Capacitance of Josephson junctions made on bicrystalline substrates of different geometries

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The electromagnetic parameters of YBa₂Cu₃O_{7-x} (YBCO) grain boundary Josephson junctions (JJs) fabricated on four different tilt bicrystal geometries: 12° [001] asymmetric, 24° [001] symmetric, 24° [001] asymmetric, and 45° [100] asymmetric, have been studied. While the Swihart velocity (\bar{c}) is slightly affected by the nature of the barrier and mainly fixed by the junction width, a notable influence of the barrier structure, of the geometry of the bicrystal substrate, on the relative dielectric constant to the barrier thickness ratio (ϵ/t) values has been found. Interesting barrier information can be deduced from the study of the dependence of the junction capacitance on the junction resistance. We have observed that the capacitance values deduced by means of Fiske steps in YBCO 24° [001] symmetric and 45° [100] asymmetric JJs scales with junction resistance in the opposite direction. This result could reveal the presence of a tunnel barrier in the YBCO 45° [100] asymmetric JJs. On the other hand, different capacitance values have been obtained by means of Fiske steps and hysteresis observed in the current-voltage characteristics in 45° [100] asymmetric JJs. An interpretation of this result can be made taking into account the contribution of the depleted YBCO layers close to the crystallographic grain boundary.

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I. INTRODUCTION

A widely used technique for the fabrication of high critical temperature Josephson junctions is the epitaxial growth of YBa₂Cu₃O_{7-x} (YBCO) thin films on bicrystalline substrates. The first studies carried out on this kind of grain boundary Josephson junctions (JJs) were mainly focused on electrical and structural properties of YBCO thin films deposited on [001] symmetric tilt bicrystals with different misorientation angles. 1-3 In the following years, because of their critical impact on research and applications of high critical temperature superconductivity, bicrystalline substrates have also been investigated with other kinds of geometries.^{4–7} At this stage, few studies on the structural and transport properties of JJs generated on substrates tilted around a-b planes have been presented.^{8,9} The properties of these geometries have been more extensively studied in JJs fabricated by a biepitaxial technique which has shown to be a good option for the implementation of a reliable technology based on the Joshepson effect. 10-12

The structure of the barrier depends on many factors like the misorientation angle between the two electrodes, the orientation of the interface plane with respect to crystallographic axes, and the meandering at the barrier, so no prediction of the exact boundary structure is possible. Although some structural analyses on JJs using techniques like transmission electron microscopy¹³ have been performed, the structural and electrical information about the barrier has been mainly derived from the transport parameters: normal resistance (R_N) and critical current (I_C) .

On the other hand, from the position of resonance steps¹⁴ (Fiske and flux-flow), supporting the idea of the junctions as

long parallel resonators for electromagnetic waves, and from the amplitude of the hysteresis observed in the current-voltage (I-V) curves of the junctions, it is possible to determine the values of the electromagnetic parameters: the Swihart velocity (\vec{c}) and the relative dielectric constant to the barrier thickness ratio (ε/t) , ^{15,16} and, as a consequence, the value of the barrier capacitance (C). Electromagnetic parameters can reveal new insight into the nature of the barrier and are a very useful tool to corroborate the information deduced from transport parameters.

Previously, 17,18 we presented a study of the electromagnetic parameters of 24° [001] tilt symmetric JJs. For this geometry, we found a new scaling law that sets a constant product $\varepsilon/t \times w$ (w is the width of the junction) for samples with the same thickness. A barrier envisaged as a disordered dielectric medium with a high density of superconducting filaments taken into account in an inductive behavior can explain such a result. Also we studied the evolution of the electromagnetic parameters when the barrier is modified by irradiation with helium at 80 keV and by annealing treatments in oxygen atmosphere.

In this paper we present a comparative analysis, in terms of electromagnetic parameters, of JJs fabricated using $SrTiO_3$ (STO) bicrystalline substrates of four different tilt geometries. We show that the value of \bar{c} is mainly fixed by the width of the junction and no big quantitative differences are observed between the four geometries. However, the ratio ε/t is more dependent on the particular structure of the barrier, and so on the kind of bicrystalline substrate used for the fabrication of the JJs. The scaling law previously reported 17 for YBCO 24° [001] symmetric JJs are not fully verified for 45° [100] asymmetric JJs, pointing to a different

kind of barrier. In this sense, in the final part of the paper we have focused on the comparative analysis of these two geometries to show that interesting information about the particular nature of the barrier can be deduced from the study of the evolution of the junction capacitance with junction resistance.

II. EXPERIMENTAL PROCEDURE

A. Fabrication of the samples

Josephson junctions were fabricated using bicrystalline substrates of STO with different symmetrical and asymmetrical tilt angles around c-axis and a-axis: 12° [001] asymmetric, 24° [001] symmetric, 24° [001] asymmetric, and 45° [100] asymmetric JJs. YBCO films 50 nm thick were epitaxially grown in a high pressure (3.4 mbar) pure oxygen dc sputtering system. In the deposition process the substrate temperature was 900 °C. Electrodes showed critical temperatures (T_C) in the range 89.5–91 K, transition widths smaller than 0.2 K, and critical current densities higher than 10⁶ A/cm² at 77 K. The accurate control of the film growth process with our sputtering system has been previously demonstrated.¹⁹ Films were patterned obtaining junction widths (w) ranging between 2 and 50 μ m. A correct control on w is important since it will become a key parameter in our discussion. In this sense, we have estimated that nonuniformity in w associated to the technological etching procedure and depletion effects in the electrode edges introduce an error in w less than 5% for $w > 2 \mu m$ and in the order of 8% for $w=2 \mu m$.

For the electrical measurements the samples have been magnetically shielded and the external magnetic field was applied parallel to the grain boundary plane. The I-V curves for different values of applied magnetic field have been measured using low-noise electronics.

B. Calculus of electromagnetic parameters: \bar{c} and ε/t

We have observed Fiske steps¹⁵ in our samples, supporting the idea of the junctions as resonators where the barrier forms the effective dielectric medium. In Fig. 1 we have plotted the I-V curves corresponding to two different geometries where Fiske steps are present. The Fiske steps have been identified because of the dependence of their intensity on the magnetic field applied parallel to the grain boundary plane. 14 From the position of the Fiske resonance steps it is possible to determine the velocity \bar{c} of the resonator using the expression $V_n = n\phi_0 \overline{c}/2w$, where *n* is the resonance number and ϕ_0 the magnetic flux quantum. 15 Although the two first steps are observed in some I-V curves, we always analyze the resonance n=1. From the value of \bar{c} it is possible to determine the ε/t ratio, and the C value, using the expression $\bar{c} = [c_0(t/\varepsilon d)^{1/2}]$, where c_0 is the vacuum light velocity and d the effective magnetic junction length $d=t+2\lambda \cong 2\lambda$. In our case, $\lambda = \lambda_L \coth(\delta/\lambda_L)$, where δ is the thickness of the films, since δ is smaller than the London penetration depth λ_L .¹⁴

The C value can also be determined from the hysteresis in the I-V curves, quantified by the Stewart McCumber parameter $\beta_C = 2\pi I_C R_N^2 C/\phi_0$. ¹⁴ I-V curves are hysteretic for β_C

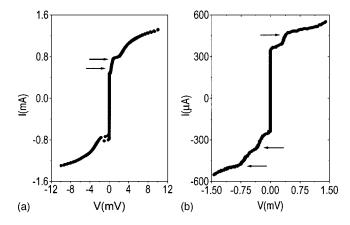


FIG. 1. (a) I-V characteristic of a [100] tilt 45° asymmetric 20 μ m wide junction with an applied magnetic field of 0.56 G. The arrows indicate the Fiske steps corresponding to n=1 and 2. The I-V curve shows hysteresis in the negative current part. (b) I-V characteristic of [001] tilt 24° symmetric 10 μ m wide junction with an applied magnetic field of 0.36 G. The Fiske steps with n=1 and 2 are observed in the negative part of the curve.

> 1, and single valued for $\beta_C < 1$. β_C can be obtained with Zappe's approximation, $\beta_C = [2 - (\pi - 2)\alpha]/\alpha^2$, in terms of the ratio $\alpha = I_R/I_C$ of the return current I_R to I_C .

III. RESULTS AND DISCUSSION

A. Electromagnetic parameters

In this section we analyze the electromagnetic parameters calculated for our four different bicrystal configurations. In Fig. 2 we have plotted \bar{c} versus w for each kind of JJs fabricated. For all the geometries the value of \bar{c} increases with w, in the same way as in 24° [001] symmetric JJs.^{17,18} For a fixed w and bicrystalline tilt geometry, even in the presence of different R_N values associated to the barrier, only a small dispersion in the calculated \bar{c} values can be observed in Fig. 2. Similar dispersion is found if we compare JJs fabricated on different kinds of substrates but with the same w. This

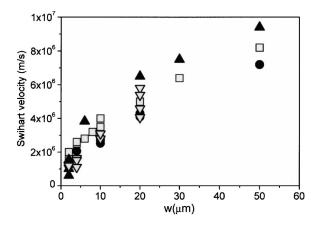


FIG. 2. Swihart velocity vs w for YBCO JJs fabricated on bicrystalline substrates of different geometries: 12° [001] asymmetric (\triangle), 24° [001] symmetric (\square), 24° [001] asymmetric (\square), and 45° [100] asymmetric (∇).

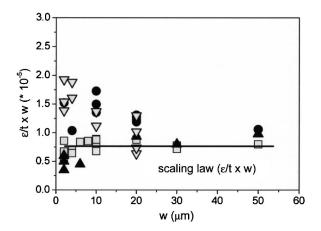


FIG. 3. $\varepsilon/t \times w$ product vs w for YBCO JJs fabricated on bicrystalline substrates of different geometries: 12° [001] asymmetric (\triangle), 24° (001) symmetric (\square), 24° [001] asymmetric (\square), and 45° [100] asymmetric (∇).

allows us to corroborate that \bar{c} is only slightly dependent on the particular properties of the barrier (then on the geometry of the bicrystalline substrate), w being the main parameter fixing its value.

In a previous published work¹⁷ we have deduced that the $\varepsilon/t \times w$ product is approximately constant for a fixed thickness of 24° [001] tilt symmetric JJs. In Fig. 3 we have plotted this product for the same junctions that we have presented in Fig. 2. For $w > 20 \mu m$, I_C becomes very large at low temperature and may exceed our current limited low noise electronics. In such a case we have to work with I-Vcurves at higher temperatures, which unfortunately implies a decrease of the Fiske steps intensity, 14 making the correct assignment of V_n difficult. We believe that any conclusion for $w > 20 \mu m$ may be uncertain. In this sense we will carry on our study for JJs with $w \le 20 \mu m$. The scaling law previously reported, verified for the 24° [001] tilt symmetric JJs, is not clearly satisfied for the junctions corresponding to the other three substrate geometries. As noted before, the systematic error affecting w and originated during the technological procedure is small and affects in the same way all our bicrystal geometries. Thus its influence on the deviation from the scaling law is discarded. In particular, the 45° [100] asymmetric JJs show the biggest dispersion, revealing a more notable influence of the nature of the barrier, of the particular bicrystal geometry, on the ε/t parameter. Although \bar{c} has a small dependence on the particular JJs (R_N and kind of substrate), as shown in Fig. 2, ε/t undergoes a larger influence because of its dependence on \bar{c}^2 . To confirm this aspect, in Table I we present a brief summary of the electromagnetic parameters of junctions of different w corresponding to 45° [100] asymmetric and 24° [001] symmetric JJs, which gives us a valuable reference because of our previous analysis. 17,18,21 As it can be seen in Table I, ε/t can show more significant variations than \bar{c} for the same w. In this sense, an analysis of ε/t , of the barrier capacitance, focused to bring information about the microstructure of the barrier and the transport mechanisms, is needed. In the next paragraph we study this point keeping as representative examples the bicrystalline geometries of Table I.

B. ε/t ratio: Comparison between 24° [001] symmetric and 45° [100] asymmetric JJs

In previous works, ^{22,23} a correlation between the capacitance per unit area (A) associated to the barrier (C/A) and the product R_NA was observed. Most data fell close to a power law $(C/A)\alpha(R_NA)^{-1}$. This behavior is common to many high temperature superconducting grain boundary experiments. Indeed, such a correlation was first observed by Moeckly et al.22 for 90° basal plane faced tilt grain boundaries in YBCO and then confirmed in YBCO 24° [001] symmetric JJs on different substrates, and YBCO step-edge junctions.²³ This scaling behavior is presented as consistent with the phenomenological filamentary model.^{24,25} The capacitance is dominated by the contributions from the superconducting regions near the interface which are separated by a short distance of oxygen deficient material. Then, if the number of superconducting filaments increases, R_N decreases and C increases.

The C/A ratio (i.e., the product $\varepsilon_0\varepsilon/t$) can be determined both from Fiske steps resonances and hysteresis of the I-V curves. However, in the case of 24° [001] symmetrical JJs, the C/A values have been determined only from the Fiske steps, because in general no hysteresis has been observed even at low temperature. For these junctions, values of β_C ranging between 0.75 and 1.3 have been estimated. Varia-

TABLE I. Electromagnetic parameters (\overline{c} and ε/t) for YBCO JJs of different values of w fabricated on 24° [001] symmetric and 45° [100] asymmetric bicrsytalline substrates.

| | 24° [001] symmetric | | | 45° [100] asymmetric | | |
|-----------|---------------------|-----------------------------------------------------|-----------------------------------|----------------------|-----------------------------------------------------|----------------------------------|
| w (μm) | $R_N \ (\Omega)$ | $\frac{\overline{c}(\times 10^{-6})}{\text{(m/s)}}$ | $\epsilon/t \ (\mathrm{nm}^{-1})$ | $R_N \ (\Omega)$ | $\frac{\overline{c}(\times 10^{-6})}{\text{(m/s)}}$ | ϵ/t (nm ⁻¹) |
| 20 | 4.6 | 5 | 4.4 | 6.8 | 4 | 6.8 |
| 20 | 3.2 | 5.5 | 3.6 | 6.5 | 5.8 | 3.3 |
| 10 | 5.2 | 3.6 | 8.5 | 13.5 | 2.8 | 14.1 |
| 10 | 3.8 | 4 | 6.8 | 11 | 3.1 | 11.5 |
| 4 | 9.5 | 2.5 | 17.3 | 37.1 | 1.5 | 48 |
| 4 | 8.5 | 2.6 | 16.1 | 27 | 2 | 27 |

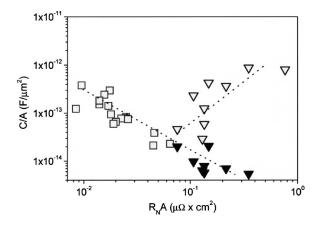


FIG. 4. Variation of junction capacitance per unit area (C/A) with junction resistance (R_NA) . In YBCO 24° [001] symmetric JJs, C/A is calculated from Fiske steps (\Box) , and in YBCO 45° [100] asymmetric JJs C/A is calculated from Fiske steps (∇) and hysteresis of I-V curves (∇) .

tions of C/A ratio with R_NA for these two kinds of JJs are shown in Fig. 4. Data relative to 24° [001] symmetric JJs follow the scaling behavior between capacitance and resistance described before, with an experimental data dispersion comparable to that reported in the literature.^{22,23} This would address a junction constituted by an array of superconductive connections separated by nonsuperconducting paths. Such a description of the barrier makes possible the interpretation of transport parameters of JJs with this geometry. 17,18 For instance, we found that transport parameters of 24° [001] tilt symmetric YBCO JJs are not clearly fixed by w: junctions with different values of w presented comparable values of R_N and I_C . ¹⁷ Following the filamentary model, ^{24,25} these parameters would be mainly fixed by the distribution and quality of the superconducting channels inside the barrier. The distribution of these filaments is basically random and exclusively dependent on the growth process of the barrier.

In 45° [100] asymmetric JJs the C/A data calculated from Fiske steps present the opposite behavior: the C/A ratio increases when R_NA increases. This tendency indicates that the enlargement of the dielectric constant (implicit in C) may be related to an increase of the concentration of defects in the barrier (implicit in R_N).²⁶ Variation of the concentrations of cations and defects mainly concerning the oxygen sublattice are the most commonly observed structural defects in JJs. However, the particular structure of the grain boundary, the interruption of the periodic lattice, may certainly depend on the particular bicrystalline substrate geometry. If 45° [100] asymmetric JJs have effectively different structural alterations at the grain boundary than 24° [001] symmetric JJs, transport measurements must reveal different features. In fact, we have found that the $I_C R_N$ product is practically constant in the 45° [100] asymmetric JJs. This phenomenological result, together with a linear relationship between the critical current density and the specific resistance values, make it reasonable to describe these junctions in a tunnel regime rather than in the filamentary framework.9 Nevertheless, more structural and chemical studies for this substrate geometry are necessary to corroborate such results.

Very interestingly, for the same samples, the C/A ratios obtained from the I-V curve hysteresis scale in opposite directions with R_NA . How can the C/A ratio obtained from hysteresis in the I-V curves scale in opposite direction with R_NA than the C/A ratio deduced from Fiske steps? In general the I-V characteristics of the 45° [100] asymmetric JJs are affected by the presence of current steps. Then, only in a limited number of cases the I-V curves are well described by the resistively shunted junction model (RSJ model). 9 Nevertheless, because of the presence of hysteresis in most of them, we consider that a calculus of the C/A ratio in terms of the β_C parameter could be also appropriated. Tarte et al. ^{23,27} found that the capacitance calculated from the Fiske resonance correlates well with that determined from the hysteresis measurements, but the analysis has mainly been performed on YBCO 24° and 36° [001] symmetrical JJs. In our case, we believe that the observed discrepancy is related to the fact that different regions of the grain boundary contribute to the C values estimated in the two different methods.

The current steps observed in the I-V curves are a consequence of an interaction of the Josephson current with resonant modes of the barrier, regarded as a transmission line. The propagation direction of the resonant modes is along the longitudinal direction of the barrier and normal to the Josephson current. It is reasonable to assume that only the most oxygen deficient regions of the barrier, very close to the crystallographic grain boundary, would form the effective dielectric medium supporting the wave propagation. Then, we believe that from Fiske steps, the calculated ε/t ratio is only related to the region acting as a transmission line. This region would be associated to the crystallographic grain boundary, although it is known that the disorder induced by the grain boundary may also affect the adjacent regions, mainly in the oxygen sublattice. The good agreement between the C values deduced from current steps and hysteresis in 24° [001] symmetrical JJs might be explained by assuming that 23,27 in symmetrical [001] tilt bicrystal junctions, with misorientation angles ranging between 10° and 24°, there is a rather sharp structural transition from good superconducting regions in the grains to the barrier. However, the smaller C values derived from hysteresis in YBCO 45° [100] asymmetrical JJs point to a serial contribution of the adjacent regions to the grain boundary, thus to a more gradual structural transition between good superconducting regions (junction electrodes) and the crystallographic grain boundary. In the framework of the band-bending theory, 28,29 the effective electronic width of the barrier is $t=t'+2l_d$, where t' is the structural width of the boundary and $l_d = (2\varepsilon_0 \varepsilon V_{bi}/en)^{1/2}$ is the width of the depletion layer, where V_{bi} is the built-in potential, n the electrode carrier density, and e the elementary charge. It has been shown that the critical current density across the grain boundaries is a function of the misorientation angle,² then because of the misorientation angle and asymmetric geometry, we expect n to be smaller, so l_d is larger in the 45° [100] asymmetric JJs. In fact, the bandbending theory has been used to explain such a relation between I_C and the misorientation angle.²⁹ The total capacitance C calculated from hysteresis can be estimated by assuming that it is the serial contribution of these three regions, i.e., $1/C_{\text{hysteresis}} = 1/C_{t'} + 2/C_{l_d}(1)$, where we associate

TABLE II. Capacitance per unit area calculated from hysteresis $(C_{\text{hysteresis}}/A)$ and Fiske steps $(C_{t'}/A)$ observed in the I-V curves. C_{ld}/A is deduced from $1/C_{\text{hysteresis}}=1/C_{t'}+2/C_{l_d}$. All data correspond to 45° [100] asymmetric JJs.

| $C_{ m hysteresis}/A \ ({ m F}/\mu{ m m}^2)$ | $\frac{C_{t'}/A}{(\mathrm{F}/\mu\mathrm{m}^2)}$ | $rac{C_{ld}/A}{({ m F}/\mu{ m m}^2)}$ |
|----------------------------------------------|-------------------------------------------------|----------------------------------------|
| 9.73×10^{-15} | 2.32×10^{-13} | 2.03×10^{-14} |
| 7.87×10^{-15} | 1.25×10^{-13} | 1.68×10^{-14} |
| 6.98×10^{-15} | 3.66×10^{-13} | 1.42×10^{-14} |
| 5.68×10^{-15} | 5.91×10^{-14} | 1.26×10^{-14} |
| 5.38×10^{-15} | 8.78×10^{-13} | 1.08×10^{-14} |
| 2.05×10^{-15} | 4.28×10^{-13} | 4.32×10^{-14} |

 $C_{t'}$ to the value determined from the Fiske resonances, and the value C_l , to each depletion layer situated at both sides of the crystallographic grain boundary, respectively. In Table II we have reported the experimental values $C_{\text{hysteresis}}/A$ and $C_{t'}/A$. C_{ld}/A is deduced from Eq. (1). Following the bandbending theory, ^{28,29} a theoretical estimation of such a ratio can be made by means of $C_{l_d}/A = \varepsilon \varepsilon_0/l_d = (\varepsilon \varepsilon_0 en/2V_{bi})^{1/2}$. The result for C_{Id}/A is 5.6×10^{-13} F/ μ m² with $\epsilon \approx 10$, eV_{bi} ≈ 0.1 eV, and $n=4.5\times 10^{21}$ cm⁻³, values used by Mannhart et al.29 for an estimation of the effective electronic width of YBCO 30° [001] tilt symmetric JJs. The parameter values are very conservative because, for example, no data is available for V_{bi} . In this sense, the fact that our calculated data (see Table II) are in the range of 10^{-14} F/ μ m² could corroborate a larger l_d value in 45° [100] tilt asymmetric JJs than in other bicrystal geometries as we have suggested. We believe that, while in [001] tilt bicrystal geometries with misorientation angles smaller than 30° it is difficult to distinguish the contribution of the adjacent regions, 23,27 in 45° [100] tilt asymmetric JJs the capacitance values deduced from hysteresis in I-V curves reveal their contribution. This is in accordance with the studies based on atomic-resolution transmission electron microscopy:30 the estimated widths of the nonsuperconducting zones adjacent to the interface increase from 0.2 to 0.9 nm (approximately) for an increase of the misorientation angle from 11° to 45°. Then, changes in the geometry of the bicrystal boundary affect the microstructure of the barrier, so that the depletion layers induced in the adjacent regions of the barrier have to be taken into account for a correct interpretation of the transport and electromagnetic parameters.

IV. SUMMARY

In this paper we have presented a detailed study of electromagnetic parameters of JJs fabricated on bicrystalline substrates with different geometries. While \bar{c} is mainly fixed by w of the junctions, the ε/t ratio can gain new insight into the nature of the junction barrier. We have previously reported a scaling law ($\varepsilon/t \times w \approx \text{const}$) for symmetrical 24° [001] bicrystalline substrates. However, other geometries like asymmetric 45° [100] do not follow this law. In this sense, a more detailed comparative study in terms of the junction capacitance reveals that the nature of the barrier may change from a filamentary model description to a tunnel context. On the other hand, we suggest that current-step resonances are mainly affected by the capacitance of regions very close to the crystallographic grain boundary (thickness t), while the hysteresis in the I-V curve may be also affected by the capacitance of the adjacent depleted layers (thickness $l_d > t$).

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