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Adjusting the Parameters of the Tunnel Barrier of the SIS Junction by Varying the Composition of the Top Electrode

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The influence of the top niobium nitride electrode in a Nb|Al-AlN|NbN multilayer structure on the parameters of the tunnel barrier (AlN) was studied. It has been shown that an increase in the nitrogen concentration in the Ar/N_2 gas mixture during the formation of NbN by magnetron sputtering leads to a decrease in the tunneling transparency of the AlN barrier.

Keywords: superconducting structures, tunneling junctions superconductor–isolator–superconductor, tunneling junction AlN, plasma chemical etching.

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1. Introduction

Parameters of a tunnel barrier in the superconductor–insulator–superconductor junction (SIS) are critical for the fabrication of terahertz receivers. For most of receiving devices the niobium based tunnel SIS-junctions are used [1-4]. The decisive factor of a barrier transparency is the way it was formed: through oxidation or nitridation. Irrespective of the way, among the barrier formation parameters are time (time of oxidation or nitridation) and working gas pressure (oxygen or nitrogen), whereas for nitridation also magnetron power and magnetron-to-substrate distance are also important [5]. This paper states that another important factor is also the SIS-junction's upper electrode material and composition, i.e. a layer of metal directly deposited on the barrier surface (thin near-barrier layer). Samples with various content of nitrogen in the upper electrode of Nb−Al−AlN−Nb(N) SIS-junction were fabricated. It is demonstrated that the higher is the nitrogen content the lower is the transparency of the tunnel barrier with one and the same junction AlN−Nb(N) formation method. The decrease of transparency is also confirmed by a tendency of growth of the tunnel barrier's major parameters (height of potential barrier and its width) calculated in accordance with J. Simmons theory [6]. Such dependence is connected with nitrogen diffusion between the layers of Al, AlN and Nb(N).

2. Fabrication of samples

The technology of fabrication of SIS structures based on niobium and its compounds using selective etching and niobium anodization (niobium nitride) is well developed and widely reproducible [3–5]. The important feature of this fabrication technology is the fact that a thin aluminum smooths up the columnar structure of niobium surface [3,4] be wetting it. This allows to get a smooth aluminum surface on the basis of which the tunnel barrier will be formed. This barrier may be oxide (AlO_x) or nitride (AlN). The type of barrier is selected depending on the specified tasks: AlN barrier allows reaching higher current densities (up to $100\,kA/cm^2$) [5,7,8] compared to AlO_x barriers (up to $15\,kA/cm^2$), which, it its turn, allows significant broadening of the receiver's input band.

The SIS-structure is formed in several stages: deposition of a three-layer SIS-structure, etching of upper electrode in this structure to form a SIS-junction area, deposition of insulation, formation of a contactor and contact pads.

The samples were fabricated on silicone substrates, with orientation $\langle 100 \rangle$, where by magnetron sputtering a layer of Al_2O_3, 100 nm thick was deposited — this layer is required to prevent the substrate etching in further process operations. By method of optical photolithography a masking of resist material was provided defining the base electrode geometry. SIS-structure Nb|Al-AlN|NbN with thicknesses 200 nm|7 nm-(1−1.5 nm)|100 nm was deposited in a single vacuum cycle in magnetron sputtering device. After that, by explosion in dimethylformamide the photoresist with a three-layer structure on it was removed. After that, on the surface of a three-layer structure by method of photolithography an area of SIS-junction was formed. For this purpose, on the photoresist mask the plasma-enhanced chemical etching of upper layer (Nb(N)) in the fluorine-containing gases (CF_4) was carried out; after that the anodization followed by insulation (SiO_2, 250 nm) deposition was performed, at the end of this stage the photoresist with a layer of SiO_2 applied on it was removed in dimethylformamide. The contactor was fabricated from Nb layer 350 nm thick by method of photolithography. Contact pads were made of a 100 nm thick gold layer. More details on the fabrication technology of Nb|Al-AlN|NbN Josephson junctions are given in article [9].

All fabricated and studied samples underwent the same cycles of structure formation. Nb and Al layers in Nb|Al-AlN|NbN structure were formed similarly for all samples. AlN tunnel barrier was formed using nitridation of Al surface in the plasm of clean nitrogen, which was initiated on the aluminum magnetron. Such method of nitridation allows avoiding unnecessary bombardment of Al layer surface that could be inevitable if plasma discharge is initiated using HF-generator supplying power directly to the holder with sample.

Parameters of barrier formation that remained unchanged: gas — nitrogen, pressure $7.9 \cdot 10^{-3}$ mbar, power supplied to HF magnetron 50 W, distance between the sample and magnetron 130 nm. The varied parameter was nitridation time: 90 s or 180 s. It shall be emphasized that aluminum nitride formation on the aluminum surface is not associated with the material deposition due-to target sputtering, but results only from the nitrogen ions interaction with the aluminum surface on the sample. Upper SIS-junction electrode was niobium nitride or niobium. The following parameters during formation of this layer were constant: power supplied to magnetron at DC current 650 W, distance between the sample and the target 50 mm, argon flow 70 sccm. The varied parameter was nitrogen flow: 6, 7, 8, 10 sccm. In case of absence of nitrogen flow the pure niobium was formed. Pressure of gas mixture Ar and N_2 in chamber at the moment of NbN deposition — $9.3{-}9.5 \cdot 10^{-3}$ mbar. Upper niobium in the SIS-structure was formed in the same way as the lower one.

3. Determination of major parameters of tunnel barrier

As noted above, during formation of the SIS-junction barrier layer a thin Al film is exposed to oxidation/nitridation with formation of an insulator layer. Aluminum film shall be 5 nm thick and more to fully close all irregularities in Nb layer [4]. The characteristic thicknesses of barriers AlO_x or AlN formed on aluminum film were about 1 nm. The method described below allows assessing the tunnel barrier parameters.

Parameters characterizing the transparency of fabricated tunnel barrier are its thickness d and average height φ. There's a universal method for determination of d and φ parameters from the current-voltage curve (IVC) of a SIS-junction measured to high voltages ~ 1V. The method was based on measurement and approximation of tunnel current density versus voltage ratio that was suggested by J. Simmons [6] and generalized by W. Brinkman [10] for the tunnel junctions within voltages limit corresponding to energies not exceeding the barrier height φ. The calculation formulae and method is described in details in paper [11]. For junctions studied in this paper parameters d and φ allowing to give a quantitative assessment of the obtained barrier transparency were calculated.

Table 1. NbN films parameters, depending on the nitrogen flow, at continuous argon flow 70 sccm: temperature of transition to the superconducting state T_c, ratio of films resistance R at temperatures 20 and 300 K, resistivity ρ at 20 K, diffusion in terms of temperature of transition to the superconducting state

№ of sample	Flow N_2, sccm	T_c, K	R_{20}/R_{300}	ρ_{20}, $\mu\Omega \cdot$ cm	ΔT_c, K
1	6	14.1	1.08	160	0.35
2	7	14.7	1.215	230	0.2
3	8	14.2	1.245	260	0.14
4	10	13.3	1.22	292	0.58

4. Measurements

Before the fabrication of each sample the sputtering of control NbN films was carried out in the same conditions that were used for SIS-junction fabrication allowing to define the major parameters of the films (Table 1, Figure 1).

For this purpose a mask was fabricated with the help of which narrow metal strips (NbN) 1 cm long and 200 μm wide with contact pads were formed by photolithography. Temperature measurements of niobium nitride films were carried out in Dewar vessel with liquid helium on a 4-contact scheme. A submersible probe with a sample was slowly put into the Dewar vessel, film resistance and thermometer resistance were measured with a period of 0.5 s.

From Table 1 we can see that resistivity growing proportional to nitrogen content in the mixture is itself a parameter greatly depending on the amount of nitrogen in the gas mixture. Another essential parameter is diffusion in terms of temperature in the area of film transition to the superconducting state (ΔT_c). From Figure 1 and Table 1 we can see that the film with higher temperature of transition to the superconducting state $T_c = 14.7$ K is featuring a more steep slope of the curve $R(T)$ in the area of transition to the superconducting state ($\Delta T_c = 0.2$ K), than the film $T_c = 14.2$ K ($\Delta T_c = 0.14$ K). This can be a decisive factor when selecting the required parameters of the film for specific applications, since the diffused transition into superconducting state indicates the heterogeneity of the film composition during its growth.

The fabricated samples with SIS-junctions were measured when setting up the voltage at a temperature of 4.2 K using a submersible probe placed in Dewar vessel filled with liquid helium. Each sample was a common electrode with 14 SIS-junctions on it; a contactor with a contact pad was connected to each junction. The sample had SIS-junctions of various diameters from 1.8 to 5.5 μm. Given that the tunnel barrier parameters (height and width) are the same for each SIS-junction within one substrate, the value R_nS

can be calculated (SIS-junction area multiplied by its normal resistance after the superconducting gap) and shall be the same for each junction. The value R_n is determined from the current-voltage curve.

4 samples were studied where bottom layer (Nb) and barrier (Al-AlN) in the SIS-junction were formed similarly, but the niobium nitride layer was formed under various sputtering conditions: the nitrogen content in nitrogen/argon mixture varied. The barrier was formed in the plasma of pure nitrogen discharge at 50 W and 180 s. The current-voltage curve for 4 samples with various R_nS, measured as described earlier are presented in Figure 2. In-spite that the barrier in each sample was formed in equal conditions different values R_nS were obtained which proves the assumption of the upper SIS-junction layer influence on its parameters.

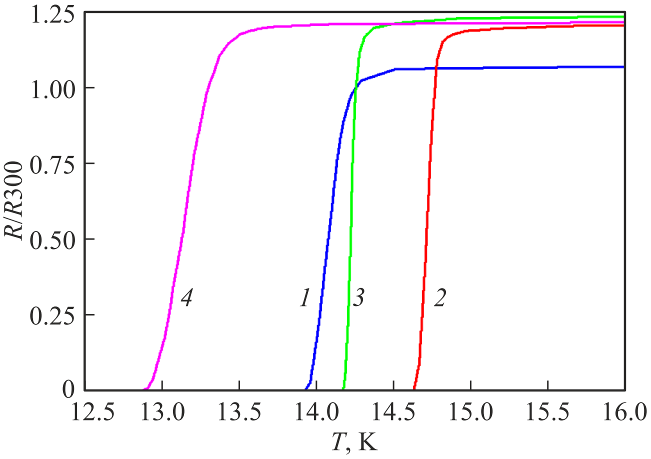

Figure 1. Curves of NbN films transition to the superconducting state, normalized resistance (R/R_{300}) versus temperature; numbers of curves correspond to the samples numbers in Table 1.

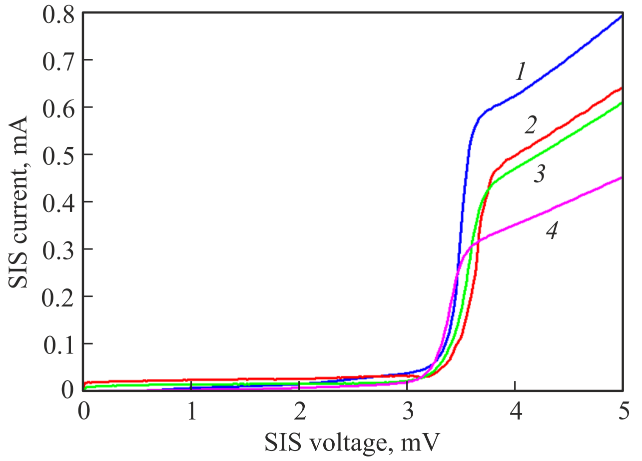

Figure 2. The current-voltage curve of SIS-junctions with different current densities for the barriers equally formed but having different nitrogen content in formation of NbN layer: curve *1* — flow $N_2 = 6$ sccm, $R_nS = 7.7\,\Omega \cdot \mu\text{m}^2$; *2* — $N_2 = 7$ sccm, $R_nS = 13.8\,\Omega \cdot \mu\text{m}^2$; *3* — $N_2 = 8$ sccm, $R_nS = 15\,\Omega \cdot \mu\text{m}^2$; *4* — $N_2 = 10$ sccm, $R_nS = 22.2\,\Omega \cdot \mu\text{m}^2$.

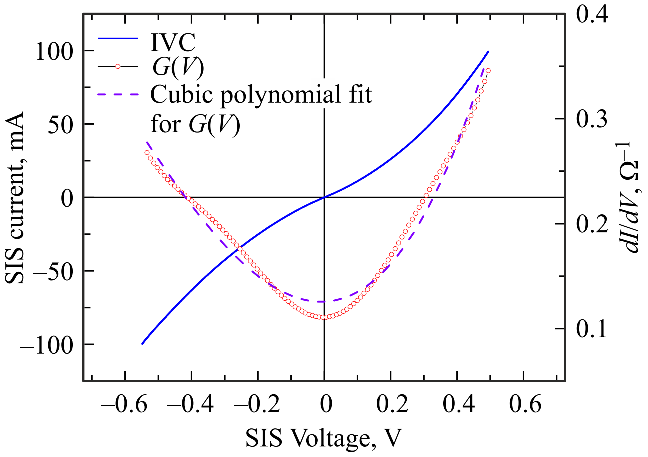

Figure 3. Current-voltage curve (IVC) of the SIS-junction for a sample with flow Ar = 70 sccm, $N_2 = 10$ sccm. Solid curve (left axis) — Current-voltage curve of the tunnel SIS-junction measured in the range of voltages ±0.5 V. The curve with circles (right axis) is the junction QD differential conductivity versus voltage. The dotted line (right axis) shows approximation with cubic polynomial for G.

To assess major parameters of the tunnel barriers of the fabricated samples a cycle of measurements was made. Figure 3 shows one of the current-voltage curves and its derivative which is itself a ratio of differential conductivity of the junction $G = dI/dV$ versus applied voltage. $G(V)$ was approximated by a cubic polynomial fit. Further, the barrier major parameters were calculated complying with method described in [11]. The barrier parameters calculation results are presented in Table 2.

As seen from Table 2, as the nitrogen concentration in gas mixture increases during nitridation both, the thickness d of insulation layer and the height of barrier φ are increasing, which proves the tunnel barrier transparency decrease with the growth of R_nS.

To study the influence of barrier formation parameters on SIS-junction, samples were fabricated with similar upper layer NbN, sputtering parameters Ar = 70 sccm, $N_2 = 7$ sccm, 650 W, but with different barrier nitridation time: 90 and 180 s. From the current-voltage curves shown

Table 2. Summary table of calculated barrier parameters for the tunnel junctions with various nitrogen concentration in gas mixture during nitridation, at the same flow of argon 70 sccm

Flow N_2, sccm	R_nS, $\Omega \cdot \mu\text{m}^2$	φ, eV	d, Å
6	8	0.29	11.9
7	14	0.3	12.6
8	15	0.34	12.5
10	22	0.37	13.8

in Figure 4, we see that the process of tunnel barrier formation significantly (by 67%) changes the parameter R_nS ($8.4\,\Omega\cdot\mu\text{m}^2$ and $14\,\Omega\cdot\mu\text{m}^2$).

For a barrier formed with parameters 50 W and 90 s a SIS-junction with Nb upper electrode instead of NbN was fabricated, the current-voltage curve of such junction is shown in Figure 5. AlN tunnel barrier was formed in similar way, however, the difference in value R_nS turned to be huge: $8.4\,\Omega\cdot\mu\text{m}^2$ for NbN and $3\,\Omega\cdot\mu\text{m}^2$ for Nb. Apparently, the niobium layer „extracts“ nitrogen from AIN layer because of diffusion processes, thus, greatly increasing the transparency of the tunnel barrier. This example illustrates great influence of upper layer in SIS-junction on the tunnel barrier properties.

Samples were manufactured with close values R_nS ($\sim 8\,\Omega\cdot\mu\text{m}^2$, that had different parameters of barrier formation (50 W, 180 s and 50 W, 90 s), different parameters of upper niobium nitride formation ($\text{Ar} = 70$ sccm, $N_2 = 6$ sccm and $\text{Ar} = 70$ sccm, $N_2 = 7$ sccm). IVC measurements for these samples are shown in Figure 6. The difference in normal resistance R_n above the superconducting gap is explained by difference in the tunnel junctions areas.

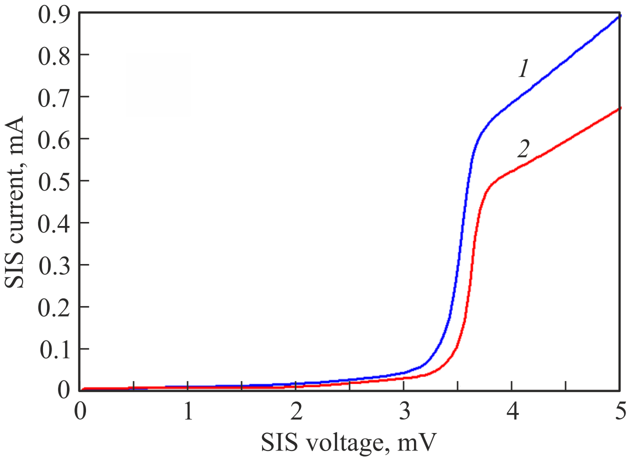

Figure 4. Current-voltage curve of SIS-junctions with similar NbN electrodes, but with different AlN barriers: on both curves the power supplied to HF-magnetron was 50 W, curve *1* — nitridation time 90 s, curve *2* — nitridation time 180 s.

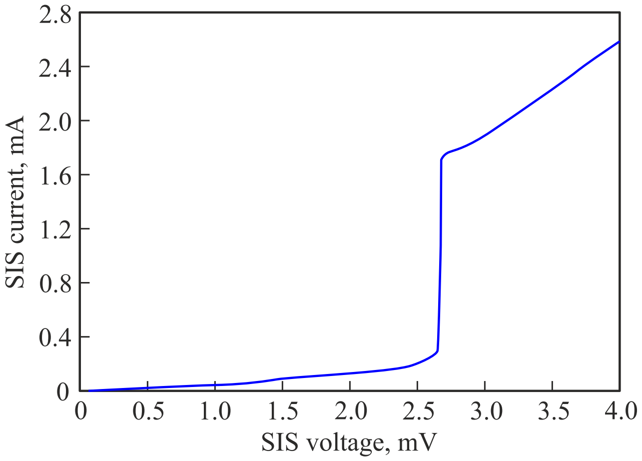

Figure 5. Current-voltage curve of SIS-junction Nb−Al−AlN−Nb with a barrier formed with parameters 50 W, 90 s.

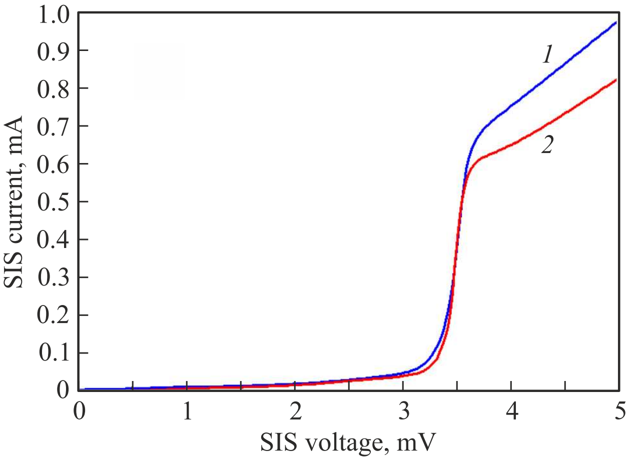

Figure 6. Current-voltage curve of SIS-junctions with the same R_nS, but with different parameters of the tunnel barrier (AlN) and upper electrode (NbN) in SIS-structure: curve *1* — barrier nitridation time 180 s, nitrogen flow for NbN 6 sccm, curve *2* — barrier nitridation time 90 s, nitrogen flow for NbN 7 sccm.

From Figure 6 we may see that despite the difference in conditions of SIS-junction formation, samples with similar R_nS ($\sim 8\,\Omega\cdot\mu\text{m}^2$) have almost similar parameters: ratio of subgap resistance to normal resistance (R_j/R_n) 23 and 24.4, blurring of the gap feature 0.418 and 0.355 mV, value of the superconducting gap V_g 3.49 and 3.47 mV. Thus, we may make a conclusion that to get the required R_nS it is needed not only to change the barrier formation parameters but also to change parameters of the upper electrode formation in the SIS-structure without compromising the quality of the junction itself. This conclusion may be especially handy when it is required to form low R_nS ($\sim 1\,\Omega\cdot\mu\text{m}^2$), while decrease of nitridation time either cannot provide to reach these values or becomes too short (< 10 s) affecting the reproducibility of such junctions.

5. Conclusion

In a SIS-junction the variation of nitrogen concentration in the upper layer of niobium nitride allows, along with known methods (change of pressure, duration of barrier formation and etc.), making a fine adjustment of the tunnel barrier transparency. It is demonstrated that increase of nitrogen concentration in the gas mixture of Ar and N_2 when sputtering the upper layer of SIS-structure leads to lower transparency of the tunnel barrier. Owing to variation of niobium nitride sputtering parameters along with selection of the tunnel barrier fabrication methods the SIS-junctions with similar parameters such as R_nS, V_g, R_j/R_n can be obtained without compromising the quality of the junction itself. Such approach to formation of SIS-structures provides brand new capabilities in control of tunnel junctions parameters and design of receivers on their base while maintaining the fabrication process reproducibility.

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Conflict of interest

The authors declare that they have no conflict of interest.

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