Study of NbTiN coatings by reactive magnetron sputtering for the production of 1.5 GHz superconducting accelerating cavities

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Abstract

The production of thin films of $Nb_{.35}Ti_{.65}N$ by magnetron sputtering is being studied for application to the realisation of 1.5 GHz accelerating cavities. Samples produced in a sputtering chamber having the same geometry as the RF resonator reached a T_c value of 15.5 K, a RRR of 1.45 and a low temperature DC resistivity of 35 $\mu\Omega$cm. With the parameters optimised in this way, a coating was produced inside a seamless copper cavity: the RF test showed a T_c for the cavity of 14.2 K, a residual resistance at low field of about 350 nΩ and a BCS resistance at 4.2 K of 55 nΩ. The latter figure, compared to the R_{BCS} of Nb/Cu and bulk Nb resonators of the same frequency (about 400 nΩ and 900 nΩ respectively), confirms the potentiality of NbTiN for applications at 4.2 K. However, improvements on the residual resistance and the dependence of the resistance as a function of the accelerating field are still necessary to get an overall performance comparable to that of standard Nb resonators.

1. Introduction

NbTiN is one of the most suitable materials for the production of superconducting thin films with T_c up to 17 K and relatively low DC electrical resistivity ρ_0 [1]. These features make it very promising for RF applications, since the BCS theory of superconductivity provides (in the "dirty limit" $l \ll \xi_o$ and for $T<T_c/2$) an approximate variation of the RF surface resistance of the form

$$R_{BCS} = \frac{A\omega^2}{T}\,\rho_0^{1/2} \quad \exp(-\alpha T_c/T) \tag{1}$$

and therefore indicates that important improvements with respect to Nb can be potentially expected from superconductors having a higher critical temperature, especially for working temperatures of 4.2 K or more.

For this reason a research programme on the development of NbTiN/Cu accelerating cavities had been undertaken at CERN during the 80's, with rather encouraging results [2]. This study, which had been stopped for some years during the series production of the LEP cavities, has been recently restarted. The purpose of this paper is to illustrate the experimental methods employed and to describe the present status of advancement.

2. The reactive sputtering of NbTiN thin films

The layout of the sputtering system is shown in Fig.1. It is based on the same cylindrical magnetron set-up developed at CERN for the production of the 1.5 GHz Nb/Cu cavities [3]. The central section of the cathode is made in this case of a $Nb_{.35}Ti_{.65}$ alloy, and is inserted between two Nb tubes which are used for the deposition of a pure Nb film inside the cavity cut-offs.

In order to determine the sputtering parameters required for the production of good quality superconducting films, a series of samples has been preliminarily produced inside a dummy stainless-steel cavity equipped with rotateable sample-holders. Then a first 1.5 GHz accelerating cavity has been coated, starting from a seamless copper cavity made by spinning and preliminarily treated with SUBU® chemical polishing.

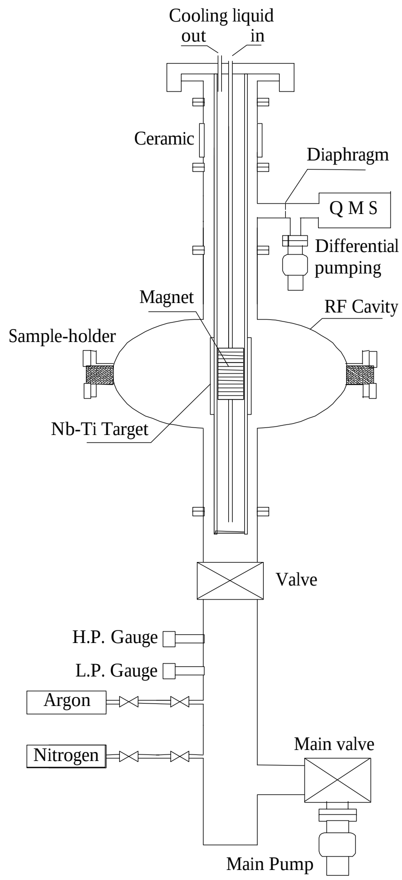

Fig.1: The sputtering system for the deposition of NbTiN thin films inside 1.5 GHz resonators.

Prior to sputtering cavity and samples are heated to 200^0C, and the discharge power then increases their temperature up to about 350^0C. In the cell the discharge is started without nitrogen, with the same sputtering parameters used for Nb coatings, namely discharge current of 3 A, discharge power of about 1.2 kW and argon pressure of 1.5 10^{-3} mbar. The NbTi layer deposited in this way is seen to improve the adhesion of the nitride film on the substrate and, by virtue of its T_c of about 9 K, provides a further barrier against the RF field penetration into the Cu substrate at possible defects of the nitride film. Nitrogen is then injected in the system through an independent line. Its flow rate is controlled by a calibrated flowmeter and its partial pressure $P(N_2)$ inside the cell is monitored by a quadrupole mass spectrometer. To this end it is essential that the latter be connected via a conductance to the upper end of the cavity, i.e. opposite to nitrogen injection, in such a way that the measurement is not altered by the pressure gradient due to discharge pumping.

It is known from the literature that during reactive sputtering the consumption rate of the reactive gas is generally related in a complicated non-linear way to the injection and to the sputtering rates [4]. In our sputtering system it is found that for a constant N_2 injection rate, optimised with respect to the initial conditions, $P(N_2)$ shows a tendency to increase steadily with time, due to a decreasing nitrogen consumption. To prevent the evolution of the above phenomenon towards an extensive nitridation of the cathode, which is found to be highly detrimental to film quality, $P(N_2)$ is kept constant at a chosen value by manually adjusting when necessary the nitrogen flow. With this method quasi-stable operating conditions can generally be approached within some minutes.

For the production of the NbTiN/Cu resonator, the sputtering system was assembled in a class 100 clean room, to prevent dust contamination. The duration of the nitride deposition was 30' which, according to the measured sputtering rates, corresponds to a thickness of about 2 μm, i.e. ten times the expected penetration depth of the RF field into the NbTiN film [1].

3. Sample characterisation

Following the procedure described in the previous paragraph, various samples have been produced with different values of $P(N_2)$, in order to optimise this parameter. It is however to be kept in mind that in this context the nitrogen partial pressure becomes physically meaningful only once all the other experimental conditions - in particular the sputtering rate - are exactly specified, and that therefore the results given below cannot be directly applied to different sputtering systems.

The values of the critical temperature, the residual resistivity ratio (RRR) and the low temperature DC resistivity in the normal state are displayed in Figs. 2-4 as a function of $P(N_2)$. These data were obtained from samples deposited onto quartz substrates, by using an apparatus that allows performing 4-point electrical measurements as a function of temperature inside a cryostat refrigerated with liquid helium. It can be seen that both T_c and RRR are maximised in a region of $P(N_2)$ around 6 10^{-4} mbar, which therefore was chosen for the production of the NbTiN/Cu cavity. In particular, the highest critical temperature is 15.5 K, which is to be compared to the best values of about 17 K reported in the literature [1] for samples deposited in a planar magnetron sputtering configuration, on substrates at 600^0C and with a higher Nb/Ti ratio.

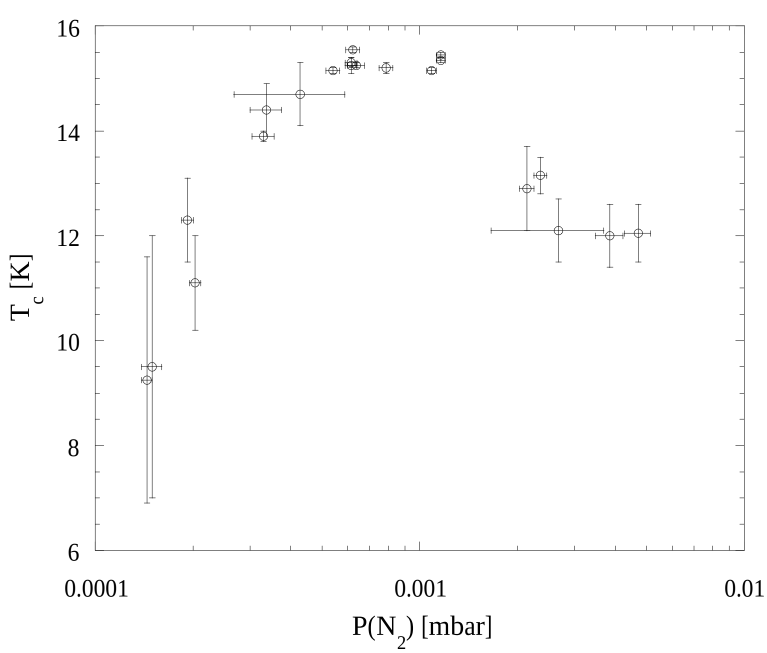

Fig. 2: T_c obtained on quartz samples as a function of the N_2 partial pressure inside the cell during the coating. Data refer to the sputtering configuration shown in Fig. 1 and the discharge parameters indicated in the text.

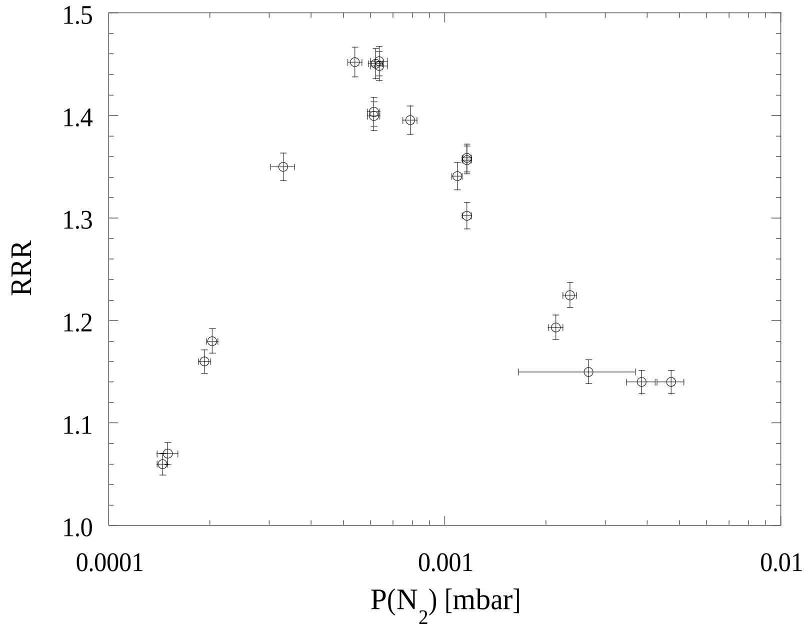

Fig. 3: The RRR for the same samples of Fig.2.

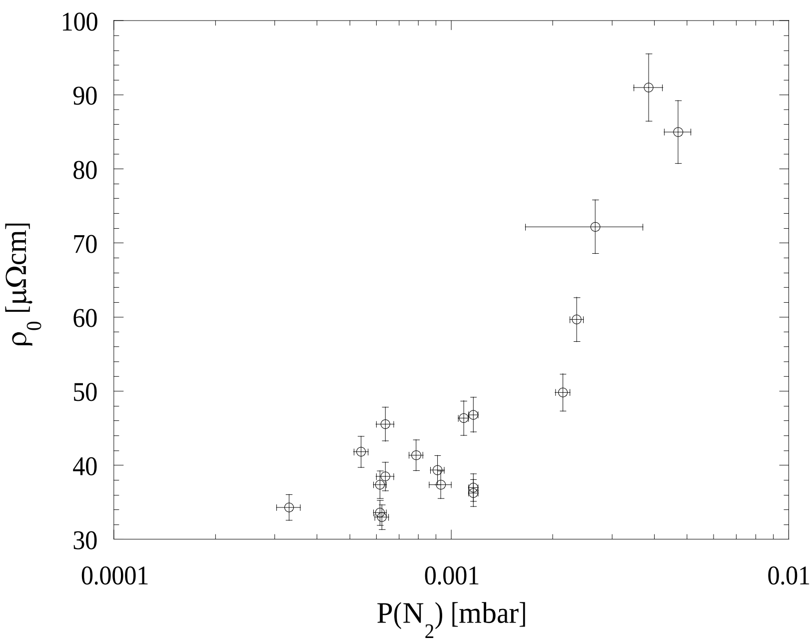

Fig. 4: Low temperature DC resistivity in the normal state for the same samples of Fig.2.

4. The RF tests on the 1.5 GHz NbTiN/Cu resonator

The NbTiN/Cu cavity was tested with the same procedure and apparatus used at CERN for the study of the 1.5 GHz Nb/Cu cavities [3]. The surface resistance R_s (related to the quality factor Q by the relation $R_s = \Gamma/Q$, with $\Gamma = 295\,\Omega$) was measured for various values of the temperature, of the accelerating field (or, equivalently, of the average magnetic field H_{rf} at the inner cavity surface) and of an external DC magnetic field H_{ext} generated by a solenoid surrounding the resonator. The T_c of the cavity was measured with Hall probes placed close to its external surface, which detect at the transition point the penetration of H_{ext} into the cavity walls. The lower critical field H_{c1} was determined by applying near the cavity wall the magnetic field generated by a superconducting coil. Finally, the zero temperature penetration depth $\lambda(0)$ was calculated from a theoretical fit of the temperature dependence of the resonator frequency. Both $\lambda(0)$ and H_{c1} are given through their ratio to the corresponding values measured with the same experimental set-up for annealed bulk Nb resonators.

Three curves of R_s vs. H_{rf} at different temperatures are displayed in Fig. 5. In Fig.6 the temperature dependence for a fixed low RF field is shown, allowing the residual and the BCS term in the surface resistance to be separated, and providing an estimation for the α parameter in eq. (1). Fig. 7 shows the additional surface resistance R_{fl} induced by the field H_{ext} as a function of H_{rf}. From the fit the parameters R^0_{fl} and R^1_{fl} are calculated, which respectively describe the linear contribution of H_{ext} to the resistance at low RF field and to the "slope" of the R_s vs. H_{rf} curve [5].

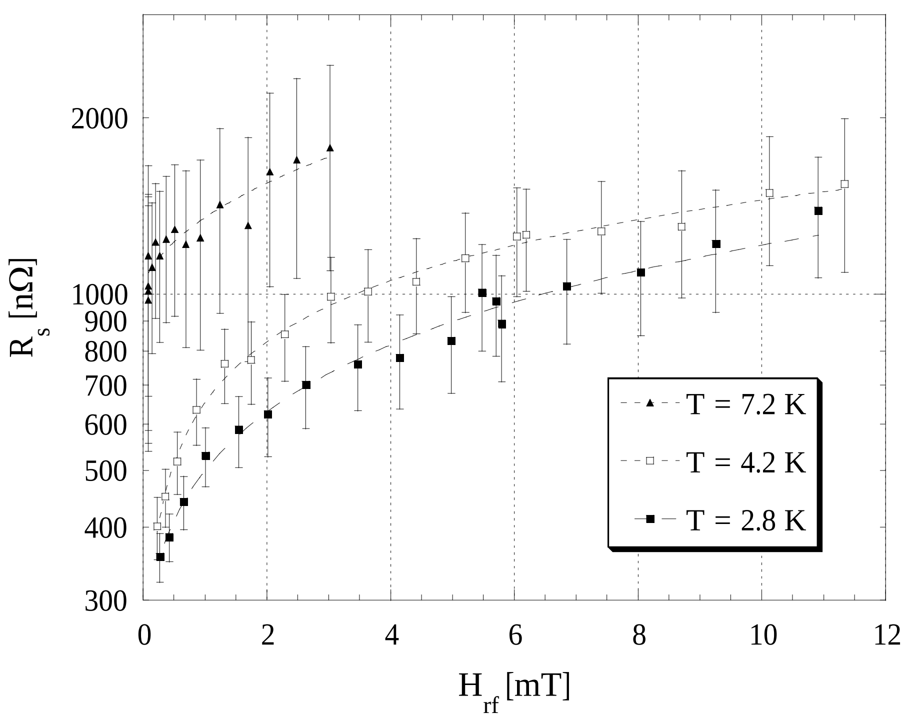

Fig. 5: Surface resistance of the 1.5 GHz NbTiN/Cu cavity as a function of the RF field at different temperatures. E_{acc} = 1 MV/m corresponds to H_{rf} = 4.55 mT.

Some of the most significant parameters obtained from the measurements are summarised in Table 1, together with the corresponding values obtained for standard Nb/Cu and bulk Nb cavities of the same frequency [3,5]. It can be seen that the expectations concerning the improvement of R_{BCS} at 4.2 K are fulfilled (55 nΩ at low accelerating field, compared to about 400 nΩ for Nb/Cu and 900 nΩ for bulk Nb), but the total surface resistance of the cavity suffers from the considerably higher value of the residual term, and moreover (see Fig. 5) the increase of R_s as a function of H_{rf} is much more pronounced than for standard Nb cavities. As a result, only at very low values of the accelerating field, the performance of this cavity becomes equivalent at 4.2 K to the best Nb/Cu cavities made at CERN.

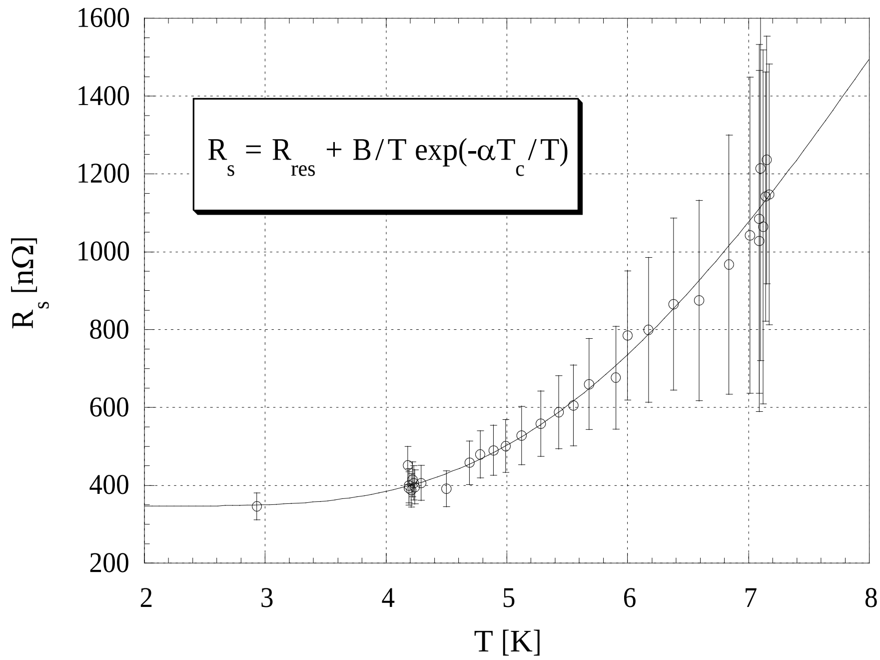

Fig. 6: Variation of the surface resistance as a function of temperature for H_{rf} = 0.23 mT. From the displayed fit, putting T_c = 14.2 K, one gets R_{res} = 350 nΩ and α = 2.3.

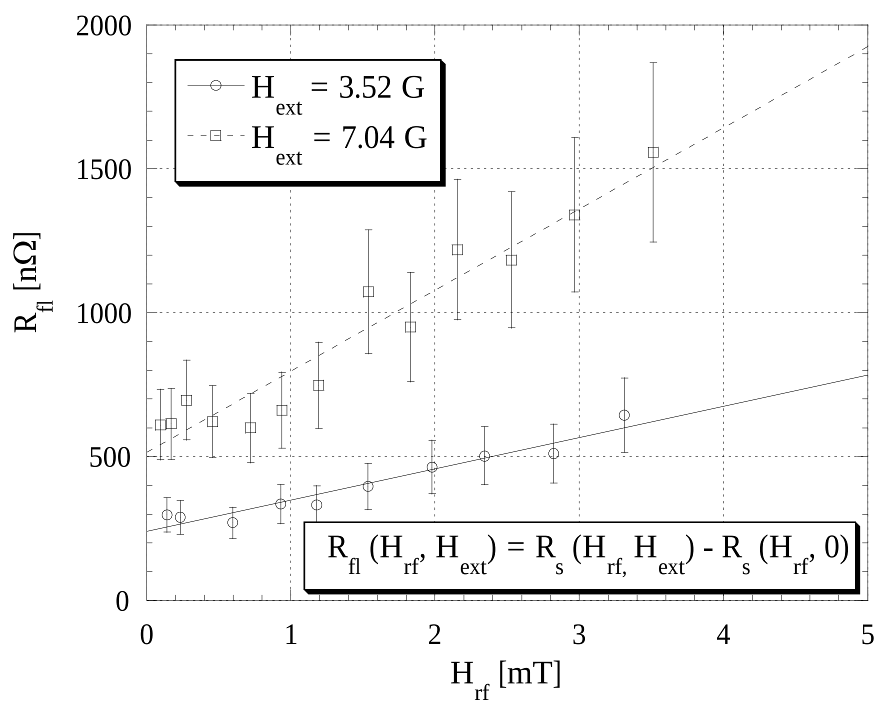

Fig. 7: Variation of the contribution induced by H_{ext} to the surface resistance of the resonator, as a function of the RF field H_{rf} for T = 4.2 K. The displayed fit is of the form: $R_{fl} = (H_{ext} - H_{th})(R^0_{fl} + R^1_{fl}H_{rf})$.

	NbTiN/Cu	Nb/Cu	bulk Nb
T_c [K]	14.15 ± 0.06	9.54 ± 0.06	9.28 ± 0.06
$\lambda(0) / \lambda(0)^{bulk\ Nb}$	4.8 ± 0.5	1.58 ± 0.09	1.00 ± 0.05
$H_{c1} / H_{c1}^{bulk\ Nb}$	0.24 ± 0.01	0.67 ± 0.01	1.00 ± 0.01
R^0_{BCS} (4.2K) [nΩ]	55 ± 15	401 ± 1	905 ± 12
$\alpha = \Delta(0)/kT_c$	2.3 ± 0.4	2.05 ± 0.04	2.02 ± 0.05
R^0_{fl} [nΩ/G]	75 ± 10	4.8 ± 0.1	127 ± 4
R^1_{fl} [nΩ/GmT]	38 ± 5	1.13 ± 0.02	1.15 ± 0.09

Table 1: Some characteristic parameters of the NbTiN/Cu cavity, compared to the corresponding values for Nb/Cu and bulk Nb cavities.

An optical inspection, performed by introducing a camera into the cavity, showed the presence of a certain number of small localised defects of accidental nature, which can account for a fraction of the observed residual resistance. Further cavity prototypes, to be produced in the frame of this research project with different sputtering parameters, substrate temperature and cathode composition, will hopefully give indications about the possibilities of overcoming the present difficulties and achieving results of relevance for of practical applications.

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