Properties of submicron [001] tilt symmetric and asymmetric 45° bicrystal grain boundary junctions

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Abstract. We have fabricated submicron YBa₂Cu₃O⁷-x [001] tilt bicrystal grain boundary junctions by a focused ion beam process. The reduction of the junction width, leading to more homogeneous grain boundary interfaces, has been proved to be very important for the study of many fundamental properties of high critical temperature superconducting junctions. In particular, experiments on 45° symmetric junctions have shown a clear evidence of midgap states, with a 0- to π-junction transition in the case of small barrier transparency; moreover, in 45° asymmetric junctions we have observed a transition from an unconventional to a Fraunhofer-like magnetic field dependence of the critical current, sign of the reduced influence of faceting on the junction properties.

Even though high critical temperature superconductors (HTS) were discovered about 20 years ago, their basic physical mechanisms are not perfectly known yet. In recent years, a number of experiments on HTS have given a clear evidence of a strongly anisotropic d-wave order parameter symmetry, with nodes along the (110) directions in k-space and a π-phase shift between orthogonal kₓ and kᵧ directions [1-2]. The most interesting results have been obtained by phase-sensitive tests, mainly based on superconducting loops containing one or more grain boundary junctions (GBJs) [1-2]. Unfortunately, some experiments related to the HTS unconventional superconducting wave function (as for example surface spontaneous currents, time reversal symmetry breaking, zero bias conductance peaks, etc.) have shown contradictory results, because of the large faceting typical of HTS GBJs and, more generally, because of the complex structure of their barrier interface, characterized by many defects and high transmissivity channels. Since these problems are strictly related to the grain boundary nature itself, they can only be solved by optimizing the fabrication technologies.

In this paper, we have followed a different approach, i.e. we have reduced the junction width to a submicron scale in order to minimize defects and barrier disuniformity. The fabrication of submicron bicrystal GBJs has been widely described elsewhere [3-5], so only an outline of the procedure is reported here.

A YBa₂Cu₃O₇-x film, 120-140 nm thick, followed by a 50 nm thick amorphous SrTiO₃ layer have been first deposited by laser ablation on [100] tilt SrTiO₃ bicrystal substrates. The SrTiO₃ layer has been used both to passivate the junction and protect the YBa₂Cu₃O₇-x surface from gallium
contamination during the Focused Ion Beam (FIB) process. Finally, a thin gold layer has been deposited by magnetron sputtering to cover the sample with a conductive layer for the FIB process and as an additional stopping layer for Ga ions. The device geometry has then been patterned by standard photolithography and a water-cooled argon ion milling etch. The junction widths has then been reduced by removing YBa$_2$Cu$_3$O$_{7-x}$ from the junction area by a FIB microscope with a Ga source.

Measurements have been performed, with very low noise electronics, in a Helium cryostat shielded by 1 thick aluminum and 3 mu-metal shields with a residual field lower than 1 mG.

Current-voltage (I-V) characteristics and magnetic field dependencies have been measured as a function of the temperature both by using a vacuum probe with a heating element, and, in helium vapors without any heaters, by carefully controlling their distance from the helium bath.

In the following two sections we show experimental data related to both 45° symmetric and 45° asymmetric [001] tilt bicrystal GBJs with widths in the range 0.3 - 1 μm.

1. 45° symmetric [001] tilt bicrystal GBJs

In figure 1, we show the temperature dependence of the Josephson current $I_c(T)$ for two 45° symmetric GBJs, 920 nm (stars) and 480 nm (circles) respectively. Both the upwards curvature of the 920 nm junction and the anomalous non monotonic dependence of the 480 nm GBJ are explained in terms of a strong contribution of midgap states [6-8], which is opposite to the continuous Josephson current. In the inset, the theoretical behaviour, calculated according to Eq.64-65 of Ref.7, is also shown for both small ($D=10^{-3}$, solid line), and high ($D=0.5$, dotted line) barrier transparency.

![Figure 1](image_url)

**Figure 1.** Temperature dependence of the Josephson current in two 45° symmetric GBJs, 920 nm (stars) and 480 nm (circles), respectively. In the inset, the theoretical $I_c(T)$ dependence, calculated according to Ref. 7, is also shown (the solid line refers to a barrier transparency $D=10^{-3}$, the dotted one to $D=0.5$).

Typically, moving from junction widths of the order of 1 μm to less than 500 nm, we always observe a transition from a monotonic to a non monotonic $I_c(T)$. As widely explained in Ref. 5, it can be accounted for by considering that a reduction of the junction width leads to more uniform barrier interfaces with a smaller number of high transmissivity channels. Low barrier transparency may then be achieved more easily, allowing the observation of the 0- to π- junction crossovers with temperature (non monotonic behaviour), as predicted by the theory (solid line in the inset) [4-5].

This transition has also been confirmed by measurements of the magnetic field dependence of the critical current in a submicron dc SQUID [4]. A half flux quantum shift in the maximum critical
current has been clearly observed by decreasing the temperature, indicating the transition from a 0-loop to a frustrated $\pi$-loop.

2. 45° asymmetric [001] tilt bicrystal GBJs

Many experiments performed on 0°-45° GBJs have shown an anomalous magnetic field dependence of the critical current $I_c(H)$, characterized by two symmetric current maxima at field values different from zero [9-11]. In addition, SQUID microscopy measurements have demonstrated the presence of fractional vortices at the grain boundary interface [9,11]. Such features have been mainly explained in terms of randomly distributed facets along the GB, with each facet characterized by an equilibrium phase difference of 0 or $\pi$, depending on the sign of the order parameter of lobes facing at that facet. An alternative explanation has also been the presence of spontaneous currents due to a broken time-reversal symmetry, which is possible in 0°-45° configurations even in the absence of a subdominant order parameter component [6].

In the following, we show that also the $I_c(H)$ behaviour has a clear dependence on the junction width, with an apparent crossover from the anomalous to a Fraunhofer-like magnetic pattern when the junction width is reduced to a submicron scale. In Figure 2a and figure 2b, we show the $I_c(H)$ dependence for two GBJs, 4 $\mu$m, and 900 nm wide, respectively. The behaviour in figure 2b is typical of all the junctions whose size is smaller than 1 micron.

![Figure 2](image)

**Figure 2.** Magnetic field dependence of the Josephson current in two 45° asymmetric GBJs, 4 $\mu$m (a) and 900 nm (b) wide, respectively.

The transition to a Fraunhofer-like dependence (or at least to a magnetic pattern with a maximum at $B=0$), observed sometimes also for junctions 2 $\mu$m wide, seems to be consistent with a reduction of the faceting in submicron junctions. Indeed, as a result of the lower interfacial energy of the (110)/(100) facet in comparison with that of the symmetrical one, the facet length in 45° asymmetric GBJs is much larger than in symmetric 45° junctions and can be of the order of hundreds of nm [12]. Submicron 45° asymmetric junctions are then characterized by a much smaller number of facets. Therefore, we may expect an average critical current density $j_0$, substantially different from zero, in contrast to micron-wide junctions where the opposite critical current contributions from the large number of facets tend to compensate, giving a very small $j_0$ (about 0).

In Figure 3, we show two $I_c(H)$ dependencies, calculated by using the model reported in Ref.13. The junction critical current density $j_c$ is modeled as a constant value $j_1$ with sign changing over randomly long facets. It is described as a Gaussian distribution with average $j_0$ and dispersion $\sigma$. Figure 3a and 3b show the dependencies expected for $j_0 = 0$ and $\sigma = 0.1$ (number of facets, N=20) and for $j_0 = 0.57 j_1$ and $\sigma = 0.1$ (N=4), respectively. In the latter case, the presence of an average current different from zero gives a clear local maximum at $B=0$, in agreement with experimental $I_c(H)$ data. In the calculations, we have reduced N to simulate a smaller junction size but the result is completely equivalent also keeping N=20.
Figure 3. Theoretical magnetic field dependence of the Josephson current in 45° asymmetric GBJs, (a) \(j_0=0, \sigma=0.1, N=20\); (b) \(j_0=0.57, \sigma=0.1, N=4\).

It is worth noting that in real junction the picture is much more complicated. Indeed, the critical current density is not constant over the different facets, because of the not uniform barrier interface. However, also in this case, we expect a better cancellation of the average critical current in larger junctions.

3. Conclusions
The presence of faceting and the intrinsic not uniformity of GB interfaces make many effects, related to the unconventional HTS superconducting wave function (like surface spontaneous currents, time reversal symmetry breaking, zero bias conductance peaks, etc.), very difficult to observe. Therefore, current fabrication techniques should be optimized in order to have more controllable and reproducible junctions. In this paper, however, we have shown that a valid alternative is the reduction of the junction width, which allows the fabrication of HTS GBJs with more uniform interfaces and a smaller influence of faceting.

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