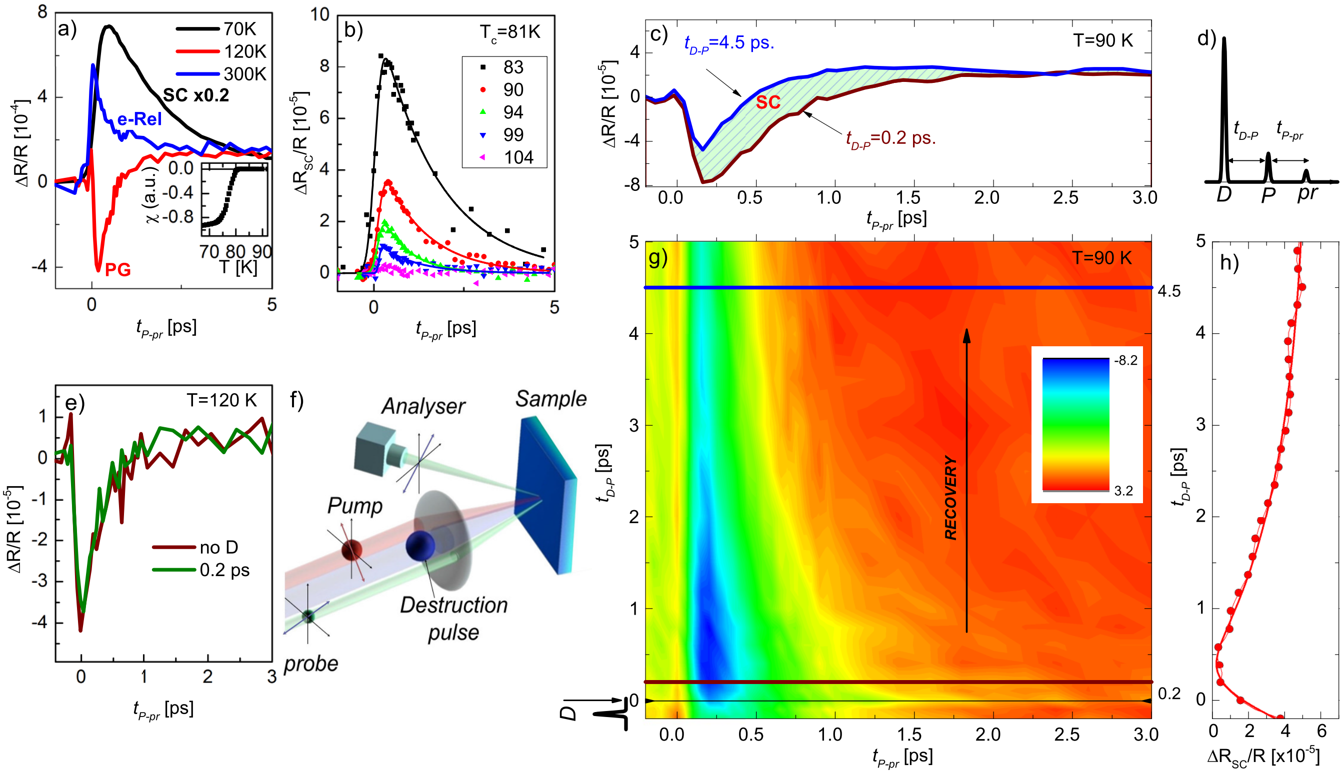

Figure 1 | Description of the 3 pulse method and pump-probe signals. Underdoped sample data: (a) Transient reflectivity signals observed at different temperatures in Bi-2212 - SC signal, the PG signal and energy relaxation of hot carriers (Inset: susceptibility curve shows $T_c = 81$ K for the underdoped sample). (b) The QP recombination at different T above T_c. (c) Two typical 3-pulse pump-probe traces at t_{D-P} 0.2 and 4.5 ps show signal with suppressed and recovered superconducting component respectively. (d) Pulse sequence and delays notation in the 3-pulse experiment. (e) The pump-probe response at 120 K in the absence of the D-pulse and 0.2 ps after the D pulse arrival. (The noise level is larger in comparison to c) due to a shorter signal accumulation time.) (f) A schematic representation of the 3-pulse experiment. Colors are schematic. Destruction (D) pulse destroys the superconductivity, Pump-probe sequence probes the recovery of the quasiparticle response. (g) A typical result of three pulse experiments at 90K shows suppression of the SC component after the D pulse arrival and gradual recovery with t_{D-P}. Readings along the blue and dark-red line are shown in Fig. 1c). The color represents amplitude of the reflectivity change. The values of the color bars indicate $\Delta R/R \times 10^{-5}$. (h) Recovery of the superconducting component amplitude with t_{D-P}.

single out the response of superconducting gap fluctuations. The approach is based on selective destruction of the superconducting state by a femtosecond laser pulse[42] 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[43] and establish proportionality of the amplitude of superconducting component in pump-probe experiment to the bare phase stiffness ρ_0, measured by THz experiments. This is directly proportional to the bare pair density n_s[23,43], 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 de-pairing 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_cs 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, $F_{th}^{SC} = 13\ \mu J/cm^2$ [48], destroys the superconducting condensate[49]. The ensuing recovery of the signal is measured by means of the 2 pulse Pump-probe (P-pr) response at a variable delay t_{D-P} between D and P pulses. There is no effect of the D pulse on the PG response within the noise level of $\sigma_{\Delta R/R} \lesssim 0.4 \times 10^{-5}$ 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 t_{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 signal is detectable up to T_{onset} = 104 K, which is 0.28 T_c above T_c but much lower than $T^* \approx 2.5\ T_c$. In Fig. 1h) we show the recovery of the amplitude of the superconducting component A_{sc} as a function of the time delay t_{D-P} after the D pulse for 90 K. The temperature dependence of the amplitude of the SC component measured by 3-pulse technique A_{sc}^{3pulse} is shown in Fig. 2b).

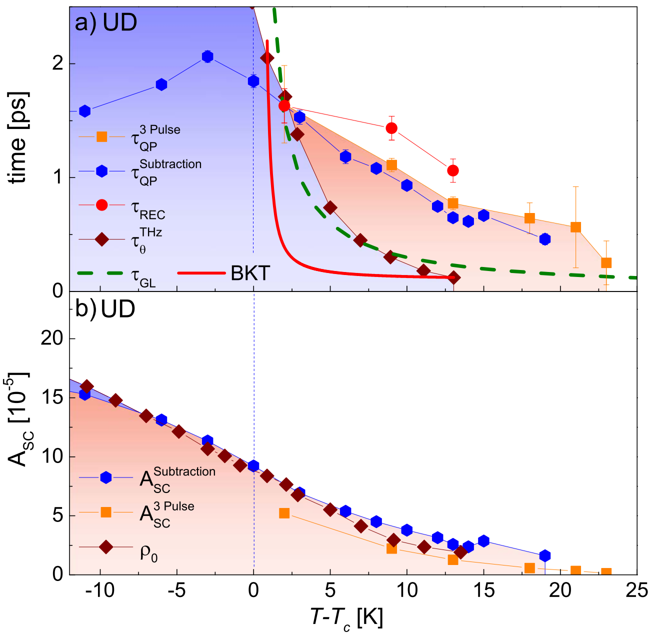

Figure 2 | Comparison of pairing amplitude and phase coherence dynamics. (a) The recovery time of the optical superconducting signal (τ_{rec}), the QP recombination time τ_{QP}^{3Pulse} measured by the three pulse technique and the QP recombination time $\tau_{QP}^{Subtraction}$ from two pulse Pump-probe measurements obtained by subtraction of the PG signal. A fit to the data using a BKT model (eq. 3 of ref. 19) is shown by the solid red line. The dashed line shows the fluctuation lifetime τ_{GL} given by the TDGL theory. The phase correlation time τ_θ^{THz} obtained from the THz conductivity measurements[43] is also shown for comparison. (b) The amplitude of the SC signal measured by the three pulse technique (A_{SC}^{3Pulse}) and the two pulse measurements with the PG signal subtracted ($A_{SC}^{Subtraction}$). The bare phase stiffness ρ_0[43] (normalized at T_c) shows remarkable agreement with the optical response.

For comparison, standard pump-probe measurements need to separate the SC relaxation from the PG relaxation by subtraction of the high temperature response extrapolated into the superconducting region. This approach suffers from the same uncertainties as other techniques such as conductivity, heat capacity, diamagnetism and ARPES. The actual T-dependence of the PG response can vary with doping, pump fluence and probe wavelength[48,50]. But, in Fig. 2b) we show that the subtraction procedure - with the use of a model[48] - gives results in agreement with the direct 3 pulse measurements. The remaining discrepancies in the amplitude can be 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 τ_{QP}^{3Pulse} 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 $\tau_{QP}^{Subtraction}$. The recovery time τ_{rec} obtained from exponential fits to the recovery of the SC response above T_c (Fig. 1 h) shows a similar T-dependence. The experiments thus show that the recovery of the SC gap 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[19,43]. The agreement between ρ_0 and A_{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 $n_s \sim |\Psi|^2$, it validates the approximation that the pump-induced changes in the reflectivity or dielectric constant ϵ for small n_s are related to the order parameter Ψ as $\delta R \sim \delta\varepsilon \sim |\Psi|^2$.

In contrast to A_{SC} and ρ_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 τ_θ^{THz} determined from the THz conductivity[19] dies out very rapidly with increasing temperature, while the T-dependence of the amplitude relaxation (τ_{QP}^{3Pulse}, $\tau_{QP}^{Subtraction}$ or τ_{REC}) is much more gradual.

Measurements on an optimally doped sample (Fig. 3c, d))) show qualitatively the same results with $T_{onset} \sim$ 102 K, which is 17 K above T_c, and slightly faster decrease of both amplitude and QP relaxation time with temperature. For the overdoped sample, F_{th}^{PG} becomes comparable to F_{th}^{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 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-

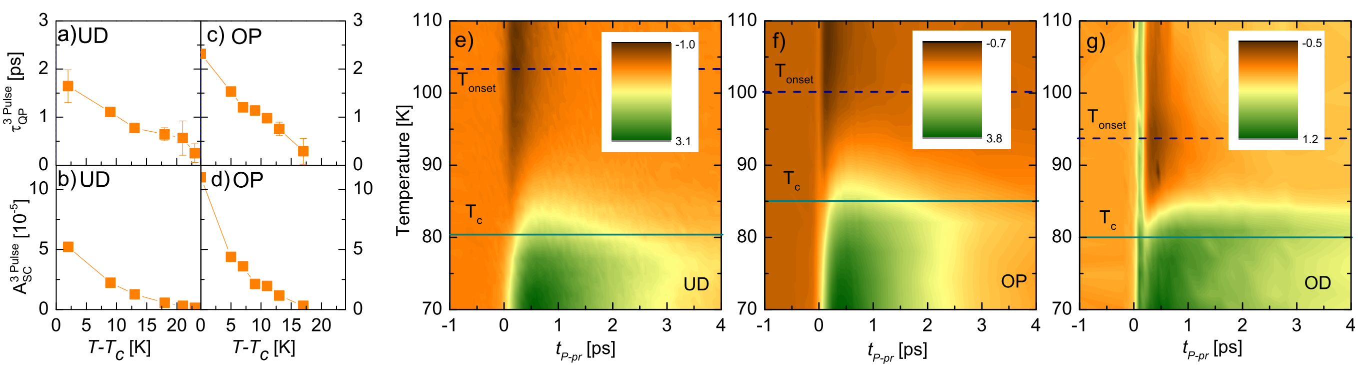

Figure 3 | Doping dependence. Comparison of the QP recombination time τ_{QP}^{3Pulse} (a) and (c) and the amplitude of the SC signal (b) and (d) measured by the three pulse technique for under-(UD) and optimally(OP) doped samples, respectively. (e)–(g) T-dependence of 2-pulse response for under-(UD), 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 $\Delta R/R \times 10^{-4}$.

field studies[51] have suggested that the PG may originate from the states inside the Mott gap, which are characterized by s-wave symmetry and very weak dispersion. Such a localized nature of the PG state excitations is consistent with previous assignments made on the basis of pump-probe experiments[39,48]. In contrast, the superconducting gap fluctuations have predominantly *d*-wave symmetry[52] and are more delocalized. This would explain the simultaneous presence of the SC fluctuations and PG components in pump-probe experiments.

Perhaps the most widely discussed model in the context of distinct pairing and phase coherence phenomena is the Berezinskii-Kosterlitz-Thouless (BKT) transition[53–55] by which decreasing temperature through T_{onset} and approaching T_c causes freely moving thermally activated vortices and anti-vortices to form pairs, thus allowing the condensate to acquire long range phase coherence in an infinite-order phase transition sharply at T_c. The bare pair density is finite to much higher temperatures (up to T_{onset}), where pairing is caused by a different mechanism, and the pseudogap is considered to be an unrelated phenomenon[56–59]. The effect is characterised in terms of a phase stiffness ρ_s, a quantity which characterizes the destruction of phase coherence by thermal fluctuations at a temperature $T_c = T_{BKT} = \pi\rho_s/8$. In cuprates ρ_s is small due to reduced dimensionality and the low carrier density. Within this approach a.c. (THz) conductivity[19,43], heat capacity[20,21], diamagnetism[26,31], ARPES[22] and Nernst effect[23–25,60] measurements were interpreted.

The BSCCO family is the most two-dimensional of the cuprate materials, so here the BKT mechanism for describing the order above T_c would be expected to be most applicable[43]. However, the data in Fig. 2a) show that the drop in τ_θ is not nearly as abrupt as the BKT model predicts. One possibility is that this broadening arises from chemical inhomogeneity of the sample, but the absence of a peak in the heat capacity at T_{onset}[20] also appears to exclude the possibility of a pure BKT transition, and implies amplitude fluctuations might be present between T_c and T_{onset} as well[21]. Thus additional mechanisms beyond BKT may also be present which would broaden the phase coherence transition, such as interlayer phase fluctuations. In this case the observed *T*-dependence would reflect the interlayer decoherence[19,43].

In more traditional approaches using time-dependent Ginzburg-Landau (TDGL) theory[61], thermal fluctuations are small for temperatures higher than $\sim$2 K above T_c[62], but can give an observable contribution to the conductivity in this temperature range[17,18,28–30]. Relaxation within TDGL theory has a "longitudinal" relaxation time τ_Δ, which corresponds to the relaxation of the magnitude of the SC order parameter, and a "transverse" relaxation time τ_θ which corresponds to the relaxation of its phase. They are related to each other in magnitude, but have the same temperature dependence near T_c, namely $\tau_{GL} \sim 1/(T - T_c)$[63]. Perhaps unexpectedly, the temperature dependence of τ_θ^{THz} nearly coincides with the behavior predicted for τ_θ by time-dependent Ginzburg Landau theory for Gaussian fluctuations. Fig. 2a) suggests that phase coherence within this system is established in a narrow, $\sim$5 K temperature interval. However, the distinctly different critical behavior of the pair amplitude dynamics speaks in favor of unconventional models of superconductivity in which pairing and phase coherence occur independently, by different mechanisms. The implication is that the observed pairing amplitude which extends to more than 25 K above T_c reflects the response of an inhomogeneous ensemble of gapped patches which are not mutually phase coherent. The weak temperature dependence of the amplitude cannot be described either by TDGL or BKT models.

Beyond the BKT vortex and TDGL scenario, other phase-locking scenarios, such as Bose-Einstein condensation of bipolarons[2,3,64] and phase-coherence percolation[4,5] may also be consistent with the observed behaviour. In both of these cases pairing and phase coherence are also distinct processes. The former comes from the condensation of pairs at T_c as preformed pair kinetic energy is reduced, while percolation dynamics is associated with the time dynamics of Josephson tunneling between fluctuating pairs or superconducting patches. The percolation timescale τ_J is given by the Josephson energy $E_J = I_c\phi_0/2\pi$, where I_c is the critical current and ϕ_0 is the flux quantum. In cuprates, $\tau_J = \hbar/E_J \simeq 300$ 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 θ is faster than relaxation of the amplitude ψ of the complex order parameter $\Psi = \psi e^{i\theta}$, the dynamics of ψ and θ being governed by microscopically different processes. It is worth remarking here that the opposite situation is found in charge density wave dynamics, where the phase relaxation is slow compared to the amplitude relaxation, and the dynamics can be described by TDGL equations for the amplitude ψ, neglecting phase relaxation θ[45].

Methods

Samples. The samples used in this work were under-, near optimally- and over-doped Bi2212 with T_cs 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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Author contributions

T.K., Y.T. and M.O. has grown the samples and done magnetic characterisation, I.M. did optical measurements. I.M. and P.K. performed data analysis. I.M., Y.T., T.M. and D.M. interpreted the data. I.M and D.M. wrote the manuscript.

Additional information

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