Interplay between structural anisotropy and order parameter symmetry effects in transport properties of $YBa_2Cu_3O_{7-\delta}$ grain boundary Josephson junctions

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Abstract. We have designed an experiment that could allow to investigate, on the same sample, the interplay between the morphological anisotropy of high critical temperature superconductors and the effects related to the order parameter symmetry. To this aim we have employed *c*-axis tilt biepitaxial grain boundary YBa₂Cu₃O_{7- δ} Josephson junctions and measured the junction properties for different junction interface orientations. The results give evidence of the possibility to tune normal state resistance and critical current density of a Josephson junction almost independently of each other. We interpret the results of this experiment as an indication that Cooper pairs and quasi-particles transport properties may be ruled by different mechanisms. Measurements apparently give evidence of the possibility to determine to some extent the presence of " π -loops" and "0" loops along the same grain boundary line.

PACS. 74.20. Rp Pairing symmetries (other than s-wave) – 74.50. +r Proximity effects, weak links, tunneling phenomena, and Josephson effects

The $I_C R_N$ factor (I_C is the critical current and R_N the normal state resistance) is one of the main quality parameters of a Josephson junction [1]. Data collected on various types of high critical temperature superconductor (HTS) Josephson junctions revealed unusual behavior of $I_C R_N$ vs. the critical current density J_C when compared with low critical temperature superconductor (LTS) Josephson junctions. While in LTS Josephson junctions, an increase of I_C is always completely counterbalanced by a decrease of R_N so that $I_C R_N$ is constant and depends only on the material (in the limit of small junction, *i.e.* junction width L less than the Josephson penetration depth λ_J , $L \ll \lambda_J$, in HTS this occurs only partially. In this anomalous feature, some aspects of the puzzling phenomelonogy of HTS Josephson junctions have been traditionally condensed. Different interpretations have been proposed to explain such a behavior. The first approaches were mainly based on hypotheses on the barrier microstructure (*i.e.* proximity effect, resonant tunneling, two-channel transport mechanism) [2]. Additional elements to explain several aspects of the phenomenology of HTS Josephson junctions have been provided by

models [3–5] based on $d_{x^2-y^2}$ -wave order parameter (OP) symmetry in HTS [3]. These approaches mainly combine effects typically related to the barrier properties or to the morphology of the grain boundary (*i.e.* proximity effect, faceting, band bending, space charge layers,...) with $d_{x^2-y^2}$ -wave OP symmetry. Nevertheless definitive experiments for a complete understanding of the transport properties in HTS Josephson junctions are still missing. The idea we pursued was to look for a type of $YBa_2Cu_3O_{7-x}$ (YBCO) grain boundary (GB), if any, whose critical current I_C and normal state resistance R_N values were not at all related to each other. We have looked for some type of junction that could clearly show, by tuning one of its parameters, that for instance by increasing I_C , R_N does not necessarily decrease. The existence of this extreme regime may imply that the mechanisms which rule I_C and R_N might be weakly related in some circumstances. This structure may be appropriate to decouple the two main effects ruling the junctions properties: symmetry of the OP and morphology/microstructure of the junction barrier region.

The solution to this problem can be offered by a junction with properties sensitive to both the $d_{x^2-y^2}$ -wave OP symmetry and to morphological anisotropy effects. The

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Fig. 1. a) The use of CeO₂ as a seed layer produces an artificial GB that can be seen as a result of two rotations: a 45° [100] tilt or twist followed by a 45° tilt around the *c*-axis of the (001) film. b) Scheme of the seed layer patterning, which allows the measurement of the junction properties for different orientations of the junction interface through the angle α . Details of the different junction configurations taking into account faceting are given in the inset.

behavior of a GB junction with the electrodes rotated by 45° tilt around the *c*-axis (in plane) with respect to each other [6,7] is known to be strongly influenced by effects of $d_{x^2-y^2}$ -wave OP symmetry [4]. If we produce an additional 45° tilt rotation of the *c*-axis (out of plane) in one of the electrodes, we also enhance effects due to GB structural anisotropy ((100) 45° tilt or twist GB). The resulting GB is shown in Figure 1a and may represent the

desired structure. It can be produced by using the biepitaxial technique, where a seed layer is used to modify the YBCO crystal orientation on part of the substrate [7]. In this case a thin layer of CeO_2 is deposited as a seed layer on a (110) SrTiO₃ (STO) substrate (see below). In this configuration we expect that d-wave induced effects might play a relevant role in some conditions. The biepitaxial technique gives the crucial advantage to provide different types of grain boundaries on the same sample. This can be easily done by changing the junction interface orientation with respect to the two electrodes *i.e.* the angle α (see Fig. 1b). For each orientation, the effects related to the order parameter symmetry and to the intrinsic structural anisotropy of the GB (tilt or twist or intermediate situations) will be in principle different. It will be necessary to consider as a term of comparison the same type of biepitaxial structure where effects due to 45° tilt in plane rotation are eliminated [8]. This kind of structure is realized by using MgO as a seed layer and their properties have been shown not to be influenced by the presence of π -loops [7]. Systematic measurements of the Josephson properties as a function of the mis-orientation angle have already been performed in GB junctions on the very first bicrystals [9] and, more recently, to investigate the order parameter symmetry [10]. In the latter case, analyzing (001) tilt GBs Ivanov et al. [10] observed anisotropy effects related to tunneling matrix elements. Nevertheless in all measurements an increase (decrease) of I_C always corresponded to a decrease (increase) of R_N respectively, demonstrating substantially that these two parameters are strongly related to each other. Another systematic study of anisotropic Josephson coupling has been performed on YBCO/Au/Ag/PbIn junctions using in-plane aligned *a*-axis films [11]. Also in these measurements I_C and R_N are strongly correlated. In this case we do have the *a*-axis HTS electrode oriented to enhance effects due to anisotropy between the a - b planes and the c-axis, but the counter-electrode PbIn is a s-wave superconductor. This prevents from observing all features related to $d_{x^2-y^2}$ -wave OP symmetry. Our junction configuration is therefore different from the designs of [10] and [11], and suitable to observe the discussed effects.

In this experiment we have used MgO and CeO₂ as seed layers, both deposited on (110) STO substrates; in all these cases the seed layers grow along the [110] direction and have a thickness of 20–30 nm. YBCO films, typically 120 nm thick, were deposited by inverted cylindrical magnetron sputtering at a temperature of 780 °C. YBCO grows along the [001] direction on both MgO and CeO₂ seed layers, while it grows along the [103]/[013] direction on STO substrates. In order to select the [103] or [103] growth and to ensure a better structural uniformity of the GB interface, we have also successfully employed vicinal substrates.

The phenomenology of the junctions based on 45° *a*-axis tilt or twist GBs (MgO seed layer) (BPMg) is quite different from the behavior of GB junctions where a 45° *c*-axis tilt accompanies the 45° *a*-axis tilt or twist (CeO₂ seed layer) (BPCe) respectively. The expected differences



Fig. 2. $I_C R_N$ values *vs.* J_C for different types of junctions, including BPMg (full circles) and BPCe (full squares). In the insets a) and b), I-V curves as a function of an externally applied magnetic field *H* for the junctions BPMg (a) and BPCe (b) are reported respectively (T = 4.2 K).

seem to be strongly correlated to the relative orientation of the OP in the two electrodes and, as a consequence, also to the absence or presence of π loops respectively. The first remarkable difference between the BPMg and BPCe junctions is the $I_C R_N$ value. For the former junctions this value is typically of the order of 1 mV at T = 4.2 K, while for the latter junctions it is about one order of magnitude less. This difference is clearly shown in Figure 2, where the $I_C R_N$ values are reported for several different BPMg and BPCe junctions as a function of their corresponding critical current density (J_C) . Data available from the literature which refer to other types of GB junctions are also reported for a comparison. $I_C R_N$ values of the BPCe junctions are close to those reported for conventional biepitaxials, while those of the BPMg junctions are of the same order of magnitude as in GB bicrystal in correspondence to the same J_C values [8].

For BPMg-type junctions, while the values of critical current density J_C and normal state specific conductance σ_N in the tilt case are quite different from the twist case, the $I_C R_N$ values are approximately the same for both [8]. In the tilt cases at T = 4.2 K, $J_C \approx (0.2-10) \times 10^3$ A/cm² and $\sigma_N \approx (1-10) (\mu \Omega \text{ cm}^2)^{-1}$ are measured respectively. Twist GBs junctions are typically characterized by higher values of J_C in the range $(0.1-2.0) \times 10^5$ A/cm² and of σ_N in the range $(20-120) (\mu \Omega \text{ cm}^2)^{-1}$ (at T = 4.2 K) [8]. We observed I_C modulations following the usual Fraunhofer-like dependence (see inset a of Fig. 2), and in all samples the absolute maximum of I_C occurs at H = 0. By suitably patterning the seed layer, we could measure the properties of a tilt junction and of junctions whose interface is tilted in plane by an angle $\alpha = 30^\circ$, 45° and 60° with respect to the *a*or *b*-axis of the [001] YBCO thin film respectively [7]. In all cases, the OP orientations do not produce a significant π phase shift along our junctions [7].

In the case of BPCeO-type junctions we observed the expected decrease of J_C , which is consistent with the fact that the lobe of the OP of the former electrode is in average facing the node of the OP of the latter electrode [3,4]. The effective current is ultimately controlled by faceting, *i.e.* the combination of conventional Josephson currents



Fig. 3. (a) Current vs. voltage (I-V) characteristics of BPCeOtype junctions at T = 4.2 K in the tilt $\alpha = 0^{\circ}$ and $\alpha = 90^{\circ}$ twist cases respectively; (b) I_C is reported for BPCeO-type junctions as a function of α for the samples BP3Ap and BP8Ap and compared with the MgO case (c) Normal state conductance σ_N of BPCeO-type junctions are reported as a function of the angle α and compared with BPMgO-type junctions (at T = 4.2 K).

and negative currents due π phase shift [4, 12]. In addition, measurements have shown that the critical current densities in the tilt ($\alpha = 0^{\circ}$) and twist ($\alpha = 90^{\circ}$) cases are quite similar ($J_C \approx (1-10) \times 10^2 \text{ A/cm}^2$ at T = 4.2 K) (Fig. 3a). They also gave clear evidence that the tilt and twist cases have lower values of the critical current density when compared with intermediate situations, that are defined by an angle α different from 0° and 90°. For angles $\alpha = 15^{\circ}$ and 75° we measured J_C values from 5 to 10 times higher than the tilt and twist cases in various samples. This is also evident from Figure 3b where J_C is reported as a function of α for the samples BP3Ap and BP8Ap respectively. J_C data in Figure 3b are also compared with the MgO case, where the critical current density increases with increasing angle in absence of effective π -loops[7]. The non monotonous dependence of J_C in BPCe-type junctions is a very peculiar feature. This behavior is consistent with the picture of a predominantly $d_{x^2-y^2}$ -wave order parameter symmetry. The tilt and the twist cases represent the most unfavorable situation for the Josephson current (see inset of Fig. 1b). From this point of view they resemble more the [001] 45° tilt asymmetric GB case [4].

For $\alpha = 0^{\circ}$ and 90° (see inset b of Fig. 2), I_C modulations do not follow the Fraunhofer-like dependence and the absolute maximum of I_C occurs at symmetric values of $H \neq 0$. This behavior is similar to the one observed in 45° tilt [001] asymmetric tilt GB junctions [4,12] and consistent with the presence of regions with π phase shift along the grain boundary. While the oscillatory behavior of I_C as a function of α is a general feature of the $d_{x^2-y^2}$ wave OP symmetry, the position of the minima $\bar{\alpha}$ can depend on the particular junction interface. Junctions in correspondence to angles $\alpha \neq 0^{\circ}$, $\bar{\alpha}$ and 90° behave more similarly to the [001] 45° tilt symmetric GB configuration. In this case, as experimentally confirmed, we expect that negative currents may be reduced. In Figure 3b, as a guide for the eye, we report the qualitative behavior of I_C vs. α expected in the case in which both electrodes are *c*-axis oriented. Due to the presence of (103) oriented regions, we expect $\bar{\alpha}$ different from 45°.

These results suggest that, by suitably patterning the CeO₂ seed layer, we can controllably determine macroscopic π loops or conventional "0" loops along the same grain boundary line respectively. This property can be used for the realization of a doubly degenerate state device, recently proposed as an element for quantum computing [13].

In Figure 3 experimental data on σ_N as a function of the angle are reported. σ_N values increased by increasing the angle α and therefore passing from the tilt to the twist case (σ_N in the range (1–10) ($\mu\Omega$ cm²)⁻¹ at T = 4.2 K) (Fig. 3c). The values of the normal state resistance are quite close to the values of the BPMg and demonstrate that in the tilt case the barrier probed by quasi-particles is mainly due to the out-of-plane rotation of the *c*-axis in the grain boundary. In the twist case the situation is more complicate. The effect of in plane rotation around *c*-axis on the normal state resistance is comparable to that of a 45° twist of the *c*-axis. In general the normal state resistance will depend on the details of the interface barrier.

In conclusion we have presented current vs. voltage characteristics (I-V) relative to (001) 45° tilt and (100) 45° tilt or twist grain boundaries Josephson junctions. Evidence of oscillatory dependence of the critical current on the angle α has been given. If this observation is combined with the angle dependence of the normal state resistance, some indications that Cooper pairs and quasi-particles may be ruled by different transport mechanisms can be inferred. This gives evidence of the possibility to tune normal state resistance and critical current density of a Josephson junction independently of each other. These results also suggest that, by suitably patterning the CeO₂ seed layer, we can controllably determine π loops or conventional "0" loops along the same grain boundary line respectively. This property can be also considered for the realization of a doubly degenerate state device [13].

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