Formation of High Quality AlN Tunnel Barriers via an Inductively Couple Plasma

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Abstract—Increasing operating frequencies of SIS receivers requires junctions that can operate at higher current densities. A major limiting factor of higher current density is the increase in subgap leakage that occurs in AlOx barriers as current densities approach and exceed 10k A/cm². AlN insulators are a promising alternative due to their lower leakage current at these high current densities. In this paper we present a more detailed analysis of the formation of AlN barriers using our previously reported inductively coupled plasma (ICP) source grown with techniqure. The ICP allows for in-depth control of ion energy and current density in the plasma. Additionally, plasmas with very low ion energy (~20eV) and a high degree of dissociation (~80%) can be achieved. The improved control allows for repeatable formation of high quality barriers. In particular, we report on the relationship between barrier thickness and plasma conditions as determined by in-situ discrete ellipsometry. Ellipsometry results were verified by fabricating Nb/Al-AlN/Nb junctions and measuring current-voltage, I(V), curves. DC I(V) curves for a range of current densities are presented.

I. INTRODUCTION

The terahertz region of the electromagnetic spectrum contains large amounts of information of interest to radio astronomers. In order for much of this information to be useful, it must be frequency down converted to lower frequencies for analysis. For frequencies below 1THz, SIS junctions offer the lowest noise temperatures; Nb/Al-AlOx/Nb junctions have been fabricated with noise temperatures of 2 to 5 times the quantum limit for frequencies below 700GHz. As the design frequency of an SIS junction increases, the desired current density increases. Unfortunately, as current densities increase beyond ~10kA/cm² the commonly used aluminium-oxide (AlOx) barrier shows an increase in leakage current. The increase in leakage current is attributed to an increase in pinhole defects in the barrier. For projects intended to operate at even higher frequencies and current densities, for example ALMA Bands 8, 9, and 10, an alternative barrier material is required to reduce subgap leakage and mixer noise temperatures.

Reference [1] first attempted to use an aluminium-nitride (AlN) barrier for SIS junctions after noticing an improvement in thermal stability after performing a nitridation of the Nb electrodes. The AlN barrier was fabricated using a process similar to that for AlOx. A thin layer of aluminium (Al) was exposed to a nitrogen plasma generated by a set of parallel plates. The plasma is necessary to crack the nitrogen molecule which will not spontaneously react with the Al layer. This work was reproduced by [2] who found that while their high jc junctions displayed excellent I(V) curves, their low jc junctions displayed poorer I(V) curves. The poor quality of these junctions was attributed to damage caused by energetic ions from the plasma striking the growing barrier. Reference [3] improved the fabrication process by moving the device wafer to the grounded electrode, a step that has been adapted by many groups [4]–[6]. Although this has yielded improved junction quality, the process still suffers from increased sub-gap leakage at higher current densities and issues of repeatability with wafers fabricated using the same processing conditions displaying current densities ranging over an order of magnitude or more.

Previously at the 2006 Applied Superconductivity Conference we reported on the first ever use of a novel technique for fabricating AlN-based tunnel junctions via a COPRA inductively coupled plasma (ICP) source [7]. An ICP enables independent control of the ion energy and current density of a plasma; this is not possible in a plasma formed via a parallel plate. Initial results of AlN formation characterized by in situ ellipsometry indicated improved control over the AlN formation dynamics when using the ICP. Reference [8] has since also adapted our ICP approach using the same COPRA ICP. By operating in a high pressure regime they have achieved reproducible, high quality AlN barriers. In this paper we present further studies of the AlN formation dynamics via ellipsometry along with dc I(V) curves of AlN-based SIS junctions fabricated using an ICP source.

II. EXPERIMENTAL METHODS

The AlN barriers described in this paper were formed using an Al over-layer process: a thin film of Al is exposed to a nitrogen plasma generated by a set of parallel plates. The plasma is necessary to crack the nitrogen molecule which will not spontaneously react with the Al layer. This work was reproduced by [2] who found that while their high jc junctions displayed excellent I(V) curves, their low jc junctions displayed poorer I(V) curves. The poor quality of these junctions was attributed to damage caused by energetic ions from the plasma striking the growing barrier. Reference [3] improved the fabrication process by moving the device wafer to the grounded electrode, a step that has been adapted by many groups [4]–[6]. Although this has yielded improved junction quality, the process still suffers from increased sub-gap leakage at higher current densities and issues of repeatability with wafers fabricated using the same processing conditions displaying current densities ranging over an order of magnitude or more.

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the loadlock of a sputtering deposition system solely used for depositing SIS trilayers. The wafer sits on a ground platter 10cm above the extraction grid of the ICP. The current density of the plasma is primarily determined by the rf power applied to the ICP. The ion energy can be varied by applying a bias to the bottom of the ICP plasma chamber through a series of capacitors. The capacitance is controlled by an energy knob with settings ranging from E(0) to E(10); E(0) is the lowest ion energy and E(10) the highest. At this moment we do not know the exact value of ion energy generated by the source, but it is believed to range from approximately 20eV to 200eV based upon measurements from the manufacturer.

A Digisel [10] discrete ellipsometer from HORIBA Jobin Yvon is attached to the loadlock of the deposition system such that measurements can be made while the wafer is sitting on the platter above the ICP during the nitridation process. The Digisel is attached at an angle of incidence of 70° and operates at a wavelength of 632.8nm. Data collection rates of up to 5 samples per second allow for real-time analysis of the AlN formation dynamics. During some sample runs the ellipsometer was set to collect data before and after the nitridation plasma was run. This was used to verify that any radiation generated by the plasma did not alter the ellipsometry data. No such alteration was found.

A. Ellipsometry Characterization

The formation dynamics of the AlN tunnel barriers were studied using in-situ ellipsometry. Ellipsometry is a non-destructive optical method to determine the thickness and optical properties (refractive index and extinction coefficient) of thin films. Ellipsometry measures the change in polarization of light reflected from the film. These values are compared to values calculated using first principles and a model of the film’s presumed thickness and optical properties. Adjusting the properties of the model until the calculated and measured polarization values agree yields the film’s properties. This method requires some prior knowledge of the films properties, such as either the film thickness or optical properties. When the optical properties are known, film thicknesses can be determined to within an angstrom when used in-situ.

Samples used for AlN formation analysis were made by first deposition ~1000Å of Al on a Si/SiO₂ wafer. At this thickness the Al layer is more than four times the thickness of the penetration depth of 632.8nm light in Al and the Al layer is opaque to the light. Thus the sample can be modelled as an Al layer upon which an AlN layer is grown (Here after the AlN formation process will be referred to as growth). Although the formation of the AlN layer does consume some of the Al layer, it is not enough to alter the model. For film thicknesses below ~100Å, the change in polarization is a product of the film thickness and refractive index. We used a value of 2.12 as found in ref [11]. This value was determined from much thicker films and to our knowledge no data is currently available on extremely thin AlN films. Ultimately, the thickness determined by ellipsometry is derived from a model and will need to be calibrated (discussed below).

B. Junction Fabrication and Testing

Nb/Al-AlN/Nb SIS junctions were fabricated using a self-aligned junction process. This process has recently been improved to yield better control over the final junction size and improved insulation around the top electrode [12]. The base electrode Nb thickness is 1650Å, the counter electrode Nb thickness is 650Å, and the Al over-layer thickness is nominally 50Å. The base electrode for these devices have been fabricated using both a lift-off and trilayer etch process. The lift-off process helps to limit stress based degradation of the junction [13] while the trilayer etch process enables the use of the ellipsometer on wafers that will be fabricated into full devices. We observed no difference in the quality of the devices fabricated by the two methods.

Measurements of the thickness of AlN barriers of fabricated devices is not possible. The thin Al over-layer is transparent and it’s thickness must be accounted for in the model. As the AlN layer forms, it consumes some Al and thus the thickness of both the AlN and Al layers are simultaneously changing. The Digisel system is only capable of monitoring a change in thickness of a single layer. However it is still possible to measure the change in polarization parameters, which do not depend on a material model, and compare these to AlN thicknesses measured on 1000Å Al films or calculated from current density measurements.

III. RESULTS

A. Barrier Thickness Measurements

The suitability of the COPRA ICP source for forming AlN tunnel barriers was first examined via in-situ ellipsometry. Plasmas with a range of ion energies and current densities were examined. Some initial earlier results are described elsewhere [7] and have been summarized in Figure 1 for the benefit of the reader. Barriers A, B, and C were grown with rf power of 150W and energy settings of E(2), E(5), and E(8); ~50eV, ~100eV, and ~150eV respectively. Barriers D and E were grown at an energy setting of E(2) with rf power of 300W and 450W respectively. The nitrogen pressure for all of the barriers was 2mTorr. The portions of the Figure to the left and right of the vertical lines (0 - 60 sec and 360 - 420 sec) represents the time before the plasma is turned on and after the plasma is turned off. It is important to note that the pre-plasma curves all closely overlap and the post growth curves are relatively flat. This indicates that radiation from the plasma is not interfering with the ellipsometry measurements and that the barriers are stable after the plasma is shut off.
As expected we see an increase in barrier thickness with both ion energy and current density after 5 minutes; higher energy ions can implant deeper into the Al layer while a higher current density provides more ions to react with the Al layer. Barriers B, C, D, and E show an faster initial growth rate that slows over time. For Barriers D and E this initial growth is very rapid yielding a ~15Å thick layer in less than 20 seconds. This is followed by a much slower growth with both barriers roughly 22Å thick after 5 minutes. This suggests the possibility that the barrier thickness may be saturating over time, but longer growth periods will need to be examined. Barrier A, formed at the lowest ion energies and current densities examined, shows a mostly linear growth curve with a thickness of 15Å after 5 minutes. Unfortunately, all of these growth curves rapidly yield thicknesses greater than the ~10Å which is likely necessary to yield high current density junctions. Additionally, the rapid increase in thickness will make it difficult to repeatedly fabricate junctions with the same barrier thickness and current density. However, the thickness of these barriers as determined by ellipsometry must be calibrated and thus these conditions can not be completely ruled out.

To achieve slower growth rates and smaller ultimate thicknesses, the applied rf power was greatly reduced. At a pressure of 2mTorr and energy setting E(2) the plasma was stable down to rf powers of 25W. By raising the pressure to 5mTorr the plasma was stable down to 10W. Figure 2 below shows the thickness-time curves for AlN barriers grown at 10W, 25W, and 50W; Barriers F, G, and H respectively.

The curves in Figure 2 are highly linear when compared with those in Figure 1. After 5 minutes the barriers are all still less than 4Å thick. Two different growth periods can be observed with the first period lasting for 5 minutes. This growth period is slightly faster than the second period with growths rates of 0.25Å/min, 0.35Å/min, and 0.62Å/min - for barriers F, G, and H respectively – compared with 0.13Å/min, 0.21Å/min, and 0.33Å/min for the following 15 minutes. Although these conditions would lead to longer growing times, the ability to control the magnitude of the rate of growth while maintain a constant growth rate should improve reproducibility with variations in the nitridation time of a few seconds having a much smaller affect on the overall thickness. Additionally, there appears to be no affect on the growth process by moving to higher pressures, although further study is needed. Importantly, with all other conditions remaining constant, higher pressures should yield lower ion energies due to thermalization in the plasma and may reduce barrier damage caused by energetic ions.

While the growth curves in Figure 2 for lower rf power show promise, there is still concern over damage to the growing barrier from high energy ions. The barriers were grown at E(2), which corresponds to an ion energy of ~50eV. This was the lowest energy setting examined previously because no conditions were found at which the plasma would start for lower energy settings. However, after developing a process for starting the plasma at higher energy settings and then raising the pressure and lowering the energy setting, stable plasmas at an energy setting of E(0) were achieved. This required a pressure of 5mTorr and rf powers of 100W or higher. These conditions should yield the lowest possible energy from the source, although because there is a small correlation between ion energy, rf power, and pressure, these may not be the absolute lowest values. In this process the plasma is started with the wafer in a separate portion of the loadlock chamber away from the plasma. After the plasma has stabilized at the desired conditions the wafer is moved into place. Thickness-time curves for barriers grown at 5mTorr, energy setting E(0), and rf powers of 100W, 200W, and 300W – Barriers I, J, and K respectively – are shown in Figure 3.
For Barriers J and K there is again an initial period with a high growth rate followed by a slower growth rate. It is likely that the two periods this still occur for Barrier I but the second period does not occur during the first 5 minutes. Although at a lower ion energy than in Figure 2, the higher rf powers yield higher growth rates such as those in Figure 1. Barriers J and K display initial rates of 6Å/min and 10Å/min followed by 0.6Å/min and 1.1Å/min. This yields barriers thicknesses of 8Å and 13Å respectively after 5 minutes. (The 0.2Å/min growth rate for Barrier I would require a nitridation times in excess of 50 minutes to yield a 10Å barrier.) The conditions for Barriers J and K are promising due to the combination of high initial growth rate to yield a thickness on the order of the desired thickness follow by a much slower growth rate to allow finer control over the final barrier thickness.

One unresolved question regarding the growth of AlN barriers is whether the barrier thickness saturates over time [3], [4]. If the AlN barrier were to saturate as a function of ion energy, the desired thickness could be controlled by selecting the necessary ion energy and thereby removing the issues of ion current density and nitridation time. Because much of the work in this area has been done using parallel plate plasmas it is difficult to isolate the contribution of ion energy to barrier thickness. Additionally, since many of the reports in the literature have been based upon current density measurements, if the barrier saturates at a thickness outside of the range useful for tunnel junctions it may not be fully observed. We examined this question with a series of long (90 min) plasma exposures. The resulting thickness-time curves are shown in Figure 4. Barriers J and K were grown at E(0) and rf power of 200W and 300W, respectively. Barriers L, M, and N were formed at E(2) and rf power of 100W, 200W, and 300W, respectively. A pressure of 2mTorr was used for all the barriers.

After 90 minutes all of the barriers continue in increase in thickness and also display thicknesses above those desirable for high current density junctions. Barriers N and M appear close to saturating and it is possible that after nitridation times of several hours they may reach a final thickness. However Barriers J, K, and L all have significant growth rates after 90 minutes. Should it be possible to reach saturation at thickness desirable for tunnel junctions, the long time that are required increase the likelihood of contamination from background gases. It is likely that the reports of saturation in the literature were for devices fabricated in the two growth periods. When comparing devices fabricated in these two periods it would likely appear that those from the second period had achieved a saturation level when compare with the rapidly changing barrier thickness and current density of those fabricated in the initial growth period.

B. dc Current-Voltage Curves

Although ellipsometry measurements can speak to the thickness of the AlN barrier, dc current-voltage, I(V), curves are necessary to speak to the quality of the barrier. Nb/AlN/Nb junctions were fabricated as described above in Section II. Junctions were fabricated with barriers grown from a range of plasmas conditions. The plasma conditions were chosen based upon the results of the ellipsometry data from figures above, beset represented by Barrier J. All the barriers were formed with a nitrogen pressure of 5mTorr and rf power of 200W. The nitridation times ranged from 15 to 25 minutes and energy settings of E(0) to E(2). As expected the current density decreased for increases in both nitridation time and energy setting. The highest quality junctions were formed with a nitridation time of 25 minutes and energy settings of E(0) and E(1). The resulting current densities ranged from 0.14kA/cm² to 32.5kA/cm². Typical I(V) curves for these junctions are shown in Figure 5.
Uniformity of current density over a 2" diameter wafer is ±15% while neighbouring junctions are nearly identical. Run to run reproducibility has shown some irregularities. Wafers A and B were fabricated with the same plasma conditions, but two months apart. Their current densities are 32.5kA/cm² and 4.2kA/cm² respectively. However, a wafer fabricated the day after wafer B with the same conditions had a current density of 4.1kA/cm². We believe that these irregularities likely arise from two sources: 1) small changes in the energy setting from run-to-run and 2) changes in the plasma conditions over time.

As discussed above the ion energy is set by turning a knob attached to a bank of capacitors. This knob does not have a physical “stop” and thus is likely to be in a slightly different location each run. Barriers made in consecutive run during which the energy knob was not moved showed thickness variation of less than 1Å. It is not possible to do this for real devices; the energy setting is increased at the beginning of a trilayer deposition for argon plasma substrate cleaning. Because nitrogen is a reactive gas it can attack the extraction grid of the ICP and cause the grid to breakdown over time. This may lead to changes in the ion energy and current density of the plasma over time despite using the settings. We have recently installed a Faraday cup plasma monitor on our deposition system and are examining both possible causes. We believe the plasma monitor will allow use the set the exact plasma conditions prior to each nitridation.

Decreasing the nitridation time from 25 minutes with an energy setting of E(0) results in very high current densities (above 100kA/cm²) and I(V) curves that display junction heating and large subgap leakage. For energy settings above E(1) the barriers shows signs of damage from the higher energy ions. Examples of both of these problems are shown above in Figure 6. Although Wafer C, fabricated with E(1) has a high quality I(V) curve, it is desirable to operate at the lowest possible energy setting to avoid the possibility of damage.

Although some questions remain about the reproducibility of this process, it is important to note the high quality of this process. Table 1 list the quality factor ($R_{op}/R_{N}$) for these devices over a range of current densities (in kA/cm²) compared with those from other groups made using both ion gun and parallel plate sources.
C. Ellipsometry Calibration

As discussed above, ellipsometry does not directly measure the thickness of a material layer but rather determines it based upon a model of the layer’s optical properties. We examined the validity of our AlN model by comparing the calculated thickness with current densities from fabricated devices. The thickness of an AlN layer formed on a thick Al layer, 1000Å, was first measured. Next an SIS junction was fabricated using the same plasma conditions. The current density of the fabricated device was then measured and compared to a theoretical current density calculated using the thickness determined by ellipsometry. The measured current density was determined from the quasi-particle rise of the dc I(V) curve. The theoretical current density was calculated using the equation derived by Simmons [16]

\[ j_C = 3.16 \times 10^{10} \frac{\sqrt{\Phi_B}}{d} \exp(-1.025 \sqrt{\Phi_B} d) \]

where \( j_C \) is the current density in A/cm², \( d \) is the barrier thickness in Å and \( \Phi_B \) is the barrier height in eV.

Figure 7 plots the measured current density of our AlN junctions based upon their measured barrier thickness along with theoretical curves for two values of barrier height. Reference [17] reported two different values of AlN barrier height depending upon the current density of the junction. Junctions with current densities above 5kA/cm² had a barrier height of 0.88eV while those with current densities below 5kA/cm² had a barrier height of 2.35eV. Our measured data fits much better with the barrier height for the higher current density which may be expected given that all of our junctions but one had current densities roughly at our above 5kA/cm². Our data can be fit with an exponential trendline defined by the equation \( j_C = C \exp(-0.99d) \) where \( C \) is a constant. This matches well to the equation \( j_C = C \exp(-0.96d) \) reported in [17].

\[ \Phi_B = 0.88 \mathrm{eV} \]

\[ \Phi_B = 2.35 \mathrm{eV} \]

Fig. 7 Current density as a function of AlN barrier thickness for measured data (triangle), values calculated for \( \Phi_B = 0.88\mathrm{eV} \) (diamond) and \( \Phi_B = 2.35\mathrm{eV} \) (square). The line is a best-fit trendline for the measured data.

The fit between the measured and calculated data can be improved by adjusting the thickness against which the measured data is plotted. An increase in the thickness shifts the curve for the measured data to the right. In Figure 8 the measured current densities are plotted with a AlN thickness increased by a factor of 1.1. This yields the best fit between the theoretically calculated data (diamond points and dotted line) and corrected data (circle points and dashed line). For very thin layers, <100Å, the change in polarization parameters measured by ellipsometry is a product of layer thickness and index of refraction. Thus an increase in thickness is proportional to a decrease in the value of index of refraction used in the model from 2.12 to 1.91. This range of values has been seen in thicker films with the variation attributed to changes in film structure [18].

\[ \Phi_B = 0.88 \mathrm{eV} \]

\[ \Phi_B = 2.35 \mathrm{eV} \]

Fig. 8 Current density as a function of AlN barrier thickness for measured data (triangle, solid line), calculated data for \( \Phi_B = 0.88\mathrm{eV} \) (diamond, dotted line), and measured data with AlN thickness adjusted by a factor of 1.1(circle, dashed line). The lines are a best-fit trendline for the measured data.

CONCLUSIONS

We presented recent results on our technique for fabricating AlN tunnel barrier for SIS junctions via inductively coupled plasma. Using in-situ ellipsometry we have studied the growth dynamics of AlN layers formed via ICP at higher pressures with lower ion energies and current densities. The ICP technique is highly versatile and capable of altering the characteristics (rate and curvature) of the AlN.

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growth curve. In the range of interest these curves can be described by two distinct growth periods: a shorter initial period with high growth rate followed by a longer period with slower growth rate. At lower plasma energy settings and rf powers both of these regions can be roughly approximated as linear. The issue of AlN thickness saturation was examined, and while it appears possible that the thickness may eventually saturate, this occurs outside the range of desirable thicknesses for SIS junctions.

We have fabricated and tested Nb/Al-AlN/Nb SIS junctions. Using nitridation conditions determined through the use of in-situ ellipsometry very high quality junctions with current densities ranging from 0.14kA/cm² to 32.5kA/cm² have been achieved with high quality factors of 25 to 15.5. Importantly even at current densities above 10kA/cm², when AIN barriers become technologically important, the quality factor of these devices remains high. Comparing the measured current density of these devices to theoretically calculated values we have confirmed the AlN thickness determined by ellipsometry to within 10%. An in-situ plasma monitor is currently being installed in the deposition system to monitor changes in the plasma over time and answer questions regarding process reproducibility.

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[10] HORIBA Jobin Yvon Inc., 3880 Park Avenue, Edison, NJ. 08820-3012. USA