

ECR NB FILMS GROWN ON CU SUBSTRATES: INFLUENCE OF ION ENERGY AND INTERFACE *

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Abstract

In the pursuit of niobium (Nb) films with similar performance with the commonly used bulk Nb surfaces for Superconducting RF (SRF) applications, significant progress has been made with the development of energetic condensation deposition techniques. Using energetic condensation of ions extracted from plasma generated by Electron Cyclotron Resonance, it has been demonstrated that Nb films with good structural properties and RRR comparable to bulk values can be produced on metallic substrates. The controlled incoming ion energy enables a number of processes such as desorption of adsorbed species, enhanced mobility of surface atoms and sub-implantation of impinging ions, thus producing improved film structures at lower process temperatures. Particular attention is given to the nucleation conditions to create a favourable template for growing the final surface exposed to SRF fields. The influence of the deposition energy on film growth on copper substrates is investigated with the characterization of the film surface, structure, superconducting properties and RF performance.

INTRODUCTION

RF fields having a very shallow penetration depth in the SRF material (~ 40 nm for Nb), one can deposit 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 [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 produced 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, thus the reduction of the lower critical field H_{c1} .

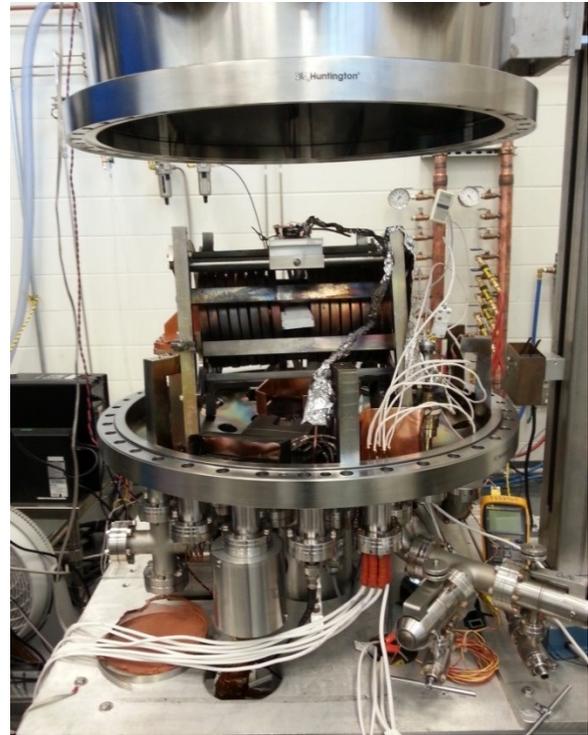


Figure 1: Inside the ECR sample coating system.

The understanding of the dependence of the final SRF performance (surface resistance, lower critical field H_{c1} ...) for Nb 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 [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 Electron Cyclotron Resonance (ECR) Nb ion source in ultra-high vacuum (UHV) (figure 1) [5]. 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. The ECR plasma produces ions with an energy of 64 eV, tunable by applying a bias voltage to the samples.

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

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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.

FILM STRUCTURE

ECR Nb films are deposited on both crystalline and amorphous Cu substrates. To achieve a crystalline interface, the native oxide layer is reduced by heating the substrate at 360 °C for 24 hours prior coating. In other hand, the amorphous Cu oxide layer can be preserved by baking and coating the samples at 200 °C or below. A series of material analyses are underway on both types of resulting Nb films and a selection is presented in this section.

Electron Back Scatter Diffraction

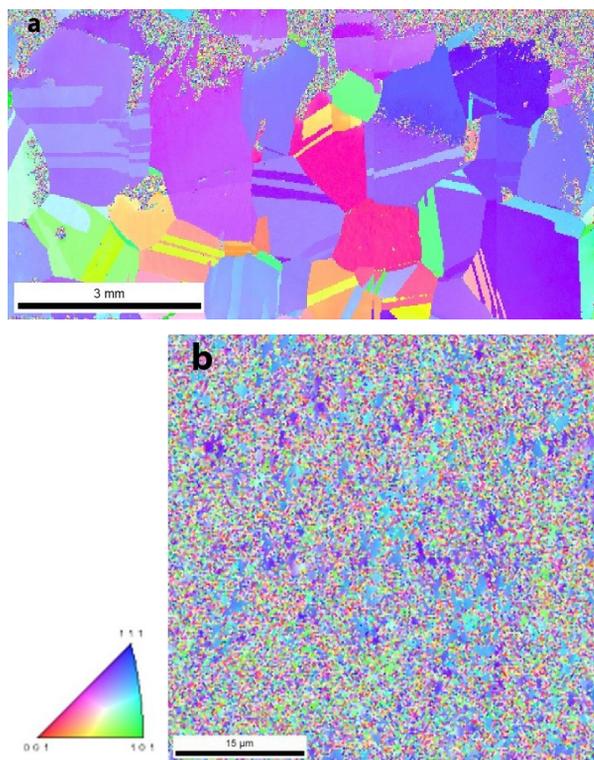


Figure 2: EBSD map for ECR films deposited on large grain Cu substrate with -120 V bias (184 eV) respectively at (a) 360 °C and (b) 200 °C (note the difference in scale).

Electron Back Scatter Diffraction (EBSD) provides microstructural information about the crystallographic nature of the films revealing grain size, grain boundary character, grain orientation, texture, and phase identity. Each data point is associated with a Kikuchi-band diffraction pattern and thus contains phase and orientation information. The EBSD data processing software can generate a variety of additional visual and analytical information. For example, Crystal orientation mapping

can be provided by using a basic RGB coloring scheme fit to an inverse pole figure (IPF). In the so-called IPF orientation map, for cubic phases, full red, green, and blue are assigned to grains whose $\langle 100 \rangle$, $\langle 110 \rangle$ or $\langle 111 \rangle$ axes, respectively, are parallel to the projection direction of the IPF (typically, the direction normal to the surface).

Figure 2 represent IPF orientation maps for films coated at 360 °C (a, crystalline interface) and 200 °C (b, amorphous Cu oxide interface) on polycrystalline Cu substrates with large crystallite sizes. Nb films coated on a crystalline interface are hetero-epitaxial, with grain size comparable to the underlying substrate (figure 2a). Nb films coated on the native Cu oxide (figure 2b) exhibit fiber growth with poorer crystal quality.

Scanning Transmission Electron Microscopy

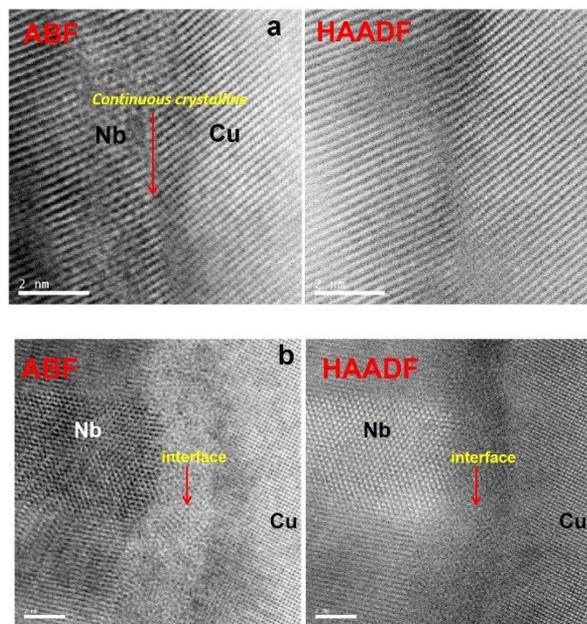


Figure 3: STEM micrographs for the Nb film/Cu substrate interface for films grown at (a) 360 °C (hetero-epitaxy) and (b) at 200 °C (fiber growth).

The atomic structure of the Nb/Cu interface for ECR films was directly imaged, with atomic-column resolution, by combining the chemically-sensitive Z-contrast imaging method (high-angle annular dark field, HAADF) with single-atom-sensitivity Electron Energy Loss Spectroscopy (EELS) in a STEM (Scanning Transmission Electron Microscope) [6].

Figure 3 shows annular bright field (ABF) and HAADF micrographs of the interface of two ECR Nb films deposited at 360 °C (a) and 200 °C (b) respectively. Coupled with EELS analyses, they reveal a continuous crystalline interface for the hetero-epitaxial film (coated at 360 °C) where the sample coated at 200 °C has an amorphous interface of about 3-5 nm between the Nb film and the Cu substrate.

CRYOGENIC MEASUREMENTS

Residual Resistivity Ratio

Residual resistivity ratio (RRR, R_{300K}/R_{10K}) measurements are conducted via the four-point probe method on a multi-sample measurement setup using calibrated CERNOX thermometers (sensitivity: 50 mK). In the case of Nb films deposited on Cu, the thick conductive Cu substrate overwhelms the measurement. To overcome this effect, the Nb films are lifted by dissolving the Cu substrate in an ammonium persulfate solution at room temperature. During the substrate removal and manipulation of the free standing Nb films, the electrical properties are likely to be degraded. Thus the RRR values obtained should be treated as lower bound values.

Table 1: RRR Values for Nb Coated at 360 °C on Cu substrates (fine grain, large grain, (100), (110), (111))

Bias Voltage	Cu fine grain	Cu large grain	Cu 100	Cu 110	Cu 111
0	51	24	35	43	50
30	57	92	78	90	80
60	--	289	82	251	85
90	115	120	57	164	98
120	118	70	83	198	138
150	82	173	99	201	245
180	97	85	--	175	62
270	101	136	55	189	72

It is found that the substrate properties and the initial growth conditions in terms of ion energy and substrate temperature are determining the final properties of the resulting Nb film [7, 8]. Table 1 shows the RRR values for hetero-epitaxial Nb films coated at 360 °C simultaneously on single crystal and polycrystalline Cu substrates. Nb (100) films grown on Cu (110) substrates have typically a higher RRR than Nb (110) films grown on Cu (100) and (111). The large grain Cu substrates have been heat treated ex-situ at 1000 °C for 12 hours and are thus fully re-crystallized. This leads to typically higher quality Nb films. To get rid of the influence of system condition variations from one coating run to another, a multi-bias sample holder has been designed and commissioned allowing the in-situ coating of samples with 8 different ion energies. A series of coating to verify the previously achieved results is under way.

Superconducting Gap Measurements

Superconducting gap measurements were performed by Point Contact Tunnelling (PCT) spectroscopy at Argonne National Lab [9] on ECR Nb films deposited on large grain Cu substrates at different temperatures. Hetero-epitaxial films have a superconducting gap similar to bulk Nb ($\Delta=1.61$ - 1.56 meV, figure 4) where fiber films grown

on the native Cu oxide have a superconducting gap of 1.3 meV.

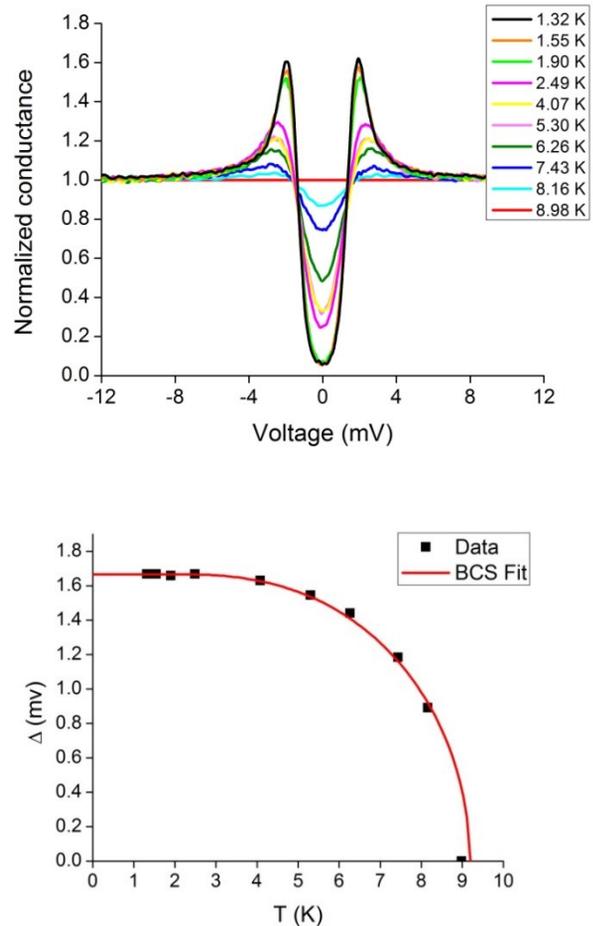


Figure 4: Superconducting gap measurement for a hetero-epitaxial polycrystalline Nb/Cu film (RRR=289).

RF MEASUREMENTS

A series of 50 mm disk samples dedicated to RF measurements have been produced and are being investigated with the 7.5 GHz TE011 sapphire loaded cavity (SIC) setup described in [10, 11]. Even though, the RF measurements need to be refined, some trend seems to emerge. Figure 5 displays the surface resistance (R_s) versus temperature for films coated with ion energies from 64 eV (no bias) to 264 eV (-200 V bias) revealing a trend of the surface resistance decreasing with increasing ion energy.

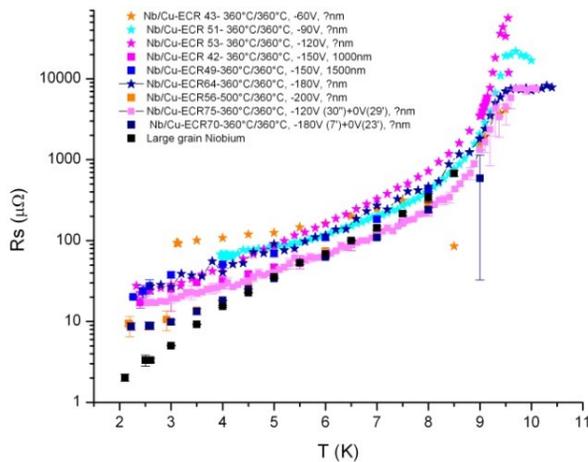


Figure 5: Surface resistance measurements for a series of ECR Nb films coated at various bias voltages and at 360 °C.

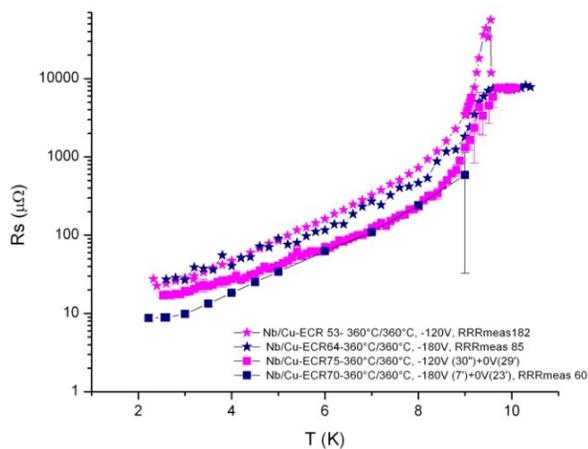


Figure 6: Surface resistance measurements for ECR Nb films coated with constant or varying ion energy during film growth.

Nucleating the film with high energy ions initially and then lowering the ion energy for the subsequent film growth by opposition to keeping the ion energy high for the duration of the coating seems also to have some effect on the RF performance. Figure 6 displays R_s curves as a function of temperature for films deposited with a constant ion energy (squares) of 184 eV (-120 V bias, pink) and 24 4eV (-180 V bias, dark blue). The second set of curves (stars) represents the surface resistance for films deposited at high energy for the initial nucleation and coalescence and then switched to 64 eV (no bias applied). One can notice a surface resistance decrease by comparison with films grown at constant high ion energy.

SUMMARY

Engineering Nb/Cu films with energetic condensation via ECR 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. Nb/Cu films can be produced with RRR and superconducting gap values similar to bulk Nb. RF performance measurements of such films reveal some influence of the incident ion energy on the surface resistance. A first attempt at engineering the film growth by varying the ion energy for the nucleation layer and the subsequent growth shows also some effect.

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