Scanning Tunneling Spectroscopy of $Bi_2Sr_2CuO_{6+\delta}$: New Evidence for the Common Origin of the Pseudogap and Superconductivity

M. Kugler and Ø. Fischer
DPMC, Université de Genève, 24, Quai Ernest-Ansermet, 1211 Genève, Switzerland

Ch. Renner
NEC Research Institute, 4 Independence Way, Princeton, New Jersey 08540

S. Ono and Yoichi Ando
Central Research Institute of Electric Power Industry, Komae, Tokyo 201-8511, Japan
(Received 3 January 2001)

Using scanning tunneling spectroscopy, we investigated the temperature dependence of the quasiparticle density of states of overdoped $Bi_2Sr_2CuO_{6+\delta}$ between 275 mK and 82 K. Below $T_c = 10$ K, the spectra show a gap with well-defined coherence peaks at $\pm\Delta_p \simeq 12$ meV, which disappear at T_c. Above T_c, the spectra display a clear pseudogap of the same magnitude, gradually filling up and vanishing at $T^* \simeq 68$ K. The comparison with $Bi_2Sr_2CaCu_2O_{8+\delta}$ demonstrates that the pseudogap and the superconducting gap scale with each other, providing strong evidence that they have a common origin.

DOI: 10.1103/PhysRevLett.86.4911 PACS numbers: 74.25.Jb, 73.40.Gk, 74.72.Hs

Many experiments provided evidence for an unusual behavior in the normal state of high-temperature superconductors (HTS) [1]: Nuclear magnetic resonance, infrared conductivity, neutron scattering, transport characteristics, specific heat, spin susceptibility, thermoelectric power, and Raman spectroscopy all showed indirect signatures which were interpreted as the opening of a gap in the electronic excitation spectrum above the critical temperature T_c, the so-called *pseudogap* [1]. This striking observation initiated an intense and challenging debate about its origin, since the answer to this key question may turn out to be essential for the understanding of high-temperature superconductivity. Two basic trends can be found in literature: Either the pseudogap has its origin in a phenomenon different from superconductivity [2] and which may possibly be in competition with the superconducting state, or it has basically the same origin as superconductivity [3] in which case it might reflect the presence of pairs above T_c.

Powerful tools to investigate these issues are angular resolved photoemission spectroscopy (ARPES) [4] and tunneling spectroscopy [5–10], since they give a direct access to the quasiparticle density of states (DOS). Probing the DOS of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) single crystals as a function of temperature clearly confirmed the presence of a gap above T_c [4–6,8]. Furthermore, ARPES demonstrated that for underdoped samples the pseudogap and the superconducting gap have a similar gap anisotropy in k space [4]. Beyond these observations, scanning tunneling spectroscopy (STS) showed that in Bi2212 the pseudogap is not restricted to the underdoped state, but that it also exists in the optimally and even overdoped case [6–8]. These studies strongly favor the idea that the pseudogap is related to superconductivity and that it might be due to precursor pairing [4–7,11]. Very recently, considerable attention has been directed to superconductor-insulator-superconductor (SIS) tunneling and, in particular, to intrinsic SIS tunneling experiments on multiple junctions in Bi2212 mesas [12]. These studies revealed a double gap structure in the tunneling conductance spectra: a sharp feature which develops on top of a much broader excitation gap. The authors thus claimed to distinguish and simultaneously observe both the superconducting gap and the pseudogap. The superconducting gap showing a strong temperature dependence and the pseudogap existing even below T_c led them further to conclude that their origins are different. The definite answer to the origin of the pseudogap thus still appears ambiguous. Note however, that in these experiments the superconducting gap at $T \ll T_c$ has about the same magnitude as the pseudogap at various doping levels, which is consistent with the above mentioned ARPES and STS studies.

A limiting experimental fact is that these recent results were all obtained on a single compound, namely, Bi2212. In order to draw more general conclusions it is crucial to investigate other HTS compounds where the superconducting parameters, such as T_c or the gap Δ, are radically different. One such compound is $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201). This material has a very low critical temperature of $T_c^{\max} \approx 13$ K [13], which is nearly an order of magnitude lower than for Bi2212 ($T_c^{\max} \approx 92$ K; see [8]). Bi2201 therefore presents an ideal stage to test the relation between the pseudogap and the superconducting gap. Here we present a scanning tunneling spectroscopy study of Bi2201 as a function of temperature.

We studied single crystals having a nonstoichiometric composition of $Bi_{2.1}Sr_{1.9}CuO_{6+\delta}$. The samples were grown by floating zone melting, yielding $T_c^{\mathrm{onset}} = 10$ K ($\Delta T = 4$ K) as measured by dc-SQUID magnetization. The temperature dependence of the Hall coefficient and the room-temperature thermopower show a characteristic behavior of overdoped samples [14] and allowed a consistent determination of the hole concentration per Cu atom, $p = 0.18$. The measurements presented in this Letter were obtained using a newly developed sub-Kelvin scanning tunneling microscope which operates under ultrahigh vacuum in variable temperatures between 275 mK and room temperature [15]. The tunnel junctions were made between *in situ* cleaved Bi2201 (001) surfaces and electrochemically etched iridium tips mounted perpendicular to the sample surface. In this configuration, the differential tunneling conductance dI/dV primarily yields an angular average over the ab plane DOS.

In Fig. 1a we focus on the tunneling conductance measured at $T = 275$ mK, showing typical spectra obtained at different locations on the sample surface. They were reproducibly observed on different crystals and after subsequent *in situ* cleavages [16]. The spectra present well-developed coherence peaks at $\pm\Delta_p \simeq 12$ meV [17]. This is a striking result, since for a T_c of only 10 K the gap magnitude is extremely large. It is by a factor 7 larger than the BCS d-wave prediction $\Delta_{\mathrm{BCS}} = 1.8$ meV [18]. This further leads to a very high ratio $2\Delta_p/k_BT_c \simeq 28$, which is even more remarkable since our sample is overdoped. For comparison, at equivalent doping, Bi2212 yields $2\Delta_p/k_BT_c \approx 10$ [6]. This result shows that in Bi2201 superconductivity is far from being BCS like, even in the overdoped regime. Because of this extreme situation it is crucial to search for signatures of a pseudogap. If the latter is related to the superconducting gap, we expect a correspondingly large pseudogap in a wide temperature interval above T_c. On the contrary, if the origin of the pseudogap is independent of superconductivity, we expect that in our overdoped samples the pseudogap may be absent or, at least, that its magnitude is completely unrelated to the superconducting gap. Harris *et al.* [17] reported an ARPES study which showed leading edge shifts of 7 meV on underdoped $Bi_{2.3}Sr_{1.7}CuO_{6+\delta}$ ($T_c < 4$ K) and of 10 meV on optimally doped $Bi_2Sr_{1.65}La_{0.35}CuO_{6+\delta}$ ($T_c = 29$ K) at temperatures above T_c. They interpreted these shifts as the opening of a pseudogap, although they could not determine at which temperature $T^* > T_c$ the pseudogap closes. Note that these values are smaller than the gap magnitude we obtained in the superconducting state on overdoped Bi2201. This observation further emphasizes the need to determine the relation between the pseudogap and the superconducting gap. As we shall see, the temperature dependence of the tunneling spectra presents a consistent picture and gives a clear answer to this issue.

Compared to Bi2212 it is notoriously more difficult to grow homogeneous Bi2201 single crystals, thus inherently resulting in locally varying tunneling spectra. To access the intrinsic DOS independently of local sample inhomogeneities, we systematically acquired 100 spectra spaced equidistantly along a 20 nm line on the sample surface at each selected temperature. Figure 2a shows the results at three characteristic temperatures. The slope of the background conductance is reproducible and topographic imaging routinely resolved atomic scale features, as shown in Fig. 1b. Hence, the variations of the spectra are not due to poor tunneling junction quality [16]. Consistently with what we observed on Bi2212, the periodic superstructure (Fig. 1b) does not lead to a spatial variation of the spectra. Figure 2b shows the average of these spectra. The peak positions scatter within ± 3 $\mathrm{mV_{rms}}$ around the average position and fine structures of the DOS tend to be smeared out. However, the average spectra clearly delineate the essential features. At $T_1 = 8.8$ K, below T_c, well-defined coherence peaks delimit the superconducting gap. At $T_2 = 24.7$ K far above T_c, a pseudogap is plainly

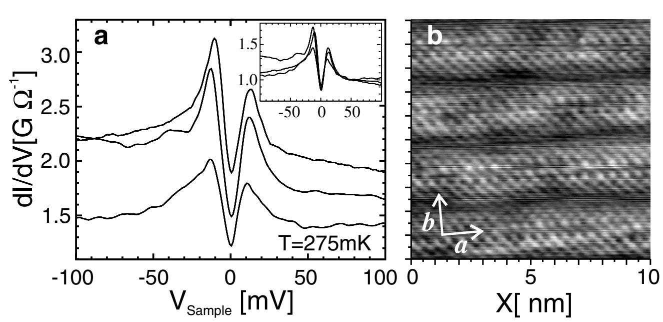

FIG. 1. (a) dI/dV spectra obtained at 275 mK at different locations on *in situ* cleaved Bi2201 ($T_c = 10$ K). In the main panel the spectra are shifted vertically by 0.2 $\mathrm{G\Omega^{-1}}$ for clarity. In the inset they are superposed and normalized. The peaks are at $\pm\Delta_p = 12$ meV. (b) 10×10 $\mathrm{nm^2}$ constant current image ($R_t = 1.5$ $\mathrm{G\Omega}$, raw data) showing atomic resolution and the 26 Å periodic superstructure along the b axis.

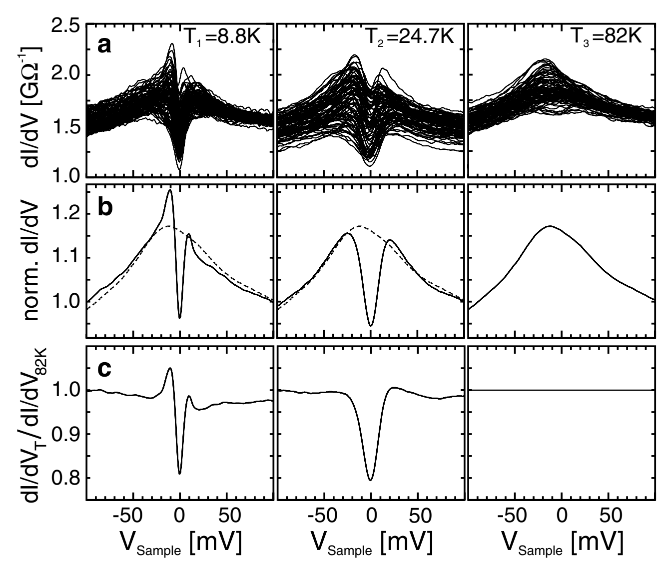

FIG. 2. dI/dV spectra acquired at $T = 8.8$, 24.7, and 82 K. ($R_t = 0.6$ $\mathrm{G\Omega}$). (a) 100 superposed spectra acquired along a 20 nm path. (b) Respective average spectra together with the 82 K data (dashed line) and normalized to the conductance at 100 mV. (c) Same spectra, but normalized to the 82 K data.

visible. Finally, at $T_3 = 82$ K, the gap has filled up and a broad hump, which is slightly offset to negative bias, develops. Assuming that the 82 K spectrum reflects the metallic normal state DOS, we normalized our data to this background conductance to highlight the spectral changes due to the pseudogap and the superconducting state.

To investigate the continuous evolution of the quasiparticle DOS, we acquired tunneling conductance spectra at various temperatures between 275 mK and 82 K as shown in Fig. 3. All spectra up to about 70 K show a gap. At the bulk T_c, the coherence peaks abruptly disappear, similar to Bi2212 [6]. This unambiguously shows that the large gap measured below T_c is the gap of the coherent superconducting state of the bulk and that the conductance peaks are related to the coherent superfluid density [19]. Furthermore, there is no sign indicating that the superconducting gap closes at T_c as predicted by the BCS theory. Instead, the superconducting gap evolves into a clear pseudogap which is roughly constant up to about 30 K ($T \gg T_c$), where the gap structure progressively becomes larger, as also observed in Bi2212 [6]. The pseudogap gradually fills up and finally vanishes at $T^* = 68 \pm 2$ K, which we defined as the temperature where the zero-bias conductance reaches 95% of the normal state conductance given here by the 82 K data.

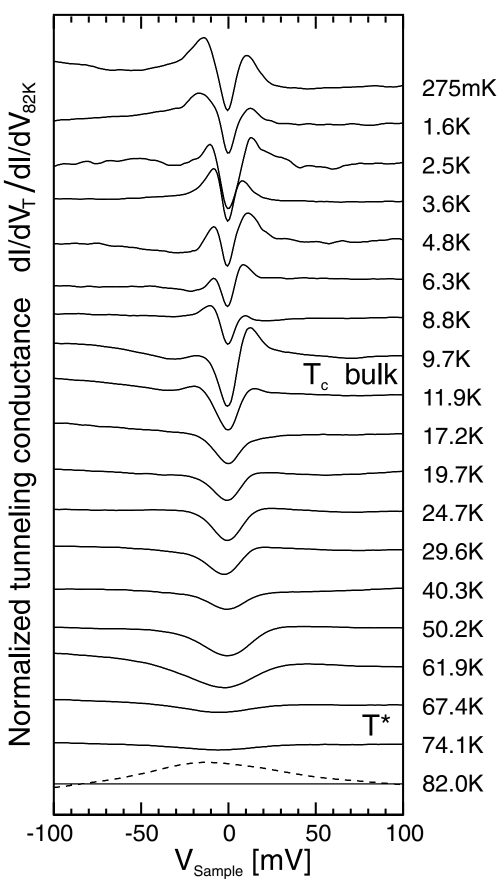

FIG. 3. Temperature dependence of the tunneling conductance of Bi2201. This representative set of spectra was acquired and normalized to the 82 K background (dashed line) as described in Fig. 2 and shifted vertically for clarity.

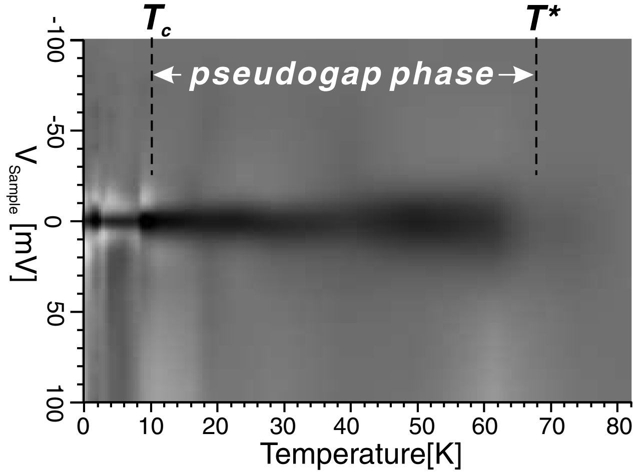

FIG. 4. Interpolated top-view representation of the spectra shown in Fig. 3. The grey scale corresponds to the normalized differential tunneling conductance.

The purpose of the top-view representation (Fig. 4) is to focus on the general trend of the overall temperature behavior, rather than on detail features. The present temperature dependence largely confirms what has been observed previously by STS on Bi2212 [6], with the central difference that Bi2201 has radically different superconducting parameters. The key observation in the temperature dependence is the extremely large pseudogap phase compared to the superconducting one. There is a factor 7 between T^* and T_c, which is precisely the same factor we observed between Δ_p and Δ_{BCS}.

A consequence of these results is that the ratio of Δ_p and T^* is the same as found in other HTS materials. In Fig. 5 we plotted T^*/T_c versus $2\Delta_p/k_BT_c$ for several HTS materials which have been investigated by tunneling spectroscopy. Oda *et al.* [11] and Nakano *et al.* [20]

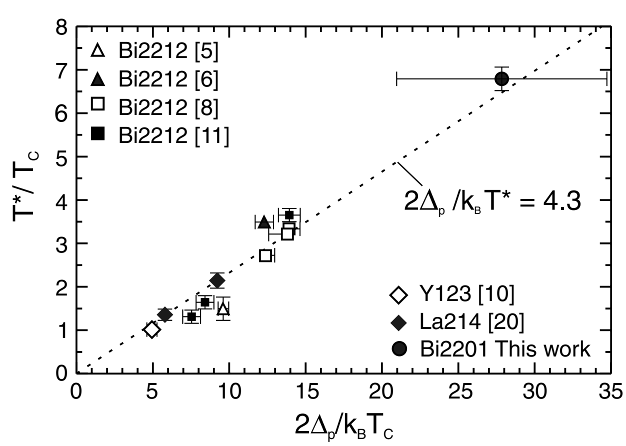

FIG. 5. T^*/T_c versus $2\Delta_p/k_BT_c$ for various cuprates compared to the mean-field relation $2\Delta_p/k_BT^* = 4.3$ [18], where T^* replaces T_c. Error bars are extracted from the given references.

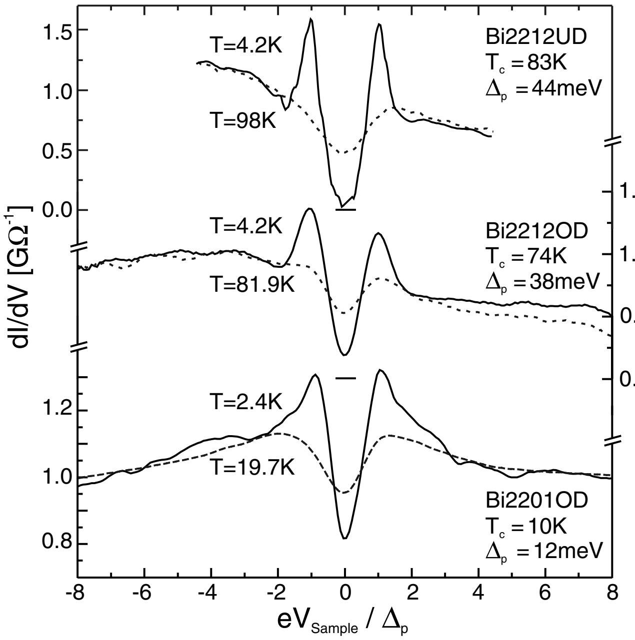

FIG. 6. Comparison between the pseudogap (dashed line, $T > T_c$) and the gap in the superconducting state (solid line, $T < T_c$) for underdoped (UD) and overdoped (OD) Bi2212 [6], and overdoped Bi2201 (present work). All curves are raw data.

highlighted that Bi2212 and La214 fall close to the BCS d-wave relation $2\Delta_p/k_BT^* = 4.3$ [18], where T_c is replaced by T^*. Strikingly, this is also true for Bi2201, although $2\Delta_p/k_BT_c$ is by a factor 2–3 larger than for the other compounds. It thus confirms that it is T^*, rather than T_c, which reflects the mean-field critical temperature of the superconductor. This consistency favors models which evoke precursor pairing as the origin of the pseudogap phase [3]. From this point of view, the fact that the pseudogap temperature T^* is almost 1 order of magnitude larger than T_c reveals that phase fluctuations are very strong in this single CuO_2 layer cuprate, even in the overdoped regime.

The central result of this Letter is related to the scaling between the superconducting gap and the pseudogap. In spite of the low T_c and the overdoped nature of our samples, we find that the ratio between the gap in the superconducting state and the pseudogap is the same as in Bi2212. Figure 6 compares the tunneling spectra above and below T_c for the present study on overdoped Bi2201, as well as for over- and underdoped Bi2212 [6]. To highlight the scaling properties, we normalized the energy scale to the respective Δ_p of each compound, thus demonstrating that the relative size of the pseudogap with respect to the superconducting gap is in all three cases the same. This consistency is remarkable since the energy scale given by Δ_p is a factor 3–4 smaller for Bi2201 compared to Bi2212. The present data thus bring a manifest confirmation that the pseudogap scales with the superconducting gap, giving a robust evidence that the origin of the pseudogap is related to superconductivity.

In conclusion, using scanning tunneling spectroscopy, we measured the temperature dependence of the DOS of overdoped Bi2201 between 275 mK and 82 K. Together with earlier results on Bi2212, we obtain a systematic picture where the pseudogap scales with the gap in the superconducting state, although both compounds show radically different superconducting parameters. Furthermore, the surprisingly large gap value observed in Bi2201 and the correspondingly large T^* add strong evidence that in these materials the pseudogap has the same origin as superconductivity.

[1] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).
[2] R. S. Markiewicz, C. Kusko, and V. Kidambi, Phys. Rev. B **60**, 627 (1999); J. M. Tranquada *et al.,* Nature (London) **375**, 561 (1995); S.-C. Zhang, Science **275**, 1089 (1997).
[3] V. J. Emery and S. A. Kivelson, Nature (London) **374**, 434 (1995); J. R. Engelbrecht, A. Nazarenko, M. Randeria, and E. Dagotto, Phys. Rev. B **57**, 13 406 (1998).
[4] H. Ding *et al.,* Nature (London) **382**, 51 (1996); A. G. Loeser *et al.,* Science **273**, 325 (1996).
[5] H. J. Tao, F. Lu, and E. L. Wolf, Physica (Amsterdam) **282C–287C**, 1507 (1997).
[6] Ch. Renner *et al.,* Phys. Rev. Lett. **80**, 149 (1998).
[7] Ch. Renner *et al.,* Phys. Rev. Lett. **80**, 3606 (1998).
[8] A. Matsuda, S. Sugita, and T. Watanabe, Phys. Rev. B **60**, 1377 (1999).
[9] N. Miyakawa *et al.,* Phys. Rev. Lett. **83**, 1018 (1999).
[10] I. Maggio-Aprile *et al.,* J. Electron Spectrosc. Relat. Phenom. **10**, 147 (2000).
[11] M. Oda *et al.,* Physica (Amsterdam) **281C**, 135 (1997).
[12] V. M. Krasnov *et al.,* Phys. Rev. Lett. **84**, 5860 (2000); M. Suzuki and T. Watanabe, Phys. Rev. Lett. **85**, 4787 (2000).
[13] S. I. Vedeneev *et al.,* Phys. Rev. B **60**, 12 467 (1999); J. I. Gorina *et al.,* Solid State Commun. **108**, 275 (1998).
[14] Yoichi Ando *et al.,* Phys. Rev. B **61**, R14 956 (2000).
[15] M. Kugler *et al.,* Rev. Sci. Instrum. **71**, 1475 (2000).
[16] The data were systematically checked for true vacuum tunneling conditions following Ch. Renner and Ø. Fischer, Phys. Rev. B **51**, 9208 (1995).
[17] J. M. Harris *et al.,* Phys. Rev. Lett. **79**, 143 (1997).
[18] H. Won and K. Maki, Phys. Rev. B **49**, 1397 (1994).
[19] D. L. Feng *et al.,* Science **289**, 277 (2000).
[20] T. Nakano, N. Momono, M. Oda, and M. Ido, J. Phys. Soc. Jpn. **67**, 2622 (1998).