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Superconducting correlations above T_c in the pseudogap state of $Bi_2Sr_2CaCu_2O_{8+\delta}$ cuprates revealed by angular-dependent magnetotunneling

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We present an angular-dependent magnetotunneling technique, which facilitates unambiguous separation of superconducting (supporting circulating screening currents) and nonsuperconducting (not supporting screening currents) contributions to the pseudogap phenomenon in layered $Bi_2Sr_2CaCu_2O_{8+\delta}$ cuprates. Our data indicate persistence of superconducting correlations at temperatures up to $1.5T_c$ in a form of both phase and amplitude fluctuations of the superconducting order parameter. However, despite a profound fluctuations region, only a small fraction of the pseudogap spectrum is caused by superconducting correlations, while the dominating part comes from a competing nonsuperconducting order, which does not support circulating orbital currents.

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How high can the superconducting transition temperature be? This principle question has been discussed for many decades [1]. The debate is fueled by an obscure border between superconducting and normal states in cuprate superconductors, caused by the presence of a normal-state pseudogap [2–8]. The pseudogap bears some similarities with the superconducting gap, which has led to a suggestion that the pseudogap is a precursor of superconductivity [6,9–15]. In this view the onset of Cooper pairing occurs at a temperature T_{c0}, significantly higher than T_c, but the macroscopic phase coherence is suppressed by strong fluctuations of the phase, but not the amplitude, of the order parameter [9]. Such precursor correlations are different from ordinary superconducting fluctuations [16,17] in a sense that the locus of ordinary fluctuations coincides with T_c and both amplitude and phase fluctuations occur simultaneously at $T > T_c$. However, a direct association of the pseudogap with the superconducting gap is confronted by evidence for the existence of nonsuperconducting competing orders in the pseudogap state [18–21]. Spectroscopic techniques (like photoemission or surface tunneling) usually measure the total gap. Separation of superconducting and nonsuperconducting contributions to the total gap is not trivial and requires a more diversified analysis [5,7,19,22]. Direct phase sensitive experiments did not reveal an appreciable Cooper pair concentration well above T_c [23]. Therefore, the question about the extent of superconducting correlation above T_c remains open.

The key character of magnetic field response of superconductors is the appearance of circulating screening currents, which eventually suppress superconductivity at the orbital upper critical field H_{c2}. However, such orbital currents are absent in the two-dimensional (2D) case when the field is applied parallel to the planes. In this case superconductivity is suppressed by Zeeman splitting of spin-singlet Cooper pairs, which occurs at a significantly higher paramagnetic field [24]. The corresponding large in-plane to out-of-plane magnetic anisotropy is generic for quasi-2D superconductors, such as $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) [25]. The strong anisotropy is not inherent to other types of orders (e.g., charge or spin density waves), which do not produce circulating screening currents in magnetic field. This is a central point that we will exploit in this work for discrimination between superconducting and nonsuperconducting contributions to the pseudogap.

In this work we develop an angular-dependent magnetotunneling technique, based on two specific phenomena for layered Bi-2212 cuprates: (i) large magnetic anisotropy caused by in-plane locking of screening currents and (ii) an interlayer tunneling mechanism of *c*-axis transport [26] facilitating intrinsic tunneling spectroscopy [27,28]. This technique enables selective probing of superconducting parts of spectra and allows unambiguous discrimination between superconducting and nonsuperconducting contributions to the pseudogap. We report that superconducting correlations persist well above T_c, but the superconducting contribution to the tunneling density of states is rapidly decaying with increasing temperature and the locus of superconducting amplitude fluctuations is bound to T_c, rather than to some higher onset temperature T_{c0}. The pseudogap amplitude is decaying much slower and well above T_c may exceed the superconducting counterpart by more than three orders of magnitude. Therefore, we conclude that the major contribution to the pseudogap phenomenon is of nonsuperconducting origin.

We study small mesa structures made from two batches of crystals with different T_c. Each sample contains several mesas with attached top electrodes, as sketched in Fig. 1(a). Details of sample fabrication and the experimental setup can be found in the Supplemental Material [29]. We studied about ten samples, all showing similar behavior. Figure 1(b) shows measured zero-bias *c*-axis (R_c, magenta) and in-plane (R_{ab}, blue) resistances versus temperature for a moderately underdoped crystal from batch No. 1. The semiconducting-type $R_c(T)$ dependence at $T > T_c$ is due to the presence of the pseudogap [8,39]. The $T_c \simeq 80$ K is determined by the onset of the *c*-axis critical current, which corresponds approximately to the middle of the in-plane $R_{ab}(T)$ transition [40].

Figure 1(c) shows the current-voltage (I-V) characteristics of one of the mesas at the same sample for zero field (black) and 10 T along the planes (red). Multiple branches at low bias, seen at zero field, are due to one-by-one switching of intrinsic Josephson junctions from supercurrent to quasiparticle branches [28]. The number of branches is equal to the number

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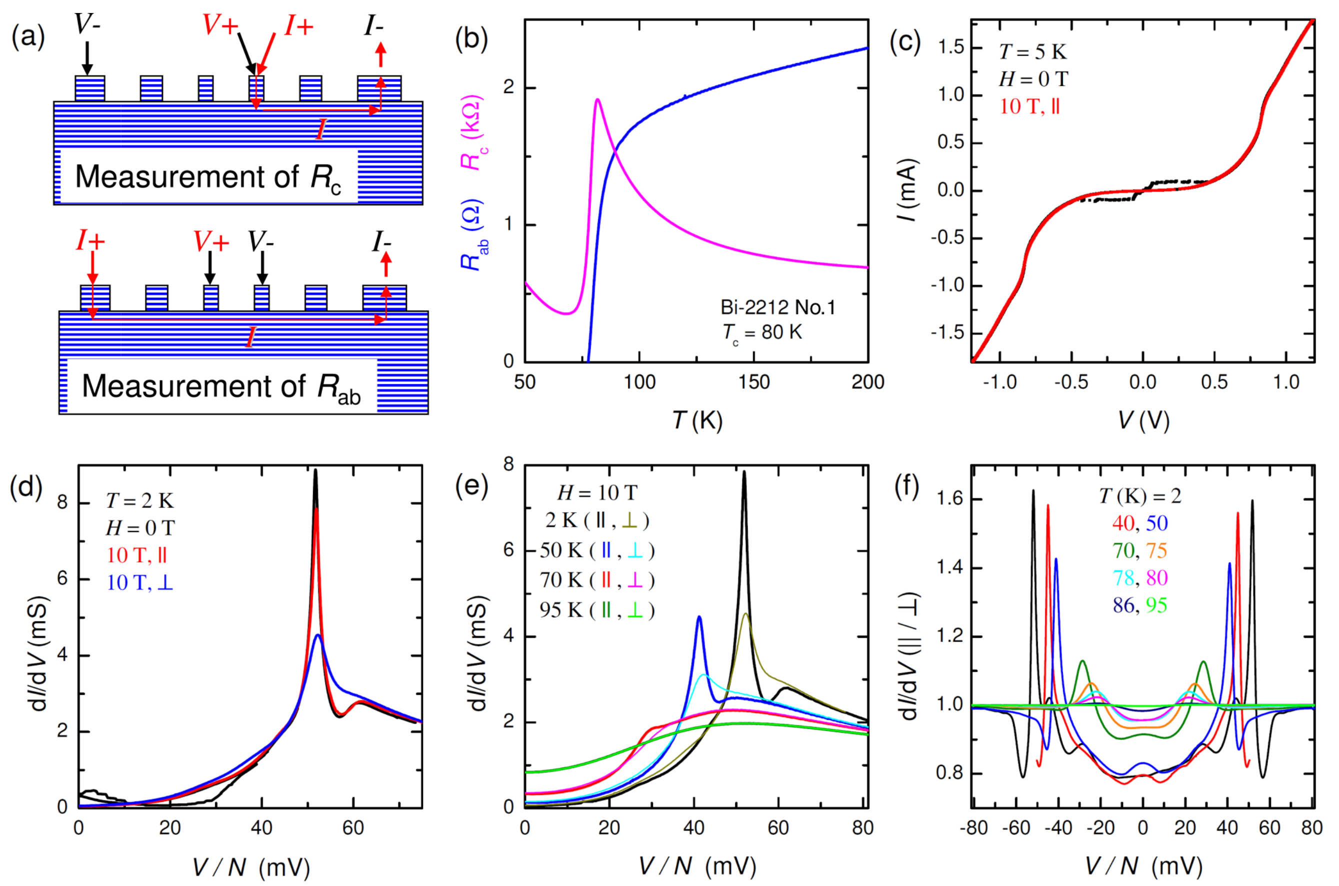

FIG. 1. (a) Sketch of a sample with contact configurations for measurements of c-axis (top) and in-plane (bottom) characteristics. (b) Resistive transitions $R(T)$ of the base crystal in the ab plane (blue) and of a mesa in the c-axis direction (magenta). (c) Current-voltage characteristics of the same mesa in zero field (black) and at 10 T parallel to the planes (red). (d) dI/dV vs voltage per junction at $T = 2$ K in zero field and at 10 T parallel and perpendicular to the planes. (e) dI/dV characteristics in parallel and perpendicular fields at different T. (f) Ratios of dI/dV characteristics at parallel to that at perpendicular field of 10 T. Such angular-dependent magnetotunneling is selective to the anisotropic superconducting contribution and allows discrimination between superconducting and nonsuperconducting parts to the spectra.

of junctions in the mesa N. At higher bias there is a sum-gap kink at $V = 2\Delta N/e$, where Δ is the superconducting gap, followed by an Ohmic tunnel resistance [8,28,40,41]. This is a typical behavior for superconducting tunnel junctions [25], which enables intrinsic tunneling spectroscopy (for a brief overview see the Supplemental Material [29]).

The ultimate resolution of our technique is limited by temperature stability during fairly long measurements [2–3 h per $dI/dV(V)$ curve]. To avoid associated thermal drifts we performed our experiment in the following way. Measurements are done in a cryostat with a superconducting magnet and a rotation stage allowing accurate alignment of the sample. First the temperature of the cryostat was thoroughly stabilized with a mK/h accuracy. After that the first measurement was done. Then the sample was slowly rotated to the orthogonal orientation, which does not disturb the thermal stability, and the second measurement with exactly the same settings was repeated.

Figure 1(d) shows dI/dV vs voltage per junction for the same mesa at zero field (black), and at 10 T parallel (red) and perpendicular (blue) to the ab planes. It is seen that the orientation of the field has a large effect. The parallel field does not affect the curves: the black and red curves practically merge in Figs. 1(c) and 1(d). It only suppresses the Josephson current, which leads to the disappearance of multiple branches in Fig. 1(c). On the other hand, the perpendicular field strongly suppresses the sum-gap peak [25]. Such a strong angular-dependent magnetotunneling effect is a consequence of the quasi-2D superconductivity in Bi-2212. The perpendicular field induces screening currents in CuO_2 layers adding to the overall energy, and thus suppressing superconductivity. To the contrary, the parallel field does not induce screening currents and thus has a negligible effect on superconductivity (provided the field is much smaller than the paramagnetic field).

Figure 1(e) shows $dI/dV(V/N)$ curves at 10 T parallel (thick) and perpendicular (thin lines) to the planes at different T. It is seen that superconducting features are rapidly washed away as $T \to T_c$. Above T_c the sum-gap peak is not seen, but zero-bias conductance remains suppressed due to the presence of the pseudogap, consistent with previous results obtained by different techniques [11,13,42] (see also the Supplemental Material [29]). It is seen that field orientation has a profound effect on the superconducting peak but not on the pseudogap hump [43]. Thus, angular-dependent magnetotunneling enables selective probing of superconducting parts of the spectra.

Figure 1(f) represents ratios of dI/dV characteristics at parallel to that at perpendicular field orientations for $T \lesssim T_c$.

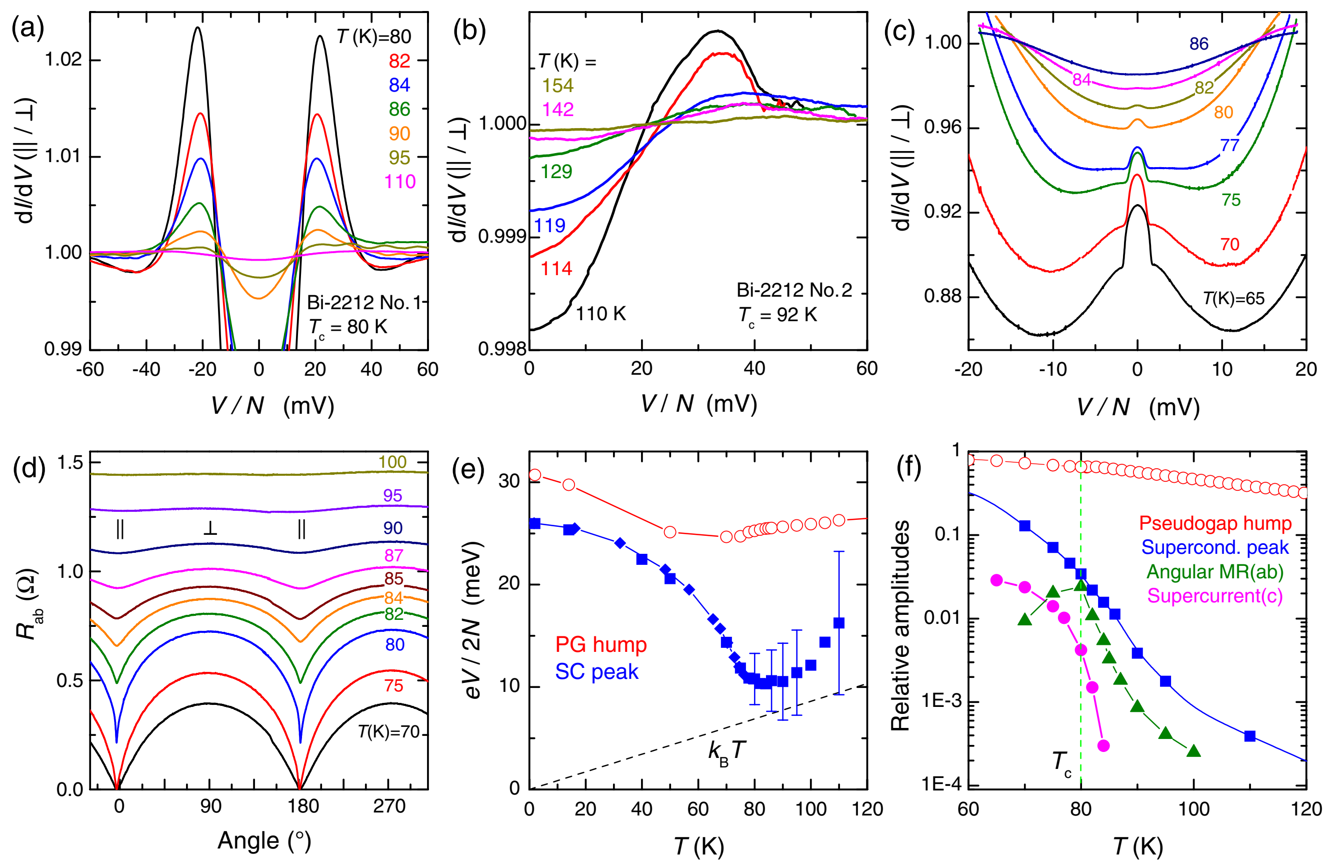

FIG. 2. (a) and (b) Angular-dependent magnetotunneling curves above T_c at $H = 10$ T for mesas at (a) Bi-2212 No. 1 and (b) Bi-2212 No. 2 crystals. (c) Low-bias parts of the curves for the Bi-2212 No. 1 mesa. The zero-bias maximum is due to an interlayer Josephson current. (d) Angular dependencies of in-plane resistances at 10 T for the Bi-2212 No. 1 crystal. The 2D-superconducting cusp at $\Theta = 0$ is distinguishable up to ~ 100 K. (e) Temperature dependencies of the superconducting (SC) peak and the pseudogap (PG) hump voltages for Bi-2212 No. 1. (f) Amplitudes of the PG hump, the SC peak, the 2D cusp in the in-plane angular magnetoresistance, and the Josephson c-axis supercurrent.

It is seen that the ratio $dI/dV(\| / \perp)$ greatly emphasizes all superconducting features, such as the sum-gap peak and the subsequent dip, and even reveals a small subharmonic singularity at $eV/N = \Delta$ [44]. Simultaneously it removes the nonsuperconducting background, as seen from an almost flat curve above T_c, which has to be compared with strongly nonlinear pseudogap characteristics from Fig. 1(e) with a dip at $V = 0$ followed by a hump. The *selective enhancement of superconducting features* in $dI/dV(\| / \perp)$ is caused by the quasi-2D nature of superconductivity in Bi-2212, which leads to the appearance of screening supercurrents only in perpendicular field but not in parallel field. Similarly, the removal of pseudogap characteristics from $dI/dV(\| / \perp)$ ratio indicates the *absence of circulating orbital currents in the pseudogap state*. This is consistent with previously reported weak magnetic field dependence of the pseudogap [25,43,45], governed by Zeeman spin splitting rather than orbital effects [46].

A rapid flattening of $dI/dV(\| / \perp)$ at $T \to T_c$, seen from Fig. 1(f), indicates that the amplitude of the superconducting condensate rapidly decreases with approaching T_c, but it does not vanish completely. In Fig. 2(a) we show data for the same mesa at $T \geqslant T_c$. It is seen that the superconducting peak survives up to several tens of degrees above T_c. Figure 2(b) shows a similar data set for a slightly underdoped mesa with higher $T_c \simeq 92$ K made on a crystal from batch No. 2. It is seen that the sum-gap peak (and a corresponding zero-bias dip) are still recognizable 50 K above T_c.

Figure 2(c) represents a detailed view of the low-bias part of $dI/dV(\| / \perp)$ curves near T_c. The central peak at $V = 0$ is a signature of an interlayer superconducting (Josephson) current. It is seen that the Josephson current is rapidly vanishing just a few degrees above T_c, where the sum-gap peak is still clearly visible. This is evidence for profound interlayer phase fluctuations leading to phase diffusion [47], which suppress the Josephson current close to T_c [23], well before vanishing of the amplitude of the superconducting order parameter.

Figure 2(d) shows angular dependencies of the in-plane resistance $R_{ab}(\Theta)$ measured at 10 T for the same Bi-2212 No. 1 sample. Below T_c the magnetoresistance shows a cusplike minimum at a parallel field orientation $\Theta = 0$, which is typical for 2D superconductors [48,49]. This is again caused by orbital currents present in perpendicular fields and absent in parallel fields. The cusp is smeared out at $T > T_c$ [50], but remains distinguishable few tens of degrees above $T_c \simeq 80$ K, clearly indicating persistence of long-range in-plane superconducting correlations.

Figure 2(e) shows voltages of the superconducting peak and the pseudogap hump for Bi-2212 No. 1. The superconducting gap is rapidly (in a BCS mean-field manner) decreasing at $T \to T_c$ [28,40]. However, our analysis reveals that it does not vanish, but persists well above T_c. The peak is smeared out with increasing $T > T_c$ [error bars in Fig. 2(e) represent the width of the peak]. The dashed line corresponds to $eV/2N = k_B T$. It indicates that the peak voltage at $T > T_c$ is essentially determined by $k_B T$ (increases with T). Therefore, the voltage of so much smeared peak at $T > T_c$, cannot be straightforwardly associated with the gap value. The residual peak simply indicates persistence of superconducting correlations, which modify the overall tunneling density of states, in general agreement with theoretical predictions from Ref. [17]. As for the pseudogap hump, it shows little T dependence [28,40] and remains visible up to much higher $T^* \sim eV_{Hump}/2Nk_B$ [8,41].

Figure 2(f) summarizes our results. It shows T evolution of amplitudes of superconducting and pseudogap features for the Bi-2212 No. 1 sample. Squares represent the sum-gap peak, $dI/dV_{\parallel/\perp}(V_{Peak})$-$dI/dV_{\parallel/\perp}(V=0)$ [see Fig. 2(a)]; solid circles the zero-bias Josephson (c-axis supercurrent) peak [see Fig. 2(c)]; triangles the strength of the 2D cusp in the ab-plane magnetoresistance $dR_{ab}/d\Theta$ ($\Theta = +0$) [see Fig. 2(d)]; and open circles represent the amplitude of the pseudogap hump $[dI/dV(V_{Hump})$-$dI/dV(V=0)]/dI/dV(V_{Hump})$. All of them decay monotonously with increasing T, but with different paces. The c-axis supercurrent is sensitive to the fragile Josephson phase coherence. Therefore, it is destroyed first by interlayer phase fluctuations, just a few K over T_c. The in-plane paraconductivity [16] depends both on the amplitude of the superconducting order parameter and on the more robust in-plane phase coherence. Therefore, the cusp in $R_{ab}(\Theta)$ survives up to a significantly higher $T \sim 20$ K above T_c. Finally, the superconducting peak in the quasiparticle density of states depends solely on the amplitude, but not the phase, of the order parameter. Therefore, it is distinguishable up to even higher $T \sim 1.5T_c$. On the other hand, the pseudogap hump shows little change in this T range. At 120 K it exceeds the amplitude of the superconducting peak by three orders of magnitude, implying that the major part of the pseudogap is not related to superconductivity. This is consistent with a disparity between superconducting and the pseudogap characteristics observed in a very low-T_c Bi-2201 cuprate [45].

To conclude, we demonstrated an angular-dependent magnetotunneling technique which facilitates unambiguous separation of superconducting and nonsuperconducting contributions to tunneling spectra of Bi-2212 cuprates. Our data indicated persistence of superconducting correlations several tens of degrees above T_c, qualitatively consistent with previous reports obtained by other techniques [10,13–15]. This can be viewed either as a BCS-like state with strong fluctuations caused by low dimensionality [16,17], or as evidence of precursor pairing above T_c, which undergoes Bose-Einstein condensation (BEC) [51–54] at T_c. The BCS-BEC crossover, however, has no distinct border [51] and the difference is rather quantitative. In the weak-coupling BCS case Cooper pairing and BEC occurs simultaneously at T_c and only rapidly decaying fluctuations remain above T_c [16]. In a strongly coupled BEC scenario the condensate amplitude also decays with increasing T, but remains large at T_c [52]. Observation of a rapid BCS-like decay of $\Delta(T \to T_c)$ [see Fig. 2(e)], suggests that cuprates are far from the unitary BEC limit and resemble more the BCS case [40], albeit with profound superconducting fluctuations. Importantly, we do not see any additional temperature, other than T_c, for the onset of pairing. Superconducting fluctuations have their locus at T_c and rapidly decay with increasing $T - T_c$ [see Fig. 2 (f)]. Although we cannot quantify the value of the superconducting order parameter above T_c, we can definitely conclude that only a small fraction of the c-axis pseudogap is caused by superconducting correlations, while the dominating part of the electronic density of states at the chemical potential is suppressed by a competing nonsuperconducting order, which does not support circulating orbital currents and, therefore, does not exhibit angular-dependent magnetotunneling effect. Most likely the pseudogap state is a combination of various types of competing charge and spin density wave or orbital orders, which were reported recently in various cuprates and by various techniques [18–21].

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