Study of NbN Josephson junctions with a tantalum nitride barrier tuned to the metal-insulator transition

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Abstract. We present recent results in developing SNS self-shunted NbN-Ta_xN-NbN Josephson junctions operating above 9 K; good control over the barrier resistivity allows us to fabricate high $R_n I_c$ product junctions. We believe that our all-nitride technology is suitable for next generation RSFQ (Rapid Single Flux Quantum) circuits. Films are sputtered on 3 inch thermally oxidized silicon wafers. An MgO buffer is used to induce a cubic NbN texture, assuring low resistivity and high transition temperature $T_c$ up to 16 K. The barrier has been characterized by using several thin Ta_xN films sputtered at ambient temperature and 300 °C, showing a resistivity behavior spanning from metallic to the insulator range. Junctions are achieved by using respectively a SNOP (Selective Niobium Overlap Process) and a SNEP (Selectively Niobium Etching Process) method. In-situ DC-Magnetron sputtered NbN/Ta_xN/NbN trilayers show very narrowed transitions up to 15.5 K. The junctions fabricated with a 7 nm thin high-resistive Ta_xN barrier show current densities in the 10 kA/cm² range and a large $R_n I_c$ product up to 0.3 mV at 10 K. Shapiro steps and Fraunhofer diffraction pattern have been observed, revealing Josephson behavior up to 14K. We also developed a complete all-NbN 10 photo-masks technology, including a common ground plane and bias resistors, devoted to RSFQ applications.

1. Introduction
Interest in RSFQ (Rapid Single Flux Quantum) logic has grown steadily among researchers, and recently this technology has been recognized as a viable alternative to the standard semiconductor-based logic [1]. Its potential applications span a large area going from very high accuracy metrology, to military and telecommunications satellite payloads, as well as teraflop computing.

Nowadays it is possible to realize RSFQ circuits counting up to a million junctions [2] and to implement complex functions such as AD-Converters and DSPs (Digital Signal Processors) [6]. Most of these achievements have been made by using a technology based on Nb/AlO_x/Nb superconducting/tunnel junctions (STJ) [2, 3]. However, this technology suffers from several inner limitations: mainly a very low working temperature (4.2 K) and the need for an external shunt resistor in order to get a non-hysteretic behavior [4].

The cryogenics problem can be eased by using superconducting materials with a higher critical temperature $T_c$. From this point of view niobium nitride films are actually one of the best candidates, showing a superconducting transition at about 16 K [5], which allows junctions operations up to 10 K. Moreover, due to its enhanced $T_c$ with respect to niobium, NbN has a greater energy gap ($2\Delta \approx 6$ mV at 4.2 K) which makes higher operating frequencies feasible.

The need for an external resistor to shunt the hysteretic STJ junctions can be fulfilled by using a metal barrier instead of an insulating tunnel barrier. In this case the quasi-particles carrying the signal across the weak-link in the AC regime will play the role of the shunting resistor [7]. Self-shunted junctions...
permit a simpler layout, hence a greater junction density; moreover, the absence of the resistor allows to achieve greater characteristic products $R_nI_c$, where $R_n$ is the junction normal state resistance, and $I_c$ the junction critical current.

The main drawback of SNS structures is the fact that conventional metals used for the barrier layer show a low-resistivity metallic behavior which weakens the Josephson coupling and limits the $R_nI_c$ product. The origin of this phenomenon should be sought in the proximity effect mechanisms [7, 8]. Alternative materials, such as highly disordered and highly resistive metals, as well as semiconductor barriers, should overcome this problem. Sputtered tantalum nitride has been recently targeted as an interesting barrier, because of its high resistivity, induced by high nitrogen flow during deposition [9, 10].

We believe that controlling the combination of these two materials, NbN for the electrodes and Ta$_3$N for the barrier, would open up a new top class of Josephson junctions which could successfully substitute the traditional Nb/AlO$_x$/Nb technology. In this paper we present our recent progresses in NbN and Ta$_3$N sputtering deposition, materials characterization, as well as junctions fabrication and testing. We will give insight into the parameters dispersion and the performances in terms of current density and $R_nI_c$ product. We will also highlight a clear relation between the nitrogen sputtering rate and the junction characteristic voltage. We will finally describe our new multilayer fabrication process, which is suitable for RSFQ applications.

2. Tantalum nitride characterization

NbN and Ta$_3$N films are fabricated by DC-magnetron reactive sputtering, which allows a direct control of all the deposition parameters. In order to characterize the barrier properties, we have deposited several Ta$_3$N films, varying the film thickness and the nitrogen partial pressure during deposition. Electrical transport measurements have been performed by using the four probes method in a Quantum Design PPMS (Physical Properties Measurement System).

The measurements in figure 1 show a clear relation between the nitrogen pressure and the film resistivity. For a nitrogen pressure $p_{N2}$ lower than 10% of the total chamber pressure $p_{tot}$, the Ta$_3$N films have a metallic-like behavior, with a superconducting transition below 4 K. The material resistivity increases at higher nitrogen partial pressures, and the metal-insulator transition takes place for $p_{N2} > 15\% p_{tot}$.

X-ray spectroscopy shows a prevalent fcc-phase orientation for 200 nm films deposited at $p_{N2} = 27\% p_{tot}$. In this case it is possible to extrapolate a lattice parameter perpendicular to the surface $a_{||} = 4.38$ Å. This value confirms a good adaptation of Ta$_3$N growing on top of NbN. Atomic Force Microscopy measurements over a 10x10 $\mu$m$^2$ 7 nm-thick sample show highly smooth and homogeneous surfaces, without holes and any particular structural strain, with a RMS-roughness of 0.35 nm.

3. Junction Fabrication

The NbN/Ta$_3$N/NbN trilayer is in-situ deposited by DC-magnetron sputtering on 3 inch thermally oxidized silicon wafers. We have shown in previous papers that a thin in-situ MgO buffer improves the superconducting properties of the NbN, inducing lower resistivity and a higher transition temperature [5]. This buffer is therefore introduced systematically before each trilayer deposition. Table 1 summarizes the typical parameters used for the trilayer fabrication.

As for the other films, the trilayers have been tested with the four probe method in order to study the evolution of resistivity as a function of temperature. The trilayers show an excellent superconducting behavior, with a critical temperature of about 15.5 K and a transition width narrower than 0.1 K. The trilayer resistivity measured at 300 K shows a dispersion less than 1% in run-to-run production, which reveals a good trilayer reproducibility.

The junctions tested in this study are mainly realized by using either a SNOP (Selectively Niobium Overlap Process) or a SNEP (Selectively Niobium Etching Process) fabrication technique [11, 12]. The material used to assure the isolation between the electrodes is a thin layer ($\approx 40 \text{ nm}$) of MgO. In the SNEP process an extra SiO$_2$ insulator layer is used to avoid short-circuits between the connection wires.
Figure 1. (a) Resistivity of Ta$_x$N films as a function of the thickness at several nitrogen partial pressures. (b) Evolution of the resistivity as a function of the temperature for several Ta$_x$N films.

Table 1. Typical trilayer sputtering parameters used during this study. (b.el.) base electrode; (barr.) barrier; (c.el.) counter-electrode; (P.) sputtering power; (T.) temperature; (t.) film thickness.

| Layer | Material | $p_{tot}$ | $p_{N_2}$ | $P$ | $T$ | t. |
|-------|----------|-----------|-----------|-----|-----|----|
|       |          | ($\times 10^{-2}$) | (%) | kW | $^\circ$C | nm |
| buffer | MgO      | 1.35      | 7.5       | 0.55 | 250  | 12 |
| b.el.  | NbN      | 1.88      | 10.5      | 1.5  | 290  | 350|
| barr.  | Ta$_x$N  | 1.15      | 40        | 0.5  | 330  | $\sim$10|
| c.el.  | NbN      | 1.88      | 10.5      | 1.5  | 290  | 210|

The wiring layer is a NbN 800 nm thick film which has been DC-sputtered at ambient temperature. The contact pads are realized either in gold or aluminum. Back-sputtering is used to improve the electrical contact before both wiring and pads deposition.

Etching techniques used during fabrication are Reactive Ion Etching in mixed $SF_6/O_2$ atmosphere, and lift-off in regular acetone bath. The photolithography facilities offer an accuracy of 1.5$\mu$m. Junctions have either a round or a square geometry, with dimensions varying from few $\mu$m$^2$ up to 100$\mu$m$^2$.

4. Junction test and discussion
We tested several SNOP junctions, fabricated with a 7 nm thick Ta$_x$N barrier which is sputtered under a 40% nitrogen partial pressure. The average critical current density is 14.5 kA/cm$^2$, and the average $R_nI_c$ product is 3.74 mV. Both values are measured at 4.2 K and show a dispersion of 15%. Figure 2 shows the current-voltage characteristic of a junction measured at 4.2K; the sample has a surface of 10.5$\mu$m$^2$ and is clearly non-hysteretic.

Junctions whose area is smaller than 4$\mu$m$^2$ have a current density about four times bigger. We believe that this is a consequence of our lithography limitations. Junctions close to the wafer edge show a
performance reduction, with a $J_c$ of 8 kA/cm$^2$ and a $R_nI_c$ product of about 1.12 mV.

The normalized junction resistance $R_nA_{SNOP}$, extrapolated from measurements at 4.2 K, is about 28 $\Omega$ $\mu$m$^2$ at the wafer center and 13.8 $\Omega$ $\mu$m$^2$ far from the center. We believe that this difference is mainly related to the high sensibility of the Ta$_x$N resistivity to the sputtering parameters close to the M:I transition [14]. Indeed, the plasma and the deposition rate can be non-homogeneous across the wafer during sputtering. From Ta$_x$N thickness measurements a barrier resistivity of 350 m$\Omega$ cm at 4.2 K at the wafer center can be inferred.

Figure 3. (a) Critical current density versus temperature for a 25 $\mu$m$^2$ SNOP junction. The curve is fitted with equation 1, following the long SNS junctions model in the dirty limit. (b) $R_nI_c$ product as a function of the temperature for different nitrogen pressures during deposition.

We studied the $J_c - T$ evolution for the junctions far from the wafer center. The curve, shown in figure 3 (a), has been interpolated with the formula which is used to describe the SNS long junctions in the dirty limit [7]:

$$J_c(T) \propto \Delta(T)^2 \sqrt{T} e^{-\left(d/\xi_n(T_c)\right)} \sqrt{T/T_c}$$

(1)

$\Delta(T)$ is the superconductor gap, which is supposed to be constant along the electrodes, because the high barrier resistivity allows to assume the rigid boundaries condition. We assumed a junction critical temperature $T_c$ of 15 K. From the fit we can extrapolate a $\xi_n(T_c)/d$ ratio of 3.6, where $\xi_n(T_c)$ is the induced Ta$_x$N coherence length at $T_c$ = 15 K, and $d$ is the barrier thickness. By using an average barrier thickness of 7 nm (measured by X-Ray reflectometry over Ta$_x$N thin films), we can infer a $\xi_n(4.2$ K) of about 3.8 nm. This value confirms the assumed long junction model and is coherent with other recently obtained experimental results [13].

Figure 3 (b) shows the $R_nI_c$ product as a function of the nitrogen deposition rate for different SNOP junctions of 25 $\mu$m$^2$ area. We notice that $R_nI_c$ increases of one order of magnitude for a nitrogen rise from 30% to 40% of the total gas pressure. This result puts in evidence the possibility of calibrating the junctions $R_nI_c$ product by acting exclusively on the nitrogen partial pressure.

We have observed a substantial difference in behavior between the SNEP and the SNOP junctions. Although the $R_nI_c$ products are of the same order of magnitude (between 1 mV and 3 mV), the $J_c$ values are much higher in the SNEP junctions, up to one order of magnitude; at the same time the barrier resistivity is higher in the SNOP case, thus compensating the $J_c$ difference. Since the trilayer parameters used in the two processes are virtually identical, we suppose that the technological process used to pattern the junctions has a strong influence on the junction yield.

In order to confirm the Josephson nature of the tested junctions, we have performed RF and magnetic field measurements at several temperatures. Shapiro steps are clearly visible for RF frequencies spanning from 7 GHz to 12 GHz; figure 4 (a) shows the I-V characteristic of a 4.9 $\mu$m$^2$ SNEP junction under a 9.4 GHz irradiation. Critical current diffraction as a function of the magnetic field has also been
observed; figure 4 (b) shows a Fraunhofer diffraction pattern obtained in a 4.9 $\mu$m$^2$ SNEP junction at 14 K. The magnetic field measurement permits to extrapolate a NbN penetration depth between 200 and 300 nm at 14 K. This value is lower than what we expected: about 400 – 500 nm as we could infer from previous measurements [15]. Therefore, a deeper investigation is needed concerning this result.

![Figure 4](image)

**Figure 4.** (a) Shapiro steps in a 4.9 $\mu$m$^2$ junction measured at 11.2 K. The irradiation source has a frequency of 9.4 GHz; 8.5 dB represents the RF power attenuation. (b) Fraunhofer diffraction of the critical current as a function of the magnetic field at 14 K for the same junction as in figure 4 (a).

5. **Next generation fabrication process**

We have recently developed a new multilayer fabrication process suitable for RSFQ applications. 10 photomask levels are needed in order to implement a common ground plane and on-chip bias resistors. Figure 5 shows a cross-section of the final process, whereas table 2 describes the different photomask levels. This will allow us to test in the next future basic RSFQ functions such as JTLs (Josephson transmission line), TFFs (Toggle flip-flop) and shift registers.

**Table 2.** Layer parameters concerning our new 10 photomasks multilayer RSFQ process. (b)=buffer layer.

| Mask   | Material                        | Thick. [nm] | Properties                        |
|--------|---------------------------------|-------------|-----------------------------------|
| M1:GND | NbN/MgO(b)                      | 400/10      | ground plane                      |
| M2:ISO1| MgO(b)/Si$_3$N$_4$              | 10/300      | $\epsilon_r = 7.5$ for Si$_3$N$_4$ |
| M3:RES | Ta$_x$N                         | 100         | $R_0 = 20$ $\Omega$ at 9 K       |
| M4:ISO2| Si$_3$N$_4$                     | 150         | $\epsilon_r = 7.5$ for Si$_3$N$_4$|
| M5:TRI | NbN/Ta$_x$N/NbN/MgO(b)          | 200/10/350/10| JJ trilayer $\lambda_{NbN} \approx 350$ nm |
| M6:JONCT |                                |             | JJ definition                     |
| M7:IA  | MgO/AlN/MgO                     | 10/20/10    | JJ self-aligned passivation       |
| M8:ISO3| MgO(b)/Si$_3$N$_4$              | 10/200      | $\epsilon_r = 7.5$ for Si$_3$N$_4$|
| M9:WIR | NbN                             | 800         | wiring                            |
| M10:CNT| Al/Nb(b)                        | 450/10      | pads                              |

6. **Conclusions**

We presented our significant progresses in fabricating NbN/Ta$_x$N/NbN Josephson junctions. Ta$_x$N films analysis showed a resistivity spanning from the metallic to the insulator region as a function of
Figure 5. Our new 10 photomask multilayer NbN/TaN/NbN RSFQ process. The table 2 summarizes the function of each photomask. A thin MgO buffer layer is used before every NbN deposition, in order to improve the superconducting quality of the nitride.

the nitrogen partial pressure during sputtering. Thin TaN films present high homogeneity and low roughness, as well as lattice parameters close to the NbN ones. NbN/TaN/NbN trilayers exhibit a high value \( T_c \) (16 K) with a narrow transition (\( \approx 0.1 \) K). Junctions with a thin high-resistivity TaN barrier are realized in both SNOP and SNEP technology. We obtained high \( R_n I_c \) products up to 3.74 mV at 4.2 K, and \( J_c \) of 14.5 kA/cm\(^2\), both suitable for RSFQ applications. The junctions fabricated by varying the nitrogen flow rate during barrier deposition show a clear dependence of the \( R_n I_c \) product as a function of the \( N_2 \) partial pressure. The \( J_c - T \) dependence has been fitted with the SNS long junction model in the dirty limit, which gives a \( \xi_n \) of about 3.8 nm at \( T = 4.2 \) K. SNEP junctions show higher current densities and lower normal resistances than the corresponding SNOP junctions, although the \( R_n I_c \) product is of the same order of magnitude: \( \approx 3 \) mV at 4.2 K. We explained this fact as a consequence of the difference between the two fabrication methods. Junctions exhibit a good RF and magnetic field response up to 14 K, confirming correct Josephson operation at a relaxed cryogenics environment. Finally, we presented our new multilayer fabrication process, including a ground plane and bias resistors, which is suitable for RSFQ applications.

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