NEW RESULTS ON NIOBIUM-SPUTTERED COPPER QUARTER WAVE RESONATORS

V. Palmieri, V.L. Ruzinov*, S.Yu. Stark*,
L. Badan, A.M. Porcellato, R. Preciso
F. Chiurlotto, M. Morvillo

INFN Laboratori Nazionali di Legnaro
Legnaro (Padua), Italy

Abstract

Niobium Sputter-coated Copper Cavities provide a refined solution for high performance resonators. The improved thermal conductivity of a few microns of Niobium sputtered onto OFHC Copper and the significant reduction of material costs make the sputtering solution an attractive alternative to bulk Niobium, even when heavy ion resonators are involved. Niobium sputtering in a Biased DC Diode configuration has been investigated for Legnaro OFHC Copper QWRs. Q-values of the order of 10^9 and accelerating fields over 6 MV/m at 7 Watt are routinely achievable. These results obtained investigating a simplified model of QWR without beam-ports has been replied without any problem when sputtering a real accelerating resonator complete of beam-ports and tuner.

* On leave from Moscow Steel and Alloys Institute, Moscow, Russia

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INTRODUCTION

In this paper, we describe the state-of-the-art and the results achieved over the past few years of investigation into Copper Quarter Wave Resonator (QWR) Niobium thin film sputtering.

Superconducting QWRs are required by the ALPI superconducting booster currently under construction at the Legnaro National Laboratories (LNL). The machine design calls for the installation of approx. 100 Lead-coated Copper 160 MHz QWRs for three different beta values: 20 resonators for the 0.056 beta section, 48 for the 0.11 beta section, and around 24 resonators for the 0.14 beta section.

In order to obtain INFN approval and financing for the new Legnaro superconducting LINAC, the *ALPI Proposal* (1986-87) suggested adopting Lead-coated cavities for the RF accelerating structure [1]. Experience in Lead technology in other laboratories confirmed the possibility to achieve positive results in short times and at low costs [2,3]. Moreover at that time, apart from CERN experience with entirely different geometrical shapes, Niobium sputtering onto such complex shapes as those of heavy ion accelerating cavities had never even been attempted.

Parallel to the construction of the ALPI machine however, the decision was made to also perform research into Niobium thin film sputtering onto OFHC Copper QWRs. The intention was to pass from Lead to sputtered Niobium technology as soon as it could be proven that sputtered QWRs provided better performance, and were easier to produce and above all, absolutely reliable.

It took us a few years to build and set up all the equipment required by the research program - the sputtering machine and the cryogenic and radiofrequency facilities - and the Copper substrate chemistry process. After that, we spent a not insignificant amount of time studying the sputtering of Niobium onto a dummy QWR, both investigating the plasma discharge in a Biased DC Diode configuration and characterizing the microstructural and superconducting properties of the coating obtained at different deposition parameters [4]. Towards mid-1991, we finally began making our first deposits onto real Copper cavities and our first RF characterizations at liquid-Helium.

Since then we have sputtered fifteen resonators. We provide the strategy adopted and the evolution of the results achieved below. The information we gathered after each RF measurement was used to modify the deposition parameters of the next cavity to sputter, in a continuous feedback process.

THE SPUTTERING CONFIGURATION

Existing literature and today's industrial practices identify magnetron sputtering as the dominant technique at the expense of others minor sputtering variants in modern coating technology. High deposition rates, low discharge potential, and low Argon pressure all make magnetron sources so attractive as to partially obscure the potential of all the other techniques. On the other hand, compared to the classical Diode sputtering, Magnetron sputtering appears to offer the following drawbacks:

i) Due to the difficulty of obtaining uniform magnetic confinement on the target, thickness disuniformity is inevitable in magnetron sputtering, unless the magnets can be moved during the process.

ii) Due to the presence of magnets or solenoids, Niobium cathodes must be generally cooled. This technical complication unnecessary in classical Diode sputtering is not without problems, due to the narrow cathode-substrate distances inside a QWR.

iii) It is well known that the superconducting properties of Niobium films are significantly modified by ion bombardment during deposition. In magnetrons, this effect is not uniform because ion density changes from place to place depending on the unbalance of the magnetic confinement, and when sputtering into a coaxial geometry closed at one end, such as into a QWR, this disuniformity is even greater.

iv) Variables such as plasma potential, floating potential, and ion density affect the ion bombardment of the film while it is growing and hence its superconducting characteristics. These variables can be easily measured from place to place using

Langmuir probes. This kind of plasma diagnostics is more complicated for magnetron sputtering than diode sputtering.

On the other hand, refractory metal high purity thin films can be deposited by DC Diode sputtering using a negatively-biased electrode. The bias promotes the ion bombardment of the growing film and washes out impurities that are weakly bonded to the crystalline lattice. When dealing with substrates having tricky shapes, the diode configuration adopted is generally the one providing the higher thickness uniformity, on condition that the cathode is designed to follow the substrate as faithfully as possible. Contrarily to the CERN shape (for which the requirement of uniform cathode-substrate distance is not possible to fulfil because of the narrow size of the cut-off bore [5]), the QWR geometry has the advantage of a removable bottom plate: this provides easy access for the introduction of a cathode designed to faithfully follow the substrate profile.

Moreover, our intention was to develop the simplest sputtering configuration possible so that once the right sputtering procedure and discharge parameters were found, the technology would be easily transferable to non-specialists or other laboratories who could install high performance resonators into accelerators without necessarily developing material science know-how or plasma coating engineering.

For the reasons above, our choice was for Biased DC Diode sputtering. We developed a sputtering target consisting of a simple Niobium cylinder of 2 mm thickness, with the right diameter to obtain an equal sputtering rate on both the central shaft and the surrounding cylinder [6,7]. For the deposition of the top-plate, there is no ring or special piece, and the target comes to an abrupt end at the top rim. Good coating thickness is ensured by a high focusing of electric field lines at the target rim, which results in an enormously high Argon ion impingement rate. Moreover, the ions hit the rim at oblique incidence angles, and as a result their sputtering yield is considerably greater than those of ions farther from the edge which arrive at normal incidence. This effect leads to an anomalously high erosion of the rim and a more than satisfactory top-plate sputtering rate.

No supplementary cathodes are used for the sputtering of the beam-ports. Niobium deposition inside the beam-port hole depends on the Argon working pressure and is a threshold phenomenon. The use of a high pressure value (0.2 mbar) prevents the formation of holes in the plasma.

Because the cavity is negatively biased, an additional electrode is required to provide the ground reference that is absolutely crucial for the quality of the film. We positioned the ground near the cavity bottom (fig.1) and not all along target as is traditionally done. In fact, at a working pressure of 0.2 mbar, the plasma is even more conductive and the ground reference is transmitted throughout the plasma.

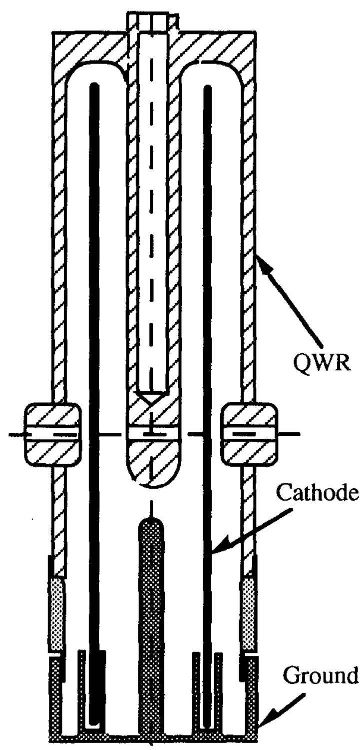


Fig. 1. The sputtering configuration we designed.

SPUTTERED QWR PROTOTYPES

While ALPI Lead-plated cavities have two holes on the resonator body for the coupler and pick-up respectively, we have transferred both to the removable bottom plate as in the JAERI QWRs. The idea was to sputter the simplest cylindrical substrate possible, and to concentrate the antennas on the removable bottom plate that is much simpler to sputter and where the lowest current density is located. During all the tests we performed, the capacitive coupler always worked excellently; therefore, such type of coupling has been accepted for all Niobium Sputtered cavities to be installed on ALPI.

In order to deal with the problems one by one, we performed sputtering onto a prototype that was identical to the real cavity, except for the fact that it had neither beam-ports nor tuner. The results we present in this paper refer to this "simplified resonator". In this way we were able to better understand the weak points in our technique and which changes in resonator geometry we needed to make in order to reach high RF performances. The beam-port problem has been dealt with separately and has been solved in different ways depending on the resonator beta value.

It is important to note that the tests performed on real high-beta cavities complete with beam-ports and tuner were equally successful: the addition of beam ports did not worsen RF performances.

Table 1 summarizes all the salient data obtained from the RF test performed on sputtered prototypes

The following data must be known to correctly interpret Table 1:

$$\Gamma = Q \cdot R_S = 29\ \Omega$$

$$H_{peak}/E_{acc} = 106\ \text{Gauss/(MV/m)}$$

$$E_{peak}/E_{acc} = 4.9$$

We found that Q versus E_{acc} very often has a pure exponential decay, $Q(E_{acc}) = Q_0\ e^{-\alpha\ E_{acc}}$. The quantity α of table 1 is the slope of decay given by the best fit of experimental data.

Figs. 2 and 3 display the Q-value at zero field and the accelerating field at 7 Watt RF power versus the date of prototype production. We can observe that better and better results were obtained test by test.

Table I

Proto-type	Q_0	E_{acc} (7W)	α (MV/m)	Curvat. **Radius**	**Notes**
Nb/Cu#1 (Jul.07.91)	4.0 x 10^7	1.25	0.11	10 mm	Plasma hole Q-switch
Nb/Cu#2 (Dec.13.91)	1.5 x 10^8	2.15	0.36	10 mm	Dark rings Small Q-switch
Nb/Cu#3 (Feb.20.92)	2.0 x 10^8	2.13	0.63	10 mm	
Nb/Cu#4 (Mar.04.92)	3.2 x 10^8	1.65	0.24	10 mm	Smaller thickness
Nb/Cu#5 (Mar.19.92)	Opening of a leak			10 mm	Argon cooling of the shaft
Nb/Cu#6 (Jul.11.92)	Test on Beamports			10 mm	
Nb/Cu#7 (Jul.16.92)	7.2 x 10^8	3.1	0.24 0.92	10 mm	Changing Slope from 1 MV/m
Nb/Cu#8 (Aug.03.92)	Test on Beamports			10 mm	
Nb/Cu#9 (Oct.31.92)	1.0 x 10^9	4.42	0.34	20 mm	Q-switch
Nb/Cu#9HP (Nov.12.92)	4.3 x 10^8	3.5	0.042	20 mm	High pressure rinsing.
Nb/Cu#10 (Jan.21.93)	1.3 x 10^9	4.1	0.48	30 mm	
Nb/Cu#11 (Feb.04.93)	2.1 x 10^9	6.7	0.21	30 mm	
Nb/Cu#12 (Feb.12.93)	9.3 x 10^8	6.1	0.16	30 mm	Deionizer failure
Nb/Cu#13 (Jun.15.93)	Test on Beamports			30 mm	
Nb/Cu#14 (Jul.11.93)	3.0 x 10^9	6.3	0.21	30 mm	Q-switch
Nb/Cu#14HP (Jul.21.93)	1.8 x 10^9	6.6	0.18	30 mm	High pressure rinsing
Nb/Cu#15 (Sep.09.93)	2.5 x 10^9	6.9	0.26	30 mm	

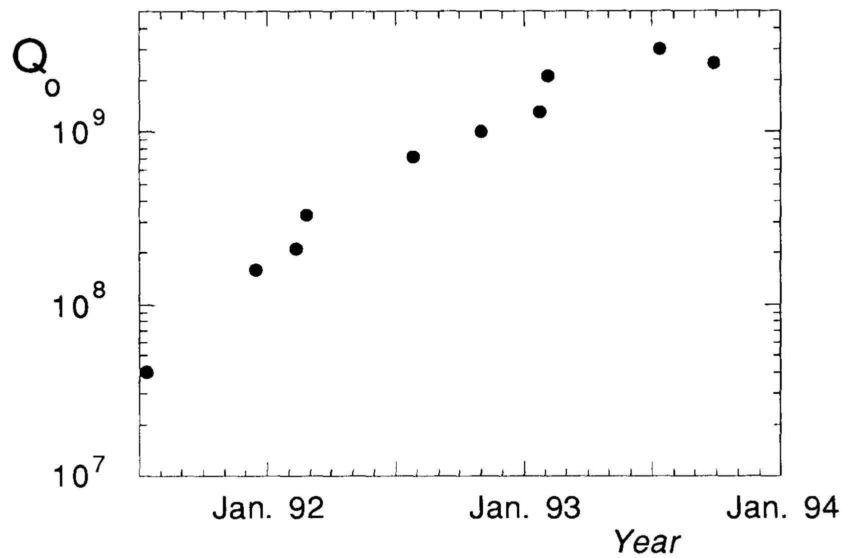


Fig. 2. Q-value at zero field versus the date of prototype production.

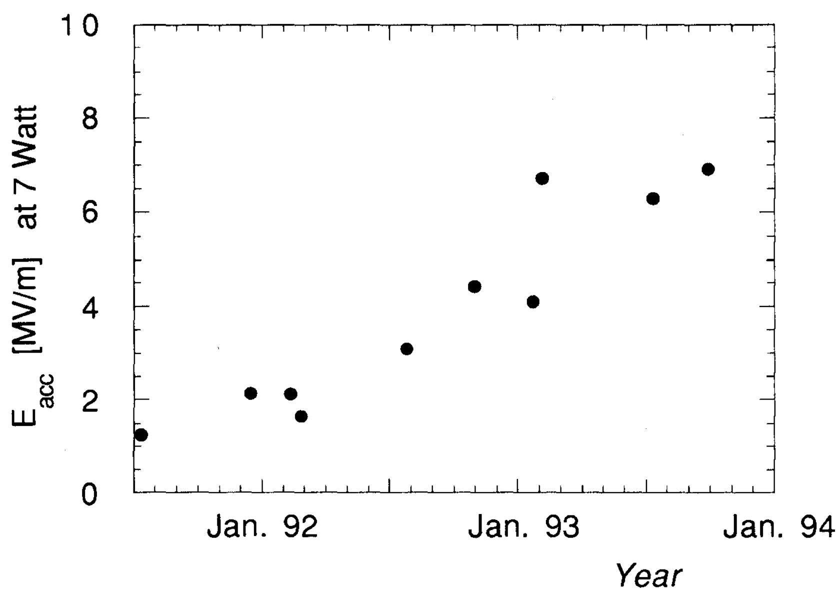


Fig. 3. Accelerating field at 7 Watt RF power versus the date of prototype production.

The history of our results up to the leap in quality that took place with Prototype Nb/Cu#11 is displayed in fig.4, where the full curves of the merit figure versus the accelerating field E_{acc} are plotted.

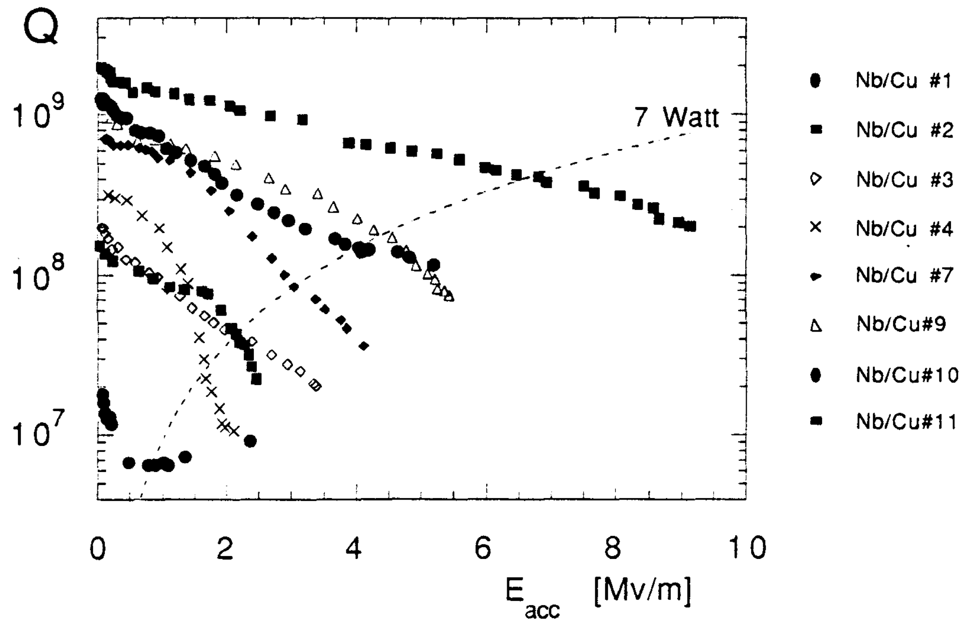


Fig. 4. The Q-value versus the accelerating field from Prototype Nb/Cu#1 up to Nb/Cu#11.

Taking as reference the best RF performances ever obtained by ALPI Lead-electroplated QWR corresponding to Qo = 2e+08, and Eacc = 3 MV at 7 Watt, we surpassed these values by optimizing the sputtering process with Prototype Nb/Cu#7. However, we achieved high quality results only with Prototype Nb/Cu#9, as soon as we made the change to a simpler resonator geometry.

An unexpected problem occurred for the first prototype, Nb/Cu#1: the Niobium coating was found to be not uniform in thickness along the shaft. Just at half length, there is a circular crown of very thin Niobium about two centimetres wide. This lack of sputtering was due to a kind of hole in the plasma between target internal surface and the central shaft. We observed that the size of such an hole was decreasing when rising the Argon pressure, up to its complete disappearance at 0.2 mbar. With the increase of the working pressure, the thickness uniformity so carefully searched for in the beginning started to fail. The discrepancy between sputtering rate onto the central shaft and rate onto the surrounding cylinder become even larger after Prototype Nb/Cu#7, when we decided to pass to a larger diameter Niobium target. Since cavity #7 in fact, sputtering rate is around 4 Å/s at the central shaft and around 2 Å/s on the outer cylinder. Of course thickness uniformity is important, but it is also a variable on which we have a certain freedom, as far as we remain within a factor two of difference and we sputter Niobium and not superconducting Nitrides.

Actually not less important than sputtering rate, there is already another parameter, the substrate temperature that introduces a big difference between sputtering onto the shaft and onto the outer cylindrical wall. Part of the power injected to the sputtering raises cavity temperature during the deposition.

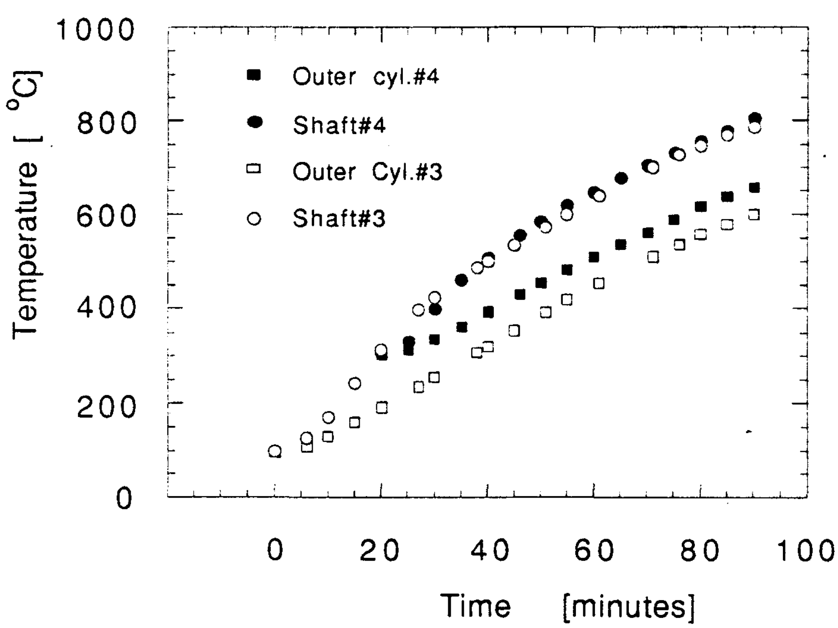


Fig. 5. The deposition temperature for Prototypes Nb/Cu#3 and Nb/Cu#4 measured in the central shaft and on the outer cylindrical wall. For the cavity #4, sputtering started with the substrate at 300°C, that corresponds to the temperature reached by a cavity in twenty minutes of sputtering starting from room-temperature.

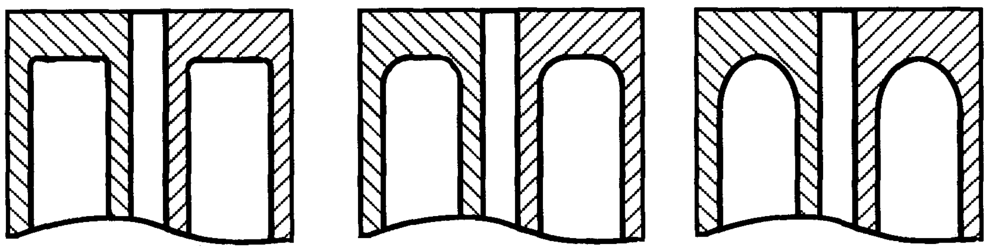
That has the advantage that the last Niobium layer deposited, just the one exposed to RF fields, has the better superconducting properties. It has the drawback, as showed in fig. 5, that temperature grows up not uniformly. In order to provide a sort of temperature regulation, we also tryed to cool the central shaft during sputtering by an Argon or by a Nitrogen flow, but the occurring of some leaks in the cooling system (Prototype Nb/Cu#5) discouraged us from using it too often.

The reason why we definitely broke the symmetry adopting a larger cathode was due to the low quality of the Niobium film at the corners connecting shaft and outer cylinder with the top plate along the whole circumference. The radius of curvature of such corners was 10 mm (fig. 6a). Two narrow dark Niobium rings, 1 cm wide, were visible in correspondence of such corners and the RRR of Niobium in such zones was not higher than 3. We observed that a 10 mm radius represented a weak point for the bias, since electric field in a sharp corner is weaker. All the impurities, expelled from film regions where the bias is active, diffuse toward places where the bias is less effective.

Sputtering tests onto small Sapphire and Copper samples located in the upper part of a dummy QWR, revealed us that this phenomenon was stronger at the outer ring rather than at the ring near the shaft. The change to a larger cathode improved somehow the film quality on the outer ring, without big effects on the inner one.

We also noticed that sputtering 5,000 Å of Niobium on these two rings, before the entire cavity deposition, gave rise to a sudden increase in RRR values of the second layer just in the two rings at issue. Prototype Nb/Cu#7 in fact was the first test providing reasonable RF performances. We have not clear up to now the real role of such buffer layer. Maybe it works as a diffusion barrier for impurities coming from the Copper substrate; maybe because quality of Niobium is poor in those corners, London penetration depth is larger than normal and a thicker film is needed; maybe due to the disordered orientation of arriving sputtered particles to the corners, a Niobium buffer layer, rather than bare Copper helps nucleation of grains of satisfactorily big size.

The goal of Qo = 10^9 was achieved only after rounding off the upper part of resonator, as sketched in figs. 6b, 6c. The radius of curvature of the connection corners of top plate was smoothed to 20 mm for Prototype Nb/Cu#9 and to 30 mm for Prototype Nb/Cu#10, where the top-part becomes completely circular.



a b c

Fig. 6. Detail of resonator geometry: a) old model with curvature radius of 10 mm; b) modifyed model with curvature radius of 20 mm; c) definitive model with curvature radius of 30 mm.

A completely rounded off top-part does not show any problem from the point of view of multipacting, moreover has the further advantages to be simple to excavate at the lathe and simple to chemical polish. After the test with the 30 mm curvature radius, we decide to adopt such a variant for the final resonator design.

Since we made the geometry change, Q-values around 2 x 10^9 and accelerating fields never lower than 6 MV/m at 7 Watt become routinely achieved. The resonator can be pushed up to 10 MV/m (corresponding to 49 MV/m of Electric Peak-field and 1060 Gauss of Magnetic Peak field) without quenching.

In fig. 7 we report a comparison of our best data on sputtering with the best data obtained: at LNL for Lead-Electroplated QWR, at LNL for Bulk Niobium and at JAERI for explosively bonded Niobium onto Copper.

Sputtered QWRs need very low time of room temperature multipacting RF conditioning, compared with Lead-electroplated resonators. Usually 2 or 3 hours of Helium-Conditioning at 5 x 10^{-5} mbar are needed to overcome emission field. Before processing the cavity, an abrupt Q-decay is found at accelerating

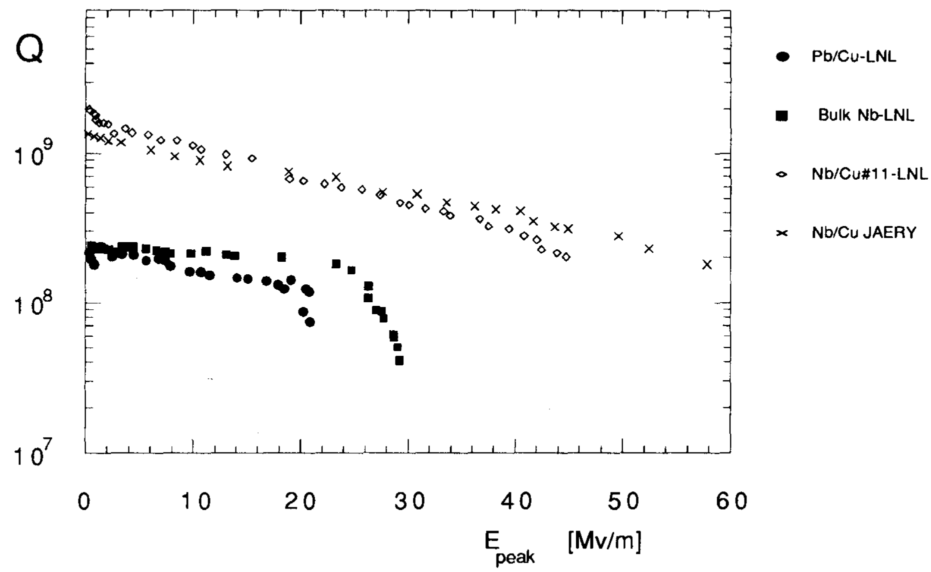


Fig. 7 Q-factor versus Electric Peak field for sputtered Nb/Cu#11 compared with results of other technologies: Lead-plated Copper at LNL, bulk Niobium at LNL, explosively bonded Nb/Cu (129 MHz) at JAERI.

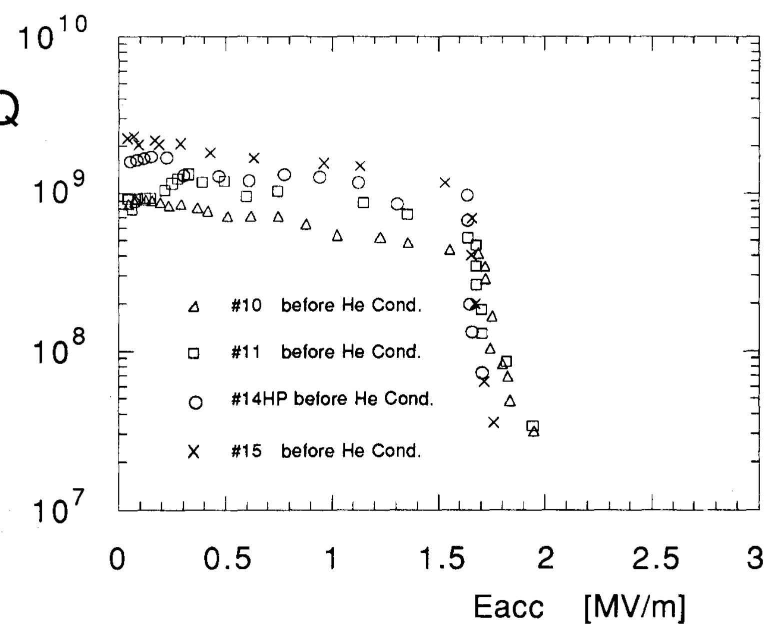


Fig. 8. Q-value versus Accelerating field, for different prototypes before He-Conditioning.

fields lower than 2 MV/m. It is worthwhile to notice that for different sputtered prototypes using the same bottom plate, the decay field was systematically the same, 1.7 MV/m. That is displayed in fig. 8; The sequence was broken when a different plate was used. That would suggest that bottom plate (maybe because of coating defects) influences the emission field mechanisms, even if low fields are located there.

As already exposed above, the resonator geometry is crucial. Nevertheless surface quality of the Copper substrate is not less important when high Q-values are desired.

After the mechanical finishing, the resonator is tumbled, electro-polished, then chemical-polished [8] and rinsed. The final rinsing is done with ultrapure water at a pressure of 100 bar. Tumbling is performed only for virgin resonators that have never been sputtered, otherwise the treatment consists of Niobium film stripping, followed by electropolishing.

We have observed that the 100 bar rinsing is extremely important especially after stripping. Generally after any chemical treatment, even the most brilliant and smooth Copper surface traps residuals of chemicals due to an imperfect washing of the surface especially in the case of tricky shapes. During the growth, impurities collected on the Copper surface may poison the Niobium film by diffusing into grain boundaries. Such a poisoning effect is more and more dangerous, the higher the substrate deposition temperature.

For Prototype Nb/Cu#12 we experienced the effect onto the film of an imperfect substrate chemical polishing. In fact during the resonator rinsing the deionizer resins failed and water conducibility inadvertently dropped. The effect was a Qo just under 1×10^9.

We have tried also the 100 bar rinsing directly onto a sputtered resonator after being characterized. The curve of Q versus accelerating field jumped down remaining parallel to the previous, maybe because high pressure water pulls away Niobium from the points where there is poor adhesion. For Prototype Nb/Cu#14, as displayed by fig. 9, after the High Pressure rinsing the Q-factor become lower, but the accelerating field at 7 Watt was increased.

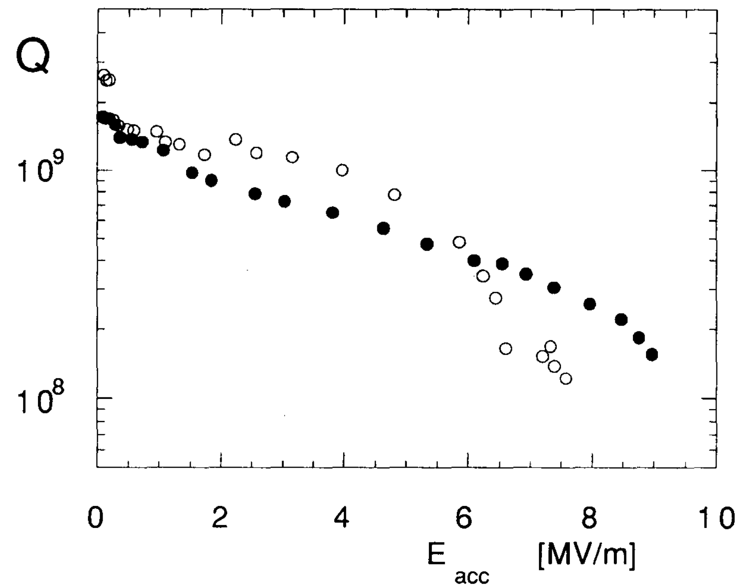


Fig. 9. Q versus E_{acc}: (o) for Prototype Nb/Cu#14, and (•) for the same prototype re-measured after High Pressure rinsing.

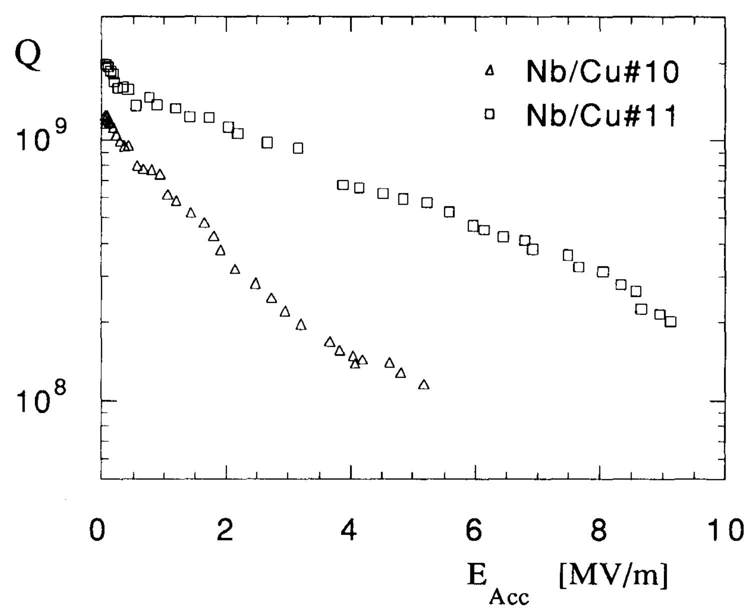


Fig. 10 Q versus E_{acc} for Prototypes Nb/Cu#10 and Nb/Cu#11. All deposition parameters were kept constant, while the coating thickness was increased of 1.5 times for the #11.

We have investigate the effect of the coating thickness on the RF performances of our sputtered resonators. In fig. 10 it is displayed the effect of increasing the coating thickness of a factor 1.5.

The two curves we report in fig. 10, as many others measured, showed a sharp exponential decay versus the accelerating field,

$$Q = Qo\, e^{-\alpha\, Eacc}.$$

The slope α passed from 0.48 to 0.21. After the success determined by the thickness increment of a factor 1.5, we insisted on the this track (see Table I), providing an increment of a factor 2 when sputtering Prototype Nb/Cu#14, and even of a factor 2.5 for Prototype Nb/Cu#15. For this last prototype we had around 8 μm of Niobium on the central shaft and around 4 μm on the outer cylinder. There is no clear indication that such big thicknesses provide further decrements of the slope α, even it seems to be a saturation effect.

Also as regards the α dependence on thickness, the problem has been not completely understood. After the fifth prototype we sputtered, in order to find a solution for the problem of "dark rings" in the cavity, we performed a long a detailed investigation on Niobium films sputtered onto small Sapphire samples plugged into the dummy QWR. One of the results of this study was an intrinsic correlation between film thickness and RRR values. This could be explained by the fact that the thicker the film is, the larger is grain size of the layer exposed to RF fields [9]. Large grain size would provide high RRR and low RF losses. Passed a certain value, a further increase of thickness would have no effect onto grain size. This would justify the above mentioned saturation.

Impurities present in the discharge would also give rise to a plausible explanation. The deposition is done in absence of presputtering. Hence all the impurities adsorbed on the cathode reach the Niobium film, that works as a sputter-ion pump. In such a way the first layer of the coating results contaminated, while increasing the thickness, the film becomes more and more pure. An other possibility is that the first film layers behave as diffusion barriers for impurities outgased from the substrate. The thicker the film is, the less impurities arrive to the surface layers. At the moment, we have no data nor evidences that can validate one mechanism in spite than another one.

The problem of the exponential Q-decay versus accelerating field is a general problem of film coated Copper cavities and its full understanding will represent a milestone in superconducting resonators technology.

Contrarily to what it happens for sputtered electron cavities, for QWRs it is evident that the exponential Q-decay is a thermal problem. Fig.11 displays the temperature raising versus the accelerating field, monitored in three different regions, the top-part of resonator, the bottom part of the outer cylinder, and the removable bottom-plate that is connected to the rest of resonator by an Indium joint.

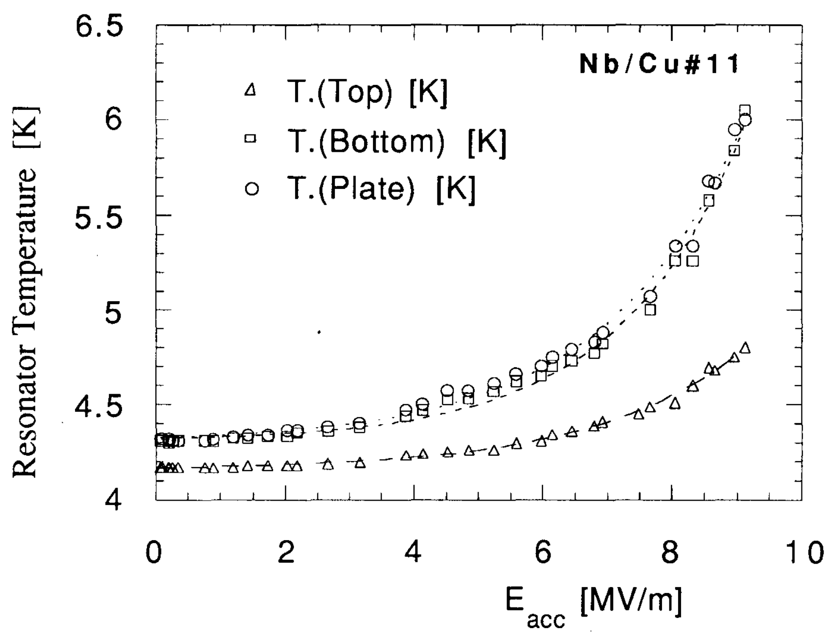


Fig. 11 Temperature dependence on the Accelerating field for three different regions of the resonator: top-part, bottom-part and bottom-plate.

The increasing of temperature suggests that the exponential slope is due to the increase with field of BCS Resistance and not of residual resistance.

CONCLUSIONS

Important progress has been achieved during last two years in Niobium sputter-coated Copper Quarter Wave resonators. Investigation of a "simplified resonator" without beam ports. has proved that high accelerating fields as high as 6 MV/m at 7 Watt of RF power can be routinely achieved. The addition of beam-ports did not change quality of results. We sputtered a real accelerating QWR (beta = 0.14) complete of tuner and beam-ports and an accelerating field of 6.7 MV/m at 7 Watt was reached as well.

The quality of the results we got and above all their reliability convinced LNL to adopt Niobium sputtered QWRs at least for the whole high beta section of ALPI LINAC.

The installation of the first cryostat containing four sputtered resonators of beta 0.14 is scheduled for mid-1994.

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