REVISTA COLOMBIANA DE FISICA, VOL. 35, No. 1, 2003

HETEROSTRUCTURES BASED ON HIGH-TEMPERATURE SUPERCONDUCTING THIN FILMS

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ABSTRACT

For many possible electronic applications as well as fundamental studies it is essential to fabricate epitaxial layered structures of insulators, semiconductors or normal metals together with high-temperature superconductors (HTS). However HTS materials have complex lattice structures and this makes it difficult to grow multilayers with sharp interfaces preserving epitaxiallity through the whole structure. In this work we describe transport measurements and microstructural analysis of $YBa_2Cu_3O_{7-\delta}$ / $PrBa_2Cu_3O_{7-\delta}$ / $YBa_2Cu_3O_{7-\delta}$ (YBCO / PBCO / YBCO) and $Bi_2Sr_2CaCu_2O_{8+\delta}$ / $Bi_2Sr_2YCu_2O_{8+\delta}$ / $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO / BSYCO / BSCCO) heterostructures deposited on (001) $SrTiO_3$ substrates by using an in situ DC sputtering technique at high oxygen pressures. Conductance measurements on this type of multilayers showed a clear quasiparticle tunneling indicating a gap structure around 25 mV in the case of YBCO compounds, 30 – 35 mV and a zero bias anomaly in the case of BSCCO materials. We will discuss also the Josephson behavior of heterostructures based on BSCCO compounds.

1. Introduction

Several electronic devices and sensors are based on planar heterostructures, i.e., multiple planar layers involving HTS and other non-superconducting materials. Planar type heterostructures are important since their method of production is compatible to the one used to produce semiconducting devices and, therefore, they may be incorporated in the production of superconducting / semiconducting mixed devices in a natural way. These devices may be produced from sandwich type structures where there are two electrodes separated by an intermediate layer that usually is not a good conductor. There are several types of planar junctions which, depending on the nature of the electrodes and intermediate materials, are called *SIS* (Superconductor / Insulator / Superconductor), *SNS* (*N* stands for Normal), *S*/*S**/*S* (*S** stands for Semiconductor), *SIN*, etc. These sandwich type tunnel junctions are very important, not only because they are natural precursor to SQUIDs and other similar practical devices, but also because tunneling measurements in such structures may provide information about the superconducting properties of the electrodes which form the junctions. Heterostructures and other types of junctions based on YBCO [1,2] and BSCCO [3,4] have been studied. The key for producing good junctions is the nature and quality of the intermediate layer and particularly, the quality of the interfaces that are formed with the electrode materials. An ideal barrier material will be one that, apart from being a good insulator and pinhole free, has a close matching of lattice parameters and similar composition to the HTS electrode to be investigated. The first condition is necessary for a stress free epitaxial growth of the top electrode and sharpness of both interfaces. The second condition is necessary to assure that foreign atoms may not interdifusse through the interface and degrade the surface of the superconducting electrodes. It is also desirable that the barrier has a similar thermal expansion coefficient.

The differential tunneling conductance vs. voltage of a tunnel junction is directly related to the density of states and provides accurate measurements of the energy gap and the electron-phonon coupling in conventional superconductors. There are no fundamental reasons why tunneling cannot provide similar information for HTS. In all cases the interface should be very clean, with composition and structure homogeneous and very sharp (within one coherence length, which may be extremely small for HTS) since tunneling is very sensitive to surface properties. Therefore, producing a good heterostructure with HTS electrodes is a formidable challenge to experimentalists. Different kinds of all-oxide *SNS* structures, including grain boundary, step edge, ramp-type, and sandwich-type junctions, have been realized by several groups for HTS device applications. In particular, YBCO / PBCO / YBCO junctions have been investigated in detail by using *a*-axis oriented [5]. In contrast with the excellent results obtained in Josephson coupled *SNS* trilayers, clear evidence for quasiparticle tunneling is difficult to obtain in all-oxide *SIS* junctions. Several attempts to realize HTS-based *SIS* structures have been reported in the literature. However, in all artificial structures, due to the difficulty of realization of good quality interfaces between the insulating barrier and the counter electrodes, quasiparticle tunneling shows a *SIN* behavior with gap-like features developed well below the measured T_c of the HTS thin 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 YBCO-based junctions with conventional superconducting counter electrodes [6] but a “complete” energy gap for the YBCO compound has not been observed. In this work we report on the successful growth of YBCO / PBCO / YBCO and heterostructures involving HTS electrodes of BSCCO with intermediate layers of BSYCO which are precursors to well behaved SQUIDs and other practical devices showing clear evidence for *SIS* quasiparticle tunneling and Josephson behavior.

2. Experimental

Superconducting, YBCO and BSCCO, and semiconducting, PBCO and BSYCO, thin films were deposited *in situ* using a high pressure DC-sputtering process. Details of this technique have been previously described [7], therefore only a short description will be given. Stoichiometry sintered YBCO, PBCO, BSCCO, and BSYCO sputtering targets of 35 mm diameter and $1\times10\times10$ mm^3 substrates of single crystals (001) $SrTiO_3$ were utilized. Pure oxygen at a pressure of 3.5 mbar was used as sputtering gas; this relatively high pressure avoids etching of the films by negatively charged oxygen ions. The substrate temperature was held constant during deposition at 850 ± 1ºC. Deposition rates, as determined by Transmission Electron Microscopy (TEM) and Rutherford Backscattering (RBS), were approximately 100 nm/h for all films. Film thicknesses were controlled by measuring the deposition time at constant sputtering rate. This deposition procedure consistently produces *c*-oriented epitaxial YBCO, PBCO, BSCCO, and BSYCO thin films with smooth and very flat surfaces as probed by several analytical techniques. Trilayers and multilayers were prepared *in situ* by sequential sputtering of the films, each of them under similar conditions. The thickness of the films was approximately 100 nm for the electrode films while the barrier thickness varied between 4 and 12 nm. Interface microstructure was determined by cross sectional lattice resolution TEM. It was observed that interfaces were very sharp and cleans with no interdifussion between layers as shown in the inset of Figs. 1-2; the intermediate film was pinhole free. RBS and channeling measurements with 2.0 MeV He ions were carried out in order to examine composition and crystalline quality of the heterostructures; as an example the RBS spectra for a trilayer BSCCO / BSYCO /

BSCCO is shown in Fig. 1. Since the back edges of the contribution to the spectrum from the different elements are quite steep, it is concluded that the interfaces are fairly sharp, that is, no smearing from interdifussion is observed. Alignment of the substrate such that the direction of the incident He beam is perpendicular to the surface results in a strong reduction of the back scattering yields; supporting the epitaxial quality of the heterostructures. Standard four probe resistivity measurements were performed. Resistivity measurements of a typical YBCO and PBCO films are shown in Fig. 2. Patterning was made photolithographically by wet chemical etching, from which 100 μm strips and 100×100 μm^2 junctions were obtained as shown in the inset of Fig. 2. Electrical resistivity of the heterostructures was made on strips and tunneling on cross type junctions. The transition temperatures of the superconducting films were not affected by the patterning process.

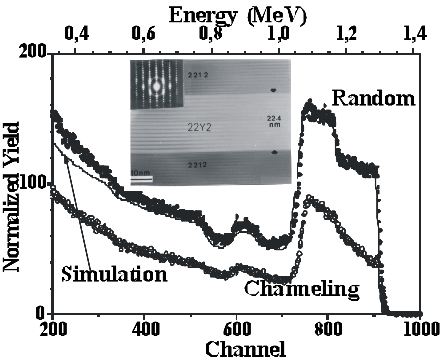

Fig. 1. RBS spectrum under random incidence and channeling condition for a trilayer BSCCO / BSYCO / BSCCO heterostructure. The inset shows a high-resolution cross-section TEM micrograph of a trilayer structure.

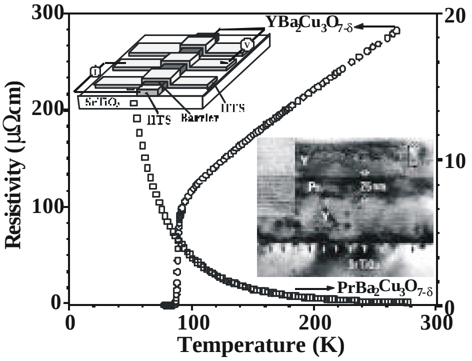

Fig. 2. Resistivity as function of temperature for a HTS YBCO film and insulating PBCO. The right hand inset shows high-resolution cross-section TEM micrograph (note the atomic sharp interfaces). The left hand inset shows the geometry of the trilayer structure after photolithography.

3. Quasiparticle tunneling in HTS heterostructures

In Fig. 3 the *I*-*V* and the d*I*/d*V* vs. *V* characteristics of an YBCO / PBCO / YBCO junction are showed at $T = 4.2$ K. Well defined maximum 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 for the all set of samples. Low zero-bias conductance values are rarely measured in planar HTS junctions. For these kind of structures, 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 [8]. 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. 3 is well related to the low measured value of the conductance at zero voltage. We notice that a certain asymmetry is observed that might reflect an asymmetry of the tunnel barrier due to not perfectly equivalent interfaces. Moreover, extrapolation to zero current of the Ohmic *I*-*V* curve of Fig. 3 seems to indicate the presence of Coulomb blockade.

The temperature evolution of *G*(0) normalized by the value of the conductance at *V* = 150 mV for Bi-based trilayers is shown in Fig. 4. *G*(0) is extracted from fitting the background conductance without considering the peak at zero bias as shown in the inset of Fig. 4. The value of *G*(0)/*G*(150 mV) starts to increase slowly and then shows two discontinuities: at 60 K a sudden increase starts to develop until T_c is reached, then it levels off at a constant value close to 1. This behavior is expected for superconducting electrodes. The *low voltage range* (voltages $|V| < 5$ mV) is characterized by a zero bias anomaly (ZBA) conductance peak; for a given temperature, the conductance decreases rapidly as bias departs from zero. The height of this peak increases as the temperature decreases. ZBAs in the tunneling conductance were already observed in metallic tunnel junctions with an oxide barrier and in a variety of measurements of tunneling junctions made of HTS, as well as conventional superconductors and semiconductor electrodes. Most of these ZBAs could be described by a theoretical model from Appelbaum [9] and Anderson [10], where a spin-spin interaction of the localized magnetic states with the quasiparticles opens up an additional tunnel channel increasing the conductance near zero bias.

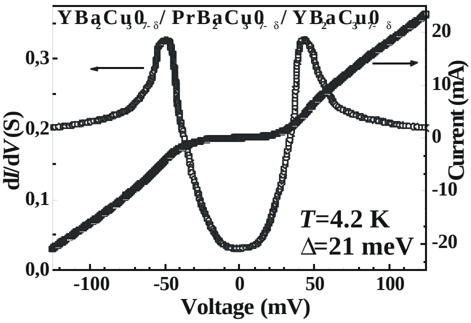

Fig. 3. Current-voltage and conductance-voltage characteristics of an YBCO / PBCO / YBCO junction.

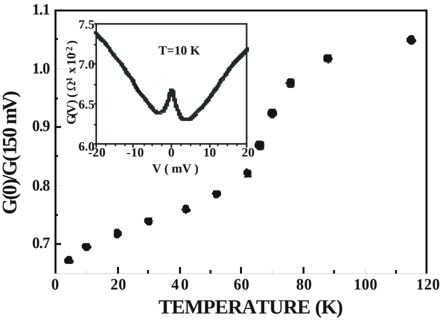

Fig. 4. Normalized conductance at zero bias vs temperature for a Bi-based trilayer. A parabolic fit (dotted line) close to zero voltage is shown in the inset, from which *G*(0) is calculated.

4. Josephson like tunneling

For heterostructures with atomic sharp *S*/*N* or *S*/*I* interface a Josephson-like tunneling can appear. To observe this effect, we have fabricated step stack Josephson junctions using bilayers of BSCCO / BSYCO epitaxial thin films as detailed described in Ref. [11]. For these heterostructures an intrinsic Josephson effect was measured. Fig. 5 shows a current-voltage (*I*-*V*) characteristic for a typical BSCCO / BSYCO step-stack junction whose structure is displayed in the inset of Fig. 5. A hysteretic behavior was clearly observed, the voltage first appeared just above the critical current I_c due probably to a flux-flow mechanism, and was followed by a large jump up to the resistive state. These *I-V* characteristics can be explained in terms of the intrinsic Josephson effect [12].

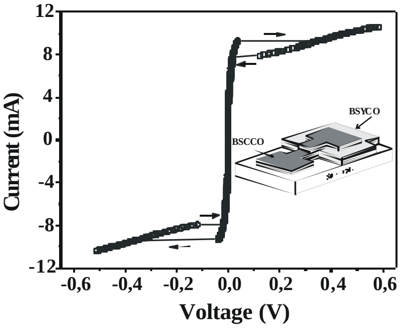

Fig. 5. *I-V* characteristics for a typical BSCCO / BSYCO step stack junction. A clear hysteretic behavior like *SIS* Josephson junction is observed. The right hand inset shows the structure of the stack-step junction and the left hand inset shows the AFM image of one BSYCO step.

5. Conclusions

In summary, we have measured an almost complete energy gap in the density of states of the YBCO compounds that have been traditionally considered the more gapless among the HTS. The conductance curves measured on BSCCO / BSYCO / BSCCO junctions show quasiparticle tunneling current with a clear gap-like structure around 30 meV. A linear conductance background above the superconducting gap structure and a ZBA, that showed strong temperature dependence understood in terms of the presence of Cu paramagnetic ions at the interface, are also observed. These junctions may also show weak-link behavior if the barrier is thin enough. One of the most striking features of this work is that the results have been obtained in all-oxide heterostructures in which *SIS* tunneling spectra are extremely difficult to achieve. Our results are very encouraged for future development of trilayer structures for HTS device applications.

Acknowledgments. This work was supported by COLCIENCIAS. The authors wish to thank K. Urban, U. Poppe, H. Soltner from IFF/IMF FZ-Jülich, Germany and A. Cucolo from University of Salerno, Italy, for support and stimulating discussions.

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