Epitaxial aluminium-nitride tunnel barriers grown by nitridation with a plasma source

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High critical current-density (10 to 420 kA/cm$^2$) superconductor-insulator-superconductor tunnel junctions with aluminium nitride barriers have been realized using a remote nitrogen plasma from an inductively coupled plasma source operated in a pressure range of $10^{-3}$ to $10^{-1}$ mbar. We find a much better reproducibility and control compared to previous work. From the current-voltage characteristics and cross-sectional TEM images it is inferred that, compared to the commonly used AlO$_x$ barriers, the poly-crystalline AlN barriers are much more uniform in transmissivity, leading to a better quality at high critical current-densities.

Quantum technology based on superconducting or magnetic metals uses nanometer thick tunnel barriers. Many routes to quantum computation are based on aluminium with aluminium oxide barriers. Niobium devices use a proximitized layer of aluminium with a similar oxide [1]. Magnetic tunnel junctions have recently moved from using amorphous aluminium oxide to epitaxial magnesium oxide with its unique spin-dependent properties [2, 3]. In quantum computation the amorphous tunnel barrier has surfaced as an important source of decoherence leading to the introduction of an epitaxial aluminium oxide barrier [4, 5]. On the other hand highly transmissive tunnel barriers are urgently needed for submillimeter mixers in order to achieve a high bandwidth [6], in electronic refrigeration to maximize the cooling power [7] and in high density magnetic memory devices [8].

It has been demonstrated that a major problem of amorphous AlO$_x$ barriers is that they are laterally inhomogeneous [9, 10]. We take this into account by using a distribution of transparencies $T_n$ by writing for the voltage-independent normal conductance:

$$G \propto \sum_n A_n T_n$$

(1)

with $A_n$ a fraction of the total area of the tunnel barrier with an assumed uniform transmissivity $T_n$. Hence, we do not assume a universal distribution of transparencies [11, 12] but one which is related to a distribution of areas with different transmissivities, resulting from the technological process. For superconducting tunnel junctions (SIS) this amounts to a situation analogous to superconducting quantum point contacts [13]:

$$I \propto \sum_n A_n j(V, T_n)$$

(2)

with $I$ the total current and $j(V, T_n)$ the voltage-dependent current-density per area $A_n$. $j$ contains contributions of different orders proportional to $T_n$, $T_n^2$, $T_n^3$, etc., reflecting multiple Andreev reflections ($j(V, T_n) = j_1(V, T_n) + j_2(V, T_n^2) + j_3(V, T_n^3) + \cdots$). For the commonly used low current-density tunnel barriers most $A_n$ have $T_n$ of the order of $10^{-4}$. Since for SIS junctions first order tunneling $j_1$ leads to a zero subgap current, the remaining subgap current is due to the higher order terms ($j_2$, $j_3$, $\cdots$), which only appear for $T_n \approx 1$. Non-uniformity, causing the emergence of areas with $T_n \approx 1$, thus leads to excessive subgap currents. Therefore the critical current density of amorphous aluminium oxide barriers is limited to 20 kA/cm$^2$ [14]. We will demonstrate that aluminium nitride barriers are superior to aluminium oxide barriers with respect to barrier uniformity.

In the work reported here a very good reproducibility is realized by using the afterglow region of a nitrogen plasma from an inductively coupled plasma source (CO-PRA) (see for example Weller [15]), from CCR technology. The plasma provides the energy to split the N$_2$ molecules into N radicals, needed for the growth of AlN. The source is mounted on a vacuum chamber. The plasma is created in the source and diffuses into the chamber. We have chosen to work in a range of high pressures ($2 \times 10^{-3}$ mbar to $1 \times 10^{-1}$ mbar), for two reasons. First, we expect at these higher pressures a larger fraction of atomic N. Secondly, the ion energies in this regime are as low as a few eV, which minimizes damage to the barrier-formation. This is different from recent work, where the plasma process not only provides the chemically active species but also creates damage by highly energetic ions [16, 17, 18, 19, 20] (although usable routes have been reported [18]). In addition many other plasma techniques suffer from instabilities, resulting in a poor process reproducibility.

The devices are fabricated on a 2 inch oxidized silicon or fused quartz substrate. All metal layers are deposited by magnetron sputtering in the process chamber of a Kurt Lesker system. First, a 100 nm Nb monitor layer is deposited, after which a ground plane pattern is optically defined. Subsequently, a bilayer of 100 nm Nb and about 7 nm Al is deposited. Without breaking the vacuum, the substrate is then transferred to a nitridation chamber, where the Al is exposed to the nitrogen plasma for several minutes, producing a layer of AlN. The substrate is then again in vacuo transferred to the
process chamber, where a top electrode of 200 nm Nb is deposited. The lateral dimensions of the multilayer of Nb/AlN/Nb are patterned by lift-off. Junctions are defined by e-beam lithography with a negative e-beam resist (SAL-601) layer and reactively ion etched (RIE) with a SF$_6$ plasma using the AlN as an etch-stop, followed by a mild anodization (5 V). The junction resist pattern is used as a self-aligned lift off mask for a dielectric layer of 250 nm SiO$_2$. A 500 nm Nb/50 nm Au top layer is deposited and Au is etched with a wet etch in a KI/I$_2$ solution using an optically defined mask. Finally, using an e-beam defined top wire mask pattern, the layer of Nb is etched with a SF$_6$/O$_2$ RIE, which finishes the fabrication process.

The fabrication process used provides a very good reproducibility. There is reproducibility within one fabrication run, illustrated by the similarity of junctions on a produced wafer. Scatter in the normal resistance $R_n$ of the junctions is caused by variation in the junction area $A$, due to uncertainties in junction definition, and variation in the barrier-specific $R_nA$ value. For 34 junctions, of which 14 are shown in Fig. 1, the standard deviation, $\sqrt{\sum_{i=1}^{m}(R_{n,i} - \langle R_n \rangle)^2/(m-1)}$, with $m$ the number of junctions, of $R_n$ has been determined to be 3.3 % relative to the average, $\langle R_n \rangle = 6.8 \Omega$. The peak-to-peak variation amounts to $\pm 6 \%$. By measuring four big junctions (two of 1 $\mu$m$^2$ and two of 2 $\mu$m$^2$), the value of $R_nA$ has been found to be 2.8 $\Omega\mu$m$^2$ (corresponding to a critical current density $J_c \approx 78$ kA/cm$^2$). Assuming perfect junction definition (which is most likely not the case), the standard deviation of $R_nA$ within one fabrication run is at most 3.3 %. Based on the average $R_n$, $A$ is 0.4 $\mu$m$^2$.

We also find a good reproducibility from run to run. We have made several batches, varying the nitridation time $t_N$ from 9 to 60 minutes. About half of the batches has been made with a low position of the chuck (30 cm distance to the plasma source) in the nitridation chamber, the other half with a higher position (15 cm distance to the plasma source). In Fig. 2 we plot the $R_nA$ product of the batches as a function of $t_N$ for the large chuck-source distance (squares) and for the small chuck-source distance (diamonds). The dashed lines indicate a dependence $R_nA \propto t_N^3$, with $k = 1.4$. Obviously, there is a systematic dependence on nitridation time, indicating a well-behaving process. By varying the nitridation time and/or the chuck position, we can realize any desired $R_nA$ value between 0.5 $\Omega\mu$m$^2$ and 10 $\Omega\mu$m$^2$.

The quality factor $Q$, defined as $R_{sg}/R_n$, where $R_{sg}$ is the subgap resistance, gives an indication of the subgap leakage through the tunnel barrier. In the inset of Fig. 2 $Q$ has been plotted for two different batches of AlN based junctions, together with data on AlO$_x$ from Miller et al. [14]. In contrast to these AlO$_x$ devices, it is evident that $Q$ is higher than 10 for all AlN devices. The lower subgap currents prove that our AlN barriers have a lower density of areas with $T_n \approx 1$, in other words a better uniformity.

As shown in Fig. 2 we reach $R_nA$ products as low as 0.4 $\Omega\mu$m$^2$, corresponding to a $J_c$ of 420 kA/cm$^2$. For such high current densities, heating effects decrease the superconducting gap voltage of the junction in the form of back-bending (Fig. 1 inset). Up to at least 130 kA/cm$^2$, this effect remains hidden, but is still present. This in-
This method shows significantly better reproducibility than other AlN growth techniques have shown in the past. Compared to the conventionally used aluminium oxide barriers, much better quality current-voltage characteristics are observed for high critical current densities, which is attributed to a spatially more uniform transmissivity of the epitaxial tunnel barrier.

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