Influence of Disorder on the Local Density of States in High-T_c Superconducting Thin Films

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Using a low temperature scanning tunneling microscope in the spectroscopic mode, we find that the disorder in a $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin film modifies dramatically the quasiparticle local density of states. Small, but well-defined superconducting regions, coexisting with dominating semiconducting areas, show well-pronounced gap structures, similar to those observed previously in high-quality single crystals. Surprisingly, between these two regions, the detailed shape of the quasiparticle spectrum is virtually identical to the pseudogap previously observed at temperatures $T > T_c$, or in the vortex core, at 4.2 K. Thus, the role of the disorder in destroying the superconducting phase is comparable to that of the magnetic field or thermal fluctuations.

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While a comprehensive model for superconductivity in the cuprates is still undecided, the experimental progress is rapidly whittling down the number of key parameters affecting the phenomenon. The temperature, magnetic field, and doping phase diagrams have been largely explored, and with a wide variety of techniques [1]. What is less established is the influence of the disorder. In conventional superconductors, the critical magnetic fields and critical current are strongly affected by disorder. For the cuprate high-T_c superconductors (HTS), the case of strongly underdoped systems is particularly interesting: The material approaches the antiferromagnetic insulating phase, with a decrease of carrier concentration, and the system is prone to the effects of disorder. In the extreme case, one could expect some dramatic modification of the local electronic structure, due to localization or magnetic disorder. Thus, scanning tunneling spectroscopy (STS) may elucidate the superconducting state on such a system.

Contrary to conventional superconductors, in HTSs the phase transition at T_c is not accompanied by the vanishing of the gap in the density of states (DOS) at the Fermi level. Different techniques have shown a local suppression of the DOS, the pseudogap, above T_c [1–6]. Measured by STS, the local DOS (LDOS) in the vortex core also revealed the pseudogap [7]. Understanding the relationship between the pseudogap and the superconducting states would be an important breakthrough.

In this Letter we show that the local quasiparticle spectrum in the presence of disorder is strikingly similar to the LDOS previously attributed to the pseudogap. The spectra change continuously from a superconducting gap to a pseudogap, as the scanning tunneling microscope (STM) tip is moved out of a superconducting region. This is a new independent observation of the low-temperature "normal" state and an experimental verification of the key role of the disorder for destroying the superconductivity on the local scale. Exploring large areas of a $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) thin film, STM/STS at 4.2 K reveals three different types of regions according to the local spectra. In the superconducting regions, the essential features of the quasiparticle DOS are evident, as shown in Fig. 1, and are typical of spectra obtained previously on high-quality single

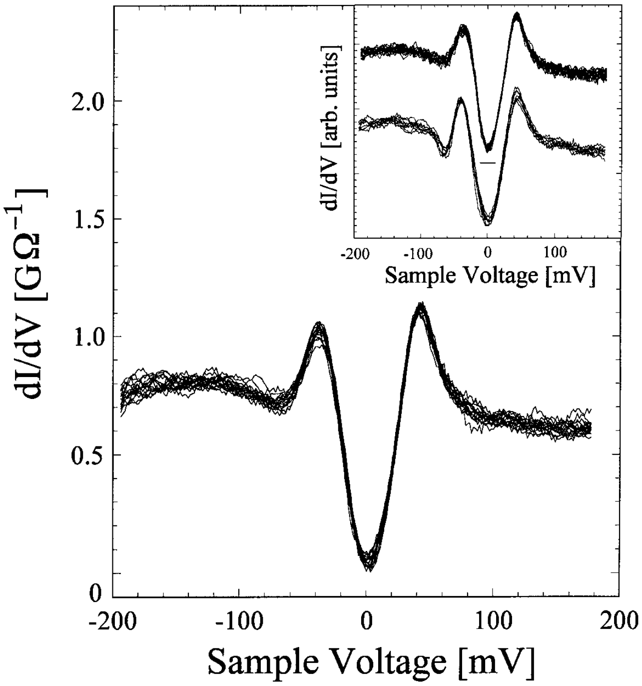

FIG. 1. Tunneling $dI/dV(V)$ spectrum obtained in a superconducting region of a Bi2212 thin film. The curves in the inset are obtained for different tip positions within the same superconducting region, showing a good reproducibility of the data. Note the slight variations in the peak and dip structures, and the small zero bias conductance.

crystals [8–12]. The inset in Fig. 1 shows the high reproducibility of the spectra at different sites of the same superconducting region. The intensity of the dip structures and the peak heights vary somewhat, from case to case. To our knowledge, this is the first reproducible STS data on a Bi2212 superconducting thin film.

In view of the numerous models of cuprate superconductivity, it is not a simple matter to interpret the tunneling spectra [13]. The curves are usually fitted using a mean-field quasiparticle DOS with a $d_{x^2-y^2}$ or anisotropic s symmetry for the gap parameter $\Delta(\theta)$ [11]. Previous STS results on single crystals [8–12] revealed only a small broadening in the best resolved spectra. However, the broadening was much larger in many other experiments. Thus, whenever a large "pair breaking" is found, it may be due to spurious scattering of the tunnel carriers on surface and tip defects. The spectra in Fig. 1 show a surprisingly small broadening, and the question remains to what extent the residual pair breaking is an intrinsic property of HTSs. If it is, a model accounting for an appropriate quasiparticle self-energy must be considered [13,14]. Another remarkable aspect of the spectra of Fig. 1 is the "dip" on the negative bias side of $dI/dV(V)$, observed previously in the best spectra on single crystals. The dip was seen at the energy -2Δ on the occupied states side of the LDOS [9], whereas in [12] two dips at both occupied states and empty states were found. In the disordered thin film, we find the dip to sometimes exist either on the negative side or on both sides, at different locations of the same sample. These signatures, absent in the BCS quasiparticle spectrum, could be an additional consequence of many-body effects [13].

It is normal that in earlier work the disorder was considered as a "bad" parameter, perturbing the experiment. In the HTSs, having a naturally defected structure and a very short coherence length, one had to overcome the effects of the disorder, particularly on the local scale of the STM measurement. This problem is precisely the focus of our study, and as will be shown below, beyond the superconducting zones, the system is characterized by a much higher structural disorder, and a dramatic effect on the quasiparticle spectrum is indeed found.

The Bi2212 superconducting films were grown to a thickness of 300 Å by molecular beam epitaxy on a (100) strontium titanate substrate. The epitaxy sequence was terminated by two nominal Bi-O layers. The chemical composition of a typical film, as measured by Rutherford backscattering spectroscopy, was $Bi_2Sr_{1.98}Ca_{1.38}Cu_{2.28}O_{8+\delta}$. The hole concentration corresponds to a slight underdoping of the samples in the bulk, as controlled by x-ray absorption spectroscopy. X-ray diffraction spectra, in Bragg-Brentano and four circle geometry, confirm the c-axis orientation and the full epitaxy within the ab plane. The Bragg peak positions are consistent with the Bi2212 phase, but high resolution transmission microscopy (HRTEM) reveals a small amount of Bi2223. The Bi2212 crystallites have a typical size of 50 nm as measured by HRTEM. Broad superconducting transitions with T_c onset of 93 K (55 K) and T_c offset of 59 K (10 K) were found from, respectively, dc-resistivity and ac-susceptibility measurements. These results indicate that we have a highly inhomogenous system.

All STS experiments were performed at 4.2 K in low-pressure He^4 exchange gas. The details of our home-built low temperature STM are described elsewhere [15]. The STS data we present were acquired only within large flat regions where the STM images showed a roughness less than 1 nm. The reproducibility of the spectra was controlled by a large number (128 or 256) of successive measurements, taken at a given tip position. To avoid any surface charge effects, tip contamination, or other artifacts, the spectra were controlled to be independent of tip-surface distance, acquisition time, or on the sign of the voltage sweep. High values of tunneling resistance (more than 1 $\mathrm{G}\Omega$) were used to ensure optimal conditions for tunneling and, in particular, to prevent tip crashes. The LDOS mapping was performed by scanning the tip along a given lateral direction while simultaneously sampling the $I(V)$ data. By reversing the scan direction, the data were verified for their correlation to the tip position on the surface. No additional smoothing or filtering of the data was done: We plot spectra that are a numerical differentiation of the direct $I(V)$ measurements.

Exploring different superconducting regions, we find that the peak-to-peak gap value 2Δ varies significantly from one region to another; from roughly $\Delta = 25$ meV to about 55 meV, while within a given region, the gap remains unchanged. Considering the influence of the oxygen content on Δ reported in Ref. [16], the gap values we find would correspond to local optimally doped and underdoped regions. A similar gap disparity exists in single crystals [8–11]. It is likely that some variations of carrier density are the cause and are more significant in thin films.

Much interest has moved away from the elusive question of the pairing mechanism in HTS, focusing on their anomalous electronic properties. Nuclear magnetic resonance [17], inelastic neutron scattering [2], and photoemission [4,5] measurements give a suppression of states at the Fermi level to exist for $T > T_c$. Angle-resolved photoemission spectroscopy [6] shows not only a persistence of the gap above T_c, but a concomitant reduced Fermi surface that returns only to the normal one at a higher temperature T^*. Finally, the pseudogap is seen directly in the tunneling spectra in a quite spectacular fashion. From Renner *et al.* [18], inset (a) in Fig. 2 shows a gap persisting beyond T_c in the underdoped single crystal ($T_c = 83.0$ K). The curve corresponding to $T = 88.9$ K is attributed to the pseudogap spectrum. The pseudogap is also seen at 4.2 K in the vortex core [7], and its form differs very little from the high temperature counterpart,

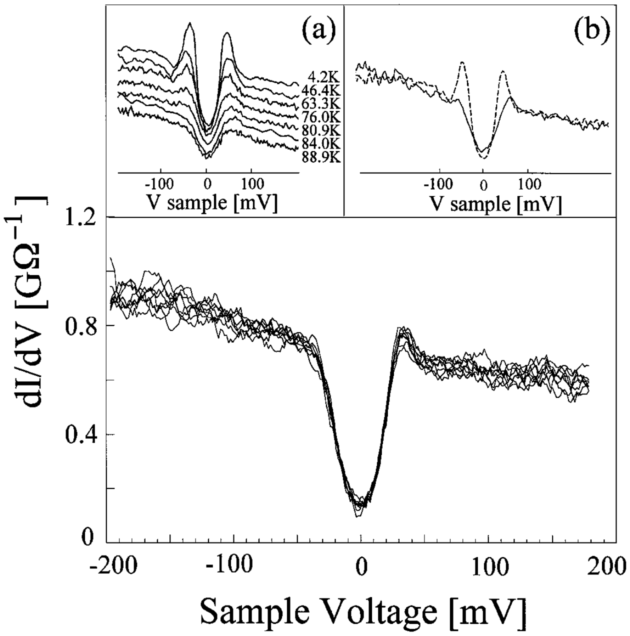

FIG. 2. Typical spectrum just beyond a superconducting region, which we attribute to the pseudogap. The spectrum is surprisingly similar to the data of Renner *et al.* [18] obtained at $T > T_c$, inset (a), and to the pseudogap structure, found at 4.2 K in the vortex core 7, inset (b) (lower curve).

as inset (b) in Fig. 2 indicates (taking into account thermal broadening). These data represent clearly the normal state spectrum, and for a high-quality single crystal. In both cases the pseudogaps were observed in the same region of the sample as the superconducting gaps and thus correspond to the same level of doping. The central part of Fig. 2 shows the typical tunneling spectrum we obtain on the Bi2212 thin film, as the tip is moved immediately beyond the superconducting regions. The particular shape of these spectra is very different from the known Coulomb gap [11] or the case of contaminated junctions [9]. The spectrum is strikingly similar to those obtained by Renner *et al.* in the two very different situations described.

In Fig. 3 we show the evolution of the dI/dV spectra as a function of the tip lateral position. As the tip is scanned out of a superconducting region, the pseudogap abruptly appears in the short distance of only a few nanometers. This fixes an estimate of the scale for the loss of superconducting phase on the order of the coherence length ξ and is thus the same as for the vortex core reported in [7]. We then find a region where only the pseudogap is revealed, as Fig. 3 shows. The scan direction can be reversed, and one finds reproducibly the superconducting gap to appear upon leaving the pseudogap region. Similar transformations are observed in different locations on the same sample and for different samples. They are less noticeable near underdoped superconducting regions. In fact, the superconducting gap is larger there, and the quasiparticle peaks are less pronounced, as recently reported by Miyakawa *et al.* [16]. Thus, the superconducting LDOS may be easily confused with the pseudogap, making the gap to pseudogap transition more difficult to observe.

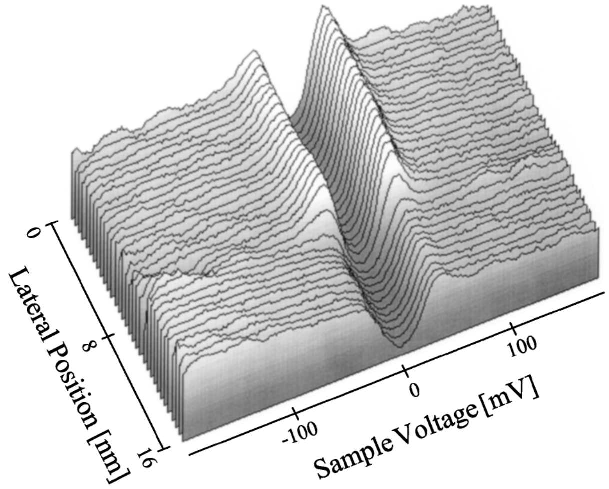

FIG. 3. Three-dimensional view of the $dI/dV(V)$ spectrum variations as a function of the tip position. The spectra change continuously from superconducting DOS to the pseudogap form. The superconducting phase is seen to vanish on the scale of a few nanometers.

When the tip is scanned further beyond the pseudogap region, the spectra gradually change into a characteristic V shape. This is illustrated in Fig. 4 in which the change is seen on the scale of 5–10 nm. The spectra vary from place to place, but their typical width is still larger than 100 meV. We term it "semiconducting" gap, since its form is typical for a semiconductor DOS. Also, it may originate from a strong localization due to disorder or from structural changes. It also might be an

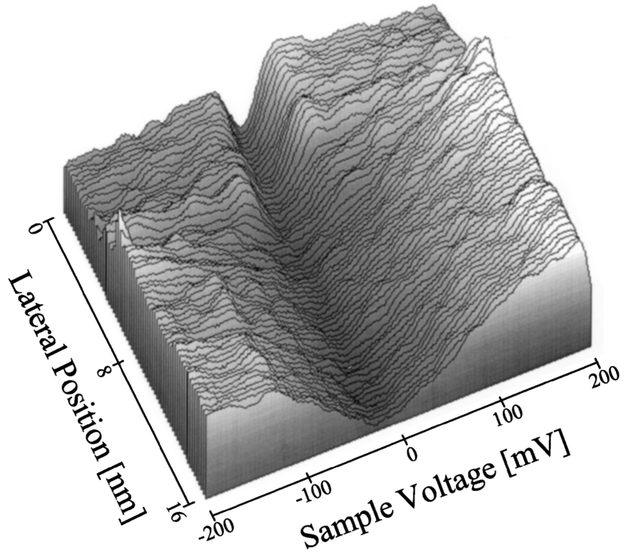

FIG. 4. The $dI/dV(V)$ spectrum variations as the tip is moved away from the pseudogap region. The changes from the pseudogap into a semiconducting form are seen.

"ultimate" pseudogap corresponding to highly underdoped, nonsuperconducting regions, as suggested in [19], according to the commonly accepted phase diagram. In summary, we find superconducting islands, bordered by pseudogap regions, dispersed on a dominating semiconducting sample surface.

We now focus on the spectral variation from the superconducting to the pseudogap form. The gap width is found unchanged within a given superconducting region, as shown in the inset in Fig. 1, and even up to its very border (Fig. 3). We conclude that no significant changes of carrier density are possible there. Furthermore, the pseudogap seen in Figs. 2 and 3 has, to a first approximation, the same value as the superconducting gap, in agreement with [7,18]. The transition seen in Fig. 3 is almost identical to that observed in [7] where the superconducting gap to pseudogap transition did not correspond to doping variations. Considering these similarities, we conclude that in our case the gap-pseudogap transition is not caused by local changes of carrier density. We rule out the possibility that a strongly underdoped region, nonsuperconducting at 4.2 K, is responsible for the observed transition. Indeed, following the accepted phase diagram, the pseudogap widths observed here (30–35 meV) correspond to nearly optimally doped regions, which should be superconducting at 4.2 K and in zero magnetic field. On the contrary, they are not superconducting, and an additional perturbation is therefore present. With respect to single crystals, the thin films are much more inhomogenous, and the disorder variations should be considered.

Numerous experiments [20] show a superconducting to insulating transition in thin films due to disorder. There, the superconducting phase is destroyed by strong disorder which localizes the electrons entirely. However, in a recent theoretical paper by Huscroft and Scalettar [21] it was predicted that, in the presence of a moderate localization potential, the disorder influences the superconducting state in a different way. They show that, in the case of increasing disorder strength, the phase coherence is lost, while the pseudogap at the Fermi level, being the signature of the pair amplitude, is not necessarily suppressed. Experimentally, the main expected change is then in the form of the DOS: The quasiparticle peaks disappear, and the states are spread over a wider energy range [21]. This is exactly the evolution of the superconducting DOS to the pseudogap form we present in Fig 3.

In this respect, the model of Huscroft and Scalettar [21] is in agreement with the idea of preformed Cooper pairs. The existence of an incoherent pair phase at $T > T_c$ (or in the vortex core below T_c) was also invoked by Renner *et al.* to explain their experimental data. As we see, the case of disordered films might also be described by such a boson model in which, as stated in [21], the "coherence of the pair phase is the central issue, and the role of fluctuations in the pair amplitude is suppressed." If this is the case, the three pseudogaps shown in Fig. 2 have the same origin, thus giving a logical explanation for the evident similarities between them.

In conclusion, we observed the quasiparticle LDOS in thin films of Bi2212 at 4.2 K. Characterization measurements indicated a highly inhomogenous system. Surprisingly, rather than continuous variation in the superconducting gaps, due to fluctuations of local doping, the STS data revealed numerous superconducting regions with stable gaps immersed in a nonsuperconducting surface. The main observation is the pseudogap regions found near superconducting islands. The STS spectra obtained there are identical to the pseudogap LDOS observed in high-quality single crystals at $T > T_c$ or in the vortex core, where the temperature and the magnetic field lead to the particular nonsuperconducting state. We suggest that the disorder plays a similar role in thin films, affecting the superconductivity by breaking the coherence of the pair phase.

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