Preparation of Continually Graded Barriers of YPrBaCuO for HTS Josephson Junctions

J. Gao, J.L. Sun, and W.H. Tang

***Abstract*—We report preparation of a novel barrier structure for high T_c superconducting multilayer Josephson junctions using a simple composite target technique. Such a barrier consists of $Y_xPr_{1-x}Ba_2Cu_3O_y$ with a continually varied concentration of yttrium. In this barrier no lattice mismatch and other incompatibility problems occur between adjacent layers. Thus the formation of interfacial defects and structural strain can be mostly prevented. The Josephson coupling takes place at the electrically formed interfaces rather than the structural interfaces. A particular feature of these junctions is that the effective thickness of the barrier strongly depends on the measuring temperature and the concentration gradient. The absence of the structural interface in the weak link region greatly enhances the reproducibility and the performance of these junctions.**

***Index Terms*—High Temperature Superconductors, Josephson Junctions**

I. INTRODUCTION

One of the major problems associated with high T_c Josephson junctions using an artificial barrier is the poor interface between the barrier and superconductor. It is believed that the interfaces between barrier and superconducting electrodes play a crucial role.[1-4] In general the interface between the barrier and superconductors is damaged and defective, which results in poor performance and reproducibility. Studies using a high resolution transmission electron microscope (HRTEM) showed that the structural, thermal, and chemical incompatibility at the interface could introduce various defects.[5-6] Also, the *ex situ* lithography process involved by either ion-milling or chemical etching further damage the interface.[1,7-9] To reduce the incompatibility between barrier and electrode, and to improve the damaged interface, various approaches like chemical etching and use of intermediate layers were reported.[7,9] However, the interfacial problems still remain although the quality of the interfaces or the lattice mismatch between barrier and superconductor could be improved. The reproducibility and performance of high T_c multilayer junctions are still not satisfactory.

In this paper, we describe the preparation of a new barrier structure with a continually graded composition of $Y_xPr_{1-x}Ba_2Cu_3O_y$ ($Y_xPr_{1-x}BCO$). High T_c Josephson junctions with such a graded barrier structure have no structural interfaces and the Josephson coupling takes place at the electronically formed interfaces. In addition, the effective barrier thickness in our junctions, which depends on the temperature and concentration gradient, can be varied. Thus the reproducibility and uniformity of these junctions are greatly enhanced.

II. PREPARATION OF THE GRADED BARRIER AND JUNCTIONS

Our junctions were fabricated on $SrTiO_3$ (STO) substrates by using off-axis RF magnetron sputtering. First, an epitaxial bi-layer consisting of the base $YBa_2Cu_3O_y$ (YBCO) electrode and the insulating layer of $PrBa_2Cu_3O_y$ (PBCO) was grown in situ using off-axis sputtering. Photoresist was then spun onto the bi-layer films as the stencil and was baked at 100 °C for 10 min. The structure was patterned by a standard lithographic process. To create a ramp edge, the samples with patterned resist were ion milled through the bi-layer and slightly into the substrate by using a Kaufinann Ar ion beam gun of 5 cm in diameter. The ion milling was performed under a pressure of (5-8)$\times 10^{-4}$ mbar, and the beam current and voltage were I = 15 mA and V = 600-800 V, respectively. To prevent overheating of the sample, the ion gun was operated in a pulse mode, which allowed a short break every 10 seconds. After the ramp edge being made, the sample was placed back into the vacuum chamber and the barrier and top electrode were then grown.

To overcome the problem caused by the damaged interface, $Y_xPr_{1-x}BCO$ was used as the barrier with the concentration x gradually changing from 1 to 0 and then from 0 to 1, as shown in Fig. 1. We first used two targets co-sputtering to make such a continually graded barrier of $Y_xPr_{1-x}BCO$. By controlling the sputter power of YBCO and PBCO targets, a continuous composition gradient could be formed. The drawbacks of this method are that the gradient of concentration x is not very uniform and the deposition system is very complicated. Moreover, it is quite difficult to control the process to obtain a smoothly changed composition in the grown barrier. Therefore we developed a composite target technique for preparing the graded barrier. A schematic diagram of our deposition system is shown in Fig. 2. The composite target consists of two semi-discs, YBCO and PBCO, respectively. It was made

Manuscript received September 16, 2000. This work has been supported by the Research Grants Council (RGC) of Hong Kong.

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simply by placing YBCO and PBCO powder in the mould, each occupying a half volume, and then pressing into a disc. It is known that the YBCO and PBCO can be sputtered under the same conditions with a similar rate. At room temperature the resistance of YBCO and PBCO are in the same order and the sputtering was operated in RF-mode. Thus the sputter power could be divided more or less equally into the two sites of the composite target, as can be seen from the shape of the sputter plasma. During the growth of barrier, the substrate was slowly rotated from the YBCO side to the PBCO side and then to the PBCO target to make a pure PBCO central layer. The reason that we insert a PBCO central layer between two graded $Y_xPr_{1-x}BCO$ layers is to ensure that no short circuits can take place even at low temperatures. After the PBCO barrier being deposited, the substrate is rotated back to the compound YBCO/PBCO target and gradually moved from PBCO side to YBCO side, and then to the YBCO target to grow the top electrode. By using such a simple technique, a smooth changing of the composition from YBCO to PBCO is obtained. A continually graded barrier without any abrupt changes in structure and composition is therefore formed.

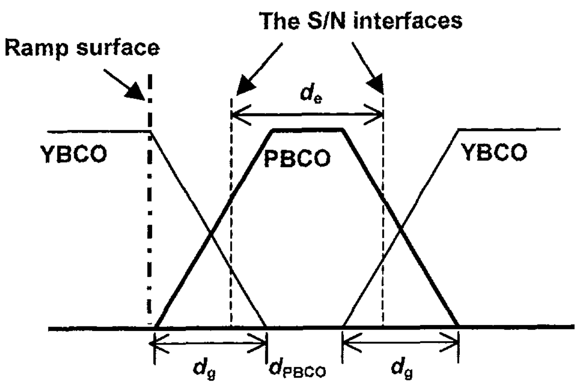

Fig.1 Schematic diagram of the continually graded barrier of $Y_xPr_{1-x}Ba_2Cu_3O_7$ with the concentration x gradually changing from 1 to 0 and then from 0 to 1. d_e is the effective barrier thickness.

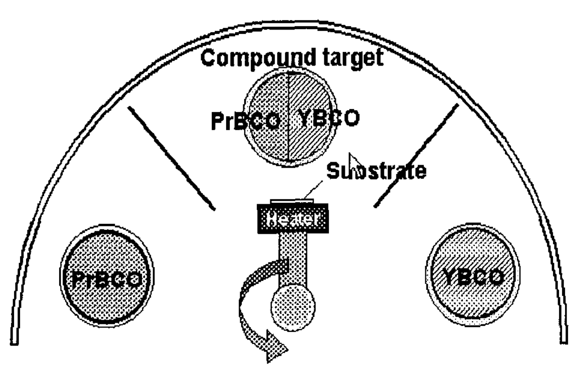

Fig2. Schematic diagram of our deposition system.

III. Results and Discussion

Fig. 3 presents the depth distribution of yttrium and praseodymium measured by x-ray photoelectron spectrometry (XPS). During the measurement, the film was sputtered through the top YBCO layer, the graded $Y_xPr_{1-x}BCO$ barrier, the bottom YBCO layer, and into the $SrTiO_3$ substrate. As can be seen in Fig. 3, the yttrium concentration x was gradually changed from $x=1$ to $x=0$ to reach a pure PBCO barrier, and then x was slowly increased from 0 to 1 to reach the YBCO electrode. Therefore the mismatching between every adjacent atomic layer would be very small and the occurrence of interface defects can be greatly reduced. Hence the reproducibility and performance of these junctions can be significantly enhanced. On the XPS spectrum the yttrium signal did not reach zero level at the center of barrier although there was a pure PBCO central layer in the barrier. The reason could be due to the penetration of the probe through the entire PBCO layer into the bottom YBCO layer as the probe depth in XPS is typically 10 nm.

In the ramp-type junctions using a conventional single PBCO barrier, the ramp surface created by ion milling is typically defective. The damaged interface can result in a large boundary resistance and low transparency for the quasi-particles at the interface. [9,10] Also, an abrupt change of lattice constant and composition at the interface can introduce strain and interfacial defects. [11] Thus the interface resistance rather than the barrier material could dominate the junction properties. In comparison, our junctions with a continuously graded barrier can mostly prevent the influence of the damaged ramp interface. It is known that the $Y_xPr_{1-x}BCO$ can be a superconductor at sufficiently low temperature, if the yttrium concentration x is larger than 0.5. The results obtained on bulk material of $Y_xPr_{1-x}BCO$ indicated that the transition temperature T_c decreased gradually as the concentration x decreased.[12]

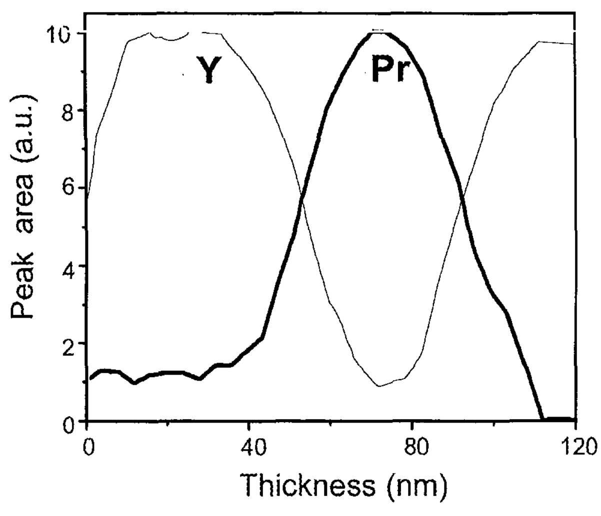

Fig.3 The depth distribution of Y and Pr measured by XPS.

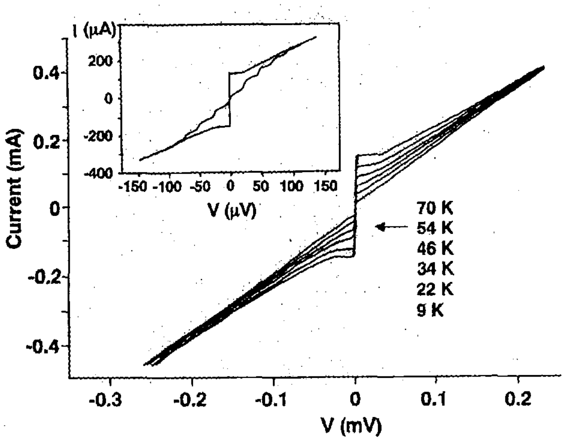

Fig. 4 I-V curves for junction with a $Y_xP_{1-x}BCO$ barrier at different temperatures (d_g= 30 nm and d_{PBCO}=4 nm). The inset shows the I-V curves with and without microwave radiation at 9 K.

Depending on the concentration x and T, the $Y_xPr_{1-x}BCO$ layer is partially superconductive. Therefore the S/I interface will be formed inside the $Y_xPr_{1-x}BCO$ layer and the damaged ramp surface is excluded from the weak link region, as shown in Fig. 1. Since the concentration x varies very smoothly there will be no abrupt change in the lattice constant and the composition. Hence no structural interface is formed within the weak link region. To assess the influence of the damaged ramp surface, which now is located within the superconducting electrode, we deposited an YBCO layer on the damaged ramp surface. It was found that the film still showed a full transition with a high T_c. We conclude that the ramp surface within the superconducting region has little effect on the performance of the junctions.

The junctions with such a graded barrier present resistively shunted junction (RSJ) characteristics. I-V curves have small excess currents (see Fig.4). Almost all of these junctions exhibited clear Shapiro steps under irradiation of a microwave field, giving unambiguous evidence of the Josephson behavior. The junctions discussed in this paper were structured into a width of either 10 or 20 μm. The normal state resistance near the transition temperature in these junctions is typically from 10^{-1} to a few Ohm. The characteristic voltages, I_cR_n, measured at 10 K are around 1 mV.

Using such a continuously graded barrier structure, the yield of successful junctions, which show a supercurrent and clear Shapiro steps, is nearly 100% (Among 40 junctions made so far, 38 exhibited clear Shapiro steps above 50 K.). In contrast, the yield of good junctions with a single barrier is typically below 50%. The uniformity of junction parameters, known as an essential issue for applications, was considered too. Damaged interfaces often cause variation in the junction properties like I_c and R_n, *etc.* In most high T_c junctions with an artificial barrier, uniformity has been influenced by the damaged interfaces leading to large variation. By using this barrier, the damaged interface is excluded from the weak link region so it can minimally affect junction performance. In our junction process, I_c variation as low as 1-σ=12% for 10 junctions with identical parameters was found at 60-65 K.

A particular feature of such a barrier is that the effective barrier thickness d_e, which is the spacing between the two S/N interfaces (see Fig. 1), could be varied even after fabrication. In our junction the barrier consists of three layers, the graded $Y_xPr_{1-x}BCO$ interlayer, PBCO layer, and another graded interlayer. The graded intermediate layers with thickness d_g can be partially superconducting depending on the temperature and the concentration x. For most samples studied in this work, the thickness of the PBCO central layer is d_{PBCO}=8 nm, and the thickness of the graded layer at the two sides was varied from d_g=6 nm to d_g=38 nm. The effective barrier thickness should be the sum of d_{PBCO} and the thickness of superconducting part within $Y_xPr_{1-x}BCO$, which is a function of the temperature and the concentration gradient. It follows that the barrier thickness in such junctions would strongly depend on the measuring temperature.

Since the d_e varies as a function of temperature, it is therefore of great interest to study the temperature dependence of the transport properties in these junctions. The normal state resistance R_nA (A is the junction area) is plotted in Fig. 5 against temperature. We found that for small values of d_g the R_nA products were quite insensitive to the temperature. It is known that PBCO is a semiconductor, whereas $Y_xPr_{1-x}BCO$ can behave as a normal metal, depending on the concentration x and the temperature. As the effective barrier thickness is also changed, the whole barrier presents a complicated system. The temperature insensitive R_nA product is a result of the combination of $Y_xPr_{1-x}BCO$ and PBCO layers. On the other hand, with a thicker $Y_xPr_{1-x}BCO$ layer R_nA is dominated by the PBCO material, which usually shows a semiconductor behavior.

The temperature dependence of I_c of these junctions was also measured and compared with that of junctions with a conventional single barrier of PBCO. Fig. 6 shows the normalized I_c as a function of reduced temperature T/T_c for junctions with various d_g (from 10 nm to 38 nm). Most of the data lie on a nearly straight line, approach a $(1-T/T_c)^1$ relationship. In contrast the I_c of junctions using a single PBCO barrier usually exhibits a $(1-T/T_c)^2$ dependence of a SNS-type

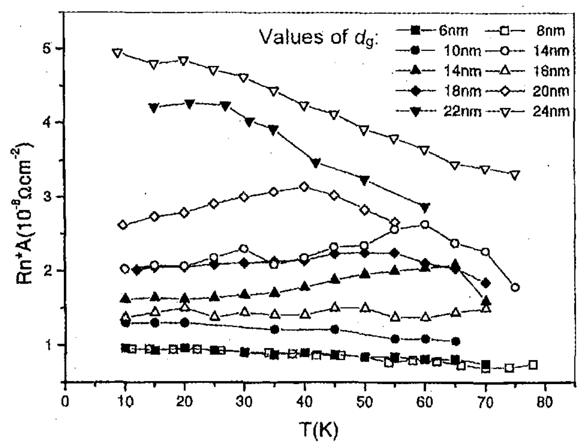

Fig. 5 The temperature dependence of the product R_nA for junctions with different values of d_g.

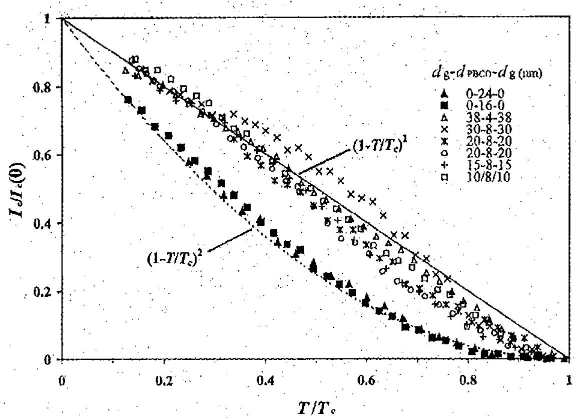

Fig. 6 The normalized I_c vs. T/T_c for various d_g. The solid symbols are data measured on the junctions with a conventional single barrier of PBCO.

junction. [1, 8] The effective barrier thickness for junction (10-8-10) is nearly the same as that with a single barrier (0-16-0), but their $I_c(T)$ dependences are far different. The power of two is characteristic of a SNS Junction. The critical current for a SNS structure can be described by [13]

$$I_c(T) = C\,|F_0(T)|^2[\xi_n(T)/\xi_{GL}(T)]^2 exp[-L/\xi_n(T)]$$

where C is a temperature dependent constant, ξ_{GL} the Ginzburg-Landau coherence length, ξ_n the effective coherence length in the normal metal, F_0 the amplitude of Cooper-pairs in electrodes and L the length of the normal metal path from one superconducting electrode to the other. As ξ_{GL} and $F_0(T)$ vary approximately as $(1-T/T_c)^{-1/2}$ and $(1-T/T_c)^{1/2}$ for $T/T_c \geq 0.5$ and ξ_n varies only slowly, the critical current I_c is proportional to $(1-T/T_c)^2$. The fact that the $I_c(T)$ of junctions with a continually graded barrier varies as $(1-T/T_c)^1$ implies that their transport process might differ from that of the SNS structure. A detailed study of the behavior of these junctions will be discussed elsewhere. [14]

IV. Conclusions

In summary, a new barrier structure with a continually graded $Y_xPr_{1-x}BCO$ has been prepared by using a simple composite target technique. In such a barrier structure the structural interfaces are absent and the Josephson coupling takes place at the electronically formed interfaces. The effective thickness of such a barrier strongly depends on the concentration gradient and temperature. The transport process in these junctions significantly differs from that of junctions with an ordinary single barrier layer and provides a very interesting system for further study. The absence of the structural interface in the weak link region can greatly help to enhance the reproducibility and performance of these junctions.

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