

GROWTH AND CHARACTERIZATION OF MULTILAYER NbTiN FILMS*

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Abstract

Theoretical interest has stimulated efforts to grow and characterize thin multi-layer superconductor/insulator/superconductor (SIS) structures for their potential capability of supporting otherwise inaccessible surface magnetic fields in SRF cavities. The technological challenges include realization of high quality superconductors with sharp, clean, transition to high quality dielectric materials and back to superconductor, with careful thickness control of each layer. Choosing NbTiN as the first candidate material, we have developed the tools and techniques that produce such SIS film structures and have begun their characterization. Using DC magnetron sputtering, NbTiN and AlN can be deposited with nominal superconducting and dielectric parameters. H_{c1} enhancement is observed for NbTiN layers with a T_c of 16.9 K for a thickness less than 150 nm. The optimization of the thickness of each type of layers to reach optimum SRF performance is underway. This talk describes this work and the RF performance characteristics observed to date.

SIS MULTILAYER APPROACH FOR SRF CAVITIES

Theoretical Proposal

A few years ago, a concept was proposed by A. Gurevich [1–3] which would allow taking advantage of high- T_c superconductors without being penalized by their lower H_{c1} . The idea is to coat superconducting radio-frequency (SRF) cavities with alternating superconducting and insulating layers (SIS structures) with a thickness d smaller than the penetration depth λ (Fig. 1). If the superconducting film is deposited with a thickness $d \ll \lambda$, the Meissner state can be retained at a magnetic field much higher than the bulk H_{c1} . The strong increase of H_{c1} in films allows utilization of RF fields higher than the critical field H_c of Nb but lower than those at which the flux penetration at grain boundaries may create a problem. The thin higher- T_c layers provide magnetic screening of the bulk superconducting cavity preventing vortex penetration. The BCS resistance is also strongly reduced because the superconducting materials used have higher gap Δ (Nb₃Sn, NbTiN ...) than Nb. With such structures, Q-values at 4.2 K could be increased two orders of magnitude above Nb values.

If a 50 nm Nb₃Sn layer is coated on a bulk Nb cavity with an insulating interlayer and if the Nb cavity can sustain fields up to 150 mT, this structure could potentially sustain

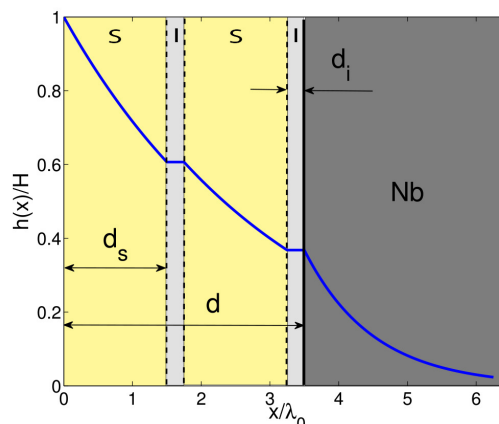


Figure 1: SIS multilayer concept with the magnetic field distribution adapted from [2].

external magnetic fields of about 320 mT and therefore reach accelerating gradients without precedent.

Choice of Materials

Superconductor Although A15 compounds such as Nb₃Sn have a higher T_c and higher superheating field, the Nb B1-compounds are less sensitive to radiation damage and crystalline disorder. B1-compounds have a NaCl structure where metallic atoms form a face centered cubic (fcc) lattice and non-metallic atoms occupy all the octahedral interstices. These compounds are characterized by the fact that they always have a certain amount of vacancies, usually distributed randomly throughout the lattice [4]. The superconducting properties of B1 compounds are very sensitive to deviation from stoichiometric composition. The phase diagram of the binary system Nb-N up to N/Nb=1 includes many different phases, characterized by different T_c . The B1-NbN superconducting phase of interest (cubic δ -phase, $a=4.388$ Å) is only thermodynamically metastable at room temperature. T_c is very sensitive to the nitrogen (N) stoichiometry and NbN suffers from a high resistivity due to the presence of both metallic and gaseous vacancies randomly distributed in both sub-lattices, in amount of 1.3% respectively. The equi-atomic composition is Nb_{0.987}N_{0.987} [4]. The ternary nitride NbTiN is the B1-compound with the highest critical temperature, 17.3 K. It presents all the advantages of NbN and exhibits increased metallic electrical conduction properties with higher titanium (Ti) percentage [5]. Ti is a good nitrogen getter, so the higher the Ti composition, the lower the number of vacancies. In contrast with NbN, the B1-TiN phase is stable at room temperature ($T_c=5$ K, $a=4.24$ Å). The two nitride phases are completely miscible resulting in a superconducting ternary NbTiN cubic phase which re-

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mains thermodynamically stable at room temperature. T_c is slightly higher for NbTiN but as for NbN, N stoichiometry is critical to obtaining the right superconducting phase.

Insulator The chosen insulator is AlN. This material is extensively used for its dielectric and piezo-electric properties for multilayer terra-hertz (THz) mixers and surface acoustic wave sensors. It can be grown with a wurtzite (hexagonal close-packed, $a=3.11 \text{ \AA}$, $c=4.98 \text{ \AA}$) or sphalerite (B1 cubic, $a=4.08 \text{ \AA}$) structure. AlN has been found to enhance the properties (T_c) of NbN and NbTiN, in particular for very thin films [6]. Its thermal conductivity (3.19 W/cm.K at 300 K) is comparable to Cu (4.01 W/cm.K).

Experimental Method

This study uses an ultra-high vacuum (UHV) multi-technique deposition system tailored to in-situ multilayer depositions and described elsewhere [7]. Multiple sample holders are available on the main chamber to allow the simultaneous deposition of witness samples to probe the quality and properties of the individual layers and allow the variation of deposition parameters during the same deposition run. All sample stages can be heated up to 800 °C and are equipped with shutters to allow the deposition in the same run of multiple sets of samples with different parameters, ensuring directly comparable environmental conditions.

The films produced are then analysed with different techniques. The crystallographic characteristics of the layers are probed via by $\theta - 2\theta$ x-ray diffraction with Cu $K\alpha$ radiation. T_c measurements are conducted via the four-point probe method on a multi-sample measurement setup using calibrated CERNOX thermometer s (sensitivity: 50 mK) [8]. RF measurements are conducted on 50 mm disk samples with a 7.5 GHz TE₀₁₁ sapphire loaded cavity (SIC) [9].

NbTiN FILMS

As a first approach, NbTiN films are grown on various substrates (Nb, MgO (100), AlN ceramics) and at various temperatures by DC reactive sputtering with an alloyed NbTi target. Bulk Nb and Nb film substrates represent the intended substrates for SIS structures in SRF applications. MgO (100), a crystalline and very smooth surface, presents ideal conditions for the film growth, whereas AlN ceramic with its distorted nanocrystalline surface is a worst-case scenario. Good quality NbTiN are typically produced both at 450 and 600 °C. Bulk-like NbTiN films, with a thickness higher than 1 μm , are reliably coated with a resulting T_c close to the bulk value of 17.3 K. Thin NbTiN films with a T_c higher than 16.6 K are produced for thicknesses higher than 50 nm.

The morphology of NbTiN films deposited on MgO at various thicknesses is illustrated in Fig. 2 with 5 $\mu\text{m} \times 5 \mu\text{m}$ AFM (atomic force microscopy) scans. Very thin films retain the roughness of the substrate. As the film thickness increases, its roughness increases as expected due to grain coarsening.

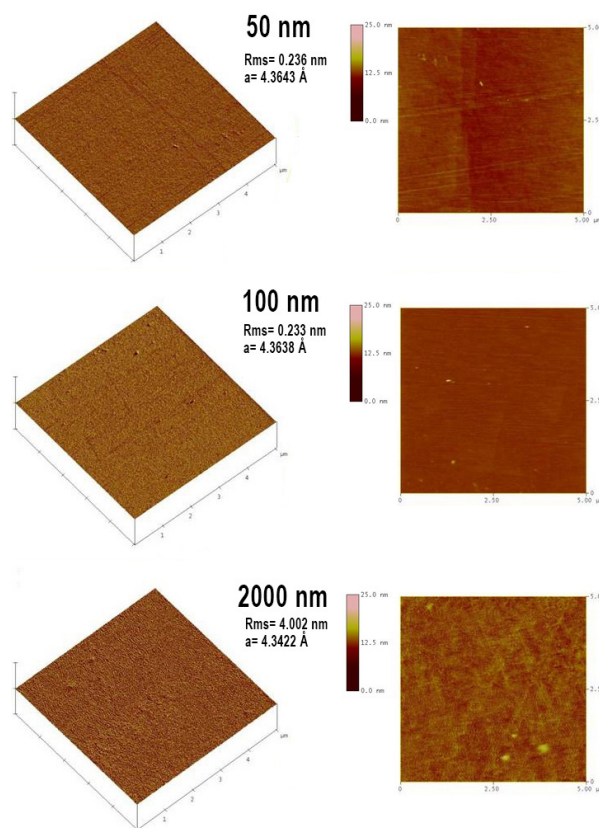


Figure 2: AFM imaging of NbTiN films deposited on MgO at 450 °C at various thicknesses.

AlN FILMS

The dielectric AlN layers are produced with N reactive sputtering of an elemental Al (99.999 %) target. The produced AlN films are fully transparent. They exhibit the B1-cubic structure when deposited at 600 °C and are polycrystalline when deposited at 450 °C. Thin AlN films deposited at 450 °C on MgO (100) were measured by spectroscopic ellipsometry. The thickness and roughness of the AlN layers were first measured by x-ray reflectivity (XRR) using a standard four-circle diffractometer with Cu $K\alpha$ radiation (respectively 30.41 nm and 28.82 nm in thickness and 0.62 nm and 0.45 nm in average roughness). The optical constants (i.e. complex-valued functions) were measured ex-situ using a J. A. Woollam variable angle spectroscopic ellipsometer (VASE) with a rotating analyzer and auto retarder, where the WVASE32 program was used to model and fit the data [10].

The fitting of the ellipsometry data represented in Fig. 3 uses a Cauchy layer model with Urbach absorption [11] to model the optical constants of the AlN film, along with bulk MgO optical constants, and a roughness layer with 50 % void fraction with ambient. The thickness and roughness of the AlN layers were taken from XRR data mentioned above. The refractive index measured for these polycrystalline films, reported in Table 1, are in the reasonable range of 1.98 -

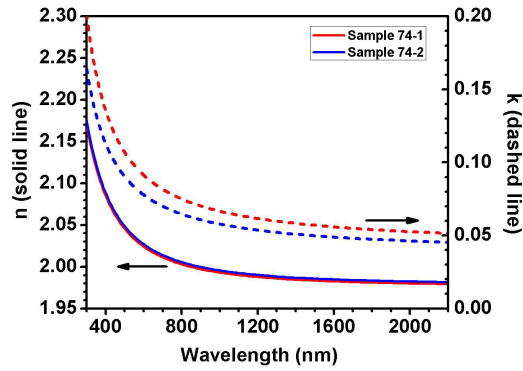


Figure 3: Ellipsometry measurement of two AlN films deposited on MgO (100) at 450 °C.

2.15, corresponding to those of polycrystalline AlN films previously reported [12].

Table 1: Measured Refractive Index at 632.8 nm for two AlN Films

Film nature	Thickness [nm]	Refractive index
Polycrystalline	30.4	2.02
Polycrystalline	28.8	2.02

NbTiN/AlN STRUCTURES

SIS structures based on NbTiN and AlN have been coated on various Nb surfaces: bulk, Nb/Cu and Nb/Al₂O₃ along with MgO and AlN ceramic substrates. The films reported here were deposited at 450 °C in-situ on bulk Nb and Nb/Cu substrates after a 24 hour-bake at 450 °C. The samples were then annealed at 450 °C for 4 hours.

Structure

Although deposition at 600 °C typically yields the best results in terms of structure and properties (T_c , refraction index...), the successive deposition of AlN and NbTiN at this temperature induces Al diffusion into the underlying Nb surface and into the NbTiN film [13].

Figure 4 represents TEM micrographs with associated individual and combined EDS maps for Nb, Ti and Al for the FIB cross-section of a NbTiN/AlN bilayer deposited on bulk Nb at 450 °C. When the coating temperature is lowered to 450 °C, the NbTiN and AlN layers still display good polycrystalline structures. There is no Al diffusion as revealed by the EDS maps.

H_{c1} Enhancement

H_{c1} measurements by SQUID magnetometry as described in [14] were performed on 150 nm thick NbTiN films deposited simultaneously on MgO and AlN ceramic and are reported in Table 2. Due to the current configuration of the system, these samples displayed inhomogeneous properties

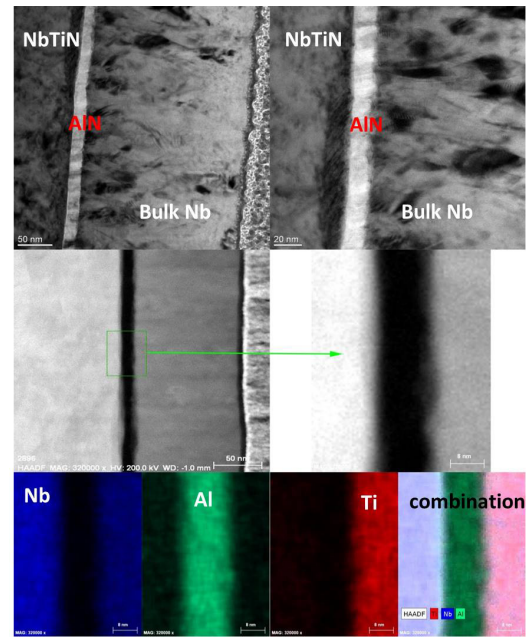


Figure 4: TEM imaging for NbTiN/AlN/Nb with associated individual and combined EDS maps (bottom) for Nb, Al and Ti.

as their stoichiometry varied according to their position on the sample holder.

Table 2: Measured H_{c1} Values for Different NbTiN Films

Substrate	Thickness [nm]	T_c [K]	H_{c1} [mT]
MgO	2000	17.25	30
AlN/MgO	148	16.66	200
AlN/AlN ceramic	145	14.84	135

The sample deposited on MgO with a T_c corresponding to the δ -phase demonstrated a field enhancement to 200 mT from 30 mT as measured for a 2 μ m bulk-like NbTiN/MgO. The sample deposited on the rough AlN ceramic, despite a T_c representative of the γ -phase, still shows a field enhancement to 135 mT. The thickness of these films is about 150 nm which is still very close to the NbTiN penetration depth. Thickness studies are under way to determine the optimum for H_{c1} enhancement.

RF Measurement

Figure 5 shows the surface resistance measurement of an NbTiN/AlN structure coated on a thick Nb film. The performance of a large grain Nb reference sample and the initial performance of the ECR Nb/Cu film are also represented for comparison. The film used as a substrate was coated ex-situ by energetic condensation [15]. The SI structure was deposited at 450 °C in-situ after a 24 hour-bake at 450 °C. The sample was then annealed at 450 °C for 4 hours. The multilayer structure shows an increase of surface resistance below 4K most likely due to the reduction and diffusion of

the Nb film oxide layer into the film bulk. However, above 4 K, the surface resistance of the SIS structure is lower than for the initial ECR Nb film and than for bulk Nb.

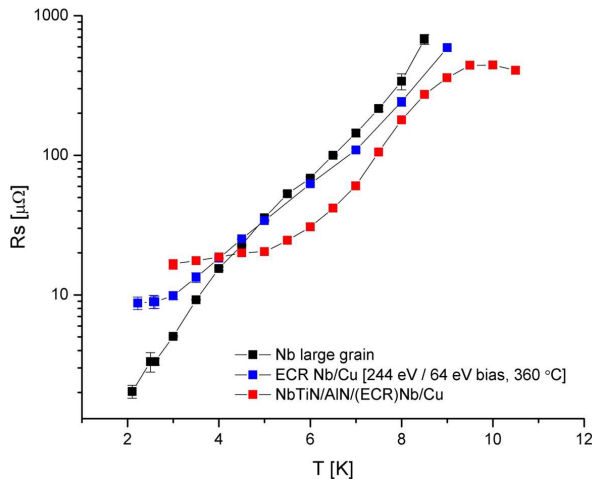


Figure 5: RF measurement in the SIC setup of a NbTiN/AlN/Nb/Cu structure compared with the original Nb/Cu film and large grain bulk Nb.

Once the optimum thicknesses for NbTiN and AlN will be defined, a series of SIS multilayers based on NbTiN will be deposited and RF tested on previously characterized QPR (Quadrupole Resonator) samples [16, 17].

CONCLUSION

Good quality NbTiN and AlN films can be deposited by DC magnetron sputtering. Bulk, i.e. thicker than 1 micron, NbTiN films are readily produced with a T_c of 17.25 K, close to the bulk value. AlN dielectric films of various thicknesses are produced with good dielectric properties. H_{c1} enhancement has been observed by SQUID magnetometry for some NbTiN films. Further studies of the H_{c1} enhancement of thin NbTiN layers are necessary both on MgO substrates and on thin AlN layer deposited on MgO. Good quality SI NbTiN/AlN layers have been produced on samples with a T_c for the NbTiN layers between 16.6 and 16.9 K. It is shown that, if the dielectric can be grown as an adequate template for the superconducting NbTiN film growth and the growth temperature is high enough to provide enough adatom mobility, the substrate macro-roughness is not necessarily detrimental to the T_c of the superconducting film. Further improvement of the interfaces is however needed. Energetic deposition via HiPIMS (High Power Impulse Magnetron Sputtering) deposition may be useful in improving the structure of both NbTiN and AlN and lowering further the deposition temperature.

The RF characterization of a NbTiN/AlN structure coated on a Nb/Cu surface shows indications that SIS coated Nb surfaces have a promise to delay flux penetration and lower RF losses for SRF cavities. A systematic study of the effect of the NbTiN film thickness in the S-I-S structure over the RF performance enhancement is under way. RF characterization

of the optimized SIS structures based on NbTiN will be conducted on bulk Nb QPR samples.

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