

# PHYSICAL VAPOUR DEPOSITION OF NbTiN THIN FILMS FOR SUPERCONDUCTING RF CAVITIES\*

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## Abstract

The production of superconducting coatings for radio frequency (RF) cavities is a rapidly developing field that should ultimately lead to acceleration gradients greater than those obtained by bulk Nb RF cavities. The use of thin films made from superconductors with thermodynamic critical field,  $H_C > H_C^{Nb}$ , allows the possibility of multilayer superconductor – insulator – superconductor (SIS) films and accelerators that could operate at temperatures above 2 K. SIS films theoretically allow increased acceleration gradient due to magnetic shielding of underlying superconducting layers [1] and higher operating temperature can reduce cost [2]. High impulse magnetron sputtering (HiPIMS) and pulsed DC magnetron sputtering processes were used to deposit NbTiN thin films onto Si(100) substrate. The films were characterised using scanning electron microscopy (SEM), x-ray diffraction (XRD), Rutherford back-scattering spectroscopy (RBS) and a four-point probe.

## INTRODUCTION

The cooling costs required to run a particle accelerator at 4.2 K are small when compared to the expense at 2 K. The drawback of running Nb SRF cavities at 4.2 K is that  $R_{BCS}$  becomes significant resulting in an unsatisfactory drop in Q factor. Superconducting materials with  $T_C > T_C^{Nb}$  can have smaller  $R_{BCS}$  than Nb at 4.2 K and therefore larger Q factors whilst still saving on cooling costs. It is possible to reduce  $R_s$  further by combining high  $T_C$  with as small a normal state low temperature resistivity as possible [3].

NbTiN is a good candidate to replace Nb as the material of choice for SRF cavity fabrication. NbTiN has a  $T_C$  of 18 K, compared to the 9.2 K of Nb, and has adequately small low temperature normal state resistivity to suggest it can operate with small  $R_s$  at 4.2 K. The development of NbTiN cavity fabrication is therefore of great interest to the SRF community.

Further uses of NbTiN technology is in multilayer SIS films. Multilayer SIS films have been suggested to increase accelerating voltages by utilising an increased first critical field ( $B_{c1}$ ) for superconducting layers with  $H_C > H_C^{Nb}$  and thickness ( $d$ ) less than the penetration depth,  $\lambda$ , to shield an underlying Nb layer. Equation 1 shows that as film

thickness and coherence length,  $\xi$ , get smaller then  $B_{c1}$  increases in the overlying superconducting layers [1].

$$B_{c1} = \frac{2\phi_0}{\pi a^2} \ln \frac{d}{1.07\xi} \quad d < \lambda \quad \text{Eq. 1}$$

The purpose of the present study is to deposit and then characterise a selection of NbTiN thin films that would be suitable for use in both SIS multilayer coatings or for operation in accelerators at 4.2 K. NbTiN was considered due to its small normal state resistivity of 37  $\mu\Omega\text{cm}$  [2], high  $T_C$  of 18 K,  $\xi$  of 3.8 nm [4] and  $\lambda$  of 150 nm. Films were deposited by reactive sputtering in a mixture of Kr and N<sub>2</sub> gas using three inch planar magnetrons utilising either an Ionautics HiPSTER 1000 high impulse magnetron sputtering (HiPIMS) power supply or an Advanced Energy Pinnacle Plus pulsed DC power supply. The HiPIMS power supply creates a characteristic peak current which is up to two orders of magnitude larger than that of the pulsed DC. High current densities at the target allow for a dense plasma with the possibility that 30 – 100 % of sputtered atoms will become ionised [5]. Variable parameters during the experiment included the N<sub>2</sub> partial pressure, the current supplied by the pulsed DC supply and the average current supplied by the HiPIMS power supply.

## EXPERIMENTAL

Thin film samples were deposited onto Si (100) substrates in a mixture of both Kr and N<sub>2</sub> sputter gasses. The base pressure of the deposition chamber was  $\sim 5 \times 10^{-10}$  mbar and the pressure was maintained at  $7 \times 10^{-3}$  mbar during deposition. Each substrate was first prepared by cleaning in ultrasonic baths of acetone then IPA and finally rinsed in deionised water. All sample substrates were set to continuously rotate at 4 rpm during deposition at 500 °C.

NbTiN films were deposited by dual magnetron sputtering. One magnetron, fitted with a Nb target, was powered by the HiPIMS power supply; whilst a second magnetron, fitted with a Ti target, was powered by the pulsed DC supply. The HiPIMS power supply was set to pulse with a repetition rate of 1000 Hz with 50  $\mu\text{s}$  pulse length whilst the pulsed DC supply was set to pulse at 350 kHz with a 50 % duty cycle. Films were first deposited with a constant current of 590 mA supplied to the Nb cathode

and 960 mA supplied to the Ti cathode whilst varying the N<sub>2</sub> partial pressure between 11 and 21 %. Next the Ti current was varied between 940 and 1020 mA with constant Nb current of 590 mA and 20 % N<sub>2</sub> partial pressure. Finally, the Nb current was varied between 510 and 590 mA with constant Ti current of 960 mA and constant N<sub>2</sub> partial pressure of 20 %.

Morphological characterisation was performed on a selection of films by SEM, XRD and RBS. A four-point probe was used to determine the superconducting transition temperature ( $T_C$ ) followed by a calculation of the normal state DC resistivity.

### VARYING NITROGEN PARTIAL PRESSURE

Samples showed no superconductivity when deposited with an N<sub>2</sub> partial pressure of 11 %. Samples were superconducting at a partial pressure of 14 % N<sub>2</sub> upwards to 21 % and the maximum  $T_C$  of  $16.67 \pm 0.01$  K occurred at 20 % (Fig 1). All  $T_C$  measurements were taken as the onset of the superconducting state where the resistance of the sample was at approximately 90 % of the normal conducting state [6]. The shape of the  $T_C$  curve displays a plateau where  $T_C$  did not vary by more than 0.3 K between 17 and 21 % N<sub>2</sub>.

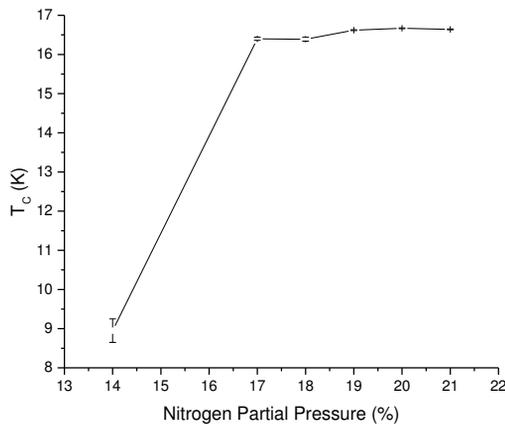


Figure 1:  $T_C$  of NbTiN thin films for varying N<sub>2</sub> partial pressure.

Table 1: Elemental Composition Of NbTiN Thin Films Deposited In N<sub>2</sub> Partial Pressures (N<sub>2</sub> P%)

N <sub>2</sub> P%	$T_C$ (K)	Nb (at.%)	Ti (at.%)	N (at.%)
20	16.67	37.7	16.2	46.1
18	16.50	41.7	16.2	42.4

A selection of the samples deposited with varying N<sub>2</sub> partial pressure were analysed by SEM, XRD and RBS. The SEM images showed that the films had a thickness of 350 nm and was expected to be consistent across all films. The XRD results indicate that the NbTiN films with the highest  $T_C$  have grown in the (111) orientation (Fig. 2). RBS measurements were only performed on two samples and are detailed in Table 1. An RBS measurement of the

sample deposited with 20 % N<sub>2</sub> partial pressure, a Nb cathode current of 590 mA and Ti cathode current of 960 mA is shown in Fig. 3. Assuming a perfect stoichiometry corresponding to atomic percentages of 25% Nb, 25% Ti and 50% N we concluded our films were deficient in both Ti and N whilst containing too much Nb.

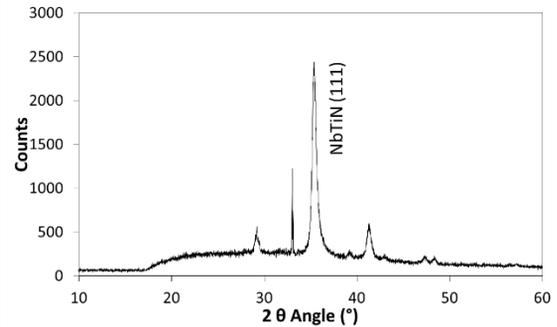


Figure 2: NbTiN thin film with  $T_C$  of 16.67 K. XRD shows the fcc phase with preferred growth orientation (111).

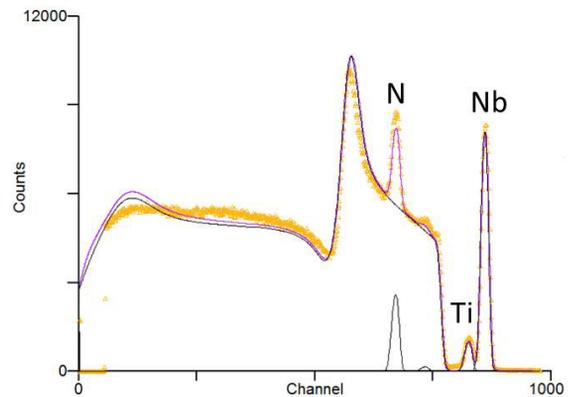


Figure 3: RBS data showing the elemental ratio of Nb, Ti and N in an NbTiN thin film.

The resistance versus temperature curves suggest that the normal state DC resistance of the NbTiN films is inversely proportional to the N content. The resistance measured across the four-point probe when passing a current of 0.1mA reduced with increasing N<sub>2</sub> partial pressure from a maximum of 0.5 mΩ at 11 % down to 0.016 mΩ at 21 % N<sub>2</sub> (Fig. 4).

### VARYING CATHODE CURRENT

The next phase of the study varied Nb and Ti cathode currents with the intention of achieving perfect stoichiometry and increasing  $T_C$ .  $T_C$  of films with either varying Ti or Nb cathode current are shown in Fig. 5. The results indicate that the  $T_C$  is largest for the Ti current at 960 mA with the sample displaying superconductivity at  $16.67 \pm 0.01$  K.  $T_C$  drops sharply at a Ti cathode current of 980 mA and above with the smallest measured value of  $8.2 \pm 0.01$  K occurring at 1020 mA.  $T_C$  of films with varying Nb cathode current exhibit a maximum  $T_C$  of  $16.70 \pm 0.01$  K at 530 mA. Other Nb cathode currents which were tested do not differ greatly.

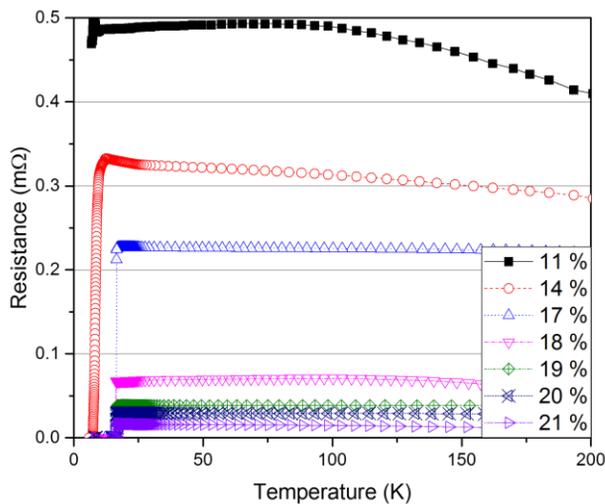


Figure 4: The resistance versus temperature curve for NbTiN films deposited with varying  $N_2$  partial pressures.

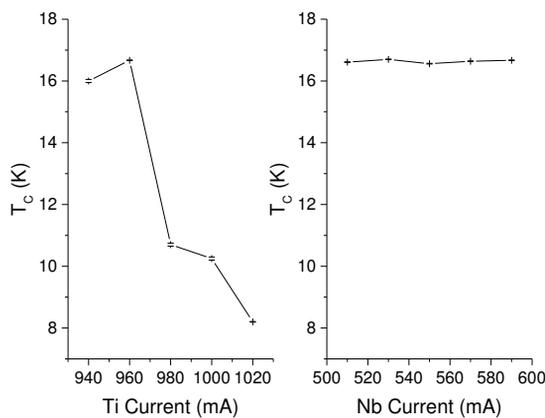


Figure 5:  $T_c$  of NbTiN thin films with either varying Ti or Nb cathode current.

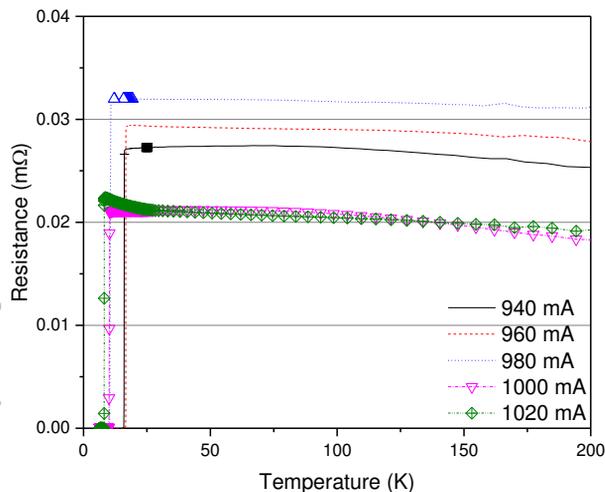


Figure 6: The resistance versus temperature curve for NbTiN films deposited with varying Ti cathode current.

The resistance versus temperature curves for the samples deposited with either varying Ti or Nb cathode currents are shown in Figs. 6 and 7 respectively. The range of measured DC resistances in the normal conducting state varied

between 0.013 and 0.032  $m\Omega$  for an applied current of 0.1 mA. Therefore, changing either the Ti or Nb content did not affect the measured DC resistance to the same extent as did changing the  $N_2$  partial pressure over the range of deposition parameters which were tested.

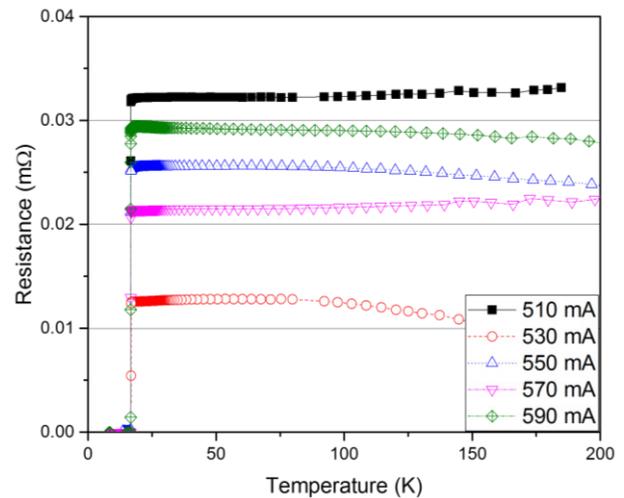


Figure 7: The resistance versus temperature curve for NbTiN films deposited with varying Nb cathode current.

## CONCLUSIONS

NbTiN films have been deposited with  $T_c$  up to  $16.70 \pm 0.01$  K. These films exhibit an approximately constant resistance with temperature and have sharp superconducting transitions. The sharp transition is assumed to be due to a uniform superconducting lattice made up of the (111) growth orientation. It is hoped that such films will perform well if tested for either Q factor at 4.2 K or for use in SIS multilayer application.

It was hoped that the  $T_c$  could be tuned to a value which was closer to the maximum value in the literature of 18 K, however this was not the case. The RBS data shown in Table 1 indicates that the stoichiometric composition was deficient in Ti and N with an excess of Nb however attempts to alter the atomic percentages of either Nb or Ti by varying their respective cathode current did not result in an appreciable increase in  $T_c$  with maximum values of between 16.5 and 16.7 K occurring for a range of different deposition parameters.

## REFERENCES

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