## Linear normal conductance in copper oxide tunnel junctions

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We have measured the tunneling spectra of a large number of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>-based junctions that show linear background conductances. When conservation of states between the superconducting and the normal characteristics is observed, we have found a linear dependence of the slope of the G(V) curves on the zero-bias conductance values  $G_0$ . Tunneling data on the Bi-Sr-Ca-Cu-O and La-Sr-Cu-O systems seem to confirm this behavior. We infer that linear background conductances reflect the normal density of states of the cuprate superconductors and we speculate that this behavior is due to the interlayer coupling mechanism between the CuO<sub>2</sub> planes.

Unlike conventional superconductors, the tunneling characteristics of the copper as well as of the bismuth oxidebased junctions often show conductance backgrounds that linearly increase in voltage for  $|V| \ge \Delta/e$ . The interpretation of such anomalous behavior presents some difficulties since the tunneling conductance depends on the density of states of the high- $T_c$  material, N(E), as well as, when the tunneling probability P(E) becomes energy dependent, on the barrier properties. A correct analysis of the linear behavior has to necessarily distinguish between the two contributions. Consequently, it is of primary importance to carefully check the junction quality in order to identify extrinsic, barrier-related effects. Unfortunately, this requirement is difficult to be achieved in high- $T_c$  superconductor (HTSC) -based junctions, and some ambiguity remains in this aspect.

There have been several theoretical attempts to explain the conductance linear behavior in terms of anomalous, normal density of states (DOS) and frequently the high-bias tunneling spectra have been interpreted in support of the resonant-valence-bond (RVB) model,<sup>1</sup> of the marginal Fermi-liquid behavior<sup>2</sup> or in terms of localized states in the superconductor.<sup>3</sup> On the other hand, the linear conductance can also result when charging effects of metallic inclusions take place in the barrier or when inelastic scattering from the magnetic moments in the cuprates occurs during the tunneling process.<sup>4</sup>

An experimental study of the systematics of the linear behavior has been carried out for the bismuth oxide superconductors,<sup>5</sup> in which high-quality tunnel junctions can be fabricated. In this class of compounds, a linear dependence of the slope of the linear background on the zero-bias conductance value seems to confirm the hypothesis of an anomalous, normal DOS. Besides, in a large range of  $T_c$  variations, a remarkable correlation between the  $T_c$  and the conductance slope has been observed.<sup>5</sup>

A similar investigation in perovskite cuprate superconductors can give rise to some ambiguity. Due to the extremely short and anisotropic coherence length, easy degradation of the surfaces, and the high degree of anisotropy, good quality tunnel junctions are difficult to achieve on these materials. An important issue is the occurrence of a reliable test to identify a "pure" tunneling process and to exclude "spurious" effects in the barrier. In this paper we show that conservation of states over the whole temperature range can be assumed to select the junction characteristics in which quasiparticle tunneling is the dominant conduction mechanism. With this constraint, we have extensively studied the behavior of more than 50 Y-Ba-Cu-O-based junctions and we have found a linear dependence of the conductance slope and the extrapolated conductance value at zero bias. We believe that this behavior is intrinsic in the DOS of the high- $T_c$  material. This idea is supported by the analysis of other authors' tunneling data in the Bi-Sr-Ca-Cu-O and La-Sr-Cu-O systems.

For pure tunneling processes, no interactions in the barrier have to take place. The thin insulating layer has to be homogeneous and continuous, and pin-hole and metallic shorts have to be avoided since they produce parallel, nontunneling conductance channels. In HTSC-based junctions, important checks can be carried out, by using a traditional, BCS superconducting counterelectrode. Observation of a well-defined, low-leakage energy gap of the low- $T_c$  material is then used as the criteria for junction quality.<sup>6</sup> However, one cannot exclude that "spurious" mechanisms are activated above the  $T_c$  of the conventional superconductor. For higher temperatures, conservation of states between the normal and the superconducting tunneling spectra, appears to be a reliable test to ensure that in the high- $T_c$  material the states are redistributed below  $T_c$ , giving rise to the superconducting energy gap.

Figure 1 shows two typical tunneling characteristics (full lines) at T=10 K and T=50 K measured in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>-Pb planar junctions obtained by exposing to the ambient atmosphere Y-Ba-Cu-O single crystals chemically etched in a 1% Br solution in methanol. The fabrication procedure of this type of junction has been reported elsewhere.<sup>7</sup> The junction of Fig. 1 showed at low temperatures well-defined Pb gap and phonon structures. From the *I*-*V* curve, we have quantified that subgap leakages were less than 0.5% at 1 K. We observe in Fig. 1 that the background conductances linearly increase with voltage over hundreds of millivolts and do not depend on temperature. This observation is typical of Y-Ba-Cu-O-based junctions<sup>7,8</sup> and has also been found in La-Sr-Cu-O (Ref. 9) and Bi-Sr-Ca-Cu-O (Refs. 10–15) tunnel structures.

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FIG. 1. Typical conductance vs voltage characteristics of a Y-Ba-Cu-O/Pb planar junction measured at T=10 K and T=50 K. The dashed lines have been calculated from expression (1). Inset: the sum rule for the superconducting (white) and normal (dashed) areas at the indicated temperatures for the same junction.

Unlike traditional materials, it is not possible to check the normal-state conductance at low temperatures by driving normal the HTSC by means of an externally applied magnetic field. However, the part of the spectra for  $|V| \ge \Delta/e$  is expected to be largely independent of whether the high- $T_c$  material is superconducting or not. Therefore the normal conductance can be estimated at any temperature by the expression

$$G(V) = \int_{-\infty}^{+\infty} (G'|E| + G_0) \left[ -\frac{\partial f(E+eV)}{\partial E} \right] dE, \quad (1)$$

where G' and  $G_0$  are, respectively, the measured high-bias conductance slope and the extrapolated conductance at zero bias. The Fermi function f(E) accounts for thermal excited quasiparticles, which contribution is observed at low energies ( $eV \approx k_B T$ ). The dashed lines in Fig. 1 have been calculated from expression (1) at T=10 K and T=50 K, respectively.

We have tested conservation of states in the whole temperature range by integrating over energy the measured conductance spectra and the inferred normal conductance characteristics. In the inset of Fig. 1 we show the histogram of the superconducting (white) and normal (dashed) areas below the curves at different temperatures for the same junction. We observe that the spread of the calculated areas is less than 2% in all the temperature range. This is an important test to exclude that barrier related effects are activated at a certain temperature.

In fact, spurious effects in the barrier, such as inelastic tunneling processes, are responsible for breaking of the Cooper pairs and result in a higher normal electron contribution to the conductance measured inside the gap structure. On the other hand, charging effects, such as Coulomb blockade, give rise to an artificially low conductance near zero bias which may simulate a complete energy gap even in the case of gapless superconductors.

To see whether the linear background is an intrinsic feature of the DOS and is not due to an energy-dependent tunnel probability, by following the fabrication procedure previously indicated, we have formed Y-Ba-Cu-O-based planar



FIG. 2. Slope of the linear conductance vs the zero-bias conductance for:  $YBa_2Cu_3O_7$  (dots),  $GdBa_2Cu_3O_7$  (open circles),  $YBa_2Cu_4O_8$  (open triangles) and Zn, Fe, and Al substituted junctions (open squares). The line through the data is a least-squares fit with a slope of 0.92. The triangle refers to a degraded Y-Ba-Cu-O/Pb junction.

junctions of different resistances by changing both the time of the air exposures and the counterelectrode materials. Pb, Au, and Bi thermally evaporated thin films have been used. Conservation of states with less than 2% variations has been considered as criterion to select reliable data.

The central result of our study is shown in Fig. 2 where the background slopes, G', versus the extrapolated zero-bias conductance values,  $G_0$ , for a large number of Y-Ba-Cu-O junctions are reported. Dots and open circles in Fig. 2 refer to  $T_c = 90$  K YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> and GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> junctions, respectively. The line through the data is a least-squares fit with a slope of 0.92. This fact strongly supports the idea that the linear background is intrinsic in the DOS and does not depend on the barrier properties. We observe that data from YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub> samples with  $T_c = 80$  K (open triangles) and from substituted YBa<sub>2</sub>(Cu<sub>x</sub> $M_{1-x}$ )O<sub>7</sub> junctions with M = A1, Fe, Zn and 75 K $\leq T_c \leq 90$  K (open squares) quite well accommodate on the same line.

The triangle in Fig. 2 refers to a "degraded" Y-Ba-Cu-O/Pb junction, for which a considerable smearing of the Pb superconducting features at low temperatures was observed. In this case the spread between the areas of the superconducting and the normal tunneling spectra was more than 10% and, as expected, the data deviate from the linear fitting.

To check the validity of this result in other HTSC's, we have considered La-Sr-Cu-O (Ref. 9) and Bi-Sr-Ca-Cu-O (Refs. 10-15) tunneling data in the literature which show linear backgrounds and undergo less than 5% variations of the normal and the superconducting areas below the tunneling characteristics at the reported temperatures. The results are shown in Fig. 3 where we have plotted the background slopes versus the extrapolated conductance at zero bias. Dots refer to 2212 Bi-Sr-Ca-Cu-O and open triangles to the La-Sr-Cu-O system. The least-squares fits (full lines) have a slope of 0.91 for the Bi-Sr-Ca-Cu-O and 0.81 for the La-Sr-Cu-O compound. The almost linear dependence of the two quantities is rather interesting since data were taken from different experiments by different techniques. Triangles in Fig. 3 refer to a La-Sr-Cu-O-based junction in which inelastic tunneling processes were clearly observed.<sup>4</sup> Once again, these data deviate from the linear fitting.



FIG. 3. Slope of the linear conductance vs the zero-bias conductance for La-Sr-Cu-O (open triangles) and Bi-Sr-Ca-Cu-O-based (dots) junctions. Data are reported from the literature (see references in the text). The lines through the data are the least-squares fits with slopes of 0.81 and 0.91, respectively. The triangles refer to a La-Sr-Cu-O-based junction in which inelastic tunneling processes were clearly observed.

The fact that for different HTSC's we have found  $G' = G_0/V^*$  seems to confirm the anomaly of the normal DOS in these compounds. We observe that  $V^*$  values for the three superconducting oxides lie in a factor of 2 or 3. In the RVB model,<sup>1</sup>  $eV^*$  is proportional to the density of holons and in the marginal Fermi liquid<sup>2</sup> represents the cutoff in the excitation spectrum. It is therefore likely that this term has origin in the electron correlations.

To conclude, we mention that in some cases constant background conductances in the tunneling spectra of HTSC's have been found. This feature is more commonly measured in Bi-Sr-Ca-Cu-O point-contact junctions,<sup>16,17</sup> and, recently, has been observed in highly coherent Y-Ba-Cu-O/ Pr-Ba-Cu-O/Y-Ba-Cu-O (Ref. 18) trilayer tunnel structures. It is our opinion that both the linear and the constant behaviors are intrinsic in the HTSC's and are related to the interlayer and the intralayer properties, respectively.

The most essential structural unit common to all cuprate superconductors are the CuO<sub>2</sub> planes which are believed to be responsible for superconductivity. The large majority of the in-plane superconducting properties of these materials are well understood and appear rather conventional. "BCS-like" tunneling spectra with constant backgrounds could be related to CuO<sub>2</sub> planes that in the majority of the cases have been directly probed by point-contact junctions. The Bi-Sr-Ca-Cu-O system, for which flat backgrounds have been more frequently measured, is considered the more bidimensional among high- $T_c$  superconductors with well-separated CuO<sub>2</sub> planes.

On the other hand, the mechanism of the  $\text{CuO}_2$  interlayer coupling remains unresolved. The *c*-axis charge transportation involves different structural units, which show a metallic or semiconducting character, as the CuO chains in the Y-Ba-Cu-O compound. When surface preparation is difficult to achieve, as in fabricating HTSC planar tunnel junctions, below  $T_c$ , the normal electron (*c*-axis) contribution is often measured, that superimposes to the superconducting (inplane) features coming from Cooper pairs.

In summary, we have performed an extensive tunneling study of the linear behavior of the normal conductance in Y-Ba-Cu-O-based junctions with different resistances. When the number of superconducting and normal states is preserved, we have found a linear dependence for the zero-bias conductance value and the slope of the linear background conductance. We have observed a similar relationship in a large number of Bi-Sr-Ca-Cu-O and La-Sr-Cu-O tunneling data reported in the literature. We speculate that this behavior is indicative of a linear, normal DOS due to the interlayer charge transportation mechanism intrinsic in the HTSC's.

- <sup>1</sup>P. W. Anderson and Z. Zou, Phys. Rev. Lett. 60, 132 (1988).
- <sup>2</sup>C. M. Varma, P. B. Littlewood, S. Schmitt-Rink, E. Abrams, and A. E. Ruckenstein, Phys. Rev. Lett. 63, 1996 (1989).
- <sup>3</sup>J. C. Phillips, Phys. Rev. Lett. **59**, 1856 (1987).
- <sup>4</sup>J. R. Kirtley and D. J. Scalapino, Phys. Rev. Lett. **65**, 798 (1990).
- <sup>5</sup>F. Sharifi, A. Pargellis, and R. C. Dynes, Phys. Rev. Lett. **67**, 509 (1991).
- <sup>6</sup>A. M. Cucolo, Int. J. Mod. Phys. B 7, 2549 (1993).
- <sup>7</sup> M. Gurvitch, J. M. Valles, Jr., A. M. Cucolo, R. C. Dynes, J. P. Garno, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **63**, 1008 (1989); J. M. Valles, Jr., R. C. Dynes, A. M. Cucolo, M. Gurvitch, L. F. Schneemeyer, J. P. Garno, and J. V. Waszczak, Phys. Rev. B **44**, 11 986 (1991).
- <sup>8</sup>H. L. Edwards, D. J. Derro, A. L. Barr, J. T. Markert, and A. L. de Lozanne, Phys. Rev. Lett. **75**, 1387 (1995).
- <sup>9</sup>T. Ekino, Physica C **205**, 338 (1993).

- <sup>10</sup>H. Tao, A. Chang, Farun Lu, and E. L. Wolf, Phys. Rev. B 45, 10 622 (1992).
- <sup>11</sup>J. J. Wnuk, R. T. M. Smokers, F. W. Nolden, L. W. M. Schreurs, Y. S. Wang, and H. van Kempen, Supercond. Sci. Technol. 4, S412 (1991).
- <sup>12</sup>S. Fujta, K. Nakao, T. Sugimoto, K. Uehara, and Y. Shiohara, Physica C **199**, 135 (1992).
- <sup>13</sup>T. Ekino and J. Akimitsu, Phys. Rev. B **40**, 6902 (1989).
- <sup>14</sup>A. Placenik, M. Grajcar, S. Benacka, P. Seidel, and A. Pfuch, Phys. Rev. B **49**, 10016 (1994).
- <sup>15</sup>A. M. Cucolo, R. Di Leo, A. Nigro, P. Romano, and M. Carotenuto, Phys. Rev. B **49**, 1308 (1994).
- <sup>16</sup>J. F. Zasadzinski, N. Tralshawala, P. Romano, Q. Huang, J. Chen, and K. E. Gray, J. Phys. Chem. Solids **53**, 1635 (1992).
- <sup>17</sup>Ch. Renner and Ø. Fischer, Phys. Rev. B **51**, 9208 (1995).
- <sup>18</sup>A. M. Cucolo, R. Di Leo, A. Nigro, P. Romano, F. Bobba, E. Bacca, and P. Prieto, Phys. Rev. Lett. **76**, 1920 (1996).