ISBN: 978-3-95450-234-9 ISSN: 2673-5504 doi:10.18429/JACoW-SRF2023-MOPMB083

INVESTIGATION OF THE MULTILAYER SHIELDING EFFECT THROUGH NbTiN-AlN COATED BULK NIOBIUM *

I. H. Senevirathne[1,2,†], D. R. Beverstock[2,3], A-M Valente-Feliciano[2], A. Gurevich[1],
J. R. Delayen[1,2], Center for Accelerator Science, Old Dominion University, Norfolk, VA, USA
[2]Thomas Jefferson National Accelerator Facility, Newport News, VA, USA
[3]Applied Science Department, William & Mary, Williamsburg, VA, USA

Abstract

We report measurements of the dc field onset B_p of magnetic flux penetration through NbTiN-AlN coating on bulk niobium using the Hall probe experimental setup. The measurements of B_p reveal the multilayer shielding effect on bulk niobium under high magnetic fields at cryogenic temperatures. We observed a significant enhancement in B_p for the NbTiN-AlN coated Nb samples as compared to bare Nb samples. The observed dependence of B_p on the coating thickness is consistent with theoretical predictions.

INTRODUCTION

Improving the performance of superconducting radio frequency (SRF) cavities, reducing their high-field Q-slope and increasing the maximum RF breakdown field are key goals of current SRF accelerator R&D. The breakdown field and the high-field Q-slope of Nb cavities is eventually limited by dissipative penetration of superconducting vortices. The high-field performance of Nb cavities can be improved by depositing Superconducting-Insulating-Superconducting (SIS) multilayers [1] which act as barriers for penetration of vortices, allowing the bulk Nb to sustain higher magnetic fields. As a result, SIS structures can increase the accelerating gradients in SRF cavities.

The field of flux penetration, B_p through the SIS structure is an important parameter of merit of the multilayer shielding effect on bulk niobium under high magnetic fields. Recently a dc magnetic Hall probe technique [2] has been developed to measure the penetration field in various materials, including bulk, thin films, and specifically SIS structures. In this work we use this dc Hall probe technique to investigate the effectiveness of NbTiN-AlN coatings on bulk niobium.

SIS MULTILAYER SYSTEM

The SIS multilayer structure consists of alternating thin layers of superconducting and insulating materials deposited on the inner surface of the niobium cavity as shown in Fig. 1.

The SIS structure provides a barrier for penetration of vortices into the bulk superconducting material. The superconducting layers are typically high-transition-temperature (high-T_c) superconductors such as niobium titanium nitride (NbTiN), niobium nitride (NbN) or niobium tin (Nb_3Sn). Thin superconducting overlayers are separated from a bulk superconductor by thin (a few nm thick) insulator layers which suppress tunnelling of Cooper pairs between superconductors.

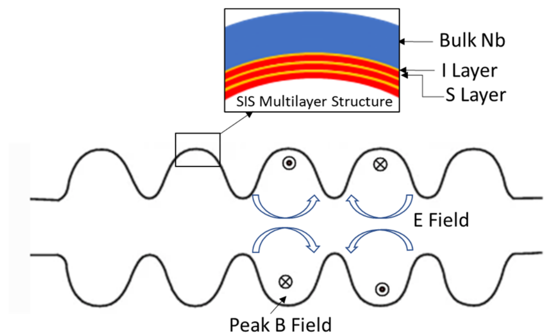

Figure 1: SIS multilayers deposited on the inner surface of the bulk Nb cavity to enhance the peak surface magnetic field and delay the vortex penetration into the cavity wall.

CANDIDATE MATERIALS

The multilayer structures comprising NbTiN and AlN as superconducting and insulating materials are promising candidates for the SIS coating of Nb cavities. Bulk NbTiN has a higher T_c as compared to Nb, and the T_c of NbTiN films can be varied from 8 to 17.3 K by tuning the stoichiometry and the film thickness. In terms of material characteristics, NbTiN offers excellent mechanical properties and robustness. NbTiN has been found to exhibit a particularly low surface resistance at RF frequencies [3], making it a desirable SRF material. Another significant advantage of NbTiN is its compatibility with established fabrication techniques. It can be deposited as a thin film using various deposition methods such as Chemical Vapor Deposition (CVD) like Atomic Layer Deposition (ALD) or Physical Vapor Deposition (PVD) like Direct Current Magnetron Sputtering (DCMS) and High-Power Impulse Magnetron Sputtering (HiPIMS).

Furthermore, AlN is a good insulator with a relatively high thermal conductivity (as compared to Al_2O_3, MgO), which helps transferring the RF heat dissipated in high-T_c overlayers through the Nb cavity wall.

THEORETICAL PREDICTIONS

The magnetic shielding of bulk Nb by SIS structures depends on the thickness of the superconducting overlayers [4, 5]. It was shown that the maximum superheating field $H_{sh}(d)$ of the SIS structure on Nb has a maximum as a function of the overlayer thickness d due to the current

*Work supported by NSF Grants PHY-1734075 and PHY-1416051, and DOE Awards DE-SC0010081 and DE-SC0019399.
† isene001@odu.edu

counterflow induced in the overlayer by Nb substrate. As a result, the maximum superheating field of a SIS structure can exceed H_{sh} of both the bulk Nb and the coating material. Figure 2 shows the contour plot of the superheating field as functions of the thicknesses of S and I layers generated using the equations of Ref. [4]. Here we took the London penetration depth of 240 nm and a coherence length of 3.5 nm for NbTiN. These calculations were done for an ideal surface and S-I interface. However, more realistic defects and surface roughness can reduce the maximum screening field of SIS structures [5].

In what follows we show the Hall probe measurements of the magnetic field at full flux penetration B_p in NbTiN-AlN coated on bulk Nb. We varied the thicknesses of the superconducting layer d, while keeping the insulating layer thickness constant at 10 nm. It turns out that the observed $B_p(d)$ has a maximum similar to that predicted theoretically for the maximum superheating field [4, 5].

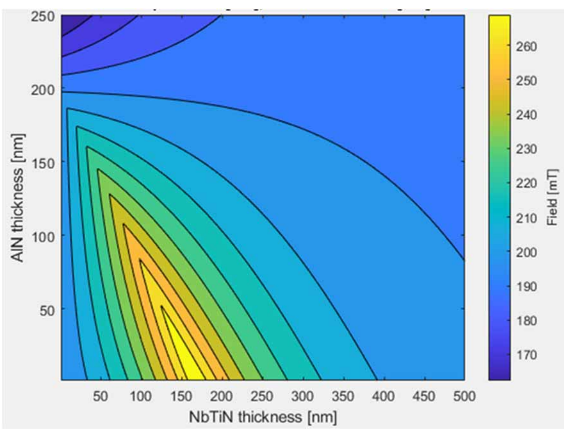

Figure 2: The contour plot of the maximum achievable peak surface field, Bv calculated using equations from Ref [4] for the London penetration depth of 240 nm and a coherence length of 3.5 nm in the NbTiN layer.

SAMPLE FABRICATION

Reactive Direct Current Magnetron Sputtering (reDCMS) was employed to deposit thin layers of NbTiN and AlN onto bulk Nb in an Ultra-High Vacuum (UHV) system with a base pressure of 10^{-10} Torr. The Nb substrates underwent buffered chemical polishing (BCP) to remove a 5 µm layer from the surface. Four Nb substrates, each with a diameter of 50 mm and a thickness of 250 µm, were utilized as substrates, accompanied by witness samples for evaluating the quality and properties of the films. Following a 24-hour bake at 600 °C, the films were deposited onto the bulk Nb at 450 °C and subsequently post-annealed for 4 hours at the same temperature. Table 1 represents the thickness of both layers for the sample set.

Figure 3 shows the crystallographic structures of deposited thin films examined by X-ray diffraction (XRD). High-angle XRD measurements give an information about the crystalline relations between the substrate and the deposited layers.

Table 1: NbTiN and AlN Layer Thicknesses of Coated Samples

Nb Substrate Treatment	Sample #	Layer Thickness (nm)	
		NbTiN	AlN
BCP	1	83	10
	2	166	10
	3	250	10
	4	371	10

The electron backscatter diffraction (EBSD) analysis reveals the multi-crystal nature of the film deposited on the bulk Nb material, as depicted in Fig. 4. The morphological characteristics of these deposited films can be observed in the scanning electron microscope (SEM) images.

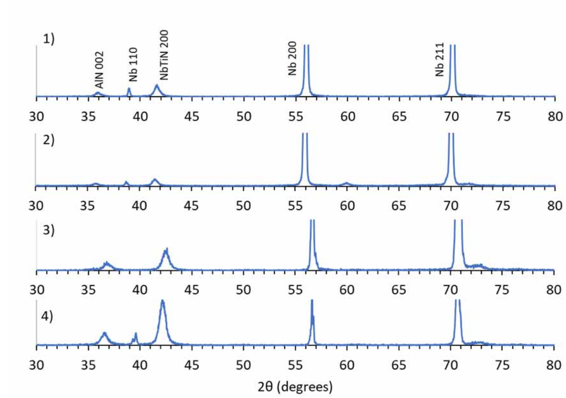

Figure 3: XRD Analysis that shows crystallographic structures of deposited thin films.

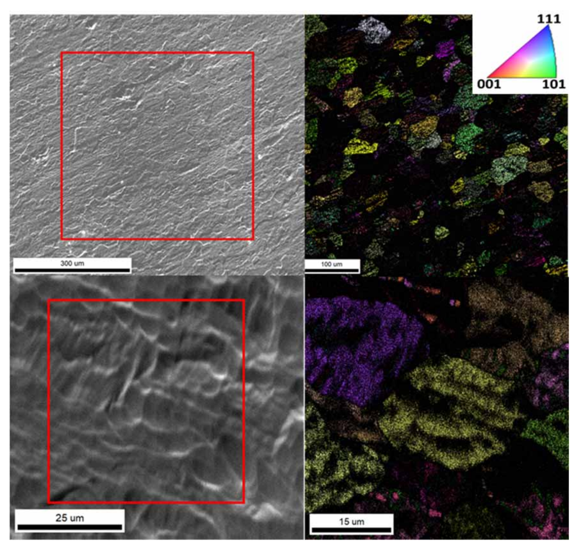

Figure 4: SEM image and relevant Inverse Pole Figure (IPF) map from EBSD showing the polycrystalline nature of the deposited film on bulk Nb (sample #2).

The resistances of the films were measured from 4.5 to 300 K using standard four-point probe method. Here critical temperature (*Tc*) ranged from 12.5 to 16 K as shown in Fig. 5. Low *Tc* values are often observed for very

ISBN: 978-3-95450-234-9 ISSN: 2673-5504 doi:10.18429/JACoW-SRF2023-MOPMB083

thin films due to their high defect density such as vacancies, dislocations, and grain boundaries. When the

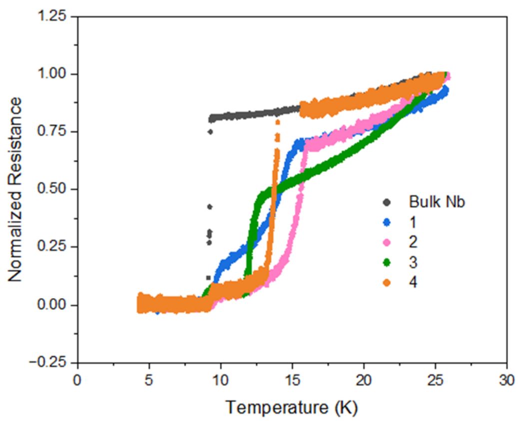

Figure 5: Resistance as a function of temperature measured by the four-point probe method.

same films were deposited on MgO substrates, they exhibited T_c above 16 K.

FIELD PENETRATION MEASURMENTS

The Magnetic Field Penetration (MFP) magnetometer, equipped with a dc Magnet and Hall probes was designed specifically for testing flat superconducting samples, whether in the form of bulk or thin films [6-8]. It serves as an instrument for the measurements of magnetic field penetration in superconducting samples [2]. Its main purpose is to identify the minimum surface magnetic field B_p at which the magnetic flux fully penetrates through the sample. This is a convenient instrument for exploring the shielding effect of multilayer structures [9, 10]. In our MFP experimental setup, a superconducting solenoid is positioned above the sample to apply a dc magnetic field to one surface of the film. This magnet can generate a magnetic field exceeding 500 mT on the surface of the sample. On the opposite side of the sample, three Hall probes are mounted along the sample radius to detect the magnetic field that has penetrated through the film.

Figure 6 illustrates the field penetration at two different temperatures, 4 K and 2 K, as detected by the Hall probes positioned at the centre, 4.4 mm and 10.0 mm from the centre of the sample. The measurements were conducted on both bare bulk Nb (without any coating) and Nb samples coated with NbTiN and AlN. Significant improvement in B_p is readily apparent across all coated Nb samples compared to the bare Nb (represented by the blue curve). This observation clearly demonstrates the screening effect of the multilayer films on the surface of bulk Nb. The magnitude of the increase, however, is dependent upon the thickness of the NbTiN layer as shown in Fig. 7(b). For comparison, Fig. 7(a) shows the maximum screening field as a function of NbTiN thickness derived from the contour plot in Fig. 2.

The observed variation of B_p with the NbTiN layer thickness is consistent with theoretical predictions of an optimal thickness for the NbTiN layer at which the

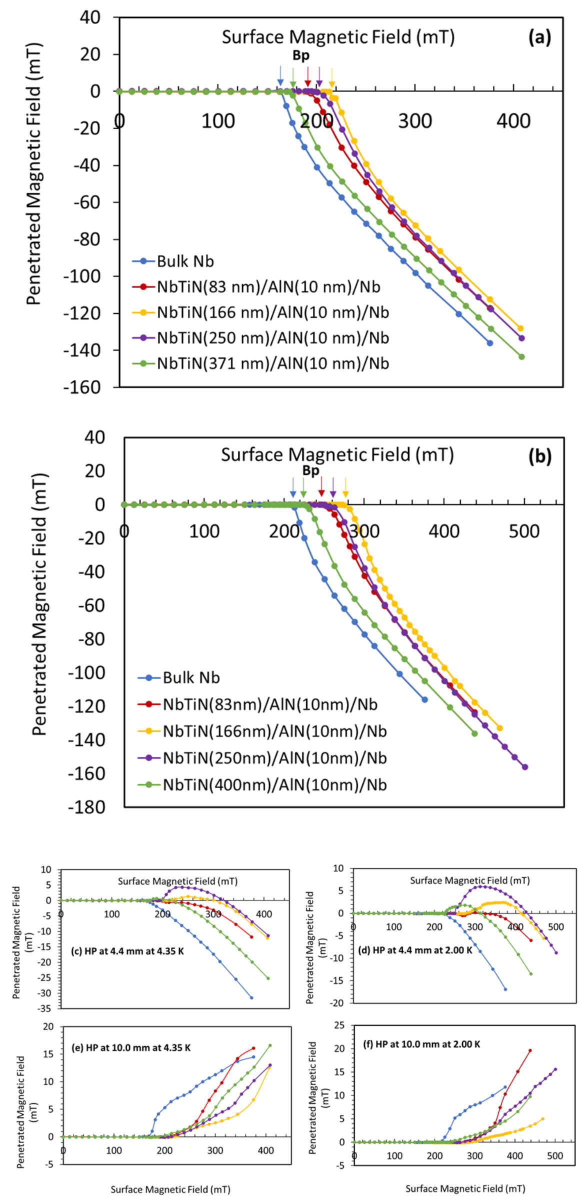

Figure 6: Magnetic field penetration measurements detected from the centre Hall probe (a, b), Hall probe at 4.4 mm (c, d) and 10.0 mm (e, f) from the centre at both temperature 4.35 K and 2.00 K.

surface breakdown field is maximum. We observed a nearly 25% increase of B_p at the optimum thickness of the NbTiN layer compared to B_p for Nb substrate.

All of these thin films were deposited on BCP Nb substrates with the surface roughness of a few microns, and other defects such as scratches and dislocations. We can anticipate further improvements in the performance of the thin films by employing smoother substrates. Smoother substrates can provide a better surface for film nucleation, minimizing growth defects and overall resulting film

ISBN: 978-3-95450-234-9 ISSN: 2673-5504 doi:10.18429/JACoW-SRF2023-MOPMB083

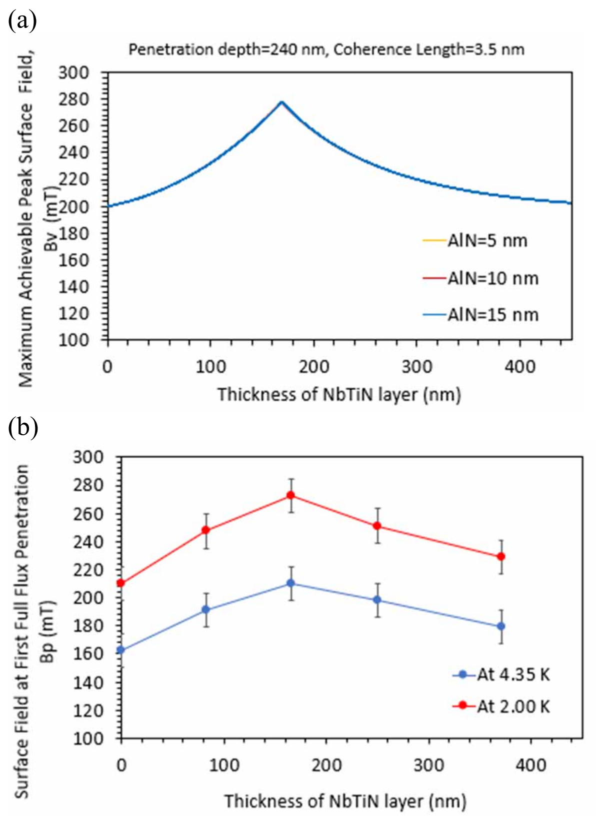

Figure 7: The variation of (a) theoretical Bv and (b) experimental Bp as a function of NbTiN thickness.

roughness which enhances the overall performance and characteristics of the thin films.

EFFECT OF THE SURFACE ROUGHNESS

In this study, the next step focuses on the production of an Nb substrate with a smoother surface. To achieve this, the mechanical polishing (MP) procedure was optimized to efficiently remove the damaged surface layer and attain an evenly polished surface with a mirror-like finish. This optimization aimed to reduce the time and effort required for the polishing process. Subsequently, electropolishing (EP) was performed on the sample to eliminate approximately 3-4 µm of material from the surface to remove any remaining material from the MP process.

However, during testing of the Nb samples at cryogenic temperatures, hydrides (lager than 100 µm) were observed at the surface. To address this issue, a new set of samples underwent a heat treatment process at 800 °C for 3 hours. After the heat treatment, the samples underwent another round of EP.

The MFP tests of the treated samples have shown a small increase in B_p compared to BCP Nb, as indicated by the red and blue cross marks in Fig. 8. However, this increase is not as substantial as expected.

Additionally, when examining the surface using backscatter images (Fig. 9), we observed that tiny hydrides still persist on the surface. These findings suggest that further improvements are necessary in the surface

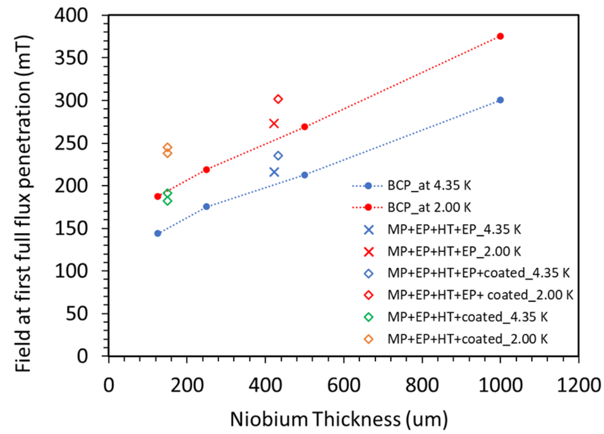

Figure 8: Field penetration measurements of Nb with different surface treatments.

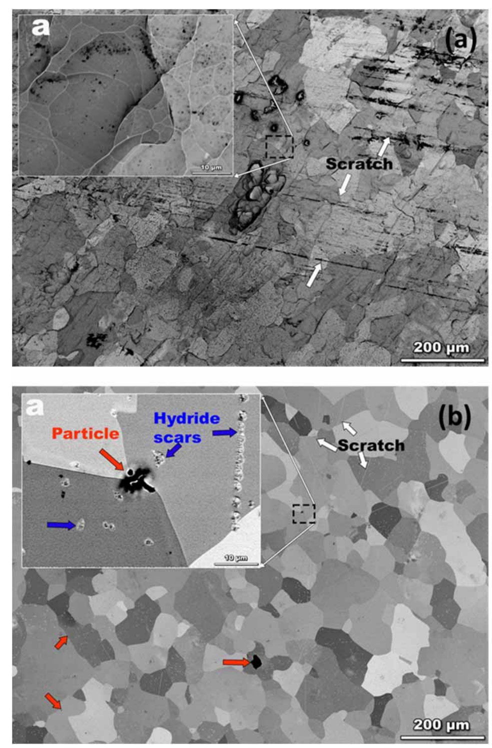

Figure 9: The Backscatter Electron (BSE) images of (a) BCP Nb and (b) MP+EP+HT+EP Nb. (MP: mechanical polishing, EP: electro polishing, HT: heat treated.

treatment process to effectively eliminate the hydrides and achieve a stronger enhancement in B_p.When a thin film consisting of two stacks of NbTiN and AlN was deposited onto a smooth Nb surface, a notable enhancement in B_p was achieved (blue and red diamonds).

To compare the effect of substrate roughness on MFP measurements, sample 3 and 4 from Table 1 were also deposited on smooth Nb surfaces. In order to prevent contamination during the heat treatment in a furnace, these Nb samples were carefully transferred to the coating chamber for deposition. Prior to the coating, a standard practice of subjecting the samples to a 24-hour bake at 600 °C was employed. This baking process is sufficient to eliminate hydrogen from the samples, ensuring a clean surface for the subsequent deposition process. Indeed, an enhancement in B_p was observed depending on the NbTiN film thickness when compared to the BCP substrate (green and orange diamonds). However, there is still room for further improvement in Bp by optimizing the surface treatments and utilizing high-purity cavity grade materials, which can lead to greater enhancements in the overall SRF performance of SIS coatings.

CONCLUSIONS

We investigated the impact of multilayer thin films on the enhancement of field at the first magnetic penetration field, B_p, in bulk Nb. Through the deposition of single layers of NbTiN and AlN on Nb substrates, we achieved a notable increase in B_p compared to bare Nb substrates. The observed enhancement of B_p depends on the thickness of the NbTiN layer, in agreement with theoretical predictions.

To obtain a mirror-finish smooth Nb surface, we optimized the mechanical polishing procedure followed by electropolishing. Heat treatment of Nb was performed to remove hydrogen to prevent hydride formation during cryogenic testing. The smoothening of the Nb surface resulted in an increase in B_p. After the polished Nb was coated with NbTiN and AlN layers, a further increase in B_p was observed compared to those layers on BCP Nb.

ACKNOWLEDGEMENT

The authors thank Shreyas Balachandran for providing the BSE images and the SRF staff at Jefferson Lab for the technical support.

REFERENCES

[1] A. Gurevich, "Enhancement of rf breakdown field of superconductors by multilayer coating", *Appl. Phys. Lett*., vol. 88, p. 012511, Jan. 2006. doi:10.1063/1.2162264

[2] I. H. Senevirathne, A. Gurevich, and J. R. Delayen, "Direct Current Magnetic Hall Probe Technique for Measurement of Field Penetration in Thin Film Superconductors for Superconducting Radio Frequency Resonators", *Rev. Sci. Instrum.,* vol. 93, p. 055104, May 2022. doi:10.1063/5.0083309

[3] A.-M. Valente-Feliciano, "Superconducting RF materials other than bulk niobium: a review", *Supercond. Sci. Technol.,* vol. 29, no. 11, p.113002, Sep. 2016. doi:10.1088/0953-2048/29/11/113002

[4] T. Kubo, Y. Iwashita, and T. Saeki, "Radio-frequency electromagnetic field and vortex penetration in multilayered superconductors", *Appl. Phys. Lett.,* vol. 104, p. 032603, Jan. 2014. doi:10.1063/1.4862892

[5] A. Gurevich, "Maximum screening fields of superconducting multilayer structures", *AIP Adv.* vol. 5, p.017112, Jan. 2015. doi:10.1063/1.2162264

[6] I. H. Senevirathne, G. Ciovati, and J. R. Delayen, "Measurements of the Magnetic Field Penetration into Superconducting Thin Films", in *Proc. SRF'19*, Dresden, Germany, Jun.-Jul. 2019. doi:10.18429/JACoW-SRF2019-THP050

[7] I. H. Senevirathne, A-M Valente-Feliciano, A. Gurevich, and J. R. Delayen, "Measurement of magnetic field penetration in superconducting materials for SRF cavities", in *Proc. IPAC'21*, Campinas, Brazil, May 2021. doi:10.18429/JACoW-IPAC2021-MOPAB396

[8] I. H. Senevirathne, A. Gurevich, and J. R. Delayen, "Magnetic Field Penetration Technique to Study High Field Shielding of Multilayered Superconductors", in *Proc. SRF'21*, East Lansing, MI, USA, Jun.-Jul. 2021. doi:10.18429/JACoW-SRF2021-SUPTEV001

[9] I. H. Senevirathne, D. R. Beverstock, A-M Valente-Feliciano, A. Gurevich, and J. R. Delayen, "Field Shielding of NbTiN Based Multilayer Structure for Accelerating Cavities", in *Proc. LINAC'22*, Liverpool, UK, Aug.-Sep. 2022. doi:10.18429/JACoW-LINAC2022-THPOGE19

[10] I. H. Senevirathne. "Measurements of Magnetic Field Penetration of Materials for Superconducting Radio Frequency Cavities", Ph.D. Dissertation, Phys. Dept., Old Dominion University, VA, USA, 2023. doi:10.25777/dq8r-g881