MATERIAL QUALITY & SRF PERFORMANCE OF Nb FILMS GROWN ON Cu VIA ECR PLASMA ENERGETIC CONDENSATION*

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Abstract

The RF performance of bulk Nb cavities has continuously improved over the years and is approaching the intrinsic limit of the material. Although some margin seems still available with processes such as N surface doping, long term solutions for SRF surfaces efficiency enhancement need to be pursued. Over the years, Nb/Cu technology, despite its shortcomings, has positioned itself as an alternative route for the future of superconducting structures used in accelerators. Significant progress has been made in recent years in the development of energetic deposition techniques such as Electron Cyclotron Resonance (ECR) plasma deposition. Nb films with very high material quality have then been produced by varying the deposition energy alluding to the promise of performing SRF films. This paper presents RF measurements, correlated with surface and material properties, for Nb films showing how, by varying the film growth conditions, the Nb film quality and surface resistance can be altered and how the Q-slope can be eventually overcome.

INTRODUCTION

RF fields have a very shallow penetration depth in SRF materials (~40 nm for Nb). Instead of the commonly used bulk Nb, one can then envision using a thin layer of Nb deposited on the inner surface of a castable cavity structure made of copper (Cu) or aluminum (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 [1–3] and the successful implementation in LEP-2 with 352 MHz cavities. Due to defects inherent to the magnetron sputtering technique used for Nb deposition, the 1.5 GHz Nb/Cu cavities suffered a significant reduction of Q at accelerating gradients above 15 MV/m [4]. 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 thus 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 H_{c1}...).

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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 chosen deposition technique. With the recent developments in deposition techniques via energetic condensation [5], 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) [6, 7] for careful investigations into the film growth dynamics. 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.

NB/CU FILM STRUCTURE

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 of film growth, but it is also strongly affected by the underlying crystal structure developed from the initial film nucleation and the substrate nature. One can approach SRF Nb film growth in three phases: film nucleation on the 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. 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. As anticipated, 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 [8]. 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 [9]. Films coated on polycrystalline Cu substrates are hetero-epitaxial, with grain size comparable to the underlying substrate. The substrate crystal quality is shown to have a strong influence on the final quality and structure of the Nb film.

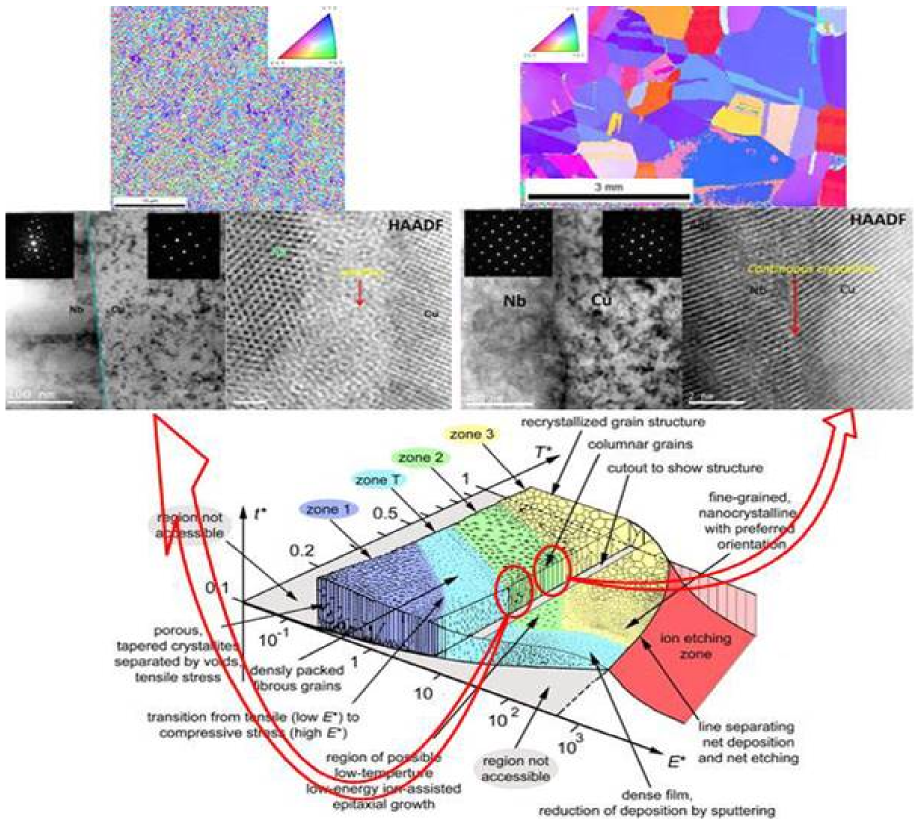

Figure 1: Illustration of the ECR Nb film microstructure variation with the generalized Structure Zone Diagram from [10].

Figure 1 illustrates how with varied deposition conditions (ion energy and substrate temperature) different structures in the structure Zone diagram can be achieved. For Nb/Cu films, a dependence of RRR on the incident ion energy is observed, at fixed coating temperature. At equivalent substrate quality, the quality of the ECR Nb films is very reproducible.

Microstructure for Hetero-epitaxial ECR Nb Films

For hetero-epitaxial growth, the samples are typically baked at a temperature higher than 360 ℃. For these baking temperature, the native Cu oxide layer is reduced and dissolved the Cu bulk, providing a crystalline surface and favoring Nb hetero-epitaxy. The samples are then coated in-situ at 360 ℃. For the film analyzed here, the substrate was large grain OFHC Cu. To produce large grain Cu substrates, OFHC Cu is heat treated at 1050 ℃, which fully re-crystallizes the material and releases strain and dislocations. The nucleation and early growth phases (first 100 nms) were performed at an ion energy of 184 eV, followed by a subsequent growth at 64 eV for the remainder of the film thickness. The idea is to provide enough energy to nucleate an adaptive Nb layer which then transitions to a relaxed Nb layer, providing a good template for homo-epitaxial Nb growth. Lowering the energy for the subsequent growth allows to continue growing a relaxed Nb, minimizing the defect density that can be associated with higher ion bombardment.

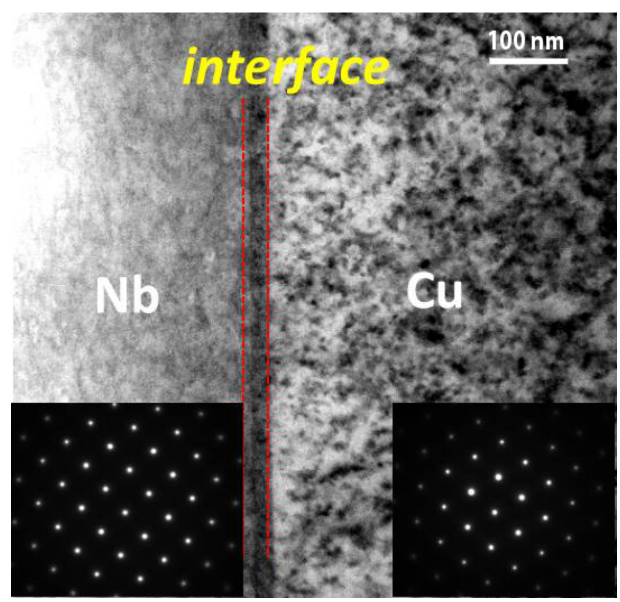

Figure 2: FIB-TEM cross section of an hetero-epitaxial film grown at 184 eV and 64 eV, at 360 ℃on large grain Cu.

FIB Cross-section and TEM/EELS Analyses

In the hetero-epitaxial growth mode, the Nb film mimics the Cu substrate and exhibit a film bulk structure with similar grain size. Although hetero-epitaxy can be achieved at lower coating temperature, Fig. 2 displays a TEM observation

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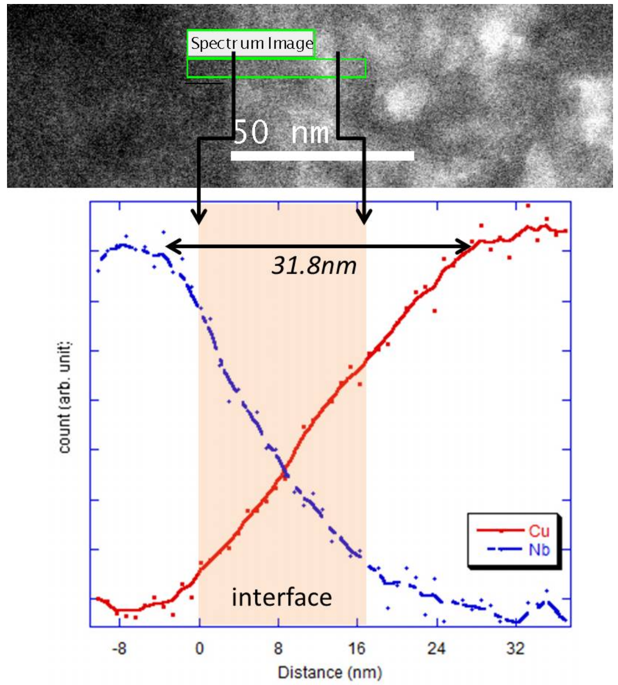

Figure 3: EELS analysis of the Nb-Cu interface of the ECR Nb film from Fig. 2

along with the diffraction patterns for the Nb film and the Cu substrate for a cross-section of the film mentioned above, prepared by FIB (focused ion beam).

Higher resolution TEM imaging reveals a continuously crystalline interface. The initial high energy ion bombardment coupled with a relatively high substrate temperature produces a dense and strong interface. The associated electron energy loss (EELS) measurement, represented in Fig. 3, shows an 30 nm crystalline interface where Nb and Cu are intermixed. For hetero-epitaxial films coated at lower energy or lower temperature, the film-substrate interface is much smaller, in the range of less than 10 nm.

Superconducting Gap Measurements

Superconducting gap measurements by Point Contact Tunneling (PCT) have been performed on some ECR samples at ANL. The technique is described in detail in [11]. The PCT measurement performed on the film represented in Fig. 2 are displayed in Fig. 4 with the junction conductance fits and superconducting gap dependence as a function of temperature. Although ECR hetero-epitaxial films generally display a narrow gap around 1.55 meV, closer to what is similarly measured for bulk Nb, this particular film displays a gap at 1.53 meV and a sub-gap at 1.38 meV. This is sometimes observed in bulk Nb cavities [12].

RF PERFORMANCE OF AN ECR NB/CU FILM

In an effort to characterize the potential RF performance of ECR Nb/Cu films and compare them with bulk Nb and films otherwise produced, a series of RF measurements

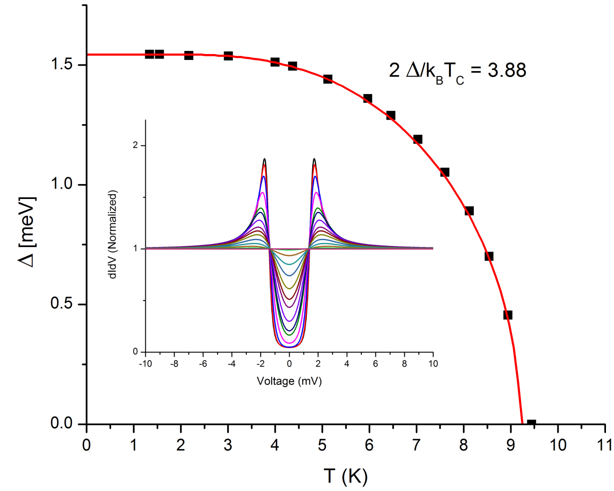

Figure 4: superconducting gap measurement by PCT for the hetero-epitaxial film from Fig. 2.

with the Quadrupole Resonator (QPR) at CERN has been launched. The main advantages for this setup [13–15] is the ability to produce detailed RF measurements at 400, 800, and 1200 MHz, frequencies comparable to bulk Nb and Nb/Cu cavity used frequencies.The surface resistance R_S can be measured over a range of fields, up to 60 mT, and temperatures. The shift in resonance frequency with temperature can also be measured allowing calculation of the penetration depth change. From these measurements, characteristic parameters can be derived such as the residual resistance R_{res} at low field, the penetration depth at 0K, λ_0, as well as the critical temperature, T_c. This section presents some of the results obtained with a film equivalent to the one presented in the previous section. The coating parameters were identical except for the Cu substrate nature. The QPR Cu sample was made of standard fine grain OFHC Cu grain whereas the film studied in the previous section was deposited on large grain OFHC Cu. The bake-out temperature was also 360 °C instead of 500 °C. The RRR measured on a simultaneously coated Nb/Cu film by the 4-point probe method [16] was 122 and T_c was 9.46 K. EBSD IPF mapping on both the QPR and witness samples shows a high quality polycrystalline Nb film with, as anticipated, a grain size of about 50 μm comparable to the Cu substrate. FIB-SEM observations show a very smooth film surface along a very smooth interface with the Cu substrate. Observation at the nm scale shows no porosity, no delamination or substructure. Derived from the various QPR measurements, the London penetration depth λ_0 is measured to be 37 nm, T_c is 9.36 K and the superconducting gap Δ is about 1.59 meV.

Figure 5 shows the performance at 4 K and 400 MHz of the ECR Nb film along with a reactor grade bulk Nb QPR sample (RRR 47), and a typical LHC magnetron sputtered cavity (RRR 10). As can be observed, R_{res} at low field for the ECR film is comparable to the LHC sputtered cavity, but the slope of R_S with field is comparable to the bulk niobium of similar RRR. The relatively high R_{res} may likely be

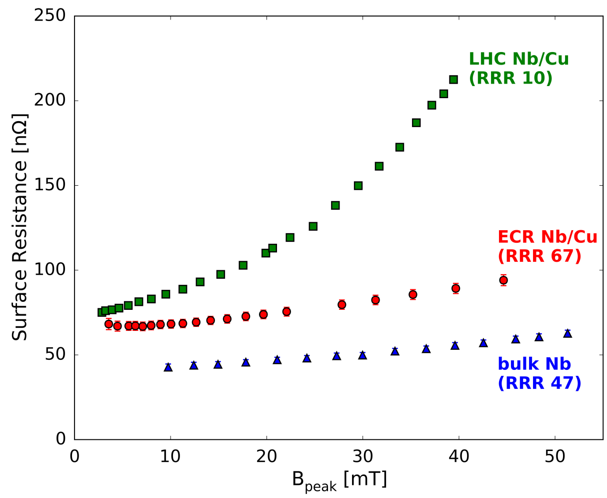

Figure 5: RF Measurement at 400 MHz and at 4 K of an ECR Nb/Cu film compared to sputtered and bulk Nb cavities.

due to contamination from the final e-beam welding of the sample to the support structure, performed post-deposition. Although this final weld is not on the coating itself, it generates heating of the substrate and the Nb film. At 2.5 K, a Q-slope is still present as compared to bulk Nb, but it is less pronounced and linear in field compared to the LHC magnetron sputtered cavity, for which R_s is proportional to exp (B_{RF}). More detailed analyses along with trapped magnetic flux and thermal cycling measurements are presented in [17].

CONCLUSION

Engineering Nb/Cu films with energetic condensation via extracted ECR plasma ions allows 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. Recent RF measurements of an ECR Nb hetero-epitaxial film reveals a behavior at 4 K and 400 MHz comparable with bulk Nb. Although an R_s increasing slope persists at 2.5 K, this shows promise for overcoming the so-far characteristic Q degradation for Nb/Cu films. To complete this work, a series of RF measurement of ECR films of various structures (hetero-epitaxial and fiber) in the Quadrupole Resonator at CERN is on-going.

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