DEVELOPMENT AND CHARACTERIZATION OF THIN FILM SUPERCONDUCTING RADIO FREQUENCY SURFACES FOR ACCELERATOR CAVITIES

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Abstract

Superconducting thin films have the potential to improve the performance of particle accelerators. Before these thin films can be implemented, a systematic study on structure-property correlations is necessary. Here, we present the characterization of niobium thin films deposited onto both ceramic and metallic substrates. In particular, the film microstructure and superconducting properties are examined. Our findings show that the strain and crystallographic orientation of niobium films can adversely impact the transition between the superconducting and normal state as well as the lower critical field.

I. Introduction

Particle accelerators are tools used by scientists to study sub-atomic particles as well as properties of materials. NASA created the Space Radiation Laboratory at Brookhaven National Laboratory for radiobiology studies that simulate exposure to space radiation [1-2]. Results from these studies will help protect astronauts from genetic mutations, cancer, and other adverse effects that result from being subjected to radiation in space.

Traditionally, particle accelerators have comprised of superconducting radio frequency (SRF) cavities made entirely out of Nb. However, the performance of these cavities has been optimized such that they are now approaching fundamental material limits of Nb such as the first critical magnetic field, $H_{C1}$. In order to overcome these limits, a model has been proposed that involves stacking alternating superconducting-insulating-superconducting (SIS) layers on a Nb surface, where the superconductor has a higher $H_{C1}$ and critical temperature ($T_C$) than Nb [3]. This structure would shield the cavity from higher magnetic fields and thus allow for an increase in the accelerating gradient. Incorporating this SIS model could increase the capability of accelerator experiments such as those at the Space Radiation Laboratory at Brookhaven National Laboratory.

Before these SIS structures can be implemented, fundamental studies on Nb thin films and the correlation between their structure and superconducting properties need to be carried out. Here, work on Nb thin films deposited on both ceramic and metallic surfaces is presented. In particular, the effects of strain at the substrate-film interface and variations of the growth parameters are explored.

II. Niobium Thin Films on Ceramic Surfaces

In order to study the growth of epitaxial Nb thin films on ceramic substrates, a correlated study of structure and superconducting properties was carried out on $\text{Al}_2\text{O}_3(11\overline{2}0)$ surfaces [4]. The films were prepared using DC magnetron sputtering in an ultra-high vacuum (UHV) system. The structure of the films was examined in-situ using Reflection
High Energy Electron Diffraction (RHEED) as well as ex-situ using X-ray diffraction (XRD) and Transmission Electron Microscopy (TEM). Superconducting properties such as transition temperature and AC susceptibility were studied using Superconducting Quantum Interference Device (SQUID) magnetometry.

Due to a lattice mismatch between the thin film and the substrate, strain is always found at the interface between the materials. Strain in the early stages of growth can affect structure properties in the entire film such as grain size, dislocations, and number of phases present. Early stages of growth in Nb films were studied using RHEED which has the capability to probe the in-plane lattice parameter of crystalline materials. During film growth, the in-plane lattice parameter was measured after each atomic layer (AL) was deposited. These measurements, shown in Figure 1, show how Nb forms a hexagonal phase for the first three atomic layers before returning to its native body centered cubic (bcc) form. While this hexagonal phase has previously been reported [5], the strain found in the bcc phases from 6-14 AL has not been reported. Nb forms a hexagonal phase followed by the strained bcc phase in order to overcome the 10.7% strain along the Nb[100] direction and the 8.3% strain along the Nb[110] in the Nb(100)/Al₂O₃(1120) system.

The out of plane strain in Nb films was studied using XRD and TEM. XRD measurements, displayed in Figure 2 (Top), show that the out of plane lattice parameter is expanded compared to bulk for thinner films, but relaxes to a bulk like value as the film thickness increases. The out of plane strain for the 30 nm, 100 nm, and 600 nm films were 1.25%, 0.36%, and 0.20% respectively. While it is clear that strain is present in these films, the measured strain is an averaged value over the entire thickness of the film. Therefore, in order to have a more precise measurement of the out of plane strain near the interface, a Nb film was examined with TEM as shown in Figure 2 (Bottom).

![Figure 1: The evolution of strain versus film thickness.](image)

Fourier transforms were then used on the TEM images to find that the spacing corresponding to the distance between Nb(110) planes was 2.37 Å, larger than the bulk value of 2.333 Å. Combining the RHEED, XRD, and TEM measurements, we can see that in Nb(110)/Al₂O₃(1120) films, there is a strained region near the interface containing both hexagonal and cubic phases as well as a more bulk like region located away from the interface.
In order to study what effect this strained region would have on the superconducting properties, the 30 nm, 100 nm, and 600 nm films were examined with SQUID magnetometry. In particular, the transition temperature and AC susceptibility were measured. AC susceptibility is defined in Equation 1, where the real part corresponds to the sample’s response to an external magnetic field and the imaginary part corresponds to losses in the system.

\[
\chi(\omega) = \chi'(\omega) + i\chi''(\omega) \tag{1}
\]

As shown in Figure 3, the AC susceptibility for the 30 nm film has a double step in the real component and a corresponding double peak in the imaginary component. The thicker films show only a single step and corresponding peak. The smaller of the two peaks seen in the 30 nm film is related to the strained region at the interface while the larger peak corresponds to the relaxed region away from the interface. While the strain is still present in the thicker films, the effect of this strained region is diminished due the amount of material that is relaxed that will dominate over any interfacial effects. These interfacial effects will have an impact when fabricating SIS devices where the film thicknesses are on the order of tens of nanometers. The transition temperatures seen in Figure 3 are lower than the bulk value of 9.2 K because there was a 100 Oe field applied during the measurements which effectively lowers the transition temperature.
In addition to $\text{Al}_2\text{O}_3(11\bar{2}0)$ substrates, Nb films were deposited onto MgO(001) surfaces which allow for either Nb(001) or Nb(110) growth depending on the growth conditions and surface preparation. The surface morphology, obtained using Atomic Force Microscopy (AFM), of Nb/MgO films is greatly dependent on which orientation that the films grows as shown in Figures 4 and 5. Nb(001) films develop surfaces with isotropic features while Nb(110) films form anisotropic features that are oriented 90° with respect to each other. These features correspond very well to the expected epitaxy that Nb(001) has one possible orientation on MgO(001) surfaces while Nb(110) films have two possible orientations that are rotated 90° from each other. Additionally, the features scale with increasing film thickness. By applying principles of general dynamic scaling and power spectral density (PSD) techniques [6], these two types of surfaces can be classified by what scaling class they fall under. PSD analysis of the AFM images show that Nb(001)/MgO(001) follows the behavior of the “new” universality class while Nb(110)/MgO(001) follows the behavior of the “super-rough” universality class [7].

The Nb surface will determine how subsequent layers will grow and ultimately their properties. Because surface roughness can be detrimental to SRF performance, subsequent studies to correlate surface roughness with critical field are needed.

#### III. Niobium Thin Films on Copper Surfaces

In addition to incorporating superconductors with higher $T_C$ and $H_{C1}$ into SRF cavities, there is also a desire to use Nb-coated Cu cavities for thermal efficiency. Cu(001) surfaces were prepared and subsequently had Nb thin films deposited under various conditions to study how the growth parameters affect the superconducting performance [8]. One parameter investigated was the growth temperature.

RHEED measurements indicated that Nb grows with a (110) orientation on Cu(001) surfaces. The surface morphology of Nb films grown on Cu displayed features similar to those seen in the Nb(110)/MgO(001) samples. The surface features on the sample prepared at RT were finer than those found on samples.

Figure 4: AFM scans of Nb(001) surfaces for a thickness of (a) 30 nm, (b) 50 nm, (c) 100 nm, and (d) 1000 nm.

Figure 5: AFM scans of Nb(110) surfaces for a thickness of (a) 30 nm, (b) 50 nm, (c) 100 nm, and (d) 600 nm. All scans are 2 µm x 2 µm.
prepared at 150 °C and resulted in a lower RMS roughness (1.98 nm for RT, 2.87 nm for 150 °C). Figure 6 shows representative RHEED patterns and AFM scans of Nb(110)/Cu(001) films.

![RHEED pattern and AFM scan](image)

FIG. 6. (a) RHEED pattern for Nb(110)/Cu(100)/Si(100) along the Si[100] and Si[110] azimuths. (b) A representative 2 µm x 2 µm AFM scan for Nb films on the Cu template.

XRD measurements on these films give a lattice parameter exhibiting less than 1% strain compared to the bulk value. For the growth carried out at RT, the average grain size was found to be 44.5 nm while for growth at 150 °C it was 49.9 nm. The increased growth temperature allowed for the formation of larger crystallographic grains.

As shown in Figure 7, a higher growth temperature lead to a sharp transition between the superconducting state and the normal state while a lower growth temperature lead to a slower transition that started as low as 7 K. The higher growth temperature produced films with an $H_{C1}$ value of 100 Oe, while growth at RT produced films with an $H_{C1}$ value of 50 Oe.

![Superconducting transition](image)

Figure 7: Comparison of the superconducting to normal state transition for Nb films grown at 150 °C and room temperature (RT).

### IV. Conclusions and Future Work

This study on Nb thin films highlights issues that need to be accounted for when preparing SIS devices to be used in SRF cavities such as interfacial strain, the correlation between structure, surface morphology and superconducting properties, and growth parameters. It also provides valuable information for modifications to theoretical work that may assume bulk behavior in the SIS structure such as the presence of multiple phases and their detrimental superconducting effects.

Ongoing studies include a more complete examination of growth parameters for Nb thin films on Cu(001) surfaces as well as extending the study to Cu(111) surfaces since both surfaces will be found in Cu cavities. In the coming year, SIS devices will be fabricated using NbN and MgO (i.e. NbN on MgO on Nb) and their performance studied with SQUID magnetometry. Preliminary studies on other potential superconductors such as Nb(Ti)N, Nb$_3$Sn, and MgB$_2$ will also be carried out.
References


