Inherent inhomogeneities in tunneling spectra of $Bi_2Sr_2CaCu_2O_{8-x}$ crystals in the superconducting state

C. Howald,¹ P. Fournier,² and A. Kapitulnik¹

¹Departments of Applied Physics and of Physics, Stanford University, Stanford, California 94305 ²Département de Physique, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K2R1

(Received 15 January 2001; published 20 August 2001)

Scanning tunneling spectroscopy on cleaved BSCCO(2212) single crystals reveals inhomogeneities on length scales of ~ 30 Å. While most of the surface yields spectra consistent with a *d*-wave superconductor, small regions show a doubly gapped structure with both gaps lacking coherence peaks and the larger gap having a size typical of the respective pseudogap for the same sample.

DOI: 10.1103/PhysRevB.64.100504

PACS number(s): 74.50.+r, 74.25.Jb, 74.72.Hs

Tunneling spectroscopy has been an important tool in the study of high-temperature superconductors since their discovery. While in the early days of high- T_c a variety of gap sizes and structures were found and introduced much controversy into the subject, later measurements yielded greater consistency among groups, revealing a more coherent picture of the surface of high- T_c materials as viewed with STM. Among many examples we note that STM studies revealed the nature of the superstructure in BSCCO,¹ the size² and *d*-wave nature³ of the gap, the effect of local impurities and the emergence of zero-bias anomalies,⁴⁻⁶ and the electronic structure of the core of vortices.^{7,8}

While in general published data concentrates on specific aspects of the properties of the superconductor such as the gap, the pseudogap, impurities, etc., very little has been published on the large-scale structure of the electronic state at the surface of high- T_c materials. Judging from photoemission measurements that reveal a \vec{k} -dependent gap and pseudogap, 9^{-12} thus asserting that \vec{k} is a good quantum number, it has been the common belief that the surface is homogeneous with only few scattering centers and that it represents the bulk properties. However, recent measurements by a variety of techniques suggest that superconductivity may not be homogeneous in high- T_c superconductors. Recent STM studies of YBCO (Ref. 13) and BSCCO (Ref. 14) show profound variations in the tunneling spectra. In particular, the BSCCO results on highly disordered films and lead-doped crystals of Cren et al. exhibited pseudogap spectra over a large fraction of the sample surfaces. The issue of homogeneity (either in the static or dynamical sense) is very important in view of recent theoretical developments that find phase separation to be an integral part of the high- T_c scenario.15-17

In this paper we discuss scanning tunneling spectroscopy measurements on the surface of $Bi_2Sr_2CaCu_2O_{8-x}$ (BSCCO) single crystals which show that superconductivity in high temperature superconductors is inhomogeneous. This inhomogeneity appears at a scale of a few coherence lengths and is not a consequence of strong scatterers or strong disorder. We argue that this inhomogeneity is likely a consequence of electronic phase separation into regions that are proximity-coupled to give a continuous variation of the tunneling spectroscopic features.

The slightly underdoped single crystals of BSCCO used in this study were grown by a directional solidification method¹⁸ and their T_c of ~80 K was measured by SQUID magnetometry. Crystals were mounted into a UHV-low-T STM system. The samples are cleaved at room temperature in a vacuum of better than 10^{-9} torr. Immediately, they are transferred to the low temperature chamber which houses the STM and presumably reaches much lower base pressure due to cryopumping. The base temperature of the microscope is 6 K, however the data shown in this paper were taken at 8 K. Images are taken with a gold tip at a sample bias of -200 mV and a setpoint current of 100 pA. The dI/dVspectra are taken using a lock-in technique with an ac modulation of 1 mV, after fixing the tip location at a height that gives 100 pA current at -200 mV. This setpoint current establishes the relatively arbitrary normalization condition used for the raw data. The quality of the surface and the scanning capability are demonstrated in Fig. 1(a) where we show a topographic scan of a 100 $\text{\AA} \times 90$ \AA region. Evident are the atomic resolution and the clarity of the superstructure¹ with average periodicity ~ 27 Å. All cleaves which yield images exhibit similar topography.

Figure 1(b) shows a line of differential conductance spectra taken over a range of 80 Å in the region shown in Fig. 1(a). By fitting the acquired spectra to a *d*-wave density of states for a maximum gap size $\Delta [\Delta(\theta) = \Delta \cos(2\theta)]$ with Gaussian broadening Γ^{19}

$$\frac{N_{\mathcal{S}}(E)}{N_{N}} = \int_{-\infty}^{\infty} \operatorname{Re} \int_{0}^{2\pi} \frac{d\theta}{2\pi} \frac{E}{\sqrt{E^{2} - \Delta(\theta)^{2}}} \frac{e^{-(E-U)^{2}/2\Gamma^{2}}}{\sqrt{2\pi}\Gamma} dU,$$
(1)

we find two important results. For most of the spectra this fit yields values of $\Delta = 42\pm 2$ mV and $\Gamma = 4\pm 2$ mV with residuals approximately the size of the noise. However, there are regions in which the spectra give poor fits to this formula, also yielding unphysically large values of Γ and Δ . The regions are 20 to 30 Å across, their areal fraction is ~20%, and their centers lie within 5 Å of the superstructure ridges. The size and shape of one of the regions is shown in Fig. 2(a), which maps the gap size (normalized by the 42 mV value) as determined by the *d*-wave fit. This map,

PHYSICAL REVIEW B 64 100504(R)



FIG. 1. Typical topographic scan of a 100 Å \times 90 Å area (a). Dotted line marks the location of the spectroscopic scans shown in the lower figure (b).

like Fig. 1(b), shows that the spectra vary continuously through each region.

This continuous variation suggests two extremal spectra: those from the center of these regions and those far away. These two extreme shapes are shown in Fig. 3 for positive bias. In particular, the spectrum taken outside the regions is consistent with the density of states of a d-wave superconductor averaged over k space (as expected for tunneling perpendicular to the Cu-O planes), while the other shows a double gap shape with no coherence peaks. In addition, the shape of the smaller of these two gaps is similar to the shape of the inner part of the superconducting spectra. Figure 2(b) shows the width at which the conductance reaches twice the zero bias value, normalized by the value of those spectra with the best d-wave fits. This plot uses the same scale as Fig. 2(a) in order to emphasize the contrast between them. The only features in Fig. 2(b) are consistent with noise. That is, while the behavior at the gap energy is inhomogeneous, at low energies the sample appears to be homogeneous. The fact that the spectra in these regions look like those of the background superconductor at low bias indicates that the



FIG. 2. Map of normalized gap size (a). High points reflect a larger gap with suppressed coherence peaks. Lower map (b) is the width of the spectrum at twice the minimum conductance G(0), indicating a homogeneous gap structure at low energies (see text).

node structure exists throughout the sample. Thus the regions maintain the fourfold asymmetry of the superconducting state despite the apparent suppression of superconductivity. In particular, this rules out these features being caused by impurities. Both theoretical^{20,21} and experimental studies^{4–6} show that potential scatterers yield entirely different behavior, characterized by subgap structure (zero-bias anomalies): the largest change in spectra near potential scatterers is in the range where we see no change.

Examination of Fig. 1 and other similar scans shows that the two extreme shapes are separated by a distance of ~ 30 Å. For BSSCO this distance is roughly twice the coherence length ξ . As will be discussed below, this suggests that the smooth variation seen may not be the product of smooth variation in the underlying character of the material, but may instead reflect a breakdown into domains with different character which are then proximity coupled. This proximity coupling would explain why these regions do not break the superconductor *d*-wave symmetry.

Although curve (b) in Fig. 3 has the same shape at low bias as a "good" superconductor, it develops a different gap structure at higher bias. The abrupt change in gap structure is an indication that the spectrum of quasiparticles changes INHERENT INHOMOGENEITIES IN TUNNELING ...



FIG. 3. Extremal dI/dV from the region shown in Fig. 2. Location of the spectra is indicated in the topograph (right). Note that the two spectra are taken at equivalent positions with respect to the superstructure. In the spectra, Δ represents the gap energy while ϵ marks the kink energy, related to the size of the "bad" superconductor region (see text).

abruptly. HTSC, and in particular BSCCO crystals of the type we are using are believed to be in the clean limit. The superconducting state is therefore characterized by a gap Δ , associated with pairing instability and "pair size" $\xi \sim \hbar v_F / \Delta$. Terminating the gap opening at energies $\epsilon < \Delta$ means a larger length scale $L \sim \hbar v_F / \epsilon$. Since the coherence peaks mark roughly the coherence length, the shoulder in curve (b) indicates a length scale that is roughly twice the coherence length. This is therefore the length at which the system crosses over from the behavior of a pure *d*-wave superconductor at long length scales to behavior which is more reminiscent of the "insulating" state. The larger gap with no coherence peaks resembles the pseudogap.

Summarizing our observations, we find that while the gap changes smoothly between the two extremes presented in Fig. 3, the system breaks into distinct domains of either "good" or "bad" superconductivity. This observation suggests that the "bad" superconducting regions are a consequence of insulating regions in proximity with the "good" superconductor. Since for high- T_c superconductors the coherence length is so short, proximity implies leakage of the superconducting wave function a distance of order ξ into the insulator.²⁴ Similarly, the superconductor weakens as well over a similar distance close to the boundary. The result is therefore the smoothly modulated behavior shown in Fig. 2. This follows from a scenario for phase separation where the initial boundaries between adjacent puddles are relatively sharp. Such a situation is most likely a consequence of a proximity to a first order phase transition that is becoming continuous due to disorder $^{15-17,22}$ as was first shown by Imry and Ma.²³ The sharpness of the initial domains is indicated by the relatively sharp kink in the spectrum of the "bad" superconductor as it crosses over from low energy superconductorlike to high energy pseudo-gap-like behavior.

The above suggestion that the inhomogeneities observed

PHYSICAL REVIEW B 64 100504(R)



FIG. 4. Spectra taken near the disordered area. The location of each spectrum is shown in the topograph (right).

by STM spectroscopy on the surface of BSCCO single crystals are intrinsic and reflect phase separation does not contradict the photoemission results.⁹⁻¹² While one might expect that inhomogeneities would destroy the k dependence observed in photoemission, the type of inhomogeneities seen here would not. The low-k behavior would measure the longwavelength homogeneity, while, since both the superconducting gap and the pseudogap have the same symmetry, averaging in regions of pseudogap would not destroy the symmetry of the gap seen in photoemission. Moreover, the above scenario is also in agreement with recent investigations of vortex structure in high- T_c superconductors. The peculiar result there is the fact that while the zero-field spectroscopy maps show an inhomogeneous pattern, application of a magnetic field results in vortices that are always surrounded by "good" superconductor as judged from the STM spectroscopy.^{7,8} Within the framework of our interpretation this implies that vortices will prefer to have their cores reside on "bad" superconducting regions. The surrounding area is then a good superconductor and since the vortex core does not proximity couple to the superconductor, the result is a homogeneous superconductor in the regions surrounding the vortex.

Having argued that the observed inhomogeneities are not the consequence of disorder, we can check this claim by intentionally disordering the surface. Since BSCCO is very sensitive to excessive current density or electric field we locally increased the setpoint current to 500 pA at -200 mV sample bias and scanned the tip over a 140 $\text{\AA} \times 90$ \AA area. Subsequent topography shows an apparent depression at the center of the "damaged" area because of decreased local density of states. The superstructure and atomic corrugation are no longer visible within this region, indicating strong disorder, though they remain visible over the rest of the sample surface. Figure 4 shows a strip image of the contour of constant integrated density of states and a line of spectra taken across the "damaged" area. Note that Cren et al.¹⁴ see qualitatively similar spectra in strongly disordered samples. Clearly at about 100 Å away from the center of that area a

C. HOWALD, P. FOURNIER, AND A. KAPITULNIK

good superconducting gap is obtained indicating only very small influence of the damaged area. As we approach the boundary of the scanned area and continue towards its center the spectra change continuously. In particular, the height of the coherence peak drops continuously to the background conductance level. However, unlike the variations in the spectra shown in Fig. 1, here the gap size does not change at all as the coherence peaks disappear. Once inside the region, the gap broadens into a pseudo-gap, and then an insulating gap. Spectra *F* and *G* near the border of the damaged area look similar to those of the pseudogap observed by tunneling⁷ and photoemission.¹²

Comparing this area with the region on the native surface, several points are obvious. Both show a continuous decrease in the coherence peak height. Also, both show pseudogap features inside the region. However, the pseudogap size varies in the intentionally disordered case while it seems to be constant in the other. This is presumably because the disorder, and perhaps doping as well, is varying throughout this region, yielding a varying intrinsic gap size. In addition, for

- ¹M.D. Kirk, J. Nogami, A.A. Baski, D.B. Mitzi, A. Kapitulnik, T.H. Geballe, and C.F. Quate, Science **242**, 1673 (1988).
- ²C. Renner, O. Fischer, A.D. Kent, D.B. Mitzi, and A. Kapitulnik, Physica B **194**, 1689 (1994).
- ³B. Barbiellini, O. Fischer, M. Peter, C. Renner, and M. Weger, Physica C **220**, 55 (1994).
- ⁴Ali Yazdani, C.M. Howald, C.P. Lutz, A. Kapitulnik, and D.M. Eigler, Phys. Rev. Lett. 83, 176 (1999).
- ⁵E.W. Hudson, S.H. Pan, A.K. Gupta, K.W. Ng, and J.C. Davis, Science **285**, 88 (1999).
- ⁶S.H. Pan, E.W. Hudson, K.M. Lang, H. Eisaki, S. Uchida, and J.C. Davis, Nature (London) **403**, 746 (2000).
- ⁷Ch. Renner, B. Revaz, K. Kadowaki, I. Maggio-Aprile, and O. Fischer, Phys. Rev. Lett. **80**, 3606 (1998).
- ⁸S.H. Pan, E.W. Hudson, A.K. Gupta, K.-W. Ng, H. Eisaki, S. Uchida, and J.C. Davis, Phys. Rev. Lett. 85, 1536 (2000).
- ⁹Z.-X. Shen, D.S. Dessau, B.O. Wells, D.M. King, W.E. Spicer, A.J. Arko, D. Marshall, L.W. Lombardo, A. Kapitulnik, P. Dickinson, S. Doniach, J. DiCarlo, A.G. Loeser, and C.H. Park, Phys. Rev. Lett. **70**, 1553 (1993).
- ¹⁰Z.-X. Shen and D. Dessau, Phys. Rep. **253**, 2 (1995).
- ¹¹ H. Ding, T. Yokoya, J.C. Campuzano, T. Takahashi, M. Randeria, M.R. Norman, T. Mochiku, K. Kadowaki, and J. Giapintzakis, Nature (London) **382**, 51 (1996).

- ¹²A.G. Loeser, Z.X. Shen, M.C. Schabel, C. Kim, M. Zhang, A. Kapitulnik, and P. Fournier, Phys. Rev. B 56, 14185 (1997).
- ¹³H.L. Edwards, D.J. Derro, A.L. Barr, J.T. Markert, and A.L. de Lozanne, Phys. Rev. Lett. **75**, 1387 (1995).
- ¹⁴T. Cren, D. Roditchev, W. Sacks, J. Klein, J.-B. Moussy, C. Deville-Cavellin, and M. Lagues, Phys. Rev. Lett. **84**, 147 (2000); T. Cren, D. Roditchev, W. Sacks, and J. Klein, cond-mat/0006039 (unpublished).
- ¹⁵ V.J. Emery, S.A. Kivelson, and H.Q. Lin, Phys. Rev. Lett. 64, 475 (1990).
- ¹⁶V.J. Emery and S.A. Kivelson, Physica C **209**, 597 (1993).
- ¹⁷S. Chakravarty, R.B. Laughlin, D.K. Morr, and C. Nayak, Phys. Rev. B **63**, 094503 (2001).
- ¹⁸P. Fournier, A. Kapitulnik, and A.F. Marshall, Physica C 257, 291 (1996).
- ¹⁹M. Oda, C. Manabe, and M. Ido, Phys. Rev. B 53, 2253 (1996).
- ²⁰M.I. Salkola, A.V. Balatsky, and D.J. Scalapino, Phys. Rev. Lett. 77, 1841 (1996).
- ²¹Y. Tanaka and S. Kashiwaya, Phys. Rev. B **53**, 9371 (1996).
- ²²E. Shimshoni, A. Auerbach, and A. Kapitulnik, Phys. Rev. Lett. 80, 3352 (1998).
- ²³Y. Imry and S.-K. Ma, Phys. Rev. Lett. **35**, 1399 (1975).
- ²⁴G. Deutscher and K.A. Müller, Phys. Rev. Lett. **59**, 1745 (1987).

PHYSICAL REVIEW B 64 100504(R)

the native defect, there remains a vestige of the superconducting gap in the pseudogapped spectra. This indicates that the smaller size of the native defect allows proximity coupling to the superconductor that is lost inside the larger manufactured defect. The effect of disorder is therefore different than the intrinsic inhomogeneities observed on the pure surface. When strong enough, the disorder "pins" the insulating phase as is the case in the middle of the damaged area. Again, this implies a "good" superconductor at the periphery of the "bad" superconductor.

In conclusion, we find that the intrinsic surface of BSCCO exhibits inhomogeneities that strongly suppress superconductivity, while maintaining the same low energy structure. These regions show spectra that are reminiscent of the pseudogap, but also appear to be proximity coupled to the surrounding superconductor.

We thank Steve Kivelson and Bob Laughlin for many useful discussions. Work was supported by AFOSR.