HEAT TRANSFER AT THE INTERFACE BETWEEN NIOBIUM AND LIQUID HELIUM FOR 6 GHZ SRF CAVITIES

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Abstract

Cavity Thermal Boundary Resistance is something extremely complex and not completely understood by the theory. Often identified with the Kapitza resistance or with the Khalatnikov acoustic phonon mismatch at the interface metal-liquid Helium, it depends on so many different and uncontrolled parameters, that its interpretation is not covered by a complete and exhaustive treatise of the phenomenon. Therefore, 99%, or even more, of the literature about superconducting cavities worries about the cavity interior, imagining every possible surface treatment inside the resonator, while almost nothing is reported on treatments applied to the exterior. In the authors' opinion, there is a lack in experimental data analysis just due to the fact that the cavity is often considered as a whole adiabatic entity interacting only with RF fields. On the contrary, the cavity is immersed in liquid Helium and the cavity behavior cannot prescind from its thermal properties. Indeed in the normal state He-I has poor thermal conductivity and high specific heat. Moreover the heat exchange at He-II obeys to further mechanisms besides the phonon mismatch. Then, driven by the hypothesis that thermal losses are dominant for ultraclean cavities, we have collected a plethora of surprising experimental results.

INTRODUCTION

The main idea at the basis of this work can be resumed in the following question: let us suppose we have an ideal cavity, made by a perfectly homogeneous mono-crystal of extremely high purity Niobium with no trapped magnetic flux, no losses on the joints, no any parasitic problem. Will then the cavity have zero residual resistance? In other words will be there any contribution due to a bad thermal exchange with the Helium bath?

A partial answer to this question comes from the analysis of the heat exchange of the Niobium surface to the Liquid Helium, limited by the appearance of a thermal boundary resistance at the Nb-Liquid He interface, and giving rise to a temperature difference ΔT_s.

Two potential contributors determine such a temperature discontinuity:

i) A thin He fluid layer of thickness d into which the heat diffuses. The temperature difference across the layer, ΔT_f, is determined exclusively by heat diffusion in the bulk fluid.

ii) A truly interfacial temperature difference ΔT_k occurring within a few atomic layers of the solid-helium boundary and attributed by Khalatnikov [1] to the mismatch of phonon heat transport between the two media.

This latter mechanism is referred to a quantity called Kapitza Conductance defined as

$$h_{K_0} = \lim_{\Delta T_s \to 0} \frac{q}{\Delta T_s} \qquad (1)$$

having a strong T^n temperature dependence with n equal to 3, but more often in literature [2-5] n is found to vary between 2 and 4.

Khalatnikov model assumes that only a fraction of thermal phonons impinging on the interface from either side is transmitted. Nevertheless the main part of the experimental results show higher Kapitza conductance values than predicted by Khalatnikov's Theory, suggesting additional mechanisms through which thermal energy is transmitted.

Kapitza Conductance is however a quantity depending on the interface between the 2 materials: not only Niobium, but also the liquid Helium wetting Niobium. Hence it is natural to ask the following question: Does the Residual Resistance, R_{RES} depends also on Liquid He rather than only on Nb material? In other words, if we cooled the cavity in ^{3}He rather than in ^{4}He, should we wait a different R_{RES}?

THE EXPERIMENTAL MEASUREMENT OF SURFACE RESISTANCE VERSUS TEMPERATURE

One of the most measured quantities worldwide measured by the SRF community is the Surface resistance as a function of the reciprocal of the Temperature $R_S(1/T)$ Nothings is more sure and well-established in Superconducting RF theory and practice than the famous statement

$$R_S (T) = R_{BCS}(T) + R_{RE} \qquad (2)$$

where for $T<T_C/2$, it can be shown that R_{BCS} can be approximated by the "modified Arrhenius exponential"

$$R_{BCS}(T) = \frac{A\omega^2}{T_0}\exp\left[-\frac{sT_C}{2T}\right] \tag{3}$$

being **s** the Strong Coupling factor, generally found equal to 3.8 for Niobium.

It must be however taken into account that the above mentioned equations hold only at very low accelerating field, i.e. at almost zero RF power. But a measurement at almost zero RF power is experimentally rather impossible. Therefore, a second question arises: When $R_S(1/T)$ is measured at low temperatures, it is more correct to acquire the experimental points at fixed accelerating field or at fixed RF power? In better words, which one of the curve is the most useful one? The $R_S(1/T)$ curve measured at constant W or the $R_S(T)$ at constant accelerating field, E_{acc}?
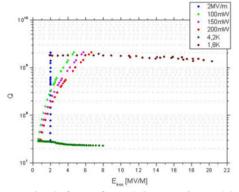

Figure 1: The Q factor for a 6 GHz cavity at 4.2 K and at 1.8 K, measured at constant accelerating field (2 MV/m) and at 3 different RF power values (100 mw, 150 mW and 200 mW).

Both type of measurement are shown in Fig. 1. Theoretically both of them should give coincident curves whenever the Q-factor is independent of accelerating field, E_{acc}. A difference arises for non-ideal curves of Q-factor versus field.

So let us consider the system of a Niobium wall facing the oscillating electromagnetic field on one side and the liquid Helium bath on the other, as in fig. 2.
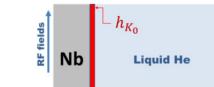

Figure 2: The Niobium cavity wall, facing the Electromagnetic RF fields at one side and the liquid Helium at the other side.

From a thermodynamic point of view, however the two different RF measurements displayed in fig. 1, at constant accelerating field or at constant power, correspond to two totally different situations: a) moving from 4.2 K to 1.8 K at constant field, it means that we acquire a serial of points of decreasing temperature and decreasing RF

power; b) moving to low temperatures instead at constant RF power, it means that we will decrease simultaneously temperature and field.

However unless of considering Magnetic/Electric field dependent problems, the constant W measurement is the only possible choice, if we desire to observe any thermal problem connected with the Kapitza conductance, by varying no other significant parameter different than temperature.

Therefore if we measure the Q-factor at different temperatures but at constant power, we observe (fig. 3) the presence of an anomaly at the lambda transition between He-I and He-II.
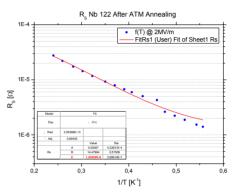

Figure 3: The $R_S(1/T)$ curve useful for separating the residual resistance from the BCS term. Around 2.18 K it is visible an anomaly.

THE JUMP OF SURFACE RESISTANCE AT THE LAMBDA POINT

After a deeper exam of the SRF technical literature, we realized that the jump discovered by the authors for 6 GHz, it was omnipresent in published papers [6], appearing both at low and at high frequency, although not recognized important.

However after a more accurate measurement across T_λ and a detailed analysis of the phenomenon, it comes out (fig. 4) that the surface resistance jump is extremely well defined and of non-little importance.
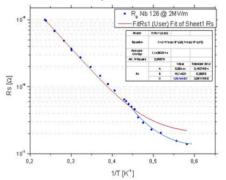

Figure 4: If the Surface resistance is measured with great accuracy, a clear step appears around T_λ, showing a lower value of the residual resistance (blue fitting line).

Figure 4 shows that if no any step would be present at T_λ, probably $R_S(1/T)$ would saturate at low temperatures

to a higher value than the one found experimentally. That means that the residual resistance is affected by the nature of the surrounding liquid Helium. This is the answer to the initial question about the possible cavity performances at 1.8 K but in liquid ^{3}He. Therefore if $R_S(1/T)$ has a transition just at 2.18 K, this happens because Liquid Helium is changing its thermal properties. As a consequence the Physics of thermal exchange cannot be neglected when interpreting the experimental data collected.
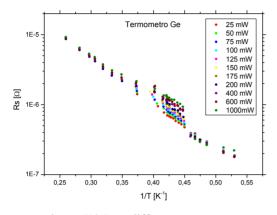

Figure 5: The $R_S(1/T)$ at different RF powers. It can be easily observed that the step at T_λ depends on power.

Moreover $R_S(1/T)$ depends on the RF Power W and figure 5 clearly shows that dependence. The more we inject RF power into the cavity, the higher the jump at T_λ will be, because it is changing the slope of the Arrhenius exponential. As far as the RF power is decreased, the jump becomes less and less visible. That explains why all the curve at Constant Accelerating field approaching the zero almost never report that jump.

In literature, this instability at the liquid helium lambda point, when noticed, sometimes appears treated simply with the data simply masked in proximity of the transition. This is substantially wrong and it brings to an overvaluation of the strong coupling factor s.

Let us consider indeed the usual realation for the BCS Surface Resistance, where T_0 is the temperature of the Helium bath,

$$R_{BCS}(T_0) = \frac{A\omega^2}{T_0} \exp\left[-\frac{sT_C}{2T_0}\right] \qquad (4)$$

Then in case of a Temperature difference ΔT at the interface of Niobium with liquid Helium, the equation becomes

$$R_{BCS}(T_0 + \Delta T) \approx \frac{A\omega^2}{T_0} \exp\left[-\frac{sT_C}{2(T_0+\Delta T)}\right] \qquad (5)$$

or simply

$$R_{BCS}(T_0 + \Delta T) \approx \frac{A\omega^2}{T_0} \exp\left[-\frac{sT_C}{2T_0}\left(1-\frac{\Delta T}{T_0}\right)\right] \qquad (6)$$

The quantity **s** is the true strong coupling factor of the Niobium and the relation (6) is the one that should be used when trying to fit a Q vs E_{acc} curve taken at constant power, when the T_λ jump is visible.

To mask the experimental points at the T_λ instability in order to use the equation (1) instead than equation (6) will then give a wrong value s^1, differing from the real **s** value of Niobium by eq. (7).

$$s^1 = s\left(1 - \frac{\Delta T}{T_0}\right) \qquad (7)$$

Moreover, since ΔT is dependent on RF power, the error on strong coupling factor evaluation is higher and higher, the higher is the RF power injected into the cavity. In other words, the $R_S(1/T)$ should be measured or at constant E_{acc}, but at almost "zero RF power" by means of extremely sensitive and not conventional instrumentation, or at constant W, and in such a case it is interesting to acquire different curves of $R_S(1/T)$ at several RF power values.

If we choose this second approach, we will measure the Surface Resistance as a function of Temperature and as a function of RF power. Then we can plot the surface resistance in a 3-dimentional graph of the type of Fig. 6.
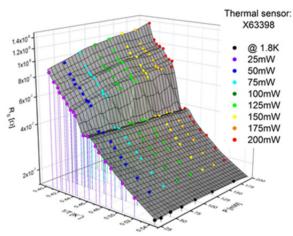

Figure 6: The tridimensional plot of Rs versus the reciprocal of temperature T and versus the RF injected power W. This 3D graph shows very clearly the transition from He-I to He-II.

The relevant information in the 3-D plot however appears also in two dimensions, if we plot Q versus E_{acc} at different values of W as in fig. 7.

Figure 7 is an extremely explicative graph. Indeed not only it clearly shows how the transition from He-I to He-II increases with power, but also contains a hidden information. Let us look the Q-factor at He-I just before the transition. It is clearly visible that the slope of the Q decay versus accelerating field is suddenly changing between 200 and 400 mW. The Q-factor decays quadratically with the Accelerating field, up to a certain level of critical power, and once overcome it the slope decreases becoming linear.
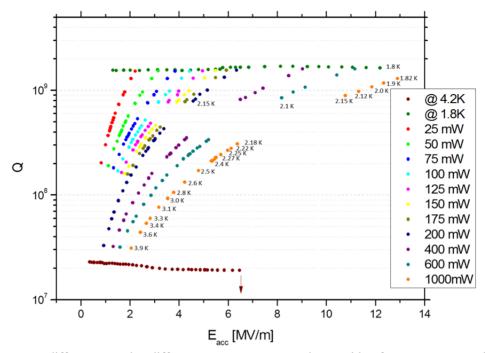
Figure 7: Q versus E_{acc} at different W and at different temperatures. Here the transition from He-I to He-II is even more clearly visible.

What kind of Physical mechanism can ever explain a dissipation that at low field is rather severe, then at higher power flux becomes less important? The answer there is and it is well known in cryogenics: at low RF power, the quadratic decay suggests the thermal nature of such dissipation problem. In that case, at a certain critical power the Helium boiling on the cavity external surface is started and the bubbles formation helps in removing heat from the surface. He-I has a poor thermal conductivity, but a great specific heat. He-II is just the opposite. So the convective motion promoted in the boiling regime by bubbles can be a very efficient heat removal mechanism in He-I.

As displayed by fig. 7, the presence of a critical power at which it is located the slope change of the Q vs E_{acc} appears at any temperature below Lambda Transition. Then in the hypothesis of the heat removal due to detachment and migration of the Helium bubbles nucleated on the Niobium external surface, it is natural to ask next question: Is any trace of this effect observable also in He-II?

The first answer to this question would be negative, since He-II has perfect thermal conductivity, no specific heat. But we must keep into account that this is true only at zero Kelvin. The operational temperature of 1.8 K is very close to the 2.18 K of T_λ, and in the two fluid model of superfluidity, $\frac{\rho_n}{\rho} = \left(\frac{T}{T_\lambda}\right)^{5.6}$, so at 1.8 K the density of normal fluid is still 34%, and it is not negligible at all.

Actually the literature is full of examples of suspect deviations from the standard quadratic decay, including the graph of the best CERN Nb sputtered Cu cavities replot by K. Saito that indicated such deviation with the question "are we really sure that this is really

multipacting?" In our hypothesis, this inflection point could be the effect of He boiling on the Niobium external surface.
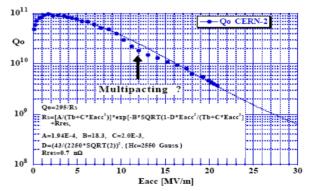

Figure 8: The Q factor versus accelerating field for the best Nb-Sputtered Cu 1,5 GHz cavity fabricated at CERN [7] and replot by K. Saito [8] that suspected the flex in the curve could not be caused by multipacting.

ACTING ONTO THE CAVITY EXTERNAL SURFACE

So in the hypothesis that the Thermal Boundary resistance is a limitation, we have tried to act on the external surface of the cavity in order to increase the cavity performances.

The first action then consisted in RF testing a 6 GHz Nb cavity at low temperatures , then extracting it from liquid Helium, warming it at room temperature and cycling it down to 1,8 K without breaking the vacuum. As shown in fig. 9 The Q-factor at 1,8 K was practically the same. That means that the RF test, done with variable

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coupling, has high reproducibility, if the cavity is kept under U.H.V. for both measurements and in beetween.
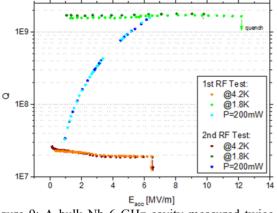

Figure 9: A bulk Nb 6 GHz cavity measured twice, just cooling the cavity down to He-II, then extracting from Helium and warming it out at room temperature without opening the cavity to air, then cooling it again to He-II. What is important is that during this double cycling from Room temperature to Liquid Helium, the cavity remains always under U.H.V.

Then we took the same cavity and we anodized the external obtaining a beautiful blue color as in fig. 10, always without breaking the vacuum. Then the cavity was rf Tested again at low temperature.

Figure 10: A 6 GHz cavity with a standard external surface (left). The same cavity after the RF test has been anodized only externally (right) and then re-measured. What is important to consider is that such operation is done when keeping the cavity on the stand always under U.H.V.

The obtained result (fig. 11) is that the external bleu anodization increases both the Q-factor and the accelerating field. But what is even more interesting is that when the anodized layer is removed we see that the Q is lowered. All operations were done without breaking the vacuum.

Our explanation considers several factors: a) the Niobium oxide has a lower Debye Temperature than pure niobium, so the Kapitza resistance will be lower; b) a specular surface will reflect phonons while the anodization produces a mat surface; c) the external oxide is more porous and rough, enhancing the possible nucleation of Helium bubbles. Most probably, the
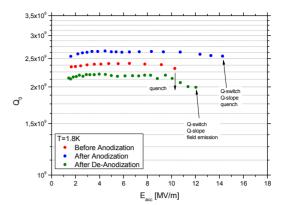

Figure 11: RF test of the cavity of fig. 9. The read curve refers to the cavity prepared in a standard way. The blue curve refers to the same cavity after anodization of the external surface. The green curve refers to the same cavity after the stripping of the external anodization. What is important to consider is that such operations were done by never exposing the cavity interior to the air.

observed effect is a linear combination of those fore different factors.

However the surface specularity seems to definitely affect the cavity performances. Fig. 12 indeed shows the Q vs Eacc Curve for a standard 6 GHz cavity before and after external electropolishing.
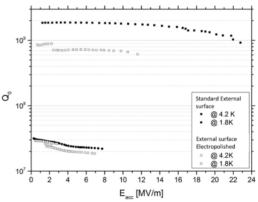

Figure 12: A 6 GHz cavity treated in standard way, and the same cavity after the electropolishing of the only external surface

The authors think that a smooth and specular surface as can result after electropolishing will have a double problem: it will reflect phonons and especially in He-I it will induce superheating in liquid Helium. In our judgment, superheating in He-I can be rather detrimental for cavity performances. Last test performed on the Nb external surface consisted in dipping a cavity before in water and then immediately cooling down to Helium the wet cavity in order to have a kind of ice film outside of the cavity. Fig. 13 reports a slight increase both in field and in figure of merit. We suppose that ice on the external wall of the cavity will promote Helium bubbles nucleation on the surface of the cavity, promoting the thermal exchange.
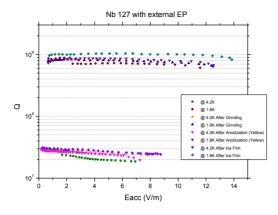
Figure 13: Q factor versus Accelerating field of a 6 GHz cavity measured after the standard surface treatment; after the grinding of the external surface, after a thicker anodization of the external surface; and after the growth of an ice film on the external surface. Again what is important to consider is that such operation is done when keeping the cavity on the stand always under U.H.V.

CONCLUSIONS

For years we have considered a cavity as an adiabatic system made by the RF fields + Niobium, because the He bath has been considered as a stable and infinite reservoir at fixed temperature. It is now the time to consider instead the adiabatic system composed by RF fields + Niobium + Liquid Helium.

We have discovered a jump in the Surface resistance versus temperature at the superfluid Lambda transition, when measuring at constant RF power. This jump, that happens when helium becomes superfluid, just proofs that thermal exchange is more important than what previously foreseen.

Starting from this consideration, we have seen that

- the external anodization of Niobium 6 GHz cavities is beneficial in terms of Q and accelerating field;
- the removal of such oxided layer is detrimental;

- a cavity electropolished outside has poorer performances;
- the superheating of liquid Helium is a not less importance of superheating of the Niobium material;
- an externally wet cavity immediately cooled in liquid He will result in better performances.

The Q variations in this paper are relatively small, but however existent. We retain that the main role of this paper is to have open a new horizon of research: the control of the external surface immersed in Liquid Helium. Working on the thermal boundary resistance will be certainly the way to a finer and more comprehensive understanding of superconducting cavity technology.

REFERENCES

[1] I.M. Khalatnikov, Sov. Phys (1951) JETP 22, 687.

[2] P.L. Kapitza, The Study of Heat Transfer on Helium II, J. Phys. (USSR) 4, 181 (1941).

[3] T.H.K. Frederking, Thermal Transport Phenomena at Liquid Helium II Temperatures, Adv. Cryog. Heat Transfer 64, 21 (1968).

[4] N.S. Snyder, Heat Transport through Helium II: Kapitza Conductance, Cryogenics Vol. 10, 89 (1970).

[5] L.J. Challis, Experimental Evidence for a Dependence of the Kapitza Conductance on the Debye Temperature of a Solid, Phys. Lett. 26A, 105 (1968).

[6] P. Dhakal, G. Ciovati, G.R. Mynemi, K.E. Gray, N. Groll, P. Maheshwari, D.M. McRae, R. Pike, T. Proslier, F. Stevie, R.P. Walsh, Q. Yang, J. Zasadzinski, Phys. Rev. Special topic, Accelerators and beams, 16, 042001(2013).

[7] V. Abet-Engels, C. Benvenuti, S. Calatroni, P. Darriulat, M.A. Peck, A.M. Valente, C.A. Van't Hof, NIM A 463 (2001).

[8] K. Saito, Q-slope analysis of niobium SC RF cavities, Proceedings of the 11th Workshop on RF Superconductivity, Lubeck/Travelmuender, Germany.