Optimization of step-edge substrates for high- T_C superconducting devices

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Abstract

To pursue step-edge for high- T_C superconducting grain boundary junctions or SQUIDs with high reproducibility and quality, we have developed two-step procedures of fabricating very good step-edge substrates. A protocol of precisely controlling step angles for Josephson junctions has been established, with which we can predict the step angles as well as get a better control of the fabrication process. The procedures can improve the step ramp quality substantially. There are no needles, waves, trenches, cascade, or other flaws on these surfaces. The step substrates present good uniformity with respect to any step angle. We have characterized high- T_C step-edge dc SQUIDs connected in series. The I – V curves of SQUID arrays show the RSJ behavior. The enhanced modulation amplitude of 110 μ V is achieved at 77 K with the step-edge dc SQUID in series. The results clearly show the high uniformity and quality of the fabricated step-edge Josephson junctions.

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1. Introduction

The high-transition temperature superconductor (HTS) step-edge Josephson junctions have

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been investigated intensively due to the promising device characteristics [1] comparable to the bi-crystal junctions. To date, most commercial applications use bi-crystal Josephson junctions made on $\text{SrTiO}_3(100)$ (STO) or MgO substrates. Although it is easy to fabricate bi-crystal junctions, the junction locations are restricted to the grain boundary of the bi-crystal substrate. It is not the case for the step-edge substrate, their locations of the junction can be chosen at your will and the pricing is only a tenth of that of the bi-crystal substrates. Moreover, the artificial bi-crystal grain boundary is vulnerable to the thermal and chemical processes in fabricating the Josephson junctions. The thermal heating during film deposition, annealing in the cooling process, or wet etching with chemicals may lead to the formation of grooves along the bi-crystal line [2–4]. The R – T curves and $I_c R_n$ product reveal sequential destructive deterioration of the junction originating from the underlying grooved substrate [2]. In addition, the long-term stability of the bi-crystal Josephson junction is susceptible to the thermal shock in cooling the device from room temperature. The above problems encountered by the bi-crystal junction can be solved with the step-edge Josephson junction [5–12].

The step-edge junction typically has two or more grain-boundaries. Several factors may affect the formation of a high-quality step-edge Josephson junction. The step-edge ramp surface must have as great an epitaxial template quality as the rest of the substrate surfaces to prevent disorder, such as additional grain-boundaries and a -axis oriented particles in the growing film. Further, the step has to be very straight to avoid the nucleation of the CuO particles [6,7]. Thus the fabrication of the substrate steps of the desirable quality puts heavy demand on the processing technique. A number of methods were proposed to produce step-edge substrate [8–10]. The purpose is to obtain a smooth ramp surface with straight step-edge lines. However, the step fabrication process to date still suffers from the problem of low reproducibility due to the strict conditions for different angle steps. Furthermore, the film growth mechanism is variable on different substrates such as STO and MgO. A step with a steep angle greater

than 60° and a clear, sharp profile is the best structure for step-edge junctions on STO. On the other hand, ramp-edge SNS junctions and junctions fabricated onto MgO step-edge substrates require low step angles. To obtain a smooth ramp surface with a straight step-edge line for different step angles is crucial in the success of the high-quality Josephson junctions and SQUIDs for large-scale integrated circuits. Some groups achieved high step angles by controlling different milling rates and re-deposition rates of the substrate with the low-milling rate Cr mask [11]. The possibility to fabricate the smooth step-edge with various angles by using simply the photoresist mask is not clear yet. In this work, we have established two-step procedures to obtain high-quality step-edge substrates. To pursue the high reproducibility for variable step angles, a protocol for fabricating step substrate for Josephson junction and SQUIDs has been established, with which we can predict the different step angles as well as get a better control of the fabrication process. Furthermore, to demonstrate that these step-edges on STO and MgO substrates are uniform, we fabricated serial SQUID arrays onto the step-edge substrates.

2. Experimental

The step-edge substrates were fabricated by ion beam etching (IBE). The IBE source comes from the Commonwealth Scientific Corporation. The fabrication of step-edge substrate requires a masking layer on substrate to define the location of the step-edge during IBE process. Here, we choose the photoresist AZ1500 as the masking layer and defined the step-edge pattern by photolithography. The patterned photoresist mask was hard baked at 100°C in an oven for 1 h in order to reduce its milling rate in the subsequent IBE process. Substrates were mounted on a water-cooled copper sample holder, and a 500 eV argon ion beam with a current density of 1.0 mA/cm^2 was used to etch the substrates. The milling rate of the substrate is about 2 nm/s for both MgO and STO substrates in all our experiments. The pressure during the etching is about $1.8 \times 10^{-4}\text{ Torr}$. The detailed process was reported in [6]. The surface morphology

and step angle of the fabricated substrates were probed by atomic force microscopy (AFM). The YBCO films were grown onto the step-edge substrates using the magnetron sputtering technique with a ratio of the step-height to the thickness of the YBCO film of about 0.7. Typically, the step-height is 200 nm and the film thickness is about 140 nm. Gold contact pads were evaporated onto the contact leads to ensure a low contact resistance. The junctions and SQUIDs were fabricated by the standard photolithography and argon ion milling. The current versus voltage (I – V) and voltage versus flux (V – Φ) characteristics of junctions and serial SQUIDs were measured using NKT's DC SQUID Catcher™ system at 77 K.

The resultant step angle on the substrate could be predicted from the following geometrical relations. In Fig. 1(a) we show the schematic plot of the correlation between the substrate and the incident ion beam direction. We defined the incident angle α to be the angle between the normal of the substrate and the direction of the incident ion beam. The rotational angle θ is defined to be the angle of the substrate rotated about the normal of the substrate. Fig. 1(b)–(d) illustrate the substrate orientations with the rotational angles θ increased from 0° to 90° while the incident angle α is fixed. For the general case shown in Fig. 1(c), it is straightforward to find the geometrical correlation between the incident angle α , the

rotational angle θ and the shading angle ϕ . The relation is

$$\tan(\phi) = \frac{h}{s} = \frac{h}{r} \cdot \frac{r}{s} = \frac{1}{\tan(\alpha) \sin(\theta)}, \quad (1)$$

where h is the thickness of photoresist mask, r is the projection of ion beam ray on the substrate surface, and s is the projection of r on the y' -axis. According to Eq. (1), the maximum shading angle ϕ is 90° when θ is zero as shown in Fig. 1(b). The minimum shading angle appears at $\theta \sim 90^\circ$ as shown in Fig. 1(d). The resultant step angle η should be a single-valued function of the shading angle ϕ for a given incident angle α . The proper incident angle α should make the ratio in milling rate of photoresist to substrate as small as possible. In addition, the rotational angle θ should not be negative in order to minimize the re-deposition of the substrate material onto the step-edge [11]. By considering all the factors above, the resultant step angle η should increase monotonically with respect to the shading angle ϕ . Accordingly, we further assumed that resultant step angle η is proportional to the normal projection of flux of the incident ion beam. In other words, η is proportional to $\sin(\phi)$ as follows

$$\eta = A \sin(\phi), \quad (2)$$

in which A is the highest possible angle we can obtain according to the relation between the milling rate and the re-deposition rate when θ is 0° [11]. Note that the highest possible angle A is a function of the incident angle α .

3. Results and discussion

According to a previous report [12], it was found the fastest milling rate of about 2 nm/s is achieve for the substrate when $\alpha = 60^\circ$. In addition, the ratio in milling rate of photoresist to substrate is minimized in this case. Hence, the incident angle α was fixed at 60° in our subsequent experiments to define the step-edge. Under a protocol ion milling processes and optimised condition of exposing the photoresist, the MgO and STO step substrates were fabricated successfully. The step-edge profile is checked on $5 \times 5 \mu\text{m}^2$ by AFM. In

Fig. 1. (a) A schematic plot of the correlation between the substrate position and ion incident direction. Schematic plot when (b) $\theta = 0^\circ$, (c) θ is between 0° and 90° , and (d) $\theta = 90^\circ$.

Fig. 2. Surface morphology of the fabricated step-edge substrates probed with AFM when (a) $\theta = 30^\circ$ and (b) $\theta = 90^\circ$.

Fig. 2 we show the surface morphology of the fabricated step-edge substrates when the rotational angles θ were kept at 30° and 90° , respectively. It is found that the ramp surface and the surface near the step-edge line were filled with the re-deposited material near the upper edge of the step. The location of re-deposition is getting far away from the step-edge line when θ is getting larger.

To suppress any re-deposition whatsoever, the second process was adapted. We rearranged the

substrate on the holder and used the ion beam to sweep away the re-deposition materials. The angles α and θ in Fig. 1(a) are set to 90° and 0° , respectively. The milling time is about 10 s. We found that the re-deposited material was removed by using the second procedure. The ramp surface becomes smoother and the step-line becomes straighter with this way. Fig. 3(b) and (d) showed the improved results of Fig. 2(a) and (b) after the second process. The two-step procedure describe above was successfully applied to fabricate the step-edge of angles varying from 30° to 70° on the MgO and STO substrates. In Fig. 3(a)–(d), we showed the surface profile of typical results. The fabricated step-edge substrates have smooth surface and straight ramp-edge. There are no needles, waves, trenches, cascade, or other flaws on these surfaces. We compare the fabricated step angles with that predicted from Eqs. (1), (2), and the results are shown in Fig. 4. When the rotational angles θ were fixed at 0° , 10° , 20° , 30° , 60° and 90° , we obtained the step angles η of 72° , 65° , 50° , 45° , 30° and 28° , respectively for MgO substrates. The same rotational angles θ give the step angles η of 72° , 64° , 50° , 43° , 28° , and 20° , respectively for STO substrates. The maximum step angle of 72° is close to the angle as described above and that in Chu's predication [11]. Comparing

Fig. 3. (a)–(d) Surface morphology of the step-edges with various step angles η . The images are taken by an atomic force microscope scanning across a $5 \times 5 \mu\text{m}^2$ area.

Fig. 4. The theoretical and experimental step angles η with respect to various rotational angles θ .

the fabricated step angles on STO and MgO substrates with those predicted from Eq. (2), we found a good consistency between the predicted data and the fabricated results.

To demonstrate the good quality of the fabricated step-edge substrates, the serial SQUID arrays were fabricated onto the MgO and STO step-edge substrates. The detailed process was reported in Ref. [6].

We fabricated serial SQUIDs on the MgO step substrates shown in Fig. 3(b). The substrates used were low angle MgO(100) substrate. The step-height was 200 nm with a step angle of 50°. The film thickness is about 140 nm. The ratio of the step-height of the substrate to the thickness of the film is 0.7. The bare SQUID were connected in series to form the serial SQUID array. The total number of SQUIDs in the array was designed to be 50. The distance between two SQUID was 15 μm . The area of the hole of the SQUID was $10 \times 20 \mu\text{m}^2$, and the junction width was 3 μm . The calculated inductance of the SQUID loop is 35 pH. 50-SQUID serial arrays were aligned along a step-edge line of 1550 μm long. Fig. 5 shows I - V characteristics for 5, 15 and 35 SQUIDs connected in series. The inset of Fig. 5 shows typical I - V characteristic of a dc SQUID at 77 K. The critical current and the shunt resistance of the bare SQUID and the SQUID array were found by fit-

Fig. 5. The inset of the plot shows that the I - V curve of a single SQUID is like the RSJ behavior. I - V characteristics of 5, 15 and 35 SQUIDs connected in series at 77 K.

ting to the RSJ model with finite thermal noise [13]. It is found that the single SQUID shows a normal resistance of 2.1 Ω and a critical current of 55 μA , corresponding to the $I_C R_n$ product of 113 μV at 77 K. The normal-state resistances for the 5, 15, and 35 dc SQUID arrays were found to be 12, 25, and 36 Ω , respectively with the critical currents of about 30 μA . In Fig. 6 we show the V - I_{mod} curves at 77 K for a bare SQUID and 5, 15, 25, 35 SQUIDs connected in series. The peak-to-peak voltage, V_{pp} , of a single SQUID is about 20 μV . The observed V_{pp} is increased as the num-

Fig. 6. V - I_{mod} curve characteristics of 1, 5, 15, 25 and 35 SQUIDs connected in series.

Fig. 7. (a) $V-I_{\text{mod}}$ curve of a 5-SQUIDs array, (b) Simulated beat phenomena, and (c) beat phenomena is a superposition of five distinct $V-I_{\text{mod}}$ curves with 3.5% difference in the modulation period.

ber of SQUIDs connected in series is increased. The observed V_{pp} is a bit smaller due to the incoherent modulation in the $V-I_{\text{mod}}$. To confirm this we replot in Fig. 7(a) the measured $V-I_{\text{mod}}$ curve of a serial 5-SQUIDs array in a wider range of the applied modulation magnetic field [14]. If we superimposed five distinct $V-I_{\text{mod}}$ curves with 3.5% difference in the modulation period and 5% difference in the amplitude shown in Fig. 7(c), we obtained the data shown in Fig. 7(b) which is similar to the measured data. We believe that the non-uniform field causes an incoherent phase relation in $V-I_{\text{mod}}$ curves in SQUIDs. Each SQUID has a small variation reveal in the difference of amplitude.

DC SQUIDs are composed of two junctions connected in series. Besides, the two junctions must be the same so as to obtain the symmetric interference effect. This is why most junctions in dc SQUIDs are fabricated as close as possible to keep the junction properties the same. Because of the above reasons, the step-line needs to be long enough to accommodate two junctions and consistent enough to keep these two junctions identical. We have fabricated 35 SQUIDs on MgO step and prove that the step is uniform up to at least 1000 μm .

Fig. 8. Flux noise of serial connected SQUIDs array at 77 K. (a) A single SQUID; (b) 5 SQUIDs; (c) 15 SQUIDs; (d) 25 SQUIDs and (e) 35 SQUIDs.

Fig. 8 shows $S_{\phi}^{1/2}(f)$ for a bare SQUID, 5, 15, 25 and 35 serial connected SQUIDs array. The white noise in $S_{\phi}^{1/2}(f)$ for a SQUID is $60 \mu\Phi_0/\text{Hz}^{1/2}$ while the serial connected SQUID show flat noise spectrum. The $S_{\phi}^{1/2}(f)$ is reduced to $18 \mu\Phi_0/\text{Hz}^{1/2}$. The low frequency $1/f$ flux noises of serial SQUIDs arrays are almost flat. The good characteristics of the low frequency $1/f$ flux noise of SQUID reveal that the step-edge substrate is good. We can obtain the same results of the STO step-edge SQUID.

Corresponding to theory the white flux noise should decrease with $1/\sqrt{N}$ where N is the number

Fig. 9. Compare the flux noise measured with predicted.

of SQUIDs. Fig. 9 showed the flux noise measured and predicted. The flux noise of serial SQUID array was bigger than designed. The cause of the bigger flux noise was the phase incoherence. The $S_\phi(f)$ of serial connected SQUID array was a little bit higher than the data predicts. The phase incoherence of each SQUID should be the cause for higher flux noise was the phase incoherence. We can obtain the same results of the STO step-edge SQUID. By the two procedures, this work also helps the preparation of the ramp surface in ramp-type junction. The results show in [15].

4. Conclusion

We have developed the protocol to fabricate high-quality step-edge substrates using two-step processes. A model of comparing the fabricated step angle with the experiment data was established. We have confirmed the high-quality step-edge substrates using high- T_C step-edge serial dc SQUIDs. The results show the high uniformity and quality of the fabricated step-edge Josephson junctions. The step-edge SQUID exhibit excellently in good result in flux-to-voltage transfer function [16–22].

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