Fabrication of High-Quality Niobium Superconducting Tunnel Junctions *

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For high-quality superconducting tunnel junctions (STJs), it is necessary to reduce leakage current as much as possible. We describe the fabrication of niobium STJs using the selective niobium (Nb) etching process and various ways to minimize the leakage current. The experiment shows that the leakage current mainly comes from shorts in the tunnel barrier layer rather than those around the junction edges. Through systematic analysis of the thin film stress, surface morphology and modified junction structures, we fabricate high-quality Nb STJs with a gap voltage of 2.8 mV and a leakage current at 1 mV as low as 8.1% and 0.023% at 4.2 K and 0.3 K, respectively.

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In comparison with other superconducting tunnel junctions (STJs) made of various combinations of superconducting and barrier materials, Nb/(Al-)AlO_x-Al/Nb STJs[1−3] have inherent high-quality characteristics such as a small leakage current in the sub-gap voltage region and a sharply defined gap voltage.[4] Moreover, its excellent stability against both long-term storage and thermal cycles is particularly useful for cryogenic experiments, and the controllability of its critical current density is convenient for designing superconducting devices and circuits.[5] Nb STJs are widely used in studies of terahertz (THz) detection and macroscopic quantum phenomena.[6−8] For astronomical observations, they can be applied as mixers to detect high-frequency weak signals because of their irreplaceable high performance.[9] Superconducting quantum interference devices (SQUIDs), the most sensitive magnetic field detectors, are mainly also based on Nb STJs. With their advantages of a high intrinsic switching speed and low power dissipation, Nb STJs are considered to be one of the best choices for the next generation of ultra-high performance computers and communications applications.[5] Therefore, it is essential to establish more reliable techniques for producing high-quality Nb/(Al-)AlO_x-Al/Nb STJs to realize the various kinds of applications of Nb STJs in the near future.

Almost all applications of Nb STJs require a low leakage current, which is one of the most important parameters of high-quality junctions. For instance, quantum control and quantum computation[10,11] are usually operated by Nb STJs and a high leakage current will make the decoherence time of the qubit shorter and therefore produce worse results.[12] When Nb STJs are used as terahertz direct detectors,[13,14] a low leakage current increases the sensitivity of detection. In this Letter, following a brief introduction of the fabrication process of Nb STJs, we report our experiments, which are designed to investigate the sources of junction leakage current and improvements of film fabrication and barrier quality.

Table 1. Parameters of reactive ion etching.

Power density	Gas	Pressure	Gas flow	Time	Thickness
0.24 W/cm^2	SF_6	4 Pa	10 sccm	60 s	750 Å

The fabrication process of Nb STJs is similar to the selective niobium (Nb) etching process (SNEP) developed by Kroger and Gurvitch *et al.*,[6,15] which is shown in Fig. 1. Firstly, trilayers 750 Å/100 Å/1500 Å were respectively deposited by dc magnetron sputtering in a main chamber and oxidation was completed in another load lock chamber. Secondly, junctions were defined by reactive ion etching (RIE) in SF_6 gas. Table 1 shows the RIE conditions. There was 2000 Å SiO_2 deposited at 50℃ by plasma enhanced chemical vapor deposition (PECVD), which has advantages over other traditional insulators. Compared with rf sputtering for the SiO_2, PECVD has no high-energy particle bombardment of Nb STJs, so this process will cause the least harm to the STJs, making it easier to control the critical current density J_c. Also in terms of the evaporation coating of SiO, a SiO_2 film prepared by PECVD is smoother. In particular, it has denser step-edge coverage,[16] which is more conducive to the fabrication of high quality Nb STJs. Finally, there is a final photolithography process and wire Nb layer deposition by the sputtering system.[2]

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The current-voltage (I–V) curve of a $6 \times 6\,\mu m^2$ Nb/AlO_x-Al/Nb STJ shows that the leakage current at 1 mV is high, accounting for over 30% of the critical current. This poor-quality STJ cannot be applied to quantum computing or for the detectors in direct detection.

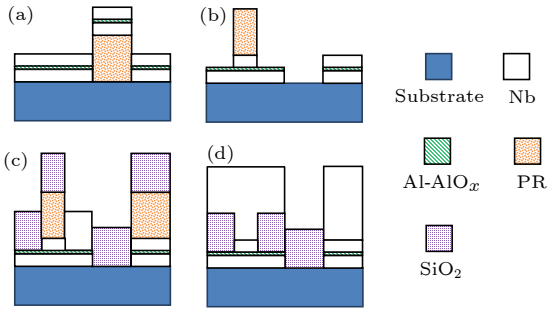

Fig. 1. Fabrication process of Nb superconducting tunnel junctions.

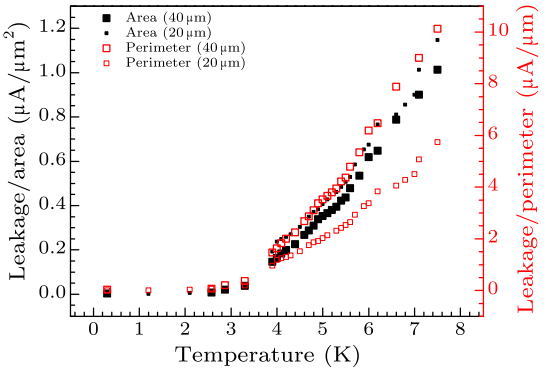

Fig. 2. Leakage current at 1 mV per unit area and per unit perimeter.

Generally, the existence of tiny pinholes in the barrier layer and the generation of shorts around the edge of junctions are the most probable reasons for a high leakage current. We measured the leakage current of two typical junctions of different sizes over temperature to verify the sources of the unexpected leakage current. The leakage current per unit area and per unit perimeter are shown in Fig. 2. As the temperature increases, comparing these two junctions, the trend of leakage current per unit perimeter is obviously different while the leakage current per unit area remains the same. The experimental results show that the leakage current is proportional to the area. Therefore, we can derive a conclusion that the leakage current is caused by the defects in the barrier layer rather than those around the edges of the junctions. These results prove that the isolation layer of the SiO_2 film deposited by PECVD has excellent quality.

To obtain a perfect barrier, the quality of the Nb film and the Al film should be improved. For superconducting materials, film stress and film surface morphology are two particularly important parameters used to evaluate the quality of films, as well as the critical temperature T_c.

During superconducting thin film deposition, residual stress can deform and degrade the barrier layer through bending around the edge, bugle holes etc. and eventually induce extra leakage current. Consequently, the base and upper Nb electrodes join together and thus the circuit is shorted. Thus it is very important to optimize Nb and Al film stress in order to minimize barrier leakage.

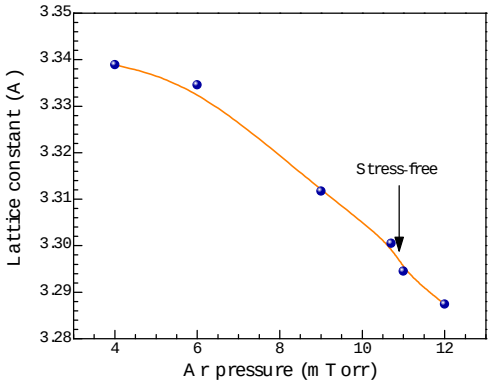

Fig. 3. Relationship of Ar pressure and Nb film lattice constants.

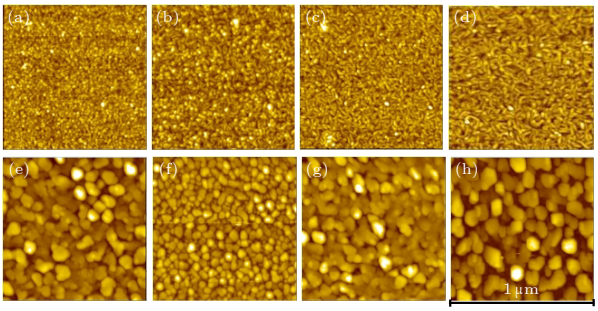

Fig. 4. AFM photographs of Nb and Al films fabricated with different powers. Nb films: scan area $1 \times 1\,\mu m^2$, (a) 200 W, (b) 220 W, (c) 245 W, (d) 200 W, reducing the distance between the target and the substrate by 5 cm. Al films: scan area $1 \times 1\,\mu m^2$, (e) 15 W, (f) 30 W, (g) 50 W, (h) 100 W.

The Ar pressure during dc magnetron sputtering is considered to be the most important factor affecting Nb and Al film stress.[17] In order to find the stress-free condition for Ar pressure, we deposited Nb films at different Ar pressures. High-resolution x-ray diffraction (XRD) was used to measure the lattice constants of the films. Figure 3 shows the relationship between Ar pressure and the lattice constants. According to a linear relationship of the lattice constants with the film stress,[4] the lattice constant of the stress-free Nb film is 3.295–3.300 Å, which is slightly smaller than that of bulk Nb, and the corresponding Ar pressure is 10.9 mTorr. In the same way, we find that the corresponding Ar pressure for stress-free Al film is

15.5 mTorr.

In addition to film stress, film surface morphology is also one of the most important parameters that directly affect the quality of the barrier layer. A rough film surface is prone to cause pinholes in the barrier layer and leads to a leakage current. Thus we need to find the optimal sputtering conditions for smooth Nb and Al films.

Figure 4 compares atomic force microscope (AFM) photographs of Nb and Al films deposited at the same Ar pressure but at different powers. As the power increases from 200 W to 250 W, the columnar structure gradually appears on the film surface, as shown in Figs. 4(a)–4(c), which can lead to pinholes in the film[4] and cause leakage current. We learn that the film in Fig. 4(a), deposited at 200 W, has the smoothest surface. When the distance between the target and the substrate is reduced by 5 cm, the columnar structure appears again in Fig. 4(d). The experimental results show that neither higher power nor smaller distance between the target and substrate is helpful for depositing high-quality Nb films in our sputtering system.

Similarly, we obtain the AFM photographs of the Al films on the same Nb film in Figs. 4(e)–4(h). We can see that the grain size is not simply linear to the power. When the power increases from 15 W to 30 W, the grain size becomes smaller and smaller; and the opposite happens when the power increases from 30 W to 100 W. This phenomenon can be explained by the deposition rate. When a low power is applied, the deposition rate is also low. Therefore, small grains are easy to aggregate into larger ones because of the effect from the base Nb film surface. However, when the power shown in Fig. 4(h) is high, the Al grains are themselves large enough to make the surface rough. Finally, the optimal power of the Al film is 30 W. All the parameters of Nb and Al film fabrication are given in Table 2.

Table 2. Fabrication parameters of the Nb and Al films.

Conditions	Nb	Al
Target	3 inch	3 inch
Base pressure	$< 5 \times 10^{-8}$ Torr	$< 5 \times 10^{-8}$ Torr
Ar pressure	10.9 mTorr	15.5 mTorr
Power	200 W	30 W
Deposition rate	500 Å/min	70 Å/min

For further improvement of barrier layer quality in order to reduce leakage current, we also tried to add an Al layer after Al oxidization in order to cover the AlO_x surface. That is to say, we changed the process from SINS into SNINS, namely a Nb/Al-AlO_x-Al/Nb structure.[1,3] The upper Al layer effectively protects the upper Nb film from the diffusion of oxygen atoms from AlO_x and also prevents the reaction between Nb and surface O-H groups,[17] which may cause micro shorted routes between the electrodes. The following experimental results show that it is easier to fabricate high-quality Nb STJs with an SNINS symmetry structure than an SINS structure.

The superconducting transition temperature T_c of 750Å Nb film was about 9.1 K. For the purpose of examining the quality of multilayers, anodization spectroscopy[18,19] was used to test the characteristics of the film interfaces in the Nb/(Al-)AlO_x-Al/Nb structure before and after improvements. The spectrum of the improved detector has a clear maximum, which means that the interface of the upper Nb film and the Al film is relatively clearer than before. Meanwhile, a steeper decrease of dV/dt than before indicates that the interface between the base Nb film and the Al film has been improved significantly.[20]

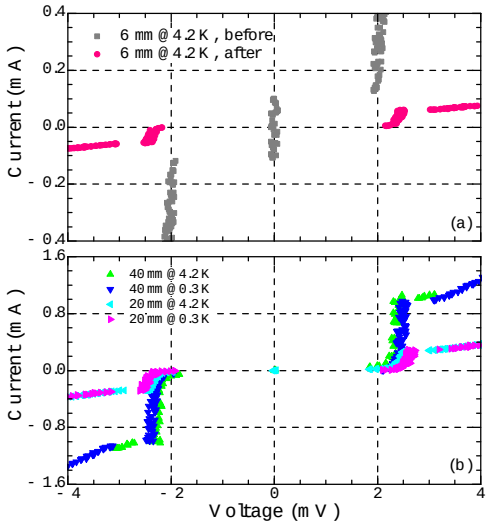

Fig. 5. I–V characteristics of the usual Nb/AlO_x-Al/Nb junctions and Nb/Al-AlO_x-Al/Nb junctions with optimized Nb and Al sputtering parameters to improve film stress and surface morphology. (a) X: 2 mV/div, Y: 0.2 mA/div, size: $6 \times 6\,\mu m^2$. (b) X: 2 mV/div, Y: 0.8 mA/div, size: $20 \times 20\,\mu m^2$ and $40 \times 40\,\mu m^2$.

As shown in Fig. 5, the I–V curves of the usual Nb/AlO_x-Al/Nb junctions and Nb/Al-AlO_x-Al/Nb junctions with optimized Nb and Al sputtering parameters to improve film stress and surface morphology have been measured. After these improvements, the leakage current of $6 \times 6\,\mu m^2$ junction at 1 mV is 25 times lower than before, as shown in Fig. 5(a). In addition, the I–V curves of $20 \times 20\,\mu m^2$ and $40 \times 40\,\mu m^2$, fabricated after improvements, as shown in Fig. 5(b), were measured at 4.2 K and 0.38 K. Comparing the ratio of leakage current at 1 mV over the critical current, we can see that the value after improvements is significantly less than that before. Taking a $20 \times 20\,\mu m^2$ junction as an example, after improvements, the leakage current accounts for 8.1% at 4.2 K and 0.023% at 0.38 K and the quality of the Nb STJ has been greatly improved. Accordingly, our efforts in film stress, sur-

face morphology and SNINS process are highly effective in reducing the leakage current. From the I–V curves, it is obvious to see that the gap voltage is about 2.8 mV.

In conclusion, we have adopted main chamber dc magnetron sputtering metal films and ex-situ barrier layer oxidation in a load locked chamber, and researched and resolved many technology problems about low-leakage-current Nb STJ fabrication. A series of studies on the leakage current issue have been carried out systematically. In order to determine the sources of leakage current, experiments about the relationship between leakage current and area or the relationship between leakage current and the perimeter are designed. We find out that defects in the barrier layer are the main reason for leakage current. Then the film stress, surface morphology and fabrication structures are optimized to improve the quality of barrier layer. The improved Nb film is smooth, stress-free and Nb STJs with stable process repeatability and performance consistency have been developed, whose gap voltage is 2.8 mV at 4.2 K. The anodization spectroscopy and I–V curves of improved Nb STJs verify that the improvements we made are very effective and that the leakage current is greatly reduced. All of these technological breakthroughs have laid a solid foundation for their future application in direct detection, quantum computing, superconducting circuits and so on.

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