MAGNETIC SCREENING OF NbN MULTILAYERS SAMPLES

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Abstract

In 2006 Gurevich proposed to use nanoscale layers of superconducting materials with high values of $H_c > H_c^{Nb}$ for magnetic shielding of bulk niobium to increase the breakdown magnetic field inside SC RF cavities [1].

We have deposited high quality "model" samples by magnetron sputtering on monocrystalline sapphire substrates. A 250 nm layer of niobium figures the bulk Nb. It was coated with a single and multi-stacks of NbN layers (25 or12 nm) separated by 15 nm MgO barriers, and characterized by X-Ray reflectivity and DC transport measurements.

DC or AC measurement of H_{C1} is an important goal for multilayer evaluation during the sample evaluation phase. A clear increase of H_{C1} at low frequency is promising indication since H_{C1} is expected to increase with frequency (see e.g. [2] and references therein). We have measured the first penetration field (H_P~H_{C1}) on DC magnetization curves in a SQUID system. H_P of NbN covered sample is increased compared to Nb alone. We have also developed a set-up that allows measuring a large range of field and temperature with a local probe method based on 3rd harmonic analysis. We have confirmed the screening behavior of a single 25 nm NbN layer placed on the top of a Nb Layer.

INTRODUCTION

The use of a superconductor allows building RF cavities with very high quality factors (Q_0 about 10^5 higher than for copper). This technology is the only one allowing having appreciable accelerating gradients (up to 40-45 MV/m) in continuous wave. Up today, Niobium is the only superconductor extensively used for this technology. Attempts to use other superconductors seem to have failed.

In principles, the limitation in accelerating field occurs when the magnetic component of the RF field reaches the RF critical field of the superconductor.

Nevertheless, the exact definition of the ultimate achievable field in a superconducting RF cavity is still in dispute.

Indeed superconducting state has been observed at fields higher than the DC measured value of H_{C1} , and one define a so-called "superheating field" applicable in RF [3]. In the 1960s, the field H_{SH} has been estimated on the basis of thermodynamic considerations related to the surface energy. Several experimental results seem to confirm this model of behavior.

According to the values of the Ginsburg-Landau (K) parameter, the « superheating » field can be approximated by the following expressions [3] :

$$H_{SH} \approx 1.2 H_c \text{ if } K \sim 1 \quad (1)$$

$$H_{SH} \approx 0.75 H_c \text{ if } K >> 1 \quad (2)$$

where H_C is the critical thermodynamic field.

For several decennia the commonly accepted explanation was that the field reverses every 10^{-9} seconds, whereas it takes 10^{-6} seconds to reach the nucleation of a normal zone. Therefore there is not enough time to trigger the nucleation.

If one follows this model, superconductors like NbN or Nb_3Sn should exhibit improved performances compare to Niobium. Only a few cavities of Nb_3Sn have been tested at various frequencies. They all exhibit a very high low field Q_0 as expected for a higher T_C material, but very soon the Q_0 degrades, and only poor performances where reached [4].

Recently the correctness of this model has been questioned [5-7]. We should consider the penetration of individual vortices, which is considerably faster (~10^{-13} s).

Moreover, at high fields and low temperature some approximations of the general theory (BCS) are no longer valid [8]. Some of the temperature corrections have been evaluated for type II superconductors in the clean limit and at low frequency. As H^{RF} approaches H_C, the normal electrons density and R_{BCS} increase due to the effect of current pair-breaking on thermal activation which in turn increases heating, making R_{BCS} nonlinear at high field. It shows that at high field the nonlinear correction increases exponentially with field and temperature, and can give rise to thermal runaway [6, 9]. This model can in particular explain the hot spots observed on cavities where bundles of trapped vortices can produce localized dissipative regions from which heat spreads over several tens of mm. The magnetic/vortex origin of the hot spots have been recently demonstrated [10].

If one consider the superheating model no vortices penetrates the superconductor until close to H_{SH}. But if on the other hand one assumes the high field dissipation are related to vortices penetration, then high field nonlinear dissipation could explain the monopoly of niobium in SRF applications since Nb has the highest H_{C1} value

(170-180 mT at 0 K) among all superconductors. High H_{C1} material is mandatory to prevent early vortex penetration on surface defects (asperities, grains boundaries…). Attempts to use higher T_C and H_{C2} superconductors have failed so far, probably due to their low H_{C1}, that allows early penetration of magnetic vortices resulting in high surface dissipation (for a recent review on that topic see [11]). In 2006 Gurevich proposed to use nanoscale multilayers of superconducting materials with high values of $H_C > H_C^{(Nb)}$ to shield bulk niobium and therefore to increase the breakdown field of Nb rf cavities [1]. Very high H_{C1} can indeed be achieved with films whose thickness d is smaller than the magnetic penetration depth λ, at least in a configuration where the field is parallel to the surface of the film [1]. So these films sustain high field without transition and without normal zone nucleation (vortex). The films screen bulk niobium and allow much higher field to be reached inside cavities.

Bulk niobium is still necessary to prevent perpendicular vortices to penetrate the film and an insulating layer (~ 15 nm) is needed to prevent Josephson coupling between coating layers and Nb substrate.

In order to test this idea, we have firstly to study the magnetic field penetration in multilayer structures. We have deposited model samples by DC magnetron sputtering on flat single crystal substrate. Preliminary dc magnetic measurements confirm experimentally this improved shielding effect.

Measuring H_{C1} challenges

One of the common ways to measure H_{C1} is SQUID magnetometry. In fact what is measured is H_P, the first penetration field, which is a combination of H_{C1}, demagnetization factor (related to the orientation of the sample compared to field) and edge effect (due to the quality of the cutting of the sample). Indeed it has been proved that depending on the cutting shape and cold work of the edge of the sample, large variation can appear on H_P. In case of ultrathin samples, the demagnetization factor is huge, and alignment becomes a concern.

Although SQUID magnetometer is a largely used technique, we feel that some ambiguity keeps in the results obtained by SQUID and we developed a local measurement that allows direct measurement of H_{C1}.

Note: other direct measures of H_{C1} like specific heat measurement require, for ultrathin films, very specific samples preparations that are hardly applicable to a large number of samples [12].

EXPERIMENTAL DETAILS

Deposition techniques

Nb/[(MgO)/NbN]n multilayer samples with n=0,1..4 have been deposited by DC magnetron sputtering on R-plane cut sapphire substrates. NbN is sputtered from a 6-inches diameter niobium target in a reactive (nitrogen/argon) gas mixture at 300°C. The same target is used for Nb deposition applying only argon pressure. Dielectric layers (MgO or AlN) are similarly RF-magnetron sputtered respectively from an MgO or an Al target. More details on the technique can be found in [13].

In this paper we will focus on the magnetic behavior of a selected number of representative samples:

i) Nb(250nm)/[MgO(14nm)/NbN(25nm)]n=1 named "SL".
ii) On a part of the wafer, the NbN top layer is further reactive ion etched to provide the bulk niobium reference sample, named "reference" or "R".
iii) A multilayer sample named "ML" Nb(250nm)/[(MgO(14nm)/NbN(12nm)]n=4

Characterization

Large angle X-rays Diffraction measurements provide information on the crystalline relations between the substrate and the deposited layers.

Low angle X-rays reflectivity gave information about thicknesses and interface roughness of the different layers.

The superconducting critical temperature T_C of each sample was measured using a Quantum Design PPMS facility.

Resulting data are summarized in table 1. Further details can be found in [6]

Table 1: summary of the X-rays reflectivity analysis

Reference Sample **R***	Thickness (nm)	Roughness (nm)	Sample **SL**	Thickness (nm)	Roughness (nm)	Sample **ML****	Thickness (nm)	Roughness (nm)
Tc = 8.9 K			**Tc = 16.37 K**			**Tc = 15.48 K**		
Nb	250	1	**Nb**	250	1	**Nb**	250	1
MgO	14	1	**MgO**	14	1	**MgO** ***	14	1
NbN	0	0	**NbN**	25	1.5	**NbN**	12	1.5

** Sample R contains only Nb capped with a layer of NbO. obtained by RIE etching of sample SL.*
*** In ML the motive MgO/NbN is repeated 4 times.*
**** Except the external, capping layer which is 5 nm*

SQUID results

For the purpose of comparison, we summarize hereafter results that have been published in details in [14]. SQUID samples are 5x5 mm^2 and were tested either in parallel or perpendicular to the magnetic field using a Quantum Design MPM S equipped with a setup which enables simultaneous measurements of M(H) for both transverse and longitudinal field orientation. Specific software developments were necessary to overcome cross talk between the two detecting loops due to misalignment. Note that without transverse measurement it is impossible to detect this effect, and slight misinterpretation of the results is possible, as in our former publication [15].

Figure 1 shows the longitudinal component of the moment of the Reference sample (250 nm Nb) measured at T= 4.5 K.

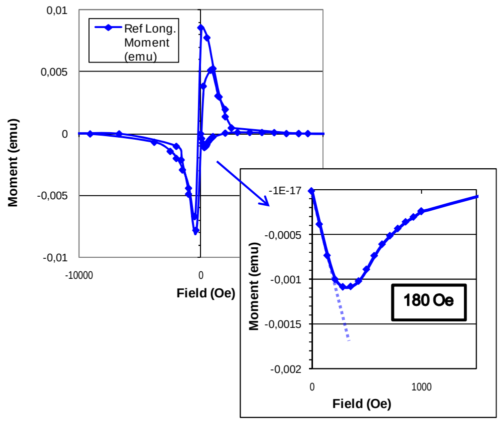

Figure 1: Magnetization curves at 4.5 K for the Reference sample (250 nm Nb): longitudinal moment at low field and determination of the first penetration field Bp. As expected for an isotropic superconductor, Bp is found to be the same in the transverse direction (not shown here).

The estimated first penetration field value is 18 mT for the Nb Reference sample, consistently with what has been observed previously in magnetron sputtered films [16]. Indeed, physically or chemically deposited films have on one hand a longer London penetration, on the other hand a reduced electronic mean free path compared to bulk Nb due to local defects inducing a reduced coherence length and so exhibiting lower B_{C1} and higher B_{C2} than the bulk material (B_{C1} ~150 mT at 4.5 K for clean bulk Nb).

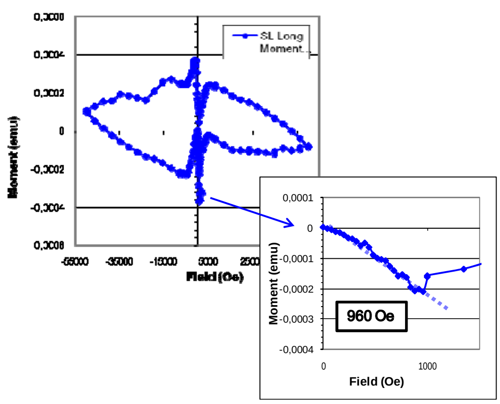

Figure 2: Magnetization curves at 4.5 K for the SL sample: longitudinal moment. Inset shows the determination of the first penetration field Bp.

Figure 2 shows the magnetization curve for the SL samples (25 nm NbN layer). In this case the longitudinal moment, exhibits a fishtail shape commonly observed on thin slabs or films of type II superconductors [15-17]. The first penetration field in SL in the longitudinal direction is greatly enhanced and reaches 960 Oe, ~ +78 mT times higher than the Reference's one. **If one could get a film this quality inside a cavity (a though experiment at this stage !), it would mean that we could gain ~ 20 MV/m in a Tesla shaped elliptical cavity with one single 25 nm layer of NbN.**

DC magnetization data on a multilayer sample Nb(250nm)/[(MgO(14nm)/NbN(12nm)]n=4 presents qualitatively the same general features as SL with a similar field enhancement (not shown here).

Nevertheless these samples exhibit a strong transverse signal, which means that they are not perfectly aligned, and that the (small) perpendicular field component is sufficient to let vortices enter inside the material. We therefore do not know the exact field configuration which is a combination of the uniform applied field and the remnant perpendicular moment. Neither are we able to quantify the edge effect and the quality of the cutting or our samples. Therefore we felt it is important to measure H_{C1} with another approach.

3RD HARMONIC ANALYSIS

Technical description

We have performed B_{C1} measurements based on ac third harmonic (V3) analysis as developed in ref. [2, 17, 18]. This technique is based on the hysteretic behavior of the magnetization in the critical state, which gives rise to none zero odd harmonics in the spectrum of the electrodynamic response of superconductors exposed to an AC magnetic field $b_0 cos\,\omega t$.

Initially these kind of facility where designed to measure λ_L or Jc close to TC, therefore they have a limited range of field and temperature. After a successful test at low field on an existing facility in Napoli, we have developed a facility at Saclay with extended performances in field and temperature.

The experimental set-ups used for our measurements [2, 18] have two particular features:

i) AC applied field is perpendicular to the sample surface (worst case situation for SRF)

ii) it is produced by a pancake coil whose diameter must be smaller than sample the size (infinite slab approximation).

This last condition makes negligible any demagnetization or edge effect.

When the sample is in the Meissner state, it acts like a perfect magnetic mirror due to screening currents. The current in the coil keeps linear and the third harmonic V3(T) is strictly equal to zero. It acquires finite values with a bell-shape temperature dependence in the mixed state below the irreversibility line. After a zero field cooling, the temperature is raised at a fixed amplitude b0 of the AC applied field until a third harmonic signal appears and it comes back to zero in the flux flow and/or normal state regimes. For further details see ref.[2].

Local magnetometry experimental results

Figure 3 shows as an example a set of third harmonic signals for various applied field for the SL sample. For each field one can determine the temperature of the first penetration of vortices (left-hand threshold of the V3 curve) and hence reconstruct the B_{C1} vs T curve.

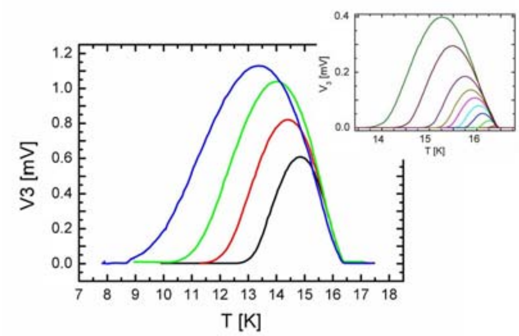

Figure 3: Set of third harmonic signals measured for an increasing series of AC magnetic field. Insert show an expansion of the scale for low fields. For each field one can determine the temperature of the first penetration of vortices (left-hand threshold of the V3 curve) and reconstruct the B_{C1} vs T curve.

Measurements at Napoli were limited to 16 mT, with no thermal regulation. Since eventually we would like to measure samples deposited onto bulk Nb, it is necessary to be able to get fields in the order of 200 mT, along with a thermal regulation that allows going below 2K. We have developed recently such facility at Saclay (see Figure 5).

A 5 mm diameter coil coiled with high conductivity copper 100 µm wire should produce > 200 mT (ultimate field not known yet) that should fit with 1.5 cm diameter samples.

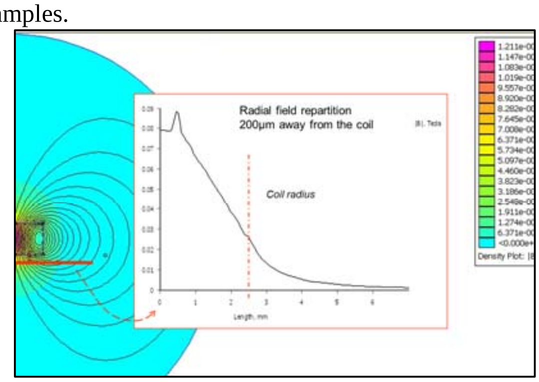

Figure 4: Field repartition for a 5 mm diameter coil. Inset shows the radial repartition 200 µm away from the coils (approximate position of the sample is expected to be closer to the coil: -~60µm).

One can note on Figure 4 that the field becomes negligible about 6 mm away from the center of the coil.

The whole facility is under vacuum to minimize the thermal transfer between the coil and the samples, and temperature can be continuously monitored between 1.9 and 70K.

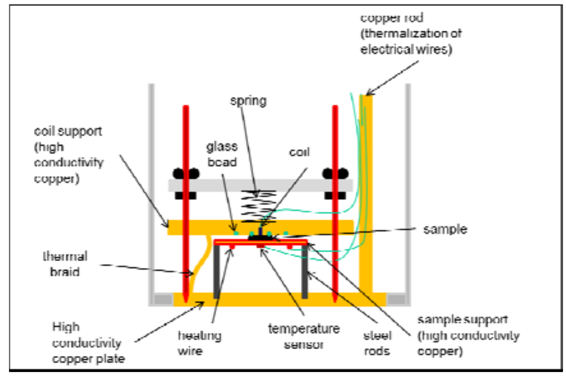

Figure 5: Scheme of the local magnetometry facility at Saclay.

The coil support is dismountable and can be replaced with one with a coil of different geometry, or even a superconducting coil, once we have assessed the third harmonic behavior of the coil itself. The calibration has been done by measuring the same sample as in Napoli, but hall sensor measurements are foreseen for confirmation. Figure 6 shows the B_{C1} curves of the SL sample compared to the Reference one. These results are very consistent with the SQUID and T_c measurements: we observe an increase of B_{C1} on the SL sample. At temperatures higher than the Nb underneath layer critical temperature (T>8.9K), the B_{C1} curve for SL is close to the curve of a single NbN layer (not shown here) whereas at for T<8.9K, when niobium is superconducting too, the B_{C1} of SL increases dramatically and becomes higher than the reference sample.

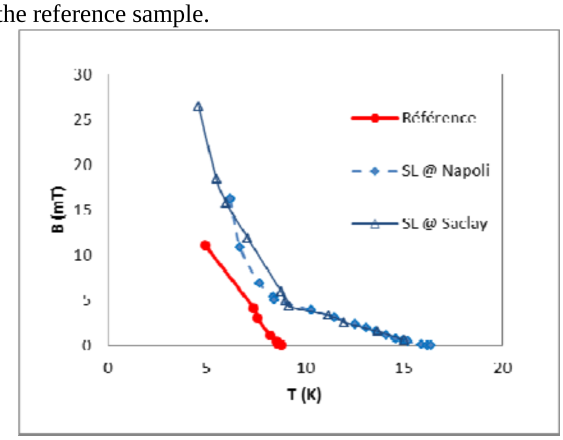

Figure 6: First penetration field for the Reference sample (red, circles) and the SL samples (blue, diamonds for Napoli experiment, triangles for the Saclay experiment) as determined with the third harmonic local probe. Around 6K, The first penetration of vortices in SL appears at a field value about twice the Reference's one.

At temperatures higher than the Nb underneath layer critical temperature (T>8.9K), the B_{C1} curve for SL is close to the curve of a single NbN layer (not shown here) whereas at for T<8.9K, when niobium is superconducting too, the B_{C1} of SL increases dramatically and becomes higher than the reference sample.

The first experimental data from Saclay have been added, showing that we still have to improve the thermal contact between the sample and the thermal sensor, but the behavior of the sample is overall the same, with a continuous increase of B_{C1} with decreasing temperature.

Higher field measurements of single and multilayer samples should be available very soon.

More interestingly, for fields low enough and T<8.9K it is possible to detect the third harmonic signal due to the niobium layer underneath the NbN one. In Figure 7 we show the B_{C1} curve obtained in this way compared to the Reference one: in the SL sample the Nb layer feels a field attenuated by the NbN layer and therefore its effective B_{C1} is higher than in the Reference sample. This observation provides a further direct confirmation of the attenuation due to the presence of NbN.

Even if the screening effect was originally predicted in the parallel field configuration, our data show that screening is effective even in the perpendicular field geometry as well. Deposition of multilayers inside RF cavities looks then promising: even if a realistic surface is not fully flat, small perpendicular field component should also be screened effectively.

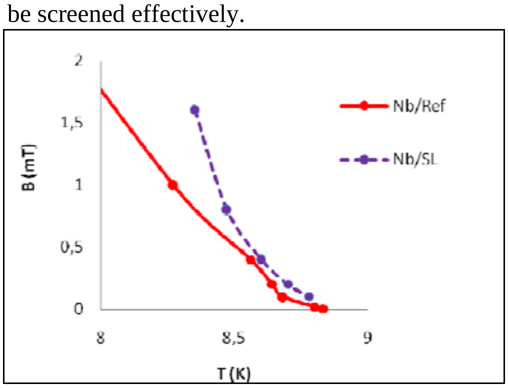

Figure 7: First penetration field for niobium in the Reference (continuous) and in the SL (dotted) (detail of the low field part of figure 5). The first penetration of vortices in niobium covered with a NbN layer (SL) appears at a apparent field values higher than the same niobium layer when directly measured in Reference. The Nb layer is effectively screened by the NbN layer, even in the perpendicular field configuration.

CONCLUSION AND PERSPECTIVES

We have established that nanometric NbN layers can screen effectively the field experienced by the underlying niobium with two different approaches: SQUID and local magnetometry. We have developed a unique facility able doing direct first penetration field of multilayer samples at high field and low temperature. Nevertheless DC measurement must be completed with RF measurement and we are looking forward to confirm these results on a TE011 cavity with demountable flat sample.

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REFERENCES

[1] A. Gurevich, "Enhancement of RF breakdown field of SC by multilayer coating". Appl. Phys.Lett., 2006. **88**: p. 012511.

[2] G. Lamura, et al., "First critical field measurements by third harmonic analysis ". Journal of Applied Physics, 2009. **106**: p. 053903

[3] H. Padamsee, J. Knobloch, and T. Hays, "RF superconductivity for accelerators". 1998: J. Willey & son.

[4] see e.g. T. Tagima, Tutorial, this conference

[5] E.H. Brandt. "Electrodynamics of superconductors exposed to high frequency fields". in International workshop on thin films and new ideas for pushing the limit of RF superconductivity. 2006. Legnaro National Laboratories (Padua) ITALY.

[6] A. Gurevich, "Multiscale mechanisms of SRF breakdown". Physica C, 2006. **441**(1-2): p. 38-43

[7] Saito. "Theoretical critical field in RF application". in SRF 03. 2003. Lubec.

[8] Voir par exemple : http://tdserver1.fnal.gov/project/workshops/RF_Materials/talks/Hasan_Sethna_superheating%20field.ppt

[9] P. Bauer, et al., "Evidence for non-linear BCS resistance in SRF cavities ". Physica C, 2006. **441**: p. 51–56.

[10] http://conferences.jlab.org/tfsrf/Tuesday/Tu2_3-Trapped_vortices_Ciovati.pdf

[11] http://tdserver1.fnal.gov/project/workshops/RF_Materials/talks/A-M_Valente-Feliciano_NewMaterialsOverview.ppt

[12] O. Bourgeois, Institut Neel (Grenoble, France), personnal communication.

[13] J.C. Villegier, et al., "Epitaxial growth of sputtered ultra-thin NbN layers and junctions on sapphire". IEEE Transactions on Applied Superconductivity, 2009. **19**(3): p. 3375-3378.

[14] C.Z. Antoine, et al., "Characterization of superconducting nanometric multilayer samples for SRF applications: first evidence of magnetic screening effect". PRST AB 2010. **13**: p. 121001.

[15] C.Z. Antoine, et al. "Characterization of superconducting multilayer samples". in SRF 2009. 2009. Berlin, Germany.
[16] N.I. Balalykin and A.B. Kuznetsov. "About the Amplitude Dependence of the Surface Resistance of Niobium Coated Copper Cavities". in SRF 97. 1997. Abano Terme (Padova), Italy.
[17] R. Russo, et al., "Niobium Coating of Cavities using Cathodic Arc". IEEE Transactions on Applied Superconductivity, 2009. **19**(3): p. 1394 - 1398
[18] M. Aurino, et al., "Discrete model analysis of the critical current-density measurements in superconducting thin films by a single-coil inductive method". Journal of Applied Physics, 2005. **98**: p. 123901.