DEGRADATION OF NIOBIUM SUPERCONDUCTING RF CAVITIES DURING COOLING TIME

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Abstract

We report evidence that Niobium cavities that spend hours at temperatures between 100 K and 200 K, before completion of cooling, build up systematically an additional resistance on their RF surfaces, destroying their quality factor. They recover totally by a warm up to room temperature. Fast cooling prevents this phenomenon to develop. An attempt is done to characterize and modelize it.

Introduction

Continuous efforts in RF superconductivity technology have given enough confidence to start building large particle accelerators using Niobium cavities.

The design values commonly accepted are an accelerating field : $E_{acc} = 5$ MeV/m and a quality factor : $Q_0 = 2$ to 3.10^9 .

Much higher performances have been obtained in laboratory tests (10 to 20 MeV/m with Q above 10^{10}). However, bad reproducibility has plagued them. Recently, erratic degradations of vacuum sealed cavities have been reported by different laboratories.^{1,2,3,4} All laboratories present at a recent meeting at Cornell have shown concern on this subject.⁵

Investigations on this phenomenon have been initiated at Wuppertal.²

We report here on systematic experiments undertaken at Saclay that prove the existence of a reproducible phenomenon. We report also on the re-analysis of previous Saclay experiments that confirm it. We finally attempt to interpret the data on the basis of a simple model, assuming the formation of a uniform layer of a weak superconductor on top of Niobium surface.

Experimental evidence

Dedicated experiments were performed during summer 1990 on a 1.5 GHz single cell accelerating cavity, and on a pill-box cavity resonating at 4 GHz and 5.6 GHz (TE011 and TE012 modes).

a) Fabrication and preparation of cavities

A systematic procedure, mostly inherited from CERN, has always been followed for fabricating and preparing the cavities at Saclay. They are made by hydroforming and electron beam welding of high purity Niobium sheets (thickness : 2 mm, RRR 200 to 300). The surface preparation consists of a buffered chemical polishing (50 μ m) in a 1:1:2 solution of HF:HNO₃:H₃PO₄ followed by rinsing in ultrapure water. Drying and assembly take place under laminar air flow in a class 100 clean room. The final mounting on the cryostat vacuum system is made in front of a dust free laminar flow. The cavity is evacuated by a turbomolecular pump, then by an ion pump down to 10^{-7} to 10^{-8} mb, before inserting in a cryostat and cooling down.

b) Dedicated tests



Fig. 1. Thermal cycles on the 1.5 GHz cavity

We have observed strong variations of the quality factor Q of the two cavities after particular thermal cycles. Figure 1 shows all the thermal cycles done on the 1.5 GHz cavity. During all cycles, the cavities remained under sealed ultra-high vacuum. The results can be summarized as follows:

1 - A fast cool down from 300 K to 1.6 K (in about 1 hour) gives always the same Q characteristics : Q value is larger than 10^{10} and does not vary significantly upon the

applied RF field up to a limit (quench or electron loading) : see figure 2, curve a.

2 - A partial warm up from 1.6 K to some intermediate temperature for a variable time duration followed by a new cool down to 1.6 K gives always degraded characteristics. Temperature plateaus were controlled by coarse manual regulation. They lasted from 1 hour to 70 hours at temperature levels ranging from 120 K to 170 K. The 70 hours long plateau did not result in a larger Q degradation than the 3 hours one. Shorter plateaus lead to smaller Q degradations. Curves b, c and d on figure 2 show different levels of Q degradation.

In such cases, one always notices a particular characteristic of the Q behaviour : starting at low field from an already low Q, it steps down to a still much lower level for a surface magnetic field of about 6 mT at 1.6 K and remains then approximately constant.



Fig. 2. Q(E) after thermal cycles. Labels refer to fig. 1.

Table 1 shows the corresponding surface resistances for 2 cavities tested at three different frequencies. R0 stands for no degradation, R1 for degradation at low field and R2 for degradation at high field.

TABLE 1 Measured surface resistances

F GHz	R ₀ nΩ	R ₁ nΩ	R ₂ nΩ
1.5	20	350	1000
4	75	7100	13800
5.6	130	16600	24000

3 - A complete recovery of the original characteristics is always observed after a room temperature warm up. An extremely good reproducibility of the Q characteristics is observed.

c) A weak superconductor ?

In order to get better insight on the phenomenon, the surface resistances have been measured as a function of temperature between 4.2 K and 1.6 K. An "additional resistance" is calculated by subtracting the Niobium resistance (as measured in the non-degraded case) from the resistance measured in the degraded case.

Figure 3 shows the "additional resistance" as a function of temperature. The change of slope at 2.8 K and the field dependence (fig. 2) suggest the presence of a weak superconductor with a critical temperature of about 2.8 K and a critical magnetic field of about 9 mT.



Fig. 3. Additional resistance for test d of figs. 1 and 2.

Analysis of previous experiments

Erratic results on the Q behaviours, common in many laboratories, were more frequent at Saclay. Some of the Q degradations were traced back to accidental pollutions, but many of them remained unexplained. Most of the puzzling results were obtained in a particular cryostat, having two possible cooling speeds. Its slow cooling procedure (LN₂ precooling) brings the cavities to the very conditions that degrade the Q.

A scan through the records of the 37 tests in this cryostat shows that :

- 11 fast cooldowns lead to 100 % success,

- 16 slow cooldowns lead to 25 % success,

- 10 unidentified tests lead to 10 % success.

A complete scan of 105 cold tests performed at Saclay with 3 cryostats confirms the disastrous effect of too long a cooling time, except for a few cases.

Discussion

We have established that some large additional resistance appears reproducibly whenever a cavity stays long enough at temperatures around 150 K. This resistance disappears totally after warm up at 300 K.

One can think of a particular resistive component appearing either in the Niobium or in the Niobium oxide outer layer. This component should be unstable at 300 K and should build up around 150 K and stop growing at lower temperatures. It is present over all the surface, as shown by the quasi-uniform heating of such degraded cavities. The additional resistance has a frequency-squared dependence. This component behaves like a weak superconductor.

In order to derive parameters of this hypothetical component, we have used a model⁶ which calculates the surface resistance of Niobium on which a thin uniform layer of a weak superconductor is superimposed. Figure 4 shows the experimental surface resistance at 4 GHz as a function of temperature together with computed values for a layer of a thickness of 28 nm and a conductivity at 4 K of $1.6 \ 10^8 \ \Omega^{-1} \text{m}^{-1}$.



Fig. 4. Surface resistance measured at 4 GHz, and fit by a thin layer model.

The Sommerfeld constant of a superconductor deduced from the relation :

$$H_C = (\gamma/2\mu_0)^{1/2} T_C$$

gives in our case a value : $\gamma = 0.17 \text{ mJ.mole}^{-1} \text{.K}^{-2}$

This value is much smaller than the one of Nb ($\gamma = 7.82$). This leads us to the idea of a polluted state of Niobium.

Roth et al² have proposed, as a candidate for the weak superconductor, a Nb₄H₃ phase, which they quote to be unstable above 220 K. This is a very interesting possibility, in spite of a quoted critical temperature of only 1.2 K.

Questions

Many questions remain to answer :

Are the elements entering in the formation of the postulated component coming from the other parts of the vacuum system or are they present permanently in the Niobium surface?

What is the exact temperature range where this component builds up? At what speed does it form?

Why did some cavities give good results in spite of a slow cooldown? What are the initial conditions that play a key role in the apparition of the phenomenon?

Why has this effect never been observed at CERN⁷ or KEK ? Would it be because of the frequency scaling ?

Conclusion

A much better control on Q characteristics of superconducting cavities is to be expected from the knowledge of this effect. An immediate application to existing cavities and accelerators can improve their performances. Cryogenics costs can be dramatically reduced for future facilities, either by including constraints on the cooling speed of the cavities, or by preventing the effect to show up.

Finding an antidote to this poison will need a thorough understanding of surface physics at low temperatures.

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