Abstract

Avenues for the production of thin films tailored for Superconducting RF (SRF) applications are showing promise with recent developments in vacuum deposition techniques using energetic ions. JLab is using energetic condensation via Electron Cyclotron Resonance (ECR) plasma and High Power Impulse Magnetron Sputtering (HiPIMS) for the development of Nb films and multilayer SIS (superconductor-insulator-superconductor) structures to reach bulk Nb performance and beyond. Nb films with RRR comparable to bulk values are readily produced. The influence of the deposition energy on the material and RF properties of the films is investigated with the characterization of their surface, structure, superconducting properties and RF response. Nucleation studies investigate the best conditions to create a favorable template for growing the final SRF surface. This paper presents a progress report on the development of Nb on Cu films and on multilayer structures based on NbTiN and AlN.

THIN FILMS APPLIED TO SRF

SRF cavities using bulk Nb are reaching RF performances approaching the theoretical limit of the material (Hc1~180mT at 2K) [1]. Any further dramatic improvement in SRF performance or system cost reduction will necessarily come via the use of improved or alternative materials. JLab with neighbouring partners is pursuing two opportunities: the engineering of Nb films for improved SRF performance and the demonstration of the phenomenology and the development of multi-layered S-I-S (superconductor-insulator-superconductor) SRF film structures following the concept proposed by A. Gurevich [2].

Nb Films

RF fields have a very shallow penetration depth in the SRF material (~40nm for Nb). One can then envision depositing a thin layer of Nb on the inner surface of a castable cavity structure made of copper (Cu) or aluminium (Al). This opens the possibility to dramatically change the cost framework of SRF accelerators by decoupling the active SRF surface from the accelerating structure definition and cooling.

The viability of SRF Nb films on Cu (Nb/Cu) technology has been demonstrated with pioneer studies at CERN on 1.5 GHz cavities [3-5] and the successful implementation in LEP-2 with 352MHz cavities. Due to defects inherent to the magnetron sputtering technique used for Nb deposition, the 1.5 GHz Nb/Cu cavities produced suffered a significant reduction of Q at accelerating gradients above 15MV/m [6].

Several material factors, highly dependent upon the surface creation conditions, may contribute to degraded SRF performance by the reduction of the electron mean free path and enabling early flux penetration. Fundamental work is required to determine the functional dependence of film-grown niobium crystal texture, intragrain defect density, and grain boundary characteristics on the resulting SRF performance (surface resistance, lower critical field Hc1,...).

SIS Multilayer Films

The SIS concept [2] proposes to take advantage of high-Tc superconductors without being penalized by their lower Hc1. SRF cavities are coated with alternating superconducting and insulating (SIS) layers with a thickness d smaller than the penetration depth λ so the Meissner state can be retained at a magnetic field much higher than the bulk Hc1. The thin higher-Tc layers provide magnetic screening of the bulk superconducting cavity delaying vortex penetration. The BCS resistance is also strongly reduced because the envisioned superconducting materials (Nb,Sn,NbTiN,...) have a larger gap Δ than Nb. With such structures, Q-values at 4.2K could be increased a couple orders of magnitude above Nb values.

Towards Optimized SRF Films

The understanding of the dependence of the final RF surface for Nb and multilayer films on the characteristics of the films produced, the nucleation, the diverse deposition parameters, substrate nature, temperature and morphology is of primary importance.

The quality of the resultant thin film is heavily influenced by the deposition technique utilized. With the availability of energetic condensation techniques [7], films with a wide range of structure and features potentially relevant to RF performance can be produced. In this context, JLab is using an ECR plasma as a Nb ion source in ultra-high vacuum (UHV) [8]. The main advantages are the production of a high flux of singly charged ions with controllable kinetic energy and the
absence of macro-particle production with which to accomplish careful investigations into the film growth dynamics. For the multilayer structures development, the deposition techniques pursued are reactive magnetron sputtering and high power impulse magnetron sputtering (HiPIMS) [9].

Film growth is approached in three phases: Film nucleation on substrate, growth of an appropriate template for subsequent deposition of the final RF surface and deposition of the final surface optimized for minimum defect density.

**TAILORED NIOBIUM FILMS ON COPPER**

The challenge is to develop an understanding of the film growth dynamics from its nucleation to the final exposed surface. The defect density within the RF penetration depth determines the electron mean free path in that layer. It is certainly affected by impurities incorporated during the final stage film growth, but it is also strongly affected by the underlying crystal structure developed from the initial film nucleation and the substrate nature. The development of every phase can be expected to depend strongly on the kinetic energy of the arriving Nb ions.

Films are produced by ECR at different bias voltages, bake and coating temperatures on Cu substrates (single crystal and polycrystalline), as well as a variety of crystalline and amorphous insulator substrates which serve as controlled systems for analysis. It is found that the substrate properties and the initial growth conditions—ion energy and substrate temperature—strongly influence the final properties of the Nb film [10].

The on-going studies show that hetero-epitaxy of Nb on Cu single crystal substrates is easily achievable with high crystalline character and at temperatures low enough to maintain the mechanical integrity of the Cu substrate [11]. Films coated on polycrystalline Cu substrates are hetero-epitaxial, with grain size comparable to the underlying substrate. For both types of substrates, their crystalline quality seems to play a predominant role in the quality of the resulting Nb film.

![Figure 1: EBSD maps of Nb films grown on Cu (a, b, c) single crystal (100), (110), (111), polycrystalline (c) large grains and (d) fine grains simultaneously [10].](image)

On insulating crystalline substrates, RRR comparable to bulk Nb values are commonly achieved. For ECR films, the maximum RRR measured so far for Al₂O₃ (11-20) and MgO (110) are respectively 488 and 424 [12]. RRR values can be varied as a function of bias voltage, baking and coating temperatures over 2 orders of magnitude [10].

For Nb/Cu films, some dependence of RRR on the incident ion energy is observed. However, additional studies are necessary due to the strong influence of substrate crystalline quality on the resulting film structure. Similar high RRR values are achieved for both single crystal and polycrystalline substrates, suggesting that the presence of grain boundaries is not necessarily detrimental (Table 1).
Table 1: RRR for ECR Nb films deposited at 360°C versus Cu substrate nature

<table>
<thead>
<tr>
<th>Cu Substrate</th>
<th>RRR</th>
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<tbody>
<tr>
<td>(100)</td>
<td>129</td>
</tr>
<tr>
<td>(110)</td>
<td>275</td>
</tr>
<tr>
<td>(111)</td>
<td>249</td>
</tr>
<tr>
<td>Polycrystalline large grains (mm$^2$)</td>
<td>289</td>
</tr>
<tr>
<td>Polycrystalline fine grains ($\mu$m$^2$)</td>
<td>118</td>
</tr>
</tbody>
</table>

The surface impedance and other parameters of these films like the London penetration depth $\lambda$ are under study with RF measurements on corresponding disk samples in a sapphire-loaded TE011 cavity [13].

MULTILAYER FILMS

NbTiN and AlN have been chosen as candidate materials to develop a proof-of-principle for the SIS multilayer concept. The ternary nitride NbTiN presents all the advantages of NbN and exhibits increased metallic electrical conduction properties with titanium (Ti) content. Its superconducting cubic phase is thermodynamically stable at room temperature [14]. $T_c$ is slightly higher for NbTiN but as for NbN, N stoichiometry is critical to obtaining the right superconducting phase. AlN films have been found to enhance the properties ($T_c$) of very thin NbTiN films [15].

NbTiN/AlN/Nb Structures

SIS structures with NbTiN and AlN have been coated at various temperatures in-situ on bulk Nb and Nb/Cu substrates in a dedicated UHV multi-technique deposition system [16]. Although the best $T_c$ was achieved at 600°C (16.5K), the deposition at 450°C of thin NbTiN/AlN layers still yields a $T_c$ of 16.05K with $\Delta T_c$ of 0.23K. Figure 2 shows the FIB cross-section of one of these SIS structures.

These structures have been coated so far only via DC sputtering (5-7eV of ion energy). The energetics then only vary through the Ar pressure, the deposition rate and the substrate temperature. Energetic condensation will next be investigated with HiPIMS [16-18] to produce films with higher density and crystalline quality [9, 18, 19]. Once the structure is optimized, 50 mm disks will be coated and tested in setups specifically designed for SIS multilayers where RF fields parallel to the structure surface can be applied [20].

SUMMARY

Engineering Nb/Cu films with energetic condensation via extracted ECR plasma ions allows the tuning of the film structure from fiber growth to equi-axial growth by varying the incident ion energy for substrate temperatures lower than if using a thermal process only. As for films deposited on insulating crystalline substrates, Nb/Cu films can be produced with RRR approaching bulk Nb values. Work on multilayer structures based on NbTiN and AlN is also underway.

![Figure 2: TEM image of a FIB cut NbTiN/AlN/Nb/Cu structure.](image-url)

REFERENCES