RF AND STRUCTURAL CHARACTERIZATION OF NEW SRF FILMS *

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Abstract

In the past years, energetic vacuum deposition methods have been developed in different laboratories to improve Nb/Cu technology for superconducting cavities. JLab is pursuing energetic condensation deposition via Electron Cyclotron Resonance. As part of this study, the influence of the deposition energy on the material and RF properties of the Nb thin film is investigated. The film surface and structure analyses are conducted with various techniques like X-ray diffraction, Transmission Electron Microscopy, Auger Electron Spectroscopy and RHEED. The microwave properties of the films are characterized on 50 mm disk samples with a 7.5 GHz surface impedance characterization system. This paper presents surface impedance measurements in correlation with surface and material characterization for Nb films produced on copper substrates with different bias voltages and also highlights emerging opportunities for developing multilayer SRF films with a new deposition system.

INTRODUCTION

Bulk Nb has been, for the past three decades, the material of choice for superconducting RF (SRF) accelerator systems. The primary reason is that Nb has the highest transition temperature (T_c=9.25K) and the highest lower critical magnetic field (H_{c1}) of any elemental superconductor. H_{c1} is the local magnetic field at which flux first penetrates into the conductor surface. The utility of a particular extended SRF material is believed to be constrained by its H_{c1}, or, perhaps its "superheated" field H_{sh} [1]. This determines the maximum accelerating field for a given cavity structure.

In recent years, SRF cavities using Nb, often in single cell cavities and occasionally in multi-cell structures, have reached RF performances approaching its theoretical limit ($H \sim H_c \sim 180$mT at 2K) [2]. Achieving the magnetic field limitation means that no further increase in the accelerating electrical field of bulk Nb cavities can be expected, limiting these accelerating gradients to about 42 MV/m. Any further significant improvement in performance or in system cost reduction to enable larger SRF accelerators operating at higher energies will necessarily come via the use of improved or alternative materials.

The SRF community strives to go significantly beyond these limitations and to achieve a factor of two or three in increased performance and/or an order of magnitude decrease in unit cost of acceleration. Two opportunities exist to accomplish these goals through the use of thin film technology to create viable superconducting RF cavity surfaces:

- The first exploits the freedom to decouple, at the system design level, the active SRF surface from the accelerating structure definition and its cooling, opening the possibility to dramatically change the cost framework of SRF accelerators.
- The second involves a concept recently proposed by A. Gurevich [3, 4] for overcoming the fundamental bulk material limitation, H_{c1}, by using a multilayer superconductor-insulator-superconductor (SIS) thin film structure.

Bulk-like Nb Films

Due to the very shallow penetration depth of RF fields (only ~40nm for Nb), SRF properties are inherently a surface phenomenon, involving a material thickness of less than 1 micron. One can then foresee the merits of depositing an Nb film on the inner surface of a castable cavity structure made of copper (Cu) or aluminium (Al).

CERN has conducted pioneered studies [5-7] in the field of SRF Nb films on Cu (Nb/Cu) applied to cavities and successfully implemented this technology in LEP-2. The following R&D work produced 1.5 GHz cavities achieving gradients up around 25MV/m [8]. However, these cavities suffered from significant losses resulting in the significant reduction of Q at accelerating gradients above 15MV/m. Some of the defects were inherent to the magnetron sputtering technique used to produce these cavities.

While tight correlation with the characterization of real materials has yet to be described, there exists a theoretical framework describing the relevant material parameters of surfaces as they influence SRF properties. Several material factors, highly dependent upon the surface creation conditions, contribute to degraded SRF performance with respect to ideal surfaces. These factors such as intra-granular impurities and lattice defect density, inter-granular impurities and oxidation, surface topography and chemistry, may lead to the reduction of the electron mean free path, thus the reduction of H_{c1}, contribute to electron scattering, "weak links" to the flow of surface super-currents and lossy sub-oxides creating non-linear loss mechanisms.

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The relative contribution of these limiting factors needs to be examined for any method used to produce SRF thin films. Fundamental work is needed to establish the correlation of detailed material characteristics with the consequent SRF performance.

Understanding the characteristics of the films produced, including systematic assessment of the RF surface impedance and other parameters like the London penetration depth λ, is the focus of an on-going study at JLab.

SIS Multilayers

Many superconducting compounds have shown higher T_c than Nb. However, none of these materials can, at present, match Nb either in terms of ease of use, or in terms of performance with increasing RF fields. Although these materials such as Nb_3Sn and NbN exhibit T_c and higher critical magnetic field, H_{c2}, higher than for Nb, their H_{c1} is lower due to their inherently small electronic mean free path, l, and consequently low coherence length, ξ. The lower critical field, H_{c1}, is the field at which magnetic vortices enter the superconductor, dramatically increasing the RF losses. In the case of multilayers, it represents an upper limit for useful SRF cavity fields, when the superconducting surface is thicker than the London penetration length, λ.

A major opportunity with a potentially enormous impact on the field of SRF accelerators is the multilayer film concept introduced by Alex Gurevich [3,4] which would allow to take advantage of high-Tc superconductors without being penalized by their lower Hc_1. The idea is to coat SRF cavities with alternating superconducting and insulating layers (SIS structures) with a thickness d smaller than the penetration depth λ. If the superconducting film is deposited with a thickness d <λ, the Meissner state can be retained at magnetic field much higher than bulk H_{c1}. The higher-T_c thin layers provide then magnetic screening of the superconducting cavity (bulk or thick film) without vortex penetration. The strong increase of Hc_1 in films allows using RF fields higher than the critical field H_c of Nb. The BCS resistance is also strongly reduced because of the use of superconducting layers with higher gap Δ (Nb_3Sn, NbTiN …). With such structures, Q-values at 4.2K of two orders of magnitude above Nb values could in principle be achieved.

Depending on the number of layers used, one may potentially attain accelerating gradients two to three times higher than that available with Nb. It is then attractive and seemingly credible to envision a 60 MV/m accelerator application operating at 4.2 K, using cast Al, pipe-cooled structures on which have been deposited multi-layer SIS films. Realization of such surfaces would greatly expand the performance reach of particle accelerators and significantly change their cost structure. Multi-layer SRF films offer promise of dramatically higher theoretical field limits.

JLab is pursuing both of these opportunities with the implementation of energetic condensation through Electron Cyclotron Resonance (ECR) and High Power Pulse Magnetron sputtering (HPPMS) and with the use of the new ultra high vacuum (UHV) multi-technique deposition system, in parallel with the use of another UHV system which offers in-situ diagnostics critical to evaluate the nucleation of the various films on their substrates.

ECR COATED NIOBIUM FILMS

Energetic Condensation via ECR

With the availability of techniques for energetic condensation in vacuum, Nb films with a wide range of microstructural properties and features believed to be relevant to RF performance degradation can be produced, characterized and RF tested. Properties such as film purity, stress, texture, and grain size can be measured over a wide range of values not accessible using conventional sputtering techniques. Various techniques are curently explored and developed by different research teams around the world [9-12]. JLab is pursuing Electron Cyclotron Resonance (ECR) Nb ion source in UHV [10] and High Power Pulse Magnetron Sputtering in the self-sputtering mode (HPPMS) [12] with energetic ions only and neutral atoms significantly blocked (method initially developed at LBNL). The main advantages of these techniques are the production of higher flux of ions with controllable incident angle and kinetic energy [13-15] and the absence of macroparticle production.

The challenge is to develop an understanding of the film growth dynamics from nucleation to final exposed surface. What matters most is the defect density (which determines the electron mean free path) within the rf penetration depth. This defect density is certainly affected by intragrain contaminants incorporated during the final stage film growth, but it is also strongly affected by the underlying crystal texture, which is in turn developed from the initial film nucleation process, which necessarily is strongly influenced by the substrate. The development of every stage can be expected to depend strongly on the kinetic energy distribution of the arriving Nb ions.

To better understand the nucleation and influence of the diverse deposition parameters, substrate nature, temperature and morphology on the final RF surface for Nb films, JLab is conducting studies on two fronts.

The study of the correlation of the surface resistance of ECR coated films with surface and material properties (structure, morphology, impurity content…) of the film as function of deposition energy and substrate temperature is ongoing.

In parallel, nucleation studies are underway within the frame work of a collaboration with the College William & Mary using a UHV deposition system equipped with in-situ reflection high-energy electron diffraction (RHEED) and scanning tunnelling microscope (STM) to monitor the in situ crystal character dependence on substrate properties and deposition parameters (temperature, working gas, intermediate annealing …). In this system, both standard magnetron sputtering and HPPMS can be implemented.

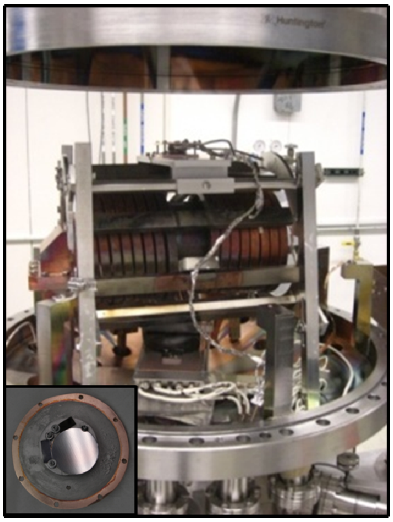

Figure 1: ECR system (a) and sample holder with a typical set of samples (b).

The objective is to grow and characterize niobium films with controlled deposition energy and substrate temperature to minimize the defect density and achieve bulk-like performance. While studying film growth in homo-epitaxy and hetero-epitaxy on both single crystal and polycrystalline substrates, particular attention is given to three sequential phases: (1) film nucleation on the substrate, (2) growth of an appropriate template for subsequent deposition of the final rf surface, and (3) deposition of the final surface optimized for minimum defect density.

The following sections present some of the standard measurements performed both for surface and material characterization and RF charcterization. For each deposition run, a set of standard samples comprising a 50 mm Cu disk and All the measurements presented are related to a Nb film produced by ECR at a bias of -90V.

Material Characterization

To inform the interpretation of the integrated film growth dynamics and influence of various deposition parameters, the standard surface and structural characterization includes X-ray diffraction (XRD), Electron Backscatter Diffraction (EBSD), Scanning Auger Electron Microscopy (SAM), high resolution Secondary Electron Microscopy (HR-SEM), high resolution Transmission Electron Microscopy (HR-TEM), and Secondary ion Mass Spectroscopy (SIMS).

In Figure 2, EBSD maps show that, for relatively high incidence ion energy (146eV), the Nb film tends to mimic the structure of the underlying Cu substrate.

Figure 3 shows high resolution bright field TEM micrographs revealing a sharp interface (a) and an oxide layer about 5nm thick. Figure 4 shows an average film roughness of about 94nm, most likely driven by the roughness of the underlying Cu substrate.

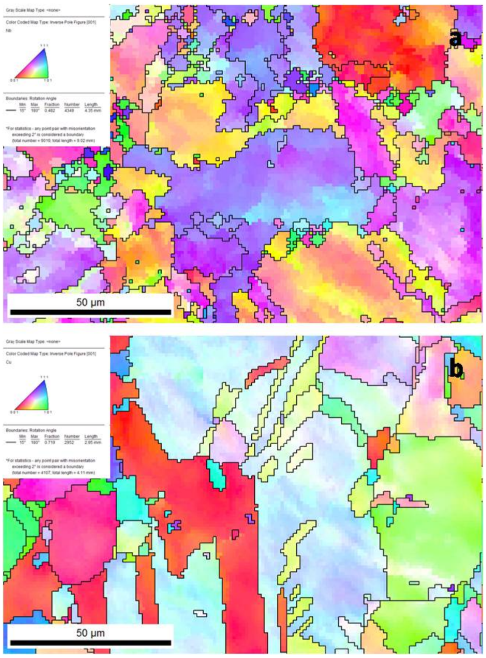

Figure 2: EBSD maps of the Nb film (a) and the Cu substrate (b). These areas are not directly superposed.

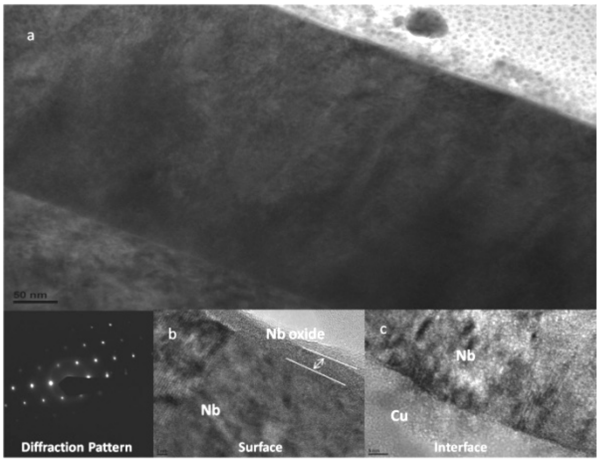

Figure 3: High resolution TEM image (a) of the ECR Nb film with corresponding diffraction pattern (b) and detail of the film surface (c).

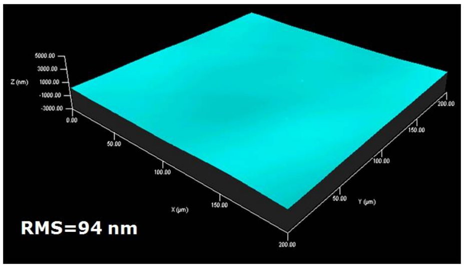

Figure 4: Surface roughness measured by profilometry.

RF and Cryogenic Characterization

The superconducting surface impedance, the penetration depth (λ) and the temperature dependence of Hc1 for each sample is investigated with the use of the Surface Impedance Characterization system (SIC) using a 7.5 GHz TE_{011}sapphire-loaded Nb cavity [16]. 50 mm sample discs will receive RF characterization in the SIC system recording the following measurements:

- Surface impedance as a function of magnetic field and temperature, from 1.9 K to the transition temperature (Figure 5 represents the surface impedance R_s for the film of interest as a function of temperature T).
- The change of superconducting penetration depth, Δλ, at 7.5 GHZ and low field will be measured by carefully tracking the cavity frequency with temperature from 2 K to right above the transition temperature while the rest of the cavity is held at 2 K.

The cryogenic performance is investigated with the 4-point-probe method to measure T_c and RRR.

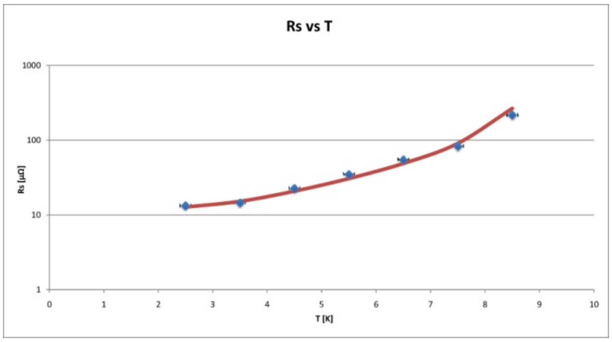

Figure 5: Surface Impedance measurement with SIC.

UHV MULTI-TECHNIQUE SYSTEM FOR SIS MULTILAYERS DEPOSITION

To implement the deposition in-situ of SIS multilayers as proposed by Alex Gurevich, JLab has built a UHV multi-technique deposition system tailored to multilayers deposition.

This system is equipped with three RF/DC magnetron heads for 50 mm targets and a RF/DC Ion source producing a 50mm beam with Ar, Kr. A self-sputter magnetron for implementation of HPPMS in self-sputtering mode is available and will be added to the system after the first phase of the commissioning is completed. A set of thickness monitors allows the thickness control of the deposited layers of different materials. Several injection lines are available on the main chamber for the injection of the sputtering working gas (Ar and Kr) and N_2 and O_2 used for reactive sputtering.

Figure 6: UHV multi-technique deposition facility.

This combination allows the implementation of DC magnetron sputtering for the deposition of metallic films or compounds like Mo_3Re. Compounds like NbN, NbTiN, as well as dielectric films can be coated via reactive sputtering. RF magnetron sputtering is commonly used for the deposition of insulator layers like aluminum oxide (Al_2O_3), MgO. With the ion source, the deposition of an insulator can be achieved with an Al_2O_3 target. The ion source can also be used to plasma etch the substrate prior to deposition, getting rid of its oxide layer for example. With the combination of magnetrons and ion source, Ion Beam Assisted Deposition (IBAD) can use energetic ions to modify film density, stress, texture, grain size, structure of the interface and other related properties.

The main chamber is pumped with a cryopump CTI-Torr 8 (1500 l/s) through a VAT gate valve with variable conductance. The base pressure for the main chamber is in 10^{-9} Torr range. During deposition, to allow a better control of the working gas pressure, the diaphragm valve aperture can be varied to adjust the conductance. The system is also equipped with a non evaporable getter (NEG) chamber to improve hydrogen pumping during the deposition process. A strip of ST707 mounted on a cylindrical support can provide pumping speeds up to 2000l/s for H_2 and CO. Baking, internal UV light exposure and plasma cleaning can be used to improve further the vacuum quality.

The vacuum quality is controlled by the combination of a convectron gauge and an UHV ion gauge in each chamber and a Residual Gas Analyzer (RGA) that can be used with differential pumping allowing the analysis of the gas composition in the vacuum system during deposition. A high-pressure ionization gauge is used in the main chamber to measure the working gas pressure during film deposition.

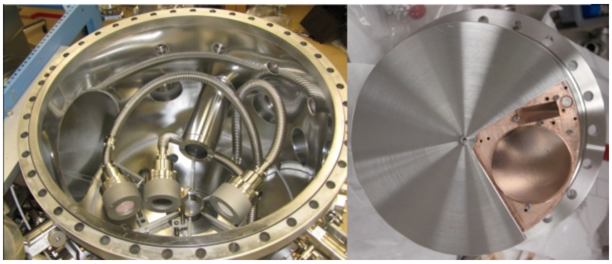

Figure 7: Setup in the UHV multi-technique deposition system main chamber and sample holder.

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 some deposition parameters during the same deposition run. All sample stages can be heated above 500˚C and are equipped with shutters to allow the deposition in the same run of multiple sets of samples using different parameters, ensuring directly comparable environmental conditions.

The system is now under commissioning with coating runs to produce Nb films, NbN, NbTiN films by reactive DC sputtering. As for the study on Nb films, nucleation studies for each type of layer will be performed in parallel with the UHV system at William & Mary. Additionally to the standard set of measurements, i.e. surface and material characterization coupled to cryogenic and RF measurements with the SIC cavity, a method is being developed to observe the onset of flux penetration in the produced SIS multilayer structures [17].

CONCLUSION

JLab in collaboration with surrounding universities is pursuing two opportunities to create viable superconducting RF cavity surfaces to reduce the cost framework of SRF accelerators and to reach higher gradients and allow operation of SRF structures at 4K.

The first approach is to understand and develop niobium films with bulk-like performance by elucidating the functional dependence of film-grown niobium crystal texture, intra-grain defect density, and grain boundary impurities on SRF performance. Studies on the correlation of the surface resistance of films produced by energetic condensation ECR with surface and material properties of the film as a function of incident ion energy and substrate temperature are underway.

The second approach is to create SIS multilayer structures following the concept proposed by A. Gurevich for overcoming the fundamental bulk material limitation, H_{c1}. To enable exploration of this effect, a new UHV multi-technique deposition system tailored to multilayer deposition has been build and is now under commissioning.

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