Electron spectroscopy analysis on NbN to grow and characterize NbN/AlN/NbN Josephson junction

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Abstract

Three layers, NbN based Josephson junction, has been growth by RF and by DC sputtering within the constrain required by the photolithography technology. An interesting superconducting film with critical temperature of $T_c = 14$ K, well above the temperature of the commercial cryocooler, has been obtained reducing sputtering power and finding a proper N\textsubscript{2} concentration in the gas mixture. The search of the new sputtering parameters has been obtained with the help of electron spectroscopy and X-ray diffraction analysis.

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1. Introduction

The properties of superconducting niobium nitride thin film are very important in their application especially for Josephson junction (JJ) devices \cite{1,2}, single photon detectors \cite{3} and devices working in the frequency range of THz. This type of material is very stable, and although the considerable development of the high-$T_c$ superconductors is still technological competitive, when used on industrial scale. JJ devices are also very attractive as radiation sensors, because of their ultra fast photo response, and high signal/noise ratio. The easiest way to make Josephson junction Superconductor–Insulator–Superconductor (SIS) is by successive sputtering deposition of NbN and AlN film.

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The NbN/AlN/NbN three layer film is deposited in a vacuum chamber, partially filled with an appropriate gas mixture of Ar and N\(_2\). The DC or RF voltage create the plasma condition to sputter the material from the target which is then deposited on a substrate. The Ar/N\(_2\) ratio of the gas mixture, the substrate temperatures, the total gas pressure and the target–substrate distance are all factors that affects the stoichiometry of the films. The fulfillment of this condition is quite critical, because the number of parameters involved makes the deposition system-dependent. Furthermore to design a commercial device it is necessary to avoid overheating of the substrate. Indeed the photolithography process requires the use of photoresist mask to design the device, and to prevent photoresist burning [4] the substrate should not overcome 120–130 °C.

The data reported in this communication have taken in consideration all these constrains, i.e. the film deposition has been performed using sputtering parameters that reduce the heating of the substrate incompatible with the presence of photoresist.

2. Experimental part

Our sputtering system is equipped with 4 in. DC and RF magnetron sources, ion gun and a grounded substrate table. The Si wafers, used as substrate, are fixed to a copper chunk with a little vacuum grease. The chunk is then attached to the rotation table by three screws. The massive support and the target–substrate distance (10 cm) maintain the substrate temperature under 50 °C during the film growth. NbN was grown in DC mode with different gas mixtures of Ar/N\(_2\) flux, (total pressure 5 × 10\(^{-1}\) Pa) while AlN was deposited in RF mode (100 W) with a gas mixture of 1:1 of N\(_2\) on Ar, the total pressure in the deposition chamber being 10\(^{-3}\) Pa. In these conditions the deposition rates for NbN and AlN were 0.6 nm/s and 0.06 nm/s respectively; the calibration of the deposition ratio have been measured ex-post by a thickness profilometer. The resistivity measurement, that assure the proper stoichiometry of the film, is performed with a four probe technique mounted in a closed-cycle helium apparatus. We observe, just after starting the cooling down of the cryostat, the trend of the resistivity vs. the temperature. If the resistivity slope is negative the film has a semiconductor nature, while the slope is positive the film has a metallic character. Indeed the slope of the resistivity can be considered as a precursor of the film quality (see the inset of Fig. 1) and if the slope of resistivity is positive there is no chance to get high-T\(_c\) NbN film. Finally, XPS measurement have been performed in a standard U.H.V.
chamber equipped with the dual-anode X-ray lamp and a double pass electron analyzer. Other measurements presented were not made in our laboratory.

3. Data analysis

The deposition of NbN and AlN face different problems. In particular the multi-phase nature of the NbN makes it more difficult to be deposited than AlN, and this is the reason why we dedicate a considerable part of our work to characterize the growth of NbN. Resistance measurements on a series of different NbN films have been performed to find the best deposition parameters.

In Fig. 1 we report the resistance value from resistivity measurement to get the NbN film with the highest $T_c$. The reported curve has been found, regardless of the compatibility with photolithography processes (300 W DC, 360 V, 0.83 A, $\text{Ar}/\text{N}_2 = 91/11$, $P_{\text{tot}} = 5 \times 10^{-1} \text{ Pa}$). Lower sputtering power and different $\text{Ar}/\text{N}_2$ ratio are required to make film compatible with photoresist and with high-$T_c$. Changing the thermodynamic sputtering parameters it is possible to obtain films with $T_c$ temperature, ranging from 9 to 15 K.

In Fig. 2 we report the X-ray diffraction of a NbN/AlN/NbN three layered film based on a good NbN i.e. with a $T_c$ of 15 K. The curve shows few peaks: (a) the Si(200) peak due to the silicon wafer substrate, (b) peaks of NbN(111) and NbN(200) cubic phase and (c) the hexagonal phase of the AlN(111), no other peaks are present indicating that the AlN and the second NbN film growth are based on the first NbN film. In Fig. 3, instead are reported the compared diffraction curves of samples grown varying the $\text{N}_2$ concentration. We note that film with the best $T_c$ is associated with the predominance of a single phase; i.e. well pronounced peaks attributed to the direction (111) and (200) cubic phase of the NbN.

Curves with peak of the hexagonal (100) and (101) phase have lower $T_c$. X-ray photoelectron spectroscopy (XPS) has been used for surface characterization of Nb and NbN thin film. In Fig. 4 the top curve shows the spectrum of the Nb film ($T_c = 9.2 \text{ K}$) where the presence of various NbOx is evident. It appears as a $3p_{1/2}$ and $3p_{3/2}$ doublet shifted toward low energy with respect to the Nb peak. In the second curve (NbN#1), the sample has been grown with 250 W DC, $\text{Ar}/\text{N}_2 = 91/3$, $P_{\text{tot}} = 5 \times 10^{-1} \text{ Pa}$ and the resulting $T_c$ is less than 8 K, the XPS data indicate that the Nb film contain N, but the thermodynamic conditions were not appropriate to form stable NbN. Indeed the NbOx peaks are still in the same position and in addition a new peak,
around 395 eV, appears. This it may indicate that some Nitrogen molecules are present inside the sample without reacting with the Nb atoms. The third curve (NbN#2) has been grown just by changing the N$_2$ concentration (Ar/N$_2$ = 91/8) and keeping equal all the other parameters, as a consequence the resulting $T_c$ is increased to 12 K. The XPS data show a little shift of the Nb$_3$p peak toward the NbN peak indicating the beginning of a weak reaction of Nitrogen with Nb. In this case NbNx film is short in nitrogen. The fourth curve (NbN#3) is referred to as a film with a much bigger concentration of N$_2$ (Ar/N$_2$ = 91/12), but the $T_c = 12$ K is still quite low. The NbN film has an excess of nitrogen; indeed the NbOx peaks are strongly reduced while the NbN peaks are dominant. Finally the film named NbN#4 has been grown with the right N$_2$ concentration (Ar/N$_2$ = 91/10) and it shows the highest $T_c$ (15 K). XPS data show a lower quantity of NbOx partially hidden below the NbN peak.

To be sure that the NbN/AlN/NbN three layer works as Josephson junction device, the stoichiometry of each layer must be correct and the interfaces must be as sharp as possible. During this work we learned how to grow high-$T_c$ NbN film in the presence of photoresist.
constrain. In particular we learned that, to avoid the overheating of the substrate, it was necessary to reduce the exposure time of the substrate to plasma temperature. For this reason thicker film requires a further reduction of the DC power. The best growth conditions to obtain NbN in the presence of photoresist material are reported below:

(a) 250W DC, Ar/N\textsubscript{2} = 91/10, P\textsubscript{tot} = 5 \times 10^{-1} Pa for a film with a thickness of 50 nm
(b) 200W DC, Ar/N\textsubscript{2} = 91/9, P\textsubscript{tot} = 5 \times 10^{-1} Pa for a film with a thickness of 100 nm.

In the case of AlN, the deposition is less critical than for NbN. Indeed since the stoichiometry of AlN does not allow different compounds and the toughness of Al is poor so that its sputtering deposition do not give much problems. The sputtering parameters used are: 100 W of RF power in a vacuum chamber pumped to a total pressure of 10^{-1} Pa, and a gas mixture Ar/N\textsubscript{2} equal to 1. The only serious problem arises from the thickness of film required for the insulator gap in the Josephson junction. Very thin film may not cover uniformly the substrate giving an uncontrolled roughness. To estimate the quality of the interface we made reflectivity measurements on a new NbN/AlN interface prepared following the same “recipe” used for the three layered device. We found that on a film 20 nm (18 + 2) thick the roughness is less than 0.5 nm. This tell us that the “recipe” used for the AlN forms quite sharp interface when deposited on NbN. Josephson junction device based on NbN have been really built by lithographic process (Fig. 5(a)), although we are not able to produce it in a large scale. In Fig. 5(b) we report a very enlarged photo of a real Josephson junction made by photolithographic process.

4. Conclusions

In conclusion, we note that the critical temperature of NbN is affected by growth conditions as shown by XPS measurements. Moreover, we have shown that, paying attention to the growing
sputtering parameters, it is possible to obtain superconductor thin films with a good critical temperature compatible with the constraints of photolithography. Finally we report on the tentative plan to grow an array of Josephson junctions by photolithography process with the perspectives to built electronic devices in the region of the “new frontier” of terahertz and other special applications like flexible electronics.

References