Elaboration and characterization of Nb and NbTiN superconducting thin films for RF applications.

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Abstract

This paper presents the last results obtained at Saclay on Nb and NbTiN coatings prepared by magnetron sputtering for making 1.5 Ghz accelerating cavities. The optimisation of the sputtering parameters was made on small samples, and the surface resistance of the films deposited on Φ 12 cm copper disks was measured using a TE011 cavity. The results obtained (R_{res}=40 nΩ at T=1.4 K and F= 4 Ghz for the best NbTiN coating) encouraged us to deposit such films in 1.5 GHz copper cavities. One Nb/Cu and four NbTiN/Cu cavities have been prepared, the copper cavities being electrochemically polished before sputtering. The Nb/Cu cavity reached 12.5 MV/m with $Q_0=10^{10}$ at low field. The NbTiN cavities could not be tested because of the blistering of the coatings.

1. INTRODUCTION

For the future accelerators, the technology of the sputter coated cavities appears as an interesting alternative to the bulk Nb superconducting cavities. Nevertheless , even if Nb sputtered cavities are already used for the 350 MHz cavities of the LEP project, important progress is still needed for the 1.5 GHz Nb and NbTiN sputtered cavities so as to become really competitive. That is why an intensive study started in 1986 at the C.E.Saclay was meant to develop and to optimize the technological process for making 1.5 GHz sputter coated copper cavities.

A magnetron sputtering setup, designed to prevent dust contamination of the cavity before sputtering was realized. In the mean time a new magnetron cathode designed so as to allow the variation of the atomic Nb/Ti ratio was also developed. Details on the setup and the cathode were previously reported elsewere [1],[2]. Several Nb sputtered monocells and pentacells cavities were prepared using this setup, and were tested at CERN. The good results obtained during this collaboration with CERN showed that the sputtering setup worked satisfactorily [2]. The selected material for the study was the NbTiN which, due to its higher critical temperature (between 16 K and 17 K), presents a lower BCS surface resistance at 4.2K than Nb [3],[4].

In this comunication we present the major results on NbTiN samples that helped to define the process parameters, the RF behavior of the NbTiN film measured using the TE011 cavity and some preliminary results on 1.5 GHz Nb/Cu and NbTiN/Cu cavities.

2. NbTiN SAMPLE RESULTS

NbTiN films deposited on polished SiO_2 substrates were used to measure the film critical temperature and its room temperature resistivity using a 4 points probe method. The film thickness was measured using a Taly-step device.The thickness of the sputtered NbTiN films was between 0.7 and 4 µm.

Chemical analysis were made for very thin films (<5000 Å deposited on polished graphit samples) using RBS and NRS technics [5], and for thicker films (1 µm to 15 µm deposited on copper samples) using GDS (Glow Discharge Spectroscopy).

All the NbTiN films have been prepared on the new sputtering setup (mentioned in the introduction) which had only been used for Nb coatings before. An exhaustive study of the sputtering parameters was thus necessary (as we did with our first setup in ref [1],[4]).

The curves on figure 1 show that there is an optimum value of the nitrogen flow for which the film has the highest critical temperature and the lowest resistivity at room temperature. Nevertheless this optimum value changes if we modify the parameters of the glow discharge.

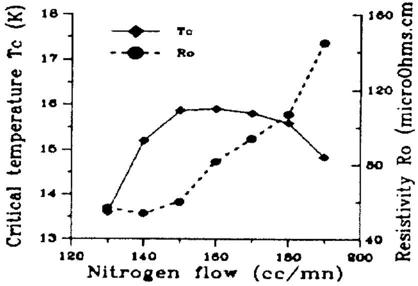

Figure 1: *typical variation of the film's critical temperature and resistivity at room temperature versus the nitrogen flow. For this example the argon flow was 250 cc/min (partial pressure of $7*10^{-3}$ mbar), the glow discharge power was P=600W, the film thickness was 1.7µm and the atomic Nb/Ti ratio was 0.52/0.48. The deposition speed is constant with the nitrogen flow (420±20 Å/min).*

We observed, like other authors [6], that the optimum value of the nitrogen flow is proportional to the power applied to the magnetron cathode. In addition we observed that the deposition speed (and thus, the metal atoms flow at the substrat's surface) is proportional to the magnetron power (figure 2). This confirms a result which is commonly observed in magnetron sputtering.

It is then clear that these two parameters, magnetron power and nitrogen flow (or nitrogen partial pressure), are the two main parameters controlling the reaction between the Nb or Ti atoms and the nitrogen ones. To obtain the good stoechiometric composition of the film (one nitrogen atom for one metal atom) the flows of metal and nitrogen atoms impinging the substrat's surface must be carefully adjusted.

Argon partial pressure, substrate temperature, magnetron cathode temperature, cathode-substrate distance and pumping speed are also important, especially to minimize the film internal stress. The high rate of internal stress may cause the coating's blistering , when the adhesion with the substrat is poor (this will later be detailed for the NbTiN films sputtered inside some copper cavities). Therefore , all parameters have to be choosen carefully and their large number makes this optimization step quite complex.

One of the basic questions in reactive sputtering is to know whether the reaction takes place on the substrate surface, on the magnetron cathode surface, or during the transport of the metallic vapor between the two surfaces. The results described hereafter seem to show that the reaction might take place on the cathode surface.

Three samples have been deposited at the same time at different distances from the magnetron cathode: d=7, 8 and 12.5 cm. The range of the deposition speed of these films (and thus the sputtered metal atoms flow on the substrate surface) varies by a factor of 3.18 following a low in $1/d^2$, whereas the nitrogen flow depends only of the nitrogen partial pressure and is therefore the same for the three samples. The critical temperatures of these films are respectively 16.15, 16.04, and 15.95 K, whereas for films deposited with the same deposition speed a 20 % variation of the nitrogen flow leads to a drastic decrease of the Tc values (shown on figure 1).

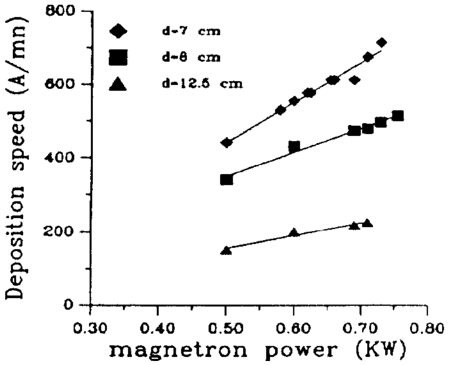

Figure 2: *the deposition speed versus the power of the glow discharge for three cathode-substrate distances (d). Argon and nitrogen partial pressures were kept constant* ($p_{Ar}=7*10^{-3}$*mbar and* $p_{N2}=2.5*10^{-3}$*mbar). A correction based on the cosine low was used each time a substrate was sputtered under an angle.*

We observed that for constant nitrogen flow, discharge power and substrate temperature, the film's critical temperature and resistivity were fairly constant regardless of the film's thickness (over 1µm).

The NbTiN films thinner than 1µm presented a lower critical temperature and a higher resistivity. This might be due to the fact that the samples reach a thermal equilibrium only after ~30min of deposition. An other possible explanation is the fact that at the begining of the sputter process the film is sligtly contaminated (O,H,C) by the adsorbed gases on the samples' surface and by the residual pressure of H_2O and CO_2. After the first micron is sputtered, the film plays a gettering role and the next microns of the layer contain less impurities.

3. RF CHARACTERIZATION USING THE TE011 CAVITY

In the next paragraphs we present the follow up of the RF characterizations made with the TE011 cavity developed at Saclay. This cavity and the method applied for the RF tests had already been described in detail previously [1]. In our last communication [1] we presented two main results:

- the first was the low residual surface resistance obtained (figure 3: R_{res}=40 nΩ for F= 4 Ghz and T=1.6 K) at low field level and the maximum RF field level B_{max}=34 mT.

- the second was the large spread of the R_{res} values measured on different NbTiN coatings: from 40 nΩ to 2000 nΩ at low field level.

We observed after the RF tests that the copper substrates had been deformed during mounting on the TE011 cavity. We increased the copper disk's thickness from 2 mm to 3 mm to diminish the deformations, and the spread of the RF results obtained on the next NbTiN coatings was considerably lowered as shown on figure 3.

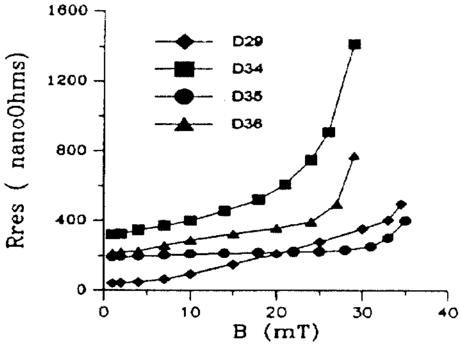

Figure 3: *Residual surface resistance of the last three NbTiN films deposited on 3 mm thick copper substrats compared to our best result previously obtained (D29)*

One hypothesis to explain the mechanical deformations' influence over the surface resistance could be the creation of micro-cracks on the NbTiN films surface. It is well known that the magnetron sputtered nitrides are brittle and cracks may appear under small deformations of the substrate. A great number of such micro-cracks could be the cause of RF dissipations. The confirmation of this hypothesis is of prime importance. It can be made by the analysis of the toughness of the films which we haven't done yet [7].

4. RESULTS ON 1.5 GHZ COPPER CAVITIES

Single cell copper cavities have been hydroformed at CERN [2]. Before sputtering, the copper cavities have been mechanically and then electrochemically polished. In figure 4 we describe the main elements of the electropolishing setup. The cavity is horizontaly placed (like on the KEK system for Nb polishing [8]), half filled with H_3PO_4 acid mixed with water and revolves around its axis at a speed of 3-5 turns per minute. The cathode, reproducing the cavity's shape, is fixed at a distance of 2-3 cm from the cavity's inner surface. A membrane, fixed all around the cathode, guides the H_2 bubbles to the upper half of the cavity. The current density is 5 A/dm^2. After the electrochemical treatment, the cavity is rinsed in ultra pure water, chemically treated with sulfamic acid and then rinsed again with utra pure water and dried in a class 100 clean air room. The film is afterwards prepared following the process described in ref [2].

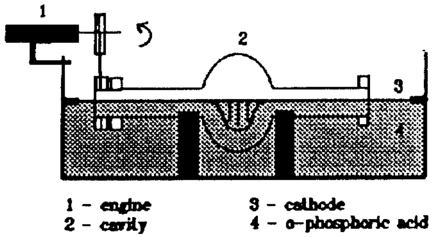

Figure 4: *electropolishing system*

4-1. Results on 1.5 GHz Nb/Cu cavities

Before sputtering any NbTiN coatings, we decided to prepare a Nb coated cavity so as to test the copper electrochemical treatment. After sputtering, the Nb coating was rinsed with ultra pure water and dried in a class 100 clean air room. The RF measurements are presented on figure 5. At T=1.6 K, $Q_0=10^{10}$ at low field level and decreases with the RF field down to 6.5 10^8 at E_{acc}=12.5 MV/m, limited by the amplifier. The cavity was not limited by Q-switches or by electrons. We will not discuss here the slope of the curve on figure 5, which is generaly observed with sputter coated cavities [9],[10] and interpreted in terms of granular superconductivity. This good result encouraged us to prepare NbTiN/Cu cavities using the copper's electrochemical polishing treatment.

4-2. Results on 1.5 GHz NbTiN/Cu cavities.

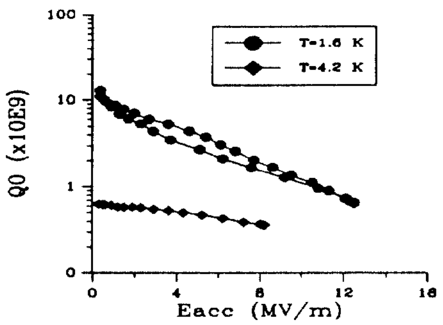

Figure.5: *RF measurements of S1-03 Nb/Cu cavity.*

Four NbTiN/Cu cavities were sputtered but unfortunately none could be tested. Indeed, the film's internal stress and probably the local poor adhesion caused the coating's blistering for all the cavities.

The process parameters were modified between the first and the last of the four cavities. External copper temperature was kept under 70°C during the process and argon partial pressure was incresed up to 10^{-2} mbars, which diminished the film's stress.

The sulfamic acid treatment was supressed and this seemed to somehow ameliorate adhesion.

Progress between the first and the last of the sputtered cavities is obvious but effort is still needed for process improvements.

5. CONCLUSION

Several sample studies have stressed the potential of NbTiN for RF applications. Process parameters were analysed and optimised working points were defined. Reproductibility of the RF surface resistance was studied on a TE011 cavity. The different sequences of the elaboration of 1.5 GHz sputter coated cavities (electrochemical polishing, rinsing, sputtering and final rinsing) were tested and work in a satisfactory manner.

We have obtained for the first Nb/Cu cavity a quality factor of $Q_0=10^{10}$ at low field level, and a maximum field of E_{acc}=12.5 MV/m without Q-switches or electron loading. The first series of NbTiN/Cu sputtered cavities could not be tested as the layer had several square milimeters blistering. Nevertheless, important progress is actualy being made.

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