A potential method to correlate electrical properties and microstructure of a unique high-$T_c$ superconducting Josephson junction

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A method to correlate microstructure from cross-section transmission electron microscopy (TEM) investigations and transport properties of a single well characterized high-$T_c$ artificial grain boundary junction is reported. A YBa$_2$Cu$_3$O$_{7-\delta}$ 45° twist junction exhibiting the typical phenomenology of high-$T_c$ Josephson weak links was employed. The TEM sample preparation is based on focused ion beam etching and allows to easily localize the electron transparent area on a microbridge. The reported technique opens clear perspectives in the determination of the microstructural origin of variations in Josephson junction properties, such as the spread in $I_c$ and $I_c R_N$ values and the presence of different transport regimes in nominally identical junctions. © 1999 American Institute of Physics. [S0003-6951(99)03404-X]

A clear understanding of the correlation between the details of the fabrication process and the electrical properties of high-$T_c$ Josephson junctions (JJ) would be required for the implementation of an optimized and reliable manufacturing technology. Such a correlation is generally quite indirect and difficult to establish. A useful approach is to divide the problem into two parts: the influence of the fabrication on the microstructure and the influence of the microstructure on the transport properties. Microstructural characterization by transmission electron microscopy (TEM) plays a crucial role in this context. Correlation between transport and structural properties could be established in a few specific cases, as on [001] tilt artificial grain boundary (AGB) junctions$^1$ and on ramp-type junctions$^2$ in YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO). This correlation is particularly complex for AGB junctions where the role of the barrier is not played by the artificially deposited layer, but by an intrinsic region at the grain boundary interface, whose nature is not fully understood. The microstructure is determined by the growth modes of the two electrodes, and is affected by factors (such as nucleation and growth of single grains or impurity phase precipitates) which are hardly controllable by the fabrication process. Two nominally identical AGB junctions may therefore present significant differences in their microstructures. As a result, the interpretation of obtained transport data is always cumbersome if the microstructure of the electrically characterized junction is not exactly known.

In order to address some fundamental issues, as the spread in properties of nominally identical AGB JJ and the analysis of single junctions exhibiting peculiar attributes, junctions with previously characterized transport properties need to be analyzed by cross-sectional TEM (XTEM) to determine their specific microstructure. Such an approach presents a very high technical difficulty from the point of view of sample preparation, since two very strict conditions are required: (i) submicron localization of the electron transparent area on the microbridge; (ii) high yield, not to destroy the unique samples formerly characterized and chosen for the analysis.

The combination of these two requirements makes conventional cross-section TEM preparation technique inappropriate. Conventionally the JJs for TEM studies are fabricated in an identical way as JJs for physical measurements. The only difference being that final patterning, to define the junction area or microbridge, is omitted leaving the junction interface to continue across the whole length of the substrate. These samples allow investigation on any location across the substrate and therefore simplify the XTEM preparation considerably. Moreover substrates containing arrays of junctions are often used to enhance the success rate and to allow for some statistical approach in the TEM analysis.

In the present letter the microstructural characterization by XTEM of a single previously characterized AGB JJ is reported here. The proposed XTEM preparation method employs precision mechanical polishing$^3$ and focused ion beam (FIB) thinning$^4$ to create an electron transparent region at the desired location on the sample, namely the barrier region of a microbridge of which all junction properties have been previously measured.

The FIB system scans a finely focused gallium (Ga) ion beam across a sample to remove material by precision ion milling and can image the sample as well. There are well
established applications of FIB for microelectronics. New applications of FIB include precision cuts and trimming, even at oblique angles. FIB thinning can be used as well for XTEM sample preparation. Clear advantages of the FIB thinning over conventional Ar ion milling are the high spatial precision, the uniform thinning of materials, the large dimensions of the obtained electron transparent area, and the high yield.

The junctions employed in the present work are YBCO biepitaxial 45° [100] or [010] twist AGB JJ fabricated on a (110) SrTiO$_3$ (STO) substrate with a MgO seed layer. YBCO grows (103)/(013) oriented on (110) STO and (001) oriented on the seed layer. Further details about the properties of the junctions and the fabrication process have been reported elsewhere. The XTEM sample preparation can be summarized as follows. A strip is cut from the substrate wafer along the microbridge and thinned by precision mechanical polishing on both sides down to a thickness of 20 μm. The strip is glued to a copper washer and mounted in the FIB. Inside the FIB system a platinum (Pt) layer of 2 μm by 20 μm is deposited to protect the junction area. Lines of material are removed on both sides of the strip with the Ga beam at normal incidence to the film surface plane. At first a large spot size is used to remove material at a high rate, reducing the FIB process time. During the initial process the sample can be tilted inside the FIB to image the different layers of the structure. When approaching the area of interest the conditions are softened to reduce damage in that region. The milling is continued line by line until only a thin foil remains intact with an electron transparent thickness. The final sample, ready for XTEM, is schematically shown in Fig. 1. The position of the various layers is indicated as well as the position of the original microbridge and the FIB beam direction. The inset (a) shows a top view of the FIB prepared membrane imaged in the FIB. The inset (b) shows a side view obtained by TEM. The localization of the feature of interest, in this specific case, was done by patterning marks on the edges of the strip and by optical inspection of the position of the marks with respect to the AGB area. This was necessary as, due to the cutting and polishing of the strip, the YBCO layer was electrically isolated and showed no contrast in the FIB image with respect to the surrounding substrate. An alternative method would be to deposit inside the FIB, a conducting layer (i.e., Pt) between the YBCO line and the Cu grid so that the YBCO layer can directly be imaged in the FIB. The combination of precision mechanical polishing to a small thickness followed by FIB ion thinning greatly reduces the FIB processing time and allows for a larger sample tilt in the TEM.

Figure 2(a) is an optical micrograph showing a set of microbridges defined across the AGB with characterized transport properties. The JJ chosen for TEM is encircled. Figure 2(b) is a scanning electron (SEM) micrograph showing the particular JJ defined with an 8-μm wide bridge. Figures 3(a) and 3(b) show the FIB prepared XTEM sample as imaged in the TEM. Figure 3(a) is a bright field image showing an overview of the AGB region. The MgO buffer layer shows up bright in the image, and the edge in MgO is clearly visible. The electron transparent area is much larger than shown in the image and the different layers with varying hardness are thinned very evenly. Figure 3(b) is a more detailed image of the AGB region. The interface plane, indicated by a dotted line in Fig. 3(b), is identified as a (100) plane, defined with respect to the (001) film. The AGB is not located exactly at the MgO edge: the (001) film extends be-

![FIG. 1. Schematical drawing of the XTEM sample, prepared from a unique microbridge after mechanical polishing and FIB thinning. The plan view SEM image (a) and XTEM image (b) of this area are given as insets. The ideal structure of the AGB is sketched. The figure is not to scale.](image1)

![FIG. 2. (a) Optical micrograph showing an array of JJ defined as microbridges across the AGB. The encircled microbridge was chosen for TEM. (b) SEM micrograph showing the width of the microbridge. In addition the difference in surface structure between the (001) and (103)/(013) films is evidenced.](image2)

![FIG. 3. (a) TEM bright field micrograph of the FIB prepared sample with the AGB area indicated by arrows. The MgO layer shows up bright. The protective Pt layer is visible on top of the YBCO film. (b) Higher magnification of the AGB area. The 1.17 nm lattice spacing in the (001) film is resolved. The interface plane between the (001) and (103)/(013) film was identified as a (100) plane, defined with respect to the (001) film and is indicated by dots.](image3)
beyond the MgO edge. This is presumably a consequence of different growth speeds in different directions and/or preferential nucleation. The actual interface structure can comprise various different interface planes, deviating from the “ideal” (100) interface plane for a [100] 45° twist AGB as evidenced from other TEM and high resolution electron microscope (HREM) studies carried out on substrates with multiple junctions. The presence of different interface planes detected in our 45° twist AGBs can be related to the different transport regimes which are found on nominally identical junctions. Other SEM and TEM observations also reveal that the junction area is composed of conducting contacts separated by secondary phase regions.

The current–voltage (I–V) characteristics of the AGB junction are reported in Fig. 4 for different values of an applied magnetic field. The critical current density \( J_c \) is of the order of \( 10^4 \, \text{A/cm}^2 \) at \( T = 4.2 \, \text{K} \) and the specific conductance \( \sigma_N \) is 100 (\( \mu \Omega \, \text{cm} \))^{-1}. The modulation of the critical current due to the externally applied magnetic field was greater than 50%. The measured period is 2.5 Gauss, which gives a factor focus of \( \sim 5 \). The maximum working temperature is 75 K and the value of critical current times normal state resistance \( J_c R_N \) is 1 meV at \( T = 4.2 \, \text{K} \). Deviations from the resistively shunted junction (RSJ) behavior, resulting in the presence of the excess current and steps at finite voltages, are evident. The voltage position \( (V_n = 200 \, \mu \text{V}) \) of these steps is not affected by the magnetic field, as opposed to the current amplitude which increases with field in correspondence to a reduction of \( I_c \). This gives evidence that the steps are related to electromagnetic wave propagation in the barrier region (Fiske steps). The presence of Fiske modes indicates in general a high quality tunneling barrier, which acts as a transmission line. Such features have been recently observed and characterized in other junctions of this type and in structures obtained by other techniques. The \( I–V \) curves demonstrate that the lack of uniformity does not seem to affect the wave propagation along the junction interface. From the position of the Fiske steps it is possible to gain information on microscopic properties, such as the phase velocity \( c \) of the electromagnetic wave in the barrier cavity and on the ratio between the barrier thickness \( t \) and its dielectric constant \( (\varepsilon_r) \). In this case \( c = 1.6 \times 10^8 \, \text{m/s} \) and \( t/\varepsilon_r = 0.008 \, \text{nm} \). The presented results indicate that a [100] 45° twist AGB determines a Josephson coupling between the electrodes.

The illustrated method for correlating microstructure and transport properties can be slightly modified to investigate microstructural variations in cross section along the width of a single microbridge. The FIB preparation method should then be combined with an extraction method to prepare various cross-section samples from one microbridge. It is possible to thin the FIB prepared XTEM sample by conventional ionmilling for very short time in order to perform detailed HREM studies at the AGB area.

It has been shown that the microstructure of a JJ with previously measured properties over a micron size bridge, can be characterized by XTEM by employing a TEM sample preparation route based on FIB thinning. This method offers clear advantages over conventional TEM preparation procedures, such as submicron area selection and a high yield. The method is illustrated here for a JJ comprising a 45° [100] twist AGB but can be applied to any kind of JJ. This technique offers a tool to investigate the microstructural origin of transport properties. A clear advantage of this method is that the junction area is still integrated in the cross section of the wire, and a large number of junctions can be measured on the same slice of the wire. One of the cases, for instance, in which this is very useful, is the determination of the critical current density, \( J_c \), which is of the order of \( 10^4 \, \text{A/cm}^2 \) at \( T = 4.2 \, \text{K} \). Deviations from the ideal behavior are clearly visible in the \( I–V \) curves of the junctions. The authors wish to thank L. Rossou and G. Stoffelen for the highly precise mechanical polishing and P. Van Marcke for the FIB specimen preparation. This work has been partially supported by the project PRA-INFM “HTS Devices” and IUAP 4-10.

7. The AGB is described by the axis and angle around which the two electrodes are rotated. We do not distinguish between [100] and [010] directions of YBCO.