Interlayer tunneling spectroscopy of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$: a look from inside on the doping phase diagram of high $T_c$ superconductors.

V.M. Krasnov

Department of Microelectronics and Nanoscience, Chalmers University of Technology, S-41296 Göteborg, Sweden

(October 30, 2018)

A systematic, doping dependent interlayer tunneling spectroscopy of Bi2212 high $T_c$ superconductor is presented. An improved resolution made it possible to simultaneously trace the superconducting gap (SG) and the normal state pseudo-gap (PG) in a close vicinity of $T_c$ and to analyze closing of the PG at $T^*$. The obtained doping phase diagram exhibits a critical doping point for appearance of the PG and a characteristic crossing of the SG and the PG close to the optimal doping. This points towards coexistence of two different and competing order parameters in Bi2212. Experimental data indicate that the SG can form a combined (large) gap with the PG at $T < T_c$ and that the interlayer tunneling becomes progressively incoherent with decreasing doping.

PACS numbers: 74.25.-q, 74.50.+r, 74.72.Hs, 74.80.Dm

Observation of an energy gap in the electronic density of states (DOS) had a decisive role in understanding of low $T_c$ superconductivity [1]. However, fifteen years after discovery of high $T_c$ superconductors (HTSC), there is still no consensus about HTSC energy gap. Several experiments revealed different energy scales in HTSC [2–7]. One of those, a normal state pseudo-gap (PG), persists at $T > T_c$ [5, 6]. The origin of the PG is an intriguing open question, which is crucial for understanding HTSC. Currently, the scientific community is divided, believing either in superconducting or nonsuperconducting origins of the PG. The resolution can be provided by a doping phase diagram, both because Oxygen-doping is the most critical HTSC parameter (HTSC can be altered from a metal to an antiferromagnetic insulator by decreasing O-content) and because distinctly different diagrams are expected for different scenarios [6]. In a superconducting scenario, the PG represents the pairing energy, which can be finite at $T > T_c$ in a strong coupling case. The smaller gap represents the energy required for maintenance of a long range coherence [5] at $T < T_c$. Those two energies should merge in the overdoped (OD) region, as the weak coupling limit is approached. If, on the contrary, the PG appears abruptly at some critical doping point $p_c$ and the PG crosses the superconducting gap (SG) at the phase diagram, it would correspond to a non-superconducting PG [6], which develops in the underdoped (UD) region at the expense of the SG.

The present state of confusion requires further studies using advanced experimental techniques. One of those is an interlayer tunneling spectroscopy, which is unique in its ability to measure properties inside HTSC single crystals. This method is specific to strongly anisotropic HTSC, like Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212), in which mobile charge carriers are localized in double CuO$_2$ layers, while the transverse (c-axis) transport is due to interlayer tunneling [3]. Interlayer tunneling has become a powerful tool for studying both electron [3–7] and phonon [8] DOS of HTSC. It has several important advantages compared to surface tunneling techniques: (i) it probes bulk properties and is insensitive to surface deterioration or surface states [3]; (ii) the current direction is well defined; (iii) the tunnel barrier is atomically perfect and has no extrinsic scattering centers; (iv) mesa structures are mechanically stable and can sustain high bias in a wide range of temperatures ($T$) and magnetic fields ($H$).

Here I present a systematic O-doping dependent interlayer tunneling study of Bi2212. The spectroscopic resolution was improved by decreasing in-plane mesa sizes, thus avoiding stacking faults and self-heating in the mesas [9]. This way it was possible to trace the SG and the PG at $T \sim T_c$ and analyze ”closing” of the PG at a characteristic temperature $T^*$. The obtained doping phase diagram exhibits a critical doping point for appearance of the PG and a characteristic crossing of the SG and the PG close to the optimal doping. This points towards coexistence of two different, competing order parameters in HTSC. Experimental data indicate that the SG can either form a combined gap with the PG or remain uncombined at $T < T_c$ and that the interlayer tunneling is predominantly coherent in OD samples, but becomes progressively incoherent with decreasing doping.

Small mesa structures, with areas $A = 10 – 30 \mu m^2$, containing $N = 5 – 12$ intrinsic junctions, were made on top of Bi2212 single crystals by a microfabrication technique [8]. The fabrication was highly reproducible: all mesas on the same crystal exhibited similar behavior, independent of $A$ and $N$. UD crystals were prepared by annealing in vacuum at 600°C.

Fig. 1 shows current-voltage characteristics (IVC’s) per junction at 4.2K for different doping. A characteristic knee in IVC’s is clearly seen, followed by a normal resistance branch $R_N$. The knee is strongly suppressed both by $T$ [4] and $H$ [4], while $R_N$ is almost $T, H$ independent. Such behavior is typical for SIS-type tunnel junctions, in which the knee occurs at a sum-gap voltage $2\Delta_{SG}/e$, where $\Delta_{SG}$ is the maximum SG. Multiple branches at low bias correspond to one-by-one switching.
of junctions from a supercurrent to a quasiparticle (QP) branch. QP branches carry important information: (i) the maximum spacing between QP branches $\delta V_{QP}$ is an additional parameter for estimation of the SG; (ii) the extent of QP branches along the vertical axis in Fig. 1 represents the $I_c R_N$ product per junction, which is a critical parameters of a Josephson junction.

Figs. 2 and 3 show tunneling conductance $\sigma = dI/dV$ curves for slightly OD and UD samples, respectively. Below $T_c$, a sharp peak, corresponding to the knee in IVC’s, is seen. The peak voltage, $V_{peak}$, decreases as $T \rightarrow T_c$. Above $T_c$ the peak disappears, but a distinctly different dip-and-hump structure remains, representing the persisting PG \footnote{3}. $T \rightarrow$ dependencies of the peak (large symbols) and the hump (small symbols + lines) voltages for four samples with different doping are shown in Fig. 4.

For OD samples, $V_{peak}$ can be clearly traced up to $T_c$ and $V_{peak} \rightarrow 0$ at $T_c$, see Figs. 2 a) and 4. Note that $V_{peak}(T < T_c)$ is substantially larger than the hump voltage $V_{hump}(T_c)$ in the OD mesa. At $\sim 150K$, $V_{hump}$ starts to decrease and vanishes at $T^* \approx 200K$ (see Figs. 2 b) and 4). Interestingly, IVC’s are nonlinear even above $T^*$, see Fig. 2 b) and $\sigma(V)$ has an inverted parabola shape, which might indicate the presence of van-Hove singularity close to Fermi level in slightly OD samples \footnote{3}. Details of the PG closing at $T^*$ are important for understanding the origin of the PG. At the first glance $V_{hump}(T \rightarrow T^*)$ resembles a BCS-like dependence, typical for a phase transition due to an onset of charge or spin density waves \footnote{13}. However, a different perspective opens when the parabolic background at $T > T^*$ is subtracted, see Fig. 2 c). In such a plot the PG simply "fills-in" at $T^* \approx \Delta_{PG}$ without a significant change in $V_{hump}$. This may indicate that there is a smooth crossover rather than a true phase transition at $T^*$.

The behaviour of the SG in UD samples at $T \rightarrow T_c$ is one of the most important and yet controversial issues \footnote{3}. For UD samples the peak is much weaker than for OD samples even at low $T$, cf. Figs. 2 a) and 3 a), and it rapidly smears out with increasing $T$. The contrast of the small peak can be increased by subtracting the background PG dip-and-hump at $T > T_c$, as shown in Fig. 3 c). Thus $V_{peak}$ can be located at $T \sim T_c$.

UD samples showed two distinct types of behavior, which I refer to as "small" and "large" gap cases, cf. Figs. 1 c) and d). $dI/dV$ curves for the large gap case are shown in Fig. 3. The behavior of large and small gaps is different: (i) for large gaps, the dip-and-hump is strongly enhanced at the expense of the peak, e.g., in Fig. 3 the PG dip-and-hump is clearly recognizable even at low $T$. For the large (UD84.4) and small (UD85) gap samples, in Fig. 4, ratios of hump to dip conductances $\sigma(V_{hump})/\sigma(0)$ at 100 K are $\sim 5.2$ and 1.8, while $\sigma(V_{peak})R_N$ at 4.2K is $\sim 1.6$ and 10, respectively. (ii) For small gaps $V_{peak} \rightarrow 0$ at $T_c$ and decreases with UD together with $T_c$, while for large gaps $V_{peak}$ remains finite at $T_c$ (even though it drops considerably at $T_c$) see Figs. 3 c) and 4, and both peak and hump voltages increase with underdoping despite the decrease of $T_c$. (iii) Noticeably, apart from gap magnitudes, other parameters, such as $\rho_c, J_c$ and $I_c R_N$ are similar, implying that the tunneling barrier is not affected.

The observed differences can be explained by the following scenarios for formation of small and large gaps,
shown schematically in insets a) and b) of Fig. 4:

The small gap is developed on top of a modest suppression of the DOS at Fermi level, i.e., when there is no true gap at \( T_c \), which might interfere with the opening SG. Therefore, the peak in \( dI/dV \) represents the bare (uncombined) SG, which vanishes at \( T_c \), while the dip-and-hump represent a "normal" background, which is hindered by the growing SG. Such behavior was observed for OD, optimally doped and UD samples with the small gap, see Figs. 2 a) and 4.

On the other hand, in the large gap case the SG is developed on top of a true gap \( \Delta_0 \), see inset b) in Fig. 4. Indeed, from Fig. 3 b) it is seen that the PG dip-and-hump flatten with increasing \( T \) in a state conserving manner, characteristic for a "true" energy gap in DOS, and \( \sigma(V) \) curves intersect in one point, indicating approximately constant value of the PG in the measured \( T \) range. Below \( T_c \) this causes formation of the combined \( (\Delta_0 \text{ and } \Delta_{SG}) \) large gap. In agreement with this assumption: (i) the large gap does not vanish, but approaches \( \Delta_0 \) at \( T_c \), see Figs. 3a) and 4. (ii) The peak completely disappears at \( T_c \) but does not transform into the hump because \( eV_{hump} > \Delta_0 \), see the dashed line in inset b). (iii) The volume of the peak (superfluid density) is small because it builds up from an initially suppressed DOS. (iv) The opening of the SG at \( T < T_c \) shifts all DOS features, including the hump, as shown in inset b)

of Fig. 4. The correlated shift of both the peak and the hump with \( T \) for UD84.4 sample, as shown in Figs. 3 and 4, is a strong argument in favor of the combined scenario of the large gap. Similarly, uncorrelated \( T \)-dependent peak and \( T \)-independent hump in the OD93 sample, see Figs. 2 and 4, suggests that the small gap represents the uncombined SG. Interestingly, if we take \( V_{peak}(4.2K) - V_{peak}(T_c) \) as a measure of the SG part of the combined gap, it will coincide with the small gap for a similar doping, as shown by arrows in Fig. 5 d). A systematic increase of the hump energy with decreasing \( T \) was observed by ARPES would have been consistent with the combined scenario of the large gap if not for the lack of correlated \( T \)-dependence of the coherence peak.

Fig. 5 shows O-doping dependencies of: a) \( T_c \), dashed line represents the empirical expression, used for estimation of \( p \); b) the critical current density, \( J_c \), and the \( I_cR_N \) product per junction; c) the tunneling resistivity at large bias \( \rho_c = R_N \) (ns) \( I_cR_N \) is an important parameter of a Josephson junction. As Bi2212 is likely to be a d-wave superconductor, the \( I_cR_N \) depends both on \( \Delta_{SG} \) and the coherence (in-plane momentum conservation) of \( c \)-axis tunneling (another highly debated issue in HTSC). The \( I_cR_N \) is maximum \( \approx \Delta_{SG}/e \) for coherent, and zero for completely incoherent tunneling. For OD mesas \( I_cR_N \approx 10mV \) is a considerable fraction \( \approx 0.6 \) of \( \Delta_{SG}/e \), indicating predominantly coherent nature of the interlayer tunneling. With underdoping,
the $I_cR_N$ decreases dramatically at a much faster rate than $\Delta_{SG}$. This indicates that the interlayer tunneling becomes progressively incoherent in UD Bi2212.

Fig. 5 d) shows the obtained doping phase diagram of Bi2212. Here I plot $1/2 V_{\text{peak}}(4.2K) \sim \Delta_{SG}/e$, $1/2 V_{\text{hump}}(100K > T_c) \sim \Delta_{PG}/e$ and $\delta V_{QP}(4.2K)$, It is seen that the small gap ($\sim$ SG) shows a similar tendency as $T_c$ and decreases both on OD and UD sides. This is also supported by a correlated behavior of $\delta V_{QP}$. In contrast, the large gap ($\sim$ PG) increases approximately linearly with underdoping, as shown by the solid line. The PG and the SG lines cross at about the optimal doping, $p = 0.16$. On the OD side the PG becomes considerably less than the SG and shows a clear tendency to vanish at the critical doping point, $p_c \approx 0.19$. This speaks in favor of a nonsuperconducting origin of the PG, consistent with earlier observations of different $T$ and $H$ dependencies of the SG and the PG. Within such a scenario, a suppression of superconductivity (decrease of $T_c$, $\Delta_{SG}$, the superfluid density, etc.) in UD HTSC is caused by appearance of the competing order parameter (PG), e.g., due to strengthening of antiferromagnetic correlations and formation of spin density waves. Note that a similar phase diagram, attributed to competition between superconducting and antiferromagnetic orders, was reported for heavy fermion superconductors [2].

At present, the reason for appearance of either small or large gaps in UD samples is unclear. However, it is not due to irreproducibility of fabrication (all mesas on the same crystal show the same behavior) or macroscopic defects (regular QP branches are observed in both cases). Presumably, the ambiguity is connected with a microscopic inhomogeneity of UD crystals [20]. The presence of ambiguity obscures identification of the genuine HTSC behavior in the UD region. However, there is no ambiguity for overdoped and optimally doped samples. Therefore, conclusions that there is a critical doping point in HTSC phase diagram and that the SG and the PG cross rather than merge near the optimal doping are robust.

In summary, O-doping dependence of Bi2212 was studied using high resolution interlayer tunneling spectroscopy. We were able to simultaneously trace the superconducting gap and the $c$-axis pseudo-gap at $T \sim T_c$ and analyze "closing" of the PG at $T^*$. The obtained doping phase diagram exhibits a critical doping point for appearance of the PG and a characteristic crossing of the SG and the PG close to the optimal doping, indicating a competing nature of two coexisting order parameters in HTSC. In UD samples, the SG can either form a combined gap with the PG or remain uncombined at $T < T_c$, but the bare SG vanishes at $T \sim T_c$ for all studied doping levels. Analysis of $I_cR_N$ vs. $\Delta_{SG}$ indicates that the interlayer tunneling is predominantly coherent in OD, but becomes progressively incoherent in UD samples.

[1] I.Giaever, Phys.Rev.Lett. 5, 464 (1960)
[2] G. Deutscher, Nature 397, 410 (1999)
[3] V.M.Krasnov, A.Yurgens, D.Winkler, P.Delsing and T.Claeson, Phys.Rev.Lett. 84, 5860 (2000)
[4] J.C.Campuzano, H.Ding, M.R.Norman, H.M.Fretwell, M.Randeria, A.Kaminski, J.Mesot, T.Takeuchi, T.Sato, T.Yokoya, T.Takahashi, T.Mochiku, K.Kadowaki, P.Guptasarma, D. G.Hinks, Z.Konstantinovic, Z.Z.Li, and H.Raffy, Phys.Rev.Lett. 83, 3709 (1999)
[5] A.V.Puchkov, D.N.Basov, and T.Timusk, J. Phys. Cond. Mat. 8, 10049 (1996)
[6] J.L.Tallon and J.W.Loram, Physica C 349, 53 (2001)
[7] V.M.Krasnov, A.E.Kovalev, A.Yurgens, and D.Winkler, Phys.Rev.Lett. 86, 2657 (2001)
[8] M.Kugler, O.Fischer, Ch.Renner, S.Ono and Y.Ando, Phys.Rev.Lett. 86, 4911 (2001)
[9] R.Kleiner, F.Steinmeyer, G.Kunkel, and P.Müller, Phys.Rev.Lett. 68, 2394 (1992)
[10] V.M.Krasnov, N.Mros, A.Yurgens and D.Winkler, Phys.Rev.B 59, 8463 (1999)
[11] M.Suzuki and T.Watanabe, Phys.Rev.Lett. 85, 4787 (2000)
[12] K.Schlenga, R.Kleiner, G.Hechtfischer, M.Mössle, S.Schmitt, P.Müller, Ch.Helm, Ch.Preis, F.Forsthofer, J.Keller, H.L.Johnson, M.Veith and E.Steinbeiss, Phys.Rev.B 57, 14518 (1998)
[13] A.Lanzara, P.V.Bogdanov, X.J.Zhou, S.A.Kellar, D.L.Feng, E.D.Lu, T.Yoshida, H.Eisaki, A.Fujimori, K.Kishio, J.I.Shimoyama, T.Noda, S.Uchida, Z.Hussain, Z.X.Shen, Nature 412, 510 (2001)
[14] V.M.Krasnov, A.Yurgens, D.Winkler and P.Delsing, J.Appl.Phys. 89, 5578 (2001); V.M. Krasnov, cond-mat/0109079
[15] C.C.Tsuei and J.R.Kirtley, Rev.Mod.Phys. 72, 969 (2000)
[16] P.W.Anderson, Science 279, 1196 (1998)
[17] Y.Tanaka and S.Kashiwaya, Phys.Rev.B 56, 892 (1997).
[18] O.K.Andersen, O.Jepsen, A.I.Liechtenstein, and I.I.Mazin, Phys.Rev.B 49, 4145 (1994)
[19] R.S.Markiewicz, C.Kusko and V.Kidambi, Phys.Rev.B 60, 627 (1999) F.Onufrieva and P.Pfeuty, ibid. 61, 799 (2000); S.Chakravarty, R.B.Laughlin, D.K.Morr, and Ch.Nayak, ibid. 63, 094503 (2001)
[20] T.Cren, D.Roditchev, W.Sacks, J.Klein, J.-B.Moussy, C.Deville-Cavellin, and M.Lagués, Phys.Rev.Lett. 84, 147 (2000)
[21] N.D.Mathur, F.M.Grosche, S.R.Julian, I.R.Walker, D.M.Freye, R.K.W.Haselwimmer, and G.G.Lonzirach, Nature 394, 39 (1998)