SUPERCONDUCTING NBN-BASED MULTILAYER AND NBTIN THIN FILMS FOR THE ENHANCEMENT OF SRF ACCELERATOR CAVITIES

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Abstract

Current superconducting radio frequency (SRF) technology, used in various particle accelerator facilities across the world, is reliant upon utilizing bulk superconducting cavities, usually made of bulk Nb. Due to technological advancements in the processing of bulk Nb cavities, the facilities have reached accelerating fields very close to material-dependent limits, ~50 MV/m for bulk Nb. One possible solution to improve upon this fundamental limitation was proposed a few years ago by A. Gurevich and it consists of the deposition of alternating thin layers of superconducting and insulating materials on the interior surface of the cavities. Some candidate superconducting materials proposed for this multilayer scheme are NbN, NbTiN and Nb3Sn. Here we present our recent results on NbN-based trilayers in coupon samples in order to further advance the multilayer approach. Since NbTiN has higher lower critical field (Hc1) and higher critical temperature (Tc) than Nb and increased conductivity compared to NbN, it is a more promising candidate material for this new method. Thus, we also present experimental results correlating film microstructure with superconducting properties on NbTiN thin film coupon samples while also comparing films grown with targets of different stoichiometries. It is worth mentioning that we have achieved thin films with bulk-like lattice parameters and transition temperatures while also achieving Hc1 values larger than bulk for films thinner than their London penetration depths.

INTRODUCTION

Linear particle accelerator facilities like the Thomas Jefferson National Accelerator Facility (JLab) in Newport News, VA rely on superconducting radio frequency (SRF) cavities made of bulk niobium. These SRF cavities benefit from their ability to be continuously operated with low power dissipation, have lower microwave surface resistance and achieve higher maximum accelerating fields than conventional copper cavities. Recent advances in Nb cavity technology have increased the maximum breakdown field (~50MV/m) approaching the theoretical limit given by the thermodynamic critical field Hc(0) = 200mT, at which the RF field induces the depairing current density at the surface of the cavity. Thus, further enhancements require a new approach.

Due to the fact that the SRF phenomenon is active a short distance into the superconducting material close to the surface, one can feasibly exploit thin films with the goal of improving cavity performance. Thin films offer several advantages over current bulk materials, since their microstructure can be tailored and have the ability to be employed in a multitude of material combinations that can result in cavities that could surpass current individual bulk properties. Therefore, in order to push the accelerating fields of SRF cavities beyond the current material limitations it is possible to utilize thin film deposition on the interior of cavities using various new materials and combinations, as well as improved deposition conditions compared to previous attempts [1].

A possible model to achieve field enhancement utilizes alternating layers of superconducting-insulating-superconducting (SIS) thin films deposited on the interior of cavities to shield the innermost surface from magnetic field vortex penetration thus enabling the possibility to even double the maximum field gradient achievable [2]. In such model, it is predicted that shielding of the inner surface is possible using layered superconducting materials, with higher critical temperatures (Tc) and lower critical field (Hc1) values than bulk Nb, such that magnetic field vortex penetration and propagation is reduced or even suppressed resulting in enhanced cavity quality factor, Q. Here, the insulating layers (I) are needed between the superconducting layers (S) to (i) suppress the Josephson currents between the screening superconducting layers so that magnetic field (H) at the interior surface of the Nb cavity is smaller than the bulk Hc1 ≅ 150 mT of clean Nb and (ii) to impede propagation of magnetic field vortices that might form to decrease losses resulting from such vortex propagation near the interior surface of the cavity. Proposed candidate superconducting materials for this system include: NbN, NbTiN, Nb_3Sn, MgB_2 and possibly other more exotic materials. The ultimate goal of the SIS structures is to reduce or even eliminate strong dissipation of vortices in a thin film geometry for which no thermodynamically stable position of a single vortex exists up to $H \approx H_{sh}$. An important feature to take into account is that it is well known that superconducting films thinner than their London penetration depths exhibit enhanced Hc1 values in the parallel field geometry compared to bulk, but – concerning screening ability, this enhanced Hc1 would be relevant primarily for the special metastable state in which the trapped field in the insulating layer is equal to the applied field H; more importantly a thermodynamically stable vortex situation with strong RF

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vortex dissipation does exist for films thicker than the London penetration which would compromise the proposed screening.

This, this enables the ability to "tailor" a lower critical field at the inner cavity surface by varying the layers constitutive materials and their thicknesses thus allowing the optimization of overall cavity performance. Therefore, the experimental implementation of the SIS model will test the shielding of the inner surface theoretical prediction, which may result in cavities with much larger maximum accelerating field gradients and improved cavity quality factor.

However, before this model can be employed we must first understand the relationship between the thin film geometry, the material microstructure, the ultimate surface morphology and the resulting superconducting properties of the materials considered in order to control the process for greatest benefit. Also, various possible deposition techniques available must be explored in order to determine the optimal method that yields the best films and yet still capable of successfully coating the interior of cavities with high quality films. Work is ongoing to successfully test the SIS model in coupon samples, and some promising results have been reported recently on test cavities [3]. However, despite this modest success, the latest experimental results on coupon samples suffer from increased surface roughness and microstructure defects whose effects are magnified in the thin film geometry. Because SRF is a surface phenomenon penetrating about one micron into the active material, the decreased surface quality has a large impact on the resulting cavity performance. Even current bulk Nb cavities must undergo extensive surface treatments to achieve the ultimate cavity quality and SRF performance.

Since thin films have one dimension that is highly constrained, achieving films with bulk-like properties is not a trivial exercise and is highly dependent on the material in question, underlying substrate, deposition parameters and the deposition method employed.

In this paper, we present recent results on NbN-based trilayers as well as our studies correlating the microstructure of NbTiN thin films with their resulting DC superconducting properties. We also compare the results of samples grown using DC magnetron reactive sputtering using a 70/30 (%wt) NbTi target to samples grown using an 80/20 (%wt) NbTi target.

EXPERIMENT

All the films and multilayered studied were prepared on single crystal MgO(100) substrates using DC Magnetron Reactive Sputtering in a high vacuum deposition system with a base pressure in the 10-8 Torr range and a fixed target to substrate distance of 13.5 cm. Prior to film growth the substrates were cleaned in an ultrasonic bath of alternating acetone and methanol and annealed at 600 C for one hour under vacuum. After growth, the structural properties of the samples (lattice parameter, grain size…etc.) were determined via X-Ray diffraction (XRD) using a Panalytical 4-circles diffractometer, while the DC superconducting properties (Tc, Hc1…etc.) were measured using a Quantum Design MPMS SQUID (superconducting quantum interference device) magnetometer.

NbN-Based Trilayers

Microstructure In order to truly assess the viability of the multilayer theory for SRF cavity applications, we must expand beyond simple single layer coupon samples. The next logical step in this process is to create multilayer samples. With this need in mind, we first set out to create adequate single layer NbN films, since this material is the simplest to transition into the multi-layer scheme. While our group had successfully done this in the past [4], we first set up to reproduce our earlier NbN results in order to grow successful NbN-based multilayer films. To this end, we first calibrated our system for NbN deposition, and once films of adequate characteristics were demonstrated, we undertook the growth of Nb/MgO/NbN trilayered films. The trilayers were grown by reactive and DCMS on 2" MgO(001) substrates and consisted of: a very thin (<5nm) MgO seed layer over the crystalline substrate in order to improve surface morphology, based on our previous work [5], a base 250nm layer of Nb, a 15nm MgO layer, and finally an 85nm NbN layer. The choice of thickness for the Nb layer was done to preserve adequate surface roughness for subsequent growth, while the choice of NbN thickness was below the London penetration depth and based on our previous work.

When undergoing this growth, of utmost concern is (1) quality of the interfaces, and (2) achieving the correct phase correlated to optimal superconducting properties. This concern arises from the fact that, while it is possible to achieve good epitaxy between the NbN and a crystalline MgO(001) substrate, strain can affect the

Table 1: Properties of NbTiN Samples prepared using Different Stoichiometry Targets

Sample	Substrate	Target Stoichiometry (Nb/Ti %wt)	Thickness (nm)	Lattice Parameter (Å)	Grain Size (nm)	Hc1 (Oe)
a	MgO(001)	70/30	50	4.318	28.19	140
b	MgO(001)	70/30	150	4.324	39.34	320
c	MgO(001)	70/30	80	4.327	34.52	140
d	MgO(001)	70/30	120	4.305	34.81	300
e	MgO(001)	70/30	80	4.324	30.98	600
f	MgO(001)	80/20	145	4.351	20.85	2000
g	AlN(cer)	80/20	145	4.346	19.81	1350

ISBN 978-3-95450-178-6

surface morphology thus affecting subsequent epitaxial growth through multiple interlayers. Figure 1 shows XRD scans for a the NbN-based trilayers, where we observe that the appropriate delta NbN(002) phase with absence of any other NbN phases was achieved. The corresponding lattice parameters for the trilayer films were around 4.38Å, i.e. within bulk value. Also, the grain sizes varied from 26 - 27nm. We track this characteristic of film microstructure, since it is known that grain boundaries are responsible for degraded superconducting thin films compared to bulk.

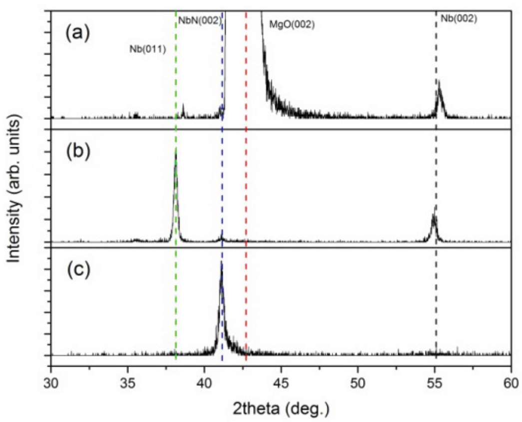

Figure 1: XRD scans for NbN Trilayer samples a) 092314 b) 100114 and c) 100714. All exhibit only the NbN(002) phase showing good epitaxy with top NbN layer and underlying substrate. Note, all scans were optimized on the NbN(002) reflection and varying misalignments of the layers leads to differing intensities of reflections between samples. Highest performing film, 092314, exhibited predominantly Nb(002) texturing, confirming previous results found by group [4].

We note that the XRD scan shows that while one of the samples exhibit only the Nb(002) phase for the Nb layer, the other sample exhibited Nb(011) and Nb(002) texturing. This is important since it has been shown previously [6] that Nb samples with (011) texturing exhibit poorer superconducting performance than those with only (002) texturing due to grain boundaries.

Superconducting Properties All of the trilayers exhibited good DC superconducting properties with Tc values around 13K, again reaffirming the XRD result of the adequate NbN phase of NbN. The samples also exhibited large enhancements in the Hc1 values compared to bulk NbN and, as shown in Figure 2, the best sample had an approximate Hc1 value of over 2000 Oe, over 10 times larger than bulk NbN value and improved over our earlier results. These results showcase the great promise of the multilayer approach while underscoring the importance of adequate control of thin film microstructure and surface parameters.

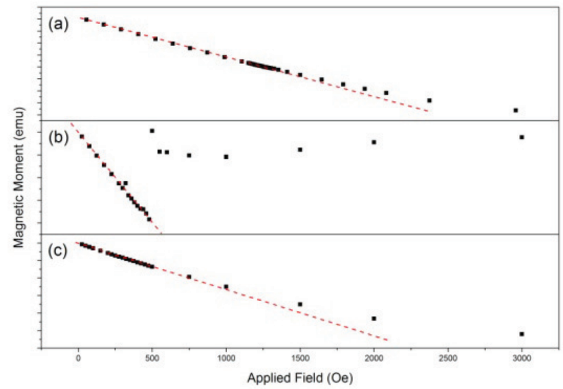

Figure 2: SQUID magnetometry data for Hc1 determination of NbN-based trilayer sample a) 092314 with an Hc1 of ~2100 Oe b) 100714 with an Hc1 of ~480 Oe and c) 100114 with an Hc1 of ~ 750 Oe. The data was acquired using the Bohmer method referenced and Hc1 was determined as the point before the deviation from the Meissner slope.

NbTiN Thin Films

NbTiN films were grown using a single two inch 70/30 (%wt) NbTi target in an Ar and N_2 gas mixture. The growth parameters of the NbTiN films were optimized by varying the total pressure, mass flow rate, partial pressure of N_2, and substrate temperature while comparing the microstructure of the resulting films aiming at the most bulk-like properties. To this end, films were grown using: ~2.5% N_2 partial pressure, a total pressure of ~4.1 mTorr, and a substrate temperature of ~600C, since it has been shown that higher temperature growth leads to films with higher Tc values [7]. Additional films were grown in a similar setup using DCMS on AlN and MgO substrates with an 80/20 (%wt) NbTi target, a fixed target to substrate distance of 80mm and with a thin AlN interlayer grown prior to NbTiN. All the films used in these experiments varied in thickness from 50nm to 2um.

Microstructure We note that NbTiN has many possible phases that may have significant effect on the resulting properties of the films if present. During deposition, the phases present in the films can be tailored by altering parameters such as pressure. In this study, it is of utmost importance to create films in the optimal superconducting phase of NbTiN, (002). Thus, the parameter of particular interest was the nitrogen partial pressure, and, in order to optimize the nitrogen partial pressure, a systematic study was carried out changing only the N_2 partial pressure while holding other variables constant during growth. The films were then analyzed using XRD to determine the resulting lattice parameters, grain size, and mosaicity.

From the XRD scans in Figure 3, our samples exhibit MgO(002) reflections from the substrate and NbTiN (002) reflections showing clear epitaxial growth of the NbTiN on the substrate. No other phases of NbTiN are observable in the range from 20 – 80 deg for our samples leading to the conclusion that our samples exhibit only the optimal superconducting phase. The grain size of our samples were obtained from FWHM of the NbTiN(002) XRD peak and using the Scherrer equation. The grain sizes varied between 28 and 40nm, while the film with the highest Hc1 value had a grain size of approximately 31nm.

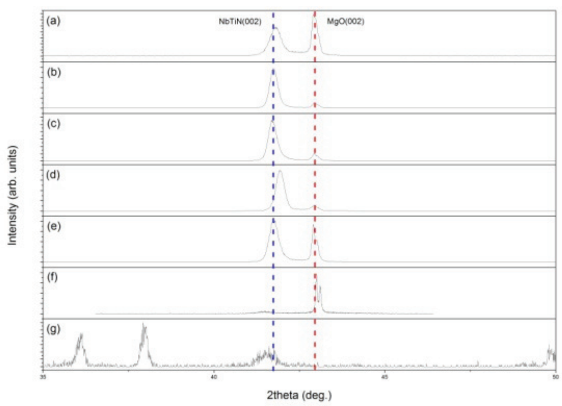

Figure 3: XRD scans from all of the NbTiN samples measured for this study. The letter labels correspond to those in Table 1. Samples f and g, made with the 80/20 target, exhibit a clear shift to lower angles corresponding to a larger lattice parameter than those made with the 70/30 composition target. The extra peaks present in scan g result from the ceramic AlN substrate used for that sample.

Adequate stoichiometry to achieve the appropriate superconducting phase in a ternary compound is very important and the film lattice parameter is one of the most important values for assessing such composition. NbTiN is a ternary alloy and hence, the actual composition of a thin film depends on adequate nitrogen partial pressure as well as target composition when using a composite NbTi target. Ultimately, the stoichiometry of a particular film determines its lattice parameter. It has been shown [8] that as you increase the Nb concentration in a NbTiN film, you achieve larger lattice parameters, however at our desired compositions, you also operate on the edge of an unstable phase of the compound. For this study, two targets of differing stoichiometry were used to investigate the dependence of the lattice parameter of the resulting films and correlate it with their superconducting properties. Films grown at W&M utilized a 70/30 (%wt) NbTi target while an 80/20 (%wt) were used in films grown at Jefferson Lab.

The films grown using a 70/30 (%wt) NbTi target resulted in lattice parameters between 4.30 and 4.327 Å with the film with the largest Hc1 value had a lattice parameter of 4.32 Å. While films grown using the 80/20 (%wt) target exhibited somewhat larger lattice parameter, between 4.34 and 4.35 Å with the film with the largest Hc1 value having a lattice parameter of 4.35 Å.

Superconducting Properties When comparing across samples in Table 1, it is seen that the samples grown using the 80/20 target had better DC superconducting properties than the 70/30 counterparts. The 70/30 samples exhibited Tc values between 12 and 14K while the 80/20 samples achieved Tc values around 16K. The Hc1 values in all samples were estimated from SQUID DC measurements carried out following a standard method described in the literature to minimize systematic errors [9].

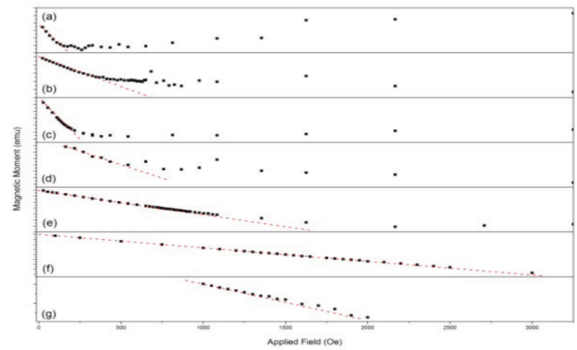

Figure 4: SQUID magnetometry data for NbTiN samples with Hc1 values of a)~140 Oe b)~320 Oe c)~180 Oe d)~500 Oe e)~830 Oe f)~2000 Oe and g)~1350 Oe. Data was obtained and analyzed using same method as mentioned for the NbN-based Trilayers above. All samples exhibited purely the NbTiN(002) and those with larger lattice parameters similarly had higher Hc1 values. The letter labels correspond to those in Table 1.

As can be seen in Figure 4, the highest Hc1 observed in the films grown using the 70/30 target was around 830 Oe while the highest for the 80/20 films was on the order of 2000 Oe. However, it is necessary to note here that all of the Hc1 values listed in Table 1 are necessarily underestimates of the true value due to geometric constraints within the measurement system. It can be shown that, since the Hc1 enhancement is only valid in the parallel DC field geometry, any component of the applied magnetic field perpendicular to the sample surface will necessarily result in a reduction in the observed Hc1 value due to early onset magnetic flux penetration, hence our clarification regarding the underestimate quality of our Hc1 measurements.

CONCLUSION

NbN-based Trilayers grown on 2 inch coupon samples were prepared and the DC superconducting results were quite successful, indicating that shielding beyond bulk Nb is possible. Structure/property correlations indicated that good epitaxy was achieved with concurrent good superconducting properties. Ongoing efforts are in place to establish the SRF performance of such trilayered samples.

NbTiN Thin Film samples were prepared and a systematic study on the thickness dependence of the superconducting properties indicated that it is also possible to achieve successful thin layers with this material. We note that the choice of deposition parameters as well as target composition further affects the superconducting properties of the films, showcasing the

ISBN 978-3-95450-178-6

path to follow for trilayer studies similar to the ones carried out with NbN based tri-layer.

ACKNOWLEDGMENT

We would like to thank: the rest of the Lukaszew Research Group, Charlie Reece and the SRF Institute at Jefferson Lab for their collaboration, the College of William & Mary for support and facilities, and the Defense Threat Reduction Agency for funding this research under grant HDTRA1-10-1-0072.

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