ISTITUTO NAZIONALE DI FISICA NUCLEARE

Laboratori Nazionali di Frascati

INFN-19-15/LNF
16 Luglio 2019

Nb superconductive thin film coating on flat Cu disks for high gradient applications

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Abstract

In this work we present the characterization of Nb superconductive films deposed on copper substrates with two different techniques: the PVD magnetron sputtering and the Pulsed Laser Ablation. In the first method Nb films $\sim 3\mu m$ thick were deposited with an average roughness of 160 nm. The superconductivity properties of these films were also determined with a 4-probe resistivity measurement. Data show a superconducting transition at $9.6K$ as expected from Nb films. With the second technique thick Nb films were deposited on copper substrates using the Pulsed Laser Ablation. In this case the Rutherford Backscattering was used to determine the thickness and the chemical state of these films that show different degrees of oxidation.

Published by
Laboratori Nazionali di Frascati

1 Introduction

Performance improvements of next generation of particle accelerators are mostly dependent on the new technological developments of accelerating cavities, i.e., mainly on the field gradients they are capable to withstand. Recent studies of the performance of radio-frequency (RF) copper structures operated at cryogenic temperatures have shown a dramatic increase in the maximum surface electric field that may be reached. These researches successfully at the frontier of achievable gradients using microwave accelerators, introduced a new technique based on the cryogenic cooling of RF cavities. Actually, the most performing resonant cavities technology is based on copper structures, cooled to tens of kelvin to reach maximum gradients ranging from 100 MV/m up to 1000 MV/m. Over this values the resonant cavity starts to reach the breakdown limit where discharges affect the surface of the cavity. Irreversible damage starts to occur together with an exponential cascade phenomenon, which makes the cavity unusable. In order to overcome these limits and possibly increase the structure performance, recent studies are focused on thin films coatings of RF cavities in order to control and improve the copper properties. This next generation of technologies will permit the design and the manufacture of compact and affordable accelerators in many research areas and for industrial applications. We have to underline also the significant improvements in the performance of the beams that can be delivered by RF photoinjectors that utilize these innovative technological approaches. Indeed the use of hard and/or cryogenic copper may reach 250 MV/m surface electric fields in S band and 500 MV/m in X band. This field levels in turn permit an increase over a factor of 25 in the beam brightness. A soft X-ray or a X-ray -FEL based on such a photoinjector would have $\sim$ 1/3 the undulator length, and 3 times the peak power. In addition, an intolerable level of dark current characterizes the present manufacturing technologies. Here we show results related to Nb coatings on copper to minimize the dark current level for cryogenic operation. Indeed, Nb-coated copper cavities, operating at superconducting temperatures (2-4 K), have been shown to provide higher stability against quench, insensitivity to trapped earth magnetic field and higher value of unloaded quality factor Q_0 for a lower cost. [1]

2 Plasma Vapour Deposition (PVD) using magnetron sputtering

The substrates we used for the sputtering process were OFHC copper discs (diameter 50 mm, thickness 5 mm). A preliminary mechanical turning of the discs was mandatory in order to remove superficial scratches and the central pin, remaining from the previous machining process. The average superficial roughness Ra, evaluated after the mechanical turning, was 85 $\pm$ 25 nm. An electro-polishing (EP) process was also applied to smooth the substrate surface. During EP, the sample was sustained with a screw fixed in its backside. The polishing involved several steps as follow:
1. ultrasonic degreasing with Rodastel-30 soap;

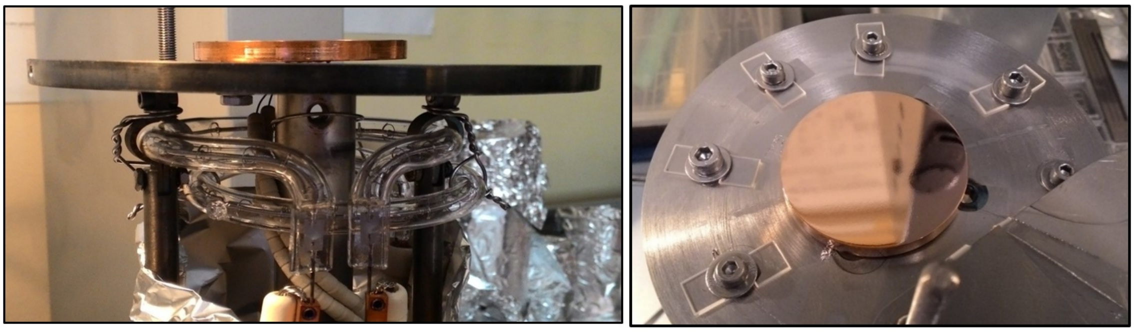

Figure 1: (a) Top plate of the support hosting the 4“ magnetron sputtering system; (b) top view of the sample holder. Ten quartz samples are placed near the copper disc to evaluate the deposition.

2. ultrasonic cleaning in deionized water;
3. electropolishing in H3PO4: butanol in the volume ratio of 3:2;
5. surface passivation with sulfamic acid;
6. ethanol rinsing and drying with pure nitrogen.

In details, the first two steps were performed at $60°C$ and lasted 50 minutes each. Electropolishing was performed at room temperature and needed a time ranging from one to three hours. After polishing, the surface of the disc was passivated in a diluted sulfamic acid solution (20 g/l). The whole process removed 100-150 μm of copper, producing a mirror like surface. The Nb superconductive coating was deposed by PVD magnetron sputtering technique. Previous experience on thin films of Nb deposed on Cu resonant cavities at CERN and at INFN suggested to deposit 3 μm of Nb while maintaining the temperature of the substrate around $500°$ C in order to achieve a high RRR value (the index of purity of the Nb deposited film) [2-7]. The deposition was performed in a UHV horizontal chamber (see Fig. 1 and 2).

The system that supports the substrate consists of a 4 mm thick stainless steel disc on top of a cylindrical copper shield, which protects two concentric IR lamps and enhances the heating efficiency.

A thermocouple in contact with the base of the copper substrate is used to monitor the temperature (Figure 1a). Several quartz samples visible in Fig. 1 were placed near the copper disc on the 4“ magnetron sputtering system in order to estimate the deposited film thickness, the deposition rate and the superconductive properties of the Nb films (RRR and Tc).

To reach ultra high vacuum (UHV) before the deposition process, 48 h of baking at $550°C$ by IR lamps were necessary. A few hours before starting the deposition, the IR lamps power was reduced to reach the temperature of $500°C$ at the thermocouple contact. The coating process was performed by running the 4” planar magnetron equipped with the Nb (RRR 300) water-cooled planar target. The base pressure was around $3 \cdot 10^{-8}$ mbar at $\sim 550°C$. During the deposition process, the Argon pressure was 5·10-3 mbar and the sputtering current was set at $2.1A$ $(27mA/cm^2)$. Five minutes of target conditioning was

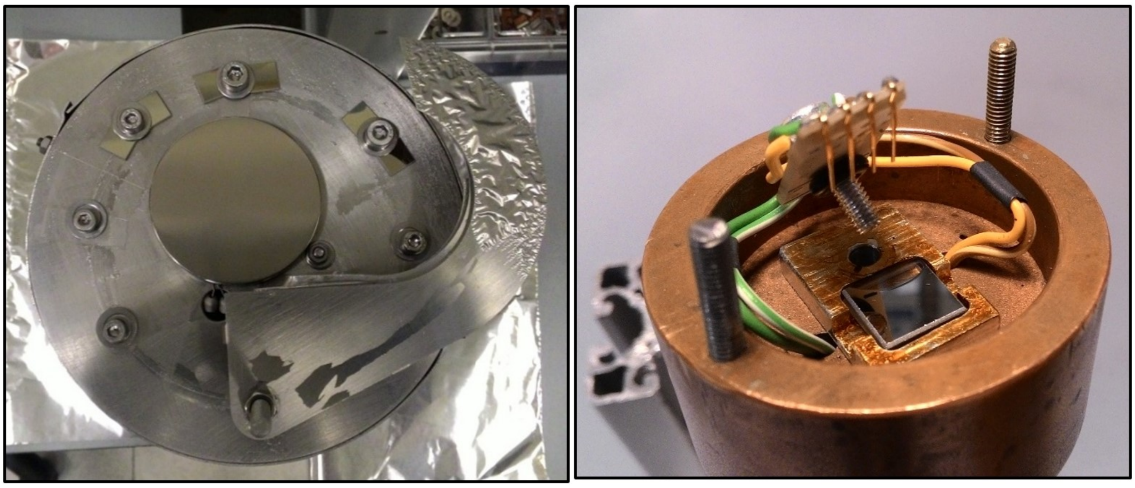

Figure 2: (a) Nb sputtered on the copper substrate; (b) four-point probe measuring system.

done prior to start the deposition. After opening the chamber, the coated samples were immediately inserted in a plastic wafer box closed under nitrogen atmosphere, ready for the next characterization and for shipping.

3 Characterization of Nb films deposed by PVD magnetron sputtering

The Nb samples were characterized in terms of roughness using the Veeco DEKTAK 8 profilometer calibrated using a spare copper disc sample, polished and coated following the same procedure of the real substrates. The real samples were not characterized in order to prevent dust contamination and scratches on the surface. The results obtained are reported in Table 1.

Treatment	Superficial Roughness R_a
Mechanical turning	$(85 \pm 25)nm$
Electro polishing	$(160 \pm 35)nm$
Magnetron sputtering coating	$(180 \pm 30)nm$

Table 1: Disc roughness after each treatment measured by the Veeco DEKTAK 8 profilometer.

A four-point probe testing platform was used to evaluate the Nb thin lm RRR parameter [8] and the critical temperature using the quartz samples placed near the copper disc inside the UHV chamber (see Fig.2). Also the thickness was measured on the masked quartz samples using the Veeco DEKTAK 8 profilometer. These Nb films show a superconductive behaviour and all have a thickness of $\sim 3\mu m$, $RRR = 26$ and $T_c = 9.36K$.

4 Nb deposition by Plasma Laser Ablation (PLA)

The experimental setup we used in the preparation of the samples using the Plasma Laser Ablation is depicted in Fig. 3. The ablating radiation was produced by a pulsed Nd-YAG laser EKSPLA PL 2143C, which can deliver up to 100 mJ at 1064 nm with a pulse length of 25 ps or 40 mJ at 532 nm and 20 ps by second harmonic generation. We have chosen this solution because, in this pulse duration regime, the multiphoton ionization mechanism dominates over the thermal vaporization at shorter wavelengths. This process leads to the formation of single ions or smaller charged clusters that could be easily accelerated by an electric field [9]. The laser radiation hits the target at an angle of $30°$ and it is focused by a BK7 lens with $f = 30cm$ in order to obtain an elliptic spot on the surface with a minor axis of $2mm$, corresponding to an average fluence of $1.1J/cm^2$ at the operating energy. The laser pulse frequency was fixed at $10Hz$ and the whole deposition process runs up to 25 min corresponding to 15000 laser shots. The target was rotated by a stepping motor to change the spot position periodically in order to avoid the formation of deep craters on the surface, which could change direction of the plasma plume.

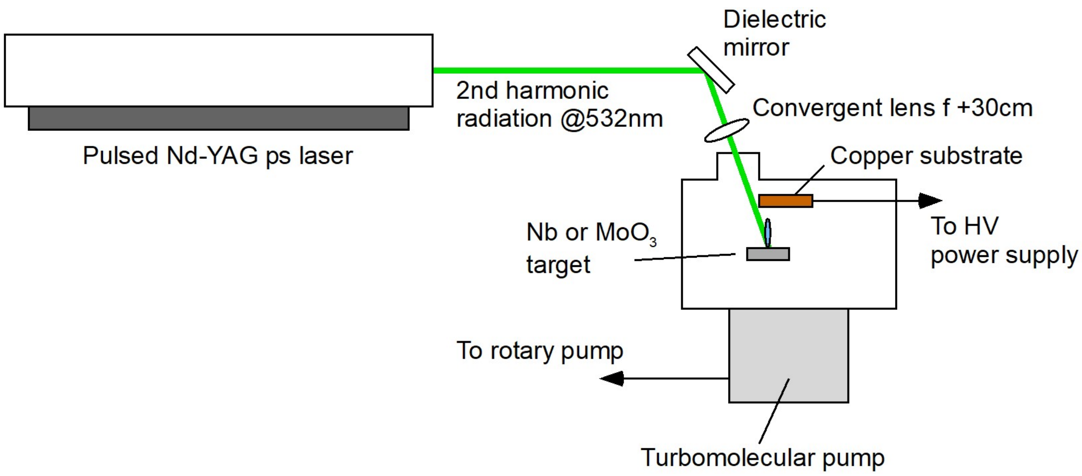

Figure 3: The experimental setup used for the growth of the PVD samples.

The copper substrate was placed in front of the target at a distance of $5cm$ and it was kept at an electric potential of $-5kV$ respect to it. The voltage has been chosen in order to avoid breakdown discharges between target and substrate through the plasma plume. No particular preparation procedures of the samples, other than a deep cleaning in isopropyl alcohol by the use of an ultrasonic washer, have been followed in this phase. The whole assembly was placed in a vacuum chamber where a turbo-molecular pump guarantees a backing pressure of $\sim 5 \cdot 10^{-6} mbar$. In the second phase of the experiment the deposition apparatus has been upgraded to allow the deposition on larger substrate area, i.e., discs of diameter up to 2” (see Fig. 4). In this second layout the discs are mounted on a 3-axis translator (travel length $= 70mm$ for each axis) with three stepping motors. The system is controlled by a software that allows grid-like paths in the horizontal xy plan at a given speed with an accuracy of few micron. In addition, we can also add a sort of ripple in the translation in order to improve the uniformity of the deposition. The target is also moved by a vacuum sealed rotating stage controlled by a DC motor. Particular attention has been paid to the cleaning of the substrates. Since we need to eliminate traces of gas trapped in the copper lattice, the substrate is clamped on a macor cylindrical block, warmed by two high power glass resistors up to $100°C$ through the application of a DC voltage, that increases the degassing rate. The ceramic holder also electrically insulates the substrate from the translator stages that are grounded together with the vacuum chamber walls. The vacuum pumping system is the same of the setup in Fig. 3. However, the better quality of the new vacuum chamber together with the use of new electrical and mechanical feedthroughs allowed reaching a vacuum level better than $10^{-6} mbar$. There is also the possibility of blowing gas in the vacuum chamber from the outside, to carry out a deposition in a controlled atmosphere or to cool the samples in an inert environment in order to prevent oxidation and to minimize contamination phenomena.

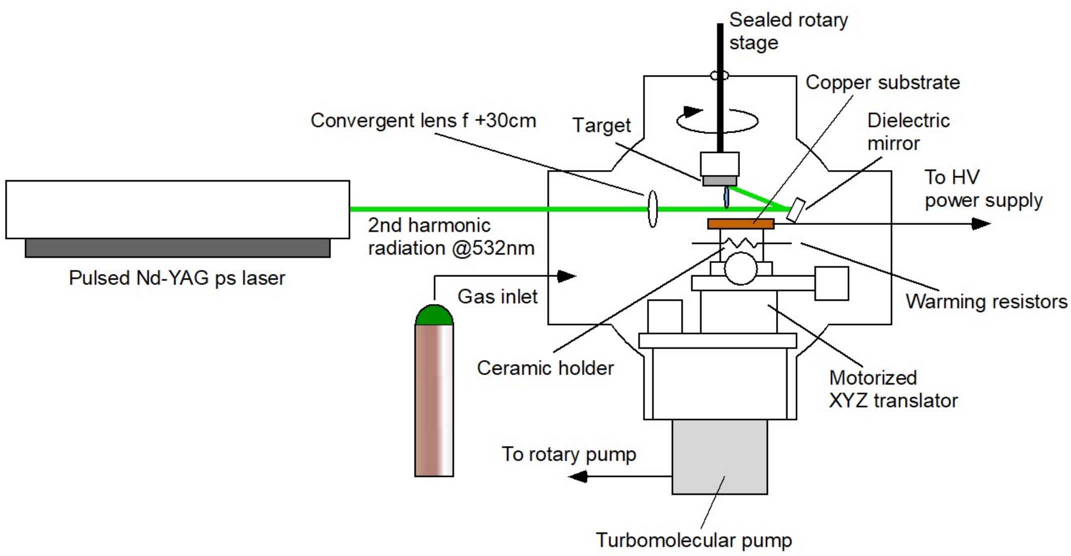

Figure 4: The experimental setup used for the coating of discs with larger substrate areas.

5 Characterization of Nb films deposed by PLA

We report here the characterization by Rutherford Backscattering Spectrometry (RBS) of two samples of niobium showed in Fig. 5 (Nb1 and Nb2) grown using Plasma Laser Ablation technique. The RBS spectra were analysed using the XRump software to evaluate the elemental composition and to estimate the surface density $[at/cm^2]$ of these samples obtained considering different layers containing Nb, O and Cu. The RBS measurements were performed with a $2.0MeV$ He+ beam working at normal incidence. The detection mode was $165°$ backscattering angle by using the $3.5MV$ HVEEE Singletron accelerator system. The beam spot was circular with a diameter of 1 mm.

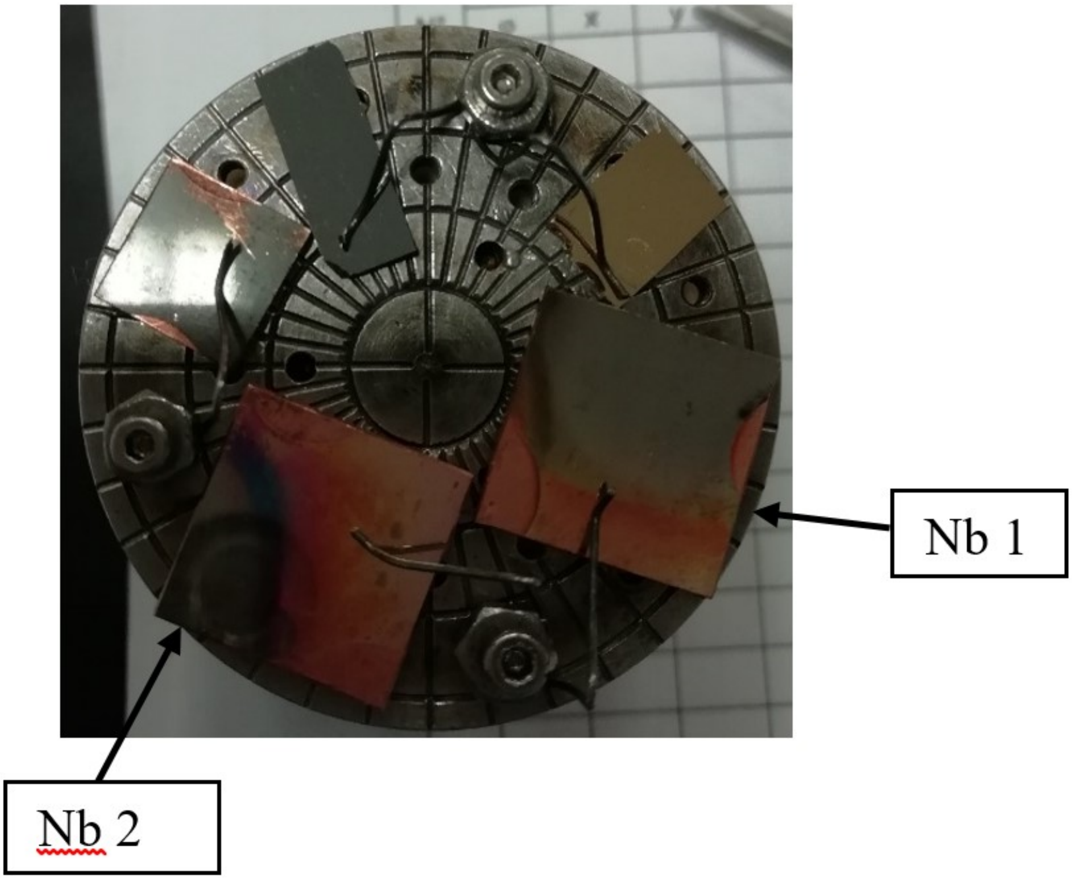

Figure 5: Photograph of the sample holder for RBS analysis with the two Nb films characterized.

The Nb1 sample was probed in two different regions: the central zone showed in Figure 6(a) and in the lateral region showed in Figure 6(b). The RBS spectra of the two areas are compared in Figure 6(c). Spectra show that between the two areas there is a difference of about 20% of surface density (at/cm2) and, therefore, a variation of 20% in the thickness occurs between the two regions. The lateral zone is thicker than the central area and the approximate thickness is $\sim 150/200nm$.

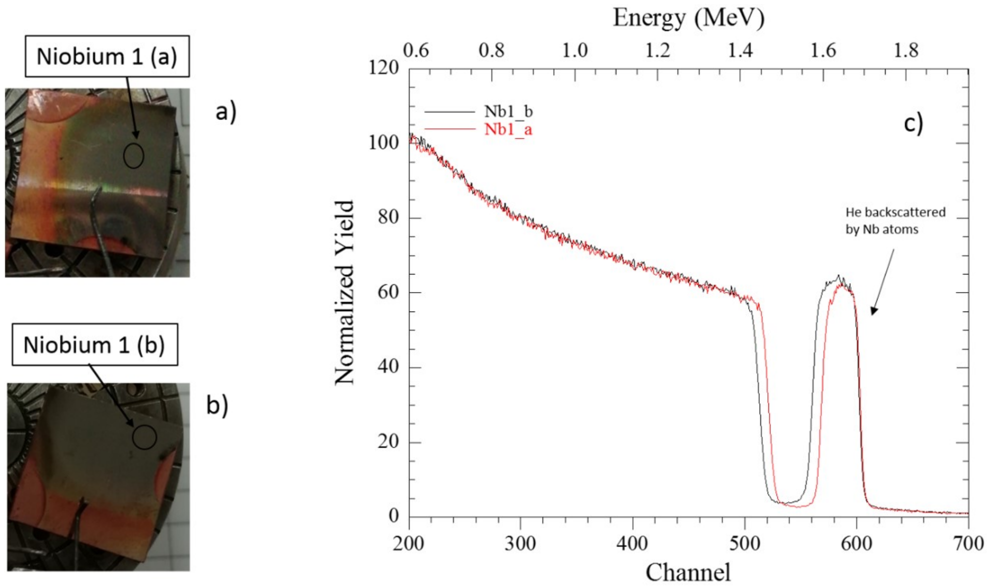

Figure 6: (left) Photographs of the areas hit by the beam on the Nb1 sample, center (a) and edge (b); (right) comparison of the RBS spectra in the two probed areas.

In particular, for the a region of the Nb1 sample, the simulation showed in Fig. 7 points out a layer on the Cu substrate with the composition $Nb_1O_{0.5}$ and a surface density $5.75 \cdot 10^17at/cm^2$. The structural view of this sample is displayed in Figure 7 (right - not in scale).

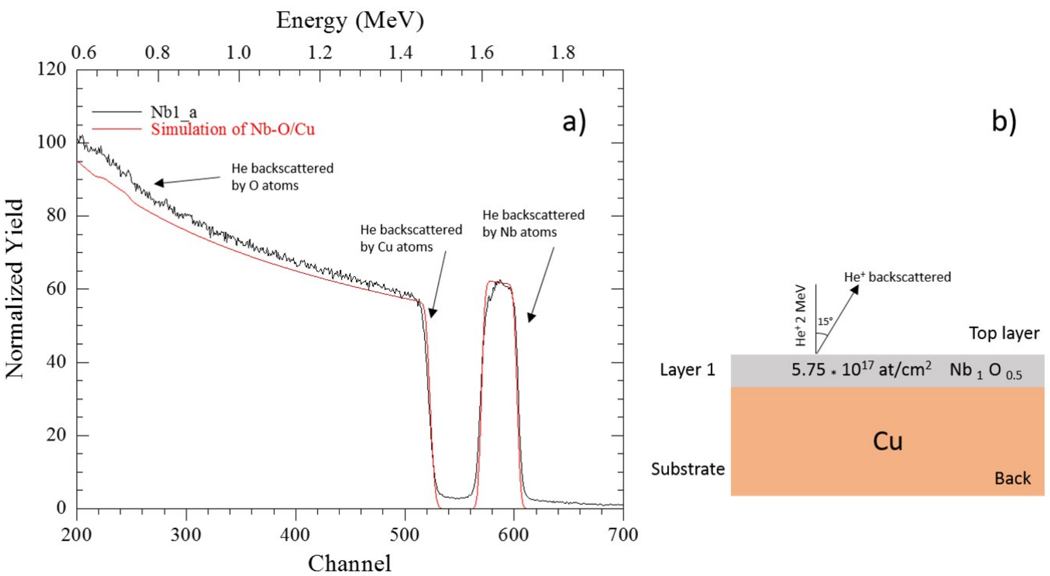

Figure 7: (left) Comparison of the RBS spectra of the sample Nb 1 in the region a (black) with the simulation (red) carried out by XRump; right) the estimated structure (not in scale) of this Nb1/Cu sample in the region (a).

In the lateral region (b) the Nb 1 sample has the same composition of the (a) region while the surface density is higher $7.3 \cdot 10^17at/cm^2$ (see Figure 8 right - not in scale).

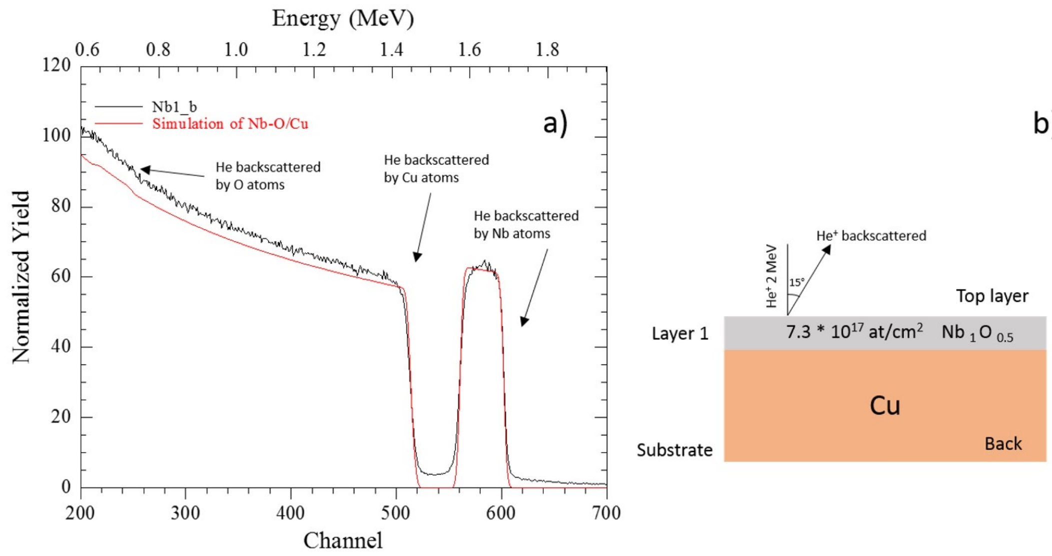

Figure 8: (left) Comparison of the RBS spectra of the sample Nb 1 in the region b (black) with the simulation (red) carried out by XRump; right) the estimated structure (not in scale) of this Nb1/Cu sample in the region (b).

The RBS spectrum of the sample Nb 2 (black) and the simulation (red) are showed in Figure 9. The XRump simulation also of this second sample has been obtained with different layers containing Nb, O and Cu. Based on the result of the simulation, the layout of this film is shown in Figure 9 (right - not in scale). In particular, in the top layer the composition is $Nb_1O_0.7$ and the surface density is $2\cdot10^17at/cm^2$. The deeper layer shows a strong oxidation and its composition is $Nb_1O_{3.8}$ with a surface density of $8\cdot10^17at/cm^2$. The approximate thickness due to the 1^{st} and 2^{nd} layer accounts to $\sim 400nm$.

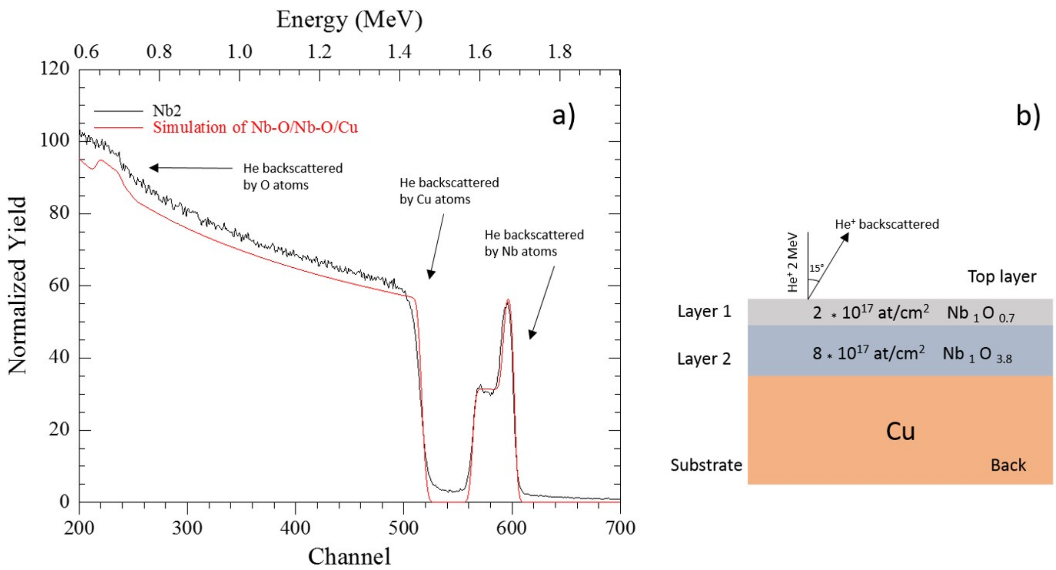

Figure 9: (left) Comparison of the RBS spectra of the sample Nb 2 (black) with the simulation (red) carried out by XRump; right) the estimated structure (not in scale) of the Nb2/Cu sample in the region (b).

5.1 Conclusions

The high power dissipation in copper cavities made the possibility of using superconducting cavities particularly attractive [10]. Indeed, the surface resistance of a superconducting material at radio frequencies tends to a low, constant, value. Most superconducting RF cavities are made of niobium, which has a critical temperature of $9.2K$.
The theoretical surface resistance of niobium at $3K$ is about $20n\Omega$. Although, the surface resistance of a superconductor increases with frequency much more rapidly than does that of copper, the very high values of Q achieved with superconducting cavities mean that the stored energy for a given accelerating gradient is much greater than in normal conducting cavities. We report here the techniques and the characterizations of Nb superconductive films deposited on polished OFHC copper substrates. We used both PVD magnetron sputtering and Pulsed Laser Ablation technique. In the first case after electropolishing we deposed on the copper surface Nb films $\sim 3\mu m$ thick with a roughness of $\sim 160nm$ and a superconductive transition of $9.6K$. With the second technique (Laser Ablation) we deposed Nb films on mechanical polished OFHC substrates with a thickness of 200 nm and 400 nm and chemical composition as measured with RBS. The characterization showed the presence of oxygens with a different degree of oxidation in these two films. From these results it is clear that in order to achieve pure, non-oxidized, Nb films is necessary to improve the vacuum pressure during the deposition process. Nb-coated copper cavities, operating at superconducting temperatures (2-4 K), have been shown [2] to provide higher stability against quench, insensitivity to trapped earth magnetic field and higher value of unloaded quality factor Q_0 for a lower cost, moreover Nb coatings on copper cavities may also minimize the dark current problem at cryogenic operation. Moreover, recent results obtained by coating with a 2 mm thick layer of fine grain niobium of high residual resistivity ratio for superconducting RF cavities strongly support the need to continue the investigation of the properties of thick metallic coatings [11].

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