Quasiparticle Tunneling Properties of Planar $YBa_2Cu_3O_{7-\delta}$/$PrBa_2Cu_3O_{7-\delta'}$/$HoBa_2Cu_3O_{7-\delta}$ Heterostructures

A. M. Cucolo, R. Di Leo, A. Nigro, P. Romano, and F. Bobba
Unità Istituto Nazionale di Fisica della Materia-Dipartimento di Fisica, Università di Salerno, I-84081 Baronissi (SA), Italy

E. Bacca and P. Prieto
Departamento de Fisica, Universidad del Valle, A.A. 25360 Cali, Colombia
(Received 7 August 1995)

We report a demonstration of *S-I-S* quasiparticle tunneling in *c*-axis oriented planar junctions consisting of YBaCuO/PrBaCuO/HoBaCuO thin film heterostructures. In the conductance curves at low temperatures well developed peaks indicative of gap structures are found at ±45 mV. The high bias conductance shows flat dependence on energy and the best quality junction has a zero-bias conductance less than 0.05 of the normal state value. We discuss our spectra in the framework of a BCS-like density of states as well as in terms of a $d_{x^2-y^2}$ symmetry of the superconducting order parameter.

PACS numbers: 74.50.+r, 74.80.Dm

Tunneling studies in high-T_c heterostructures composed of superconducting-insulating-superconducting (*S-I-S*) layers are of great interest from both a fundamental and a technological point of view. Junction characteristics in fact can provide important information on the quasiparticle density of states and on the mechanism responsible for superconductivity which still are open questions about high-T_c superconductors (HTS). On the other hand, trilayers are the basic structure for high-frequency transmission lines, for filters, and for the realization of Josephson junctions for high-T_c integrated circuits.

Problems associated with multilayer structures arise from the insulating barriers and interfaces. Barrier materials need to have a close lattice match and compatible deposition conditions with the HTS in order to propagate epitaxial growth. Formation of good quality interfaces is also important. The roughness of the surfaces can produce the mixing of properties of different directions in these anisotropic materials. In addition, due to the extremely short coherence length ξ, the superconducting properties are easily degraded at the interfaces. To overcome this last difficulty, superconductor-normal-superconductor (*S-N-S*) junctions have been developed for Josephson devices, since, due to the proximity effect, they show Josephson coupling even if the metal barrier thickness is greater than ξ.

Recently, different kinds of all-oxide *S-N-S* structures, including grain boundary, step edge, ramp-type, and sandwich-type junctions, have been realized by several groups for high-T_c device applications. In particular, $YBa_2Cu_3O_{7-\delta}$/$PrBa_2Cu_3O_{7-\delta'}$/$YBa_2Cu_3O_{7-\delta}$ junctions have been investigated in detail by using *a*-axis oriented [1–3] and, more recently, (103)-oriented films [4].

In contrast with the excellent results obtained in Josephson coupled *S-N-S* trilayers, clear evidence for quasiparticle tunneling is difficult to obtain in all-oxide *S-I-S* junctions. Several attempts to realize HTS-*I*-HTS structures have been reported in the literature. $Ba_{(1-x)}K_xBiO_3$/$BaBi_2O_y$/$Ba_{(1-x)}K_xBiO_3$ sandwich junctions [5], $YBa_2Cu_3O_{7-\delta}$/$PrBa_2Cu_3O_{7-\delta'}$/$YBa_2Cu_3O_{7-\delta}$ ramp-type junctions [6], $HoBa_2Cu_3O_{7-\delta}$/$PrBa_2Cu_3O_{7-\delta'}$/$HoBa_2Cu_3O_{7-\delta}$ heterostructures [7], and $Bi_2Sr_2CaCu_2O_8$/$Bi_2Sr_2YCu_2O_8$/$Bi_2Sr_2CaCu_2O_8$ junctions [8] have been measured. However, in all these structures, due to the difficulty of realization of good quality interfaces between the insulating barrier and the counterelectrodes, quasiparticle tunneling shows a *S-I-N* behavior with gaplike features developed well below the measured T_c of the HTS films.

Some encouraging results have been obtained in junctions with natural barriers. The simultaneous presence of the Josephson effect and quasiparticle tunneling has been reported in YBaCuO-based junctions with conventional superconducting counterelectrodes [9] but a "complete" energy gap for the YBaCuO compound has not been observed. Coexistence of both effects has also been found in BaKBiO junctions with both natural and artificial barriers [10,11].

In this Letter we report on the successful growth of $YBa_2Cu_3O_{7-\delta}$/$PrBa_2Cu_3O_{7-\delta'}$/$YBa_2Cu_3O_{7-\delta}$ and $YBa_2Cu_3O_{7-\delta}$/$PrBa_2Cu_3O_{7-\delta'}$/$HoBa_2Cu_3O_{7-\delta}$ trilayer structures in which evidence for *S-I-S* quasiparticle tunneling is observed. The trilayer structures were realized by sequential deposition of the films from sintered stoichiometric targets by using a high pressure dc sputtering process in pure oxygen. Details on the fabrication procedure have been reported elsewhere [12]. (001) $SrTiO_3$ substrates have been used. Cross-type junctions of $0.5 \times 0.5\ mm^2$ have been prepared by depositing the films through $SrTiO_3$ shadow masks. The vacuum chamber was opened to change the masks for the deposition of subsequent layers and only one junction at a time on each substrate was fabricated. Barrier thickness ranged between 100 and 300 Å.

 0031-9007/96/76(11)/1920(4)$10.00

Trilayer characterization was carried out by means of x-ray analysis, scanning electron microscopy (SEM), and energy dispersive spectroscopy (EDS). These analyses showed high-quality c-axis oriented heterostructures with correct stoichiometry. Electrical resistivity was measured that indicated for both the bottom and top layers metallic behavior in the normal state and sharp superconducting transitions between 89 and 91.6 K with $\Delta T_c \leq 2$ K.

We have fabricated 12 trilayer junctions, among them four showed S-I-S behavior, two S-I-N features, while the remaining samples gave no clear evidence for quasiparticle characteristics. In this Letter, we concentrate on the results obtained on $YBa_2Cu_3O_{7-\delta}/PrBa_2Cu_3O_{7-\delta'}/HoBa_2Cu_3O_{7-\delta}$ junctions with a 100 Å barrier layer for which the best quality S-I-S' tunneling characteristics have been measured.

In Fig. 1 the I-V and the dI/dV vs V characteristics of a YBaCuO/PrBaCuO/HoBaCuO junction are showed at $T = 4.2$ K. Well defined maxima in the conductance curve are observed at about ± 45 mV while the low bias tunneling conductance shows more states in the gap than predicted by the BCS theory. The ratio between the zero-bias conductance and the conductance value at 150 mV is less than 0.05 in this sample and ranged between 0.05 and 0.4 in the other junctions. Low zero-bias conductance values are rarely measured in planar HTS junctions. They have been mainly observed in point contact and break junctions [13–15]. Recently, variation of this feature in BiSrCaCuO vacuum tunneling spectra has been related to the changing characteristics of the junction topmost layers [13].

In contrast with previous reports on the YBaCuO system [16,17], a flat background instead of a linear conductance is measured at high biases. By means of vacuum tunneling spectroscopy [13], it has been demonstrated that in BiSrCaCuO single crystals flat backgrounds are associated to highly reproducible spectra with sharp gap features, while, on the same sample, linear backgrounds reflect a degraded surface stoichiometry. In BaKBiO-based junctions, flat backgrounds have been found in all-oxide heterostructures [11] while a linear behavior has been measured in junctions with native barriers [18]. In this last case, a universal relationship has been reported between the conductance slope and the conductance value at zero bias: $G(V) = G(0)(1 + \alpha V)$ as predicted by the marginal Fermi liquid model [19]. We have found a similar dependence in YBaCuO-based junctions with natural barriers [20]. These observations seem to indicate that also the high-bias conductance behavior depends on the properties of the closest layers to the barrier. In this sense the flat background observed in Fig. 1 is well related to the low measured value of the conductance at zero voltage.

We notice that a certain asymmetry is observed in the curves of Fig. 1 that might reflect an asymmetry of the tunnel barrier due to not perfectly equivalent interfaces. In fact, in HTS heterostructures, realization of a superconducting counterelectrode and a good-quality second interface is very difficult to achieve [5–8]. Moreover, extrapolation to zero current of the Ohmic I-V curve of Fig. 1 seems to indicate the presence of Coulomb blockade. However, we consider that the effect on the evaluation of the derivation maximum positions of our spectra is negligible, since the intercepts at zero from both high positive and negative currents give voltage values well below the gap edges [21].

In Fig. 2 we show the $G(V)$ normalized to $G(150 \text{ mV})$ for the same junction of Fig. 1 at temperatures both above and below T_c. In all our junctions we have found that the high-bias conductance depends on temperature, decreasing by approximately 1 order of magnitude from 4.2 to 100 K. A similar effect has been found in BaKBiO/BaBiO/BaKBiO sandwich junctions [5] and, although less pronounced (10%), in YBaCuO-based junctions with natural barriers [16]. As it can be observed from the figure, up to 40 K, the conductance peaks are progressively smeared and move to higher voltages as the temperature is increased. This effect is qualitatively similar to that found in tunnel junctions with isotropic, BCS superconducting electrodes as a consequence of thermal smearing. Some anomaly in the conductance behavior is observed for temperatures above 50 K.

We have investigated this aspect more closely and in Fig. 3 the normalized conductance as a function of $V^{1/2}$

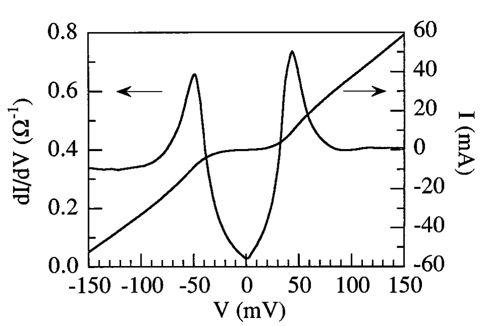

FIG. 1. I-V and dI/dV vs voltage characteristics of a YBaCuO/PrBaCuO/HoBaCuO junction at $T = 4.2$ K.

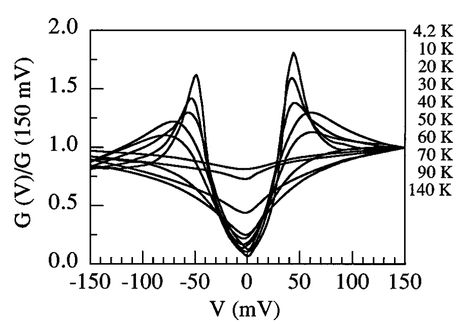

FIG. 2. Normalized conductances vs voltage at different temperatures.

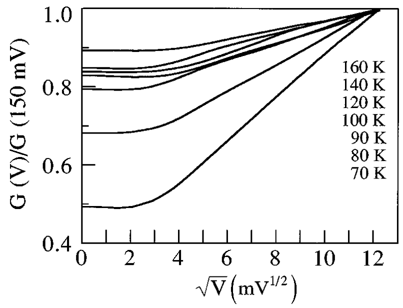

FIG. 3. Normalized conductances as a function of $V^{1/2}$ at temperatures above 70 K.

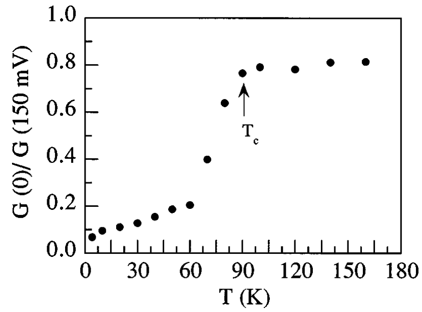

FIG. 4. Normalized zero-bias conductance vs temperature.

is reported for temperatures between 70 and 160 K. The deviation at low voltages can be ascribed to the effect of thermal smearing. A square root dependence on energy has been predicted by McMillan [22] for the density of states of systems close to the metal-insulator transition and has been experimentally observed in disordered, three-dimensional systems [23,24]. Since the PrBaCuO compound is near to the metal-insulator transition, we do not exclude that some disorder might drive to the metallic state few interface layers, resulting in a square root dependence of the density of states on energy. This process depends on temperature and may modify the transport from a tunneling to a diffusion mechanism, as studied in Nb/amorphous Si/Nb tunnel barriers [25].

To conclude that the whole set of data of Figs. 1–3 reflects the intrinsic YBaCuO density of states, it is important to investigate both the status of the interfaces as well as the nature of the transport mechanism in the barrier. Charge carriers are generally localized in PrBaCuO and, at least for large thickness (>1000 Å), transport properties are dominated by a variable range hopping mechanism [7]. In principle, one cannot exclude that also tunneling electrons proceed via an intermediate state in the barrier.

We carried out some controls on the quality of our junctions [26] and in Fig. 4 we report the temperature dependence of the normalized zero-bias conductance. As can be observed, superconducting structures first appear at $T_c = 90$ K where there is a discontinuity in the $dG(0)/dT$ similar to what one would expect due to the opening of the energy gap at T_c in a conventional, BCS superconductor. This is a strong indication that, at least at one interface, there is no thick (on the scale of ξ) reduced-T_c layer. A second discontinuity is seen around 60 K that could be related to some reduced T_c of the HoBaCuO bottom layer and/or to a variation of the transport mechanism through the barrier. However, a double discontinuity in the $G(0)$ vs T dependence has also been observed in YBaCuO/I/N junctions with natural barriers [16,17], in which gap features appear at $\pm\Delta$. In the present work the $G(0)$ vs T behavior is very similar while the tunneling spectra shows S-I-S structures at $\pm 2\Delta$. This seems to be reasonable evidence that superconductivity of the electrodes is preserved at both interfaces.

Besides, conservation of states between normal and superconducting spectra is an important issue to identify barrier-related effects. We have found that integration over energy of the 4.2 and 140 K curves of Fig. 2 yields variation of the number of states less than 4%.

After these observations, in Fig. 5 we compare the tunneling data of Fig. 1 (open circles) with the theoretical curve of a S-I-S junction with isotropic, BCS-like superconductors (dashed line). It can be observed that, if some pair breaking is accounted for, a quite satisfactory fitting of the peak amplitude and width is achieved. However, the experiments measure more states in the gap. The dashed line in the figure was computed by using for both electrodes a modified BCS density of states:

$$N(E) = \mathrm{Re}\{(E + i\Gamma)^2/[(E + i\Gamma)^2 - \Delta^2]\}^{1/2},$$

where Re stand for the real part and Γ is a phenomenalogical parameter introduced by Dynes, Narayanamurti, and Garno [27]. In the fitting we have used $\Delta_{\mathrm{YBCO}} = \Delta_{\mathrm{HBCO}} = 21.5$ meV and $\Gamma_{\mathrm{YBCO}} = \Gamma_{\mathrm{HBCO}} = 4.5$ meV.

In Fig. 5 we report (full line) the low bias conductance behavior predicted for a S-I-S junction with $d_{x^2-y^2}$ symmetry of the superconducting order parameters. An almost quadratic dependence on energy is expected in this case [28] that is close to the low bias behavior of the

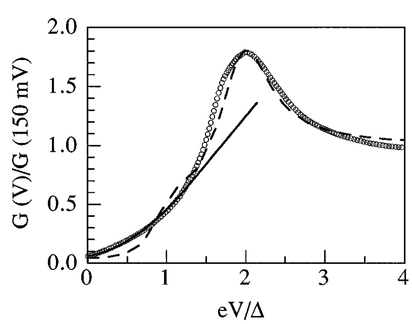

FIG. 5. The same data as in Fig. 1 (open circles). BCS fit with a pair-breaking contribution: $\Delta = 21.5$ meV, $\Gamma = 4.5$ meV (dashed line). Asymptotic low-bias behavior predicted for a $d_{x^2-y^2}$ wave symmetry (full line).

experimental data. Moreover, the amplitude of the measured peaks is compatible with the tunneling conductance of a *S-I-S* junction with $d_{x^2-y^2}$ wave electrodes without invoking any pair breaking mechanism in the barrier.

In summary, we have measured an almost complete energy gap in the density of states of the YBaCuO and HoBaCuO compounds that have been traditionally considered the more gapless among the high-T_c superconductors. Some of the literature's most debated issues have been discussed in the paper such as the unusual presence of a flat background conductance in the YBaCuO tunneling spectra, as well as the possibility of a $d_{x^2-y^2}$ wave symmetry of the superconducting order parameter. It is our opinion that one of the most striking features of this work is that the results have been obtained in all-oxide heterostructures in which *S-I-S* tunneling spectra are extremely difficult to achieve. Even if some anomaly might depend on the use of a nonconventional barrier, we consider our results of great encouragement for future development of trilayer structures for HTS device applications.

The authors thank M. Carotenuto and L. Falco for the technical assistance. This work was supported by the Consiglio Nazionale delle Ricerche (CNR) under Project "Interface studies in high-T_c based planar junctions" and by COLCIENCIAS under Contract No. 1106-05-197-95.

[1] J. B. Barner *et al.,* Appl. Phys. Lett. **59**, 742 (1991).
[2] T. Hashimoto *et al.,* Appl. Phys. Lett. **60**, 1756 (1992).
[3] T. Umezawa *et al.,* Appl. Phys. Lett. **63**, 3221 (1993).
[4] H. Sato, H. Akoh, and S. Takada, Appl. Phys. Lett. **64**, 1286 (1994).
[5] S. Martin *et al.,* Phys. Rev. B **47**, 14 510 (1993).
[6] T. Becherer *et al.,* Phys. Rev. B **47**, 14 650 (1993).
[7] N. Kabasawa *et al.,* Phys. Rev. Lett. **70**, 1700 (1993).
[8] A. M. Cucolo *et al.,* Appl. Phys. Lett. (to be published).
[9] A. G. Sun *et al.,* Phys. Rev. Lett. **72**, 2267 (1994).
[10] A. N. Pargellis *et al.,* Appl. Phys. Lett. **58**, 95 (1991).
[11] R. L. Fink *et al.,* Appl. Phys. Lett. **62**, 3360 (1993).
[12] P. Prieto *et al.,* Solid State Commun. **83**, 195 (1992).
[13] Ch. Renner and O. Fischer, Phys. Rev. B **51**, 9208 (1995).
[14] D. Mandrus *et al.,* Europhys. Lett. **22**, 199 (1993).
[15] H. L. Edwards, J. T. Markert, and A. L. de Lozanne, Phys. Rev. Lett. **69**, 29 671 (1992).
[16] M. Gurvitch *et al.,* Phys. Rev. Lett. **63**, 1008 (1989).
[17] A M. Cucolo, C. Noce, and A. Romano, Phys. Rev. B **46**, 5864 (1992).
[18] F. Sharifi, A. Pargellis, and R. C. Dynes, Phys. Rev. Lett. **67**, 509 (1991).
[19] C. M. Varma *et al.,* Phys. Rev. Lett. **63**, 1996 (1989).
[20] A. M. Cucolo *et al.,* in *High Temperature Superconductivity,* edited by M. Acquarone (World Scientific, Singapore, 1995), p. 363.
[21] P. J. M. Van Bentum *et al.,* J. Magn. Magn. Mater. **76&77**, 561 (1988).
[22] W. L. McMillan, Phys. Rev. B **24**, 2739 (1981).
[23] R. C. Dynes and J. P. Garno, Phys. Rev. Lett. **46**, 137 (1981).
[24] G. Hertel *et al.,* Phys. Rev. Lett. **50**, 743 (1983).
[25] Y. Xu, A. Matsuda, and M. R. Beasley Phys. Rev. B **42**, 1942 (1990).
[26] A. M. Cucolo, Int. J. Mod. Phys. B **7**, 2549 (1993).
[27] R. C. Dynes, V. Narayanamurti, and J. P. Garno, Phys. Rev. Lett. **41**, 1509 (1978).
[28] H. Von and K. Maki, Phys. Rev. B **49**, 1397 (1994).