Technology for $YBa_2Cu_3O_7$ SNS- and SIS-Josephson Junctions

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***Abstract*—Ramp-edge Josephson junctions from the high-temperature superconductor $YBa_2Cu_3O_7$ require a multilayer preparation process. In order to increase the reproducability of the junction characteristics we investigate the subprocesses using statistical methods for the design of experiments. The optimization of the process parameters enables us to prepare ramp-edge Josephson junctions with $PrBa_2Cu_3O_7$ as barrier material which exhibit tighter barriers. Adapting the growth conditions for $YBa_2Cu_3O_7$ on MgO films we prepared Josephson junctions with a 10 nm MgO barrier.**

I. Introduction

The ramp-edge technology is a convenient concept for the realization of Josephson junctions from thin films of $YBa_2Cu_3O_7$ [1]. It allows for the free placement on the chip and requires no specially prepared substrates as with bicrystal or step-edge junctions. Furthermore, the characteristics of the junctions can be controlled by the choice of the barrier material and the thickness of the barrier.

A considerable problem of the ramp-edge technology is its need for a well defined barrier growth on the edge in order to obtain a high reproducibility. At least two deposition steps with their respective patterning procedures contribute to the variation in the characteristics of the Josephson junctions.

II. Experimental details

A. Design of experiments

For our study of the preparation process we use statistical methods for the design of experiments (DOE) [2]. This enables us to separate the significant parameters and to give a quantitative description of their effects and the effects of the interactions between the parameters. Although statistical methods in the design of experiments provide information with a comparatively small number of experimental runs, the total number of parameters involved in the preparation of ramp-edge Josephson junctions is too large to allow the study of the entire process at once. We therefore investigated the subprocesses of film deposition, photolithography, and etching separately, assuming them to be independent.

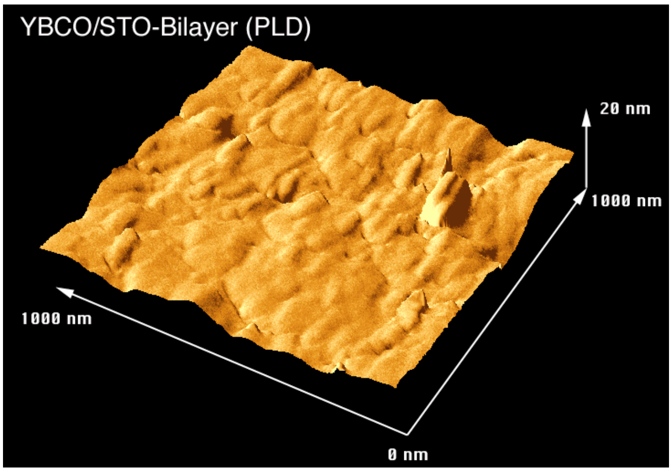

Fig. 1. Surface of a bilayer from 200 nm $YBa_2Cu_3O_7$ and 50 nm $SrTiO_3$ measured by atomic force microscopy. The RMS roughness is 1 nm.

B. *Deposition of* $YBa_2Cu_3O_7$ *and* $SrTiO_3$

We employ pulsed laser deposition (PLD). The KrF-eximer laser (λ =248 nm) with a pulse width of 25 ns is focused onto the target on an area of 16 mm^2 under 45° angle of incidence. Target materials can be changed in situ. The targets are polycrystalline pellets with a diameter of 18 mm which are rotated during deposition. Their surfaces are preconditioned by polishing with abrasive paper and ablating with 300 laser pulses, hitting every spot approximately 20 times. The target-substrate distance is fixed at 100 nm on-axis. The substrates are (100)-oriented $SrTiO_3$-single crystals with a

TABLE I
Deposition Parameters for $YBa_2Cu_3O_7$ and $SrTiO_3$ Films

Parameter		$YBa_2Cu_3O_7$	$SrTiO_3$
substrate temperature	(°C)	815	815
oxygen pressure	(Pa)	8	40
energy density on target	(J/cm^2)	2.0	1.7
laser pulse frequency	(Hz)	3	3

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size of 10 mm × 10 mm × 1 mm. They are glued with silver paste to ceramic carriers by which they are mounted to the heater. A pyrometer is used to measure the substrate surface temperature. The oxygen pressure is controlled dynamically by regulating the oxygen flow.

An investigation with DOE of the deposition process for $YBa_2Cu_3O_7$ films revealed the optimal parameter set given in Table I. This parameter set yields $YBa_2Cu_3O_7$ films which grow c-axis and in-plane oriented with critical temperature $T_c = (88.6 \pm 1.6)\,\mathrm{K}$ and smooth surfaces with less than 2500 outgrowth per square millimeter [3]. $SrTiO_3$ films were optimized with respect to good overgrowth conditions for $YBa_2Cu_3O_7$, a good thickness homogenity and smooth surfaces. With the parameters listed in Table I we obtain smooth $YBa_2Cu_3O_7/SrTiO_3$ bilayers with an RMS roughness of 1 nm over an area of $1\,\mu\mathrm{m}^2$ which allow subsequently deposited $YBa_2Cu_3O_7$ films to grow in the same quality as on the substrate [4]. Such a bilayer is shown in Fig. 1.

C. Patterning

The patterning process consists of conventional photolithography and argon-ion etching in a parallel-plate reactor. For the photoresist process its dimensional stability, the height of the resist structures, smoothness of the edges, and their angle to the substrate surface are the investigated characteristics. An earlier investigation with Hoechst resist AZ5214 and developer AZ312 revealed a dominating sensitivity of all the resist characteristics to variations of the developer concentration between 50% and 60%. To avoid the implied problems for reproducible results we chose the Shipley resist S1813 and developer MF319 which can be used undiluted. For this resist-developer combination no significant influence of the development time is found. The dimensional stability depends mainly on the exposure time which exhibits a strong interaction with the substrate, ranging from an optimal value of 8 s on a fresh $SrTiO_3$ substrate to 12 s on a 200 nm $YBa_2Cu_3O_7$ film. This effect can be attributed to the reflectivity of the surface. Accordingly, the exposure time has to be adapted for multilayers of various film thicknesses. The resist thickness is unaffected by exposure and developing time, indicating that no removal of unexposed resist occurs, variing between $1.75\,\mu$m and $1.55\,\mu$m for rotation speeds between 4000 RpM and 5000 RpM. The roughness of the resist edges is neglegible and unaffected by parameter variation. Also no significant change in the angle between the resist edges and the substrate prior to postbaking was found, ranging from 60° to 65°. That slope softens to angles between 45° and 55° after baking the resist structures for 5 minutes at temperatures between 120°C and 140°C on a hotplate. Table II lists the optimal parameters for the photoresist process.

TABLE II
PARAMETERS FOR PHOTORESIST AND ETCH PROCESS

Parameter		Setting	
rotation speed	(RpM)	4000	
softbake temperature	(°C)	100	
softbake time	(min)	30	in oven
exposure intensity	(mW/cm^2)	25	at 365 nm
exposure time	(s)	8-13	substrate dependent
developing time	(s)	50	
roundbake temperature	(°C)	140	
roundbake time	(min)	5	on hotplate
HF power	(W)	140	
argon pressure	(Pa)	2	

TABLE III
DEPOSITION PARAMETERS FOR $PrBa_2Cu_3O_7$ FILMS

Parameter		investigated range	optimal setting
substrate temperature	(°C)	815 – 850	815
oxygen pressure	(Pa)	3 – 8	8
energy density on target	(J/cm^2)	1.7 – 2.3	1.7
laser pulse frequency	(Hz)	1 – 5	1

The argon-ion etching process was investigated with respect to the slope of the edges in $YBa_2Cu_3O_7$ and the smoothness of the etched surfaces. Variing the HF power from 60 W to 140 W and the argon pressure from 0.5 Pa to 2 Pa we find that the surface roughness is mainly due to the roughness of the etched film and not significantly depending on the etching parameters. The angles of the $YBa_2Cu_3O_7$ edges range from 12° to 21°, predominantly as a function of the etch rate of the $YBa_2Cu_3O_7$ which increases with HF power and argon pressure. Since the angle of the resist structures shows no significant influence we assume that the resists experiences further rounding through heating from the plasma, leading to uniform angles independent of the prior treatment. As a result the angle of the $YBa_2Cu_3O_7$ edges α_{YBCO} is controlled entirely by the etch parameters via the ratio of the etch rates of the resist and $YBa_2Cu_3O_7$, r_{resist} and r_{YBCO} and the angle of the resist structure α_{resist} according to $\tan\alpha_{\mathrm{YBCO}} = \frac{r_{\mathrm{YBCO}}}{r_{\mathrm{resist}}} \tan\alpha_{\mathrm{resist}}$. With the parameter set given in Table II we obtain $\alpha_{\mathrm{YBCO}} = (20 \pm 1)°$ and roughnesses of well below 5 nm RMS on the etched substrate surfaces.

III. JOSEPHSON JUNCTIONS WITH $PrBa_2Cu_3O_7$ BARRIER

A. Optimization of $PrBa_2Cu_3O_7$ *films*

$PrBa_2Cu_3O_7$ is a convenient barrier material for ramp-edge Josephson junctions due to the similar crystal structure with $YBa_2Cu_3O_7$, allowing for good overgrowth conditions. A problem often encountered is the tendency to grow a-axis oriented which would cause the top $YBa_2Cu_3O_7$ layer to grow a-axis oriented as well. Accordingly, the investigation of the $PrBa_2Cu_3O_7$ growth parameters was carried out with respect to the crystalline and morphological quality of the films, deposited with a

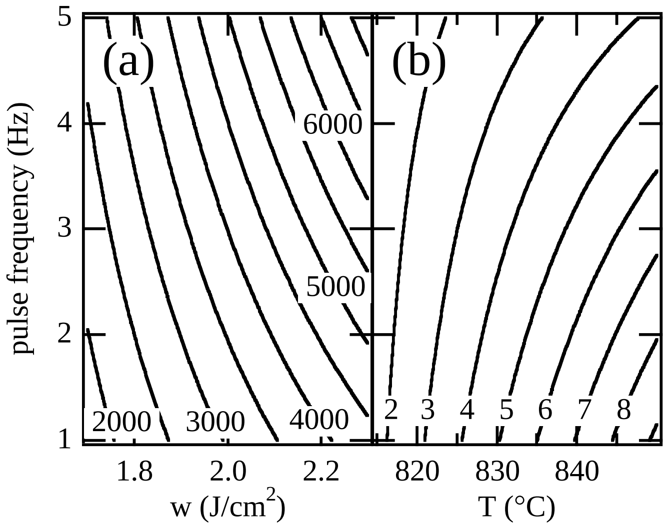

Fig. 2. a) Outgrowth density in outgrowth per square millimeter and b) RMS roughness in nanometer of $PrBa_2Cu_3O_7$ films, in dependence of their significant parameters pulse frequency and energy density w and substrate temperature T, respectively.

fixed number of 1000 Pulses. The investigated parameter range and the optimal settings are listed in Table III. The epitaxial quality was determined by Raman spectroscopy. All the films are almost completely c-axis oriented with an in-plane oriented fraction of above 90%, variing within the error of the measurement of 5% [5]. The outgrowth densities increase with the energy density on the target and the pulse frequency, yielding less than 2000 outgrowth per square millimeter for the optimal setting. The film thickness is a function of the energy density, the oxygen pressure, and a strong interaction between them, resulting in a deposition rate of 136.5 nm per 1000 pulses for the optimal parameter set. No correlation between the outgrowth density and the film thickness was observed. The surface roughness on an area of $25\,\mu$m increases substantially with the substrate temperature, exhibiting a significant interaction with the pulse frequency to the effect that low settings of these parameters improve the RMS roughness from 10 nm to 2 nm. A positive effect of a higher oxygen pressure is less significant, but a strong interaction with the substrate temperature can be observed. At 3 Pa and 850°C parts of the film surfaces exhibit film roughnesses exceeding the generally obtained values by two orders of magnitude, independent of the energy density and pulse frequency. This qualitative change in the surface morphology is obviously due to partial melting. The dependence of the outgrowth density and the surface roughness on their most significant parameters is depicted in Fig. 2. Fig. 3 summarizes the relative importance of the investigated parameters for the film characteristics.

B. Junction characteristics

In order to evaluate the effect of the independently optimized parameter sets on the junction characteristics a series of chips was prepared at a fixed barrier thickness of 40 nm $PrBa_2Cu_3O_7$. At 77 K we measured the critical current I_c and the normal state resistance R_N of various ramp-edge Josephson junctions with a width of $5\,\mu$m. The obtained values of the critical current of $I_c = (6.7 \pm 3.6)\,\mu$A and the normal state resistance $R_N = (1.3 \pm 0.4)\,\Omega$ are within the expectations for Josephson junctions prepared with the unoptimized process [6], but exhibit a reduction in the spread by more than one order of magnitude. The observed spread in I_c and R_N is partially due to the uncertainty in determining these values from noise rounded V-I characteristics of Josephson junctions operated close to their critical temperature. The resistance vs. temperature measurements reveal for all the junctions a residual resistance of about 1 Ω between the transition temperature of the $YBa_2Cu_3O_7$ films of above 86 K and temperatures close to 77 K. This foot-like structure could already be observed prior to the optimization, reaching to less than 3 K below the transition temperature of the $YBa_2Cu_3O_7$ electrodes. Despite thermal rounding the V-I characteristics generally exhibited a RSJ-like shape, an observation rarely occuring with the unoptimized process.

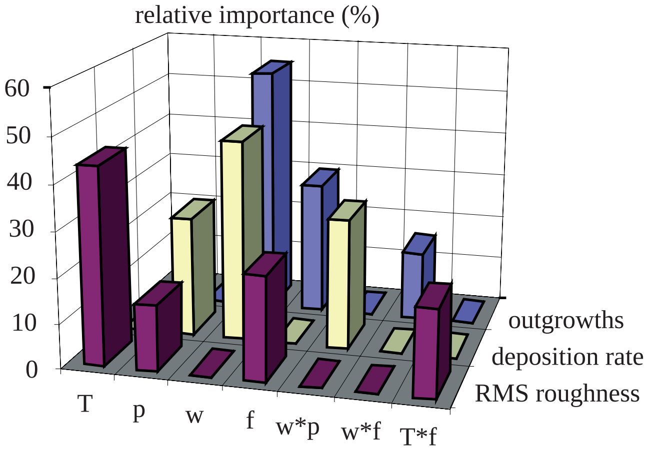

Fig. 3. Summary of the effects of the investigated parameters substrate temperature T, oxygen pressure p, energy density w, pulse frequency f, and their interactions on the outgrowth density, deposition rate, and RMS roughness of $PrBa_2Cu_3O_7$ films.

We conclude from the reduction in spread, the extended residual resistance, and the improvement towards RSJ-like behavior of the junctions that the optimized process yields effectively tighter barriers. Nevertheless, the spread in the junction characteristics has to be reduced further. A study of the interaction of the ramp preparation and its overgrowth by the barrier and top electrode is necessary, giving up the assumption of independent subprocesses.

IV. JOSEPHSON JUNCTIONS WITH MgO BARRIER

A. Deposition of $YBa_2Cu_3O_7$ *on* MgO *films*

The use of MgO promises an insulating behavior of the barrier in ramp-edge Josephson junctions. In contrast to the other materials MgO films are deposited reactively,

TABLE IV

DEPOSITION PARAMETERS FOR MgO AND $YBa_2Cu_3O_7$ ON MgO FILMS

Parameter		MgO	$YBa_2Cu_3O_7$
substrate temperature	(°C)	700	750
oxygen pressure	(Pa)	0.1	12
energy density on target	(J/cm^2)	2.5	2.0
laser pulse frequency	(Hz)	3	3

ablating from metallic targets. The material grows as MgO in an atmosphere of about 0.1 Pa oxygen [7]. The overgrowth of the barrier layer by the top $YBa_2Cu_3O_7$ electrode had to be adopted. With the standard deposition parameters given in Table I the $YBa_2Cu_3O_7$ films grow *a*-axis oriented with a rough morphology. An investigation of the $YBa_2Cu_3O_7$ deposition process with DOE, variing the substrate temperature and the oxygen pressure, showed that a significant improvement is achieved by reducing the deposition temperature and increasing the oxygen pressure, the latter assuring *c*-axis oriented growth. Table IV lists the deposition conditions for smooth MgO films and their overgrowth by $YBa_2Cu_3O_7$ films.

B. Junction characteristics

A chip prepared with a 10 nm MgO barrier and the adopted growth conditions for the top $YBa_2Cu_3O_7$ electrode exhibited V-I characteristics as depicted in Fig. 4. At zero bias a differential resistance of approximately 500 Ω was observed over a temperature range from 80 K down to 30 K. At 4.4 K near 9 mV a hysteretic jump towards the normal conducting line occurs, shifting to lower voltages and smoothing with increasing temperature.

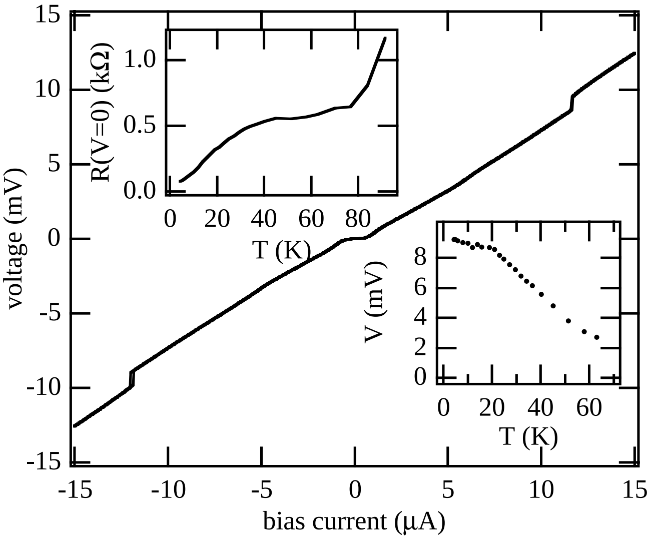

Fig. 4. V-I characteristic of a ramp-edge Josephson junction with a 10 nm MgO barrier at 4.4 K. The insets show the residual differential resistance of about 500 Ω at zero bias between 30 K and 80 K and the temperature dependence of voltage of the hysteretic jump.

V. CONCLUSIONS

We have investigated the multilayer preparation process for ramp-edge Josephson junctions using statistical methods for the design of experiments. Assuming them to be independent, the subprocesses were examined separately. The optimization with respect to the quality of $YBa_2Cu_3O_7$ and $SrTiO_3$ films and bilayers, a stable patterning process, and the growth of smooth $PrBa_2Cu_3O_7$ barriers yield junctions with significantly tighter barriers and a reduced spread in the junction characteristics. An adaption of the barrier thickness and a further reduction of the spread is necessary for applications as integrated magnetometers [8], along with an investigation of the overgrowth of the ramp edges, for which independence of the subprocesses can no longer be assumed. For MgO barriers the required adaption of the overgrowth process for the top $YBa_2Cu_3O_7$ electrode was obtained. A junction with a 10 nm MgO barrier exhibits characteristics suggesting a SIS-Josephson junction with a high density of localized states in the barrier material.

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