NbTiN thin films for RF applications

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Abstract

This paper presents some results of the study of $[Nb_{(1-x)}Ti_x]N$ superconducting coatings prepared by reactive magnetron sputtering for making accelerating cavities at 1.5 GHz. A series of films deposited on samples have been prepared using a new magnetron cathode developed at Saclay. The RF tests have been performed with a cylindrical TE011 cavity made out of massive niobium, with a dismountable end plate. One test demonstrated that NbTiN films can have a very low residual surface resistance at 1.6 K, comparable to that of the bulk niobium; and at 4.2 K, a surface resistance about a factor of 5 lower than that of bulk niobium due to its higher Tc, 16.2 K.

Introduction

Superconducting cavities made out of bulk niobium perform remarkably: $Q_0 \geq 5.10^{10}$ and Eacc $\geq$ 25 MV/m for monocell cavities. At the frequency of 1.5 GHz used at Saclay, the superconducting properties of niobium (Tc=9.2 K) dictate an operating temperature below 2 K in order to get a reasonable RF surface resistance (typically 20 nΩ). The high cost of the cryogenic installations used for these cavities led us to explore the possibility of making superconducting cavities working at a higher temperature (4.2 K). Previous results [1,2] showed that NbTiN coatings prepared by reactive magnetron sputtering are good candidates for this purpose.

At first, we prepared NbTiN samples at the CE Grenoble using a planar magnetron sputtering device [3]. The RF surface resistance of these films deposited on copper discs was measured using a cylindrical TE011 cavity made out of bulk niobium with a dismountable end plate. The first results obtained were encouraging: at 4.2 K and 4 GHz, a surface resistance Rs=1600 nΩ was measured, compared to 5200 nΩ for Nb. The residual surface resistance R_{res}, though, remained high: 400 nΩ compared to 70 nΩ for niobium, and the maximum field level attained before quenching was 28 mT (equivalent to Eacc=7 MV/m).

With the purpose of further improving these results, we have developed a new magnetron cathode for a second sputtering installation at Saclay.

In this paper we present the results of the first $[Nb_{(1-x)}Ti_x]N$ coatings prepared using this new cathode. The RF characterizations of these superconducting NbTiN coatings deposited on copper discs have been performed with the same TE011 cavity.

Elaboration and characterization of sample coatings

The samples were prepared by reactive magnetron sputtering. The NbTi magnetron cathode used at Saclay is 10 cm long , 2 cm in diameter and has a 3 mm wall thickness. The magnetic field is generated by water cooled permanent magnets and is set at 20 mT parallel to the cathode surface. The Nb/Ti ratio of the films can be changed by adding Nb rings of different heights in the $Nb_{0.36}Ti_{0.64}$ magnetron cathode tube.

The magnetron cathode can be retracted from the sputtering chamber. That allows us to prepare the cathode by sputtering its surface outside the deposition chamber before coating the samples, or to protect it when the sample cleaning, by reverse sputtering ion bombardment, is in progress.

The sputtering apparatus is equiped with a 140 l/s turbomolecular pump and a 12 m^3/h primary pump . High purity (99.9995 %) argon and nitrogen gas flows are regulated by flowmeters.

The film thickness has been measured with a Taly-step gauge. The chemical analysis uses ion beam techniques, RBS and NRS [4] for very thin films ($\leq$ 5000 Å), and GDS (Glow Discharge Spectroscopy) for thicker films (1 to 15 µm).

With the reactive magnetron sputtering technique we can vary many parameters to optimize the superconducting properties of the coatings: argon pressure; nitrogen pressure; discharge power; and substrate temperature. These

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parameters are strongly correlated to the characteristics of the setup: geometry; pumping speed; magnetron cathode surface; cathode geometry; and the magnetic field at the cathode surface. This is why we first made a rough parametric study of the sputtering setup based on an analysis of the nitrogen consumption and the magnetron target voltage during sputtering [3,5,6]. Thus we determined the range of the elaboration parameters which gave the highest Tc films. Then a more refined analysis could be made and we fixed the distance between the substrates and the cathode surface (8 cm), the argon pressure (5.10^{-3} mbars), and the discharge current (1.5 A corresponding to power values ranging from 600 to 680 W). With these conditions the glow discharge was very stable, and that allowed us to investigate the dependence of the Tc on the nitrogen partial pressure during sputtering. Fig 1 represents a typical evolution of Tc versus the nitrogen gas flow rate F(N2). The maximum Tc value obtained for $(Nb_{0.55}Ti_{0.45})N$ films deposited on SiO_2 substrates is Tc=16.0 K with ΔTc<0.1 K for f(N2)=8.75 cc/mn (corresponding to a nitrogen partial pressure of 1.5 10^{-3} mb without the glow discharge). The thickness of these coatings was 2.2 µm.

The determination of the dependence of Tc on the Nb/Ti ratio of the films, on the argon partial pressure, on the intensity of the glow discharge, and on the thickness of the films is now in progress.

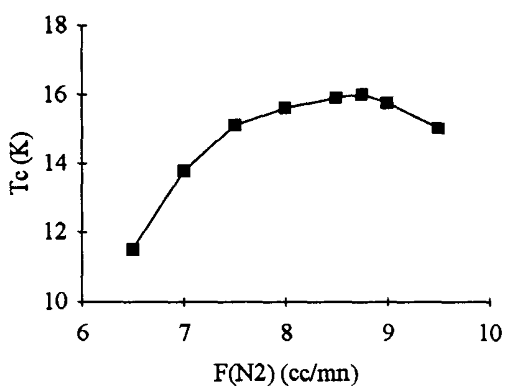

Fig 1 Tc-value vs nitrogen flow rate $F(N_2)$ for $(Nb_{0.55}Ti_{0.45})N$ / SiO_2 coatings.

Copper substrate surface preparation

For the RF tests (described below) the NbTiN films are deposited on 12 cm diameter copper discs. The surface preparation of the copper is essential to the adhesion of the thin film in order to have good thermal contact between the copper and the film.

The samples were first mechanically polished (~12µm), then cleaned in hot soap, and rinsed in demineralized water. Lastly, an electrochemical treatment was made just before the sputtering of the NbTiN film. The thickness removed before the sputtering was about 20 µm.

The electrochemical treatment tested was done in an electrolytic solution of orthophosphoric acid. The current density was 230 A/m^2. The rate of attack was about 0.5 µm/mn. This treatment gave a mirror like surface, and the roughness determined by Ra values is of the order of 0.1 µm.

We verified, in water with ultrasound, the good adhesion of the NbTiN coatings on the copper surfaces prepared with this electrochemical treatment.

The cleaning of the copper sample by ion bombardment was then made "in situ" just before the sputtering. This treatment seemed to be necessary to prevent the creation of tiny uncoated spots on the copper surface. The power used for the sputter cleaning was about 1 W/cm^2 during 15 mn at an argon pressure of 2.10^{-2} mb.

RF characterization of the NbTiN films

We made the RF characterizations on NbTiN films deposited on copper removable end plates mounted on our cylindrical TE011 massive niobium cavity equipped with a variable input coupler [7]. Before testing, all the discs were rinsed with filtred ultra-pure water (18 MΩ) and dried in a class 100 clean air room.

We have operated at the TE011 and TE012 modes, without any electric field at the surface, with resonance frequencies of 4 GHz and 5.7 GHz, respectively . The quality factor Q of the cavity was measured by the usual decrement method.

Using this cavity we could determine the RF surface resistance of the superconducting thin film by comparing measurements made using a massive Nb (RRR=180) end plate with the measurements made using a NbTiN sputtered copper end plate. We obtained the surface resistance of the coating by subtracting the contribution of the cavity body from the total surface resistance of the cavity.

The method assumes that the surface resistance of the niobium cavity remains invariant from one test to another. This reproducibility was confirmed in the first experiments carried out with niobium discs without any chemical treatment of the cavity. Nevertheless, the niobium surface resistance of the cavity is periodically checked by performing one test with a Nb disc after two tests with NbTiN / Cu discs.

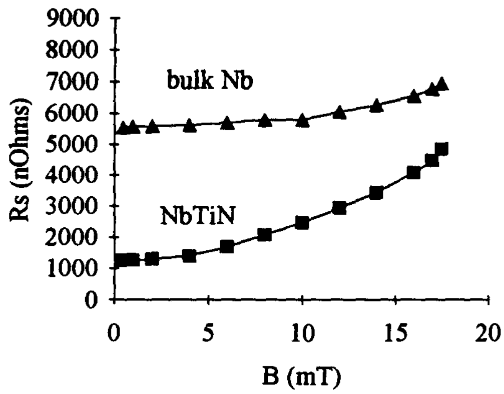

Fig 2: Surface resistance of the D29 $(Nb_{0.55}Ti_{0.45})N$ / Cu film at 4 GHz and 4.2 K.

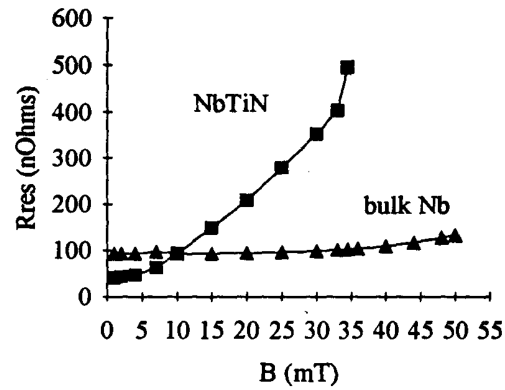

Fig 3: Residual surface resistance of the D29 $(Nb_{0.55}Ti_{0.45})N$ / Cu film at 4 GHz and 1.6 K.

At T=1.6 K, in these conditions, the major part of the error on the coating R_{res}, which is determined by the uncertainty on the Nb R_{res}, is +/- 20 nΩ at 4 GHz, and +/- 50 nΩ at 5.7 GHz.

At 4.2 K the R_s error results from each of the errors on the two Q measurements successively performed with the Nb and the coated discs. The Q error is estimated to be +/- 4% with our experimental precautions. The error becomes very large when the coating surface resistance is lower than the Nb one.

We also obtained the critical temperature and the penetration depth for the films (if thick enough) from quality factor and resonance frequency measurements as a function of temperature from 10 K to 17 K.

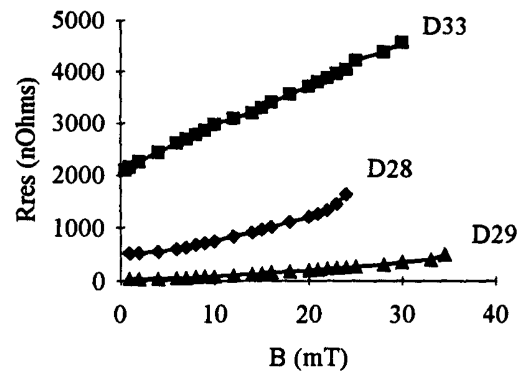

Fig 4: R_{res} at T=1.6 K and F=4 GHz for NbTiN coatings of Tc 16.17 K (D29), 16.31 K (D28), and 16.0 K (D33)

Results of the RF tests.

The best result we obtained is presented on fig 2 and fig 3. At T=1.6 K and F= 4 GHz the R_{res}, 40 nΩ, is as low as that of bulk niobium at low field level; it increases with the field level and reaches 500 nΩ at 34 mT (limited by a quench). At T=4.2 K the Rs is much lower than for bulk niobium and then measurements errors become very large.

However, for T < 1.8 K, we have obtained a large spread of R_{res}. For instance (fig.4) the last three comparable NbTiN/Cu discs, which had Tc between 16 and 16.3 K, had R_{res} from 40 to 2100 nΩ at a low field.

For these discs the RF magnetic field limit due to quench reached values from 24 to 34 mT.

In using a program based on a model developed by H. Safa [8], we found a penetration depth in the range of 300nm, in good agreement with the values obtained by S. Calatroni at CERN [9].

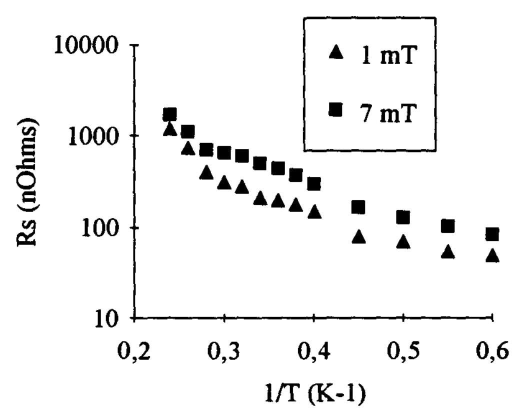

Fig 5: Surface resistance as a fonction of 1/T for the D29 NbTiN coating.

As shown on fig 5, below T=2.4 K (1/T=0.42 K^{-1}) the variation of the Rs with the temperature is rather slow, probably because of the high Tc value (greater than 16 K). In spite of the fact we had only a few experimental points with large errors between 4.2 K and 2.4 K, we could fit them by the BCS resistance computed by Safa's program [8] with a very low gap parameter $2\Delta/kTc$ of value 1.7 (to be compared to $2\Delta/kTc$=3.56 obtained for bulk Nb by the same calculation). This probably results from the granular structure of the NbTiN films.

The large R_{res} increase with magnetic field (fig.3 and 4) is also certainly due to the granular aspect of the superconducting coatings deposited by sputtering techniques [10].

The behaviour of the surface resistance as a function of the frequency is not clear. Whereas for bulk niobium the R_{res}, as well as the BCS Rs, depends on F^2; for our last NbTiN films this F^2 dependence is only roughly observed for R_{res}; Rs at 4.2 K seems to be invariant at our operating frequencies of 4 GHz and 5.7 GHz.

The limitations observed with the NbTiN coatings subjected to RF power seem to be quite different from those observed with the niobium coatings we had also prepared on monocell copper cavities [11,12]. One of the main limitations of the Nb/Cu coatings is a "Q-switch" (sudden Q drop with the field increase caused by a superconducting to normal phase transition) due, for example, to a poor thermal contact to the copper underneath. With our NbTiN coatings we have never observed such a Q-switch. The spread on R_{res} values obtained at low field are also lower for Nb coatings than for NbTiN ones. Many efforts are going on to understand these differences.

Conclusions

We have shown that NbTiN films are a very promising material for accelerating cavities. They can have a low residual surface resistance value, comparable to that of the bulk niobium. We obtained R_{res}=40nΩ at F=4 GHz and T=1.6 K at a low field level for one coating tested with a TE011 cavity. In addition, these films have critical temperatures higher than 16 K, and consequently their surface resistance at T=4.2 K is much lower than that of bulk niobium (five times lower for the best result). A maximum field level of 34 mT has been reached; value which corresponds to an accelerating field of 8.5 MV/m.

The main problems we still have to solve are the R_{res} spread that we obtain on films prepared under the same conditions, and the R_{res} increase with magnetic field. We will also start soon to prepare sputter coated 1.5 GHz monocells cavities.

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