Direct test of pairing fluctuations in the pseudogap phase of underdoped cuprates

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We report on a direct test of pairing fluctuations in the pseudogap regime of underdoped superconducting cuprates using a Josephson junction. In this experiment, the coupling between a rigid superconducting pair field and pairing fluctuations produces a strong specific signature in the current-voltage characteristics. Our results show that fluctuations survive only close to Tc (Tc < 15K) and therefore cannot be responsible for the opening of the pseudogap at high temperature.

The normal state of high-temperature superconductors changes qualitatively as a function of the temperature and the doping level of charge carriers, suggesting different ground states [1]. In particular, in the underdoped regime, a loss of spectral weight in the electronic excitation spectrum, the so-called pseudogap (PG), is observed above Tc, and below a characteristic temperature T* [2]. The origin of this phenomenon is hotly debated: is it related to superconductivity or to a competing hidden order? Many believe that the answer to this question may hold the key to the understanding of high-Tc superconductivity.

As first observed in the spin channel by NMR, the PG has also been observed by most of the one particle probes of electronic excitations [2]. Angle Resolved Photoemission Spectroscopy [3] and Scanning Tunneling Spectroscopy [4] showed that the charge channel is also affected but, more importantly, displayed a characteristic energy of the PG which merges with the superconducting gap when the temperature is lowered below Tc. Moreover, ARPES data also showed that the pseudogap has the same anisotropy as the superconducting gap in momentum space [2]. All these observations reveal a smooth crossover rather than a sharp transition line between the pseudogap regime and the superconducting state, and have led to the superconducting precursor scenario. As opposed to the conventional BCS transition where pairing and condensation occur simultaneously at Tc, in underdoped cuprates fluctuating pairs may form at T*, with no long range coherence, and condense in the superconducting state at Tc [2, 3, 4]. Difficulties in confirming (or invalidating) this scenario arise from the fact that most of the experimental techniques used to investigate the pseudogap are sensitive to the one-particle excitations only, and therefore cannot provide a test of pairing above Tc.

Due to its ability to probe the properties of the superconducting wave function, the Josephson effect is a natural way to address the fluctuation issue.

We have designed an original Josephson-like experiment to directly probe the fluctuating pairs in the normal state by measuring the imaginary part of the pair susceptibility. In a second order phase transition, the susceptibility is given by the linear response of the order parameter to a suitable external field. In the case of the superconducting phase transition, Ferrel and Scalapino showed that the role of the external field could be played by the rigid pair field of a second superconductor below its own Tc [8]. In a Josephson junction in which one side of the junction is the fluctuating superconductor of interest above its Tc, while the other side is a superconductor below its Tc, the coupling between the pairing fluctuations and the well established pair field gives rise to an excess current proportional to the imaginary part of the frequency and wave number-dependent pair susceptibility χ(ω, q).

For a conventional superconductor above its Tc [9],

\[ χ^{-1}(ω, q) = N(0)e(1 - iω/Γ_0 + ξ^2(T)q^2) \],

where \( Γ_0 = (16k_B/\hbar)(T - T_c) \) is the relaxation frequency of the fluctuations, \( ξ(T) \) the superconducting coherence length, \( N(0) \) the quasiparticle density of states at the Fermi level and \( ε = (T - T_c)/T_c \). The frequency \( ω \) is related to the dc bias voltage \( V \) across the junction through the Josephson relation \( ω = 4eV/h \) and the wave number \( q \) is related to a magnetic field applied parallel to the junction. In the absence of magnetic field, the excess current is given by [9]

\[ I_{ex}(V) = A\frac{ω/Γ_0}{ε[1 + (ω/Γ_0)^2]} \],

where \( A \) depends on the coupling through the barrier and on the characteristics of the superconductors. An explicit calculation has been done by Ferrel [5].

Let us emphasize that this dc measurement is sensitive to the pair fluctuations at any frequency (the voltage sets it) and that its temperature dependence is mainly controlled by the distance to Tc through ε and Γ0. In the 1970s, Anderson and Goldman observed gaussian fluctuations just above the transition temperature of conventional superconductors [10]. They measured an excess dc current through tin/tin oxide/lead junctions (for Tc(Sn)<T<Tc(Pb)), in qualitative agreement with relation (2). Janko et al, proposed a similar experiment, where the superconductivity of an optimally doped (OD) cuprate is used to probe the superconducting fluctuations.
in the PG regime of an underdoped (UD) cuprate with a lower Tc [11]. They predicted that an excess current in the junction should persist up to T* if, according to their model, incoherent pairs are responsible for the PG phase. Independently of their respective theoretical framework, all the scenarios involving pairing fluctuations formed at T* should lead to the same conclusion. On the contrary, for a standard BCS-like transition, the contribution of pairing fluctuations should be limited to the vicinity of Tc,4UD.

Josephson-like structures involving two different materials have to be made with thin films. Since high-Tc compounds grow at high temperature where diffusion is fast, underdoping cannot be obtained by changing the oxygen concentration in one layer only. For the coupling to be strong enough, interfaces have to be of very high quality, and therefore an epitaxial structure has to be used: the barrier must have the same crystallographic structure as the superconductors. Only a few materials can fulfill these requirements. We have chosen : - NdBa2Cu3O7 (NdBCO) as optimally doped compound since it grows smoother than the yttrium compound -YBa2Cu3O7 (YBCO(Co)) as underdoped material: Co substitutes Cu in the chains, leading to underdoping with minor disorder in the CuO2 planes [12] -PrBa2Cu4O8 (PBCGO) as the barrier: PrBa2Cu4O7 is a weak insulator, and doping with Ga increases its resistivity. For this experiment we used mainly 30 and 50 nm thick barriers. c-axis trilayer structures YBCO(Co)/PBCGO/NdBCO have been grown on SrTiO3 (100) substrates by pulsed laser deposition and covered by an in-situ gold layer. UV photolithography and ion irradiation have been used to design trilayer junctions whose dimensions range from 40×40μm2 to 5×5 μm2 within a wafer (Fig 1) [13]. The details of this completely new process developed on purpose will be given elsewhere [12]. Its reproducibility provides the basis for the reliability of these experiments.

Figure 1 b) displays a typical resistance versus temperature curve of a junction made with a 30 nm thick barrier. Below Tc,4OD=90K, the high resistance of the barrier (15Ω) and the equipotential gold layer on the top of the mesa guarantee that the current flows homogeneously along the c-axis in the junction, and that the voltage drop measured in this experiment is dominated by the barrier. At T=61K the UD compound becomes superconducting as expected, and as also observed by measuring its magnetic susceptibility by SQUID magnetometry. A Josephson coupling occurs between the two layers and the resistance of the junction drops to zero. Before describing the temperature regime of main interest (61K→90K), we first establish that both dc and ac Josephson effects do occur when both electrodes are in the superconducting state. This is of great importance since the excess current in the fluctuating regime has the same origin as the Josephson one at low temperature. Below the two Tc, current-voltage characteristics display a typical Josephson RSJ-like behaviour with an I2R product of 2 meV at 4.2K (Fig 2). The current-voltage characteristics exhibit clear Shapiro steps at fixed voltage Vn = n2e f/h (n=0, ±1, ±2) when the junction is irradiated with microwaves.
of frequency $f$ (Fig 3 right inset). We have checked that the width of the steps satisfies the linear relation with frequency and that the current height of the steps modulates with the microwave power as expected (Fig 3 left inset) [15]. Such a Josephson effect through PBCO (or PBCGO) barriers has been reported by several groups. This material is known to contain localised states which control the transport; the Josephson effect takes place by direct or resonant tunneling through localised states in the barrier [16, 17, 18]. At finite energy, quasiparticle transport occurs by hopping through these states [18, 19]. In our junction, the background conductance of a 30 nm thick barrier for $T>T_{cOD}$ has a weak dependence with energy, as expected for one or two localized states in the barrier. The conductance follows the characteristic law $G = G_0 + aV^{4/3}$ while junctions with a 50 nm thick barrier exhibit the power law $G = G_0 + aV^{2/3} + bV^{5/2}$ expected for three localized states [18]. Since the transport includes non-elastic hopping, no clear spectroscopic signatures are expected compared to tunnel junction with conventional superconductors. It must be stressed that tunneling is not a requirement for this Josephson-like experiment. Doping PBCO with Ga reduces the number of localized states, and allows to use barriers with a very few of them, but thick enough to avoid microshorts due to extrinsic inhomogeneities.

We now focus on the intermediate regime of temperature ($T_{cUD}<T<T_{cOD}$), the one of main interest here. In order to increase the sensitivity of the experiment, we measure the dynamic conductance $G = dI/dV$ of the junction as a function of the bias voltage. Figure 3 a) displays typical results. At high energy ($>10\text{meV}$) the background exhibits a weak dependence with energy and temperature indicating that the quasiparticle transport is dominated mainly by one or two localized states. In addition to the quasiparticle background, an excess conductance peak emerges from the Josephson current at zero energy when the temperature crosses $T_{cUD}$, and reduces rapidly when the temperature is increased further. It disappears 14K above $T_{cUD}$, well below the characteristic temperature expected for the PG in our compound ($T^*\approx250\text{K}$ estimated from resistivity measurement), as opposed to Janko’s prediction. The peak presents all the characteristics expected from standard gaussian fluctuations above $T_{cUD}$, as calculated and observed in conventional superconducting transitions [10].

Figure 3 b) displays the result of $G_{ex}$ calculation as a function of temperature, in qualitative agreement with the experimental data, both in energy and amplitude, provided the phase fluctuations introduced by voltage noise in actual junctions is properly taken into account. This is simply due to Johnson noise in this rather high temperature experiment. Following reference [20], $G_0$ has to be replaced by $\Gamma = \Gamma_0 + \Gamma_1$ where $\Gamma_1 = 4e^2Rk_B\delta \omega^2/\hbar$ and $R$ is the resistance of the junction. As expected, thermal noise cuts the low energy part of the fluctuation spectrum.

The excess conductance peak is strongly suppressed by microwave radiation; this can be used to get suitable background to extract the excess conductance due to fluctuations. The result is shown in the inset figure 3 b) : the overall shape of the curve is in good agreement with the calculation. The energy scale $h\Gamma/4eV$ (voltage corresponding to $G_{ex}=0$) is close to the expected value from the gaussian model, albeit a little larger. This is an important issue, since $\Gamma$ is related to the characteristic relaxation frequency of the fluctuations. The small discrepancy may originate from the details of the transport (localized states, d-wave symmetry), and the actual choice of $T_{cUD}$, since there is a finite transition width. As microwaves may also affect the quasiparticles transport, this method is not accurate enough to make a full quantitative comparison of the $G_{ex}$ data with the model, and to determine $\Gamma$ for each temperature.

The data do not follow Janko’s predictions. The extra-contribution due to fluctuating pairs is expected to move towards high energy when the temperature is in-
increased (typically 10meV at $T/T_{cUD}=1.1$) : this is not observed here. The broad feature, which extends up to 10 meV is seen in all the samples but can not be attributed to fluctuations since it is already present at low temperature (where no fluctuation are present) and evolves continuously through $T_{cUD}$ up to $T_{cOD}$, where it disappears. Following Devyatov et al. [21], we therefore attribute this feature to Andreev Reflection in the presence of localized states. G(V) is highly symmetric and doesn’t show any evidence of the particle-hole asymmetry suggested by Janko.

The temperature dependence of the pairing peak is a key point. We have already ruled out the barrier conductance itself as a possible explanation for the temperature dependence of the observed signal. We can exclude any contribution of the UD layer itself since the $c$-axis conductance of underdoped YBCO increases with temperature in this temperature range [22], and since no specific energy dependence is expected. The temperature dependence of the OD layer properties has to be addressed. It must be emphasized that in this linear response experiment, the current is directly proportional to $Im\chi$, which is independent of the strength of the external field. The full calculation of the linear coefficient $A$ shows that the excess current is weakly sensitive to the temperature dependence of the fully established superconducting pair field in this temperature range (see Fig 3 a) inset). This contribution has been properly taken into account in the present calculation. Therefore, the decrease of the excess conductance peak cannot be trivially attributed to the reduction of the superconducting pair field in the optimally doped layer. The excess current is observed in the temperature range where gaussian fluctuations are expected in cuprates, i.e. roughly 15K above $T_{cUD}$ given their short coherence length and the weak anisotropy of YBCO. As an example, the Lawrence-Doniach calculation of the paraconductivity above $T_c$ leads to less than 5% of excess conductivity in this range of temperature.

The strength of our experiment is that it relies only on the presence of pairing fluctuations and the Ginsburg-Landau theory. An attempt to indirectly detect pairing above $T_{cUD}$ by high frequency measurements has been made previously in a restricted range of frequency, and within a precise theoretical framework [23]. Fluctuations have been observed up to 95K only, far below $T^*$, in rather good agreement with our result. Our broadband measurement reveals the presence of gaussian fluctuations with no further theoretical assumption.

In summary, by probing the pairs directly, we have found that in the normal phase of an underdoped cuprate, pairing fluctuations survive only in a reduced regime of temperature above $T_{cUD}$, which is consistent with a standard model of gaussian fluctuations. This result is in contradiction with the precursor superconducting scenario as an explanation of the pseudogap. Consequently, focusing on alternative scenarios seems to be a reasonable approach to investigate the underdoped regime of superconducting cuprates.

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