Superconducting properties and surface roughness of thin Nb samples fabricated for SRF applications

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Abstract. Using a thin Nb layer on Copper substrate has several advantages compared with the bulk Nb in construction of Superconducting Radiofrequency accelerating cavities (SRF) for particle accelerators. We were evaluating the properties of two series of Nb layers deposited on Cu substrate, mainly by determining the start of magnetic flux penetration into the sample – the first magnetic flux entry field $B_{en}$, proportional to the $B_{c1}$. The values of $B_{en}$ are compared with the surface roughness and surface morphology of the Nb layers, which have a strong influence on $B_{en}$. The surfaces of the samples were also treated by Nd:YAG laser depending on laser irradiation dose. The results of $B_{en}$ and surface roughness before and after laser polishing are compared.

1. Introduction

In superconducting radiofrequency (SRF) cavities, the surface quality of the inner wall has significant impact to performance of the cavities. The quality of the surface roughness influences the superheating field $H_{SH}$, which is a limitation factor of the cavities, therefore a great attention is paid to the surface treatment [1-2]. In the work [3] is demonstrated how the presence of defects or grain boundary induce flux penetration and cause quench of superconductivity at fields much smaller than $H_{SH}$. It is well known that surface of the material provides energy barrier against the field penetration [4-5].

Bulk niobium is the material mostly used in SRF cavities for particle accelerators due to its high critical temperature $T_c = 9.25$ K and high superheating field $\mu_0 H_{SH} = 200$ mT [6]. Creation of a thin Nb film or multilayer structures on high purity substrate may improve the performance of the cavity beyond the fundamental limits of bulk niobium [7-13]. The production of thin Nb film on Cu substrate in applications of SRF cavities provides technological and cost advantages, but it has not achieved the performance of the bulk Nb yet.

Since the thin Nb film cannot be chemically treated, great attention is paid to surface preparation of the Cu substrate. The surface quality of the Cu substrate is very critical because its roughness and morphology are replicated by the Nb growing film. However, thin Nb film can be treated by laser irradiation to reduce its surface roughness.

In this work, two series (L20 and L16) of the thin Nb films deposited on Cu planar substrates were studied. The difference between them is in used Cu substrate polishing. The L20 substrate was treated...
by chemical polishing using SUBU solution. The L16 substrate was polished by a two-step process consisting of Electropolishing (EP) and subsequently the application of SUBU solution. On both substrates 3 μm Nb films were deposited using the same deposition conditions. The substrate polishing and Nb depositing procedures were carried out at Legnaro National Laboratories (INFN).

Afterwards they were cut into 5 samples: one reference sample and 4 of them polished using different laser radiation doses. Cutting and laser polishing processes were carried out at Riga Technical University (RTU). The results of superconducting characterizations, surface roughness and surface morphology analysis were done at Institute of Electrical Engineering (IEE) Bratislava to evaluate properties of the coated Nb films before and after laser polishing.

2. Experimental

2.1. Sample production

This section describes about the production processes of the Nb films on Cu substrates (L20 and L16 samples). The production process of the samples started from OFE copper sheet (rolling machining) which were cut at CERN to sizes of 53×53 mm².

These Cu parts were then sent to INFN, where they performed surface polishing and deposition process. The copper part of the L20 sample was treated by chemical polishing using SUBU solution, what is a mixture of sulfamic acid (5g/l), hydrogen peroxide 32% (50ml/l), n-butanol 99% (50ml/l) and ammonium citrate (1g/l); the working temperature was 72°C.

L16 sample was polished by double process, first a deeply etching with Electropolishing and then by SUBU for the surface finishing. The electrolyte used for EP was a mixture of phosphoric acid and butanol in a volume ratio of 2:3 performed at room temperature. Standard polishing protocol and more details about polishing techniques are described in [14].

From a simple optical inspection significant difference are visible (Fig. 1). The SUBU sample (L20) present a mirror like surface. In the EP+SUBU sample (L16), EP created a mirror like surface with a diagonal texture due to the oxygen evolution during the EP process and the absence of a bath agitation. Consequently, SUBU process reduced the texture, but did not remove it completely.

![Fig. 1: Copper surface after the polishing treatments](image)

After Cu polishing processes were both samples coated with a thin 3 μm thick Nb film via DC magnetron sputtering at the same deposition conditions. Used parameters for the coatings of Nb thin films were: deposition temperature 650 °C, Base pressure < 9×10⁻⁸ mbar, Current density 27 mA/cm², Target power ≈ 750 W, Discharge gas Argon, Discharge pressure 5×10⁻³ mbar.

After deposition process were both samples sent to RTU, where they were cut to 5 samples of approximately 2×2 mm², one reference sample and 4 of them polished using different laser radiation doses.

2.2. Laser post treatment

The Nb surfaces of the measured samples were irradiated by Nd:YAG laser (λ = 1.064 μm, the pulse duration τ = 6 ns and intensity I = 200.0 MW/cm²) in scanning mode with 5 μm step in Ar atmosphere. Four parts from L20 and four parts from L16 samples were irradiated with four different doses of photons: D1=1.4×10²⁰ phot/cm², D2=2.2×10²⁰ phot/cm², D3= 4.4×10²⁰ phot/cm², D4=44×10²⁰
phot/cm². Thus, we have 10 measurement samples: L20 (non-irradiated), L20-D1 (irradiated by laser dose D1), L20-D2, L20-D3, L20-D4, L16 (non-irradiated), L16-D1, L16-D2, L16-D3 and L16-D4.

2.3. Superconducting properties
The superconducting properties of the Nb films were characterised using Vibrating Sample Magnetometer (VSM) option of the commercial PPMS system from Quantum Design Inc.

DC magnetization curves of the samples were measured after cooling down the sample below $T_c$ in zero applied field (zero-field-cooled conditions, ZFC). The magnetic moment ($m$) of the measurement sample was then measured in dependence on monotonically increasing applied field $B = \mu_0 H$, recording the so-called virgin magnetization curve. The applied field was oriented perpendicular to the flat face of the sample.

The main characteristic determined from the magnetization curves was the so-called first magnetic flux entry field $B_{en} = \mu_0 H_{en}$. It is the applied field at which the magnetic flux starts entering into the sample’s volume. It was detected as the field at which the virgin magnetization curve starts to deviate from the linear dependence that the virgin curve follows in the initial part starting from the zero applied field. The field $B_{en}$ was determined employing a 2% relative difference criterion, i.e. as the applied field at which the relative difference between the virgin magnetization curve and the initial linear trend reaches 2%.

The first flux entry field $B_{en}$ is proportional to the first (lower) critical field $B_{c1}$ through a geometrical factor that depends on the dimensions of the sample. The geometrical factor has not been determined, however, as the dimensions of all the magnetization measurements samples were almost identical, the relative comparison of $B_{en}$ between the samples still gives a valid information.

The cutting process created certain damage at the edges of the measurement samples, however, the same procedure was followed and the same cut-off machine was used in preparation of all the investigated samples. We thus assume the damage was comparable in all the cases and the determined $B_{en}$ values can be used to relatively compare the samples’ properties.

2.4. Nb morphological characterization
The roughness of the Nb surface layers were performed using Atomic Force Microscope (AFM). Before AFM measurement the samples were cleaned in Ultrasonic bath in liquid isopropyl alcohol (IPA) for 2 minutes, then rinsed by acetone and consequently dried by nitrogen gas. Measurements of the roughness were evaluated scanning four different spots of the Nb coated sample. Scan area was $10 \times 10 \mu m^2$. Resulting roughness was calculated as an average value of four measurements.

After AFM measurements, the measured samples were placed into Scanning Electron Microscope (SEM) to analyse the Nb film morphology.

3. Results and discussion
Critical temperature of the samples has been determined, as well. In the measurement of the critical temperature the constant magnetic field of 5 mT was applied perpendicular to the flat face of the sample and the temperature was gradually decreased from 12 K to 4.22 K. The magnetic moment ($m$) of the sample was measured at 0.1 K steps and the superconducting transition temperature $T_c$ was taken as the onset of magnetic moment change.

Critical temperatures of all investigated samples are in the range $9.3 \pm 0.1 K$. The samples treated by laser polishing did not show a change of critical temperature with respect to non-irradiated samples.

The results of first magnetic flux entry field $B_{en}$ for the Nb coated samples before and after laser irradiation are summarized in Table 1. The $B_{en}$ results of the samples show different behaviour between L20 and L16 series after laser polishing.
Table 1: First mag. flux entry field $B_{en}$ determined at 4.22 K for the Nb coated samples before ($B_{en0}$) and after laser polishing

| Sample | $B_{en}$ [mT] | $B_{en}/B_{en0}$ [-] |
|--------|---------------|-----------------------|
| L20    | 26.6          | 1                     |
| L20-D1 | 17.0          | 0.64                  |
| L20-D2 | 15.4          | 0.58                  |
| L20-D3 | 13.9          | 0.52                  |
| L20-D4 | 17.7          | 0.67                  |
| L16    | 12.8          | 1                     |
| L16-D1 | 17.4          | 1.36                  |
| L16-D2 | 18.4          | 1.44                  |
| L16-D3 | 14.1          | 1.1                   |
| L16-D4 | 16.7          | 1.3                   |

All the samples irradiated by laser from L20 series show a large decrease of the first flux entry field $B_{en}$. The relative decrease of laser irradiated samples to non-irradiated one ranges from 33% in case of sample L20-D4 up to 48% in case of the L20-D3.

In the case of L16 series, there is an increase in $B_{en}$ after the laser irradiation for all four samples. The relative increase of $B_{en}$ ranges from 10% in the case of L16-D3 up to 44% in the case of the L16-D2 sample.

We can see that critical temperatures $T_c$ of the samples were not change after laser polishing, but $B_{en}$ values were dramatically changed. So, we can assume that large change of $B_{en}$ in both series after laser polishing is primarily due to change of the Nb surface quality.

The AFM characterizations of non-irradiated and laser irradiated samples are summarized in Tab.2.

Table 2: AFM results of roughness $R_a$ compared between Nb coated samples before ($R_{a0}$) and after laser polishing

| Sample | $R_a$ [nm] | $R_a/R_{a0}$ [-] |
|--------|------------|-----------------|
| L20    | 8.8±2.1   | 1               |
| L20-D1 | 10.2±2.8  | 1.16            |
| L20-D2 | 9.0±2.7   | 1.02            |
| L20-D3 | 9.1±0.8   | 1.04            |
| L20-D4 | 7.9±1.3   | 0.9             |
| L16    | 14.9±2.1  | 1               |
| L16-D1 | 7.5±3.2   | 1.36            |
| L16-D2 | 8.3±1.5   | 1.44            |
| L16-D3 | 6.4±1.3   | 1.1             |
| L16-D4 | 8.8±2.9   | 1.3             |

The AFM results of the non-irradiated samples show different surface roughness between L20 and L16 samples. Sample L20 has average surface roughness $R_a$ of 8.8±2.1 nm and L16 14.9±2.1 nm. The higher value of the roughness for L16 sample is caused by different substrate polishing process. Diagonal texture of the uncoated substrate surface after the EP process influenced resulting roughness of the Nb surface. However, in both polishing techniques was used SUBU solution, which leads to creation of pitting on surfaces (also visible in Fig. 6 and 9) and this could lead to deviation of the roughness values.

Ratio between $R_a$ and $B_{en}$ of laser irradiated samples compared with non-irradiated one are shown in Fig. 2 and Fig. 3, for L20 and L16 respectively. On all measured values standard deviations are shown as error bars. The standard deviation of $B_{en}$ was reaches 10% level for all samples.
The AFM results of the roughness for L20 series show very little change in surface roughness after the laser irradiation. In the cases of L20-D1, L20-D2 and L20-D3 there is little increase in surface roughness 16%, 2% and 4% respectively. In the case of L20-D4 sample, there is little decrease of the surface roughness, for about 10%. However, in the surface roughness evaluation, there is standard deviation up to 25%, so we can assume that laser radiation of the L20 samples didn’t change their surface roughness or very little.

The results of L16 series show that the surface roughness decreases dramatically after the laser irradiation, also with assuming standard deviation of the roughness measurements. Decrease of the surface roughness ranges from 41% in case of L16-D4 sample up to the biggest decrease of 57% in case of L16-D3 sample.

On the Fig. 4 and Fig. 5 are shown AFM pictures of the typical surface morphology of non-irradiated samples (a) and the most irradiated D4 samples (b). The images scale size 10×10 μm².

Fig. 4: AFM images of the Nb surfaces for L20 non-irradiated (a) and L20-D4 samples (b). The Nb surface roughness \( R_a \) of the L20 non-irradiated sample is 8.8±2.1 nm, for L20-D4 it is 7.9±1.3 nm.
The surface morphology of the L20 non-irradiated sample consists of Nb polycrystallise grains with sizes ranging from 300 nm to 2 µm (Fig. 6). The surface of this sample is also covered by pits, which was caused by SUBU substrate polishing. The diameter of pits ranges from 200 nm to 5 µm.

In the case of laser irradiated L20-(D1-D4) samples, the grain boundaries become thinner and less visible. The size of grains stayed the same like in case of non-irradiated sample. The surfaces look smoother after laser polishing, but some other defects appear. All L20 laser irradiated samples contain cracks on their surfaces. The highest density of cracks appears surface of the L20-D2 sample (Fig. 7).

All four L20 laser irradiated samples also appear high-hills on their surfaces, visible in Fig. 8. This image shows the surface of the sample L20-D3, where is the largest density of the high-hills on the surface. These high-hills could be caused by boiling of Cu under the Nb layer.
Fig. 8 SEM image of the Nb surface for L20-D3 sample

The L16 non-irradiated sample shows visible grains and pitting appears on its surface (Fig. 9), similar like in the case of L20 non-irradiated sample.

Fig. 9: SEM image of the Nb surface for L16-D1 samples

The SEM image of L16-D1 structure (Fig. 10) shows that Nb surface becomes smoother after the laser irradiation and pitting mostly disappeared. All four L16 laser irradiated samples show similar surface morphology and roughness, which was also confirmed by AFM analysis.

Fig. 10: SEM image of the Nb surface for L16-D1 sample
Decrease of the surface roughness $R_a$ is most probably caused by melting of the Nb film by the laser irradiation and disappearance of pitting. Melting of the Nb film also smoothed the diagonal texture created due to electropolishing of the substrate.

4. Conclusion
Superconducting properties, surface roughness and surface morphology were investigated on 2 types of coated Nb samples. Both types consist of the same copper sheet and the same Nb deposition conditions. Difference between them was in used Cu substrate polishing: L20 sample was polished by SUBU and L16 by double process EP+SUBU. Both samples were cut to 5 samples and 4 of them were irradiated by laser at different laser doses (D1-D4).

Surface roughness inducing local field enhancements can trigger premature vortex penetration and causes decrease of $B_{en}$. The investigation shows that reduction of the surface roughness can lead to improvement of the first flux entry field $B_{en}$.

The first magnetic flux entry field of the L16 series had increased in all four cases after the laser polishing, primarily due to improved surface quality and disappearance of the pitting from the surfaces. Laser irradiation leads to decrease of surface roughness maximal from $14.9 \pm 2.2$ nm to $6.4 \pm 1.3$ nm in the case of L16-D3 sample.

In the case of L20 series, the surface roughness stayed approximately the same after laser polishing, but $B_{en}$ decreased what could be caused due to large number of defects on their surfaces after laser irradiation. Creation of the high-hills, cracks and other defects could trigger premature field penetration.

The differences between results L20 and L16 series were caused due to different polishing process of the substrate and different response to laser irradiation what confirms AFM and SEM analysis.

Laser irradiation is a very promising post-treatment to reduce the surface roughness and to increase the first flux penetration field. The mechanism of defect creation, which can cause decrease of $B_{en}$ (case of L20 series) is still under investigation.

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