Fabrication of aluminum nitride barrier SIS mixer devices using nitrogen plasma diluted by noble gases

Akira Endo\textsuperscript{1,2,3}, Hirofumi Inoue\textsuperscript{2}, Shin’ichiro Asayama\textsuperscript{1}, Takashi Noguchi\textsuperscript{1}, Matthias Kroug\textsuperscript{1} and Tomonori Tamura\textsuperscript{1}

\textsuperscript{1} National Astronomical Observatory of Japan, 2-21-1 Osawa, Mitaka, Tokyo 181-8588
\textsuperscript{2} Institute of Astronomy, the University of Tokyo, 2-21-1 Osawa, Mitaka, Tokyo 181-0015
\textsuperscript{3} JSPS Research Fellow

E-mail: akira.endo@nao.ac.jp

Abstract. Nb/Al–AlN/Al–AlO\textsubscript{x}/Nb SIS junctions have been fabricated by various RF plasma nitridation techniques. The reproducibility of the $R_N A$ product from batch to batch was improved by diluting the N\textsubscript{2} plasma with He or Ar. For the He+N\textsubscript{2} process, a power-law relation between the $R_N A$ product and the nitridation time was found to hold along $10^6-10^8 \Omega \mu \text{m}^2$, which implies that the thickness of the barrier grows logarithmically during the time it is exposed to the plasma. By applying a pulse-time-modulated RF power, we were able to slow down the nitridation significantly. Finally, we present the results of preliminary RF measurements. The Nb/Al–AlN/Al–AlO\textsubscript{x}/Nb SIS mixers were tested at around 150GHz and 350GHz, and showed noise performances comparable to those of Nb/Al–AlO\textsubscript{x}/Nb SIS mixers.

1. Introduction

Superconductor-Insulator-Superconductor mixers (SIS mixers) are widely used in low-noise, high sensitivity heterodyne receivers for radio astronomy. Two important aspects of an SIS mixer’s performance are its noise temperature and relative RF-bandwidth. The “quality factor” of an SIS junction, which is usually defined by the ratio of the junction’s subgap resistance and normal resistance, $R_{sg}/R_N$, is generally required to be at least $\sim 15$ for low noise performance. To achieve a broad RF-bandwidth, the junction must have a very high current density $J_C$ or a small $R_N A$ product [1], where $A$ is the area of the junction. For example, $J_C \geq 15 \text{ kA cm}^{-2}$, or $R_N A \leq 15 \Omega \mu \text{m}^2$, is required to cover the 790-950GHz band with good atmospheric transparency ($\Delta \omega/\omega = 19\%$).

Thus the technology to fabricate SIS junctions with both small leakage current and low $R_N A$ products is the key in the development of high-sensitivity, broadband SIS mixers. While the classical Nb/Al–AlO\textsubscript{x}/Nb barrier SIS junctions tend to be leaky at $R_N A < 20 \Omega \mu \text{m}^2$ [2–4], AlN\textsubscript{x} barrier SIS junctions can have $R_N A$ products as low as $1 \Omega \mu \text{m}^2$ with still acceptable quality, e.g., $R_{sg}/R_N > 10$ [5]. This can be understood as a result of the lower barrier height of AlN\textsubscript{x} barriers compared to AlO\textsubscript{x} barriers ($\phi_{\text{AlN}_x} \sim 0.9 \text{ eV}$ [6], $\phi_{\text{AlO}_x} \sim 2 \text{ eV}$ [7]).

From the practical point of view, the reproducibility of the $R_N A$ product is important in order to increase the device yield. If the process is sufficiently slow, the $R_N A$ should be controllable by the nitridation time of the Al film. Though there have been a number of reports on the nitridation time-dependence of the $R_N A$ product, they do not necessarily coincide with each other. While pioneering studies applied linear fits to their data ($R_N A \propto t_N$)[8; 9], subsequent studies have
Table 1. Conditions used for RF plasma nitridation

| experiment | gas composition | \( \text{N}_2 \) flow (sccm) | \( \text{Ar} \) flow (sccm) | \( \text{He} \) flow (sccm) | pressure (Pa) | power (W) | \( t_{\text{on}} \) (ms) | \( t_{\text{off}} \) (ms) |
|------------|-----------------|------------------|-----------------|-----------------|--------------|----------|----------------|----------------|
| (a)        | 100%\( \text{N}_2 \) | 20-50            | 0               | 0               | 13           | 1        | –              | –              |
| (b)        | \( \text{Ar}+10\%\text{N}_2 \) | 5                | 45              | 0               | 13           | 1        | –              | –              |
| (c)        | \( \text{He}+10\%\text{N}_2 \) | 5                | 0               | 45              | 13           | 1        | –              | –              |
| (d)        | \( \text{He}+6\%\text{N}_2 \) | 3                | 0               | 47              | 0.3          | 1        | –              | –              |
| (e)        | \( \text{He}+50\%\text{N}_2 \) | 25               | 0               | 25              | 13           | 1        | –              | –              |
| (f)        | \( \text{He}+90\%\text{N}_2 \) | 45               | 0               | 5               | 13           | 1        | –              | –              |
| (g)        | 100%\( \text{N}_2 \) | 50               | 0               | 0               | 2            | 10       | 0.1            | 0.9            |
| (h)        | 100%\( \text{N}_2 \) | 50               | 0               | 0               | 2            | 100      | 0.01           | 0.99           |

Col. (1): Sign for each condition; Col. (2): Composition of the process gases; Col. (3): Flow of nitrogen; Col. (4): Flow of argon; Col. (5): Flow of helium; Col. (6): Total pressure; Col. (7): Applied RF power; Col. (8): The time the RF power is turned on during one cycle; Col. (9): The time the RF power is turned off during one cycle.

reported exponential relations \( (R_N A \propto e^{kt_N}) \) [10; 11], or peculiar relations involving plateaus [12]. The growth of the physical thickness of the AlN layer has also been monitored in situ by some studies [10; 13], which have reported growth curves saturating roughly logarithmically. Meanwhile, no theory to our knowledge has been established which predicts the growth rate of the thickness of the AlN\( x \) layer during low energy plasma nitridation.

2. Experiment

We fabricated Nb/Al–AlN\( x \)/Nb SIS junctions with various nitridation conditions, using a process similar to that described in Ref. [14].

The SIS-trilayer was fabricated using the ULVAC CS200ET sputterer. This instrument has a load-locked chamber and a sputtering chamber, which have base pressures of \( 1 \times 10^{-4} \) and \( 1 \times 10^{-5} \) Pa, respectively. The chuck table in the load-locked chamber is round and has a diameter of 360mm. It is connected to an RF source that is used for nitridation and wafer cleaning. The films are deposited by DC magnetron sputtering. The chuck tables in both chambers and the metal targets are cooled by water.

The films were deposited on fused quartz wafers, 35mm in diameter and 0.3mm in thickness. The SIS trilayer consists of a bottom-Nb electrode (200nm), Al-AlN\( x \) bilayer (~10nm) and the top-Nb electrode (100nm). After the deposition of the trilayer, the junction was defined using the Canon FPA-3000 i5+ i-line stepper, and the surrounding trilayer was etched down to the Al layer, leaving the bottom-Nb layer to serve as a groundplane. The Nb wire layer (500nm) was isolated from the groundplane by a layer of sputtered SiO\(_2\), and was connected to the top electrode of the junction by a via-hole either lifted off or etched by ICP. Additional information about the process and instruments used can be found in our previous papers [11; 15].

Between the deposition of the Al and the top-Nb electrode, the wafer was transferred to the load-locked chamber for RF plasma nitridation. The various nitridation conditions (a)-(h) are summarized in Table 1. In condition (a), where pure \( \text{N}_2 \) was used as the process gas, the small power (1W) used for nitridation was not sufficient for igniting the plasma, so a larger power (~6W) was applied at the beginning. In conditions (b)-(f), where the \( \text{N}_2 \) gas was diluted by either Ar or He, only the noble gases were introduced into the chamber for plasma ignition. Once the plasma was ignited, the power was lowered to 1W and nitrogen was introduced.
The "nitridation time", $t_N$ was measured from the moment we opened the nitrogen valve. In conditions (g) and (h), pure N$_2$ gas was used, and the RF power source was programmed to quickly switch on and off with a cycle of 1ms: The power was turned on for 10% and 1% of the 1ms cycle in experiments (g) and (h), respectively.

3. Results and discussion
3.1. Effect of diluting the nitrogen plasma with noble gases
The time-dependence of the $R_NA$ product on nitridation time is presented in Fig. 1. In this section, we compare the results of nitridation conditions (a), (b) and (c) to evaluate the effect of diluting the N$_2$ plasma with Ar or He. In condition (a), where pure N$_2$ was used, the $R_NA$ had very poor reproducibility: The $R_NA$ of junctions nitridized for 200-300s had a large variety of 10$^{-10}$ Ω µm$^2$. As for condition (b) using Ar + 10%N$_2$, a correlation between nitridation time and $R_NA$ can be seen, though the trend of the relation is not so clear. However, the results of condition (c), where He + 10%N$_2$ was used, show a tight power-law relationship between $R_NA$ and $t_N$ of the form $R_NA \propto t_N^k$, which holds for six orders of magnitude in $R_NA$: 10$^{-10}$-10$^6$ Ω µm$^2$.

The better reproducibility achieved by the composite plasma of N$_2$ and noble gases can partly be explained by the way the plasma was ignited. Because in experiment (a) there was nitrogen already in the chamber before the ignition, we can expect that a significant amount of nitridation took place during the unstable period of ignition. On the other hand, in experiments (c) and (d), there was only the nonreactive noble gas in the chamber at the time of ignition, so the nitridation did not begin until we started flowing N$_2$ into the chamber.

3.2. Dependence of the nitridation rate on the composition ratio and the total pressure
In this section, we concentrate on the difference between the results of experiments (c)-(f), also presented in Fig. 1. In all four of these experiments, a composite gas of He and N$_2$ was used.
The major difference between conditions (c) and (d) is the total pressure. By comparing the fitted lines in Fig. 1, we can see that condition (d) with the lower pressure (0.3Pa) exhibits a slower nitridation rate: It takes roughly twice the time to reach a certain $R_N A$ in process (d) with respect to process (c) with a pressure of 13Pa. Interestingly, the difference in the pressure seems to add an offset to the growth line in Fig. 1, rather than affecting the index of the power-law relation. Notice that the power-law index of the trend is considerably smaller in process (b), where Ar was used as the solvent gas.

Next, if we compare conditions (e) and (f) with (a), we see that the nitridation rate of conditions (e) and (f) are similar to that of condition (a), at least at $t_N = 60s$. This indicates that the composition ratio, $N_2/(N_2+He)$, has only a minor effect on the nitridation rate. This should be confirmed by additional experiments with longer and shorter nitridation times.

3.3. Nitridation with a pulse-time-modulated RF input

Fig. 3. Relation between the $R_{sg}/R_N$ factor and the $R_N A$ product. The circles are the results of Nb/Al–AlN$_x$/Nb barrier SIS junctions fabricated with various processes described in Table 1. The triangles and circles are data of Nb/Al–AlO$_x$/Nb barrier SIS junctions fabricated with the same sputterer and a different sputterer in our laboratory, respectively.

The final two experiments presented in Fig. 1 are (g) and (h), where a pulse-time-modulated RF power was used to discharge the pure nitrogen gas. In condition (h), the instantaneous RF power was 10 times of that of condition (g), while the on-time was reduced to 1/10. In both experiments, the time-averaged RF power was 1W and the dc bias voltage was similar to that of condition (c): 5-10V. Though we have only two reliable results so far, we can see that by using this technique the nitridation rate can be made significantly slower. By optimizing the on-time/off-time ratio and the instantaneous RF power, we could probably realize an even slower process.

3.4. Growth mechanism of the AlN$_x$ barrier

From tunneling theory, the $R_N A$ product is an exponential function of the physical thickness of the barrier, $d_{AlN_x}$ [16]. Therefore, a semi-logarithmic plot of the $R_N A$ v.s. $t_N$ relation is homologous to the growth curve of the AlN$_x$ barrier. The results of experiments (c) and (d), the conditions for which we have the most data points, are re-plotted onto a semi-log scale in Fig. 2. The scale on the left ordinate is in units of thickness, which has been calculated from the $R_N A$ product assuming that the barrier height is the same as that measured by Wang et al. [6]. As can be seen, the results of experiment (c) and (d) are fitted well by logarithmic growth curves, which is consistent with optical in situ measurements that have been reported in the past [10; 13].

The growth curves can provide us with information about the growth mechanism of the barrier. For example, a logarithmic growth ($d \propto \log t$) is observed when thin Al films are oxidized at room temperature or below, and is theoretically explained by migration of the metal cations
[17]. On the other hand, oxide layers can grow parabolically ($d \propto \sqrt{t}$) at relatively high energy states (e.g., thermal oxidation of metals at temperatures above $10^3$ K): This is theoretically explained by thermal diffusion [18; 19]. In fact, Iosad et al. have carefully considered the energy of various nitrogen species in the plasma, and concluded that thermal diffusion is the dominant mechanism of nitridation in their process for nitriding Al films [12]. As for our process, both logarithmic and parabolic curves seem to fit the results fairly well, and we cannot at this point determine the dominant mechanism of the nitridation.

By comparing the results of experiments (d) and (g) in Fig. 2, applying the RF power in short pulses seems to be a more efficient way to slow down the nitridation, compared to lowering the pressure of the reactive gas. This is probably because the nitridation becomes intermittent. The slowness of the reaction could be useful in improving the $R_N A$-reproducibility.

3.5. Leakage current
The $R_{sg}/R_N$ ratio, so-called the quality factor, of junctions on each wafer is plotted against the $R_N A$ product in Fig. 3. The subgap resistance, $R_{sg}$, and the normal resistance, $R_N$, are defined as the dc resistance at 2 and 4mV, respectively. Each point in the figure represents the best junctions on the wafer, where the yield varies from one to another. So far, the leakage current of AlN$_x$ barrier junctions seem to increase at $R_N A < 100 \ \Omega \ \mu m^2$ in a similar way to that of AlO$_x$ barrier junctions. Moreover, there is no clear difference in quality when we compare the results of different nitridation conditions. This could be indicating that the quality of our junctions is limited by some factor other than the nitridation/oxidation condition itself, for example the surface condition of the bottom-Nb electrode, onto which the thin Al layer is deposited.

3.6. RF test
The RF performance of a Nb/Al–AlN$_x$/Nb SIS mixer was tested in double sideband (DSB) mode in the 133-170 GHz frequency range. The mixer block was a scaled model of that described in Ref. [20]. The noise performance of the receiver was measured by the standard Y-factor method. Figure 4 presents the dc $I$-$V$ curve and the IF power output from the receiver with the beam pointing at hot (300 K) and cold (80K) absorbers. The measured DSB receiver noise temperature at 135-165 GHz was typically 20-60K, with a minimum of 22K at 149GHz. It is noted here that the measured receiver noise temperature of the Nb/Al–AlN$_x$/Nb SIS mixer is comparable with those of Nb/Al–AlO$_x$/Nb SIS mixers obtained at the same frequency band. A similar RF experiment was also done at the 350GHz band, and again a similar noise performance to those of Nb/Al–AlO$_x$/Nb SIS mixers was obtained.
4. Conclusion
Nb/Al–AlNₓ/Nb SIS junctions were fabricated by various RF plasma nitridation conditions. The reproducibility of the $R_NA$ product from batch to batch was improved by diluting the $N_2$ plasma with He or Ar. A power-law relation between the $R_NA$ product and the nitridation time was found to hold along $10^{6}-10^{8}$ $\Omega \mu m^2$, which implies that the thickness of the barrier grows logarithmically. By applying a pulse-time-modulated RF input, the nitridation was found to slow down significantly. Finally, we showed that the Nb/Al–AlNₓ/Nb SIS junctions fabricated by this process can indeed be used for SIS mixer application, with performances comparable to those of Nb/Al–AlOₓ/Nb mixers at frequencies around 150 and 350GHz.

Acknowledgments
The authors would like to thank the members of the ALMA Band 4 group at NAOJ and the ASTE group at IoA for their technical assistance in measuring the RF performance of the mixers.

References
[1] Kawamura J, Miller D, Chen J, Zmuidzinas J, Bumble B, LeDuc H G and Stern J A 2000 Appl. Phys. Lett. 76 2119–2121
[2] Kleinsasser A, Rammo F and Bhushan M 1993 Appl. Phys. Lett. 62 1017–1019
[3] Kleinsasser A, Miller R, Mallison W and Arnold G 1994 Phys. Rev. Lett. 72 1738–1741
[4] Miller R and Mallison W 1993 Appl. Phys. Lett. 63 1423–1425
[5] Torgashin M Y, Koshelets V P, Dmitriev P N, Ermakov A B, Filippenko L V and Yagoubov P A 2007 IEEE Trans. on Appl. Supercond. 17 379–382
[6] Wang Z, Terai H, Kawakami A and Uzawa Y 1999 Appl. Phys. Lett. 75 701–703
[7] Tolpygo S, Cimpoiasu E, Liu X, Simonian N B, Polyakov Y A, Lukens J E and Likharev K K 2003 IEEE Trans. Appl. Supercond. 13 99–102
[8] Shiota T, Imamura T and Hasuo S 1992 Appl. Phys. Lett. 61 1228–1230
[9] Kleinsasser A, Mallison W and Miller R 1995 IEEE Trans. Appl. Supercond. 5 2318–2321
[10] Dolata R, Neuhaus M and Jutzi W 1995 Physica C 241 25–29
[11] Endo A, Noguchi T, Matsunaga T and Tamura T 2007 IEEE Trans. Appl. Supercond. 17 367–370
[12] Iosad N N, Ermakov A B, Meijer F E, Jackson B D and Klapwijk T M 2002 Supercond. Sci. Tech. 15 945–951
[13] Cecil T W, Weikle R M, Kerr A R and Lichtenberger A W 2007 IEEE Trans. Appl. Supercond. 17 3535–3528
[14] Bumble B, LeDuc H G and Stern J A 2001 Proc. of the 9th Int. Symp. on Space THz Tech. 295–304
[15] Endo A, Noguchi T, Kroug M and Tamura T 2007 Proc. 18th Annual Symp. Space Terahertz Tech. in print
[16] Simmons J 1963 J. Appl. Phys. 34 1793–1803
[17] Cabrera N and Mott N F 1948 Rep. Prog. Phys. 12 163–184
[18] Deal B E and Grove A S 1965 Journal of Applied Physics 36 3770–3778
[19] Wagner C 1969 Corrosion Science 9 91–109
[20] Asayama S, Ogawa H, Noguchi T, Suzuki K, Andoh H and Mizuno A 2004 Int. J. Infrared and Millimeter Waves 25 107–117