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Tunneling spectroscopy into $YBa_2Cu_4O_8$: Intralayer and interlayer analysis

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We have measured the superconducting and normal tunneling characteristics of quality controlled planar junctions based on $YBa_2Cu_4O_8$ (Y124) single crystals. Our data show that the in-plane properties are similar to the $YBa_2Cu_3O_7$ (Y123) system. A higher value of the zero-bias conductance is measured in the Y124, indicating an increased number of normal electrons due to the presence of the double chain structure. Linearity of the background conductance is observed in both compounds. Less steep slopes are found in Y124 that might indicate a reduced interlayer coupling in this system.

The most essential structural unit common to all cuprate high-T_c superconductors (HTS) are p-type or n-type doped CuO_2 planes which are believed to be responsible for superconductivity. Doping is induced by the neighboring structures, making these planes evolve from antiferromagnetic insulators (undoped case) to quasi-two-dimensional conductors.

Each HTS system can be labeled by the number n of adjacent CuO_2 layers in the unit cell. In the La-Sr-Cu-O compounds these planes are fairly isolated from each other ($n=1$) while in other materials they can cluster in groups of two or three as in the Y-Ba-Cu-O and in the 2:2:1:2 Bi-Sr-Ca-Cu-O phase ($n=2$) or in the 2:2:2:3 Bi-Sr-Ca-Cu-O and in the Tl-Ba-Ca-Cu-O series ($n=3$). It has been found that the critical temperature increases with the number of adjacent CuO_2 layers, each homologous series of compounds being characterized by the value of its maximum T_c, $T_{c\max}$. Recently, a universal relationship between the normalized $T_c/T_{c\max}$ and the hole concentration has been reported for p-type high-T_c cuprates.[1]

While the in-plane properties of the layered HTS are generally well known, the mechanism of the c-axis charge transport and the CuO_2 interlayer coupling remains unresolved.

To better understand the role of the interlayer coupling, Y-Ba-Cu-O/Pr-Ba-Cu-O superlattices have been mostly used,[2,3] however also the Y-Ba-Cu-O series appears to be a specific candidate.[4] In fact, it is well known that the CuO chains between CuO_2 planes are unique to these compounds. Their particular stacking along the c axis produces different members of the homologous series. Among these, the well-known $YBa_2Cu_2O_7$ (Y123) with $T_c \approx 90$ K has a single chain parallel to the b direction in the unit cell while the $YBa_2Cu_4O_8$ (Y124) with $T_c \approx 80$ K has two chain elements. This double chain causes a parallel shift by $b/2$ and zig-zag along the b axis in the Y124 and therefore doubles the unit cell lattice constant c with respect to the Y123 system.[5] Since the double chains are stable up to high temperatures ($\approx 850\,°C$), Y124 has excellent thermal stability of the oxygen stoichiometry and has no orthorhombic-tetragonal phase transformation, i.e., no twins are produced during crystal growth.

The Y124 oxide superconductor was first observed as lattice defects in partially decomposed Y123 powders and as a component of multiphase thin films. Synthesis of the bulk compound has then been achieved at high oxygen pressure[6] as well as at ambient oxygen from hydrated nitrades reactions.[7]

In this paper we report tunneling measurements on the Y124 system. Differences and similarities with the Y123 compound are discussed in order to investigate the different role of the interlayer coupling in these materials.

Our single crystals were grown at high pressure from mixtures of polycrystalline Y123, $BaCuO_2$ and CuO, as described in Ref. 5. Susceptibility measurements indicated onset T_c of about 80 K. The width of the transitions was typically less than 1 K. The tunnel junctions on Y124 and Y123 we refer in this paper had natural barriers formed by a short air exposure (≈ 30 min) of chemically etched (1% Br in methanol) single crystals and were completed by thermally evaporating a 5000-Å Pb thin film counterelectrode. The area of the cross-type junctions was about 0.1 mm^2.

In Fig. 1 we present the typical conductance characteristics of a Y124/Pb junction, the data being normalized to the conductance value at $V=100$ mV. At $T=4.2$ K, curve (a), well-defined Pb gap and phonon structures can be observed,

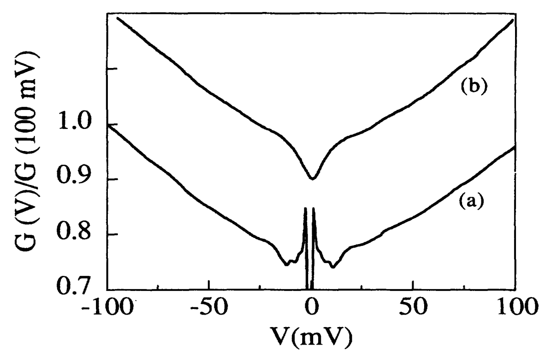

FIG. 1. Normalized conductance vs voltage for a Pb/Y124 junction, at $T=4.2$ K, curve (a), and at $T=8$ K, curve (b). The vertical axis has been shifted for clarity.

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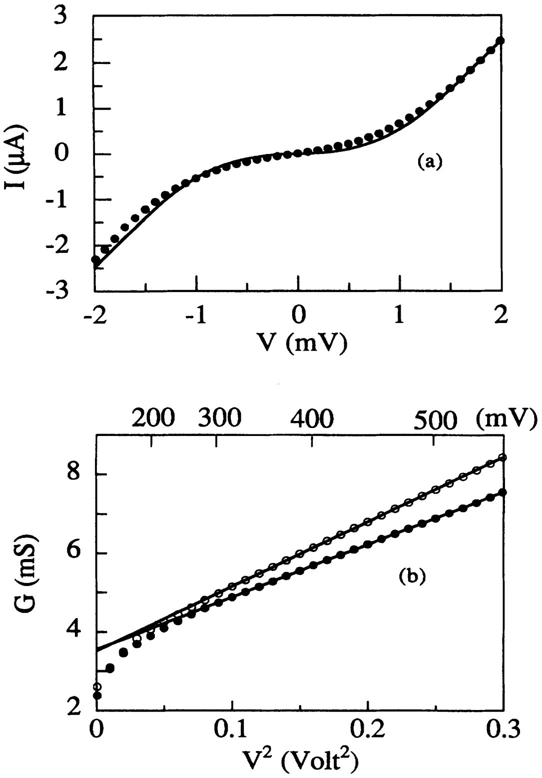

FIG. 2. Low and high bias quality controls for a Pb/Y124 junction at $T=4.2$ K. (a) *I-V* characteristic (dots) and theoretical fitting (full line) for the current flowing through a Pb-*I*-*N* junction at the same temperature. (b) Conductance as a function of voltage squared for positive (dots) and negative (open circles) biases. Full lines have been obtained from the least-squares best fitting parabola. Voltage range is indicated above the curve.

while at $T=8$ K, curve (b), the Pb superconducting features disappear and the conductance closely reproduces the Y124 density of states (DOS) provided that quality controls guarantee reliability of the measured tunneling spectra.[8]

We carried out both low and high bias controls and representative results are reported in Figs. 2(a) and 2(b). At $T=4.2$ K, with Pb in the superconducting state, curve (a), a fairly good agreement between the measured low bias *I-V* characteristics (dots) and the theoretical fitting (full line) for the current flowing through a Pb-*I*-*N* tunnel junction at the same temperature, is observed. In the fitting, the HTS was considered as normal due to the finite conductance value in the studied energy range. This analysis indicates that subgap currents are essentially due to thermally excited quasiparticles. Leakages are negligible through the barrier that has been further tested up to high bias voltages.

In Fig. 2(b) we show the high bias conductance data as a function of V^2 for both positive (dots) and negative (open circles) biases. Positive bias corresponds to positive HTS base electrode. We observe that the traditional parabolic dependence on voltage is exhibited for 200 mV$<V<$550 mV with a linear term ranging for $V<$200 mV. From the least-squares best fitting parabola (full line), an average barrier thickness $\bar{d}=15$ Å and average height $\bar{\phi}=1.9$ eV are inferred. These values are typical of our Y124 junctions and are consistent with the results obtained in conventional as well as in other HTS based junctions.[8,9]

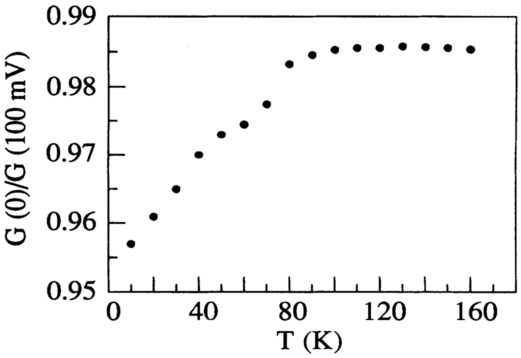

FIG. 3. Normalized zero-bias conductance vs temperature for a Pb/Y124 junction.

Both kinds of controls reported in Figs. 2(a) and 2(b) when routinely carried out indicate that a single step tunneling process is the main conductance mechanism through the barrier.

Back to Fig. 1, we observe that, in comparison with the tunneling characteristics of the Y123 system,[10] the double structure at about ±18 and ±36 mV is not well resolved, but only a broadened structure of reduced amplitude appears centered at about ±27 mV. In addition, a higher value of the zero-bias conductance is measured in the Y124 compound. We think that the increased anisotropy due to the zig-zag chains can account for the smearing of the superconducting features while an increased contribution of normal electrons can be responsible for the high $G(0)$ value. We remark that the Y123 low-energy structures reported at about ±5 mV in Ref. 10 are not present in the Y124 tunneling characteristics of Fig. 1.

In Fig. 3, the normalized zero-bias conductance $G(0,T)/G(100\text{ mV},T)$ as a function of temperature of another Y124/Pb junction is reported. As it has already been observed,[10,11] the discontinuity at $T=80$ K is indicative of particle condensation in the superconducting state. The fact that the "surface T_c" measured by tunneling spectroscopy corresponds to the "bulk T_c," of the single crystals is a strong indication that no degraded layer, on a scale of the superconducting coherence length ξ, exists at the junction interface with the tunnel barrier.

We observe that in this junction the variation of $G(0,T)$ is less than 10% in all the measured temperature range. In about 20 measured junctions, we have found that modulation of this quantity never exceeds 0.2. Controls such as those reported in Fig. 2(a) assure us that leakages are not relevant in these junctions while reproducibility of this effect makes unlikely any explanation in terms of normal grains at the barrier interface. Therefore, at the present, we have to seriously consider that this more than 80% "normal" contribution is an intrinsic feature of the Y124 compound. The variation of the same quantity in the Y123 based junctions is about 50%.[10]

To go further with the analysis of the superconducting behavior, in Fig. 4 we show the numerically computed conductance derivative vs voltage at $T=8$ K for the junction of Fig. 1 (full line) and for a Y123 based junction (dashed line). When subtracted from the data, the Y-Ba-Cu-O energy gap

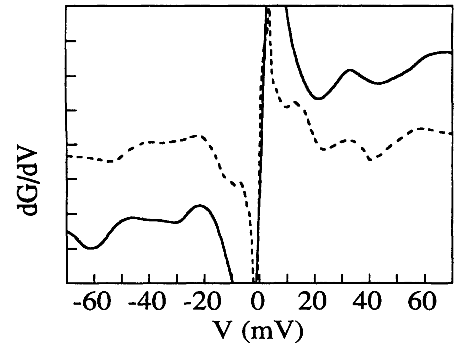

FIG. 4. Conductance derivative (arb. units) vs voltage at $T=8$ K for a Pb/Y124 junction (full line), and for a Pb/Y123 junction (dashed line).

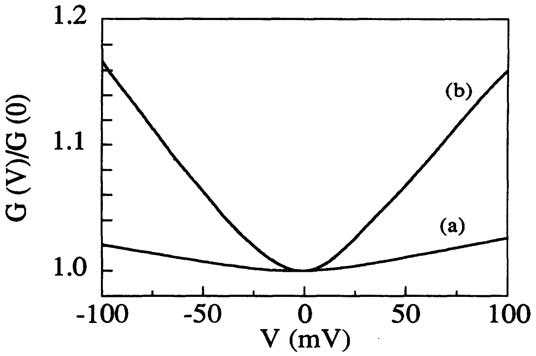

FIG. 5. Normal state conductance vs voltage for a Pb/Y124 junction, curve (a), at $T=90$ K and for a Pb/Y123 junction, curve (b) at $T=100$ K.

$\Delta \approx 18$ meV, both curves, with a lower resolution, reasonably reproduce the main features of the Y123 low-frequency excitation spectra reported in Refs. 12 and 13.

The similarity of the conductance derivatives in Fig. 4 indicates that the superconducting energy gap as well as higher-energy structures are similar in the two Y-Ba-Cu-O systems. In agreement with other experiments which show that superconductivity is mainly confined in the CuO_2 planes, our data seem to confirm that the in-plane properties undergo irrelevant modification in these compounds. As we already mentioned, the only relevant difference in the curves of Fig. 4 remains the low-energy structure present at about ±5 mV that, among cuprate superconductors, is peculiar of the Y123 compound.

Many authors have related this feature to a superconducting energy gap in the planes of the chain structures,[10,11,14] the metallic character of the chains being proved by optical and transport measurements.[15,16] If the chain superconductivity was intrinsic, an enhancement of the low-energy structure should be expected in the Y124 material. Disappearance of this feature is consistent with normal metal chains. In the Y123 material with a single CuO layer, chain superconductivity is induced from the CuO_2 adjacent planes by a kind of internal proximity effect.[11,14] The extremely short value of the c-axis coherence length, makes this effect disappear (or to be dramatically reduced) in the Y124 system. A reduced strength of the interlayer coupling in this material can then be assumed.

To further investigate this hypothesis, we have compared the normal state properties of both Y124 and Y123. In Fig. 5 typical normal state conductances are reported. The curves have been normalized at $V=0$ mV and the measurements have been carried out at $T=90$ K, for the Y124 system, curve (a), and at $T=100$ K, for the 90-K phase, curve (b). These data sets are typical of the respective compounds and junction variations in the same compound are much smaller than the differences measured between the Y124 and Y123 systems. We have found that the conductance slopes are quite insensitive to temperature variations in the range 4.2–300 K. Figure 5 indicates that linearity is preserved in both materials and a slower variation in energy of the Y124 normal conductance is observed.

Anomalous HTS normal state properties are the object of intense debate in literature, and particular attention has been devoted to the analysis of the tunneling linear conductance data. There have been several theoretical attempts to understand this behavior in terms of either elementary excitations in the material[17,18] or inelastic tunneling processes in the barrier.[19]

Recently, a detailed experimental study in the rather low T_c lead bismuth oxide superconductors has been reported and it has been demonstrated that the slope of the linear background is proportional to T_c in those compounds.[20] It has also been suggested that the value of the slope is a measure of the strength of the coupling mechanism responsible for superconductivity.[20] Our results seem to confirm that also in layered cuprate Y-Ba-Cu-O superconductors, a steeper slope is found in the higher-T_c compound while we speculate that slopes can give a measure of the interlayer coupling in these materials.

From the analysis of the above results, it is clear that both intralayer and interlayer coupling have to be taken into account to exhaustively describe the superconducting properties of layered high-T_c materials. The present study suggests that the in-plane superconducting properties remain unaltered in Y124 and Y123, while effects of a decreased strength of the interlayer coupling are observed in the Y124 system. As for the bismuth oxides, normal state properties seem to confirm a close relation between the T_c value and the slope of the linear background conductance and we hope that our investigation can stimulate a more quantitative analysis of this topic in the layered cuprate superconductors.

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[1] H. Zhang and H. Sato, Phys. Rev. Lett. **70**, 1697 (1993).
[2] Q. Li *et al.*, Phys. Rev. Lett. **64**, 3086 (1990).
[3] T. Terashima *et al.*, Phys. Rev. Lett. **67**, 1362 (1991).
[4] S. L. Cooper *et al.*, Phys. Rev. Lett. **70**, 1533 (1993).

[5] B. Dabrowski *et al.*, Physica C **202**, 271 (1992).
[6] J. Karpinski *et al.*, Nature (London) **336**, 660 (1988).
[7] R. J. Cava *et al.*, Nature (London) **338**, 328 (1989).
[8] A. M. Cucolo, Int. J. Mod. Phys. B **7**, 2549 (1993).
[9] A. M. Cucolo *et al.*, Physica C **207**, 21 (1993).
[10] M. Gurvitch *et al.*, Phys. Rev. Lett. **63**, 1008 (1989); Phys. Rev. B **44**, 11 986 (1991).
[11] A. M. Cucolo *et al.*, Phys. Rev. B **46**, 5864 (1992).
[12] J. J. Rhyne *et al.*, Phys. Rev. B **36**, 2294 (1987).
[13] V. M. Svistunov *et al.*, Fiz. Tverd. Tela (Leningrad) **30**, 3515 (1988) [Sov. Phys. Solid State **30**, 2022 (1988)].
[14] S. Takahashi and M. Tachiki, Physica C **170**, 505 (1990).
[15] Z. Sclesinger *et al.*, Phys. Rev. Lett. **65**, 801 (1990).
[16] T. A. Friedmann *et al.*, Phys. Rev. B **42**, 62 171 (1990).
[17] P. W. Anderson and Z. Zou, Phys. Rev. Lett. **60**, 132 (1988).
[18] C. M. Varma *et al.*, Phys. Rev. Lett. **63**, 1996 (1989).
[19] J. R. Kirtley and D. J. Scalapino, Phys. Rev. Lett. **65**, 7981 (1990).
[20] F. Sharifi *et al.*, Phys. Rev. Lett. **67**, 509 (1991).