THERMAL CONTACT RESISTANCE AT THE NB-CU INTERFACE

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Abstract

Niobium thin film sputtered copper cavities are strongly limited for the application in high field accelerators by the unsolved "Q-slope" problem. In the present paper, we examine the different contributions of the niobium film, the copper substrate, the Helium-Copper interface and the Niobium-Copper Interface, proposing the hypothesis that main cause of losses is due to an enhanced thermal boundary resistance R_{Nb/Cu} at the Nb/Cu interface, due to poor thermal contact between film and substrate. So, starting from different Q vs Eacc experimental curves from different sources, and using a typical "inverse problem" method, we deduced the corresponding distribution functions generating those curves. Assuming that only a small fraction of the film over the cavity surface is in poor thermal contact with the substrate (or even partially detached), due to bad adhesion problems, we propose as a possible solution of the problem, the possibility to use higher temperatures of deposition and the adoption at the interface of a buffer layer of a material that alloys both with Copper and with Niobium.

INTRODUCTION

Superconducting Niobium sputtered Copper cavities were successful in nineties at CERN for the construction of the Large Electron Collider (LEP), and then at Legnaro National Laboratories of the INFN for the Quarter Wave Resonators (QWRs) of the ALPI heavy ion accelerator [1-2]. Recently the Nb/Cu technology has been retrieved again at CERN for the QWRs fabrication of the ISOLDE ion beam Facility [3].

Unfortunately, the use of Nb sputtered Copper Cavities in particle accelerators is not as common as desired, because, if compared to bulk Nb cavities, Nb thin film sputtered Cu cavities present a severe Q decay problem as a function of the RF accelerating field, as displayed in Fig. 1.

The Q-slope affecting thin film cavities prevents their use in any accelerator, where high fields are required.

The reason underlying the strong decay of the Q-factor versus the accelerating field in thin film cavities is still unknown and, the understanding of the reasons under the Q-drop would be the first step to solve the problem and in this

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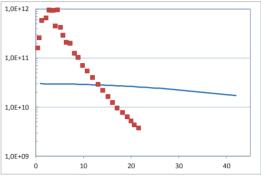


Figure 1: Typical behaviour of Q-factor versus the accelerating field for bulk Niobium cavities compared to Nb film sputtered cavities. The Q factor displayed by red squares is referred to a 1.5 GHz Nb Sputtered Cu cavity measured at 1.7 K [4]. The Q factor displayed by the blue line, is referred to 1.3 GHz bulk Niobium cavities measured at 1.8 K.

way to access at low cost, high performance thin film cavities.

A Q decay effect is also present, at a lower extent, in bulk niobium cavities and many researchers have proposed several models in order to explain the Q-slope (or Q-drop) mechanism [5], mostly trying to justify the difference between the film and bulk cavities on the basis of the lower film *RRR* [6].

One of the main mechanism on which the Q-slope effect is based is the so called "thermal feedback model" [7]. In few words, the model writes the surface resistance in Taylor expansion:

$$R_{s}(T) = R_{s}(T_{o}) + \left(\frac{\partial R_{s}}{\partial T}\right)\Big|_{T_{o}} \Delta T + O(\Delta T^{2})$$
(1)

 ΔT is the temperature difference between the inner superconducting cavity surface and the Helium bath, and it is proportional to the overall thermal boundary resistance R_B and to the rf power P_d dissipated in the cavity to sustain the accelerating field. The dissipated power depends on $R_S(T)$, being

$$P_d = \frac{1}{2} R_s(T) H_{RF}^2,$$
 (2)

where H_{RF} is the peak amplitude of the surface magnetic field. This induces a thermal feedback, since, fixed the H_{RF} value, the power leads to a temperature increase followed by a surface resistance increase and by a further power increase. The overall effect is a moderate Q-slope at low fields rapidly increasing approaching the thermal

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runaway field, where the dissipated power diverges and the cavity undergoes a global quench.

The scope of the present paper is to introduce a new, effective model based on the presence of local high thermal resistances between the Nb sputtered film and the Cu bulk cavity, due to partial film detachment in small portions of the cavity surface.

A SYSTEM MADE OF ONLY FOUR ELEMENTS: FILM, SUBSTRATE AND TWO INTERFACES

At last a Niobium sputtered Copper cavity is a system composed of only four elements: The niobium film; the copper substrate, the Helium-Copper interface and the Niobium-Copper Interface.

For years, researchers have being trying to improve the quality of the Niobium film according several techniques [8-10], last but not least the High-Power Impulse Magnetron Sputtering (HIPIMS) [11]. However no one of such efforts improved the results of Fig. 1.

The attention was also focused onto the copper substrate, °from the copper substrate was also considered.

Several attempts were done at CERN to block hydrogen from the substrate, by depositing a Titanium under-layer between Cu and Nb in order to getter hydrogen, but again no result on the Q-slope. The niobium films behaved in all respects like standard films coated on simple copper [12].

Oxygen was also suspected to diffuse from Copper into the film. However, there are experimental evidences that for Nb film cavities, an oxidized Copper surface works even better than a pure Copper surface [13]. The Q-drop was consistently larger for Nb films grown on fully oxide-free copper substrate than for standard ones, proving that the oxide layer is even beneficial rather than poisonous.

In other words, all the experimental attempts to reduce the Q-slope in thin film cavities by improving the film quality or by reducing the contamination from the substrate, essentially failed.

Referring to the Copper helium interface, nevertheless Copper has a higher Debye temperature of Niobium, and nevertheless in our previous paper we have shown that the status of the cavity external surface has also a role, the effect of the Kapitza thermal resistance between the cavity external wall and the He bath should be, in principle, negligible.

In this paper we will analyze in detail the thermal boundary resistance R_B at the Nb/Cu interface, showing that in Nb/Cu thin film cavities, thermal feedback effects could be relevant in respect to bulk cavities.

THE NB/CU INTERFACE

Before proceeding in our analysis, it is important to underline that Nb/Cu cavities realized by explosively bonded Nb/Cu bilayer [14] give excellent results (Q factors over 10^{10} and maximum accelerating fields close to 40 MV/m). Now, the differences between a Nb clad Cu

cavity and a Nb sputtered Cu cavity lay in only two aspects: the purity and thickness of the Niobium and the quality of Nb/Cu interface.

In the above mentioned hypothesis that any improvement of the film quality does not result in an improvement of the Q-slope, the full penetration bonding of the Nb clad Cu could play a key role, whenever compared to the weaker adhesion of a sputtered Niobium film onto Copper.

Moreover, there is no miscibility range between copper and niobium in the equilibrium phase diagram in Fig. 2 [15] at the temperatures usually adopted for sputtering. Indeed, the Nb-Cu system is considered as a classical example for non-miscible systems.

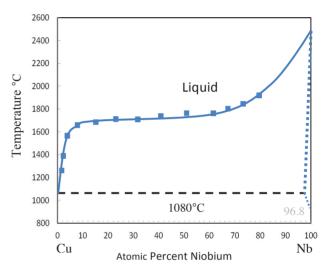


Figure 2: The Cu-Nb phase diagram, after D.J. Chakrabarti and D.E. Laughlin [15].

The problem of the Nb film adhesion on a Cu substrate has been poorly investigated and not well understood. In some cases the Nb film can peel off immediately after the sputtering and, when this happens, the fault is generally attributed to a non-perfect chemistry treatment of the copper substrate. In some other cases, for the author's experience, the Nb film can partially peel off even after tenth of years, and this can be attributed to stress release inside the film.

Other similar evidence is given by the fact that film peeling was found in some 352 MHZ 4-cell elliptical cavities when dismounted from LEP at CERN [16] several years after of their operation.

Moreover also Nb/Cu cavities deposited by filtered cathodic arc plasma were not successful, because the Nb film was randomly peeling [17-18].

The poor Niobium copper adhesion could also explain the experimental result found at CERN that was above mentioned, i.e. that the Nb film sputtered onto an oxidized copper surface gave performances systematically better in respect to films sputtered onto onsite pre-sputtered oxide-free copper. This result could be consistent with the hypothesis of a monotectic reaction

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occurring in the Cu-Nb system induced by oxygen impurities [19].

In conclusion we consider that the problem of film poor thermal contact to the substrate could be far more important that commonly believed.

THERMAL MODEL FOR THE NB/CU INTERFACE IN THE PRESENCE OF ADHESION PROBLEMS

Modeling the heath flow between the Nb film and the Cu substrate is far from being trivial. Several models were developed in the Sixties [20-21] to describe the situation of two solid sheet surfaces set in contact in vacuum with heath flowing normally from one body to the other. The two solid surfaces are assumed to touch only in a limited number of spots (n contact points per unit area) and the heath conductance is treated as a point contact conductance. In this case a contact thermal boundary conductance h_c is defined through the relation:

$$\dot{Q} = h_c \Delta T_c \tag{3}$$

where \hat{Q} is the heat flow per unit area and ΔT_c is the rising temperature gap at the interface.

For a regular matrix of multiple contacts, if a low conductance h_c can appear between two nominally flat surfaces in contact under vacuum, and since also the Kapitza contribution is in serial to the contact resistance, at low temperatures the overall Nb/Cu thermal boundary conductance $h_{Nb/Cu}$ will be given by:

$$h_{Nb/Cu}^{-1} = (h_c)^{-1} + (h_k^{eff})^{-1}$$
(4)

and the effective Kapitza contribution is $h_k^{eff} = h_k \frac{A_{eff}}{A}$,

that represents the contact occurring only over an effective area $A_{\rm eff}.$

Though the model discussed above was developed for two metal surfaces in contact, a physical situation that is somewhat different from that of a film deposited on a substrate, nevertheless it gives clear indications that a loose interface physical contact of different origins can lead to high values of the thermal contact resistance, strongly varying along the contact surface area.

The occurrence of high values of $R_{\rm Nb/Cu} = 1/h_{\rm Nb/Cu}$, the thermal boundary resistance between Nb and Cu in some spots of the cavity surface due to the weak contact of the Nb film to the Cu substrate, as described above, is in our hypothesis, the main factor determining the high Q slope observed in Nb thin film coated Cu cavities.

EFFECT OF THE NB/CU INTERFACE ON THE QUALITY FACTOR IN THE PRESENCE OF ADHESION PROBLEMS

In Nb bulk cavities (sheet thickness d) the thermal resistance is due to the Nb conductance (k)and the Nb/HeII Kapitza resistance, i.e. $R_B=d/k+R_K$ The Kapitza

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resistance R_K strongly depends on the cavity external surface morphology, ranging approximately in the interval 1-10cm²K/W at 1.8K, dominating the system thermal resistance for high RRR cavities [22-23].

For thin film Nb/Cu cavities the overall thermal boundary resistance should be $R_B=d_{cu}/k_{cu}+ d_{Nb}/k_{Nb}+ R_K$ + $R_{Nb/Cu}$. The first term has a value of 7E-2cm²K/W; the second of 4E-3cm²K/W; the third term is the Kapitza resistance at the Cu/HeII interface. The literature data at 1.8K give $R_K=2-4$ cm²K/W, i.e. in the same range of the Nb/HeII Kapitza resistance values. As discussed in the previous Section, if adhesion problems at the Nb/Cu interface are not considered, the thermal contact resistance should be:

$$R_{Nb/Cu} = 1/h_k \approx 0.3 cm^2 K/W$$

and would be negligible in respect to the Cu/He Kapitza resistance.

However if we will have a loose contact at the Cu-Nb interface, the effective Kapitza contribution $h_k^{eff} = h_k \frac{A_{eff}}{A}$, occurring over an effective area A_{eff}, must be taken into account.

However if we consider that the Nb film is indeed weakly bonded to the Cu substrate at least in very limited cavity surface portions, the Nb/Cu boundary resistance can become pretty high in those areas and an important value of ΔT can appear at the Cu-Nb interface, fully dominating the heath conduction.

In this hypothesis, locally, the temperature distribution along the direction normal to the cavity surface (in a "one-dimensional" approximation) is reported.

The local value of $R_{Nb/Cu}$ will not be constant over the cavity surface : indeed, in the simple model presented in the previous paragraph, it will depend on the number (n) and dimension (c_m) of the effective thermal contact spots in the partially detached film regions, that will have some statistical distribution over the cavity surface.

However, as foreseen already by the thermal feedback model, if the adhesion of the film is not perfect in some areas, a ΔT at the Nb-Cu interface will immediately increase the temperature of the film, and since the BCS resistance is temperature dependent, the rf losses of the film will increase, determining a further ΔT_i , that again will give rise to a further increase in rf losses, that will produce a smaller ΔT_i , and so on, determining a thermal runaway of the film as described by Eq. (5). Indeed in the dirty limit approximation and for T < T_C/2,

$$R_{S}(T + \Delta T) = \frac{A\omega^{2}}{T_{0} + \Delta T} \exp\left[-\frac{\Delta_{0}}{K_{B}(T_{0} + \Delta T)}\right] + R_{o}$$
(5)

for relatively small values of ΔT it is $\Delta T = R_B P_d$ where R_B is the overall thermal resistance (from the inner cavity surface to the He bath) and P_d is the rf power dissipated per unit area at the inner cavity surface, given by $P_d = \frac{1}{2}R_s(T_o)H_{RF}^2$. Therefore:

$$\Delta T = R_B \frac{1}{2} R_s(T) \left(\frac{k}{\mu_o}\right)^2 E_{acc}^2 \tag{6}$$

(H_{RF} is proportional to the accelerating field E_{acc} through the relation $H_{RF} = (k / \mu_o) E_{acc}$, with k= 4,5mT/(MV/m) [24]).

The converging value of $R_S(T+\Delta T)$ versus E_{acc} can be easily numerically calculated. So, after a few iterations, as in Fig. 3, the film will immediately arrive to an overheated steady state, that will more and more depend by the thermal boundary resistance for increasing R_B values.

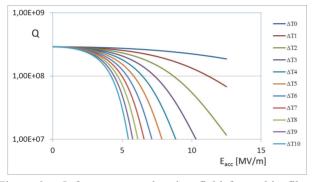


Figure 3a: Q-factor vs accelerating field for a thin film Nb/Cu 6GHz cavity at T=1.8K, after 10 interactions. The parameter used for calculation were $R_0=0.8\mu\Omega$; $R_B=6cm^2K/W$; $A=6e-3\Omega$; $\Delta_s/K_s=17,5K$;

However in the hypothesis of local area film detachment, it is very improbable that the "weak areas", were all of the same size. It is very much probable instead that they follow a statistical distribution over the whole cavity surface. We will call $f(R_{Nb/Cu})$ the statistical distribution function of the $R_{Nb/Cu}$ values, that will satisfy the following conditions:

$$\int_{0}^{\infty} R_{Nb/Cu} f(R_{Nb/Cu}) dR_{Nb/Cu} = \overline{R_{Nb/Cu}}$$
(7)

where $\overline{R_{Nb/Cu}}$ is the average value of the Nb/Cu thermal boundary resistance over the cavity surface.

Of course the integral of the distribution function will be normalized to 1.

$$\int_{0}^{\infty} f(R_{Nb/Cu}) dR_{Nb/Cu} = 1$$
(8)

For Nb/Cu cavities we can write $R_B=R_K+R_{Nb/Cu}$ with R_K being the value of the Cu/He Kapitza resistance at the temperature T_o and $R_{Nb/Cu}$ the value of the Nb/Cu contact resistance. Since $R_{Nb/Cu}$ changes over the cavity surface as described by the statistical distribution function $f(R_{Nb/Cu})$, the average value of the surface resistance, R_s that will determine the Q value will be given by the integral equation

$\overline{R_s(T_o, E_{acc})} = \int_0^\infty R_s(T_o, E_{acc}, R_B) f(R_{Nb/Cu}) dR_{Nb/Cu}$ (9)

This indicates that the Q(E_{acc}) curve in Nb/Cu cavities will critically depend on the $R_{Nb/Cu}$ distribution function f($R_{Nb/Cu}$) that would depend on the distribution over the cavity surface of the quality of the effective thermal contact spots in the partially detached regions.

However from the experimental Q(E_{acc}) curve we directly have $\overline{R_s(T_o, E_{acc}, R_B)}$ and then by inverting Eq. (9) (that belongs to the class of first type Fredholm integral equations), we can infer the statistical distribution function f(R_{Nb/Cu}). This is indeed a classical "inverse problem" [25].

In Fig. 4 we present one example of fitting procedure based on the solution of Fredholm integral of the first type. The data points at T=1.7K refer to the best Nb/Cu 1.5 GHz cavity ever measured CERN [6]. The dashed curve represents the calculation, the fitting procedure starting, from the parameters reported in Table 1.

Table 1

To	$A\omega^2$	Δ_0/K_B	R ₀	R _{sn}	R _K
1.7 K	$2*10^{-5}$	18.4 K	$0.8n\Omega$	0.005Ω	$3 \text{ cm}^2 \text{K/W}$
	Ω/K				

The corresponding function $f(R_{Nb/Cu})$ is :

$$f(R_{Nb/Cu}) = 1.52 \cdot 10^6 R_{Nb/Cu}^{-(6.21-0.225 \ln R_{Nb/Cu})}$$
(10)

and it is reported in Fig. 5. The function is only reported for $R_{\rm Nb/Cu}$ values above $250 {\rm cm}^2 {\rm K/W}$: again lower $R_{\rm Nb/Cu}$ values would affect the Q-slope above the maximum measured field ($E_{acc}=22 {\rm MV/m}$), so that the function f cannot be determined below that value. In this case the precise value assumed for the Cu/He Kapitza conductance is unessential, since the thermal effects are much less efficient for low frequency cavities and the Kapitza resistance alone in this case would determine no observable Q-slope effect in the considered field range.

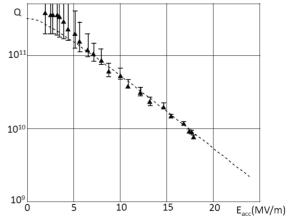


Figure 4: Q-factor vs E_{acc} for a thin film Nb/Cu 1.5GHz cavity at T=1.7K realized at CERN [4]. The data are the experimental points of the highest curve in Fig. 2 for E_{acc} >2MV/m. The dashed line represents the data fitting using the distribution function $f(R_{Nb/Cu})$ in Fig. 5.

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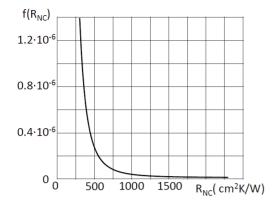


Figure 5: Distribution function $f(R_{Nb/Cu})$ got by inverting Eq. 9 using as input data the Q-E_{acc} data reported in Fig. 4.

DISCUSSION AND CONCLUSIONS

The integral I under the represented $f(R_{Nb/Cu})$ curve in this case is extremely low (I=0.0003), and it is indeed sufficient to assume that 0.03% of the Nb film in bad thermal contact with the copper substrate (or o partially detached from it) significantly contributes to the observed Q-slope.

The strong Q slope at low fields, within this model, is linked to the high $R_{Nb/Cu}$ values "tail" of the statistical distribution function. High values of $R_{Nb/Cu}$ imply locally high thermal resistance (R_B) values and therefore low local quench fields. Increasing the field, the Nb film areas in loose contact with the Cu substrate will be gradually driven into the normal state, characterized by a high surface resistance, so that the typically high Q-slope of the Nb/Cu cavities is simply due to this progressive "micro-quench" process. Of course these phenomena are typically of the Nb/Cu cavities and cannot occur in bulk Nb cavity.

Therefore in the hypothesis of a point-like lack in adhesion of the Niobium film to the Substrate, the authors think it is worthwhile to invest in the improvement of that interface. The main strategies on which we desire to put attention are: i) the increase of deposition temperature up to the cavity mechanical rigidity limit, and ii) the deposition of an intermediate buffer layer between Copper and Niobium.

Both strategies have been preliminary studied at LNL of the INFN for the preparation of Nb sputtered Cu 6 GHz cavities, giving promising results. In particular the deposition of a sub-micrometric intermediate layer of Palladium between Copper and Niobium has given a little, but encouraging, increase of both Q-factor and accelerating field, as displayed in fig. 6. This result is absolutely preliminary, but it encourages us to further proceed toward a focused investigation.

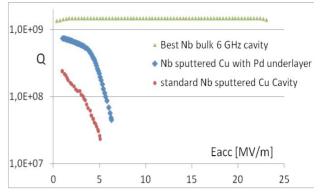


Figure 6: - Q vs E_{acc} at 6 GHz, for a standard Nb sputtered Cu cavity(red circles), for a sputtered Cavity with a Pd underlayer, and for comparison the best result obtained for bulk Nb at 6 GHz.

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